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TRANSURANICS IN NATURAL ENVIRONMENTS

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A SYMPOSIUM AT GATLINBURG
OCTOBER, 1976

Nevada Applied Ecology Group
U.S. Energy Research &
Development Administration

PRINTED IN THE UNITED STATES OF AMERICA
AVAILABLE FROM:
NATIONAL TECHNICAL INFORMATION SERVICES, NTIS
U. S. DEPARTMENT OF COMMERCE
5285 PORT ROYAL ROAD
SPRINGFIELD, VA. 22161

PRICE: PRINTED COPY: $16.25
MICROFICHE: $3.00

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TRANSURANICS IN NATURAL ENVIRONMENTS

JUNE 1977

EDITED BY
M. G. White and P. B. Dunaway

NEVADA APPLIED ECOLOGY GROUP
UNITED STATES
ENERGY RESEARCH & DEVELOPMENT ADMINISTRATION
LAS VEGAS, NEVADA

TABLE OF CONTENTS

PAGE

PREFACE v

HISTORICAL REVIEW

Early Studies. *J. N. Stannard.* 1 6.4912

Off-Site Medical Activities, Nevada Test Site and the Medical Liaison Officer Network: A Historical Review. *E. van der Smissen* and *M. E. Kaye.* 17 6.4913

TRANSURANICS IN TERRESTRIAL ENVIRONMENTS

SOILS RESEARCH

Sampling of Soils for Transuranic Nuclides: A Review. *E. B. Fowler* and *E. H. Essington.* 25 6.4914

Distribution of Transuranic Radionuclides in Soils. A Review. *E. H. Essington* and *E. B. Fowler.* 41 6.4915

Factors Influencing the Chemical Extractability of ^{241}Am From a Contaminated Soil. *H. Nishita* and *M. Hamilton.* 77 6.4916

Plutonium Association in Soils. *T. Tamura.* 97 6.4917

Distribution of Plutonium and Americium at a Former Los Alamos Waste Disposal Site. *S. Fried, A. M. Friedman, J. Hines, G. Schmitz,* and *M. Wheeler.* 115 6.4918

MICROORGANISM RESEARCH

The Relationship of Microbial Processes to the Fate of Transuranic Elements in Soil. *R. E. Wildung, H. Drucker,* and *F. H. F. Au.* 127 6.4919

TRANSURANICS IN TERRESTRIAL ENVIRONMENTS (Cont.)

RESUSPENSION AND PARTICLE RESEARCH

Estimation of Aerosol Plutonium Transport by the Dust-flux Method: A Perspective on Application of Detailed Data. *J. H. Shinn.* 171

Plutonium and Tracer Particle Resuspension: An Overview of Selected Battelle-Northwest Experiments. *G. A. Sehmel.* 181

A Review of Resuspension Models. *J. W. Healy.* 211

Brief Survey of Particle Work Relevant to the Test Sites. *M. W. Nathans* and *L. Leventhal.* 223

Measurements of Americium and Plutonium in the Atmosphere. *D. W. Efurd, G. L. Merrill, Jr., J. T. Phelps,* and *K. D. Rosenlof.* 249

Development of Specialized Instruments and Techniques for Resuspension Studies. *P. L. Phelps* and *L. R. Anspaugh.* 273

VEGETATION RESEARCH

Plutonium Contamination of Vegetation in Dusty Field Environments. *E. M. Romney* and *A. Wallace.* 287

Foliar and Root Pathways of Plutonium Contamination of Vegetation. *R. C. Dahlman* and *K. W. McLeod.* 303

Root Uptake of Transuranic Elements. *R. K. Schulz.* 321

Retention, Absorption, and Translocation of Foliar Contaminants. *D. A. Cataldo* and *B. E. Vaughn.* 331

Chronic Radiation Induced Chromosomal Aberrations in Native Shrubs at Nevada Test Site. *D. M. Varney* and *W. A. Rhoads.* 351

TRANSURANICS IN TERRESTRIAL ENVIRONMENTS (Cont.)

ANIMAL RESEARCH

Transuranic Elements in Terrestrial Animals and the Environment: An Introduction. *G. D. Potter.* 379

FIELD STUDIES

Plutonium and Other Transuranics in Small Vertebrates: A Review. *W. G. Bradley, K. S. Moor,* and *S. R. Naegle.* 385

Review of Grazing Studies on Plutonium-Contaminated Rangelands. *D. D. Smith.* 407

LABORATORY STUDIES

Application of the Artificial Rumen and Simulated Bovine Gastrointestinal Fluids Procedure in the Study of the Bioavailability of Transuranics. *J. Barth.* 419

Transport of Plutonium Via Food Products of Animal Origin. *W. W. Sutton* and *A. A. Mullen.* 435

TRANSURANICS RESEARCH IN AQUATIC ENVIRONMENTS

Transuranic Speciation in the Environment. *E. A. Bondietti* and *F. H. Sweeton.* 449

Distribution Patterns and Transport of Plutonium in Freshwater Environments With Emphasis on Primary Producers. *L. D. Eyman* and *J. R. Trabalka.* 477

Plutonium-237: An Important Research Tool for Studies of Environmental Plutonium Kinetics. *J. R. Trabalka* and *L. D. Eyman.* 489

Distribution of ^{238}Pu and ^{239,240}Pu in Aquatic Macrophytes From a Midwestern Watershed. *C. W. Wayman, C. E. Bartelt,* and *J. J. Alberts.* 505

^{238}Pu and ^{239,240}Pu Distribution in Fish and Invertebrates From the Great Miami River, Ohio. *G. E. Bartelt, C. W. Wayman, S. E. Groves,* and *J. J. Alberts.* 517

Effects of Naturally Occurring Organics on Plutonium-237 Uptake by Algae and Bacteria. *J. P. Giesy, D. Paine,* and *L. W. Hersloff.* 531

ANALYSIS METHODOLOGY FOR TRANSURANICS

Historical Review of Transuranic Analytical Methodology. *R. A. Wessman* and *L. Leventhal.* 545

TRANSURANICS STATISTICS AND MODELING

Some Design Aspects of Transuranic Field Studies. *R. O. Gilbert* and *L. L. Eberhardt.* 575

Some New Statistical Concepts for Quality Control. *R. R. Kinnison* and *A. N. Jarvis.* 593

Ratio Estimation Techniques in the Analysis of Environmental Transuranic Data. *P. G. Doctor* and *R. O. Gilbert.* 601

Nevada Applied Ecology Group Model for Estimating Plutonium Transport and Dose to Man. *W. E. Martin* and *S. G. Bloom.* 621

LIST OF AUTHORS AND PARTICIPANTS 707

OTHER NAEG PUBLICATIONS *

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*Inside back cover.

Preface

PREFACE

Perhaps the most important step in any organized scientific effort is the periodic review and evaluation of the known and the unknown aspects of a problem. Identification and acknowledgment of relationships and consideration of alternative courses of investigation are extremely important steps in attempt to reach solutions to a problem; or, at best, to gain information and guidance for moving out into the unknown.

The Nevada Applied Ecology Group Symposium on the Dynamics of Transuranics in Terrestrial and Aquatic Environments was held in October, 1976, at Gatlinburg, Tennessee. The symposium was planned to bring about a concerted review and evaluation of the current status of a major environmental problem and the associated body of information available to investigators involved in studies of environmental transuranics.

Papers selected for publication in this document are coauthored by scientists and other technical and professional people from several national laboratories, academic institutions, private corporations, and government agencies. It is recognized that not all aspects of environmental transuranics are covered in this publication. Reports by M. Wahlgren, Argonne National Laboratory (read by J. Alberts, ANL), and J. Pinder, Savannah River Ecology Laboratory, were not received for publication, and a report by A. L. Boni and R. W. Taylor, SRL, E.I. DuPont, "Plutonium Isotope Distribution in U.S. Surface Waters," was not cleared for publication. It is hoped that those areas of environmental transuranics research not presented in this document will be included in publications currently underway or planned by other research groups.

An addition to environmental transuranics literature since the Gatlinburg symposium was the two-volume NEVADA APPLIED ECOLOGY GROUP PROCEDURES HANDBOOK FOR ENVIRONMENTAL TRANSURANICS, NVO-166. The NAEG procedures handbook (dated October, 1976, released in May, 1977) includes most of the NAEG standard procedures used in applied environmental plutonium and other transuranics sampling at the Nevada Test Site, with related laboratory and statistical procedures. The handbook is available from the National Technical Information Services, NTIS, Springfield, Virginia.

Our gratitude is extended to the symposium authors and participants; to arrangements personnel (ORNL); to the headquarters, field, and laboratory people who encouraged and contributed to the symposium, especially Gordon Facer, HQ ERDA/DMA; Mahlon E. Gates, Manager, ERDA/NV; Roger Ray, Assistant Manager for Environment and Safety, ERDA/NV; Henry B. Gayle, Paul G. Noblitt, Timothy M. Catt, and the Word Processing Center, Holmes & Narver, Inc., Las Vegas; Winnie Howard and Don L. Wireman, NAEG staff; and to certain "senior" scientists who shared with the participants their perspective of many years, J. Newell Stannard, Eugene van der Smissen, and Eric B. Fowler.

The cover design indicates the volcanic nature of the high mesa area as related to lower Mojave Desert playas and Cane Springs on the Nevada Test Site.

In addition to serving as a statement of the status of research on associated problems, hopefully this document will assist in evaluation and guidance for future efforts and priorities for environmental transuranics investigations, reaching toward a better environment for man.

Mary G. White
Scientific Program Manager
Nevada Applied Ecology Group

Paul B. Dunaway
Chairman, Steering Committee
Nevada Applied Ecology Group

Historical Review

EARLY STUDIES

J. N. Stannard
University of Rochester, Rochester

INTRODUCTION

The organizers of this symposium anticipated that it would attract a number of younger radiation ecologists who might benefit from, perhaps even enjoy, a short discussion of the early beginnings of biomedical work on plutonium. It was originally planned that this paper would review the early laboratory studies and would be followed by a similar review of field tests and field studies. The speaker who was to present this aspect could not be present but this fact was not known in time to modify the substance of the present paper. Therefore, the title should be read as "Early Laboratory Studies."

It does happen that the older field studies were reviewed in part, but not in any sense completely, in a chapter on "Plutonium in the Environment" which I prepared for the review volume, Uranium, Plutonium, and Transplutonic Elements (Hodges *et al.*, 1973). I would be glad to try to provide copies of the pertinent sections to those interested. There are, of course, other and more exhaustive reviews of the field work but not many are in the open literature (i.e., nongovernmental literature with broad distribution).

The term "early" in the title above may be regarded as "early early" by many of you since it reaches back to the work done under the auspices of the Manhattan District during World War II beginning in 1943. It emphasizes the biomedical work done during those years on through the 1950s and just into the 1960s under the aegis of the U.S. Atomic Energy Commission and counterpart agencies abroad. Much of it has been reviewed before (see, for example, Langham and Healy, 1973) but not in references generally available to members of this audience. Hence, I will proceed as if you were meeting the information for the first time, but with apologies to some of the old-timers also present here who know the story as well as I do or better. Some of you may be surprised at the amount and quality of the work done under great pressures of time and what would be regarded now as slow and cumbersome methodologies.

THE FIRST BIOMEDICAL STUDIES

In early 1943, there existed about 2 milligrams of plutonium produced at the Berkeley cyclotron. This was distributed over the entire research program and was as Seaborg (1970) said, in retrospect, "a quantity so precious we couldn't afford to ingest any of it." (3) The potential external and internal radiation hazards associated with the production of large quantities of fission products in the newly developed chain-reacting pile demanded the full attention of the Plutonium Project's Health Division headed by R. S. Stone, and this miniscule quantity of plutonium had to be ignored.

However, the situation changed very rapidly. The Clinton reactor ("Site X," i.e., Oak Ridge) began producing milligram quantities of plutonium isotopes beginning in late 1943. In early 1944, gram amounts began to be produced with the start-up of the Hanford reactors ("Site W") and by mid-1945, kilogram quantities were in existence.

The potential health hazards of plutonium seem to have been first appreciated by Glenn Seaborg. As described in detail in the Langham and Healy (1973) paper already cited, in January, 1944, Seaborg (1944a) expressed his concerns to Dr. Stone in a letter dated January 5, 1944, and ten days later Seaborg (1944b) suggested that 10 mg. of plutonium be made available to Joseph G. Hamilton and his associates at the Crocker Laboratory at Berkeley. Their initial job was to determine its metabolism, i.e., its distribution and excretion in contradistinction to its effects. This was done forthwith using laboratory mice, and in a remarkably short time, results were reported. The plutonium was delivered to Hamilton on February 10, 1944, and the laboratory's progress report for February (Allison, 1944) contains the following:

> "Product studies--oral absorption of all valence states is less
> than 0.05%
> Lung retention is high
> Absorbed material is predominantly in the skeleton
> Excretion is very small in urine and feces

How is that for promptness? Today, the preliminary paperwork would have only begun in one month.

Thus, it appeared that plutonium acted like radium in that it was a boneseeker but it was less readily absorbed and more avidly retained, especially in lung. It was this behavior analogy to radium which became the basis for the first attempt to set an exposure limit for plutonium.

CONTINUING METABOLIC STUDIES

As more plutonium became available, these first observations were extended in Hamilton's laboratory and with the organization and development of laboratory programs, notably at the Chicago "Metallurgical Laboratory" and the project at the University of Rochester School of Medicine and Dentistry, more detailed information was obtained and the several species added. In late 1946, R. D. Finkle from Chicago and reports from the group at Rochester confirmed the basic facts enumerated by Hamilton (Finkle, 1946). But it also began to be clear that the analogy to radium was only a general one. Many differences between plutonium and radium became apparent. Chief among these was the detailed behavior in bone. Largely from autoradiographic studies in Hamilton's laboratory, it was clear that plutonium was much less uniformly distributed in bone than was radium (which is not uniformly distributed itself). Particularly, it appeared that plutonium was much more likely to collect on surfaces and interfaces in bone and did not enter the mineral matrix except very slowly and it did not exchange for calcium as does radium.

The significance of this for toxicity was appreciated and added impetus to the direct measurements of toxicity in animals already begun at Chicago and Rochester.

We will return to this point. But first we must examine what was being done to learn about the metabolism of plutonium in man.*

STUDIES OF METABOLISM IN MAN

While the laboratory studies were providing some of the needed information, the rapid growth in numbers of workers potentially exposed forced quick decisions regarding maximum permissible exposures. As always, the question persisted as to whether or not man handled plutonium in roughly the same way as the laboratory animals. This was approached in two ways--directly through the administration of what were then regarded as tracer quantities of plutonium to hospitalized patients considered to have short life expectancies and indirectly by meticulous studies of plutonium excretion by workers.

The study with patients was performed in 1945. It involved 16 individuals over 45 years of age who, by the nature of their disease, were considered unlikely to live long enough for the plutonium to exert any toxic effect. The

---

*Important animal laboratory studies have been conducted in both the United Kingdom and the Soviet Union. But both the work and their publication are much later than the period of primary focus for this review.

majority of the patients were at Strong Memorial Hospital in Rochester, New York, where there was not only an active biomedical project under the Manhattan District, but an active and well-managed Metabolism Unit in the Department of Medicine. The four others were at California and Chicago with one individual at Oak Ridge.

The experiment was done entirely to study metabolism, not effects, and involved primarily excretion. The results became available fairly soon in the classified literature and indicated sufficient agreement between the excretory patterns of man and the laboratory animals to reassure those concerned with standard setting that there was not a huge species difference. But the data obtained on tissue distribution were meager and the highly classified nature of the effort prevented any general dissemination of the information for a considerable period.

As a sidelight on how some of these things had to be done, most of the plutonium analyses on the Rochester patients were done at Los Alamos and only the heroic trundling back and forth of the late Wright Langham between Los Alamos and Rochester, by train arriving at Rochester in the inevitable snowstorm, enabled completion of the project. The report (Langham *et al.*, 1950) contains the names of those who assisted at Rochester, but it was a Los Alamos report and essentially no identification of the laboratories was discernable. Because of the sensitive nature of the subject, including the fact that some of the patients defied their prognoses and have lived well into their eighties, that report remained classified at various levels down finally to "Official Use Only" until very recently. Fortunately, Pat Durbin has gone back over all of these data recently and reanalyzed them in the light of current knowledge. Thus, you can read a modern interpretation of the meaning of these pioneer efforts.

Short reports of distribution and excretion studies in man at the California and Chicago laboratories appeared as "CH" reports (Crowley, *et al.*, 1946, and Russell and Nickson, 1946). I do not know how long these data remained sequestered.

The other primary source of early data on the metabolism of plutonium in man was the group of workers at Los Alamos. As you know, some of these have remained available for the long-term study--a facet beyond the scope of this presentation. But there were many more than these and all were subject to routine collection of excreta as a means of bioassay. (Remember that the crystal counters we now employ to detect the low energy photons from plutonium by external measurement had not yet been invented.)

These studies of excretion in workers and correlation with animal studies led to the formulation of the so-called Langham (1957) equation for expressing excretion as a function of time. Used for many years before publication, it has served health physicists faithfully, if not always precisely, for many years.

We now have modifications of the equations and methodology and computer programs to check the fit of the data. But I submit that the construction and use of the original concept and technique was remarkable when one considers how little plutonium is excreted except in feces soon after inhalation.

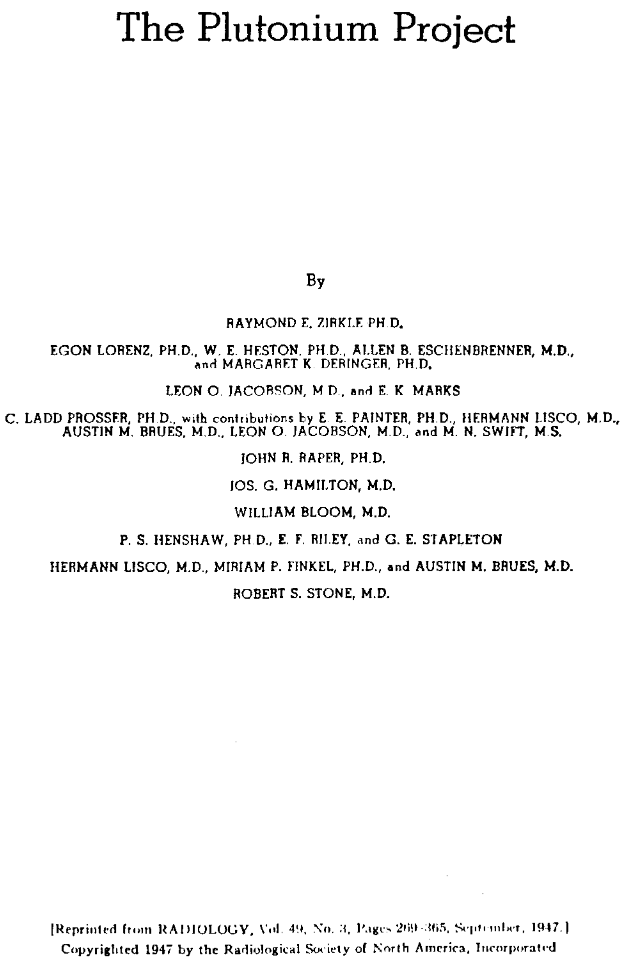
The fact that the microdistribution of plutonium in bone was found early not to be similar at all to that of radium has already been emphasized. This led to some speculation that its toxicity--or more specifically, its effects in bone--might not be proportional to its energy and half-life relative to radium. But first it needed to be known if plutonium would be carcinogenic in the same sense as radium and to what degree.

The answer came primarily, or at least first, from the Chicago group. Among the first of the postwar open literature publications which revealed what had been going on in "The Plutonium Project" was a special issue of Radiology (Anon., 1947) entitled "The Plutonium Project." The title page and table of contents of this now classical document are reproduced here as Figures 1 and 2. Note first that much of the work concerns external radiation and aspects of the project other than plutonium itself. But note also the presence of a summary paper on the metabolism of the fission products and the heavy elements by Joseph G. Hamilton and, especially for our purpose here, the paper on carcinogenic properties of radioactive fission products and of plutonium by Hermann Lisco, Miriam P. Finkel, and Austin M. Brues. This paper established for the first time in the open literature the carcinogenic potency of plutonium. The paper by Bloom in the same issue summarized other effects and touched upon carcinogenicity (see Section E. also), but it was the Finkel, Brues, *et al.* team which followed through in determining the relative effectiveness of different nuclides in producing bone cancer, as will be described presently.

Although the work at Rochester was under way at about the same time, it was not officially part of "The Plutonium Project" and reached the open literature somewhat later. Besides the metabolic studies in animals and in humans referenced earlier, the Rochester group carried out two laborious studies, one quite large, comparing the toxicity of polonium-210, plutonium-239, and radium-226 in rats. The doses were given intravenously, were relatively large, and the endpoint was lethality. Although published later, it is clear that these experiments established at the time of their performance the fact that plutonium-239 was more toxic even as measured by lethality in a relatively short time than radium-226 (Fink, 1950).

Because of the isolation of the various projects for security purposes, it is hard to say that these experiments confirmed the surmises of others based upon the autoradiographic evidence that since plutonium deposited and remained nearer to living cells in bone, it might be more toxic than per unit of dose than its prototype radium-226. But from the vantage point of our present knowledge, the facts are quite consistent with such a view.

Full confirmation of the difference in carcinogenicity in bone between plutonium and radium came in 1953 in a now classic paper by Miriam P. Finkel (1953). She compared the average probability of the CF1 female mouse dying with a malignant bone tumor as a function of isotope dose. And she studied not only



The Plutonium Project

By

RAYMOND E. ZIRKLE PH.D.

EGON LORENZ, PH.D., W. E. HESTON, PH.D., ALLEN B. ESCHENBRENNER, M.D.,
and MARGARET K. DERINGER, PH.D.

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C. LADD PROSSER, PH.D., with contributions by E. E. PAINTER, PH.D., HERMANN LISCO, M.D.,
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ROBERT S. STONE, M.D.

[Reprinted from RADIOLOGY, Vol. 49, No. 3, Pages 269-365, September, 1947.]
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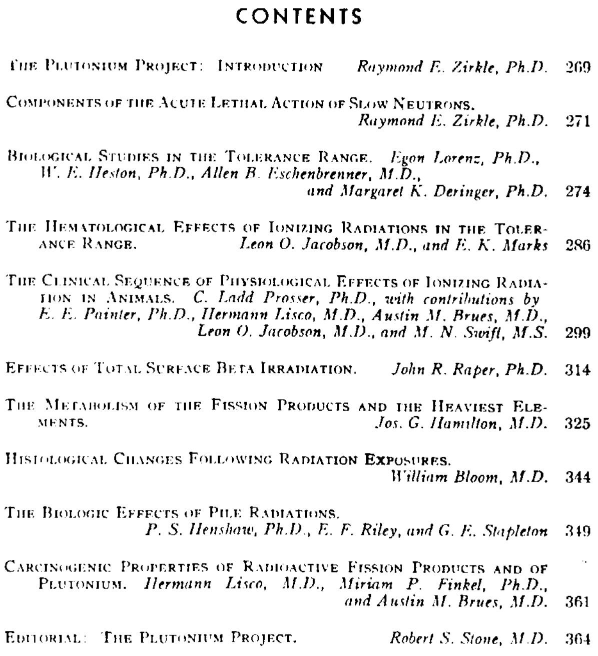
Figure 1. Title page of the first open literature report on the work of "The Plutonium Project" during World War II.



CONTENTS

THE PLUTONIUM PROJECT: INTRODUCTION *Raymond E. Zirkle, Ph.D.* 269

COMPONENTS OF THE ACUTE LETHAL ACTION OF SLOW NEUTRONS. *Raymond E. Zirkle, Ph.D.* 271

BIOLOGICAL STUDIES IN THE TOLERANCE RANGE. *Egon Lorenz, Ph.D., W. E. Heston, Ph.D., Allen B. Eschenbrenner, M.D., and Margaret K. Deringer, Ph.D.* 274

THE HEMATOLOGICAL EFFECTS OF IONIZING RADIATIONS IN THE TOLERANCE RANGE. *Leon O. Jacobson, M.D., and E. K. Marks* 286

THE CLINICAL SEQUENCE OF PHYSIOLOGICAL EFFECTS OF IONIZING RADIATION IN ANIMALS. *C. Ladd Prosser, Ph.D., with contributions by E. E. Painter, Ph.D., Hermann Lisco, M.D., Austin M. Brues, M.D., Leon O. Jacobson, M.D., and M. N. Swift, M.S.* 299

EFFECTS OF TOTAL SURFACE BETA IRRADIATION. *John R. Raper, Ph.D.* 314

THE METABOLISM OF THE FISSION PRODUCTS AND THE HEAVIEST ELEMENTS. *Jos. G. Hamilton, M.D.* 325

HISTOLOGICAL CHANGES FOLLOWING RADIATION EXPOSURES. *William Bloom, M.D.* 344

THE BIOLOGIC EFFECTS OF PILE RADIATIONS. *P. S. Henshaw, Ph.D., E. F. Riley, and G. E. Stapleton* 349

CARCINOGENIC PROPERTIES OF RADIOACTIVE FISSION PRODUCTS AND OF PLUTONIUM. *Hermann Lisco, M.D., Miriam P. Finkel, Ph.D., and Austin M. Brues, M.D.* 361

EDITORIAL: THE PLUTONIUM PROJECT. *Robert S. Stone, M.D.* 364

Figure 2. Table of contents of the special issue of Radiology devoted to The Plutonium Project. Note that the work extended far beyond plutonium itself.

^{239}Pu and ^{236}Ra, but ^{232}U, ^{233}U, ^{90}Sr, and ^{45}Ca.* Figure 1 from her paper is reproduced herewith as Figure 3. The greater effectiveness of ^{239}Pu on a μCi/kg injected dose basis is very clear. Others have calculated the relative radiation doses and while the ratios change somewhat, the basic relationships between plutonium and radium are the same on either an administered dose or calculated radiation dose basis. In Figure 4 (Dr. Finkel's Figure 2), the comparative lethality is shown using radium-226 as the baseline.

It was this information which, more than anything else, influenced the decision in 1950 to consider plutonium-239 as at least five times as effective as radium-226 in producing osteogenic sarcoma (Langham, *et al.*, 1973).

These toxicity studies led directly to the founding of the beagle dog experiment at the University of Utah. After all, there was still a large gulf in probable effective doses, lifespan, tumor susceptibility, etc., between the CF1 female mouse and man. The beagle experiment was a much needed intermediate step. But while its inception date might fit the caption of "early work," the results have been coming to full flower only recently. Suffice it to say here that the basic relationships seen in the mouse have appeared also and held in the dog. Also, the addition of ^{228}Th to the Utah experiment helped to sustain the suspicion, based on earlier radiochemical data, that plutonium and thorium might have more in common than plutonium and radium-226. A brief glimpse at some of the Utah results on comparative toxicity is shown in Figure 5, taken from Dougherty and Mays (1969).

INHALATION STUDIES

We are now accustomed, particularly in considering environmental exposures to plutonium, to regard inhalation as the most likely route of entry. Nearly all of the above toxicity studies involved intravenous injection. Was there anything in the early work pertinent to our present concerns regarding the production of lung cancer by plutonium?

Admittedly, inhalation studies were slower in coming about because of the large technical difficulties of preparing and administering controlled aerosols of radioactive materials. There were some rather crude inhalation experiments performed using hot wires to produce the aerosols. But the primary early information was obtained by the technique of intratracheal intubation. We now know that this direct instillation of material is not a satisfactory substitute for true inhalation experiments. But the results were useful in giving first approximations regarding pulmonary retention and clearance, even effects.

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*The fact that ^{232}U and ^{233}U were employed in this work has only recently been fully appreciated. The relationships shown here are entering into current thinking about uranium standards.

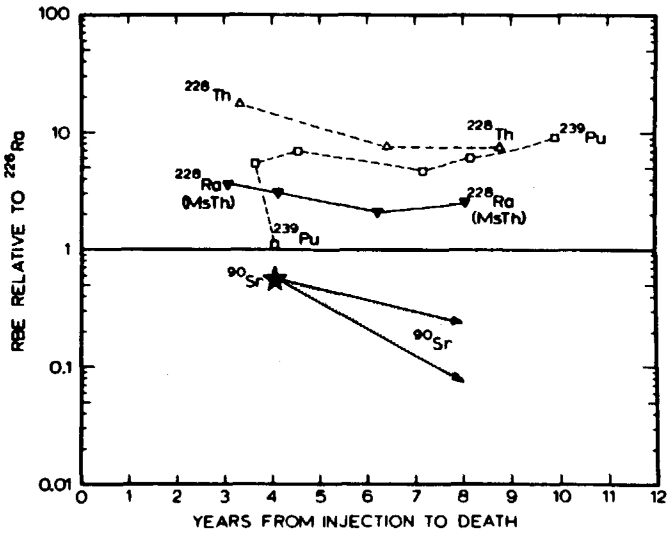


Figure 3. Bone tumor induction in the CF1 female mouse. From Finkel (1953).

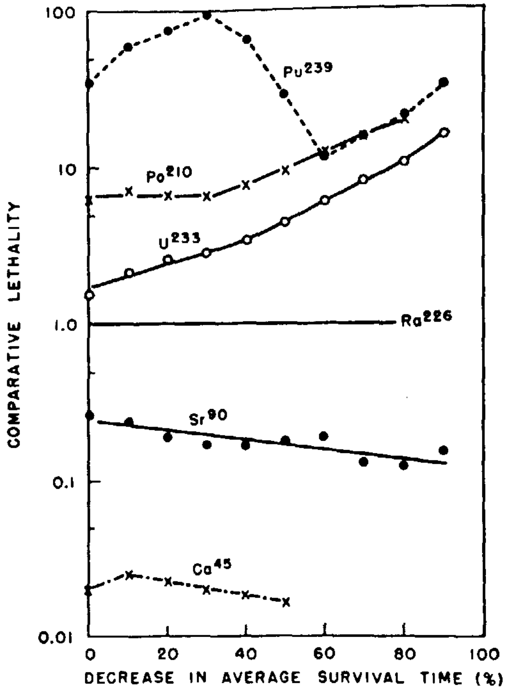
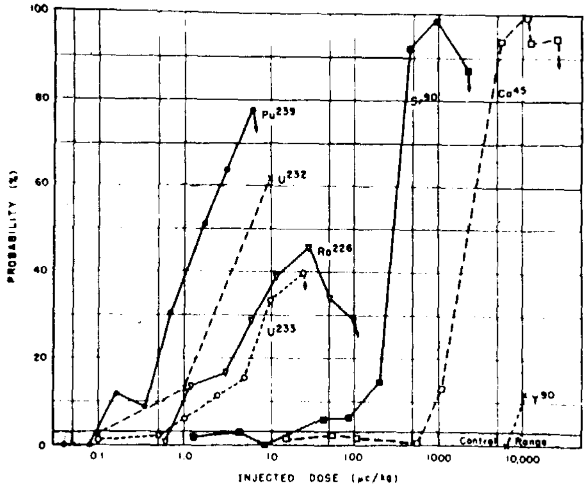


Figure 4. Comparative lethality from bone tumor induction in the CF1 female mouse. From Finkel (1953).



Average probability of dying with a malignant bone tumor as a function of isotope dose.

Figure 5. Comparative lethality of several elements of the beagle dog. Bone tumor development was the primary cause of lethality. The effects are expressed as relative biological effectiveness (RBE) relative to ^{226}Ra. From Dougherty and Mays (1969).

The laboratories at Rochester and Hanford were most active in inhalation studies, although some were also done at Chicago. The Rochester group turned its efforts primarily to uranium inhalation and to some work with inhaled polonium-210 but did not begin work with plutonium aerosols until much later than the period of this review.

The early statement from Hamilton that "lung retention is high" was confirmed in the work at Hanford. However, it became apparent that the nature of the compound made a large difference, with plutonium oxide the most avidly retained form and some others relatively soluble.

Credit for the first demonstration of a lung tumor induced by plutonium goes to Ralph Wager (1956), who did his work in an old Quonset hut on the Hanford reservation. He gave PuO_2 by intratracheal instillation to mice. After one year, 3 of 10 animals which had received 0.06 Ci of PuO_2 showed readily identifiable lung tumors. (Details as to mouse strain and type of tumor must be deferred to a more complete analysis.) This finding was the beginning of the enormous and fruitful efforts in inhalation toxicology of plutonium at the centers mentioned plus more recent entrants such as the Lovelace Group in Albuquerque.

THE NATIONAL NUCLEAR ENERGY SERIES

With the gradual lifting of classification restrictions of the late 40s and early 50s, it was realized that a set of books summarizing the bulk of the declassifiable information would be of enormous value. To accomplish this, the Atomic Energy Commission and the McGraw-Hill Book Company cooperated in getting out the equivalent of a Dr. Elliot's Five Foot Shelf for Nuclear Energy. This was called the National Nuclear Energy Series (NNES). Each of the primary research groups became a "Division" and a very ambitious assembly of titles and editors was produced. The subject matter included chemistry, physics, instrumentation, engineering, and biology and medicine. An enormous amount of information was assembled and published and the series is a fine source of information on the sort of thing I have been discussing this morning. Indeed I am surprised to find that many young investigators do not know of this series or use it as the starting point for their literature surveys.

Unfortunately, the interest of potential authors and editors began to lag by the mid-50s. They had more pressing current work to do. The excellent editorial board and staff had to be disbanded before all of the volumes could be completed. Thus, despite the NNES, a significant amount of detailed information remains sequestered in old files and on microfilms.

NOTE: For the oral presentation, the title page and table of contents of some of the NNES volumes most pertinent to biomedical laboratory work with plutonium were shown. To save space, only the references will be given here. They are Bloom (1948), Fink (1950), and Stone (1951).

CONCLUDING REMARKS

This summary has had to be truncated at many points. Also, for the reasons given at the outset, it has neglected almost entirely the early field and ecological type studies. While not, insofar as plutonium is concerned, as extensive or intensive as the laboratory studies, I wish there were time to at least outline them. But there is not and I will console myself with the thought that you are more likely to have heard of these than of the early laboratory studies.

Let me close by exercising one of the prerogatives of a teacher of more years than many of you are years old and share one of my pet peeves. This is the student who begins a graduate seminar on a given subject (and I have been to more such seminars than I can count) with a statement something like this: "I will begin my presentation by giving you a short history of the development of this field beginning with the classical studies of Jones and Smith in 1969." 1969 Indeed! Does he have no appreciation of the long line of his scientific forebearers who struggled mightily to establish the foundations for the present work?

I do hope you have the feeling that we have a solid foundation of biomedical work with plutonium and the other radioactive actinides, going back approximately three and a half decades to say nothing of the efforts of the chemists and physicists who predicted the existence of these elements.

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OFF-SITE MEDICAL ACTIVITIES, NEVADA TEST SITE
and the
MEDICAL LIAISON OFFICER NETWORK: A HISTORICAL REVIEW

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ABSTRACT

The "off-site" was originally defined as "that area surrounding the Nevada Test Site (NTS) for a radius of about 300 miles." Prior to 1954, the off-site radiological safety activities were conducted by the Atomic Energy Commission. In 1954, the Public Health Service was given the responsibility for off-site monitoring, and, in addition, a physician was also on temporary assignment. This physician, in addition to functioning as a monitor, also functioned part time as a physicial liaison in regard to possible or alleged radiation injury. Medical concern was based upon two crude guidelines: (1) possible radiation "overexposure" based upon extrapolation from surface and air radiological monitoring; and (2) determination of actual radiation injury based upon signs and symptoms among people alleging radiation injury.

The area of concern expanded to 13 areas surrounding the Nevada Test Site, and in 1956, the first Medical Liaison Officer Network (MLON) was initiated. Over the years, MLON increased to a point where there was a representative from every state in the Union; the area of concern expanded to include the entire United States, parts of the South Pacific, Hawaii, and Alaska; and sophisticated methods of evaluation were added--urine sampling, thyroid scanning, blood counts, and whole-body counting. Epidemiological studies were initiated on body burdens of radionuclides and certain disease clusters.

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HISTORICAL REVIEW

The history of medical concern in the off-site area and the Medical Liaison Officer Network (MLON) is tied closely to the history of the NTS. It developed from the point of extrapolating "what might happen" from data of surface and air monitoring, to delineation of body burdens of radionuclides by biological sampling and whole-body counting.

While this growth was occurring, there was also development of knowledge of radiation injury by the selected physicians of MLON. By 1968, most of the MLON representatives had more knowledge of radiation injury than did 99 percent of the "experts" in 1957. In 1957, there were less than 100 physicians in the entire United States who had any significant training in radiation injury, and only a few of these physicians had ever seen a case of acute radiation injury, much less treated one. Medical knowledge became more and more specialized; and, like the Phoenix bird of mythology, we saw the demise of the radiation injury generalist and the evolvement of the radiation injury specialist.

In 1954, the Public Health Service was given the responsibility for radiological safety in the off-site area (that area surrounding the NTS for a radius of about 300 miles). Two PHS officers were permanently assigned to the Las Vegas Branch of the Albuquerque Operations Office of the AEC. In addition, a number of personnel were assigned on a temporary basis as monitors. One PHS physician was also on temporary assignment. He, in addition to functioning as a monitor, also functioned part time as a physician liaison in regard to possible or alleged radiation injury in the off-site area.

By the fall of 1956, plans had been worked out for the first MLON with representation from 13 areas surrounding the NTS. The physicians in this group were mostly those in practice in the area.

In establishing this Network, it was described as primarily an epidemiologic function. The description stated:

"The primary function of physicians participating in this Network is that of liaison. It is contemplated that the Liaison Officers will serve as a link between the local physician who sees the patient with a suspected or possible radiation injury, and a qualified expert in radiological health. Responsibility of the Liaison Officers will not extend beyond a preliminary investigation of the reported circumstances. His work will be, in effect, that of an epidemiologist. In any case where more than superficial investigation is desired, the Liaison Officer will be expected to refer the case to the Public Health Service Headquarters Group for follow-up by the Public Health Service and the Atomic Energy Commission."

These early MLON representatives were not expected to have either the training or the experience to qualify them as medical expert witnesses in the area of radiation injury, and they were not to be classified as such experts. The MLON Liaison Officers were expected to have some knowledge of radiation and radiation effects.

In establishing this first group of MLON representatives, those chosen were given two weeks of indoctrination and training at Mercury, Nevada, in February, 1957. This training provided information in radiological health activities, including data and current scientific knowledge of long-term acute hazards, and the relationship between radioactive fallout and industrial and medical exposures. The Coordinator for this group, except for a brief time, functioned through offices at PHS Headquarters in Washington, D.C.

In the 1957 modification to the memorandum of understanding between the Atomic Energy Commission and the Public Health Service, a change was made to officially establish the Medical Liaison Officer Network. This paragraph is as follows:

"Scope B--Liaison Officer Network

1. Establish a network of Liaison Officers throughout the Continental United States and its territories to assist the AEC in the investigation of reported incidents, complaints, and claims related to the AEC's test activities. The location of these Liaison Officers shall be in accordance with an operations plan to be prepared by the PHS and approved by the Albuquerque Operations Office.

2. The PHS shall be responsible for the organization of the Liaison Officer Network and indoctrination of its members."

In 1957, the operational plan for the MLON was quite brief, describing the administrative plan in which the composition of the Network and security requirements were established. This Network had representatives in each of the then eight regional offices, and in only thirteen states. This plan also established the mechanism by which investigations would be carried out and the mechanism of reporting the results of investigations.

In 1957, a full-time Medical Officer was detailed to the Nevada Test Organization with the responsibility of medical contact with the physicians in the off-site area. During the six months' test period in 1957, this officer traveled over 25,000 miles making contact not only with the physicians of the area, but also, on their request, with certain of their patients. This same physician also handled a part of the MLON Operations which included the investigation of a number of cases either personally or by telephone, including one case in Mexico. Because of the information which this medical officer was able to supply to local physicians in the off-site area, many of the potential complaints could be handled by the local physicians based upon the knowledge which he had received.

The types of cases investigated ranged from those that could possibly be due to radiation--leukemia, thyroid nodules, depigmentation, depilation--to some which had no possible connection with radiation--green and red sweat, cracked teeth, muscle spasm. Cases of flash blindness, allegedly due to viewing the fireball, were also investigated. Some of these cases carried with them very interesting stories.

One young boy had died of leukemia in the winter of 1957. This investigation included interviews with the parents, and also with the Reno physicians who had cared for the patient. The parents claimed that the boy had developed leukemia as a result of eating radioactive snow. Monitoring records showed no "hot spots" or high levels of radiation detected in the area during the period in question. Vital statistics for Nevada and Utah were also negative for any unusual number of cases of leukemia that year, or the preceding year. The exact type of leukemia was not determined. The Reno physicians were of the opinion that this was not radiation induced. The conclusion of this investigation was that one could not determine whether or not a specific case was radiation induced or not, but based upon the evidence, this did not appear to be radiation-induced leukemia. This latter statement was made on a television broadcast, and the father of the boy threatened to shoot the investigator if he ever again set foot on his property.

One claim on which reimbursement was actually made occurred in an elderly gentleman about 80 miles from the test site. This might be considered a "freak" blast injury. At the time of detonation of the nuclear device, the gentleman was just getting dressed in his home. Apparently, through a channeling mechanism in the atmosphere, the blast from the detonation was channeled through the atmosphere and its effect came down at this man's home, striking the large picture window and causing a startle reaction in the man causing him to twist his neck and develop a spasm in the muscles of the neck. This man was sent to a physician in Salt Lake City, and his expenses were paid.

Cases claiming flash blindness or retinal injury were sent to an ophthalmologist for evaluation, again at government expense. Investigation of time and location of these incidents indicated a possibility of the patient having seen the fireball, but the ophthalmologist found no injury.

The case of "green and red sweat" was sent to a dermatologist. The patient had claimed to have driven through a fallout cloud and then developed the red and green sweat. This patient was sent to a dermatologist, at government expense, and it was determined that the coloration of the sweat was due to a type of bacteria producing the color. He was treated for this and had no further trouble.

Several dermatological cases were investigated and a positive diagnosis made. This diagnosis was not related to radiation.

The basic concept on which MLON worked was positive: "Do not simply say, this is not radiation injury; say, this is not radiation injury because it is ____, and make a positive diagnosis." At all times, we sought the best medical consultation when there were any questions concerning the diagnosis.

During 1957, we performed one autopsy. A man had died near Ely, Nevada, and the local community, and there was even some question by the local physician, started circulating the rumor that the man had died of acute radiation injury. Fortunately, we had some pathologists available at the Test Site who were from Walter Reed Army Medical Center. This team of pathologists was flown to Ely and performed the autopsy. They found no evidence of radiation injury.

Nineteen fifty-seven also saw the start of biological sampling. Besides the autopsy, urine specimens were obtained from a group of people in the off-site area and these specimens were analyzed at Walter Reed for radioactivity.

During the fall of 1957, near the end of Operation PLUMBBOB, the U.S. Public Health Service collected 24-hour urine samples from some of the persons living in the off-site area, and from some of the persons engaged in the off-site activities in the same area. A total of 28 persons was sampled. The areas where samples were obtained were Lincoln Mine, Alamo, and Mercury, all in Nevada. Because radioactivity was so low in the individual samples, a series of five pools was made of the samples. Samples were pooled in the following groups: (1) Mercury--four males; (2) Alamo residents--four males and one female; (3) Alamo nonresidents--two males, one female; (4) Lincoln Mine residents--two males, three females; and (5) Lincoln Mine nonresidents--two males. The samples from each pool were also pooled for one large composite sample.

These specimens were analyzed for the Public Health Service by the Walter Reed Army Institute of Research. The samples were specifically analyzed for strontium-90 (actually Yr^{90}, daughter of Sr^{90}), cesium-137, and iodine-131.

The average level of strontium-90 in a 24-hour urine sample was about 1.0 dpm (disintegrations per minute), which was considerably below the maximum sensitivity of the analytical method used. The five pooled samples showed no significant activity above background which could be identified as the Yr^{90}, daughter of Sr^{90}. Since the pooled samples represented four to ten man-days of urine, the mean excretion levels for individuals would be less than 2.5 and 1.0 dpm per man-day, respectively.

We assumed an excretion rate of 0.1 percent of body burden per day for Sr^{90}, thus the 2.5 dpm per day level suggested a total body burden of no more than 1.1 x 10^{-3} microcuries. This was several times lower than values for fetuses in the Chicago area, and in view of the proximity of these people to the NTS was quite surprising. Low annual rainfall and ingesting foodstuffs grown elsewhere appeared to the investigators to be the overriding mechanisms responsible, despite the close proximity of Alamo and Lincoln Mine residents, for the lower levels.

The mean excretion levels of cesium-137 for Mercury, Alamo, and Lincoln Mine were 51, 118, and 187 dpm per liter. Assuming a 24-hour urine volume of 1,500 ml per day, and an excretion rate of 0.46 percent of total cesium burden per day, estimated cesium body burdens were:

Mercury:	7.5 x 10^{-3} microcuries
Alamo:	1.74 x 10^{-2} microcuries
Lincoln Mine:	2.77 x 10^{-2} microcuries

It is of note that the highest body burden recorded in this group was from a nonresident at Mercury who had 0.187 percent of the maximum permissible (NBS Handbook 68 Industrial standards) body burden. This person was counted in a whole-body counter 14 months later (September, 1958) and at that time his body burden of cesium was within the so-called "normal limits" of the average U.S. population.

The individual excretion levels for cesium-137 varied by a factor of about 10 for each locale, yet the mean body burden at Mercury was exceptionally close to the mean level of 6 x 10^{-3} microcuries for U.S. citizens measured during 1956-57. Although at Lincoln Mine the mean was slightly higher, it was only twice the natural potassium-40 activity in man and about 1/300 of the recommended maximum allowable activity for cesium-137 in nonindustrial personnel.

Although the iodine-131 excretion levels measured in these Nevada subjects represented only a short interval of time, the values were not remarkably different from I^{131} measurements during Operation Teapot. The highest I^{131} activity measured in our study group was in an Alamo resident, and was 1.7 percent of the "maximum permissible thyroid burden" (NBS HB-68 standards).

Thyroid burden can be estimated crudely by assuming that 30 percent of the urinary activity represented thyroidal release at a rate of about 2 percent per day. The mean adult thyroid burdens of I^{131} are then:

Mercury:	5.6×10^{-4} microcuries
Alamo:	1.5×10^{-3} microcuries
Lincoln Mine:	1.6×10^{-3} microcuries

The 1960s saw a marked increase in the use of radiation by industry and physicians, with a comparable increase in the possibility of a radiation incident occurring. It was believed desirable to include all types of radiation emergencies in the training sessions of the MLON.

Several investigators carried out studies of leukemia clusters and thyroid disease in certain areas of Utah and Arizona. The MLON participated in some of these investigations carried on in the sixties.

One of these investigations concerned leukemia deaths in the area surrounding St. George, Utah. Rumors were circulating among the citizens that certain deaths, and particularly an increase in leukemia deaths in 1962, were due to fallout occurring from some 1955 tests. During those tests, one cloud was estimated to have given the residents of a small town near St. George about 5 rad total body exposure, with higher doses to the thyroid. Review of the vital statistics for the area did show a small peak of cases in that year (1962). This was a "peak" of three cases, whereas the average for several years preceding and subsequent to 1962 was less than one case per a year. Statistical evaluation indicated that if four cases had occurred, it would have been a statistically significant peak, but three was still within the statistical variation.

We had also received some complaints of chronic anemia and thyroid nodules in residents in northern Arizona. Investigation produced no significant findings in this area.

Cases investigated by MLON in 1968-69 had very strong psychiatric overtones. One such patient called the USPHS and/or the AEC every 24-48 hours. She was hospitalized on several occasions with the final diagnosis being chronic paranoid schizophrenia. This diagnosis was arrived at only after all possible causes (organic) of her symptoms had been eliminated.

This MLON case presented a difficult, and different, investigative and management problem. Psychiatric illness is common in the United States and was

certainly encountered in MLON investigations. Pertinent problems in investigation and management were reviewed with MLON representatives, and some general recommendations for management of similar situations were developed.

Nineteen sixty-eight also saw the institution of a program for doing whole-body scans and counting among representative residents of the off-site area. This program has continued and, along with the studies of the Alaska natives, has become one of the main methods of monitoring body burdens of radionuclides in residents of the off-site area.

Genetic studies and evaluation of chronic radiation injury have never been entered by the MLON. MLON investigations have generally concentrated on acute illnesses, or problems, and have always tried to make a positive diagnosis rather than simply stating "that is not radiation injury." In all the years of MLON investigation, we have yet to find a true case of radiation injury which could be attributed to radioactive fallout.

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Transuranics in Terrestrial Environments

SAMPLING OF SOILS FOR TRANSURANIC NUCLIDES: A REVIEW

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ABSTRACT

A review of the literature pertinent to the sampling of soils for radionuclides is presented; emphasis is placed on transuranic nuclides. Sampling of soils is discussed relative to systems of heterogeneous distributions and varied particle sizes encountered in certain environments. Sampling methods that have been used for two different sources of contamination, global fallout, and accidental or operational releases, are included.

INTRODUCTION

An accurate assessment of the amount and distribution of transuranic nuclides in soils is necessary for the evaluation of environmental contamination resulting from accidental or operational releases and for establishment of baseline or background levels. One difficulty in sampling of soils for transuranic nuclides is the generally heterogeneous distribution of radionuclides, which can result in a high potential for cross-contamination during sampling. The heterogeneity results from the tendency for most transuranic nuclides to form or associate with particles, from the multiplicity of sources, and from the variable nature of the soil. No one sampling procedure has been found that will provide a representative sample and adequately address all of the problems inherent in sampling of soils for transuranics.

This report reviews the literature pertaining to sampling methods used for estimating transuranic nuclide content of soils. Comparisons of sampling procedures or techniques are made when information is available. Some of the unique problems associated with sampling are discussed.

HISTORICAL REVIEW

Historically, soil sampling techniques originated from agriculture and engineering projects in which a major objective was detailed characterization of soils; the components of interest were distributed somewhat uniformly both on a micro- and macroscale. Many of the procedures used for sampling of transuranic nuclides in soils were modifications of those procedures.

The most important facet of sampling of soils is proper definition of mission and objectives. The objective describes the overall result or answer desired, while the mission sets the quality or acceptability of the result. The mission should address such important factors as: (1) the intrinsic variability of the medium to be sampled; (2) the required accuracy and the number of replicates necessary to provide defensible results; (3) the choice of procedure needed to minimize cross-contamination; and (4) the cost of sampling, sample preparation, and analysis.

The importance of those factors was recognized early in the history of soil sampling. For instance, Vinson *et al.* (1919) discussed the importance of horizontal subdivision of the sampling area consistent with soil type; this work is one of the earliest to stress proper sampling design with due consideration for cost and objectives. Marbut (1921) and Fillinger (1931) cautioned against contamination and/or cross-contamination of samples. A form of cross-contamination recognized in later studies was expressed by Cline (1944): "Many conflicting results that appear in the literature are due to mixing unlike horizons." Veatch (1925) stressed the importance of vertical subdivisions within the same soil type and discussed those subdivisions as discrete populations that differ chemically and physically. Those subdivisions--or horizons, in the broad sense--are common to a particular soil type. Munch and Bidwell (1928) emphasized the importance of a proper sampling method to accurately represent the total population being sampled. Cline (1944), in reviewing the work of Munch and Bidwell, noted the need for replicate samples that can be evaluated separately to provide a statistical basis for sound sampling design. Jewell (1936) and Kerr and von Stieglietz (1938) investigated the requirements to be placed on sample replication to obtain reliable estimates of a particular unknown. They emphasized that too often the number of sample units is chosen arbitrarily and generally is insufficient to detect the wide variability associated with the soils. Hossack (1936) also discussed the importance of an accurately defined mission that accommodates the degree of variability of the matrix, numbers of samples, and cost.

One of the more complete reviews of soil sampling is that of Cline (1944), who treated such topics as: (1) concept of sampling, (2) statistics, (3) cross-contamination, (4) sampling tools, (5) compositing of samples, and (6) sample preparation. Cline's statement relative to composited samples and the establishment of a specific mission is important and bears repeating: "If any statistic other than the mean is required, a single composite sample is completely inadequate."

SAMPLING THE ENVIRONMENT FOR RADIONUCLIDES

Sampling efforts for radionuclides, including transuranics, have been identified as (1) those for worldwide nuclear explosion fallout, and (2) those for accidental or operational releases of radionuclides. The missions for those two types of sampling may be different and thus require sampling procedures appropriate to the difference in missions.

Global Fallout

A program of extensive sampling for fallout radionuclides was initiated in 1953 by Alexander *et al.* (1960). Within five years, the program was global in scope (Harley, 1972). Emphasis through 1967 was on sampling for ^{90}Sr; however, the sampling techniques developed for ^{90}Sr have been adopted for the more recent programs of sampling for plutonium and other radionuclides. The sampling method described by Harley (1972) defined each sampling site to be flat, grassy, and undisturbed. Both surface and core samples are taken; the surface sampler is normally 8.9 cm in diameter and 5.1 cm deep, often designated as a "cookie cutter." Subsurface samples are obtained with a barrel auger to the desired depth. Ten samples are taken on a transect at 30-cm intervals and samples from respective depths are composited. Harley emphasized that soil sampling will provide data for current status but is not applicable to studies that involve measurement of short-term changes. This is because the short-term changes in radionuclide concentration are generally small compared to the variability in local radionuclide distribution. The sampling efforts have been directed predominantly toward global radioactive fallout where areal distribution and particle size were more nearly uniform and concentrations were low. Under such conditions, a complex sampling procedure was not required as indicated by the following experiment conducted by Harley (1972). Two sampling sites, separated by distances of 0.1 m to 100 km, were chosen at each of five different geographical areas. Radioactivity in each sample was measured and means were calculated for each area. The area means differed by 2-18%, and the average percentage deviation of the pooled means for all five areas was 10.6%; there was considerable uniformity in the sample pairs.

Mamuro and Matsunami (1964) have reported the collection of fallout particles in Japan as large as 20 μm diam from Chinese nuclear weapons tests and particles as large as 17 μm diam from earlier Russian tests. Uranium and plutonium were identified in some of the fallout particles. A limited number of highly radioactive particles may appear in global fallout, which may result in a high degree of variability in radioactivity associated with soil samples. However, no reports have been found that document a high degree of variability attributable to the presence of highly radioactive fallout particles.

Hardy *et al.* (1973) discussed the efforts to determine the distribution of ^{238}Pu deposited on the earth's surface as a result of the atmospheric burnup of a SNAP power generator. Undisturbed areas at 65 sites around the world were sampled. Each sample consisted of ten 8.9-cm diam cores taken to a 30-cm depth, representing a surface area of 622 cm^2. No further information on the method of sampling or quality of results relative to the adequacy of the sampling method were given.

The New Zealand Department of Health (1974) has reported on the deposition of ^{238}Pu from the SNAP-9A atmospheric burnup. The method of sampling the soil surface was that used by Health and Safety Laboratory (HASL) as previously discussed by Alexander *et al.* (1960).

Michels (1974) reported on sampling in the Red Desert Basin of Wyoming using a 10-cm square "jig" to cut the perimeter of the sample. Profile samples at the 0- 5-cm and the 5- 10-cm depth intervals were then obtained with a flat-bottomed scoop. Although sampling in this case was for ^{137}Cs content, the procedure has been used by others in sampling the soil surface for transuranics. It is comparable, except for configuration, to the ring method used by the Nevada Applied Ecology Group (NAEG) as discussed in the next section.

Aarkrog and Lippert (1974) sampled a variety of soils in Denmark for ^{90}Sr, ^{137}Cs, and "gamma radioactivity." Samples were obtained with a Van Veen sampler; however, that sampler did not well define the depth sampled. For profile samples, a "HAPS" bottom core tool was used. The HAPS sampler was designed for sampling bottom sediments but was tested on terrestrial soils. It is of interest that the authors did compare results obtained by the two methods and concluded that the same degree of accuracy was obtained with samples collected by both methods to a 4-cm depth. Aarkrog and Lippert (1975) later reported on sampling to a depth of 50 cm with a HAPS bottom corer. The nature or description of both the Van Veen and HAPS samplers could not be found except that they both appear to be modifications of coring tools.

Hardy (1974) has used a sampling procedure similar to the NAEG trench procedure to obtain profile samples to determine depth of penetration and the distribution of global fallout ^{239,240}Pu in soils. He cautioned against using the procedure where rocks and stones are prevalent and where cross-contamination of lower profile cuts with higher radioactive material from above could occur. The cross-contamination effect of worms and burrowing insects, cracks from swelling, and other biological and physical disturbances were alluded to but not discussed.

Accidental or Operational Releases

Sampling techniques used in evaluating the accidental or operational releases of radionuclides depend on the specific mission and may or may not be similar to those used for global fallout. Differences in the methods are dictated by the nature of the radionuclide distribution in the soil, the range of particle sizes, and the generally higher levels of radioactivity associated with accidental or operational releases compared to global fallout.

Fowler *et al.* (1968) initiated an extensive soil sampling program at Palomares, Spain, after the accidental destruction of two nuclear devices. The mission designated sampling for plutonium to a depth of 45 cm to determine inventory and to estimate vertical distributions. A core tube (King tube) was used and efforts were made to divide the profile into discrete segments. Results among replicates at common depths obtained from areas of high contamination were considerably variable (up to two to three orders of magnitude); hence, data were treated *en masse* rather than as discrete sections of profiles.

The high probability of smearing as a result of using the coring tool was recognized and caused the authors to conclude that coring, as a sampling method, should not be used if reliable data are to be obtained for profile analyses.

It is of historical interest to note that a group of scientists interested in environmental radioactivity met in Washington, D.C., in October, 1970, at the request of U.S. Atomic Energy Commission to address problems associated with sampling for plutonium. The objective of the working group was to provide guidance relative to sampling techniques. Essentially the group recommended the HASL method of sampling (Harley, 1972), but recognized sampling problems associated with accidental or operational releases that could not be covered by the HASL method. The meeting occurred after the experience of cleanup of plutonium from two nuclear devices accidentally destroyed at Palomares, Spain, but prior to the initiation of studies at the Nevada Test Site (NTS) by NAEG. The full impact of the problems associated with accidental or operational releases was probably not recognized.

Poet and Martell (1972) discussed the measurement of plutonium and americium contamination levels in the environs of the Rocky Flats plutonium plant and elsewhere in the greater Denver area of Colorado. Their sampling procedure was not discussed in detail but consisted of collection of both surface samples to a depth of 1 cm and soil profiles to a depth of 14 cm. Surface samples were collected from even, undisturbed sites that had not been subjected to irrigation or plowing. Where the history of the site was not known, samples were taken along old fence lines midway between posts. Each soil sample was collected over an area of 1000 cm^2 by spatula to the desired depth as uniformly as practical. Profile sampling was not discussed except that precautions were taken to avoid contamination from the shallow soil horizons.

In 1970, the Soils Sampling and Analysis Committee of the NAEG outlined a sampling technique to be used in areas at NTS where plutonium, americium, and uranium were dispersed in localized areas. A detailed discussion of that sampling technique was presented by Fowler *et al.* (1974). In brief, sampling points are selected randomly and may be grouped into strata (regions of similar radioactivity levels, or of a similar physical nature) in order to reduce the variability. The surface sampling tool is a steel ring 12.7 cm in diam and either 2.5 or 5 cm deep. A lip is welded to the upper edge to provide rigidity, a surface for application of pressure, and to assure constant depth of penetration. The 12.7-cm diam of the ring is dictated by the diameter of a radiation detector used to obtain *in situ* measurements of associated radioactivity (in this case, the 60 keV gamma emissions of ^{241}Am). Surface area and depth define the volume; the weight of that volume of dry soil defines the field bulk density. Those data are necessary to convert radioactivity per unit weight measured by analytical techniques to radioactivity per unit surface area. Soil adjacent to the exterior of the ring is removed to the depth of the ring; the confined volume is then transferred to a plastic bag using an appropriate tool. Depth profile samples may be drawn using this method by repeating the steps for each subsequent depth to be sampled. Removal of soil exterior to the ring is necessary to minimize the possibility of cross-contamination of deeper, less radioactive fractions; rings and tools must be decontaminated or replaced after each use.

The technique applied by NAEG specifically for profile sampling is a modification of the profile sampling method used in soil surveys (Soil Survey Staff, Dept. of Agriculture, 1951); its origin appears to be lost in history. A trench of appropriate size for ease of access is opened adjacent to the point to be sampled. The face of the trench adjacent to the area to be sampled is shaved from side to side to the depth of each sample; the debris is then removed from the trench. A metal flat-bottomed scoop, preferably stainless steel, 10x10x5 cm deep with the opened edges sharpened, is inverted and pressed into the soil surface. The back of the scoop just contacts the exposed, shaved face of the cut. Adjacent soil exterior to the scoop is removed to the 5-cm depth; the scoop is carefully withdrawn, inverted, and pressed into the exposed face so as to encompass the exposed block of soil. A template is pressed into the soil vertically at the front (open end) of the scoop to isolate the block of soil, which is then transferred to the sample container. Tools are decontaminated or replaced and the procedure continued to the desired total depth. Every attempt is made to avoid cross-contamination. The procedure works well in sandy loam, loam, or loamy sand. It does not work well on soils with more than a few small stones.

Bernhardt (1976) surveyed the literature for soil sampling and analytical techniques for environmental concentrations of plutonium. Bernhardt mistakenly stated that the NAEG sampling technique is "intended for sandy and rocky soils that cannot be sampled by core techniques" and that for surface samples, "a minimum number of five separate samples should be taken along a straight line transect and composited for analysis." He also mistakenly stated that for profile samples, "a minimum of five samples should be taken from separate trenches along a straight line transect. Composite the samples for analysis." While NAEG neither endorses nor disapproves transect sampling and/or compositing for its operations, these methods have not been used for routine inventory and distribution sampling by NAEG. The procedures do not work well in very rocky soils but do work well on most other nonhighly compacted soils. The procedures used by NAEG are in lieu of core sampling techniques in an effort to reduce cross-contamination. Transects have been used as preliminary site surveys for the purpose of approximating the location of radioactivity at a site. Compositing of samples has not been done; the value of compositing as a general procedure is open to question.

Techniques for the sampling of soils at Enewetak in a recent extensive sampling effort were described by Lynch and Gudiksen (1974). The soil matrix was coral sand and coral with varied amounts of organic matter and the parent material was coral limestone bedrock. Two types of soil samples were taken; surface and profile. At surface sampling locations, two samples were collected: one was a 30-cm^2 x 15-cm-deep core, and the second was a composite of two 30-cm^2 x 5-cm-deep cores. Profile samples were obtained using the trench method described above for NAEG profile sample collections. Comparisons of methods were not discussed.

Schneider (1974) sampled soils at the DuPont, Savannah River Complex as part of a study of soil-plant relationships. A "large hole" (dimensions not given) is dug to a depth of 5 cm. Soil to the 5-cm depth is retained as the surface sample. Inside the "large hole," a "smaller hole" is dug to the 15-cm depth; soil from that hole is retained as the 5- to 15-cm section of the profile.

The intention of the large and small holes appears to be an avoidance of cross-contamination.

Eberline Instrument Co. (1974) reported on soil sample collection for Project Wagon Wheel in western Wyoming. Samples of 100 cm^2 x 1.25-cm deep were collected at the surface, at the 7.6-cm depth, and at the 15.2-cm depth. Elements of interest were plutonium and uranium; however, the sampling method was not given.

Fix (1975) cited sampling of the soil surface at the Hanford, Washington, Reservation for the determination of plutonium and uranium content in a program related to the environmental status and potential buildup of radioisotopes. Five plugs of 10 cm diam and 2.5 cm deep were collected from a 10 m^2 area and composited. The method is similar to that of HASL for obtaining surface soil samples (Alexander *et al.*, 1960).

Nyhan *et al.* (1976) have reported on the use of a hand-driven core sampler for obtaining soil samples to various depths from Trinity Site in New Mexico. The purpose of the investigation was to determine $^{239,240}Pu$ distributions in those soils. The coring tool was a sharpened plastic tube that retained the entire sample. The tube was cut into sections for profile analysis. The problem of smearing, as discussed by Fowler above, was not treated.

Krey *et al.* (1976) have taken samples for plutonium at the Rocky Flats complex using both a template and a coring method. Briefly, the mission was to provide greater detail (relative to earlier sampling efforts) in the plutonium concentrations in soil in and around the Rocky Flats complex, and to relate plutonium concentrations in air to the local soil contamination on a per unit surface area basis. Sampling sites were selected to provide an estimate of the areal distribution of the surface plutonium surrounding each of the air particulate sampling stations, as a measure of the plutonium available for reentrainment. The coring and template methods cannot be compared directly since the sampling sites were different, ranging from about 1 to 3 km from the plant. Reproducibility of aliquots taken by the core method was tested and a range of 1% to 30% was determined for the data presented. A similar comparison of reproducibility for the template method was not made. For the template method, a 1000-cm^2 area was outlined with an aluminum template and the soil removed with flat chisels and scoops to a depth of 5 cm. Soils were stony; problems, if encountered, were not detailed.

Carrigan and Pickering (1967) used a Swedish Foil Sampler to obtain bottom sediments in the Clinch River for measurement of fission products originating at the Oak Ridge National Laboratory complex in Tennessee. Although the Swedish Foil Sampler was designed for coring bottom sediments, its use is discussed here because of the unique design, a design that could reduce the degree of cross-contamination of core samples. Narrow, thin steel foils are attached to a piston and drawn up the inside of the 6.3-cm diam coring tool as it is pressed into the sediments. The core barrel is thus progressively lined and smearing is greatly reduced or eliminated.

SUBSAMPLING

Related to the problems of sampling in the field are the problems of subsampling in the laboratory. The sample presented to the analyst must be of an acceptable configuration, which implies subsampling of a large mass--often kilograms of soil. Subsampling and sample preparation are interdependent to the extent that they must be discussed as an entity.

Cline (1944) discussed subsampling of soil masses and stated that the mixture must be reduced in size and subsampled in such a way that a single analysis will produce an unbiased estimate of the mean of the entire sample. He stated further that the accuracy with which a subsample can be drawn depends on its size and degree of heterogeneity. Harley (1972) discussed sample preparation and subsampling of soil masses for the determination of plutonium in global fallout. The method employed crushing, grinding, and sieving to obtain a mass acceptable for subsampling. However, as pointed out earlier, global fallout is relatively homogeneous in particle size and distribution and is not subject to the vagaries encountered in the accidental or operational releases. The preparation and subsampling techniques outlined by Harley may not be adequate for samples representing accidental or operational releases. The radionuclide distribution may be heterogeneous and contain particles of a wide range of sizes. The particles are difficult to reduce to a size which can be uniformly distributed throughout the sample.

As with sampling, much work remains to be done on subsampling. It is probable that multiple subsampling and multiple analyses are the only techniques available at the present time to adequately define the radionuclide content in the sample.

DISCUSSION

The importance of careful sampling techniques appears to have been given an unjustifiably low priority in the reviewed literature. Much of the literature on sampling related to sampling for fission products with only a few references directed toward transuranics and uranium. Evaluation of individual sampling methods or the comparison of methods for sampling of soils for transuranics could not be accomplished since detailed procedures and results often were not given. In many cases, the only sampling information given was: "samples were obtained" or "samples were taken."

All too often, sampling is conducted with insufficient attention to the means by which the objectives are to be met. Proper specification of (1) the intrinsic variability of the medium to be sampled; (2) the number of replicates necessary to provide defensible results; (3) the choice of procedure needed to minimize cross-contamination; and (4) the cost of sampling, sample preparation, and analysis must be defined by the mission in terms of the objective to be fulfilled.

In some cases, data obtained for one mission--more often for an objective with an undefined mission--have been used by other workers in the field to support conclusions related to an entirely different mission. Such a practice may lead to erroneous or unsupportable conclusions.

Sampling of soils with a coring tool has been quite popular. A problem associated with almost all coring devices is that of smearing, which introduces a variable not easily controlled, particularly when sampling large differences in radionuclide concentration within the soil profile. The coring tool may be used for determination of total radioactivity content from the surface to some predefined total depth. An example of the smearing problem (Table 1) was reported by Fowler *et al.* (1968). Soil cores were collected at the Palomares, Spain, site from an untilled hillside plot in the pathway of local fallout, about 200 m from the point of impact of a nuclear device. The area had received about 5 cm rainfall per year. Five months had elapsed between the event and sampling. The 3.7 fold increase from the 5.0- to 15-cm depth to the 15- to 25-cm depth is explained as contamination of the core, probably by particulate plutonium dragged from the surface into deeper soil fractions by the coring tool. Transuranic concentrations in soils are generally low, but their distribution in space may have a range of several orders of magnitude and cross-contamination is a problem which must be addressed.

Accidentally- or operationally-released radionuclides may be distributed heterogeneously, not only on a areal basis, but also as a wide range of particle diameters and types. Those particular types of heterogeniety have been observed at several locations. Sampling of the heterogeneous distributions of particles requires special care that all particle sizes are adequately represented in the samples. An example of a heterogeneous distribution of highly radioactive particles is shown in Fig. 1. The autoradiograph was prepared from soil obtained from an area near the point of impact of one of the nuclear devices at Palomares, Spain. The wide range of particle diameters and associated radioactivities is suggested. If one assumes a single spherical particle of PuO_2 0.6 μm-diam, the resultant radioactivity amounts to about 0.2 dis/min/particle; an 18 μm-diam particle will produce about 4700 dis/min/particle.

In the case of a sample from an area receiving accidentally- or operationally-released radionuclides, the heterogeneous distribution of radioactive particles of varied sizes poses serious problems associated with subsampling. Fowler *et al.* (1968) discussed subsampling and the variability in observed radioactivity due to a heterogeneous radioactive particle distribution. Seven one-gram aliquots were taken from a single sample; the radioactivity of dry soil corrected for background ranged from zero to 778 dis/min/g (Table 2). There is no technique that will effectively reduce PuO_2 particles of the typical sizes observed by Fowler to a population of subparticles that can be uniformly distributed throughout the sample mass. Faced with such a dilemma, the researcher turns to replicate subsampling. In the NAEG program, ball-milling, sieving, and replicate analyses have been employed. Analytical data from different laboratories on subsamples containing radioactive particles often vary by factors of 2-3.

Table 1. Concentration of Plutonium in Untilled Soil From Palomares, Spain, Plot 2-1 Sampled With a King Tube

Core Segment Depth (cm)	Plutonium Activity (pCi/g)
0-5	2.5
5-15	0.8
15-25	3.0
25-35	1.2
35-45	2.1

(Adapted from Fowler *et al.*, 1968)

Table 2. Variation in Observed Activity in 1-g Aliquots From a Single 25-g Soil Sample, Plot 2-2, Palomares, Spain

Aliquot	1	2	3	4	5	6	7
dis/min/g	455	778	0	28	8	53	45
Mean =	195						
s =	303						
s/N = SE =	115						

(Adapted from Fowler *et al.*, 1968)

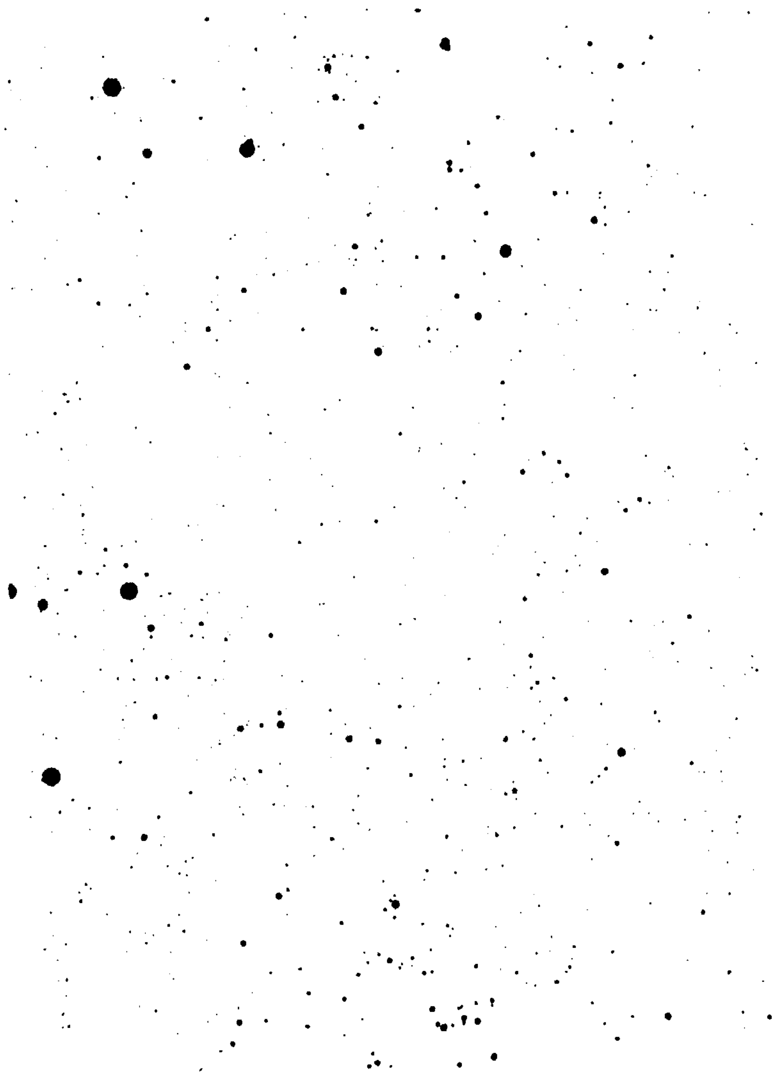

Fig. 1. Autoradiograph of soil from a Nuclear Device Impact Area, Palomares, Spain, Showing Discrete Particles Containing Plutonium (Reproduced from Fowler *et al.* 1968).

Information relative to comparisons of methods for sampling of soils would be desirable. A statistical treatment of data indicating the applicability of sampling methods for various matrices or forms of a matrix could serve as a guide in the development of sampling programs. However, of the applicable literature reviewed, in only one case was there a reference to a comparison of methods for sampling, and in that case no data were presented. The importance of the sampling methods has not received due consideration: in fact, not only data but also methods of sampling are open to legal scrutiny.

A comparison of sampling methods is complicated by factors other than those associated with matrix and/or technique. Sample preparation, aliquoting, and analysis all contribute various levels of variability to the final result and must be sorted out in comparing sampling methods. One way to evaluate the variability at each step is to prepare and analyze a number of replicates; the number of replicates would depend on the degree of precision needed to make the evaluations. Analyses are expensive; hence, a number of replications on a large number of samples necessary to test sampling methods becomes cost prohibitive. Such factors as the particle problem and low radioactivities introduce large errors which, in turn, pose questions relating to interpretation of apparent differences in sampling methods. The specific difference necessary to conclude that sampling methods produce the same or different results is thus dependent on the variables associated with matrix and sampling, preparation, aliquoting, and analyses. With present state of the art, it follows that only large differences among sampling methods can be interpreted as such and that small differences should not be considered as absolute.

SUMMARY

The literature for sampling of transuranic nuclides in terrestrial environments was reviewed. Although many reports are available on various aspects of transuranics in the environment, very little is said about specific sampling methods and the adequacy of sampling procedures to fulfill a specified or implied mission. In most cases, the mission was not discussed.

Two general methods are used for soil sampling: one is based on coring and the other is based on use of a template. Because of the potential for smearing associated with coring, resulting in cross-contamination, the coring method appears to be applicable only where total radioactivity to a defined depth is to be determined. Coring may not be applicable to sampling for vertical distributions of transuranic nuclides in soils unless precautions are taken to prevent cross-contamination.

Comparisons of sampling procedures for transuranic nuclides in terrestrial environments were not found. At present there is no assurance that cross-contamination or other problems with the sampling methods used did not exist.

The presence of radionuclides in the environment has become a sensitive issue. Each worker in the field should assure that the sampling procedure used is that which will result in interpretive data consistent with a well-defined mission. Sampling procedures should be detailed or referenced.

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DISTRIBUTION OF TRANSURANIC NUCLIDES IN SOILS: A REVIEW

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ABSTRACT

The literature is reviewed to ascertain the degree of movement and the distribution patterns for transuranic and uranium nuclides in soils. Typical plutonium and uranium profiles are presented and an attempt is made to identify unique characteristics causing deviation from an ideal distribution pattern. By far most of the distribution observations are with plutonium and little is reported for uranium and other transuranic nuclides.

INTRODUCTION

In the past decade interest has been growing in the environmental impact of certain transuranic radioisotopes because of their potentially hazardous nature to man. The potential for dispersion of these transuranics due to accidental release from components of the nuclear fuel cycle, improper radioactive waste management, accidental or purposeful nuclear weapon incidents, and further redistribution of existing global fallout and specific loci of contamination is being studied. There has been specific emphasis, during the past few years, on determining the degree of vertical migration of certain transuranic radioisotopes in soils, both in the laboratory and under field conditions. Further investigation on the environmental fate of transuranic elements is urgently needed in order to assess their long term distribution patterns.

This paper reviews past documentation on the vertical distribution of transuranic nuclides and uranium in the terrestrial environment under natural or near natural conditions. Observations made thus far will aid in predicting future redistribution rates and indicating where additional evaluations are needed.

REVIEWS

Francis (1973) and Price (1973) reviewed the literature through 1971 on plutonium and other transuranics in soils, plants, and animals. They found several works on plutonium and americium migration or distribution in soils, but information on other transuranics was lacking. Several papers described laboratory leaching studies on the movement of radioactive transuranic waste materials through soils. In general, these early papers showed that plutonium and americium can move through soils and are influenced by such factors as nature and oxidation state of source material, soil pH and organic matter content, presence of complexing agents, and cultural practices. It was suspected that, under natural conditions, most of the plutonium moved as colloidal or discrete dense plutonium oxide particles and not as solubilized species, the soluble plutonium being tightly bound by exchange mechanisms in the soil.

Wilhelmi (1974) reviewed the literature through 1973 and listed all the known sources of plutonium and transuranics added to the environment. Plutonium movement through the soil was discussed, and typical profiles, where available, were presented for fallout plutonium and for accidental plutonium releases at Rocky Flats, Colorado. Fallout plutonium was reportedly found to depths exceeding 30 cm, although the proportion of total profile plutonium found below 30 cm was only several percent. Wilhelmi (1974) pointed out that plutonium was strongly bound to soil material and migrated slowly; however, plutonium was found to migrate rapidly through cracks and fissures in geologic materials underlying soils. The review also revealed that there was little information on the influence of such factors as physical and chemical nature of soil and soil organic matter.

SOURCES OF TRANSURANICS

The source of transuranic nuclides in the environment, for purposes of discussion, can be categorized into global fallout, local fallout, safety shots, and accidental releases. Wilhelmi (1974) summarized the sources and noted that except for atmospheric nuclear explosions and the atmospheric burnup of a SNAP-9 power generator, almost all of the other sources were localized.

Global fallout is produced primarily from nuclear weapons explosions in the atmosphere, where substantial quantities of the radioactive debris are injected into the stratosphere. This material is slowly added to the environment over long periods of time and appears somewhat as a continuous source.

In contrast to global fallout is the local fallout associated with nuclear explosions and low level chronic releases from nuclear fuel processing. The local fallout from nuclear explosions and stack effluents from processing are deposited onto the soil surface soon after release and enter into the soil weathering processes immediately.

A number of nuclear weapons tests were conducted at the Nevada Test Site (NTS) and Tonopah Test Range (TTR) which resulted in little or no fission yield. These tests were chemical high explosive detonations of, or associated with, components of nuclear devices to determine the degree of safety or stability of the devices to the impact of an explosion. These tests were called "Safety Shots." The conduct of these tests resulted in the limited dispersion of the nuclear fuel, mostly plutonium and some americium as an impurity but in some cases natural or enriched uranium. The nuclear material was dispersed in the form of discrete metallic or oxide particles formed from the burning of the metal in air. The high temperatures attained in a nuclear explosion were not present; thus, the dispersed material had different characteristics from those it would have had if it had been involved in a nuclear explosion.

There have been a number of accidental releases of transuranic nuclides. Typical of accidental releases are the occasional low level releases from waste disposal facilities; accidental destruction of nuclear weapons as that which occurred over Palomares, Spain; and possible localized leaks of stored transuranic contaminated materials. Wilhelmi (1974) summarized all known accidental releases and noted that there were very few observations of transuranic nuclide distributions in soils associated with the accidental releases.

Expanding efforts to provide energy from nuclear sources will increase the inventory of transuranics and may increase the potential for an additional burden of transuranics in the environment. Those transuranic radionuclides of greatest concern are shown in Table 1 and Table 2, as compiled by Perkins (1975) and Langham (1971), respectively. Not all transuranic nuclides are considered hazardous, either because of the small amounts produced or the short radioactive half-lives.

A projected 100-fold increase in total curies of transuranics in waste over the next 24 years indicates the urgency for more information. However, this does not mean that there will be a 100-fold increase in the environmental burden of these transuranics.

DISTRIBUTION OF TRANSURANICS IN SOILS

There have been a number of studies dealing with the distribution of plutonium in various environmental situations, and only a few laboratory

Table 1. Activity of Heavy Elements in Accumulated Waste for Entire Nuclear Industry

	Normalized to 1 for ^{239}Pu						Decay Half-Life (y)
Isotope	1972	1976	1980	1986	1990	2000	
^{237}Np	0.22	0.22	0.20	0.15	0.14	0.76	2.1×10^6
^{239}Np	11.5	11	14.5	43.1	31.7	7.84	6.4×10^{-3}
^{236}Pu	0.00092	0.00075	0.00056	0.0036	0.010	0.012	2.85
^{238}Pu	36.1	49	55.6	167	283	361	8.64 x 10
^{239}Pu	1	1	1	1	1	1	2.44×10^4
^{240}Pu	1.54	1.7	1.89	4.45	5.33	3.14	6.58×10^3
^{241}Pu	315	300	295	403.3	367	255	1.32 x 10
^{242}Pu	0.0043	0.0044	0.0047	0.0099	0.011	0.011	3.79×10^5
^{243}Pu	--	--	--	--	--	--	--
^{241}Am	96.1	96.5	103	178	141	77.2	4.58×10^2
^{242m}Am	5.69	5.5	7.23	19.5	14.2	5.68	1.52×10^2
^{242}Am	5.69	5.5	7.23	19.5	14.2	5.68	1.82×10^{-3}
^{243}Am	11.5	11	14.5	43.1	31.7	7.84	7.95×10^3
^{242}Cm	5637	3020	2624	3183	953	278	4.45×10^{-1}
^{243}Cm	2.31	2.25	2.34	4.03	3.00	2.16	3.2 x 10
^{244}Cm	1561	1510	2441	9813	6464	919	1.76 x 10
^{245}Cm	0.23	0.23	0.50	2.64	1.92	0.33	9.3×10^3
^{246}Cm	0.046	0.046	0.072	0.49	0.35	0.063	5.5×10^3
^{247}Cm	--	--	--	--	--	--	--
TOTAL CURIES	7.2×10^5	1.4×10^7	7.9×10^7	9.3×10^8	1.2×10^9	1.8×10^9	

Adapted from Perkins (1975).

Table 2. Plutonium Economy of the Future

	Annual Production and/or in Use (kg)		
	1970-1980	1980-1990	1990-2000
^{239}Pu			
Power Production	20,000	60,000	80,000
^{238}Pu			
Space Applications	10-20	100	--
Medical Applications		5	6,000
Transplutonium Isotopes			
^{244}Cm	40	180	200
^{252}Cf	0.1	0.8	3.5

Adapted from Langham (1971)

studies designed to predict migration rates of plutonium and americium in specific soils. Of the several sources listed in the previous section, global fallout offers the most uniform source of plutonium and probably will offer the best source for long term migration studies.

Global Fallout

deBortoli and Gaglione (1969) sampled sandy or loamy sand soils from the environs of the Euratom Ispra Establishment, Ispra, Italy, in 1966 to determine the depth of fallout ^{239}Pu (^{239,240}Pu) penetration. The ^{239}Pu distribution in two profiles collected some distance apart are shown in Fig. 1, trace A. The ^{239}Pu was found to a depth of 15-20 cm in samples collected with a coring tool.

In 1970, Hardy and Krey (1971) measured ^{239}Pu (^{239,240}Pu) to a depth of 25-30 cm in a sandy soil in Brookhaven, NY (Fig. 1, trace B). This profile was also collected with a coring tool and compares closely with the profiles taken in 1966 from Ispra. In 1972, Hardy (1974) measured ^{239,240}Pu in a soil profile from North Eastham, MA., the first of a proposed time series of profiles (Fig. 1, trace C). This soil was not sampled by a coring method; rather, samples were obtained using a modification of the trench method used by the Nevada Applied Ecology Group (NAEG) at NTS (Fowler *et al*, 1974).

The use of coring tools for the purpose of collecting soil profile samples introduces a potentially serious cross-contamination variable. It is possible that both the similarity of profiles A and B, Fig. 1 and the amount of plutonium reported at depth, resulted from cross-contamination introduced by the coring tool. Profile C may reflect a smaller degree of cross-contamination; however, it has not been shown that the trench method prevents cross-contamination.

Hardy *et al*. (1972) reported ^{239}Pu (^{239,240}Pu) in soil profiles from widely separated locations within the United States. Less than 5% of the total ^{239}Pu was found in soil profile fractions below 30 cm. Some of these same profiles were reported by Bennett (1976).

McLendon (1975) reported the results of studies to determine the vertical distribution of ^{238}Pu and ^{239}Pu (^{239,240}Pu) in the on-plant and off-plant environs of the Savannah River Plant, Aiken, SC. In 1973, soil profile samples were collected by coring and slicing the cored soils into 0- to 5-, 5- to 15-, 15- to 22.5-, and 22.5- to 30-cm increments. Results indicated that in most cases the ^{238}Pu and ^{239}Pu were localized in the top two increments and remained relatively immobile.

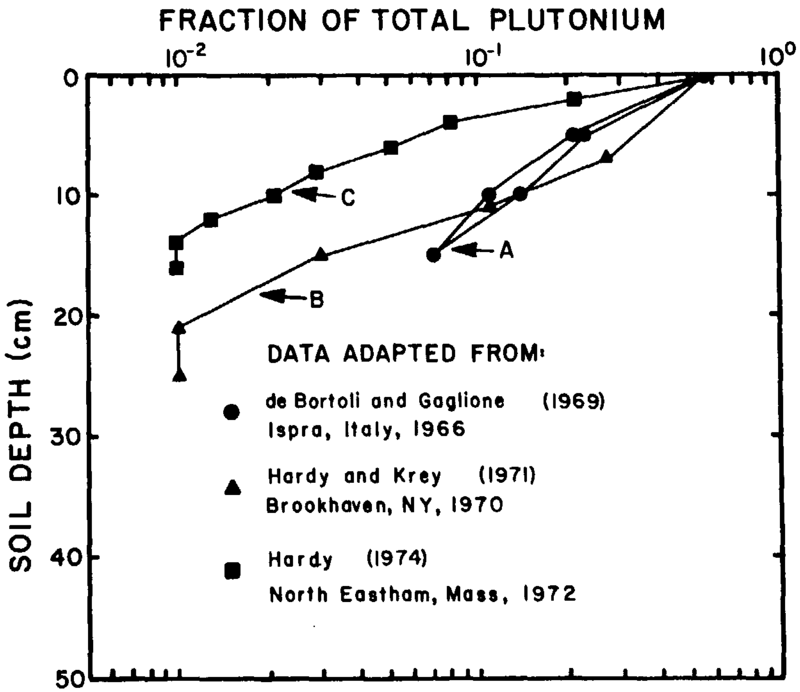

Fig. 1. Global Fallout Distribution in Sandy Soils.

To date no information has been found on ^{241}Am distribution in soils from global fallout. However, interest in the distribution of ^{241}Am in soils should be forthcoming, since the importance of ^{241}Am related to plant uptake and biological hazards has been recognized (Fowler and Essington, 1974).

Local Fallout

The Trinity event was the first atmospheric nuclear explosion. Plutonium distributions in soils near the Trinity Site, Alamogordo, NM, were reported for samples collected more than 20 years ago by Olafson *et al.* (1957). A 1972 study on this site was made by Hakonson and Johnson (1974) and plutonium distributions have been studied in more detail by Nyhan *et al.* (1976).

Olafson (1957) reported measurable downward plutonium movement occurring only where previous surface contamination was high and that the plutonium distribution followed the same trend as that found for fission products. Samples collected in 1948, three years after the event, showed the plutonium localized in the top 2 in. of soil. Hakonson and Johnson (1974) reported ^{238,239}Pu distribution in soil profiles collected near Ground Zero (GZ) and at various distances along the fallout pathway to 56.4 km. Soil profile samples were obtained using a coring method resulting in 0- to 2.5-, 2.5- to 7.5-, and 7.5- to 30-cm increments. Positive total plutonium (^{238}Pu plus ^{239,240}Pu) levels were found in the 7.5- to 30-cm increments in nearly all the profiles investigated out to 56.4 km. This suggests that some of the plutonium initially deposited 27 years ago had migrated to a depth of 7.5-30 cm in the soil. Except for the GZ profile, the plutonium concentrations in the 0- to 2.5-cm increments generally increased toward the outer end of the sampling transect. This observation is consistent with the fallout zone mapped by Olafson (1957), where the highest plutonium concentrations in soils, vegetation, and small mammals outside the GZ area occurred about 45 km from GZ. The vertical distribution of plutonium was relatively uniform in most of the core samples between GZ and the 24.1-km station. More distant profiles reflected a preponderance of plutonium in the surface 2.5 cm similar to the original observations of Olafson (1957) and Olafson and Larson (1961). Hakonson and Johnson (1974) suggested that many factors could have accounted for Olafson's observations including differences with distance from GZ in the chemical and physical form of the plutonium and differences in the chemical, physical, and biological makeup of the environment.

Nyhan (1976) carried out a soil profile sampling program at Trinity in 1974 and 1975 to determine the maximum depth of ^{239,240}Pu penetration and the variability of ^{239,240}Pu penetration with location. Four sites were chosen: a control site 4.8 km south of GZ and three sites 1.6, 16, and 44 km northeast of GZ within the fallout pathway. Plutonium-239,240 at the control site, which represented global fallout, was too low to be detected below the 2.5- to 5.0-cm increment. On the other hand, ^{239,240}Pu was found to a depth of about 30 cm at the 44-km station. Figure 2 reproduces four of the profiles reported by Nyhan (1976) and is representative of the high degree of variability in ^{239,240}Pu distributions with depth. There was no consistent distribution pattern that could be related to obvious physical, chemical, or biological factors. The maximum depth of penetration, however, appeared to be related to the average maximum rain water penetration into the soil.

Nyhan (1976) observed that samples collected at greater distances from GZ reflected increased amounts of ^{239,240}Pu associated with <100 μm-diam. particles. Various factors were noted as possible reasons for the increased ^{239,240}Pu migration at the 44-km site. This site receives 20% more rainfall than the other sites. Soils in this area contain gypsum, which could enhance the displacement of ^{239,240}Pu from soil binding sites due to the high concentration of soluble calcium ions and the enhanced water percolation due to flocculation of soil clays by the calcium ions. Another consideration was soil disturbance by the digging activities of badgers and other ground dwelling small mammals.

Atmospheric nuclear explosions also took place at NTS. Unfortunately, there is no record of measured transuranic nuclide distribution in soil profiles for any of those atmospheric nuclear explosions.

Two areas where atmospheric nuclear weapons testing has been conducted, and for which the vertical distribution of plutonium and americium has been measured, are the Enewetak and Bikini Atolls. Lynch and Gudiksen (1973) measured ^{239}Pu (^{239,240}Pu) and ^{241}Am in the coral soils and sands of Enewetak Atoll. Soil profile samples were collected to total depths of 35 to 185 cm. Concentrations of ^{239}Pu were reported in all profiles; ^{241}Am analyses were performed on many of the samples, but ^{241}Am data were not interpreted by the authors. Profile samples showed a wide range of activity distributions as a function of depth on different parts of the atoll, as shown in Fig. 3. Although meaningful generalizations regarding the ^{239}Pu distribution could not be made, Nervik (1973) commented on several of the groups of profiles observed. On the southern islands of the atoll, activity levels were usually low throughout the full range of depths sampled; some sampling locations showed ^{239}Pu concentrations decreasing somewhat from the surface through the first 10 or 20 cm of soil (Fig. 3, note trace A). Soil profiles inland on the islands subjected to fallout but not to construction or other GZ earth moving activities, showed a rapid and fairly steady decrease of ^{239}Pu levels from the surface to total depth, as shown in Fig. 3, trace B. In contrast, ^{239}Pu profiles from beaches and other exposed areas showed uniform or slowly decreasing ^{239}Pu activity levels from the surface to total depth (Fig. 3, trace C). Occasionally, distribution patterns showed accumulation of ^{239}Pu at

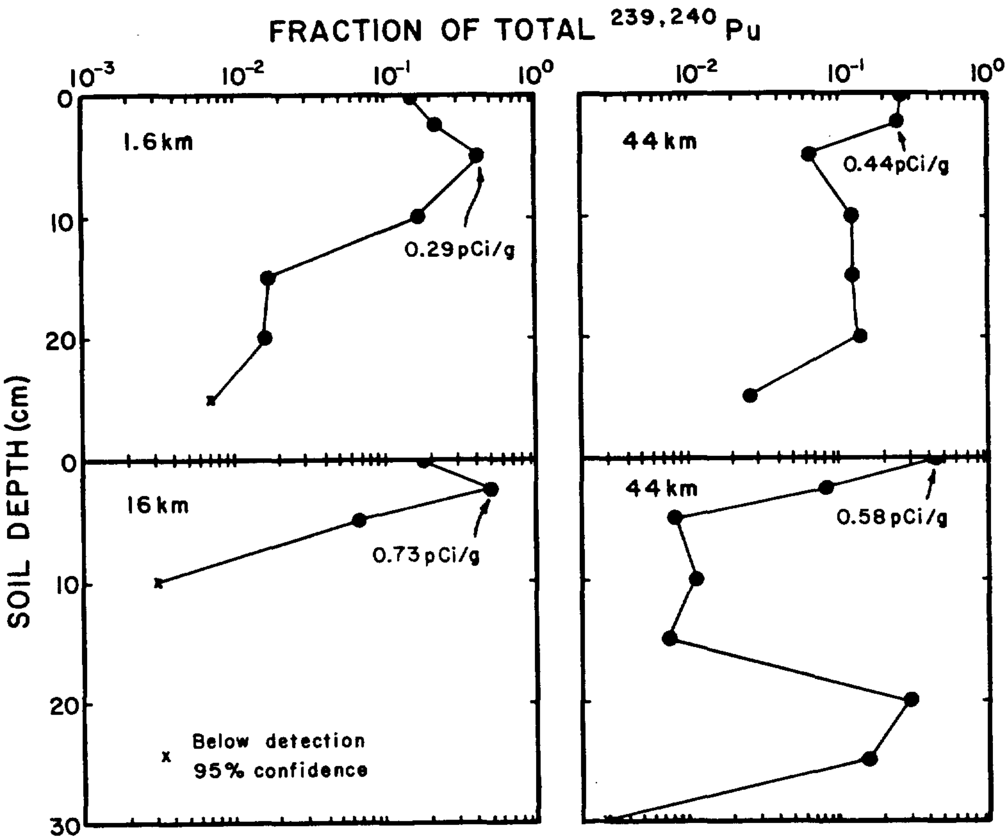

Fig. 2. ^{239,240}Pu Distribution in Trinity Site Soils (Adapted from Nyhan *et al.*, 1976).

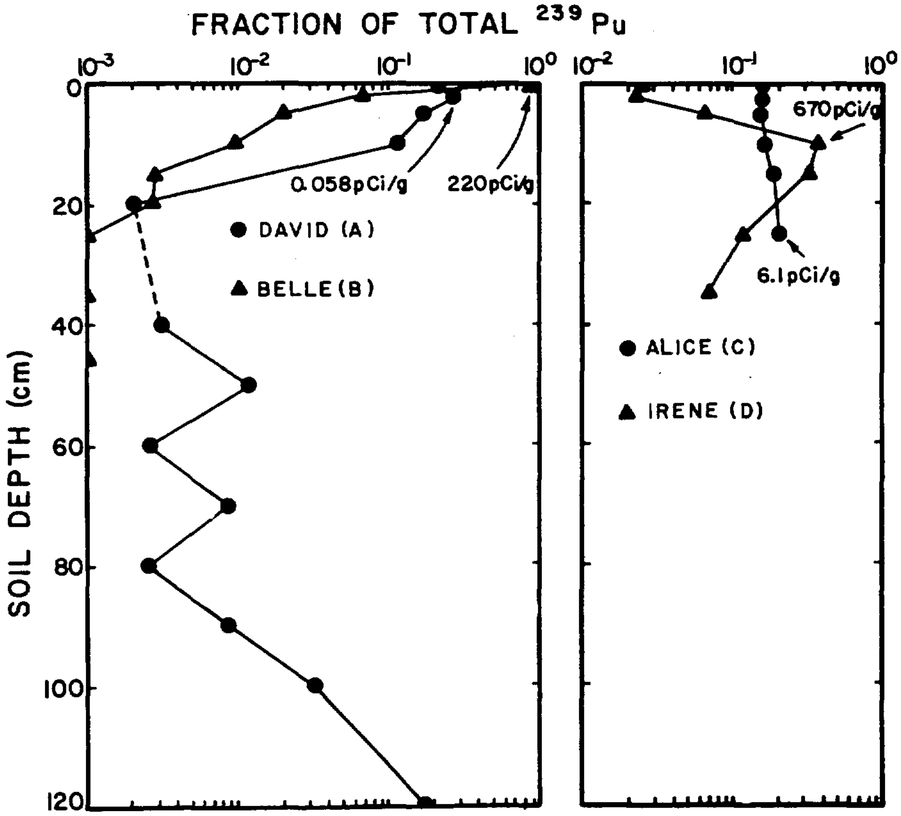

Fig. 3. Distribution of ^{239}Pu in Enewetak Atoll Soils (Adapted from Nervik, 1973).

some depth (Fig. 3, trace D); these profiles generally correlated with areas which had been subjected to testing, construction, or earth-moving activities. Several of the profiles indicated that ^{239}Pu may have migrated beyond the depth sampled, thus the total depth of penetration of ^{239}Pu remained unknown. A 1974 study by Noshkin *et al*, (1976) of radionuclide concentrations in the ground water reservoir of one of the islands revealed that ^{239,240}Pu had migrated to a depth of 80 m below the island surface (Fig. 4). Plutonium-239,240 was detected at all intermediate depths sampled, suggesting that a portion of the ^{239,240}Pu was very mobile throughout the water-saturated coral-sand environments. As was shown in the soil profiles presented by Lynch and Gudiksen (1973), Noshkin (1976) also observed that the vertical distributions of ^{239,240}Pu at any one test well were unrelated to the distributions found on other islands or at different test wells on the same island.

Nevissi *et al*. (1976) discussed the distribution of ^{239,240}Pu in soils of Bikini Atoll. Plutonium-239,240 was detected as deep as 100 cm in soil profile samples with the distribution shown in Fig. 5. Trace A is a profile characteristic of a disturbed soil, possibly due to earth movement, construction, or clean up efforts. Traces B and C are two additional plutonium profiles from Bikini Atoll. It was noted that all the profiles exhibited ^{239,240}Pu distributions similar to those of ^{90}Sr measured on the same samples.

According to Nevissi (1976) there are three possible transport mechanisms for plutonium in Bikini soils: solution transport, biological transport, and suspension transport. Based on the soil and biological conditions at Bikini, it was concluded that solution transport and biological transport were not the major methods by which plutonium migrated downward. The suspension and resuspension of plutonium and plutonium-bearing particles was the principal mode of plutonium transport in these soils. Nevissi (1976) noted the observation of Held *et al*. (1965) that the algal crust of the atoll soils tend to retain a greater portion of all radionuclides. Although it was not possible to predict the mechanism of plutonium retention by the organic crust, the properties of the algal layers and the large adsorptive surface area may be one explanation. It was concluded that the present levels of ^{239,240}Pu and their distribution at Bikini, are not likely to change significantly in the immediate future unless mechanical disruption of the soil occurs.

Safety Shot Sites

A program is in progress under the auspices of NAEG to determine the inventory, distribution, and biological fate of transuranics and other prevalent radionuclides at the Safety Shot Sites of NTS. To date, this program has generated data on soil plutonium, americium, and uranium distributions for areas where safety tests of nuclear weapons have occurred.

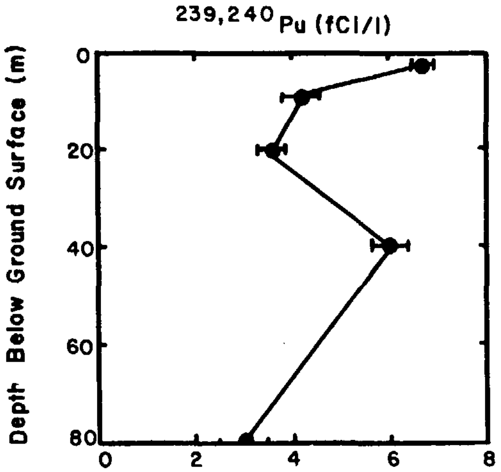

Fig. 4. Vertical Distribution of ^{239,240}Pu in a Test Well, Enewetak Atoll (Adapted from Noshkin *et al*., 1976).

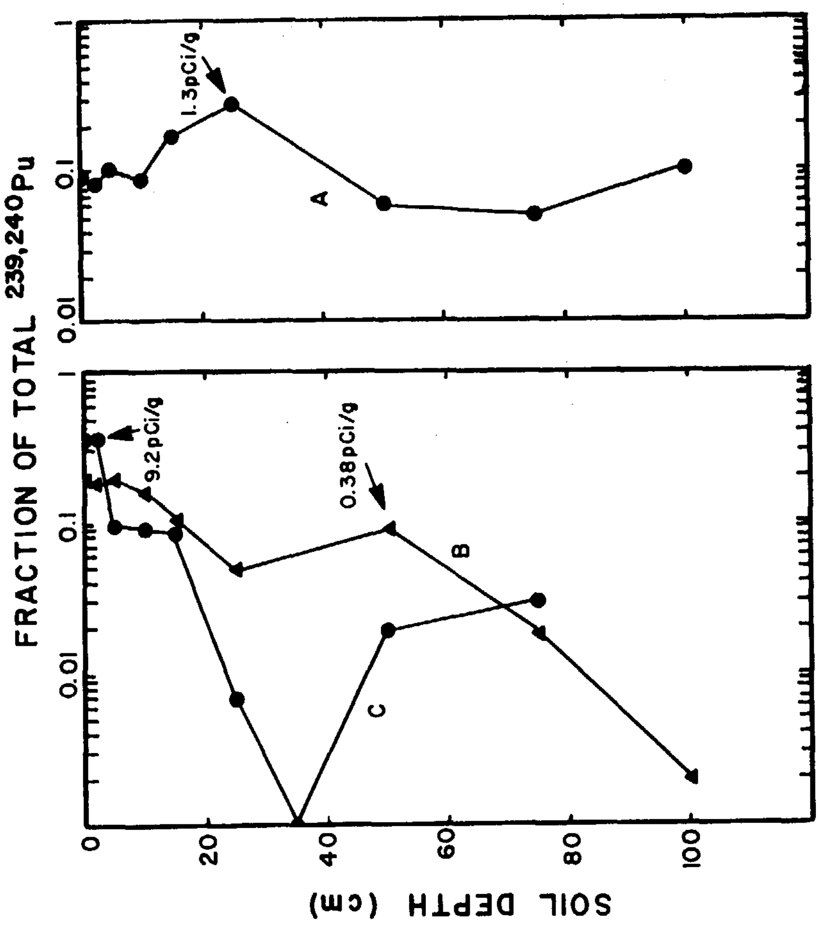

Fig. 5. Distribution of 239,240Pu in Bikini Atoll Soils (Adapted from Nevissi *et al.*, 1976).

Romney *et al*. (1970) were the first to investigate the vertical distribution of ^{239}Pu (^{239,240}Pu) in the Safety Shot areas. Some ^{239}Pu was found to have migrated to 9- to 12-cm depths only 1.3 years after the event. Two profiles taken 10.8 years after the event showed similar distributions and indicated that deeper sampling would probably reveal additional ^{239}Pu penetration.

Between 1972 and 1973 a concerted effort was launched by NAEG to describe the inventory and distribution of uranium, plutonium, and americium at the Safety Shot Sites. This provided a more detailed evaluation of the vertical distribution of the nuclides. Essington *et al*. (1976) evaluated some 70 soil profiles from 7 test areas to determine the maximum depth of ^{239,240}Pu and ^{241}Am penetration and their distribution within the soil profile. As with the observations at Enewetak and Bikini, the ^{239,240}Pu distributions were extremely variable but could be categorized into three groups: normal, disturbed, and abnormal (altered by the physical and chemical conditions of the soil). Figure 6, trace A shows a normal or unaltered ^{239,240}Pu profile where the concentrations decreased with depth in a regular fashion described, perhaps, by a multiple exponential distribution. Figure 6, trace B shows a disturbed ^{239,240}Pu profile and differs from the Profile A in the large residual activity in the lower portions of the profile. This profile represents the ^{239,240}Pu distribution in a streambed area, where it is presumed that intense storm runoff has mixed the streambed materials and effectively distributed the ^{239,240}Pu throughout the depth sampled. Similar profiles were reported by Lynch and Gudiksen (1973) and Nevissi (1976) for highly sorted beach materials at Enewetak and Bikini Atolls. Other evidence of disturbance included: that of small mammal digs, post-event decontamination efforts, and the growth and reduction of blow sand mounds prevalent in the desert environment. Figure 6, trace C represents a rather widespread observation reflecting the effects of soil formation on ^{239,240}Pu distribution. The small increase in ^{239,240}Pu was correlated with the top of the "B" horizon, the zone of clay accumulation.

A very important aspect of plutonium migration into soils is the effect of time. Unfortunately, no experiments were designed nor were any measurements taken which would allow a detailed comparison of short and long term downward movement of plutonium. However, one of the profiles collected by Romney (1970) in 1958, 1.3 years after the safety shot event, was compared to a profile collected in 1973 by NAEG from a point 50 m away (Essington, 1976). The two profiles are shown in Fig. 7, normalized to fraction of total ^{239,240}Pu in the profile. The two profiles are similar, however the 1973 profile indicates a slightly deeper penetration of plutonium. The 1958 profile indicates that most of the plutonium found in the soil migrated shortly after deposition, and only a small amount may have moved to greater depths thereafter. In comparing these two profiles one must consider the sampling and analytical variability associated with each sample. Analytical variabilities (specifically radioactive counting statistics) for the 1973 samples are in the order of the symbol size; however, the variability for the 1958 samples was not given.

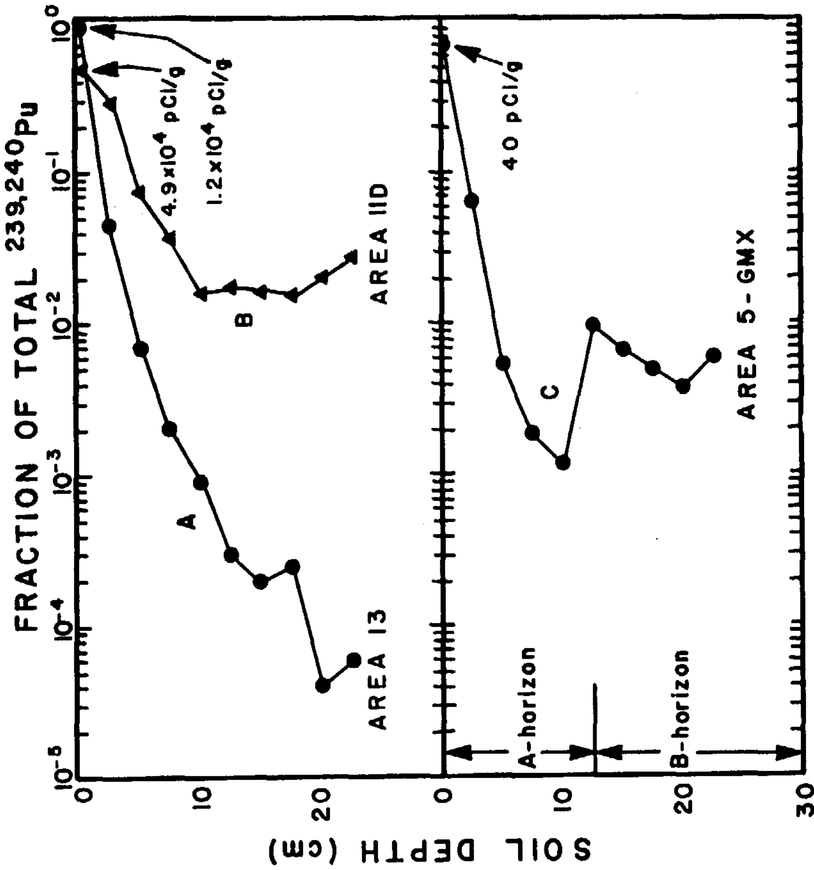

Fig. 6. Distribution of $^{239,240}Pu$ in Soils of the Nevada Test Site (Adapted from Essington *et al.*, 1976).

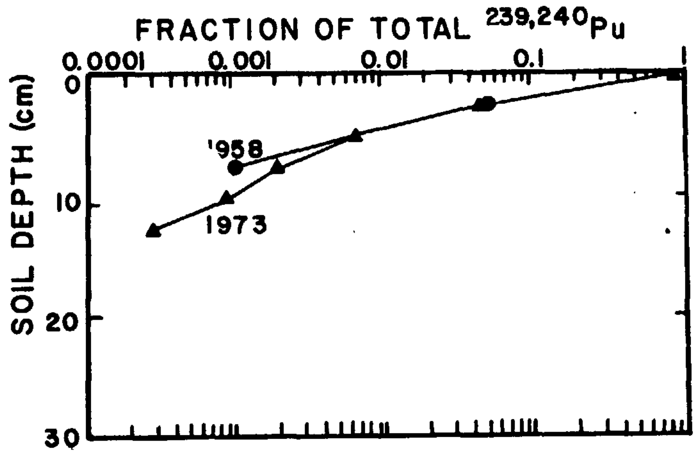

Fig. 7. Distribution of $^{239,240}Pu$ in Two Soil Profiles Collected in 1958 and 1973 at NTS (Adapted from Essington *et al.*, 1976).

Very few measurements have been made of ^{241}Am movement in soils. Differential movement of americium and plutonium appears to have occurred in some of the soil profiles collected by NAEG. Essington (1976) presented a profile of ^{239,240}Pu/^{241}Am which indicated that small amounts of ^{241}Am appeared to be moving faster than ^{239,240}Pu at the Safety Shot sites. It should be pointed out, however, that not all of the profiles taken showed this pattern; ^{241}Am and ^{239,240}Pu appeared to move at the same rates in many of the profiles.

Accidental Releases of Plutonium

Fowler *et al.* (1968) reported results of soil sampling for plutonium at Palomares, Spain, after the accidental destruction of two nuclear weapons in that farming community. After considerable cleanup effort, a series of plots were located for the purpose of following the dispersion of the remaining low levels of plutonium from virgin land and from cultivated land. Figure 8 shows two such plutonium profiles which are the result of averaging nine fractional cores from each site. The effects of plowing the soil can be seen in the highly variable plutonium profile from the plowed field. A coring method was used to collect the soil profile samples from the two Palomares plots. In reviewing the data from this experiment it was noted that the distribution of plutonium predicted from the analytical results might be influenced by the cross-contaminating effect of the coring tool. Small particles of plutonium or plutonium oxide may have been dragged from the more radioactive surface soil into deeper profile samples, giving the appearance of a constant level of plutonium with depth.

The other major accidental release of plutonium for which soil profiles were investigated occurred at the Rocky Flats, CO, Nuclear Fuels Re-Processing Plant. A series of fires occurred over the years possibly releasing small amounts of plutonium to the environment, but probably the largest concentrated release occurred when drums filled with plutonium-laden oil and cuttings began leaking. To determine the distribution and amount of ^{239}Pu around the environs of the Rocky Flats plant, Krey and Hardy (1970) began a sampling program which included evaluation of the vertical distribution of ^{239}Pu (^{239,240}Pu) in the local soils. A number of soil profiles were taken in 1970, two of which are shown in Fig. 9. Plutonium-239 was found to a depth of 13 cm although the distributon was widely variable, presumably due to differences in soil chemical properties. In some cases as much as 60% of the ^{239}Pu was below 5 cm.

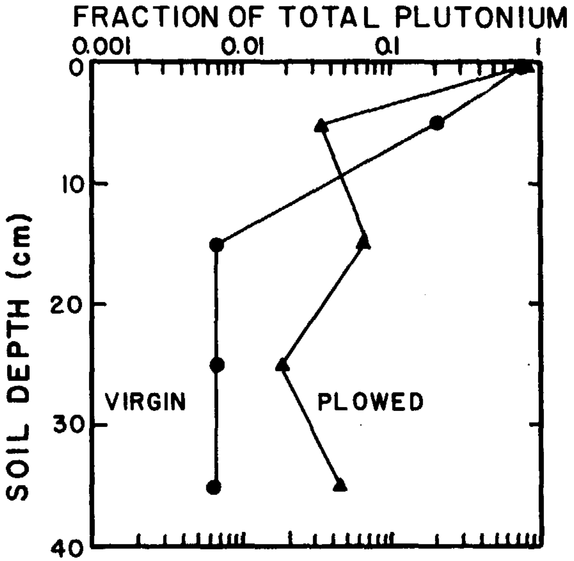

Fig. 8. Distribution of Plutonium in Palomares Soil (Adapted from Fowler *et al.*, 1968).

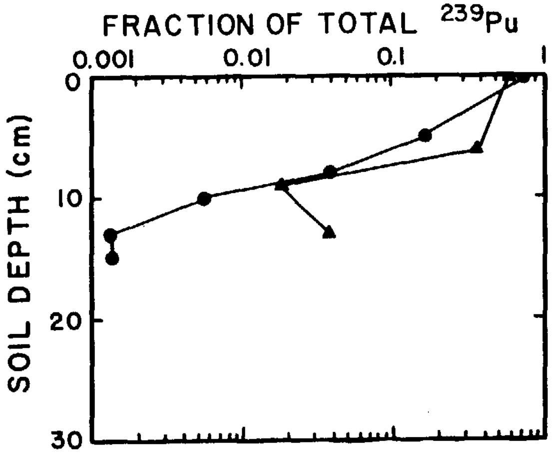

Fig. 9. Distribution of ^{239}Pu in Rocky Flats, Colorado Soil (Adapted from Krey and Hardy, 1970).

Special Studies

There have been a number of special studies dealing with migration of transuranics in soils, particularly with respect to laboratory investigations and radioactive waste disposal problems.

Routson *et al.* (1975) reported on a study to evaluate soils from eastern Washington and South Carolina in their capacity to sorb ^{237}Np and ^{241}Am. In this study the effects of Ca^{++}- and Na^{+}-ion concentrations on the radionuclide distribution coefficient, K_d, were determined. Distribution coefficient values for ^{237}Np decreased in all cases of increased Ca^{++}- and Na^{+}-ion concentrations, as shown in Fig. 10. Values for ^{241}Am also decreased with increasing Ca^{++}- and Na^{+}-ion levels in the South Carolina soil. However ^{241}Am-K_d values from the Washington soil were greater than 1200 and were not influenced by different concentrations of either Ca^{++} or Na^{+}.

The distribution coefficient can be related to ion migration rate in soil, making the proper assumptions that equilibrium is attained during leaching and that the species measured in the laboratory determination of K_d are the same as those in the leaching system and that the system is saturated. For example:

$$v_r = v_w \; \theta / [(1-\theta) \rho \; K_d + \theta]$$

where v_r = velocity of the radionuclide in soil,
v_w = velocity of the leaching water,
θ = fractional porosity,
ρ = grain density, and
K_d = measured distribution coefficient.

Using the above relationship and data of Fig. 10, ^{237}Np, as a soluble ion, can migrate much faster than ^{241}Am, particularly at low Ca^{++}- and Na^{+}-ion concentrations. Of course the specific physical and chemical characteristics of the soil will govern the actual degree of movement.

Glover *et al.* (1976) conducted equilibrium sorption measurements on 17 soils from the United States with plutonium and americium nitrates. This experiment was designed to discover relationships between plutonium and americium and certain physical and chemical characteristics of the soils. Among the factors investigated were cation exchange capacity, soluble carbonate, soluble salt content, organic carbon, pH, Eh, and particle size distribution. Distribution coefficient values for americium at a concentration of 10^{-10} $\underline{M}$ ranged from 82 to 10,000, and plutonium K_d values ranged from 35 to 14,000 for a concentration of 10^{-6} $\underline{M}$. Regression analysis was used to determine correlations between chemical/physical parameters and the degree of radionuclide sorption. Cation exchange capacity and clay and sand contents were found to be the most important, which indicates that a conventional ion exchange process was responsible for the sorption of plutonium and

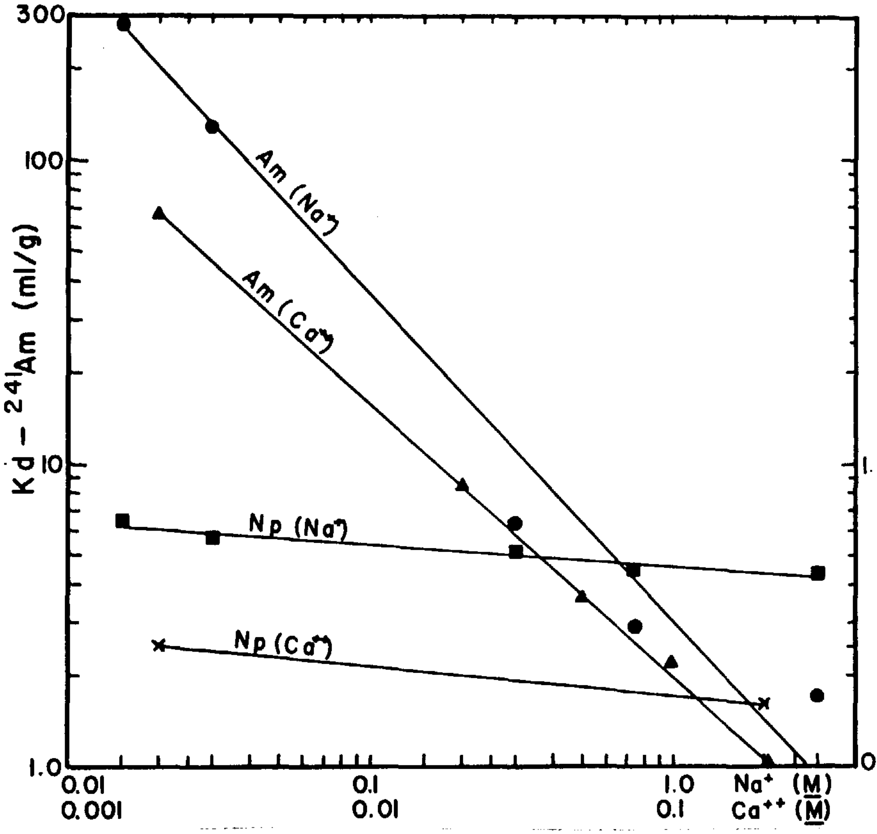

Fig. 10. ^{241}Am and ^{237}Np Distribution Coefficients (K_d) for South Carolina Soil (Adapted from Routson *et al.*, 1975).

americium. During the same study, elution characteristics of plutonium were determined in several of the soils for which K_d had been measured. Figure 11 shows the distribution of plutonium in three of those soils. As the K_d increased so did the retention of plutonium in the uppermost portions of the soil column.

Considerable amounts of ^{238}Pu are presently being used as heat sources for power generators particularly in space applications. Patterson *et al.* (1976) conducted an experiment to determine the fate of metallic ^{238}Pu under environmental conditions. Within environmental test chambers, a ^{238}Pu heat source specimen or fragments thereof, were placed on soils under simulated climatic conditions. The first "rain water" that percolated through the test soil contained a small amount of ^{238}Pu. This very rapid ^{238}Pu breakthrough indicated that some of the material that spalled from the fuel was very small particulate or colloidal. Auto-radiography of the percolated rain water revealed plutonium oxide particles of 0.05 to 0.8 µm diam. with a count median diameter of .095 µm. Nearly all of the plutonium oxide particles carried through the soil by water were less than .05 µm diam., except for the occasional presence of a particle as large as 4 µm. Patterson presented a number of soil profiles taken in the test chamber at the conclusion of the long-term leaching experiments. Using auto-radiography, it was observed that each plutonium oxide particle was associated with one microscopically visible soil particle. This indicated that trapping of plutonium oxide particles by soil was not a matter of filtration of the fine oxide particles, but that there was agglomeration of individual oxide and soil particles.

A recent, highly specialized, study on the distribution of actinides under waste disposal trenches at the Hanford Reservation in Washington was reported by Price and Ames (1976). This program was established to assess the future radiological impact of actinides in the ground underlying retired trenches and to develop methods for the long term control of contaminants. Samples were taken at 5 cm, 50 cm, 2 m, 4.5 m, and 9 m below the trench floor using a core sampler driven 0.5 m or 1 m for each profile increment. Both ^{239}Pu (^{239,240}Pu) and ^{241}Am were determined on the sediments from the core samples. One of the profiles is reproduced in Fig. 12. Examination of the core samples revealed both particulate and nonparticulate plutonium that may have been responsible for the observed distribution. The particulate fraction consisted of discreet plutonium particles, 2 to 25 µm diam., and was restricted to the top portions of the sediment columns. Price and Ames (1976) discussed in some detail hydrolysis and the possible soil-mineral-plutonium reactions which may have been responsible for sorption of plutonium in the lower profile fractions.

Similar coring studies by Holcomb *et al.* (1976) are in progress at Savannah River Plant.

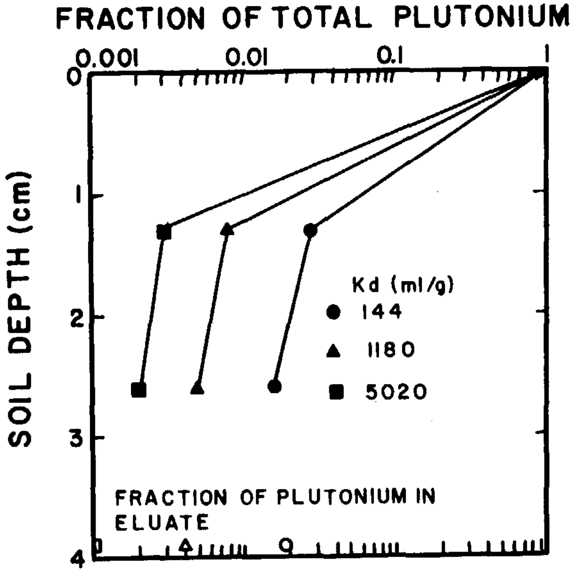

Fig. 11. Distribution of Plutonium in Soils Exhibiting Varying Plutonium Distribution Coefficients (K_d) (Adapted from Glover *et al*., 1976).

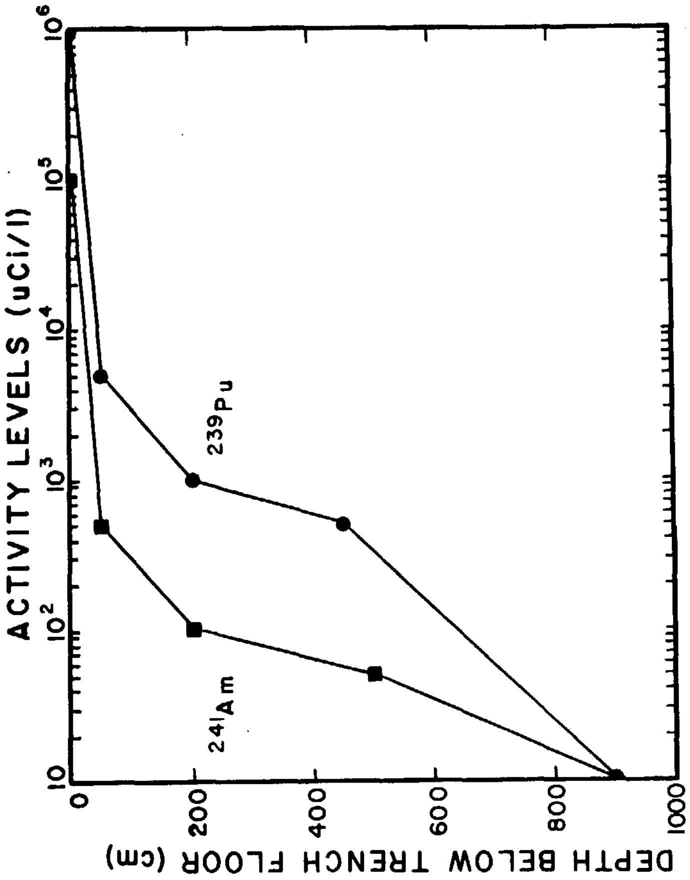

Fig. 12. Distribution of ^{241}Am and ^{239}Pu Under Waste Disposal Trench at the Hanford Reservation, Hanford, Washington (Adapted from Price and Ames, 1976).

MOVEMENT OF URANIUM

Although uranium is not a transuranic, certain uranium isotopes in the nuclear fuel cycle have been identified as potentially hazardous and warrant discussion.

Till (1975) discussed the recycle aspects of ^{233}U bred from ^{232}Th in the High Temperature Gas-Cooled Reactors. The recycle ^{233}U can contain up to 1200 ppm ^{232}U, which was stated to be very radiotoxic, as having a relatively high specific activity, and as accounting for 82.1% of the total ^{232,233}U radioactivity. Table 3 lists the assumed activities of various uranium isotopes in 1 g of ^{233}U fuel. It can be seen that ^{232}U can account for greater than 50% of the total uranium activity present. Because of the potential environmental hazard of ^{232}U, an evaluation of uranium migration through soils is in order.

Natural uranium is distributed throughout minerals and sediments the world over. Hansen and Stout (1968) noted that uranium and thorium appeared to accumulate in soils as a result of rock weathering and soil formation. Maximum natural thorium concentrations tended to be in the "B" horizon, whereas the maximum uranium concentrations were most often in the topsoil. This suggests that thorium moves down the soil profile more rapidly than uranium. Generally, uranium and thorium concentrations are highest in the soil clay fractions; however, clays developed from carbonate rich soils show relatively low uranium and thorium concentrations. Hansen (1970), in a subsequent study, suggested that 600,000 years after initial deposition, most of the nuclides of the uranium and thorium families are in the finer textured soil fractions and that the uranium that is retained by the soil is more likely to be fixed in some immobile form.

Probably the only literature citation revealing uranium migration in environmental soils on a short-time basis was presented by Essington (1976). During the nuclear testing program discussed earlier in the section entitled "Safety Shot Sites," there was an explosive test of a ^{235}U enriched nuclear device. A soil profile taken from near the ground zero area (Fig. 13) showed the vertical distribution of ^{239,240}Pu ^{238}U, and ^{235}U. The ^{238}U levels reflected the natural concentrations in the mineral structure of the soil. However, when ^{235}U was distributed over the soil surface it did migrate into the soil profile as evidenced by the ^{235}U distribution. This added ^{235}U appeared to behave very much like ^{239,240}Pu, also distributed at approximately the same time.

Table 3. Assumed Radioactivity of Uranium Isotopes in 1 gram of ^{233}U Fuel

Isotope	Equilibrium recycle atom fraction	Equilibrium recycle activity (Ci)
^{232}U	.001	2.14×10^{-2}
^{233}U	.613	5.82×10^{-3}
^{234}U	.243	1.51×10^{-3}
^{235}U	.080	1.72×10^{-7}
^{236}U	.063	3.99×10^{-6}

Adapted from Till (1975).

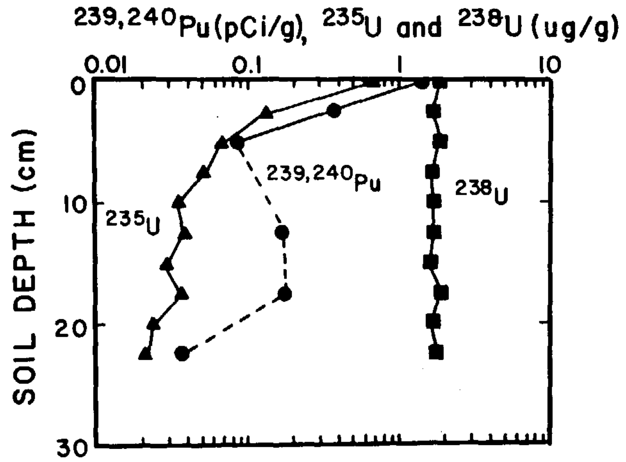

Fig. 13. Distribution of ^{239,240}Pu, ^{235}U, and ^{238}U in NTS Area 11-A Soil (Adapted from Essington *et al.*, 1976).

PREDICTIONS OF TRANSURANIC MOVEMENT IN SOILS

Attempts have been made to describe the movement of transuranics through soils on a mathematical basis. Lester *et al.* (1974) investigated the hypothetical band and impulse releases of transuranic parent-daughter chains through a soil column (that column which follows the ground water flow path) to a surface water body from an underground geologic nuclear waste disposal site. Numerical resolutions of analytical (mathematical) solutions revealed that differences in adsorption characteristics between soluble chain members, axial dispersion, and radioactive decay all act to reduce radionuclide discharge rates at the exit of the soil column. This method draws on a uniform, one-dimensional model and requires evaluation of the distribution coefficient, K_d, for each soil-radionuclide combination. Thus, this technique has limited application to predicting movement of transuranics in terrestrial soils where radionuclide K_d values can vary over wide ranges along the transport path. Other mathematical models to predict the distribution of radionuclides in soils have been presented by Cohen (1975) and Aleksakhin (1963).

A direct application of predicting transuranic distributions in a soil profile was presented by Jakubick (1976), who attempted to reconstruct plutonium concentrations in soils of south-west Germany. By correlating the few ^{239,240}Pu measurements available in Germany with the fallout data from Ispra, Italy, the probable history of plutonium fallout was reconstructed. From these data the plutonium input function in the vicinity of Heidelberg was derived. The plutonium distribution in a soil column from fallout in the United States was used in conjunction with the calculated cumulative fallout input function to predict the plutonium distribution and turnover rate in Heidelberg soils. Results of this study imply that the migration of fallout plutonium takes place completely, or to a great extent, in the form of discrete particles probably weakly aggregated to soil particles. The removal of fallout plutonium particles (analogous to plutonium oxide particles) from a given soil layer seemed to obey an exponential law, with a characteristic turnover time of 5-6 years for a 5 cm thick partially saturated soil layer.

DISCUSSION

This review has pointed out that there are many and diverse observations of transuranic distributions in soils; however, almost all of the observations are limited to plutonium. Some information on the distribution of americium has been produced, but generally only at nuclear weapons testing areas. Measurements on the distributions of the other transuranics in environmental soils is lacking. Although the literature on transuranic chemistry in soils was not discussed, many of the papers reviewed either hypothesized, concluded, or in isolated cases measured the fact that plutonium had migrated into the soils and probably was in particulate form, either as plutonium oxide particles, hydrated plutonium oxide colloids, or plutonium particles aggregated to soil particles.

Even though the plutonium distributions in soil are quite variable, there has been an attempt at a rough categorization. It appears that irrespective of the source, plutonium profiles in soil can be categorized into three groups: normal, disturbed, and abnormal.

Normal profiles exhibit no obvious profile development and the plutonium distribution appears to reflect a complex exponential function. This type of profile is more likely to be associated with an undisturbed soil. Only a limited number of observed plutonium profiles can be categorized as normal.

The disturbed plutonium profiles result from activities associated with construction, burrowing animals, water and wind erosion, and redeposition after the initial deposition of plutonium. The plutonium distributions in these disturbed soils are quite varied. They might show a uniform distribution of plutonium with depth, that is, as deep as samples were collected, as was found on beaches and open areas of Enewetak and Bikini Atolls, or in a desert wash area where runoff from intermittent but severe rainstorms actively mix the top layers of material.

Abnormal plutonium distributions are generally unexplainable and a series of hypotheses have been used to suggest the possible mechanisms responsible for individual observed distributions. These possibilities range from faulty analyses of some fraction (or fractions) to unidentified mechanical disturbances, to the physical and chemical interactions of plutonium with the soil constituents. Understanding of the abnormal distributions may be a key to describing the physical and/or chemical interactions effective in the redistribution of plutonium or transuranic nuclides within soils. For instance, the accumulation of plutonium in specific soil zones which are related to soil forming factors, such as A and B horizons or carbonate lenses, has been recognized. The filtering action of specific soil horizons for small colloids containing plutonium or plutonium particles, and the potentially different chemistry among the zones, may be the long term governing factors for plutonium migration rates in soils. The authors are aware of no study currently in progress to address these problem areas.

Early in the history of plutonium distribution studies, it was thought that plutonium was so tightly bound to soil, that plutonium could not penetrate into the soil in the short time that has elapsed since its first introduction into the soil. This may be true for the migration of soluble ionic plutonium since it may be tightly bound by soil particles. However, the migration of plutonium as soluble complexes, or in conjunction with soil particles also occurs to a sufficient degree, and some plutonium does migrate rapidly and to great depths, as evidenced by the ground water study at Enewetak.

Close examination of many of the published profiles indicates that the true front of plutonium migration was not observed due either to insufficient depth of sampling or inadequate detection capability. The implication is that a small amount of plutonium may have migrated to greater depths than those measured.

The few inferences that can be drawn from observations of the duration of migration suggest that for a short period of time after deposition onto soil there is a rapid penetration of plutonium into the soil. Indications are that a small amount of plutonium moves rapidly, and to some depth, into the soil as though the plutonium were complexed and relatively uneffected by the attractive forces of soil particles. For large concentrations of freshly deposited plutonium this effect may ultimately present a significant environmental hazard. Studies should be conducted to assess the magnitude of the possible mechanisms involved.

The turnover rate of fallout plutonium has been calculated to be 5-6 years for each 5 cm of depth giving rise to the exponential depth profiles in undisturbed soils. This observation appears to be somewhat independent of the total rainfall of the area.

Many of the early soil profiles were collected by coring techniques which may have introduced a cross-contamination variable, predicting higher-than-real concentrations of plutonium at depth. However, this problem was recognized and much of the later work used techniques (such as the "trench method") which provided some control over that variable. One cannot argue at this point with definitive data that the coring method is faulty, only that the variable does exist and remains unanswered.

ACKNOWLEDGEMENT

The authors wish to thank Mr. T. Ensminger, Oak Ridge National Laboratory (ORNL) for his assistance in querying the *Nevada Applied Ecology Information Data Base*, for references dealing with the distribution of transuranic nuclides in soils.

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FACTORS INFLUENCING THE CHEMICAL EXTRACTABILITY OF ^{241}Am FROM A CONTAMINATED SOIL

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ABSTRACT

Factors influencing the extractability of ^{241}Am from an artificially contaminated soil were investigated. This was done with an equilibrium batch technique using a CH_3COOH-NH_4OH and HNO_3-NaOH extracting systems. The influence of several soil components was determined indirectly by selectively removing them from the soil. The effect of water- and HCl-soluble salts and organic matter on ^{241}Am extractability was small. The most marked effect was due to the soil organic fraction that was not water- or HCl-soluble. This organic fraction was influential under both low and high pH conditions, but its influence was particularly marked under low pH conditions. The free iron-oxides had an appreciable effect under low pH conditions, but no observable effect in the high pH range. Though to a lesser extent, the free silica and alumina, amorphous alumino-silicate, and possibly residual organic matter also showed some influence. These results provide some implications on the conditions that influence the movement of ^{241}Am in soils and its availability to plants. A review of the literature on the behavior of Am in soils is included.

INTRODUCTION

It is well recognized that with the increasing use of nuclear energy for different purposes, particularly power generation, there is an increasing need to improve our understanding of the fate and the effects of the radionuclides that might be released into the biosphere. The major transuranium elements that are important from the long-term hazard standpoint are Np, Pu, Am, and Cm (McKay, 1961; Thomas and Perkins, 1974; Pigford, 1974; Pigford and Ang, 1975). The hazard stems from the fact that these elements are extremely toxic (Denham, 1969; Stannard, 1973; Bair and Thompson, 1974) and several of their isotopes are long-lived.

This paper deals with an aspect of the chemistry of transuranium elements in soils. In order to limit its scope, it deals mainly with Am. More specifically, it is concerned with the chemical extractability of ^{241}Am from an artificially contaminated soil as a function of pH and several different soil components. This study is an extention of our previous work with ^{238}Pu and ^{242}Cm (Nishita, 1976). As it will be discussed later, this paper provides several implications on the movement of ^{241}Am in the soil and its availability to plants.

LITERATURE REVIEW

Studies on the behavior of Am in soils are still limited. A recent review of Am in soil and plant systems by Brown (1976) indicates that more work has been done with plants. Since the present study pertains to the chemistry of Am in soils, this review is constrained to the information that may serve as background and may be helpful in assessing its behavior in soils.

Origin of Am. Among the isotopes of Am, there are two that are relatively long-lived, the 433-year ^{241}Am and the 7,370-year ^{243}Am. Of these, ^{241}Am is the more important. The isotope ^{241}Am is produced in nuclear reactors by multiple neutron capture (Pigford and Ang, 1975):

$$^{239}\text{Pu}\ (n,\gamma)\ ^{240}\text{Pu}\ (n,\gamma)\ ^{241}\text{Pu} \xrightarrow[15y]{\beta^-} {}^{241}\text{Am}$$

The isotope ^{243}Am is produced by more complicated successive series of neutron capture. In a nuclear detonation, these isotopes can be produced directly in another manner, i.e., through the formation of very high mass U isotopes by extremely rapid succession of neutron capture (Diamond *et al.*, 1960; Thomas and Perkins, 1974). These U isotopes then decay rapidly to form a spectrum of transuranium elements. The proportion of radionuclides produced by these pathways differ somewhat. Whereas ^{239,240}Pu and ^{241}Am are the main transuranium α activities from nuclear detonations, ^{238}Pu, ^{241}Am, and the Cm isotopes are the principal α activities from nuclear reactor operations (Thomas and Perkins, 1974).

The major source of worldwide contamination by ^{241}Am is ascribed to the occurrence of ^{241}Am in the fallout from nuclear detonations. The ^{241}Pu/^{239,240}Pu activity ratios of global fallout present in soils as of January 1, 1970, was 8.2 (Krey and Krajewski, 1972). The ^{241}Am, from ^{241}Pu decay, is increasing in the global environment (Poet and Martell, 1972; Harley, 1975; Livingston, Schneider, and Bowen, 1975; Krey *et al.*, 1976). Krey *et al.* (1976) calculated that ^{241}Am content of the present fallout in the soil will peak in the year 2037 and will represent 42% of the ^{239,240}Pu activity. The ^{241}Am/^{239,240}Pu activity ratio of integrated global fallout in February 1974 was around 0.22. According to Poet and Martell (1972), the maximum radioactivity contribution of ^{241}Am would probably occur 70-80 years after the environmental release of ^{241}Pu.

Contamination Level. The actual levels of fallout ^{241}Am in surface soil in the United States are estimated to be in the range of 2.4 fCi/g (calculated from total Pu level of 20 fCi/g (Harley, 1975) by taking Am/Pu ratio into consideration). At Bikini Atoll, which was exposed to close-in fallout, ^{241}Am values from 1.2 to 45 pCi/g were observed in surface soil samples collected in 1972 (Nevissi, Schell, and Nelson, 1976). The surface soil (0-1 cm) collected near the Pu processing plant at Rocky Flats in 1969 contained from 0.01 to 0.14 pCi/g. Americium-241 values from 2×10^{4} to 10.4 nCi/g have been observed in the surface soils (0-5 cm) from Nevada Applied Ecology Group study areas at Nevada Test Site (NTS) (Romney, 1975). Measurements of the vertical distribution of ^{241}Am in certain soil profiles at NTS indicate detectable amounts of it are present to a depth of 25 cm (Romney, 1975; Essington *et al.*, 1976). From inferences made from profile distribution of Pu (Nevissi, Schell, and Nelson, 1976), Am might be detectable as deep as 75-100 cm in some soil profiles at Bikini Atoll. Americium and Pu parallel one another in behavior because of their chemical relationship.

Chemical Properties. The chemical properties of Am that one, perhaps, should be cognizant of in considering its behavior in soils follow:

Americium has the valence states of (0), (III), (IV), (V), and (VI) (Keenan, 1959; Penneman and Keenan, 1960; Keller, 1971). Americium is found in four oxidation states, Am (III), Am (IV), Am (V), and Am (VI), in aqueous solutions (Keller, 1971). The trivalent, pentavalent, and hexavalent Am occur as hydrated Am^{+++}, AmO_2^{+}, and AmO_2^{++} ions in the absence of complexing agents. The trivalent state is the most stable one encountered in aqueous solution. The tetravalent Am is stable only in concentrated fluoride and phosphate solutions (Yanir, Givon, and Marcus, 1969), undergoing rapid disproportionation in all other solutions. The pentavalent and hexavalent Am are reduced slowly to lower valence state by the radiolysis products formed by the action of their own α-radiation (Gel'man *et al.*, 1962; Myasoedov *et al.*, 1974). Americium (V) disproportionates in acid solutions to form Am (VI) and Am (IV), and the latter is immediately reduced to Am (III). Americium (V) and Am (VI) are susceptible to reduction by many common reagents (Stokely and Moore, 1967). Americium is relatively stable in strong acid, but is reduced by water if the acid concentration falls below about 0.01 M (Coleman *et al.*, 1963). Under alkaline condition, Am (VI) solution is not stable, but is reduced completely to Am (V) in 1-2 days (Cohen, 1972). In the form of simple hydrated trivalent ion, it reacts similarly to trivalent lanthanide ion and forms sparingly soluble trifluoride, hydroxide, phosphate, iodate, oxalate, and double sulfate (Penneman and Keenan, 1960; Gel'man *et al.*, 1962; Keller, 1971). The readily soluble Am (III) compounds include the perchlorate, nitrate, sulfate, and halides (Gel'man *et al.*, 1962). The tendency of Am (III) to form complexes with the anions of inorganic acids are in the following order: $F > SO_4^{2} > NO_3 > Cl > ClO_4$ (Gel'man *et al.*, 1962; Keller, 1971). It forms chelates with synthetic chelating agents such as DTPA (diethylene triaminepentaacetic acid) (Wallace, 1972a, 1972b) and very likely with a number of soil organic components (Mortensen, 1963; Kononova, 1966; Schnitzer and Khan, 1972; Stevenson, 1972). Stability constants of a number of Am chelates have been compiled by Keller (1971). The tendency of Am to form complexes with organic and inorganic ligands relative to other transuranium elements is Pu (IV) > Am (III) = Cm (III) > Pu (VI) > Np (V) (Keller, 1971).

Hydrolysis is an important property that needs to be considered, since the state of hydrolysis of hydrolyzable ions is one of the controlling factors on their adsorption and desorption. For example, Matijević *et al.* (1961) studying the Th adsorption of AgI sols found that the hydrolyzed species of Th are more strongly adsorbed than the simple hydrated ion. According to the ion adsorption model of James and Healy (1972), hydrolysis could bring about conditions favorable to the adsorption of lower charged species that are formed as it progresses. This is explained by the fact that as hydrolysis proceeds, the solvation energy term in their model changes in magnitude much more than the coulombic energy term. The hydrolysis of microamounts of Am depends on its concentration and on the chemical environment such as the kind and the concentration of the acid or base electrolyte (Korotkin, 1973b, 1974). At present, the effect of an electrolyte is attributed to the change of the activity of the ions due to the change of their state of hydration. Samartseva (1969), using the technique of sorption on Pt, determined the beginning of hydrolysis of ^{241}Am (10^{-10} M) to be at pH = 2.0-2.5. Shalinets and Stapanov (1972), by electromigration technique, found the beginning to be in the similar pH range for Am at 10^{-5} M concentration. Korotkin (1973a, 1973b, 1974), using distribution chromatography and electromigration techniques, determined it to be at pH = 0.5-1.0 for ^{241}Am concentration range of 10^{-10}-10^{-6} M. Obviously, there are some discrepancies, part of which may be due to experimental conditions. In any case, it would be helpful to know the state of hydrolysis of Am as a function of pH in a complex system as a soil.

Along with hydrolysis, the degree of polymerization under various conditions needs to be considered also. The beginning pH for the polymerization of Am in a complex system such as a soil does not appear to have been defined. Some experiments with simple aqueous solutions, however, have been done. An investigation by Samartseva (1969) on the sorption and desorption of ^{241}Am from Pt showed that up to pH = 4.0-4.5, the sorption process was reversible, indicating that ^{241}Am was ionically dispersed, whereas at pH = 5, irreversibility of ^{241}Am sorption, which is a characteristic of the colloidal state, occurred.

Sorption and Desorption in Soils. Sorption studies on Am in soils is still limited. Routsen, Jansen, and Robinson (1975) determined the distribution coefficients (K_d values) of ^{241}Am in subsoils of two different soils, one from the arid western United States and the other from the humid southeastern United States. The first soil, which was of neutral reaction and higher in $CaCO_3$ content and cation exchange capacity, showed considerably higher K_d values (> 1,200) than the second soil, which was acidic and higher in clay content.

Few comparisons of sorption with respect to other transuranium elements have been made. Routsen, Jansen, and Robinson (1975) showed that ^{241}Am (III) was adsorbed to soil stronger than ^{237}Np (V). By inference from the quantitative difference of behavior between Am and Pu, it appears that in general Am may be less strongly sorbed to soils than Pu. For example, measurements of the vertical distribution of ^{241}Am and ^{239,240}Pu in NTS soil profiles have found that in some profiles, a decrease in the ^{239,240}Pu/^{241}Am ratio with depth occurred, indicating greater vertical movement of ^{241}Am relative to ^{239,240}Pu (Fowler and Essington, 1974; Gilbert *et al.*, 1975; Essington *et al.*, 1976).

Resident Am in a soil collected beneath a waste storage crib was leached by groundwater and by 1 N $NaNO_3$ to a considerably greater extent than the resident Pu (Hajek, 1966). In the chemical extraction of soil, collected in the Pu-contaminated area at the Rocky Flats Plant and assumed to be contaminated by Am and Pu in the dioxide form, the Am extraction coefficients were generally higher than the values for Pu by one or two orders of magnitude (Cleveland and Rees, 1976). Also, plant uptake studies have shown strong indication that Am is more readily taken up by the plant than is Pu (Cline, 1968; Price, 1972; Romney *et al.*, 1975; Adams *et al.*, 1975). The comparative environmental behavior of Pu and other transuranium elements have been discussed in an excellent review by Dahlman, Bondietti, and Eyman (1976).

Few experiments that relate to the desorption of ^{241}Am in soils have been done. Hajek (1966) found that 7.5% of the resident Am in a soil collected under a waste storage crib was leached by 1 N $NaNO_3$. Knoll (1969) in a soil column experiment found that the soil had little or no effect on removing the Am from any of the tagged organic compounds applied, and certain untagged organic compounds (di-(2-ethylhexyl) phosphoric acid and hydroxyacetic acid) completely leached the ^{241}Am from the tagged soil. Wallace (1972a) reported that 100% of ^{241}Am applied to a loam soil was extracted by DTPA, but not by EDDHA (ethylenediamine di-ortho-hydroxyphenol acetic acid). Apparently, certain chelates are more stable to exchange in the soil than others. Cline (1968) showed a soil type effect. In soil columns leached with 100 inches (254 cm) of irrigation water, in excess of 98% of the Am was retained in the top 1 cm of the acid soil, whereas only 76% was retained in the top of the alkaline soil.

MATERIALS AND METHODS

The soil used for this study was Aiken clay loam (Ultisol) and its chemically treated forms. The chemical properties of these soil materials are shown in Table 1.

The influences of the various soil components on the extractability of ^{241}Am were determined indirectly by chemically removing selected components and determining the effect of their removal. Except for free alumina, silica, and amorphous alumino-silicate, the procedures for the removal of the various components have been reported previously (Nishita, 1976). In essence, treatment I was the $CaCl_2$ treatment of the virgin soil. This treatment removed the water-soluble salts and organic matter, reduced the amount of exchangeable Na, K, and Mg, and retained Ca as the major (> 50% saturation of the cation exchange complex) exchangeable cation. Treatments II through V were done in sequential manner. Treatment II was the digestion of the virgin soil with dilute HCl. This treatment removed the HCl-soluble organic matter and salts and carbonates, if present. Treatment III, which was the digestion of the samples in 30% H_2O_2, brought about the decomposition of organic matter, the dissolution of Mn oxides, and some residual carbonates, if present. Treatment IV

Table 1. Chemical characteristics of treated and untreated Aiken clay loam.

Soil Material	pH	Organic-C** %	Cation Exchange Capacity** meq/100 g	Extractable Cations Na	K	Mg	Ca	Free Iron Oxides	HNO$_3$ Extractable Mn
				meq/100 g				%	
Untreated	6.57	2.18 ± 0.01	19.91 ± 0.20	0.05	1.16	1.49	10.73	14.43	0.18
Treatment I	6.90	2.08 ± 0.04	19.75 ± 0.64	0.04	0.32	0.12	13.72	14.55	0.15
Treatment II	6.16	2.31 ± 0.02	20.45 ± 0.42	0.04	0.32	0.09	10.28	14.87	0.15
Treatment III	7.47	0.49 ± 0.01	16.53 ± 0.25	0.04	0.18	0.04	10.73	15.29	0.02
Treatment IV	7.61	0.43 ± 0.002	17.98 ± 0.68	0.27	0.51	0.04	12.09	1.03	0.01
Treatment V	8.73	0.32 ± 0.003	14.99 ± 0.09	1.20	0.15	0.07	18.01	0.87	0.01

* Treatments: Untreated = virgin soil, I = H_2O-soluble OM and salt removed, II = HCL-soluble OM and salts removed, III = OM, Mn oxides, and residual carbonates removed after treatment II, IV = free iron oxides removed after treatment III, V = free silica, alumina, amorphous alumino-silicate, and residual OM removed after treatment IV.

** With standard deviation.

was the digestion of samples in $Na_3C_6H_5O_7$-$NaHCO_3$-$Na_2S_2O_4$ mixture to remove free iron oxides. Treatment V was the boiling of samples in 2% Na_2CO_3 to remove partially the free alumina and silica and amorphous alumino-silicates. Some residual organic matter that was protected against decomposition by H_2O_2 is also removed. With this treatment, the samples were washed five times with acidified 1 N NaCl to remove the carbonates before the $CaCl_2$ treatment.

The soil materials were artificially contaminated by $^{241}Am\ (NO_3)_3$ and extracted by the equilibrium batch method as described before (Nishita, 1976). Equilibration in the present experiments was done over a 48-hour period with continuous mixing on a mechanical mixer. Two extraction systems were used, e.g., CH_3COOH-NH_4OH and HNO_3-NaOH systems. To obtain the appropriate pH levels, CH_3COOH-NH_4OH extraction solutions were prepared by titrating 1 M CH_3COOH with varying amounts of NH_4OH. Since 20 ml of these solutions were diluted to 25 ml in each tube with distilled H_2O, the actual concentration of the CH_3COO^- ions in the tubes was constant at 0.8 M throughout the pH range studied. With the HNO_3-NaOH system, the appropriate pH level was attained by adding varying amounts of HNO_3 or NaOH. The pH values reported are those of the supernatant liquid from the soil suspensions (soil extracts) after the equilibration period. The preparation of the soil extracts for radioassay and counting was done in the same manner as before with ^{238}Pu and ^{242}Cm (Wood *et al.*, 1976).

RESULTS AND DISCUSSION

CH_3COOH-NH_4OH System. The effects of CH_3COOH-NH_4OH system on the extractability of ^{241}Am from treated and untreated Aiken c. l. are shown in Fig. 1. With the untreated soil (control), the ^{241}Am extractability increased only slightly, if at all, from pH = 2.5 to pH = 3.8 and then increased noticeably to pH = 4.8. Between pH 4.8 and pH 7.7, it decreased rapidly to 0.3% of dose and then remained very low (0.04-0.3% of dose) to pH 10. Above pH 10, it increased appreciably.

Treatment I, which removed the H_2O-soluble organic matter and salts, significantly reduced (at the 95% significance level) the extractability of ^{241}Am relative to that of the untreated soil at pH 11.6. At other pH levels, there was no significant difference. Treatment II, which removed the HCl-soluble materials, significantly reduced the ^{241}Am extractability relative to the control at pH 6.5, but otherwise it also did not cause any significant difference. Since the significant differences were small, it was concluded that the two treatments, in general, had only slight effect on the extractability of ^{241}Am.

The general pattern of the ^{241}Am extractability curves were similar to those obtained with ^{239}Pu and ^{242}Cm, and its intrinsic nature has been discussed previously (Nishita, 1976). In essence, the initial increase of ^{241}Am extractability to pH = 4.8 was ascribed to increasing complexation of ^{241}Am by the acetate ions. As increasing amounts of NH_4OH were added to increase the pH,

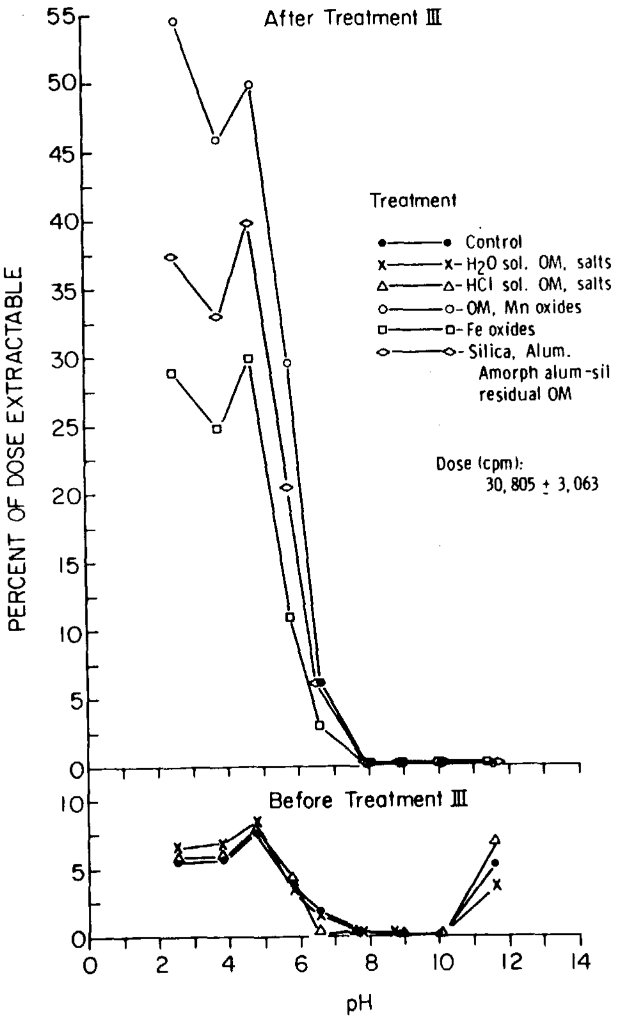

Fig. 1. Effect of pH on the extractability of ^{241}Am from contaminated Aiken clay loam chemically treated to remove various components (CH_3COOH-NH_4OH System)

increasing amounts of HN_4^+ and CH_3COO^- ions were formed. The NH_4^+ ions exchanged a fraction of ^{241}Am ions off the ion exchange complex, while the CH_3COO ions formed complexes with them. At pH = 4.8, the common ion effect became effective and the sorption of the hydrolysis products of ^{241}Am became the more dominant factor. According to the ion adsorption model of James and Healy (1972) conditions favorable to the sorption of hydrolysis products may be brought to fore as hydrolysis proceeds. Concomitant with the hydrolysis of ^{241}Am, increasing amounts of the pH-dependent sorption sites (cation exchange capacity) probably were formed with increasing pH and thus favored the sorption. The latter condition arises from increasing ionization of mineral hydroxyls and hydrous oxides and humus colloids with increasing pH (Buckman and Brady, 1969). At the higher pH levels, americium hydroxide probably was formed, but its state of polymerization and the nature of its association with the soil solid phase is not known. As evidenced by the increasing coloration of the soil extract from pale brown to very dark brown with increasing pH, the increasing extractability of ^{241}Am above pH 10 was due mainly to the extraction of the alkali-soluble organic matter. The extractability of soil organic matter by alkalies is well known (Mortensen, 1965; Kononova, 1966). The influence of organic matter became more evident after treatment III.

Treatment III significantly increased (at the 95% significance level) the ^{241}Am extractability relative to the control and treatments I and II in the pH range below pH = 7.7 and significantly reduced it at pH 11.6. The ^{241}Am extractability above pH = 7.7 ranged from 0.0 to 0.04% of dose. Although a portion of manganese oxides were removed (Table 1), the major cause of the enhanced extractability was the removal of the organic matter. This is evidenced by the very low ^{241}Am extractability above pH 10 after the removal of soil organic matter. Furthermore, manganese oxides that were formed under natural conditions are likely to be only slightly soluble under alkaline conditions. The results indicate that the high ^{241}Am extractability above pH 10 before the removal of organic matter was due to the association of ^{241}Am with alkali-soluble organic matter and that its extractability was very low from the inorganic fraction of the soil and/or the precipitate of americium hydroxide under strongly alkaline condition. Part of the enhanced extractability below pH 7.7 may be due to the reduction of the sorption sites with the removal of the organic fraction. The cation exchange capacity was decreased from 20.45 me/100 g to 16.53 me/100 g (Table 1) by the treatment. The major cause of the effect, however, was considered to be due to the strong association of ^{241}Am with the organic fraction of the soil before its removal. From another point of view, it appears that ^{241}Am ions are quite readily exchanged off from the inorganic fraction by H^+ ions under acidic condition.

Treatment IV significantly reduced the ^{241}Am extractability relative to that by treatment III in the pH range below pH = 7.7. Above this pH level, there was no significant difference between the two treatments. The latter effect has been discussed above. Not all of the reactions causing the marked reduction of ^{241}Am extractability in the acidic pH range are known, but part of it might be due to the exposure of additional inorganic and organic sorption sites by the removal of free iron oxides. This is supported by the fact that the cation exchange capacity of the soil was increased from 16.53 me/100 g to 17.98 me/100 g (Table 1) and that some residual organic matter (0.43% organic C) was present. Iron oxides are known to occur as cementing materials and partial coatings on the surface of the clay micelles and as discrete oxide particles (Carroll, 1958; Mehra and Jackson, 1960; Jenne, 1968; Anderson and Jenne, 1970).

Treatment V significantly increased the ^{241}Am extractability relative to that by treatment IV in the acidic pH range below pH ≅ 7.7. Above pH 7.7, the extractability was low as treatments III and IV. The former effect indicates that the partial removal of free silica and alumina, amorphous alumino-silicates, and residual organic matter had some effect. As to which of these materials played the dominant role was not determined, but in view of the marked influence of organic matter, the partial removal of the residual organic matter must certainly have had some influence. The residual organic C was reduced from 0.43% to 0.28% (Table 1). There was also a reduction of the cation exchange capacity from 17.98 me/100 g to 14.99 me/100 g.

HNO$_3$-NaOH System. The effects of the HNO$_3$-NaOH system on the extractability of ^{241}Am are shown in Fig. 2. The ^{241}Am extractability for the control and the soil after treatments I and II were not appreciably different. In general, the extractability of ^{241}Am decreased at a very rapid rate between pH 0.65 to pH ≅ 2.2 and then at a slower rate between pH 2.2 to pH ≅ 3.5. Above pH 3.5, its extractability remained low to pH ≅ 7.5, increased between pH 7.5 to pH ≅ 12.3, and decreased above pH 12.3. The decrease of its extractability at about pH 12.3 suggests a change of state of the tracer, organic complexing system, and/or the sorption complex. The precise nature of this change is at present not known.

After treatment III, the ^{241}Am extractability decreased at a rapid rate between pH 0.65 to pH ≅ 4 and then at a slower rate to pH ≅ 6.5. Above pH 6.5, its extractability remained very low (0.01 to 0.09% of dose). Its extractability after treatments IV and V varied to some extent relative to that after treatment III, but they generally paralleled each other. The low extractability of ^{241}Am in the alkaline range indicated again that its extraction before the removal of organic matter was due to the extraction of alkali-soluble organic matter. The results showed again that ^{241}Am extractability from the inorganic fraction of the soil was very low under alkaline condition. On the acidic side of the pH range, the removal of organic matter increased appreciably the extractability of ^{241}Am, indicating again that ^{241}Am was strongly associated with soil organic matter under acidic condition. Also, below pH 4, ^{241}Am was readily exchanged by H$^+$ ions from the inorganic fraction of the soil.

CH$_3$COOH-NH$_4$OH vs HNO$_3$-NaOH Systems. Fig. 3 shows the relative effect of the two extracting systems used. The HNO$_3$-NaOH system is a strong acid-strong base system that is much less complexing compared to the CH$_3$COOH-NH$_4$OH system. An overview of the figure shows immediately that wider pH range was covered by the HNO$_3$-NaOH system. Before the removal of organic matter, the CH$_3$COOH-NH$_4$OH system extracted more ^{241}Am than the HNO$_3$-NaOH system on the acidic side roughly to pH 7 and vice versa above pH 7. The greater ^{241}Am extraction by the CH$_3$COOH-NH$_4$OH system in the acidic range point to the well-known fact that complexing action is an important factor in the extractability

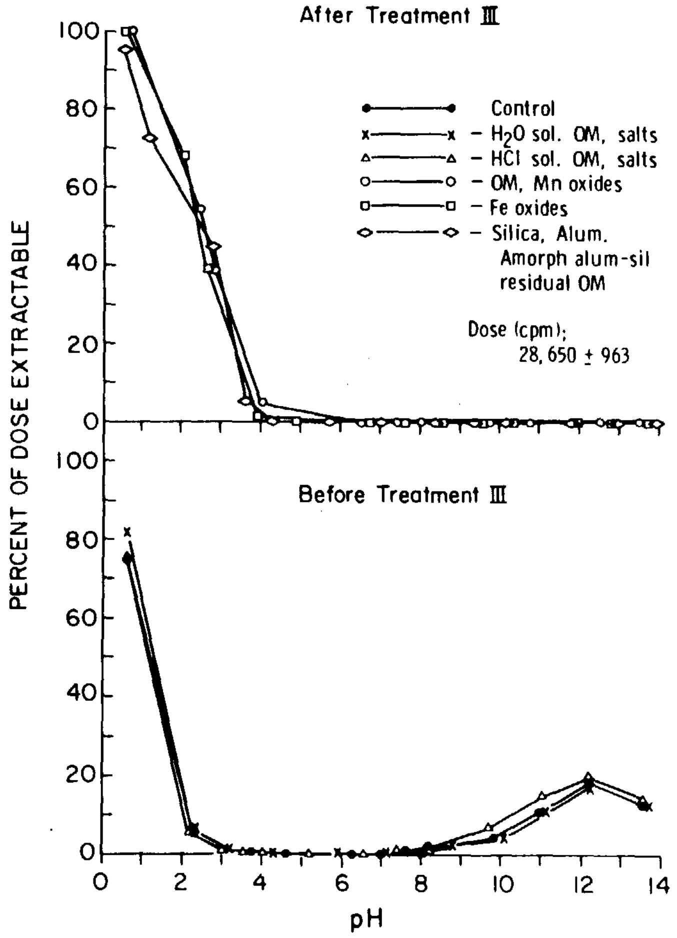

Fig. 2. Effect of pH on the extractability of ^{241}Am from contaminated Aiken clay loam chemically treated to remove various components (HNO$_3$-NaOH System)

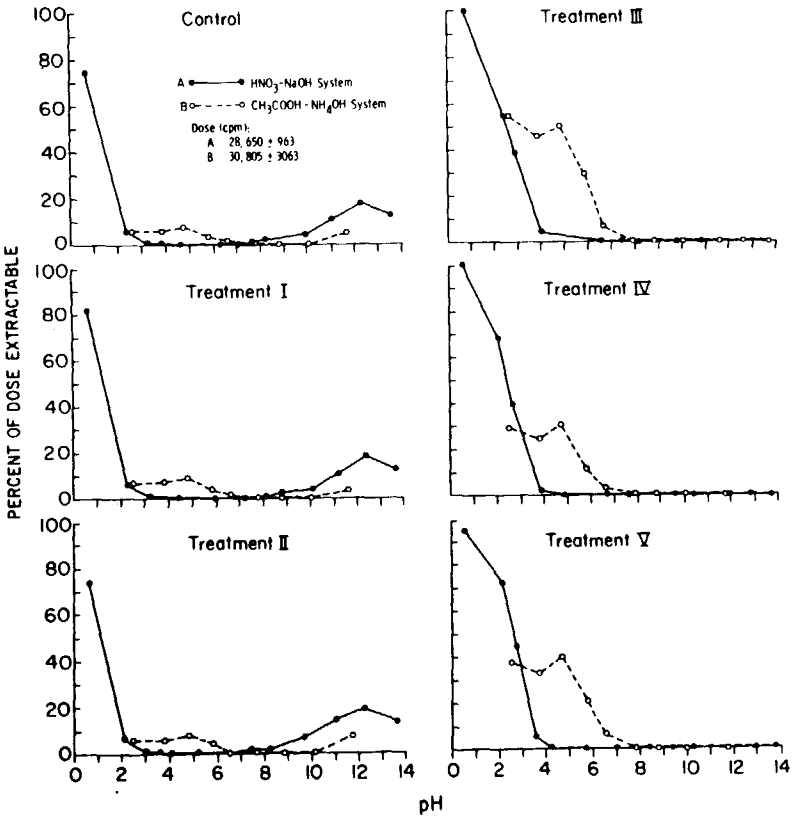

Fig. 3. Relative effect of extracting systems on the extractability of ^{241}Am from contaminated Aiken clay loam chemically treated to remove various components

of metal ions. The greater ^{241}Am extraction by the HNO_3-NaOH system in the alkaline pH range is a reflection of the fact that NaOH is a more effective extractant of soil organic matter than NH_4OH.

After the removal of organic matter, the effectiveness of the CH_3COOH-NH_4OH system in extracting ^{241}Am increased. Apparently, the stability constant of some ^{241}Am-soil organic matter complexes were greater than that of ^{241}Am-acetate complex.

Distribution Coefficients. It is well recognized that sorption and extractability are inseparably interrelated. This relationship is usually expressed by distribution coefficients (K_d), i.e., the ratio of activities in the solid phase to that in the liquid phase. Since our emphasis here has been the extractability rather than the sorption, the distribution coefficients presented (Table 2) are defined as the ratio of the activities in the liquid phase to that in the solid phase, i.e.,

$$K_d' = (A_l/A_s) \quad (m/v)$$

where A_l = equilibrium activity in the solution phase, A_s = equilibrium activity in the solid phase, m = mass in grams, and v = volume of solution in milliliters. The K_d values (Table 2) reflect, the curves shown in Figs. 1 and 2. With the HNO_3-NaOH system, the minimum K_d value occurred at pH 7.58, whereas it was at pH 10.20 with the CH_3COOH-NH_4OH system. The occurrence of the minimum extraction at the higher pH level by the latter system was considered to be due to the lower effectiveness of NH_4OH compared to NaOH in extracting soil organic matter.

GENERAL DISCUSSION

The results provide several implications on the possible behavior of Am in soils. In a kaolinitic soil containing appreciable amounts of organic matter, Am is likely to move fairly readily by the leaching action of water under strongly acidic (pH < 3) or alkaline (pH > 8) condition. In the intermediate pH range (pH 3-8), the movement of Am is expected to be low, but it is likely to be higher on the alkaline side due to the solubilization of alkali-soluble organic matter. With the addition of a relatively weak complexing agent such as the CH_3COO ions, the movement is likely to be appreciably increased in the acidic side, but not necessarily in the alkaline side of the pH range. Our unpublished results of the extraction of ^{239}Pu have shown that complexing action of CH_3COO ions is very weak compared to citrate, tartrate, or oxalate ions. In view of this, the addition of a strong complexing or chelating agent probably will increase the Am movement on the alkaline side of the pH range as well. This, however, remains to be determined.

In a kaolinitic soil, which is devoid or very low in organic matter content, Am is likely to move quite readily under strongly acidic condition (pH < 4), but only slightly under alkaline condition (pH > 7). In the intermediate pH

Table 2. Distribution coefficients (K_d') of untreated Aiken clay loam under HNO_3-NaOH and CH_3COOH-NH_4OH systems.

HNO_3-NaOH System		CH_3COOH-NH_4OH System	
pH	$K_d' \times 10^{-4}$*	pH	$K_d' \times 10^{-4}$*
0.67	2462.4 ± 841.3	2.55	45.9 ± 2.5
2.32	44.8 ± 0.1	3.81	46.3 ± 2.0
3.20	7.7 ± 0.4	4.76	65.3 ± 0.7
3.75	2.7 ± 0.5	5.77	31.8 ± 0.02
4.61	1.2 ± 0.06	6.54	13.1 ± 0.5
6.27	0.6 ± 0.1	7.69	2.7 ± 0.4
6.96	0.5 ± 0.1	8.76	0.5 ± 0.2
7.58	6.0 ± 3.1	10.02	0.4 ± 0.1
8.16	16.1 ± 0.0	11.57	44.0 ± 0.4
9.84	39.4 ± 3.9		
11.00	39.4 ± 3.9		
12.19	171.1 ± 2.6		
13.55	113.5 ± 6.3		

*Average of 2 samples with standard deviation

range (pH 4-7), the movement is likely to be slow, but it is likely to increase with decreasing pH. In the minus organic matter soil, the addition of a complexing agent is likely to enhance the movement of Am to a much greater extent in the acidic pH range than in the soil containing appreciable amounts of organic matter.

Since the movement of Am is related to its availability to plants, the statements made above apply also to Am uptake by plants, insofar as they pertain to the pH range for normal plant growth. As pointed out previously (Nishita, 1976), the pH range for productive plant growth is between pH 3 and pH 10 (Arnon, Fratzke, and Johnson, 1942; Arnon and Johnson, 1942; Small, 1954). At both ends of this pH range, the plants generally encounter adverse soil conditions for normal growth, so the optimal range is usually narrower.

In summary, several soil components, particularly the organic matter, were found to influence the extractability of ^{241}Am as a function of pH. The implications provided by this study appear to be interesting to test under environmental conditions.

FUTURE PLANS

The study described here will continue with ^{239}Pu and ^{237}Np.

ACKNOWLEDGMENTS

The authors wish to thank R. M. Haug and T. Rutherford for the chemical analysis of soil materials and A. J. Steen for counting the samples on the alpha spectrometer. These studies were supported by Contract E(04-1) GEN-12 between the U.S. Energy Research and Development Administration and the University of California.

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PLUTONIUM ASSOCIATION IN SOILS

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ABSTRACT

This paper reviews the association of plutonium in the soils and sediments of several contaminated environmental samples. The plutonium-matrix association based on selected chemical treatments is reviewed. The association of plutonium with the soil particle sizes is reviewed and a soil plutonium index is derived for several contaminated sites to permit comparison of the potential transfer to man via the inhalation pathway. The soil plutonium index accounts for the fraction of the total activity in the resuspendible sizes, the relative concentration of the activity on the soil particle, and the potential lung deposition upon inhalation.

The derived soil plutonium indices for four sites range from 0.52 to 1.26 from the least to the most severely contaminated site. The order of increasing site severity is the Nevada Test Site (0.52), Oak Ridge National Laboratory (0.69), Mound Laboratory (1.18), and Rocky Flats (1.26). Limitations of the soil factor are discussed.

INTRODUCTION

Plutonium is an element of mixed blessing. On the one hand, it is a valuable potential energy resource; on the other, its radiological properties make it a potentially hazardous element if mismanaged or mishandled. Its future potential will be determined, in part by understanding the properties, behavior, and fate of this radioelement after dispersal in the biosphere. The papers in this symposium volume provide information on the many environmental aspects of plutonium which are being investigated. As the information base and understanding regarding the various reactions of plutonium grow, sound decisions can then be made of the role of plutonium in mankind's future.

This paper reviews the association of plutonium in contaminated soils and sediments. The above-background contamination which exists at several nuclear facilities is the result of defense-related activities.

These sites, which present no immediate hazard to man, provide opportunities for understanding plutonium behavior under natural environmental situations. Major emphasis in the paper is placed on the association of plutonium on soil particle sizes and the size relationship to potential deposition in the lung. As an initial approach to relate the relative importance of plutonium in soils when the pathway to man is through resuspension and inhalation, a "soil plutonium index" is derived for each site.

CHEMICAL ASSOCIATIONS

Once plutonium comes in contact with soil and sediment particles, it becomes firmly attached to the host particle. This strong attraction is exemplified by the high adsorption coefficients exhibited by soils and sediments in laboratory studies with soils (Rhodes, 1957a and b; Prout, 1958), in sediments from fallout (Wahlgren *et al.*, 1976), in sediments from accidental release of soluble plutonium (Rogers, 1975), and in soils from bomb testing activities (Noshkin *et al.*, 1976). The distribution coefficient, Kd, which is defined as the ratio of the adsorbed plutonium per unit weight to that in solution per unit volume, ranged from about 1,000 in laboratory studies to about 100,000 in field situations.

The high Kd naturally leads to the question of mechanism(s) of the attraction. One attempt to define the Pu associations in a natural system was that by Edgington *et al.* (1976). Sediments from Lake Michigan contaminated at fallout levels were sequentially treated to remove plutonium. Selected results from their study are included in Table 1. The $MgCl_2$ treatment was used to remove easily exchangeable forms. The citrate-dithionite treatment was used to remove the reductant-soluble forms (those which might be associated with iron and manganese oxides), and the NaOH treatment was used to extract the organically associated forms. Not included in the table is the crystalline phase determined by fusion analysis. From the result obtained, Edgington *et al.* (1976) concluded that most of the plutonium is associated with iron and manganese oxides. They also changed the sequence of extractions in order to evaluate the effect of preceding treatments on the succeeding treatment. The major effect reported by them was that the organic associated plutonium increased slightly (Table 1).

One of the modifications in treatment was extraction with heated sodium citrate without the sodium dithionite. This treatment extracted 35% of the plutonium. Tamura (1976b) also treated several contaminated soils and sediments with unheated sodium citrate; the amount extracted ranged from 7 to 14%. These same samples, when extracted with unheated citric acid, released 23 to 59%. One might suspect that if citric acid had been used to extract the plutonium from the Lake Michigan samples, a higher percentage of the plutonium would

Table 1. Distribution $^{239,240}Pu$ in Selected Lake Michigan Sediments Modified After D. N. Edgington *et al.*, 1976.

Samples	Total (pCi/g)	$MgCl_2$ %	Citrate-Dithionite %	NaOH %	% Total Recovery
(B) Grand River downstream of the power plant	0.017	3.8	95.9	0	99.7
(A) 2 miles offshore Lake Michigan	0.016	0.6	80.0	0.17	80.8
(C) 6 miles offshore Lake Michigan	0.013	0	80.0	0	80.0
(I) Station 5A (85 m depth)	0.017	0	45.7	0	45.7
(H) Station 5B (84 m depth)	0.045	0	89.7	0	91.2*
(H) Station 5B**		5.6	96.3	5.0	106.9

*Including 1.5% recovery from fusion analysis; all other samples showed on plutonium.

**NaOH treatment before Citrate-Dithionite.

have been extracted. Since the citrate does not reduce the iron and manganese in the samples under the extraction conditions, the citrate-extractable plutonium may not be bound with those elements. Iron and manganese form surface oxide coatings on particles; thus the dithionite treatment coupled with citrate would serve to remove the surface coatings and possibly remove surface-sorbed plutonium as well.

Bondietti *et al.* (1976) evaluated the amount of organically associated plutonium in a floodplain soil in Oak Ridge. They used a resin technique and estimated that about 15% of the plutonium in the soil may have been associated with soil organic material. Sodium-resin, prebuffered at the soil pH, was used to maintain constant pH and decalcify the soil organic matter. This is in contrast to sodium hydroxide extraction of organic matter which raises the pH. Destruction of organic matter with sodium hypochlorite released 82% of the plutonium. They concluded that since the alkaline hypochlorite treatment would not dissolve the iron oxides, most of the plutonium was surface sorbed rather than intimately associated with iron. Since the floodplain soil and the Lake Michigan sediments represent different matrices, the mode of association could be different as postulated by these investigators. It would be informative if the different treatments employed by the investigators could be applied in a similar manner to both samples so that deductions regarding the association could be evaluated.

The high Kd's of over 100,000 associated with aged (years) sources described above, as well as with the groundwater particulates at Enewetak Atoll and the sediment from the canal near Mound Laboratory, compared to the lower Kd of about 1,000 for shorter term (days) simulated tests in the laboratory suggest a biogeochemical process which increases retention. The process of increasing retention has implications in reduction of uptake by plants as well as in the resuspension process, both in terms of the size of the particles being resuspended and the depositional pattern in the lungs.

PLUTONIUM SIZE CHARACTERISTICS

In discussing plutonium particle size, two immediate considerations become apparent. First, when Pu releases occur from stacks, the particles to be considered are pure metal or metal oxides. When these particles deposit on the ground surface, interactions with the soil particles occur. Depending on the character of the initial source, the intensity of the association with soil particles may be different. This paper is concerned with the plutonium in soils and sediments, hence it does not consider the initially released particles.

That plutonium attaches itself to host soil particles has been verified directly by microscopic and alpha-track measurement techniques (Hayden, 1976; Nathans and Holland, 1971; Nathans *et al.*, 1976). Mork (1970) studied the size association of plutonium in Yucca Flat on the Nevada Test Site (NTS) and showed that most of the plutonium was associated with soil particles greater than 44 μm diameter. Tamura (1974, 1975, 1976a) also studied soil samples from safety shot areas in NTS. These studies of particle size association included separation of different silt and clay sizes and showed that the association was primarily with the coarse silt (50 - 20 μm) and fine sand (125 - 50 μm) sizes.

The particle size segregation of Pu-contaminated soils by Tamura (1974) was made without benefit of chemical or physical treatment to induce particle dispersion. This approach was taken in order to evaluate the degree of association of plutonium as it exists in the soil. Later data were reported on size association of the plutonium with and without ultrasonic treatment for dispersion (Tamura, 1976b). In addition to comparing the effect of ultrasonic treatment on two NTS soils, data was included on a soil from the floodplain in Oak Ridge (ORNL) and a segment of bottom sediment from the canal at Mound Laboratory (ML). The same data are reproduced in Tables 2 and 3.

The ultrasonic treatment increased the plutonium contribution of the finer size fractions of the total soil (Table 3). In general, the increase in the plutonium contribution in the finer resuspendible sizes was also accompanied by an increase in mass contribution in these finer sizes (Table 2). This suggests that the plutonium contribution in the finer sizes was primarily due to plutonium-soil particle disaggregation and not necessarily to plutonium detachment from the soil particle. Considering the less than 5 μm sizes, the ultrasonic treatment increased the percentage contribution by two to five times, although the total percentage contribution in the NTS samples was still less than 10%. In the sample from Mound Laboratory, the ultrasonic treatment increased the Pu in the less than 5 μm size from 34% to 81%. More significantly, the Pu in the less than 2 μm size fraction in Mound Laboratory sample increased from 17 to 70%. The importance of the association of the plutonium with the fine size of soil particles will be discussed further in the later section of this paper.

Muller and Sprugel (1977) determined the distribution of plutonium in three soils located within 50 miles of Mound Laboratory in Ohio. Two of the soils were "background" samples reflecting fallout levels; the third was a soil sample taken one mile east of the Laboratory, containing sufficient ^{238}Pu originating from the operations to differentiate it from fallout levels. The samples were treated with a carbonate-oxalate dispersant and the fractions were analyzed for plutonium. Between 60-75% of the plutonium was associated with particles less than 4 μm size, and about 90% of the activity was associated with particles less than 45 μm (silt and clay). For comparison, a bottom sediment sample taken in the canal at Mound Laboratory showed very similar distribution with 81% of the Pu in the less than 5 μm and 100% in the combined silt and clay fraction (Tamura, 1976b).

Plutonium associations with different particle sizes in soils of Rocky Flats have been reported by Little *et al.* (1973). The mode of segregation involved sieves with only mechanical shaking of oven-dried samples. The particle size distribution showed that the less than 104 μm size particles in the soil contributed 3.7% in one sample and 11.2% another. The 3.7% and 11.2% weight fractions reported by Little *et al.* (1973) contributed 11 and 37%, respectively, to the total soil activity. The low amount of fine materials likely caused by the oven drying which would tend to "cement" particles, the short shaking

Table 2. Particle Size Distribution of Selected Samples in Percent From NTS, ML, and ORNL Without (A) and With (B) Ultrasonic Treatment (Tamura, 1976b)

Size Range (μm)	NTS		NTS		ML		ORNL	
	Sample Code							
	A13 - 1A		A13 - 1B		8C - 43-47		IRB - 1	
	A	B	A	B	A	B	A	B
> 2000	19.5	19.4	2.7	2.7	0	0	---	---
2000 - 840	7.2	7.2	3.0	2.9	0	1.9	---	---
840 - 250	24.3	22.1	20.2	19.6	6.9	4.0	---	---
250 - 125	21.1	18.7	29.6	24.7	6.0	2.8	---	---
125 - 53	17.6	16.3	33.6	28.1	6.4	3.7	0.7*	1.2*
53 - 5	5.2	6.3	8.5	10.1	28.8	22.2	23.3	19.8
20 - 5	2.4	3.3	1.1	3.2	32.2	28.4	42.9	38.4
5 - 2	1.4	2.7	0.5	3.0	9.5	7.2	10.8	9.8
< 2	1.3	4.1	0.6	5.4	10.3	29.7	19.6	28.6
Total	100.0	100.2	99.8	99.7	100.1	99.9	99.3	99.78

* Percentage greater than 53 μm.

Table 3. Plutonium Contribution (%) by Particle Size Without (A) and With (B) Ultrasonic Treatment (Tamura, 1976)

Size Range (μm)	NTS		NTS		ML		ORNL
	A13 - 1A		A13 - 1B		8C - 43-47		IRB - 1
	A	B	A	B	A	B	B*
2000 - 840	0.5	0.3	0	0.1	---	---	---
840 - 250	1.5	0.2	1.5	0.1	3.9	---	---
250 - 125	1.4	0.3	0.9	0.2	1.8	---	---
125 - 53	26.7	8.3	14.9	5.9	5.6	---	---
53 - 20	44.4	50.9	75.1	73.2	24.8	6.4	19.3
20 - 5	22.0	32.7	6.3	14.9	29.8	12.3	32.0
5 - 2	2.6	4.5	0.8	4.2	16.9	11.8	8.8
< 2	1.0	2.9	0.4	1.5	17.1	69.5	39.9
	100.1	100.1	100.1	99.9	99.9	100.0	68.0

*The A series of ORNL sample was not analyzed.

time (20 min.), and the absence of water as an agent to maintain dispersion. Bernhardt (1976) also segregated a surface soil sample from Rocky Flats using sodium metaphosphate as a dispersant. Particles less than 100 µm averaged 43% by weight. This fraction contained 75% of the total soil Pu activity.

Tamura size-segregated and analyzed the plutonium in the 5-10 cm depth sample of a soil from Rocky Flats. The activity in the surface sample (0-5 cm) of the soil contained approximately 20,000 dpm/g. Because this sample was highly contaminated, size segregation, which originally requires about 50 grams, was not performed. Segregation of a 5-10 cm depth sample was performed with and without ultrasonic dispersion treatment. The size distribution, plutonium content, and the contribution of each size fraction to the total soil activity are shown in Table 4. In contrast to the dry sieving technique reported by Little *et al.* (1973), these samples were wet-sieved. The less than 125 µm size fractions composed 41.5% of the weight and 82% of the activity in the soil without ultrasonic treatment, and 51.2% of the weight and 91% of the activity in the ultrasonified soil. The results of the nonultrasonified sample were similar to the results reported by Bernhardt. The fractionation reported by Bernhardt (1976) included only the less than 100 µm sizes; and the 10 µm notation referred to by Bernhardt is the size based on 1 g/cm^3 particle density. For silicate density of 2.65 g/cm^3, the corresponding size is 6.1 µm. The 100 µm size is based on sieve opening size and is thus not affected by the choice of density. The sizes reported by Muller and Sprugel (1977) and Tamura (1976b) are based on the silicate density.

The importance of the size association of plutonium with host particles has been recognized by many investigators (Volchok *et al.*, 1972b; Healy, 1974; Anspaugh and Phelps, 1974; Engelman and Sehmel, 1976). The interest stems from the difference in the resuspendible nature of plutonium associated with different particle sizes. When resuspended particles are inhaled, the depositional percentage of the particles in the lungs is a function of size and increases with decreasing size (Task Group, 1966). The following sections consider the different size associations of plutonium, with emphasis on the smaller sizes, and presents an initial attempt to develop an index for comparing the relative hazard of the four contaminated sites.

SOIL PLUTONIUM INDEX

Since the soil serves as a reservoir for resuspendible plutonium, the necessity for considering both soil and plutonium properties is evident in assessing any contaminated site. However, it should be remembered that many factors enter into the resuspension of soil into the atmosphere including climatologic, meteorologic, topographic, and soil factors. In this paper, the plutonium-soil particle size association, the lung depositional potential, and the resuspendible fractions are considered as modifying the potential severity index. Other soil properties are important in resuspension and must be considered before any final judgment is rendered in assessing the severity of the contamination. Chepil (1945a) lists properties of structure, specific gravity, and

Table 4. Particle Size Distribution (by Weight) With (A) and Without (B) Ultrasonic Treatment and Plutonium Distribution by Size in a Soil From Rocky Flats (5-10 cm Depth)

Soil Range (µm)	Size Distribution (%) A	Size Distribution (%) B	Activity (dpm/g) A	Activity (dpm/g) B	Activity (%) A	Activity (%) B
2000 - 850	15.1	18.6	13	27	0.5	1.0
850 - 250	22.2	25.5	99	140	3.6	5.0
250 - 125	8.5	11.3	132	331	4.8	11.9
125 - 53	13.4	11.3	332	372	12.0	13.4
53 - 20	11.3	12.0	394	429	14.2	15.4
20 - 5	9.8	11.3	634	796	22.9	28.7
5 - 2	4.2	2.7	387	263	14.0	9.5
< 2	12.5	4.2	782	419	28.2	15.1
Total	97.0	96.6	2773	2777	100.2	100.0

moisture content as important soil factors; and cover, roughness, obstructions, temperature, and topographic features as important ground factors in soil resuspension. The structure of the soil in turn is affected by organic matter, lime content, and texture.

Although from the standpoint of inhalation the fine sizes (generally limited to the less than 7 μm diameter at 1 g/cm^3 density) are considered most hazardous, the coarser soil particles cannot be ignored in resuspension. Chepil (1945a) reported that the resuspension of fine dust in an air current is mainly the result of movement of larger grains in saltation. Saltation occurs mainly in grain size of 100 to 500 μm (Chepil, 1945b). He also found that single grains or aggregates greater than 3 mm in diameter were not moved by wind velocity of 30 miles per hour at 6-inch height; and that in soils containing a range of particle sizes, the surface became covered with a bed of aggregates too coarse to be moved by the wind as the finer sizes eroded. At the Nevada Test Site (NTS), the occurrence of gravels (2 mm to 3 inches in diameter) is common as a surface cover; and they serve to stabilize the surface from wind erosion. If any decision is made to remove the contaminated soil such as at NTS, considerations must be given to minimize the erosion of the new surface which has yet to become stabilized. At Rocky Flats (RF) in Colorado, the soil surface is covered with varying amounts of grassy vegetation. The resuspension potential of the soil is influenced by the "protection" afforded the surface (Chepil, 1945a). These examples are cited to emphasize the importance of those factors which were not considered in developing the index. On the other hand, when reliable resuspension factors are determined for the contaminated soils from the different sites, the index may be used to evaluate the potential hazard by way of the inhalation pathway.

Three considerations were included in developing the soil plutonium factor. The first considers the concentration of plutonium relative to the soil particle size concentration; the second considers the depositional character of the soil-associated plutonium in the pulmonary compartment of the respiratory tract; and the third factors the fraction of the total soil plutonium in the resuspendible size fractions. Two soil properties associated with plutonium and which have not been factored into the index should be mentioned since they can greatly modify the final index. One is the erodibility of a soil which is dependent on several properties including soil structure, specific gravity, and moisture content. The other is the potential solubility of the plutonium associated with the soil particles. Inclusion of these properties into the derivation of the index must await results of further research.

The derived soil plutonium index (SI) is the product of the soil activity factor (SA), lung depositional factor (LD), and the resuspendible activity factor (RA). Thus one can define it as:

$$SI = SA \times LD \times RA$$

In the following sections, each of the factors is discussed and examples of the factors derived for samples from the four sites are given. These four sites include the Nevada Test Site (NTS), Rocky Flats (RF), Mound Laboratory (ML), and the Oak Ridge National Laboratory (ORNL). The examples given should be considered as preliminary since the number of samples investigated from each site was limited.

Soil Activity Factor. Soil particles which undergo resuspension are those less than 100 μm in diameter. Particles greater than 100 μm move by saltation and creep; Chepil (1945b) reports that particles between 100 to 500 μm move mainly by saltation. Of the particles less than 100 μm in diameter, a further distinction is made for particles less than 10 - 7 μm mass mean aerodynamic diameter (mmad) (Volchok *et al.*, 1972a). These latter sizes are based on the aerodynamic diameter which is based on particle density of 1 g/cm^3. In terms of silicate mineral density, the diameters would correspond to 6 - 4 μm. Particles less than 10 - 7 mass mean aerodynamics diameter (mmad) are commonly referred to as respirable particles. This delineation was made to segregate that portion of the particles which deposits in the nonciliated part of the lung (Volchok *et al.*, 1972a).

As discussed above, several investigators have made soil particle size segregations and determined the plutonium in the different size fractions (Tamura, 1976b; Muller and Sprugel, 1977; Bernhardt, 1976). In terms of potential resuspension, the higher the plutonium activity in size fraction relative to the fractional mass of the soil particles, the higher the activity of the particles when resuspended. For example, a soil with 1% of the soil mass in the less than 2 μm size and 10% of the activity associated with it will have a higher activity per unit mass in the air than a soil with 1% mass of < 2 μm particles containing 1% of the total soil activity. Conversely, a soil with 10% of the soil mass in the < 2 μm fraction and 1% of the activity will have less activity in the air when equally resuspended than a soil with 10% soil mass and 10% of the activity. To obtain the relative activity as distributed in the soil sizes, the activity fraction is divided by the mass fraction and identified as the soil activity factor.

The different particle size ranges of the resuspendible fractions and the fractions of the activity associated with the respective size fractions are given in Table 5. The size ranges are derived in terms of the silicate density for the 5 - 2 μm and the less than 2 μm sizes. Since the sieve used to segregate the particles had 125 μm openings, the upper limit is approximately 25 μm larger than 100 μm normally considered.

The ratio of the activity to the mass is defined as the soil activity factor (Table 5). The sum of the soil activity factor ranges from 3.14 for the sample from Oak Ridge National Laboratory (ORNL) to 7.27 for Rocky Flats (RF). The Nevada Test Site (NTS) sample and the Mound Laboratory (ML) sample have values of 4.27 and 4.62, respectively. Note, however, that the ML sample is assumed to have all particles less than 125 μm. Data from Muller and Sprugel (1977) reported particle sizes up to 45 μm, and the larger particles were reported as being greater than 45 μm. If the soil activity factor for ML is limited to particles between 4-45 μm, the factor is 0.64 for that size range.

Another interesting observation is the distribution of plutonium in the less than 2 μm fraction of NTS and RF. Although only 3% of the activity is in this fraction of the NTS sample and 28% in the RF sample; the soil activity factor is only 3 times larger in the RF sample rather than 10 times based only on activity distribution. Thus if a particle in this size range is resuspended, then the particle from RF will have 3 times the activity as the same size particle from NTS. If the soil activity factor alone is considered, then the

Table 5. Soil Activity Factor Calculated From Soil Mass (% by Weight) and Plutonium Activity (% of Total Activity) for Resuspendible Fractions

Soil (μm)	Soil Fraction	Activity Fraction	Soil Activity Factor
Nevada Test Site - Area 13			
< 2	0.04	0.03	0.75
2 - 5	0.03	0.04	1.33
5 - 125	0.43	0.92	2.19
	0.50	0.99	4.27
Rocky Flats			
< 2	0.12	0.28	2.33
2 - 5	0.04	0.14	3.50
5 - 125	0.34	0.49	1.44
	0.50	0.91	7.27
Mound Laboratory*			
< 2	0.19	0.45	2.42
2 - 4	0.09	0.14	1.56
4 - 125**	0.72	0.40	0.56
	1.00	1.00	4.54
Oak Ridge National Laboratory			
< 2	0.29	0.40	1.38
2 - 5	0.10	0.09	0.90
5 - 125	0.59	0.51	0.86
	0.98	1.00	3.14

*Data from Muller and Sprugel, 1977.

**Assumes particles greater than 45 μm to be less than 125 μm.

ORNL sample appears to have the least potential hazard and RF the highest potential hazard.

Lung Depositional Factor. The next consideration which must be taken into account is the inhalation and depositional character of the potential resuspendible particles. In modifying the size associated plutonium in the respiratory tract, the model deposition in the pulmonary region as defined by the Task Group on Lung Dynamics (1966) was considered. Although the nasopharynx, trachea, and the bronchial tree do filter resuspended particles, the rapid clearance of the particles reduces the potential hazard in these regions (Task Group, 1966). On the other hand, several models as reported by Volchok (1972a) ignore the particles larger than 10 - 7 μm (mmad); the task group model includes the deposition in the pulmonary region of particles up to 100 μm (mmad).

To modify the soil activity factor, the values selected for depositional fraction in the pulmonary region and the soil activity factors were multiplied. The depositional fractions used in these calculations were 0.40 for the less than 2 μm size, 0.12 for the 5 - 2 μm size, and 0.03 for the 125 - 5 μm size. The value of 0.03 was derived by determining the mean particle size of the three fractions (125 - 53; 53 - 20; and 20 - 5 μm sizes); the mass median aerodynamic diameter was 55 μm which correspond to 3% deposition (Task Group, 1966 (Fig. 14)). The value of 0.12 corresponds to 3.5 μm mass diameter or approximately 5 μm aerodynamic diameter. The value of 0.40 corresponds to the deposition of particles with 0.2 μm aerodynamic diameter. Although the value is arbitrary, changing the size to 1 μm or to 0.1 μm aerodynamic diameter changed the difference in the modified soil activity factor of the two extreme values (NTS and RF) by about 10%.

In Table 6, the modified soil activity factors of the three resuspendible sizes are shown. The major effect of the depositional modifier is to minimize the importance of the plutonium associated with the coarsest size. Thus the RF, ML, and ORNL samples show a 6 to 7 fold reduction in the contribution of the 125 - 5 μm size to the modified index; and the NTS sample show a 4 fold reduction. These reductions are compensated for by an increase in contribution to the modified factor by the finest size fraction.

Although the overall numerical value of the modified soil activity factor is decreased as compared with the soil activity factor, the factors are on a relative scale and therefore should not affect the intercomparisons of the different sites.

Resuspendible Activity Factor. The final factor to be considered in developing the overall soil is the amount of the total soil plutonium index activity in the resuspendible fraction. The soil activity factor considered the relative activity on the resuspendible sizes and the lung depositional factor took into account the depositional character of the particle sizes. It is also necessary to account for the amount of the total soil activity which is found in the resuspendible fraction of soil. Thus, the factor for a soil with 100% of the plutonium associated with the resuspendible size needs no further modification; whereas for a soil with only 50% of the activity in the resuspendible size, less plutonium is potentially susceptible to resuspension. Hence, the resuspendible activity factor is simply the fraction of the total activity in the

Table 6. Plutonium Index Derived From the Pulmonary Deposition and Resuspendible Fraction

Size (μm)	Soil Activity Factor	Lung Depositional Factor	Modified Soil Activity Factor	Resuspendible Activity Factor	Soil Plutonium Index
Nevada Test Site - Area 13					
< 2	0.75	0.40	0.30		
2 - 5	1.33	0.12	0.16		
5 - 125	2.19	0.03	0.07		
	4.27		0.53	0.99	0.52
Rocky Flats					
< 2	2.33	0.40	0.93		
2 - 5	3.50	0.12	0.42		
5 - 125	1.44	0.03	0.04		
	7.27		1.39	0.91	1.26
Mound Laboratory					
< 2	2.42	0.40	0.97		
2 - 5	1.56	0.12	0.19		
5 - 125	0.56	0.03	0.02		
	4.54		0.18	1.00	1.18
Oak Ridge National Laboratory					
< 2	1.38	0.40	0.55		
2 - 5	0.90	0.12	0.11		
5 - 125	0.86	0.03	0.03		
	3.14		0.69	1.00	0.69

resuspendible size fractions. These fractions are shown in Table 6. The only significant change is noted in the Rocky Flats sample which showed 91% of the total activity in the resuspendible size fraction.

Although most of the activity in these samples were found in the resuspendible size ranges, it should be noted that the size and activity distributions were determined using water as a suspending agent (i.e., wet sieving and water sedimentation). The resuspension of particles using air forces and media may be different from the water media used here. As noted earlier, Little *et al.* (1973) reported only 3.7 and 11.2% of the particles to be less than 100 μm in two soils from Rocky Flats and 11 and 37% of the activity in these fractions, respectively. He used mechanical sieving without water. In the sample reported for Rocky Flats in this paper, 91% of the activity was in the less than 125 μm sizes. The segregation not only used water but the sample (as with all others) was ultrasonified for five minutes. Without ultrasonic treatment, 82% of the activity was in less than 125 μm size fractions of Rocky Flats (Table 4).

The choice of pretreatment to segregate the soil particles and to determine the plutonium size association should be given serious consideration. The technique applied should not only consider the actual state of association but should be relatively simple without exposing the experimenter to undue hazard.

Potential Application of the Soil Plutonium Index. The soil indices derived in Table 6 represent the relative potential hazard of the soil in the inhalation pathway. To show how to apply the values, one can consider the standard reported for the state of Colorado (Johnson *et al.*, 1976) of 2 dpm/g of soil. The value for Rocky Flats in Table 6 is 1.26. The value for NTS is 0.52, which means that the soil at NTS potentially provides less lung activity than the soil at Rocky Flats. Thus, to conform to the 2 dpm/g for Colorado, the soil in NTS could have 5 dpm/g (1.26/0.52 x 2 = 5). Similar calculations for Mound Laboratory and ORNL give 2 and 3 dpm/g, respectively.

Healy (1974) suggested 500 dpm/g for bare soil without reference to particular site. If one uses the value as a general guide, the allowable concentration for NTS is 960 dpm/g (1/0.52 x 500 = 960). Similarly for RF, ML, and ORNL, the values are 395, 425, and 615 dpm/g, respectively.

The soil plutonium index derived is not a soil activity standard. It is primarily aimed at evaluating the inhalation hazard of the contamination as it exists in the soil. The attraction of setting a soil level standard is the ease of obtaining samples and performing analyses. Any standard must consider the pathways to man and biota. Although the soil plutonium index considered the inhalation pathway, similar considerations must be given to factors involved in uptake by plants and eventually ingestion by man and biota. The intent of this derivation is to provide a basis for arriving at an assessment of the contamination in soil. It should be obvious that the data presented are limited; the final soil factors should consider data from many more samples, providing appropriate statistical significance to the soil factors.

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DISTRIBUTION OF PLUTONIUM AND AMERICIUM AT A FORMER LOS ALAMOS WASTE DISPOSAL SITE

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ABSTRACT

A thirty year old liquid waste disposal site at Los Alamos Scientific Laboratory has been cored and a portion of each six inch increment analyzed for plutonium and americium. The site received unprocessed low level waste solutions from 1945 to 1952, and treated waste effluents from 1952 to 1967. The actinide content of each core increment was determined by low energy photon spectroscopy utilizing the characteristic L x-rays of each nuclide. The data obtained are presented as a histogram and compared to gross activity measurements made at the site.

A laboratory simulation of the site was also prepared using simulated waste solutions and a specimen of disposal site's geologic material, tuff, a welded volcanic ash. This simulation was scaled and proportioned according to archival records of the original site. The results of this experiment are compared with measurements from the actual core sample.

INTRODUCTION

One of the most serious problems confronting the nuclear energy industry is the long term isolation of radioactive wastes. Transuranic elements contained in such waste contribute significantly to the hazard, both in toxicity and persistence. Whatever method is finally chosen to contain these wastes, access of the actinides to the environment must be restricted. Deep terrestrial burial with adequate buffer zones appear to be one of the most promising approaches. Details of such a method have not been worked out but it seems reasonable to assume high level process waste solutions will be calcined and converted to ceramics or glasses before loading into retrievable containers. These containers would then be inserted into selected geologic formations at appropriate depths (∿1000 meters) in isolated geographic locations. In spite of these precautions, one must consider the rock or other geological stratum in which the waste filled containers have been buried as the ultimate barrier. Corrosion and leakage may violate the integrity of any container over the 10^5 year time span required for safekeeping.

An understanding of the migrational behavior of actinides in geological material is therefore extremely important. Since there are no operating disposal sites as described, the information obtained up to now has been the result of supposition, theoretical models and in our own case small scale laboratory models. Important as these results are, they do not eliminate the need of a large scale pilot plant on-site experiment.

However, several sites exist which have received low level waste. One such has been selected for this study; a liquid waste disposal trench at Los Alamos Scientific Laboratory that was used for waste disposal starting in 1945. This site does not match the description of an ideal disposal method since liquid was pumped directly into a trench excavated in porous rock and allowed to percolate downward. However, it can be considered as a rapid acceleration of what could occur in a deep repository in which water intruded.

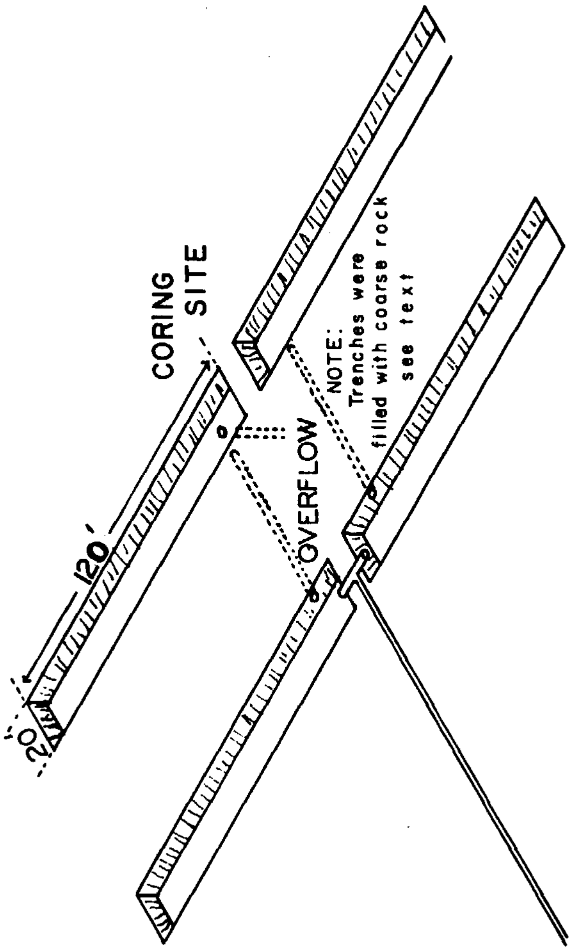
MATERIALS AND METHODS

The original disposal site (fig. 1) as constructed in 1945 at Los Alamos consisted of four trenches of trapezoidal cross section 120 feet long, 20 feet wide and 6 feet deep. The four trenches were arranged in a square configuration with the first two connected to the process plant by a tee pipe. The primary trenches were connected by overflow pipes to the other two trenches. It was near the discharge end of one of the overflow pipes that coring was done for the samples reported on here. Each trench was filled with coarse rock debris, grading from large boulders in the bottom to sand at the surface.

Unprocessed waste solutions were discharged to these trenches from 1945 to 1952. Estimates of the chemical composition and plutonium content are based primarily on data collected towards the end of that period. Beginning in 1952, through about 1967, the trenches received varying annual volumes of effluent from a liquid waste treatment plant; no unprocessed wastes were discharged after 1952. The estimated chemical composition and plutonium content of the waste discharged to the trenches is indicated in Table 1.

Some additional fill was added to the area during the late 1960s and 1970s, raising the ground surface to a meter or so above the top of the trenches. Precipitation falling on the area is predominantly reevaporated to the air, or

FIGURE 1



LIQUID WASTE DISPOSAL SITE

TABLE 1
COMPOSITION OF EFFLUENTS

Date	Volume (gallons)	Chemical Composition	Plutonium Concentration
1945 - 1950	1.4×10^7	160 ppm F^- 13 ppm NH_4^+	60 c/m/ml
1951 - 1952	1.0×10^4	"Conc" ammonium citrate 200 ppm F^-	7×10^3 c/m/ml
1953 - 1967	4.0×10^6	0.1M $NaNO_3$	Trace

produces immediate surface runoff. A small fraction, perhaps less than 5%, of the annual precipitation of 460 mm infiltrates the waste trenches. The actual distribution of the waste solutions within the four trenches is not known. For modeling purposes, it was assumed that the trench from which the core was collected received ten percent of the total discharged.

Cores were taken at six inch intervals to a depth of 20 feet. Drilling was accomplished without water for lubrication or cooling. A portion of each core was packaged in small polyethylene bottles and shipped to Argonne National Laboratory. Each sample was ground in a new mortar which was discarded after use to prevent cross contamination, dried four hours at 110° C., and loaded into a sample holder. Sample holders were constructed from 2 mm thick cardboard squares 4 cm on a side with a 5/8 inch hole in the center. A single strip of cellophane tape on each side formed a thin, wafer-shaped cavity which contained 0.4 ml of powdered material. The average weight of each sample was 0.35 g. Several analytical methods for determining actinide content were considered before choosing low energy photon spectroscopy. Chemical separation followed by alpha proportional counting was rejected because of difficulty in dissolving samples to insure complete recovery of the actinides. This method, however, offers the greatest sensitivity. Neutron activation analysis was also tried and, although promising, offers no greater sensitivity than our chosen method.

The spectrometer consists of a planar lithium drifted germanium detector 2 cm^2 by 5 mm thick. Its shallow design results in a low background from high energy photons. Its small diameter is a compromise between high resolution, 350 eV in the L x-ray region, and good geometric efficiency, about 8%. The entire detector-cryostat assembly was contained in a large 6 inch thick steel background shield with a graded x-ray liner. The resulting background in the L x-ray region is 0.27 c/m. A multichannel analyzer coupled to a computer controlled real time data acquisition system completed the spectrometer.

In operation, all samples were counted for a minimum of 15 hours. Data processing was accomplished by initial subtraction of a normalized background count from a sample spectrum. ^{241}Am then was determined by integrating under the 60 keV photopeak and that value used to normalize a standard ^{241}Am spectrum which in turn was subtracted from the sample spectrum. This step eliminated the Am x-ray contribution from that portion of the spectrum due to the Pu x-ray. ^{239}Pu was then determined by summing the integrals of the three L x-rays. The spectrometer efficiency was calibrated using synthetic standards prepared in an identical manner to the samples.

A laboratory simulation of this site was prepared primarily as a test of the validity of our modeling techniques (fig. 2). A specimen of Los Alamos tuff taken near the site was cut into a cylindrical plug 18 x 77 mm. The curved surface was waxed to restrict lateral flow of liquid. The cylinder of rock was attached to a glass tube which served as a reservoir for liquids which were to percolate the rock. Waste solutions were synthesized from ^{241}Am and ^{237}Pu tracer; the latter in place of ^{239}Pu to facilitate low level determination.

Solution volumes and concentrations were proportional (as closely as possible from available Los Alamos records) to permit an arbitrary scale of 2.2 ft/cm. The synthetic waste solutions were then delivered to the test specimen in the same sequence as at the site at a controlled rate of 2 ml/hour using a metering pump. Disposal at the site, however, was not continuous but rather sporadic. Since no record of the cycle is available, no attempt was made to reproduce it. We fully realize the complications such simplification of the model would produce but felt the chance to compare even a crude model to an actual site justified this approach. The effluent of the column was collected and prepared for analysis. At the conclusion of the experiment, the specimen was removed from the apparatus, dried, cut into 0.5 cm sections, and also analyzed by gamma spectrometry to determine the distribution of the actinides in the rock.

RESULTS

Examination of the actinide content in the core (fig. 3) showed that detectable amounts of these nuclides did indeed migrate downward under the prevailing conditions to 20 feet below the bottom of the trench. Elevated concentrations of both nuclides are found at less than 1 foot and at 13 feet. The bulk of the americium is centered around the 9 foot mark. Noteworthy is the abrupt diminution of plutonium and americium below 13 feet; in fact, only 0.2% of the detected plutonium wss below that level. On the other hand 45% of the plutonium was 1 foot or less from the bottom of the trench.

The distribution of actinides in the simulated model (fig. 4) is in excellent agreement with our earlier modeling work using tracers and simulated rain (Fried *et al.*, 1976). Characteristic of these data are strong retention of actinides at the near surface with exponential tailing downward. In addition, plutonium exhibits a second concentration band beyond this tail. The percent of plutonium in this band is a function of the prior chemical treatment of the tracer material. The chemistry of plutonium is complex, existing in a IV, V, and a VI oxidation state. Polymeric states are also known at high pH. The strongly retained large peak at the surface is most likely Pu(IV). The faster moving species is not so readily characterized. The amount of fast moving plutonium in the model here depicted is much smaller than earlier models, reflecting, perhaps, the lower pH of the feed solution. It appears in the histogram as a slight rise at 6 cm with a rapid drop. An examination of the eluent (fig. 5) from the model shows no readily discernable pattern. It contains no americium and only 5% of the plutonium. It does indicate the mobilizing influence of the various waste solutions upon plutonium. Most interesting is the rapid drop in the elution of plutonium once water has rinsed the waste solutions from the rock and is the only eluting agent. This agrees with previous work which shows that distribution of plutonium is dependent upon the conditions of loading on the rock such as chemical composition and rate. Once plutonium is adsorbed by the rock, the actual water induced migration is extremely slow.

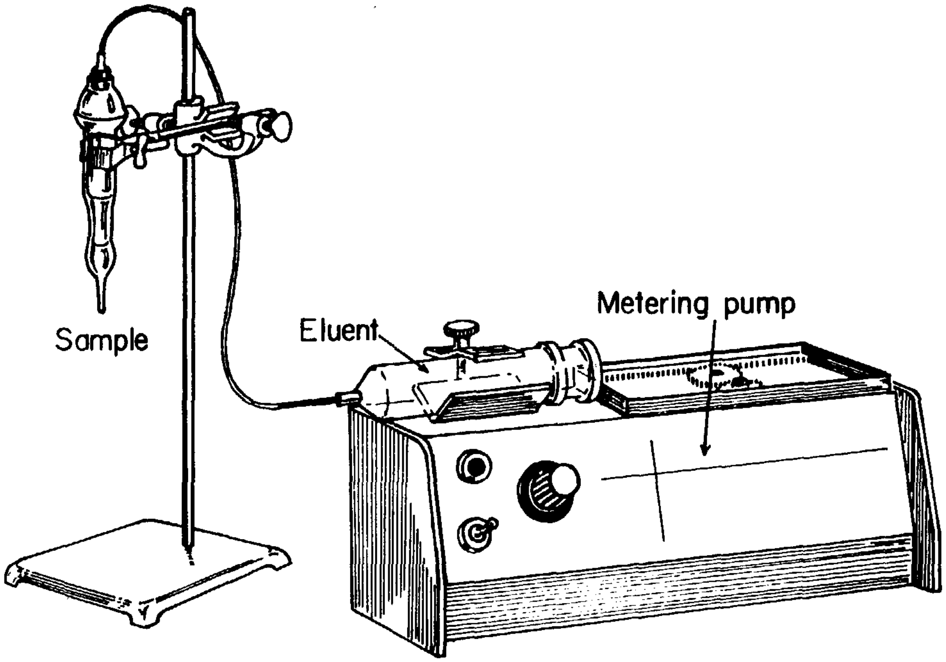
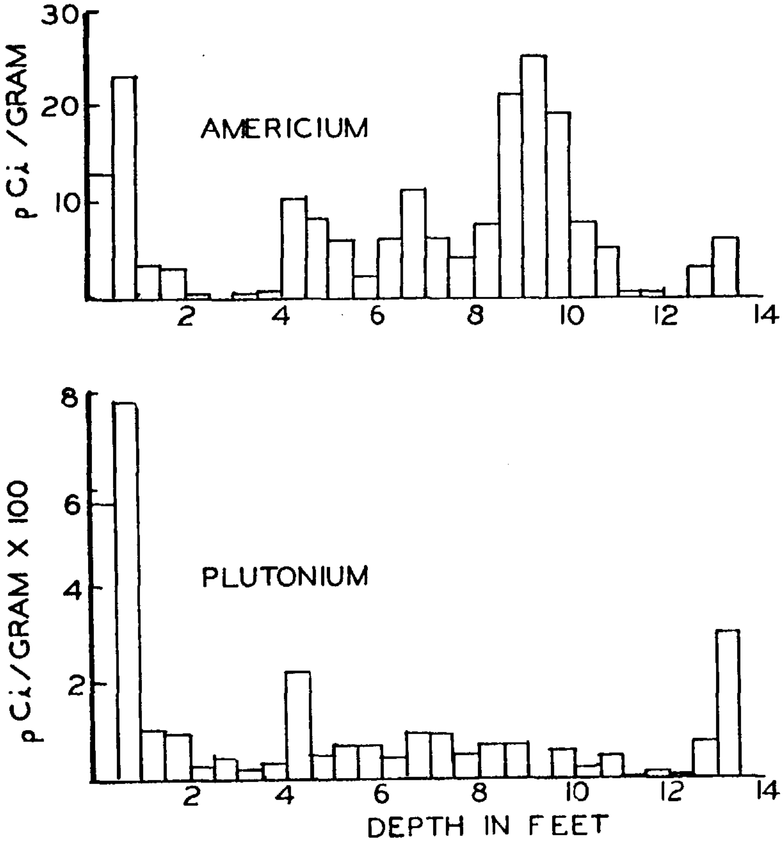
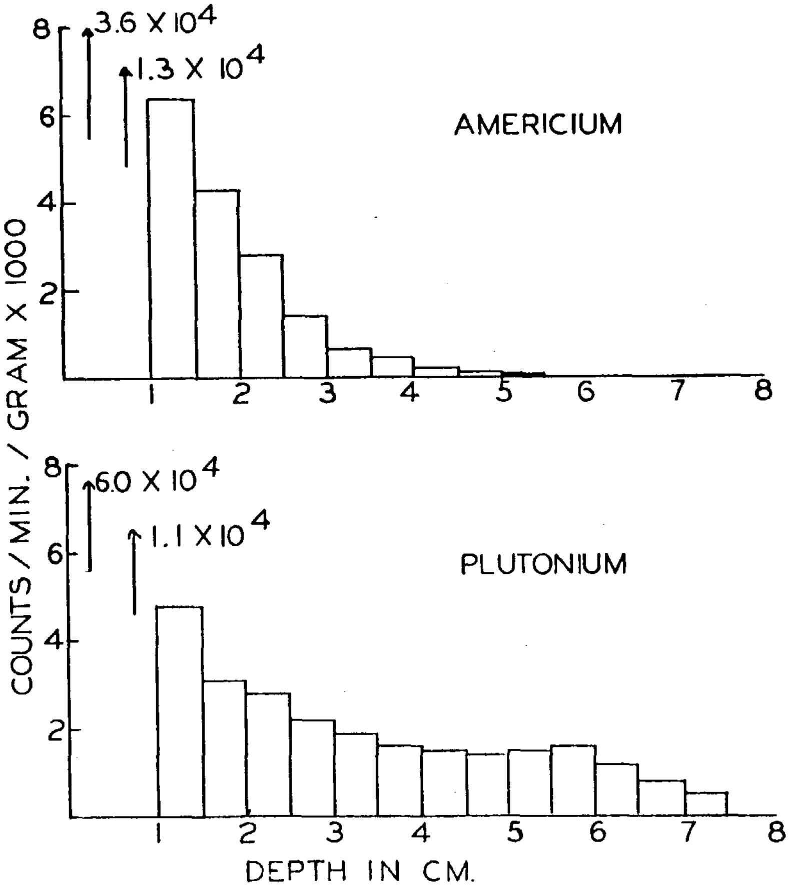


FIGURE 2. Experimental modeling apparatus.



ACTINIDE DISTRIBUTION AT DISPOSAL SITE

Figure 3



ACTINIDE DISTRIBUTION IN LAB MODEL

Figure 4

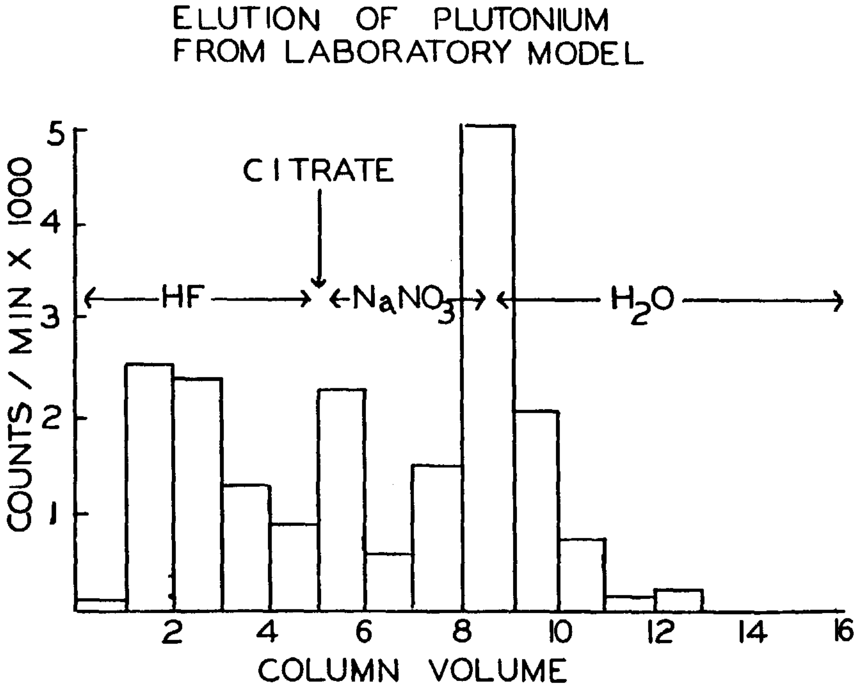


Figure 5

CONCLUSIONS

In order to compare results of the model to results from the actual site, certain reservations must be understood. The model constructed in the laboratory was primarily a chemical model. It could not reproduce geologic conditions of the site, the most prominent of which are fractures in the rock which can greatly accelerate or retard the flow of solutions through a stratum depending upon whether they are void or clay filled. Such geologic features contribute to the character of the site data and, in fact, may outweigh the chemical effects. For example, examination of the core indicated the presence of a clay-filled fracture at approximately 14 feet. This fracture appeared to be restricting downward flow of moisture, possibly accounting for the reduced activity below that depth in the core. Nevertheless, the model correctly predicts the large surface concentration for plutonium as well as a maximum depth of penetration of 14 feet. The latter is based on the modeling scale of 2.2 ft/cm. Our predictions for americium did not fare nearly so well. Instead of the near surface peak predicted by the model, americium was found at the site in a diffuse band near 9 feet. The danger of inferring too much from a single coring experiment as obvious. Therefore, more corings and evaluations are planned and under way with the hope of resolving the discrepancies and confirming the predictions.

ACKNOWLEDGMENT

This work was supported by the U.S. Energy Research and Development Administration, Division of Nuclear Fuel Cycle and Production.

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THE RELATIONSHIP OF MICROBIAL PROCESSES TO THE FATE OF TRANSURANIC ELEMENTS IN SOIL

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ABSTRACT

This review considers the influence of soil physiochemical and microbial processes on the long-term solubility, form, and plant availability of plutonium and other transuranic elements important in the nuclear fuel cycle. Emphasis is placed on delineation of the relationships between soil chemical and microbial processes and the role of soil microorganisms in effecting solubilization and transformation of elements considered largely insoluble in soils strictly on the basis of their inorganic chemical characteristics.

Soluble, diffusible Pu in soils (usually less than 0.1% of total) appears to be largely present as particulates of hydrated oxide, but several lines of evidence suggest that microorganisms may influence the solubility of Pu and that the nonparticulate plant-available fraction is stabilized in solution by inorganic or organic ligands of limited concentration in soil. The role of soil microorganisms in influencing the solubility, form and plant-availability of the transuranics is discussed on the basis of the (1) known chemistry of organic ligands in soils, (2) effects on the soil microflora, and (3) principal microbial transformation mechanisms, including direct alteration (valence state, alkylation), indirect alteration (metabolite interactions, influence on the physiochemical environment), and cycling processes (biological uptake and release on decomposition of tissues).

The toxicity of Pu to microorganisms depends on Pu solubility in soil. However, soil microorganisms are generally resistant to Pu, with toxicity apparently due to radiation rather than chemical effects. Highly resistant bacteria, fungi, and actinomycetes have been isolated from soil, and these organisms have been shown to be capable of transporting Pu into the cell and altering its form in the cell and in solution. The resulting soluble Pu complexes tend to be of higher molecular weight than simple complexes (Pu-DTPA) and negatively charged. The form of Pu, although not well-defined, is dependent upon organism type, carbon source, and time of Pu exposure during growth. These factors, in turn, are a function of Pu source, soil properties, and soil environmental conditions. Knowledge of the relative influence of these factors serves as a valuable basis for predicting the long-term behavior of Pu and other transuranic elements in the terrestrial environment.

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INTRODUCTION

In the terrestrial environment, soils represent the principal repository of the transuranic elements over geologic time. The major factor governing transuranic availability to plants in soils will be the solubility of the transuranics associated with the solid phase, since in order for root uptake to occur a soluble species must exist adjacent to the root membrane for some finite period. The form of this soluble species will have a strong influence on its stability in soil solution and on the rate and extent of uptake, and perhaps, mobility and toxicity in the plant. Furthermore, it is the solubility and form of the element which largely governs mobility in soil. Thus, any assessment of the long-term behavior of the transuranics in the terrestrial environment must be based on determination of the factors influencing solubility and form of soluble species in soil. These factors are illustrated in Fig. 1 and include the concentration and chemical form of the element entering soil, soil properties, as these influence the elemental distribution between the solid and liquid phase, and soil processes, such as microbial activity, as these influence the kinetics of sorption reactions, transuranic concentration, and the form of soluble and insoluble chemical species.

The transuranic elements of principal importance in the nuclear fuel cycle (Pu, Am, Cm, Np) may enter the soil through several avenues (Vaughan *et al.*, 1976) including (1) fallout from atmospheric testing, (2) possible airborne particulates passing a filter and liquid effluents, both during reprocessing of spent fuels and fuel fabrication and (3) leaching from waste storage facilities. The major sources of the transuranics may be classified according to expected initial solubility in soil (Fig. 2). Particulate oxides of the transuranics initially may be expected to be largely insoluble in the soil solution. Ultimately, solubility is expected to be a function of the composition, configuration and equivalent diameter of the particle as well as soil properties and processes. Oxide particles of the highest specific activity and containing the highest concentrations of impurities in the crystal lattice may exhibit greatest solubility. The combination of configuration and equivalent diameter as reflected in surface area exposed to solution will be the other main factor governing oxide solubility. Once solubilized, the transuranic elements will be subject to the chemical reactions governing soluble salts. Hydrolyzable transuranics entering the soil in acid solutions sufficiently concentrated to maintain soluble ions may be expected to be rapidly insolubilized due to hydrolysis on dilution and subsequent precipitation on particle surfaces. These include Pu(III, IV, and VI), Am (III, IV), Cm (III, IV), and Np (III, IV, VI). Conversely, transuranics not subject to marked hydrolysis may be initially more soluble. These include Pu (V) and Np (V). Immobilization of these chemical species (PuO_2^+ or NpO_2^+) may occur through cation exchange reactions with particulate surfaces. Complicating this situation, disproportionation and complexation reactions may occur concurrently.

Transuranics entering the soil as stable organocomplexes as might occur in the vicinity of a spent fuel separation facility may be initially highly soluble (Wildung and Garland, 1975). The duration of solubility and mobility in the soil will be a function of the stability of the complex to

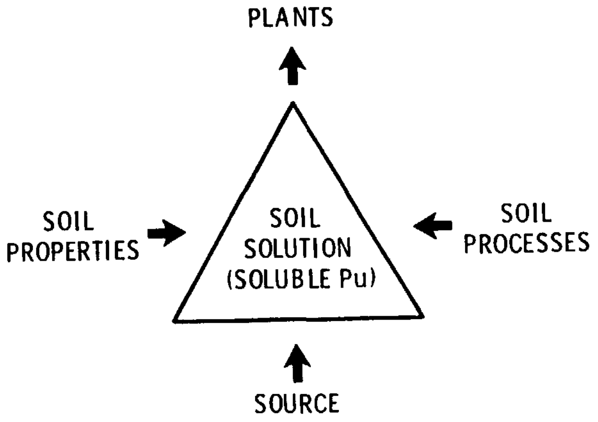

Fig. 1. Factors influencing transuranic behavior in the terrestrial environment.

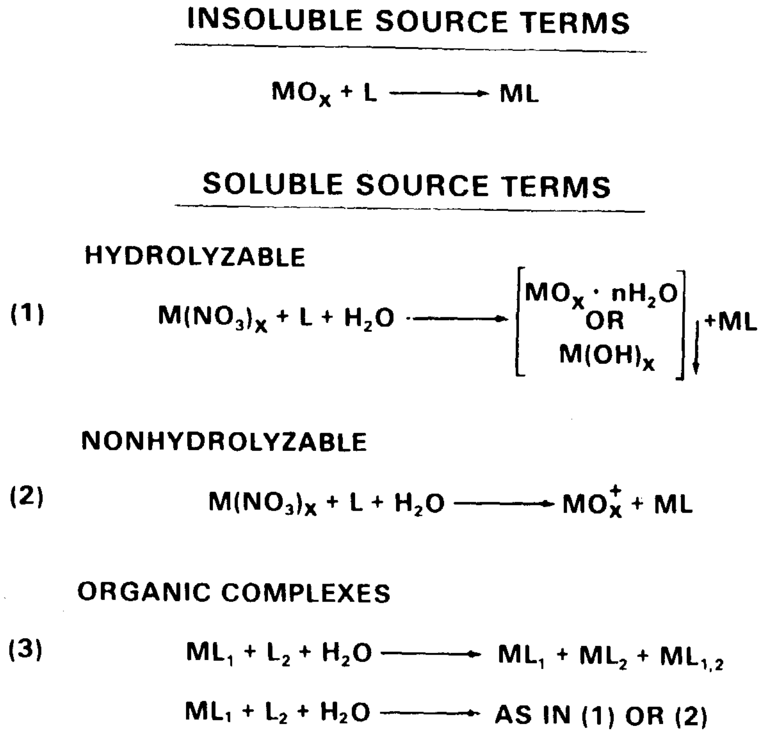

Fig. 2. Influence of source term on transuranic behavior in soil. Transuranic elements are represented by M, and L represents inorganic and organic ligands capable of reacting with transuranic elements and forming soluble or insoluble products.

substitution by major competing ions, such as Ca and H (Lahav and Hochberg, 1976; Lindsay, 1972; Norvell, 1972) and the stability of the organic ligand to microbial decomposition (Wildung and Garland, 1975). The disruption of the complex may lead to marked reduction in transuranic solubility through hydrolysis and precipitation reactions as described for acid solutions on dilution. A portion of the ion released may react with other, perhaps more stable, ligands in soil. The mobility of the intact complexes, in turn, will be principally a function of their chemical and microbiological stability and the charge on the complex which will govern the degree of sorption on soil particulates.

Further generalizations of transuranic behavior on the basis of source terms are complicated by the overwhelming importance of soil properties and processes in influencing transuranic behavior on a regional and local basis. This review will consider, in detail, the influence of soil properties and abiotic and biotic processes on the long-term solubility of the transuranics entering soils. Principal emphasis will be directed toward the role of soil microorganisms in this phenomenon. Microorganisms, in intimate association with soil particles, are known to play an important role in effecting solubilization of elements considered insoluble in soils strictly on the basis of their inorganic chemistry. To date, studies of the microbiology of the transuranic elements have been limited principally to Pu. This review will emphasize Pu, but, where possible, the available information will be used as a framework for broader discussions encompassing the long term behavior of other transuranic elements.

TRANSURANIC CHEMISTRY IN SOIL

Plutonium

The principal chemical reactions likely influencing Pu behavior in soil are summarized in Fig. 3. Plutonium ions may commonly exist in aqueous solution in valence states III, IV, V(PuO_2^+) and VI(PuO_2^{+2}). Other valence states are known (II, VII) and predicted (VIII) but these occur under unique conditions (Cleveland, 1970). Disproportionation reactions are common, and due to kinetic factors, Pu is unique among the chemical elements in that it may simultaneously exist in all of the common valence states. The tendency of Pu to hydrolyze in aqueous solutions of low acidity follows the order $Pu^{+4} > PuO_2^{+2} > Pu^{+3} > PuO_2^+$ (Cleveland, 1970). Hydrolysis, which occurs in a stepwise fashion, is likely the major mechanism whereby Pu is insolublized in the environment. At high (g/l) Pu concentrations, hydrolysis of Pu^{+4}, may lead to the formation of a collodial Pu polymer. At these concentrations, the polymer is characterized by a distinct absorption spectrum. Although the polymer has not been fully characterized, it is generally thought to be an intermediate hydrolysis product of Pu^{+4} containing oxide or hydroxide bridges, with an absorption spectrum different than $Pu(OH)_4$. However, studies by Lloyd and Haire (1973) have indicated that the polymer may be aggregates of small, discrete, amorphous or crystalline, primary particles of 5 to 20 Å in diameter. It is of interest that x-ray diffraction patterns of the polymeric Pu and that of $Pu(OH)_4$ (Ockenden and Welch, 1956) both showed a pattern characteristic of the cubic PuO_2 lattice, suggesting that the polymer and the hydroxide of Pu^{+4} may be hydrated PuO_2 with differences occurring in primary particle size and crystallinity (Lloyd and

- FOUR OXIDATION STATES

  $Pu^{+3}, Pu^{+4}, PuO_2^{+1}, PuO_2^{+2}\ (Pu^{+2}, Pu^{+7})$

- DISPROPORTIONATION

  $Pu^{+4} + PuO_2^{+1} \rightleftharpoons Pu^{+3} + PuO_2^{+2}$

- HYDROLYSIS

  $Pu^{+4} + 4H_2O \rightleftharpoons Pu(OH)_4 + 4H^+\ (K_{sp} \approx 10^{-56})$

- COMPLEX FORMATION

  $2Pu^{+4} + 3\ DTPA \rightleftharpoons Pu_2DTPA_3 \quad \log K \approx 26$

Fig. 3. Chemical reactions influencing plutonium behavior in soil.

Haire, 1973). The formation of the hydrated PuO_2 is likely directly related to Pu^{+4} concentration and inversely related to the acid concentration.

Plutonium also tends to form many complexes with a range of stabilities. The strongest complexes are generally formed by reaction of organic ligands with Pu^{+4}. However, many inorganic complexes and organic complexes of all valences may be stable under appropriate conditions. The presence of organic ligands in soils likely influences the equilibrium and concentration form of Pu in solution through complexation and subsequent inhibition of hydrolysis, polymerization, or disproportionation. It is these reactions in various highly complex combinations resulting from differences in source term, soil properties and processes that govern Pu solubility in soil and availability to plants.

Soil chemical reactions are important in governing the behavior of the various forms of Pu entering soil. Initially soluble forms entering soil have the potential for undergoing a range of chemical transformations (Fig. 1-3). Insoluble Pu, such as high-fired oxide, entering soil likely will be solubilized with time, provided soluble, stable complexes are formed (Fig. 3). However, regardless of the form of Pu entering soil, its ultimate solubility will be controlled by its aqueous chemistry and by soil factors. Soil physiochemical properties may be expected to have complex, interdependent effects on Pu solubility. The long-term behavior of Pu in soil will be a function of the kinetics of these reactions.

On the basis of research with other trace metals, recently summarized by Keeney and Wildung (1976), and limited information on the transuranic elements it may be concluded that the soil physicochemical parameters most important in influencing the solubility of the transuranics include: (1) solution composition, Eh and pH, (2) type and density of charge on soil colloids, and (3) reactive surface area. These phenomena will in turn be dependent upon soil properties, including particle size distribution, organic matter content, particle mineralogy, degree of aeration and microbial activity. The delineation of the influence of these factors on Pu solubility is difficult due to the complex chemistry of Pu.

Perhaps the simplest approach to the study of the chemistry of Pu in soil is to direct initial attention to the factors influencing its solubility in soil. However, it is difficult to define Pu solubility in soil because solubility will depend upon the method of measurement and because solubility must be arbitrarily evaluated due to the sorption of Pu on submicron clay particles or to the formation of submicron particles of hydrated Pu oxide which are difficult to centrifuge and may pass membrane filters. These effects may be illustrated by comparison of the differences in the solubility of Pu in soils [100 days after amendment as $Pu(NO_3)_4$] as determined by water extraction and subsequent membrane filtration using membranes of different average pore sizes (Table 1). The major fraction of the Pu added was sorbed on the soil, as a maximum of 10% of the extracted Pu passed through the 5 μ membrane. Successive filtration through membranes with decreasing pore size resulted in decreases in Pu concentration in the filtrate. Thus, Pu in the aqueous extract appeared to be in a wide range of particle sizes. Although membranes with pore sizes of 0.45 μ are commonly used to separate soluble

Table 1. Solubilities of Plutonium in Water Extracts of a Ritzville Silt Loam as Determined by Filtration with Membranes of Different Pore Sizes (Garland and Wildung, 1977).

Membrane Pore Size, μm	Plutonium Solubility, pg/g*
5	60,000
0.45	20,000
0.01	4,000
0.0015	1,000
0.0012	300
0.0010	50

*Plutonium added at a level of 620,000 pg/g of soil.

from particulate matter, it is evident that Pu in these filtrates may be in collodial forms. The Pu in the 0.0010 μ filtrate appeared soluble, was stable in solution, and approximated the quantity of Pu taken up by plants (Wildung and Garland, 1974). Of the soluble Pu forms likely to enter soils (previous section), Pu(NO$_3$)$_4$ and Pu-DTPA likely represent, in their respective chemistries, the range in soil behavior likely to occur. The water solubility (< 0.01 μ) of ^{238}Pu and ^{239}Pu amended to a Ritzville silt loam (organic C content 0.7%, pH 6.2) in the Pu(NO$_3$)$_4$ and Pu-DTPA forms differs markedly (Wildung and Garland, 1975). The DTPA complexes of both isotopes were water-soluble in soil and appeared to be stable over the first 40 days of incubation (Fig. 4). After 7 days of incubation, the ^{238}Pu-DTPA appeared to be slightly less soluble than the ^{239}Pu-DTPA. After 95 days of incubation, both isotopes, initially added as the complex, appeared to decrease in solubility; perhaps as a result of microbial degradation of the organic moiety and the development of new chemical equilibria.

Equilibrium concentrations of soluble Pu added as the nitrate were not obtained until 7-10 days. The solubility of ^{238}Pu and ^{239}Pu added to the soil as nitrates was much lower than the DTPA complexes, likely reflecting hydrolysis to the largely insoluble hydrated oxide. It is clear that organic ligands may have a pronounced effect on Pu solubility in soil. The rate of decrease in solubility of each isotope added as the nitrate was similar. However, in contrast to the slightly lower solubility of the ^{238}Pu-DTPA compared to the ^{239}Pu-DTPA, ^{238}Pu added as the nitrate was a consistent factor of 2-3 times more soluble than ^{239}Pu initially added as the nitrate. This difference probably resulted from the formation of larger hydrated oxide particles at the higher Pu concentration (^{239}Pu), but it may also have reflected the presence of soil components such as organic ligands, which stabilized Pu in solution but were present in limited concentrations and became important only at lower Pu concentrations.

The water solubility of ^{238}Pu, when incorporated in relatively large Pu oxide particles (>1 μ), would be expected to be greater than the solubility of ^{239}Pu oxide particles of similar size due to crystal damage and radiolysis arising from the greater specific activity of the ^{238}Pu (an approximate factor of 270). However, the behavior of the two isotopes in soil on solubilization of the oxide might be expected to follow a course similar to that exhibited by the nitrates (Fig. 4).

Equilibrium solubility after 6 days of incubation (Garland *et al.*, 1976) of Pu, added as Pu(NO$_3$)$_4$, in soils of different properties occurred after approximately 20 hours (Fig. 5). The quantities of Pu soluble at equilibrium in water and 0.01 M CaCl$_2$ differed with soil type. In the CaCl$_2$ solution, solubility was lowest in the Muscatine soil which exhibited higher silt and clay content than the other soils. Importantly, at equilibrium there was more Pu extracted by water than 0.01 M CaCl$_2$ in the Muscatine soil. The Hesson and Ritzville soils did not exhibit this property. This may be related to a difference in the dispersibility of fine colloids in this soil and/or the presence of higher concentrations of stabilizing ligands. However, the lack of a proportional dilution effect (not shown in Fig. 5) in the water extractability of Pu at lower solution to soil ratios in this soil as compared to the Ritzville and Hesson soils, provided

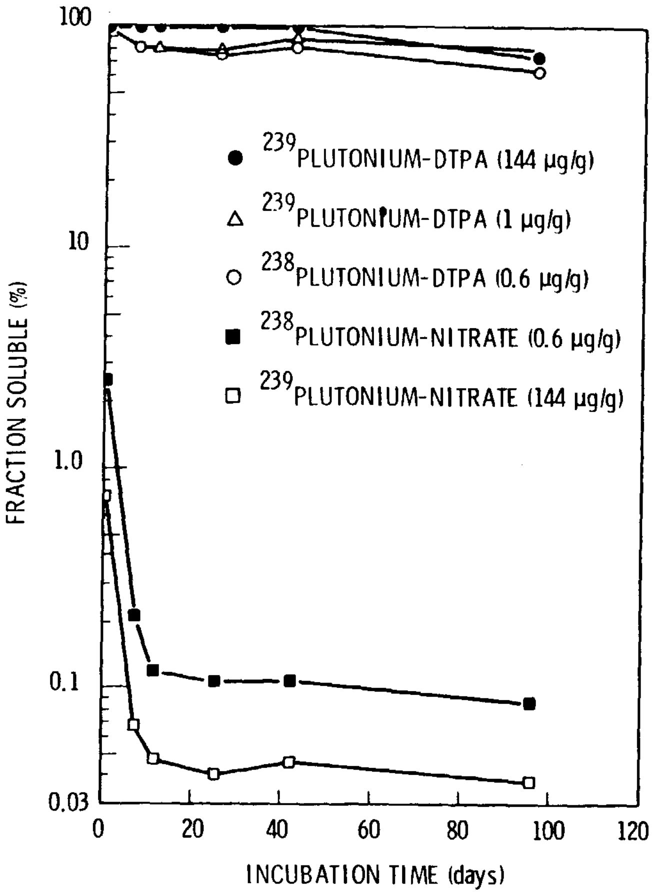

Fig. 4. Solubility of plutonium with time after addition to soil as the nitrate and the DTPA complex (Wildung and Garland, 1975).

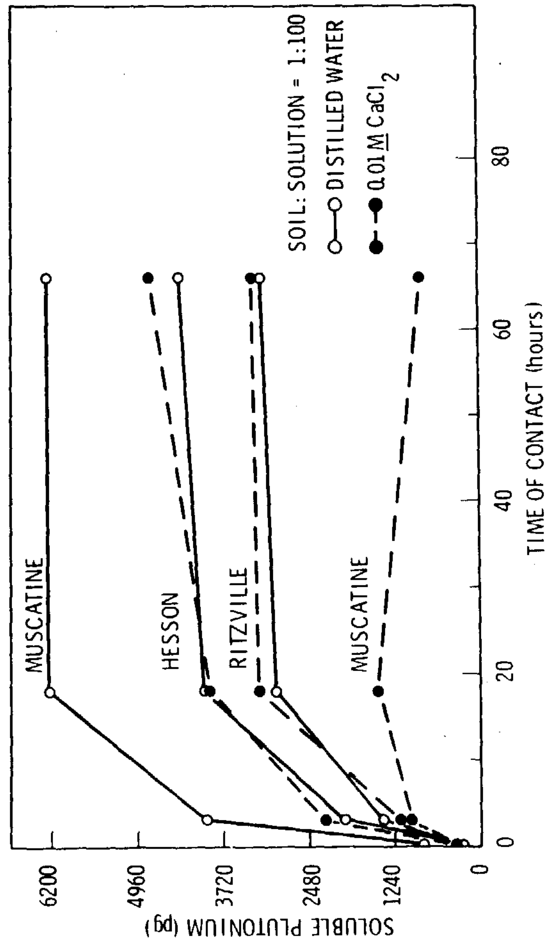

Fig. 5. Quantity of soluble plutonium removed from three soils by 0.01 M CaCl$_2$ (Garland *et al.*, 1976).

presumptive evidence for the presence of a dispersible ligand in higher concentration in the Muscatine soil.

Applying diffusion principles to characterization of mobile Pu species in soils, Garland and Wildung (1977) estimated the concentrations and molecular weight of mobile Pu in five surface soils representing a range in particle size distributions, pH (4.4-6.2) organic C (0.7 to 12.5%) and cation exchange capacities (14 to 45 meq/100 g). The diffusion coefficients calculated for the most mobile species in the five soils varied from 1.5 to 3.0 x 10^{-6} cm^2/sec (Table 2). Estimated concentrations and molecular weights of the most mobile Pu components in the five soils ranged from 9 to 55 pg/g and from 5000 to 21,000, respectively. Thus, estimated concentrations of the most mobile Pu species were of the same order of magnitude as those observed by water extraction and subsequent ultrafiltration through the 0.0010 μ membrane (Table 1). This membrane retained Pu-DTPA (molecular weight 1700). Hypothetical globular peptides of molecular weights less than 500 would pass through this membrane. However, if the molecule was a hydrated PuO_2 sphere of similar dimensions, it would have a molecular weight between 10,000 and 25,000 approximating the molecular weights of the most mobile Pu species as determined from diffusion coefficients. This fraction, therefore, likely consisted of small particles of $Pu(OH)_4$ or hydrated oxide.

The estimated diffusion coefficients for the least mobile Pu components ranged from 2.3 - 3.1 x 10^{-7} cm^2/sec with corresponding soil concentrations of 150 - 1200 pg/g (Table 2). This concentration of Pu in soil approximated the quantity of water-soluble Pu passing the 0.0015 μ ultrafiltration membrane (Table 1). Hypothetical globular proteins in this size range would have average molecular weights < 10,000. Particles of $Pu(OH)_4$ or hydrated oxides would have molecular weights of 200,000 to 500,000. Estimated molecular weights for these least mobile species calculated from diffusion coefficients were between 600,000 and 900,000. Thus, it would appear, as in the case of the most mobile species, that the least mobile species of Pu were particulate $Pu(OH)_4$ or hydrated oxides.

The comparison of filtration and diffusion data indicates that the mobile Pu in incubated soils was in the form of hydrated oxide of hydroxide in a continuum of sizes. If it can be assumed that Pu in particulate form was not available to plants, it is possible that the small fraction of Pu taken up by plants was present in soil as reaction or dissolution products with insufficient stability and/or concentration to be detected by the methods employed. Insight into this possibility was not provided by comparison of Pu behavior in different soils, as might be expected, because the estimated concentrations and molecular weights of the mobile species were not related to the soil properties measured.

Several conclusions may be drawn from studies of the soil chemistry of Pu which have important implications in terms of the potential role of the soil microbiota in influencing Pu behavior in soil. Definition of Pu solubility by filtration or diffusion alone is complicated by Pu chemistry, but, in conjunction, the measurements suggest that mobile Pu is largely particulate. However, a fraction of the mobile Pu is available to plants.

Table 2. Estimated Concentrations and Molecular Weights of Mobile Plutonium in Soils from Measured Diffusion Coefficients (Garland and Wildung, 1977).

Treatment	Most Mobile Species			Least Mobile Species		
	Diffusion Coefficient x 10-6	Molecular Weight, g/mole	Soil Concentration, pg/g*	Diffusion Coefficient x 10^{-7}	Molecular Weight, g/mole	Soil Concentration, pg/g*
Control						
$Pu_2(DTPA)_3$	5.8	1,700		53	1,700	
Soils						
Ritzville	3.0	5,000	24	2.3	0.9 x 10^6	150
Quillayute	2.5	7,200	47	2.7	0.7 x 10^6	1,200
Hesson	2.4	8,100	9	2.7	0.6 x 10^6	330
Salkum	1.5	21,000	55	2.3	0.8 x 10^6	340
Muscateen	1.9	13,000	36	3.1	0.5 x 10^6	170

*Plutonium added at a level of 620,000 pg/g of soil.

This material is obviously not particulate, but is present in insufficient concentration for characterization using current methods. The question remains--what is the form of the small quantity of Pu available to plants? This information is essential to understanding the mechanisms whereby Pu may be resupplied to solution from the solid phase in a range of soils and to predictions of the long-term availability of Pu to plants. From investigations of Pu valence state in a neutral, 0.0004 M NH_4HCO_3 solution equilibrated with PuO_2 microspheres and in burial ground leachates, Bondietti and Reynolds (1976) concluded that Pu(VI) may be stable in significant quantities in solution and suggested that monomeric Pu(VI) and its complexes may be important in Pu mobilization. In the present studies, several lines of evidence were presented suggesting that that Pu ions are stabilized in soil solution by inorganic or organic ligands for subsequent uptake by the plant. Furthermore, equilibration of weathered Pu-contaminated soil with chelating resins has been shown (Bondietti *et al.*, 1975) to result in significant desorption of Pu from the solid phase. It is known that organic ligands result in the most stable Pu complexes. Soluble organic ligands in soil are generally derived from microbial processes. The organic complexation reactions and the microbiological factors potentially influencing Pu behavior in soil will be discussed in subsequent sections.

Other Transuranic Elements

Principal isotopes of other transuranics of concern in the nuclear fuel cycle include ^{241}Am, ^{243}Am, ^{242}Cm, ^{243}Cm, ^{244}Cm, and ^{237}Np. Although detailed studies of the interaction of these elements with soils are lacking, some information has become available in recent years. Furthermore, the aqueous chemistries of these elements have been fairly well established (Katz and Seaborg, 1957). The most stable ions of Am and Cm in aqueous solutions are the cations (III); Np is most stable as the oxyion (NpO_2^+). Disproportionation is not common with these elements. Thus, major differences in their environmental behavior as compared to Pu, would be expected. Hydrolysis reactions may still be a primary factor governing the environmental behavior of Am and Cm but greater mobility and plant availability in soils might be predicted on the basis of greater solubility of the hydroxides in comparison to $Pu(OH)_4$. The Np oxycation should not be subject to significant hydrolysis at environmental pH values (Burney and Harbour, 1974). The environmental behavior of Np has been least studied of the transuranics but because of its chemical characteristics it may be the most available to the biota. A comparison of Pu, Am, and Np sorption in several soils (Routson *et al.*, 1975) indicated sorption in the order Pu > Am > Np. The chemistry of Cm should be very similar to Am if present at equal mass concentrations. The potential for complexation of these elements will be discussed in a subsequent section.

Organic Complexation Reactions

Research to date on the chemistry of the transuranic elements in soil has pointed to the importance of understanding transuranic organic complexation reactions in soil; particularly in surface soils and aquatic sediments where organic matter content is generally highest or in subsoils where the

transuranics may be dispersed in conjunction with synthetic complexing agents. Very little information is available concerning the interaction of the transuranic elements with the soil organic fraction. However, despite the difficulties in characterization of soil organic complexes, much is known both theoretically and experimentally regarding the interactions of metals with functional groups of soil organic matter (Keeney and Wildung, 1976). Much of this information concerns micronutrients of greatest agronomic importance (B, Co, Cu, Fe, Mn, Mo, Se, Zn) and this research has been the subject of a number of excellent reviews over the last two decades (Mitchell, 1964, 1972; Mortensen, 1963; Hodgson, 1963; Stevenson & Ardakani, 1972). In general, earlier studies emphasized metal interactions with intact soil or with the higher molecular weight humic components of soil whereas more recent studies have emphasized the more soluble components of soil.

It is practical to categorize metal complexes in soil in terms of their solubility since, in general, it is this factor, as previously noted, which most influences their mobility and plant availability. Three principal categories have been proposed (Hodgson, 1963) although the complexity of the soil system results in considerable overlap between categories. These include (1) the relatively high molecular-weight humic substances containing condensed aromatic nuclei in complex polymers derived from secondary syntheses which have a high affinity for metals but are largely insoluble in soil, (2) low molecular weight organic acids and bases, classified as nonhumic substances, and derived largely from microbial cells and metabolism which demonstrate relatively high solubility in association with metals, and (3) soluble ligands which are precipitated on reaction with metals.

Humic Substances. Humic substances are generally divided into three categories based on their solubilities (Felbeck, 1965). The humin (alkali and acid insoluble) fraction is soluble only under drastic conditions and is apparently of the highest molecular weight. The humate (alkali soluble, acid insoluble) and fulvate (alkali and acid soluble) fractions of soil may constitute up to 90% of the soil organic fraction (Kononova, 1966). The humates and fulvates are characterized, in part, by a high charge density due to acidic functional groups (Stevenson & Ardakani, 1972; Felbeck, 1965). This property leads to a high degree of reactivity and these materials exhibit a strong pH-dependent affinity for cations in solution and are likely strongly bound to soil minerals and other organic constituents in soil (Greenland, 1965). The acidic functional groups consist principally (in general order of acidity) of carboxyl, hydroxyl (phenolic, alcoholic), enolic, and carbonyl groups (Broadbent and Bradford, 1952; Felbeck, 1965; Schnitzer *et al.*, 1959). Total acidity has been estimated to range between 500 to 900 and 900 to 1,400 meq/100 g for humic acids and fulvic acids, respectively (Stevenson & Butler, 1969). The acidic H of humic acids was differentiated by Thompson (1965) into three groups at 100 to 200, 500 to 700, and 1,000 to 1,200 meq/100 g using nonaqueous titration methods. Basic functional groups, likely amides and heterocyclic nitrogen compounds (Bremner, 1965), probably also contribute to retention of metals but are of much less importance than acidic groups at most soil pH values.

In batch equilibration studies (Bondietti, 1974), Ca-saturated humates removed greater than 94% of the Pu(IV) from pH 6.5 aqueous solutions (compositions not given). It is unclear whether the humates represented a surface for precipitation of hydrolyzed species or were involved in complexation of Pu. However, in studies of Pu desorption from humates and reference clays, citrate removed 10-30% of sorbed Pu from the clays but less than 1% from the humic acids. Ligands forming stronger complexes with Pu (DTPA and EDTA) were required to remove significant quantities (up to 30%) of the Pu from the humate complex.

Although humic and fulvic acids likely account for most of the metal immobilization attributed to the soil organic matter, (e.g., Hodgson, 1963; Stevenson & Ardakani, 1972), they have the potential for formation of soluble complexes with metals, particularly in dilute solutions. Small quantities of metal fulvates, thought to be of lower molecular weight than the humates, may be present in soil solution. A nondialyzable material with infrared absorption spectra and elemental analyses similar to fulvic acids was isolated from a dilute salt (0.01 M KBr) extract of a mineral soil by Geering and Hodgson (1969). The material exhibited a concentration equivalent to 2.5% of a dialyzable fraction but was more effective in complexing Cu and Zn.

Nonhumic Substances With Potential For Metal Complexation. Lower molecular weight biochemicals of recent origin have been implicated in metal complexation and solubilization in soil. These materials represent (1) components of living cells of microorganisms and plant roots and their exudates and (2) the entire spectrum of degradation products which ultimately serve as the building units of the soil humic fraction. The quantity and composition of these materials will vary with soil, vegetation, and environmental conditions (Alexander, 1961, 1971). Readily decomposable wastes disposed to soil under conditions appropriate for microbial growth may, for example, result in immediate and marked increases in organic materials identified in category (1) and longer term increases of materials in category (2). Conversely, toxic materials may have the opposite effects. The specific compounds produced will be dependent upon the properties of the waste and soil environmental conditions after disposal (Routson and Wildung, 1969).

Although the concentration of the transuranics and other metals soluble in the soil solution or in mild extractants is low, often near minimum detectable levels, the major portion of Cu and Zn have been shown to be associated with low molecular weight components. Most of the titratable acidity of this fraction has been attributed (Geering & Hodgson, 1969) to aliphatic acids (< pH 7.0) and amino acids (> pH 7.0).

The production, distribution, and action of organic acids in soil has been reviewed by Stevenson (1967). A wide range of organic acids are produced by microorganisms known to be present in soil. These include (1) simple acids such as acetic, propionic, and butyric, produced in largest quantities by bacteria under anaerobic conditions, (2) carboxylic acids derived from monosaccharides, such as gluconic, glucuronic, and α-ketogluconic acids produced by both bacteria and fungi, (3) products of the citric acid cycle such as succinic, fumaric, malic, and citric acid, which are common metabolic excretory products of fungi, and (4) aromatic acids such as p-hydroxybenzoic, vanillic, and syringic acids thought to be fungal decomposition products of plant lignins. A variety of organic acids have also been reported in root exudates.

The other important group of compounds identified in significant quantities in the soil solution by Geering and Hodgson (1969) which may be expected to exhibit strong affinity for metals are the amino acids. The qualitative and quantitative aspects of amino acids and other nitrogenous components in soils have been reviewed by Bremner (1967). It was concluded that soil acid hydrolysates do not differ greatly in amino acid composition but quantitative differences may occur with differences in soil, climatic, and cultural practices. A number of acidic and basic amino acids have been reported in soil. However, it appears that the major portion of amino acid-N that is present in hydrolysates is in (1) the neutral amino acids glycine, alanine, serine, threonine, valine, leucine, isoleucine and proline, (2) the acidic amino acids, aspartic acid and glutamic acid, and (3) the basic amino acids, lysine and arginine. Most of the amino acids detected in soil hydrolysates have also been shown to exist free in small quantities in soils with levels seldom exceeding 2 μg/g. In the soil solution (Geering & Hodgson, 1969), neutral amino acids also appeared to predominate. Basic amino acids were not detected although two acidic amino acids (aspartic and glutamic acids) were present.

Stevenson and Ardakani (1972) concluded that organic acids and amino acids, while present only in small quantities in soil, were present in sufficient quantities in water-soluble forms to play a significant role in solubilization of mineral matter in soil. Small quantities of a number of other complexing agents, such as nucleotide phosphates, polyphenols, phytic acid, porphyrins, and auxins, also exist in soil (pertinent references have been summarized by Mortensen, 1963). However, it is unclear at present, whether these materials would be present in sufficient quantities in the soil solution under most soil conditions to affect transuranic solubility over the long-term.

MICROBIAL TRANSFORMATION OF THE TRANSURANIC ELEMENTS IN SOIL

Potential Mechanisms of Transformation

From the results of limited studies of soil chemistry, microbiology and plant availability of transuranics in soils, and by inference from studies of complexation of other trace metals in soils (as discussed above) it may be concluded that the soil microflora will play a significant role in transformations governing the form, and ultimately, the long-term solubility and behavior of transuranic elements in soil. There are four general mechanisms whereby microorganisms may alter the form of trace metals in soil (Alexander, 1961; Wood, 1974). These include (1) indirect mechanisms resulting from metal interactions with microbial metabolites, or changes in pH and Eh, (2) direct transformations such as alkylation and alteration of the valence state through microbial oxidation (use of the metal as an energy source) or microbial reduction (use of the metal as an electron acceptor in the absence of oxygen), (3) immobilization by incorporation into microbial tissues, and (4) release of metals on decomposition of organic residues.

All of these mechanisms may be operational in transformations of transuranic elements in soils. However, on the basis of present knowledge, it is not possible to draw conclusions as to their relative importance in affecting the long-term behavior of the transuranic elements. Since there is a paucity of information available, rather than discuss these mechanisms individually they will be addressed around a framework of current information, limited principally to Pu.

Microbial Alteration of Solubility in Soil

To provide a preliminary assessment of the potential for microbial alteration of Pu solubility in soil under aerobic conditions, Wildung *et al.* (1973, 1974) measured soil respiration rate (an index of soil microbial activity), microbial types and numbers and Pu water solubility in sterile (gamma irradiation) and nonsterile soils, which contained 10 μCi Pu/g of soil [added as $Pu(NO_3)_4$]. Carbon dioxide evolution was utilized as a measure of soil respiration rate. To measure Pu solubility, the soil was subsampled at intervals during incubation over a 30 day period, and the subsamples (1g) suspended in 1 liter of distilled water. After a 4 hr equilibration period, an aliquot of the soil suspension was filtered through 5, 0.45 and 0.01 μ millipore filters. The Pu in the 0.5 and 0.01 μ filtrates was designated water soluble although it was recognized that Pu likely was present as fine colloids (previous section).

Changes in the soil respiration rate and Pu solubility during the initial 30 day incubation are shown in Fig. 6. The concentration of Pu in the 0.45 μ filtrate during the incubation period ranged from approximately 0.5 to 1.5% of the Pu initially applied. Solubility was initially higher in the sterile soil than in the nonsterile soil but was relatively constant with time in the sterile soil. The initial increase in solubility in the sterile soil was anticipated in view of the known increases in soluble organic matter which result from gamma irradiation of soil.

The Pu solubility (< 0.45 μ) in the nonsterile soil, while initially lower, increased by a factor of 3 with incubation time to 14 days and remained significantly higher than the sterile soil during the incubation period. This increase generally followed the accumulative CO_2 curve, and maximum solubility occurred at the end of logarithmic growth for all classes of organisms. The concentration of Pu in the 0.01 μ filtrate, which represented a Pu level less than 0.2% of that applied, did not change significantly with treatment.

In an ancillary experiment, incubation was continued for 65 days until the CO_2 evolution rate reached a constant level. The Pu-containing sterile soil was then inoculated with the Pu-treated nonsterile soil and the respiration rate and solubility of Pu in the inoculated soil measured for a period of 30 days. When the sterile soils were inoculated with nonsterile soil, CO_2 evolution increased at a much more rapid rate without a lag phase, and this was accompanied by a factor of 2 increase in water solubility (< 0.45 μ)

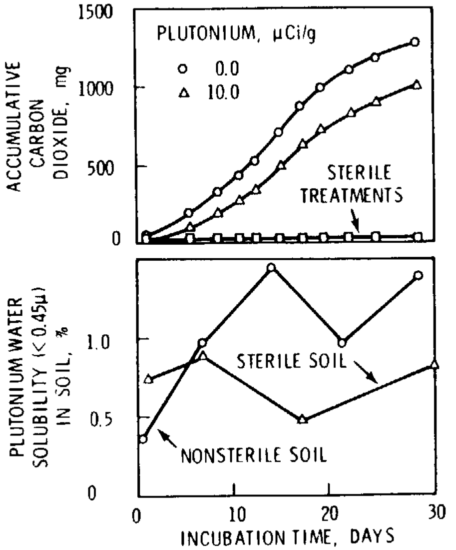

Fig. 6. Changes in soil respiration rate and solubility of applied plutonium with time of soil incubation (Wildung *et al.*, 1973, 1974).

of Pu after only 4 days of incubation, suggesting the development of a microbial population particularly capable of alteration of Pu solubility. Again, there was no change in the < 0.01 μ fraction which amounted to approximately 10% of the Pu present in the < 0.45 μ fraction.

At least under the conditions of this study, the evidence strongly suggested that the solubility of Pu in soil was influenced by the activity of the soil microflora. The potential mechanisms effecting the change in solubility include mechanisms (1) and (2) described above, i.e. indirectly through the production of organic acids that may complex Pu or the alteration of the solution pH and/or Eh near the soil colloid (or Pu polymer) without measureable effects on the overall soil pH; or directly through the transformation of Pu. Microbial effects may have occurred at the colloidal level since detectable increases in solubility did not occur in the < 0.01 μ filtrates. This appears to implicate the indirect mechanisms since direct changes generally occur at the molecular level and these would be reflected in changes in Pu concentration in the < 0.01 μ filtrates provided polymerization did not occur during extraction. Of course, a combination of the mechanisms is possible, i.e., alteration of affinity for organic ligands through a change in valence state. If the mechanism of solubilization was indirect, the results might be applicable to other transuranic elements, e.g., from consideration of the aqueous chemistry described in a previous section, a reduction in pH would be expected to increase the solubility of the other transuranic elements as well as Pu.

Increased water solubility of Pu on incubation under optimum conditions for microbial activity may be expected to increase Pu uptake by plants provided the limiting factor is not discrimination at the root membrane. In order to determine if the increased solubility on incubation resulted in increased Pu uptake by plants, the soils, incubated as previously described, were planted to barley and cultured using a split-root technique which allowed measurement of the uptake, sites of deposition and chemical forms of Pu in plant shoots and roots (Wildung and Garland, 1974). The results were compared to the results of similar plant studies in which the soils had not been incubated.

Prior incubation, which in microbial studies was shown to increase the solubility of Pu in soil, increased Pu uptake by shoots compared to the unincubated controls. The effect was greatly accentuated in the case of the soil-free roots and incubation increased the soil to plant concentration ratios by up to 37 times relative to the unincubated control, depending upon Pu soil concentration level. Thus, plant uptake measurements tend to verify measurements of increased Pu solubility in the < 0.45 μ fraction in the incubated soil. However, if, as previously discussed, Pu in particulate form (i.e., > 0.01 μ) was not available to plants, and if increases in the concentration of Pu in the 0.01 to 0.45 μ fraction resulting from microbial activity were not due to procedural artifacts arising from polymerization, the colloidal particles may be serving as a reservoir of plant-available Pu manifested only when the equilibrium concentration of Pu in solution is reduced by plant uptake.

Effect on the Soil Microflora

Soil organisms may be expected to be present at highest levels in the immediate vicinity of soil colloids (Alexander, 1961). From the aqueous chemistry of the transuranics and on the basis of recent information on transuranic chemistry in soil (previous section), the transuranic elements in soil are likely to be associated mainly with colloids. Thus, soil microorganisms may be exposed to relatively high concentrations even when total transuranic soil concentration is low. It is therefore necessary to determine the toxicity of the transuranic elements to soil microorganisms, as microorganisms exhibiting resistance to the chemical effects of the transuranics may have the highest potential for participating in alteration of transuranic form. However, the transuranic series does not contain stable isotopes and organisms chemically resistant to these elements must exhibit a degree of radiation resistance which is dependent, in large part, upon the radiochemistry of the isotope. Resistance to the chemical effects of transuranics may occur by three general mechanisms including (1) inability of the transuranics to produce a toxic effect on cell metabolism at the cytoplasmic or exocytoplasmic levels (2) inability of organisms to transport the transuranics or (3) ability of the organisms to convert transuranics, by the direct and indirect mechanisms discussed in a previous section, to a form that is either less capable of entering the cell or is not toxic to the cell. It is the latter mechanism which is likely most important in alteration of transuranic form in soil.

Effect on Microbial Types, Numbers and Activity. The effect of soil Pu concentration on the soil microflora has been measured as a function of changes in microbial types and numbers and soil respiration rate (Wildung *et al.*, 1973, 1974a). A noncalcareous Ritzville silt loam (pH 6.7) was amended with ^{239}Pu(NO$_3$)$_4$ at levels of 0.05, 0.5 and 10 μCi/g and with starch, N and water to provide optimal microbial activity. Subsamples of soil were periodically removed to determine the changes in types and numbers of soil microflora with time. During this period, soil respiration rate was monitored by continuous collection of soil-evolved CO$_2$.

The growth curve of fungi (Fig. 7) was generally typical of the growth response for other classes of microorganisms. Total microbial numbers were compared at the end of logarithmic growth. The organisms generally reached this stage after 8 to 14 days of incubation. Growth rates were compared over the intervals of maximum microbial growth for each organism at each Pu concentration. The results are summarized in Table 3.

The Pu did not generally affect the rate of growth but decreased the total numbers of all classes of microorganisms at levels as low as 0.05 μCi/g or 7 μg/g. The fungi were the exception, differing from the controls only at a Pu concentration of 10 μCi/g or 144 μg/g. Thus, the Pu did not affect maximum generation rate but rather affected the lag period or onset of the stationary phase, limiting microbial numbers.

The accumulative CO$_2$ curve generally corresponded to the growth curve of the fungi. In the case of the other classes of organisms, maximum logarithmic growth occurred before the rate of CO$_2$ evolution reached minimum

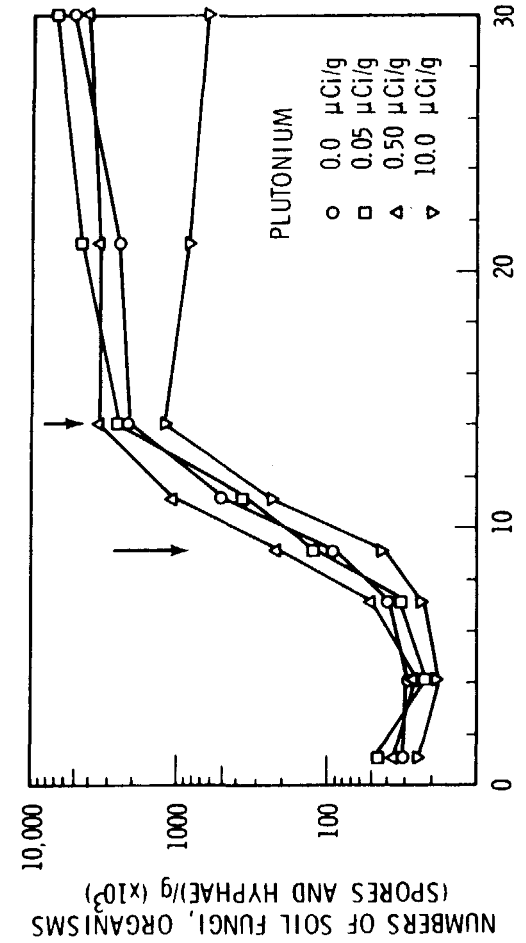

Fig. 7. Influence of plutonium concentration on the growth of fungi in soil. Arrows denote time intervals at which growth rates and total numbers were compared with other microbial types (Table 3).

Table 3. **Summary of the Effects of Plutonium at Several Soil Concentration Levels on the Distribution of Microorganisms in Soil Relative to Controls (Wildung *et al.*, 1973, 1974).**

Microbial Type	Effect ($p < 0.05$)* of Plutonium on Growth Rate at Plutonium Concentrations (μCi/g) of 0.05	0.5	10.0	Total Numbers at Plutonium Concentrations (μCi/g) of 0.05	0.5	10.0
Bacteria						
Aerobic and Microaerophillic						
Nonspore Formers	0	0	0	+	+	+
Spore Formers	0	0	0	+	+	+
Anaerobic and Facultative Anaerobic						
Nonspore Formers	0	+	0	+	+	+
Spore Formers	0	0	0	+	+	+
Fungi	0	0	0	0	0	+
Actinomycetes	0	0	+	+	+	+

*A positive sign denotes a significant effect. A zero indicates that there was no significant effect.

levels. The rate of CO_2 evolution and cumulative CO_2 over the incubation period were significantly reduced only at the 144 μg/g level of Pu amendment, although numbers of all classes of organisms except the fungi were depressed below this level (Table 3). This is in marked contrast to the results of studies with a number of other heavy metals (Drucker *et al.*, 1973) such as Ag and Hg, in which respiration rate was a sensitive measure of metal effect at levels as low as 1 μg/g in soil. Differences in the effects of the metals may be related to differences in soil solubility as well as toxicity, as discussed below. It should also be noted, however, that the effect on respiration rate was dependent upon the magnitude of the soil respiration rate in Pu treated soil relative to untreated controls, which, in turn, was dependent upon the initial level of microorganisms in soil. In soils exhibiting a higher CO_2 evolution rate, the reduction of respiration rate due to Pu amendment was more pronounced.

Studies of the toxicity of other transuranic elements to soil microflora have not been conducted.

Mechanism of Effect. To understand the long-term effects of microorganisms on transuranic form, it is important to distinguish, where possible, chemical and radiation effects of the transuranics on soil microorganisms. Pronounced initial chemical toxicity, as noted above, may result in the development of special pathways of detoxification leading to alteration of transuranic form. The lack of chemical toxicity may imply chemical modifications of the transuranic elements through interaction with cell metabolites. In contrast, radiation resistance is associated with an enhanced ability to repair radiation damage to key macromolecules without development of new biochemical pathways leading to alteration of transuranic form. However, the possibilities for indirect alteration of transuranic form would be higher for a radiation resistant organism than for an organism which did not exhibit either radiation or chemical resistance since, due to competitive advantage, these organisms may be expected to be present in larger numbers in the vicinity of transuranic colloids than less resistant organisms.

The effects of Pu on soil microorganisms may be due largely to radiation damage. Schneiderman *et al.* (1975) measured the effects of Pu form and solubility on soil metabolic activity and on the types, numbers, and resistance of soil fungi and actinomycetes in soil separately amended with ^{239}Pu (1 to 145 μg/g) and ^{238}Pu (0.6 μg/g) in soluble nitrate and DTPA complex forms, and with C, N, and water to provide optimal microbial activity. Subsamples of soil were removed over a 95-day aerobic incubation period to determine changes in numbers of fungi and actinomycetes and relative water solubilities (< 0.01 μ) of the Pu forms. Comparisons of soil fungal numbers in the presence of ^{238}Pu and ^{239}Pu at common radioactivity levels, but at different mass concentrations, indicated that Pu toxicity was due to radiation rather than chemical effects (Fig. 8). Solubility of Pu in soil influenced Pu toxicity to microorganisms with the more soluble Pu-DTPA forms resulting in greatest reductions in numbers. Similar studies have not been conducted with other transuranic elements.

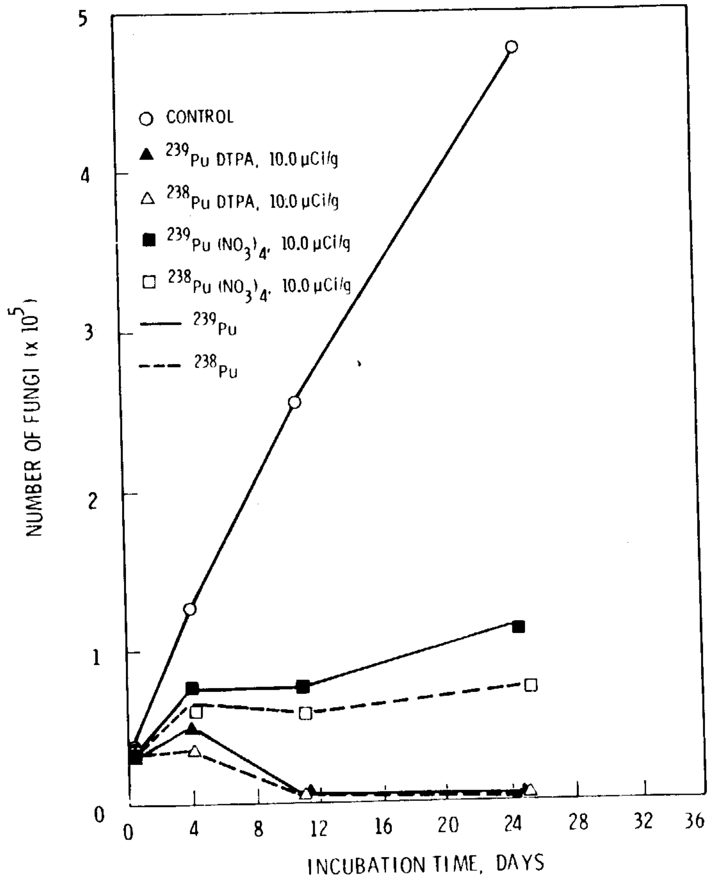

Fig. 8. Effect of different isotopes of plutonium on survival of soil fungi (Schneiderman *et al.*, 1975).

Isolation of Resistant Organisms. The study of complexation of trace metals in soils is extremely difficult due to the complexity of the soil system (Keeney and Wildung, 1976). In fact, although much information is available regarding organic ligands in soil (previous section), an organometal complex has never been isolated intact from soils. A logical approach to the study of microbial transformations of the transuranic elements is to isolate, from soil, resistant organisms most likely to alter transuranic form, study the transformation *in vitro* and validate the results in the soil system using techniques specifically tailored to metabolites identified from the simpler *in vitro* systems.

Application of enrichment techniques to the isolation of Pu resistant fungi, which have been demonstrated (previous section) to be the most resistant class of microorganisms, and actinomycetes from soil using starch as a C-source (Schneiderman *et al.*, 1975) resulted in the isolation of 14 fungal cultures and 13 cultures of actinomycetes distinct in colonial morphology. Of these, 7 of the actinomycetes and 5 of the fungal isolates were capable of growth at 100 μg/ml Pu as the soluble DTPA complex. There appeared to be a succession of actinomycetes types in the soil during incubation as indicated by the different colony morphologies obtained from enrichments after 4 and 25 days incubation. Although this phenomenon may have resulted from changes in the soil arising from the production of metabolites or chemical degradation products, it may also have resulted from a response to the presence of Pu. Only one actinomycete isolate was found which was common to enrichments from both incubation periods and this organism was present at all Pu concentrations in the media. In contrast, the fungal isolates exhibited 6 common morphological types regardless of incubation period.

Subsequent enrichment studies (Pelroy, 1976; personal communication) have resulted in the isolation of 30 distinct cultures of bacteria from soil. Of these 11 were resistant to Pu at concentrations as high as 100 μg/ml. These studies also indicated that C source as well as soil Pu concentration will play a role in determining the types and numbers of Pu resistant microorganisms present in soil, providing presumptive evidence that microbial metabolites, which will differ with C source, may play a role in Pu resistance. This subject will be discussed in the next section. The presence of Pu resistant organisms is apparently related to factors that may be expected to vary with soil type and environmental conditions. Again, similar studies have not been conducted with other transuranic elements.

Microbial Transformations

As previously discussed, there are several means whereby microorganisms may transform trace metals in soil. These may be generalized to (1) direct mechanisms such as alteration in valence state or alkylation (2) indirect mechanisms such as interactions with normal metabolites or microbial alterations of the physicochemical environment and (3) cycling mechanisms such as uptake during cell growth and release on cell decomposition. In the latter case, any combination of indirect and direct methods of alteration may be operational. Although there have been no studies conducted to date which would allow the unequivocal separation of these mechanisms, studies have been conducted which demonstrate the alteration of Pu form *in vitro* by soil microorganisms and provide evidence for both direct and indirect transformation of Pu.

Direct Transformations. The potential for direct transformation of the transuranic elements through alteration of valence state or alkylation is difficult to assess. Although, as previously discussed, the transuranics have the potential for existing in aqueous solution in several valence states, information is not available to assess the role of soil microflora in direct alteration of valence. More information is available regarding the mechanism of metal alkylation.

Alkylation of metals involving the alkyl donor methyl cobalamine and other alkyl cobalamines has been clearly demonstrated for Hg, As, and Pt (Wood *et al.*, 1968; McBride and Wolfe, 1971; Taylor and Hanna, 1977). The methyl derivative of Hg may be present in significant quantities in soils (Beckert *et al.*, 1974). Wood (1974) suggested that methylated derivates of Hg and As are important in governing their behavior in the environment. McBride (1977) also suggested that these reactions occur abiotically. The process of biochemical methylation of metals may be described as an overlap between the chemistry of methyl cobalamine (an intermediate in methane synthesis by anaerobic bacteria and methionine synthesis in aerobic bacteria) and the chemistry of the metals. In the case of the transuranics, particularly Pu, it is the complexity of the aqueous chemistry that has limited research into alkylation phenomena.

It is unknown whether an ionic species of Pu is capable of reacting *in vitro* with an alkyl cobalamine. Further, if a mechanism for biological alkylation of Pu, similar to the Hg, As, Pt alkylation reaction did exist, it would be of importance in influencing environmental behavior only if the alkylated molecule exhibited stability (Wood, 1974), i.e., a half-life in soils and sediments of hours rather than seconds. Considering the coordination chemistry of the actinides, Marks (1976) noted that U- and Th-C linkages are formed in organic solvents and the complexes are relatively stable thermally though highly sensitive to oxygen. Meaningful microbial studies await the development of an understanding of the chemical speciation of transuranics in aqueous solutions at environmental concentration levels.

Indirect Transformations. The potential for indirect transformation of the transuranic elements may be greater than for direct transformation. The potential for Pu interaction with microbial cells and metabolites has been demonstrated and many of the other transuranics form stable complexes with known microbial metabolites.

Beckert and Au (1976) demonstrated the uptake of ^{238}Pu, applied initially to malt agar in nitrate, citrate and dioxide forms, by a common soil fungus, *Aspergillus niger*. Using a specialized spore collection method, the Pu was shown to be present in the fruiting bodies. Subsequent washing to remove external contamination indicated that the major portion of the ^{238}Pu was incorporated into the spores. Similar results were obtained in preliminary studies with U, Th, and Po in mine tailings. The order of uptake was related to pH and expected solubility of the Pu added with Pu in the initially soluble nitrate and citrate forms exhibiting a factor of 2 to 3 greater uptake than the dioxide (Table 4). Availability to microorganisms of the Pu in citrate and nitrate might be expected to be considerably higher than the oxide from solubility considerations at the pCi/ml level. The relatively high microbial availability of Pu as the oxide is highly significant, and further studies are warranted to determine the mechanisms of solubilization and uptake and the significance of microorganisms in recycling processes.

There is a growing literature on organic acids and bases, capable of complexing heavy elements, which are produced directly or by secondary syntheses by a variety of microorganisms. These products may be expected to be present in soils (discussed in detail in a previous section). Their concentration and form will be dependent upon the environmental factors influencing microbial metabolism, such as C-source, (previous section), and their residence time will be dependent upon subsequent chemical and microbiological stability.

In preliminary (unpublished) studies by the senior authors and others, mixed cultures of soil organisms, isolated from soil on the basis of C requirements and Pu resistance, were analyzed as to their ability to transport Pu into cells and to alter Pu form in the cellular and exocellular media. In addition an experiment was conducted to distinguish complexation reactions resulting from Pu interactions with metabolites arising from normal metabolic processes and Pu interactions with metabolites arising from Pu resistance. To make this distinction soil microorganisms were isolated from soil in the absence of Pu and Pu added at the stationary growth phase of an enriched culture, and, transport and complexation were compared to microbial cultures isolated from Pu-containing soil and grown in the presence of Pu.

After growth for 96 hr, the cultures were separated into cellular and exocellular fractions. The cell fraction was, in turn, homogenized into intracellular soluble and cell debris fractions. The results of studies in which Pu was added at the stationary growth phase of cultures of fungi or bacteria grown on mixed organic acids or sugars are summarized in Table 5. These cultures, selected only on the basis of their ability to grow on either of two C sources, differed to a first approximation, in their interactions with Pu. In general, the majority of Pu was associated with the exocellular fraction, but significant quantities were insoluble and associated with the cell wall and membrane fractions. However, the distribution of Pu between fractions was dependent upon microorganism type and C-source. In the case of fungi, the exocellular fraction of organisms grown on the organic acid C source contained less Pu than when mixed sugars were utilized as a C source. The reverse of this relationship occurred with the bacteria.

Differences in Pu distribution as a function of C source used in enrichment were also found in cultures grown in the presence of Pu throughout incubation (Table 6). The fungal cultures grown on mixed organic acids exhibited larger concentrations of Pu both in the exocellular fraction and

Table 4. Plutonium Transport to the Spores of *Aspergillus niger* (Beckert and Au, 1976).

Chemical Form	pH	Specific Activity of Culture Medium (dry), pCi/g	Specific Activity of Spores* pCi/g	Transport Factor
Dioxide	2.5	3.34×10^3 3.34×10^4 3.34×10^5	4.7 32 383	1.4×10^{-3} 0.96×10^{-3} 1.1×10^{-3}
	5.5	3.39×10^3 3.39×10^4 3.39×10^5	9.4 101.3 976	3.4×10^{-3} 2.98×10^{-3} 2.88×10^{-3}
Nitrate	2.5	1.67×10^3 3.34×10^3 6.69×10^3	3.8 11 15	2.3×10^{-3} 3.3×10^{-3} 2.24×10^{-3}
	5.5	1.69×10^3 3.38×10^3 6.76×10^3	8.3 26 23	4.9×10^{-3} 7.7×10^{-3} 3.4×10^{-3}
Citrate	2.5	1.67×10^3 3.34×10^3 6.69×10^3	7.1 11 18.5	4.3×10^{-3} 3.3×10^{-3} 2.8×10^{-3}
	5.5	1.69×10^3 3.38×10^3 6.76×10^3	6.7 26 56	4.0×10^{-3} 7.7×10^{-3} 8.3×10^{-3}

*Arithmetic mean of 3 samples.

Table 5. Distribution of Plutonium in Mixed Microbial Cultures Exposed to Plutonium at Stationary Growth Phase and Grown on Different Carbon Sources (Senior authors, unpublished).

Fraction	Distribution* of Plutonium (%) in Cultures			
	Fungi		Bacteria	
	Mixed Sugars	Organic Acids	Mixed Sugars	Organic Acids
Exocellular Medium	75	42	39	89
Intracellular Soluble	0.49	0.068	8.3	2
Cell Debris	10	42	28	8.7

*Cultures were not replicated. Analytical precision was < ± 10% (1σ).

Table 6. Distribution of Plutonium in Mixed Microbial Cultures Continuously Exposed to Plutonium and Grown on Different Carbon Sources (Senior authors, unpublished).

Fraction	Distribution* of Plutonium (%) in Cultures			
	Fungi		Bacteria	
	Mixed Sugars	Organic Acids	Mixed Sugars	Organic Acids
Exocellular Medium	29	54	46	88
Intracellular Soluble	4.2	0.24	2.7	4
Cell Debris	29	39	31	3.5

*Cultures were not replicated. Analytical precision was < ± 10% (1σ).

bound to the cell debris fraction; the cultures grown on mixed sugars contained a higher fraction of added Pu in the intracellular soluble fraction. In the bacterial cultures, the situation was somewhat different, in that higher concentrations of Pu occurred in the exocellular fraction of the culture grown in organic acids; less Pu was associated with the cell debris fraction as compared to cells grown on sugars.

In general, the continuous presence of Pu during growth did not have pronounced effects on the distribution of Pu in the cultures (compare Tables 5 and 6). Rather, the metabolic properties of the mixed cultures as determined by C source appeared to be the major factor resulting in the observed differences. Under both sets of culture conditions, there was a high concentration of Pu bound to cell wall and membrane fractions and thus insoluble. As these materials are degraded by lytic enzymes, e.g., proteases and chitinases, soluble Pu compounds may be formed.

Preliminary characterization, using gel permeation chromatography, of the mixed culture of fungi isolated from soil and grown in sugars indicated that Pu form was altered during fungal growth (Fig. 9). The exocellular and intracellular soluble fractions obtained from organisms exposed to Pu in a single exposure and in continuous exposure contained a majority of Pu in compounds of molecular size greater than Pu-DTPA, which was used as the source of soluble Pu. Furthermore, there appeared to be a difference in Pu chemical form comparing Pu complexes formed on simple interaction of Pu with metabolites (single exposure) and Pu complexes formed on interaction after continuous Pu exposure of the culture. This suggests either that the culture grown in the continuous presence of Pu contained metabolites capable of interacting with Pu which were different chemically from those produced by the culture grown in the absence of Pu or that the culture grown in the presence of Pu contained different organisms capable of adaptive response to the element leading to the synthesis of compounds relatively specific to detoxification of Pu.

Further chemical characterization using thin-layer chromatography and electrophoresis verified differences in Pu form. Several solvents of different polarities and pH values were employed to provide a range of chemical conditions for separation. Solvent systems included: A, butanol-pyridine, a system used in resolution of amino acids; D, pentanol-formic acid, a system used in separation of sugars and sugar acids; and G, water-acetic acid, a solvent utilized in resolving keto-acids and sugars. These systems were used to resolve Pu as Pu-DTPA, and Pu in the soluble exocellular and soluble intracellular fractions of the above cultures (Fig. 10). Thin-layer chromotography using solvent A indicated that the exocellular fraction contained one component of chromatographic mobility different than the added Pu-DTPA but the complex remained present in detectable quantities. The intracellular soluble fraction contained a component of lesser chromatographic mobility but there was no evidence of Pu-DTPA. Solvents D and G did not provide good resolution. Solvent D did not mobilize Pu-DTPA or other possible complexes, Solvent G mobilized Pu-DTPA and indicated the presence of immobile Pu components in the exocellular and intracellular fractions but these were not resolved.

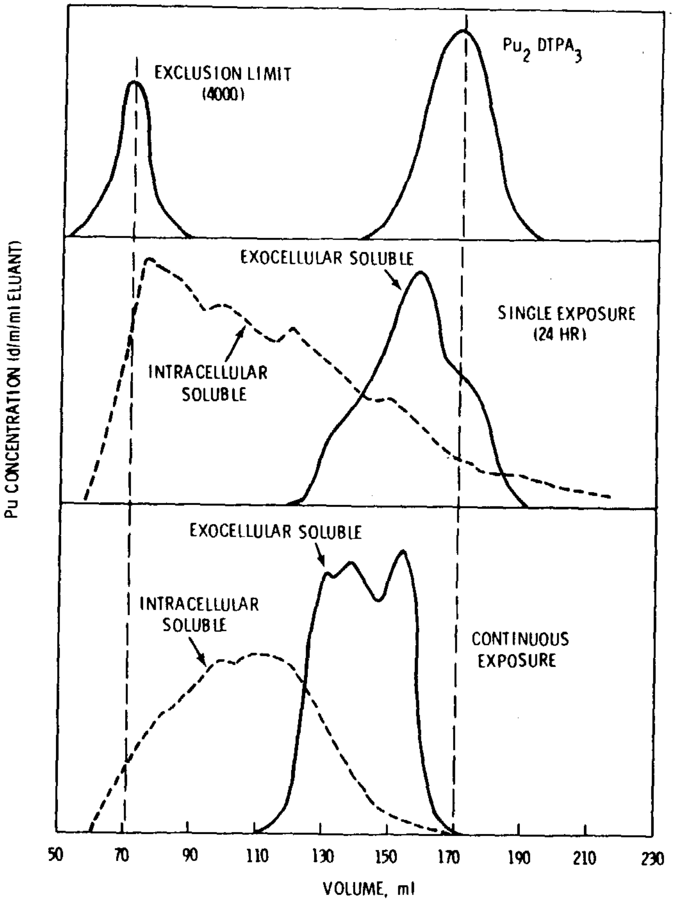

Fig. 9. Separation of soluble plutonium complexes in microbial cultures by gel permeation chromatography (Senior authors, unpublished).

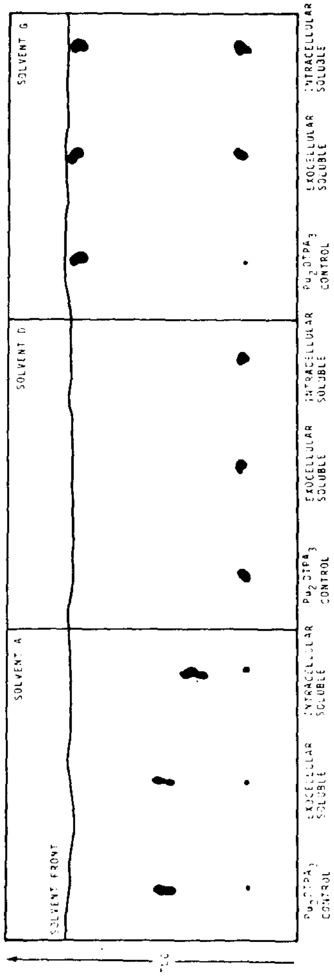

Fig. 10. Thin-layer chromatographic behavior in three solvent systems of plutonium complexes separated by gel permeation chromatography (Senior authors, unpublished).

Application of thin-layer-electrophoresis (pH 6.6, pyridine-acetate buffer system; cellulose support) indicated the presence (Fig. 11) of a relatively large amount of material of greater negative charge than Pu-DTPA in the exocellular fraction along with Pu-DTPA. The Pu ligands in the intracellular fraction were either neutral in charge in this buffer system or were of a molecular size too large to migrate under the conditions of electrophoresis. Similar alterations of Pu form by a single Pu-resistant fungus exposed continously to Pu during growth have also been reported (Robinson *et al.*, 1977).

Several phenomena may have been responsible for the observed changes in chemical form of Pu. The organism may have synthesized compounds which either bind Pu-DTPA or bind Pu more tightly than DTPA, thereby successfully competing for Pu in the presence of DTPA. Alternatively, the organism may degrade or modify the DTPA moiety allowing Pu transfer to ligands arising from microbial synthesis and degradation.

The number of known compounds with the potential to bind Pu more strongly than DTPA appears to be quite limited although hydroxamate derivatives, (Emergy, 1974) catechol derivatives, (Tait, 1975) and tetrapyrrole ring systems (Balker, 1969) may exhibit this property. If modification of the Pu-DTPA occurred prior to ligand transfer, then a myriad of microbially-produced compounds, e.g., phenolic acids, peptides, and carboxylic acids have potential for binding Pu (see previous section; also Alexander, 1971). In either case, the Pu was not in the form initially added. Thus, applications of gel permeation chromatography, thin-layer chromatography and thin-layer electrophoresis indicate that cultures of soil microorganisms were capable, through simple expression of the metabolic potential of microorganism present in soil, of changing the chemical form of Pu-DTPA with the resulting formation of a number of Pu complexes exhibiting a range in chemical properties. Differences in Pu distribution in microbial systems and in Pu form resulted from both simple interaction with metabolites and perhaps, more specific processes. These differences were dependent on organism type, metabolism and Pu resistance.

Although published information on transuranic elements other than Pu is limited, it is likely that similar transformations will occur. The extent of these transformations will be dependent upon the solubility of the element, its availability to microorganisms, its toxicity to microorganisms and its potential for complexation. While microbial interactions remain to be elucidated, the solubility (discussed in a previous section) and potential for complexation may be preliminarily assessed from known chemistry (Table 7). It is evident that the transuranic elements form DTPA complexes with stabilities similar in magnitude to Pu-DTPA over environmental pH ranges. It may be concluded that complexation with organic ligands produced by soil microflora is highly probable and investigations to identify and characterize the indirect processes and the ligands responsible for complexation of Pu in soil are equally applicable to other transuranic elements.

Cycling During Decomposition. A final process whereby the soil microflora may play a role in transformation of the transuranic elements involves the

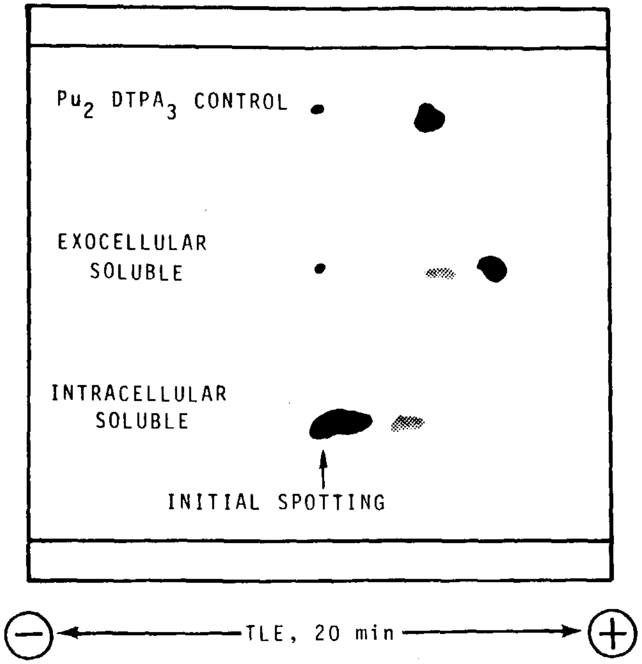

Fig. 11. Thin-layer electrophoretic behavior of plutonium separated by gel permeation chromatography (Senior authors, unpublished).

Table 7. Stability of DTPA Complexes with the Transuranic Elements (Hafez, 1969).

Complex	Stability Constant	Stable pH Range
Neptunium		
Np^{+3}	-**	-**
Np(IV) DTPA	10^{24}	0.5 - 5.8
$[Np(IV)]_2DTPA_3$	10^{20}	>5.8
Plutonium		
Pu(IV) DTPA	10^{24}	1.0 - 5.8
$[Pu(IV)]_2DTPA_3$	10^{18}	5.8 - 8.5
$Pu(IV)\ DTPA_2$	10^{14}	>8.5
Americium		
[Am(III)] DTPA	10^{20}	1.8 - 6
Am(III) DTPA	10^{23}	>6

* Curium may be expected to form complexes of stabilities similar to americium.

**Unstable in oxygenated solutions.

biological uptake (plants, microorganisms) of the elements and subsequent release on decomposition. Several studies have demonstrated plant uptake of Pu and Am and incorporation into above ground tissue. These tissues, deposited on soil either through litter fall or agricultural incorporation of crop residues will be subject to microbial decomposition. Furthermore, recent studies (Wildung and Garland, 1974) have indicated that barley roots (uncontaminated with soil particles), contained 3-8 times more Pu than the shoots. The roots of plants are in intimate contact with the soil and may be expected to decompose rapidly (weeks) under appropriate conditions of temperature and moisture, even in arid regions (Wildung *et al.*, 1975). Relatedly, microorganisms, due to their distribution in soil and large absorptive surface, compete efficiently with plants for ions in soil (Alexander, 1961). Studies described in a previous section demonstrated the association of Pu with microbial cells. Growth of microbial cells, a significant portion of the soil biomass, may therefore represent an important mechanism for biological incorporation of the transuranic elements. Decomposition of microbial cells generally proceeds at a more rapid rate than plant tissues.

Little is known of the form of the transuranics in plant or microbial tissues; of the form, rate, and extent of the transuranics released on decomposition of these tissues; or of the chemical reactions governing transuranic solubility after decomposition. However, considering the known products of microbial metabolism of organic substances, including a number of strong complexing agents (previous section), and the susceptibility of a number of the transuranic elements to complexation (previous section), it may be concluded that the transuranics, initially immobilized through biological uptake, may be at least as soluble and perhaps more soluble on decomposition.

In preliminary studies (Wildung and Garland, unpublished), Pu-amended soil containing largely undecomposed roots from a previous barley crop was leached with water and Pu solubility compared to a fallow soil containing Pu at similar levels. The results indicated that soluble Pu was initially immobilized by incorporation into roots, decreasing by a factor of 10 after root growth. Root decomposition studies are in progress. Previously observed (Romney *et al.*, 1970) increases in Pu uptake from soils by plants with increased time, generally attributed to increased root development, may have been due to increased availability through a recycling process on decomposition of plant roots. The importance of the process will be dependent upon transuranic availability to different plants and microorganisms, the turnover rate of this tissue in soils under different conditions and the stability, chemistry and biological availability of transuranic metabolites. Until this information is developed, the long-term effects of recycling processes will remain unknown.

ACKNOWLEDGEMENTS

Grateful appreciation is extended to Thomas R. Garland for his invaluable technical contributions and advice during the preparation of this manuscript.

This paper is based on work performed under U.S. Energy Research and Development Administration Contract E(45-1):1830 and Memorandum of Understanding No. AT(26-1)-539.

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ESTIMATION OF AEROSOL PLUTONIUM TRANSPORT BY THE DUST-FLUX METHOD: A PERSPECTIVE ON APPLICATION OF DETAILED DATA.

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ABSTRACT

Two methods of dust-flux measurements are discussed which have been utilized to estimate aerosol plutonium deposition and resuspension. In previous studies, the methods were found to be sufficiently detailed to permit parameterization of dust-flux to the erodibility of the soil, and a seventh-power dependency of dust-flux (or plutonium flux) to wind speed was observed in worst case conditions.

The eddy-correlation method is technically more difficult, requires high-speed data acquisition, and requires an instrument response time better than one second, but the eddy-correlation method has been shown feasible with new fast-response sensors, and it is more useful in limited areas because it can be used as a probe. The flux-gradient method is limited by critical assumptions and is more bulky, but the method is more commonly used and accepted. The best approach is to use both methods simultaneously.

It is suggested that several questions should be investigated by the methods, such as saltation stimulation of dust-flux, simultaneous suspension and deposition, foliar deposition and trapping, erodibility of crusted surfaces, and horizontally heterogeneous erodibility.

INTRODUCTION

One of the key issues facing us in assessment of the health hazards due to plutonium aerosols is the manner in which practicle deposition and resuspension is rate limited by the underlying surface. We have made progress in the past by measuring the ambient Pu concentrations and by utilizing resuspension factors, but these methods lack detail in both the time and space resolutions possible with the present state of the art. For example, we are not yet able to determine vertical and horizontal Pu aerosol concentration over a Pu-contaminated site with sufficient detail to determine the resuspension rate at low wind speeds.

If it is known, however, what the Pu host-particle characteristics are both in terms of number-size distribution in the total suspended aerosol, and in terms of total aerosol activity, it turns out in many cases to be better to study the host-particles rather than the Pu activity directly. If we assume the host-particles are dust, originating from the contaminated soil below, we find that the dust-flux method gives considerable degree of detail and allows a more intimate study of the processes of transport and the rate limiting factors at the ground surface. The purpose of this presentation is to review two approaches to measurement of dust-flux, to describe experiments in which the methods have been applied, to show the limitations and assumptions of the methods, and to summarize some new questions about Pu transport which have been revealed by our experiences in applying the methods.

THEORY OF ATMOSPHERIC DUST-FLUX AT THE SURFACE

Dust concentration is a scaler quantity which obeys laws of classical micrometeorology near the ground. These laws are explained in several standard texts and apply to gases which are exchanged with the surface (Sutton, 1953; Priestly, 1959; Chamberlain, 1975). Dust particles behave like gases unless they exceed about 20 μm diameter, above which their gravitational settling-velocity becomes increasingly important in determining their rate of deposition or suspension; let us confine our discussion to the respirable range of sizes below 20 μm. Given an "ideal" detector during a high mass-loading observation period in the atmosphere, we find that dust concentrations (and therefore Pu concentrations) will have the same apparent random variations as does a sensitive wind speed detector; see Fig. 1. The vertical flux of dust (F) is the mass passing through a given horizontal area per unit time and can be defined by an eddy-correlation:

$$F = - \overline{\chi' w'} \qquad (1)$$

where χ' is the instantaneous deviation in dust concentration from stationary mean (e.g., ten minute mean on Fig. 1), w' is the instantaneous vertical wind velocity component, and the overbar denotes a time average of the covariance product. The negative sign indicates a loss from the ground surface (either suspension or resuspension). By analogy to molecular transport processes, the flux-gradient equation is:

$$F = - K \frac{d\chi}{dz} \qquad (2)$$

where K is a diffusion coefficient and $d\chi/dz$ is the vertical gradient of dust concentration. It is generally accepted that we can estimate K from boundary layer theory, in an analogy to momentum-flux:

$$K = u^* k' z \qquad (3)$$

where u^* is the friction velocity proportional to horizontal wind speed, k' is Karman's constant adjusted for atmospheric stability, and z is the vertical height ordinate.

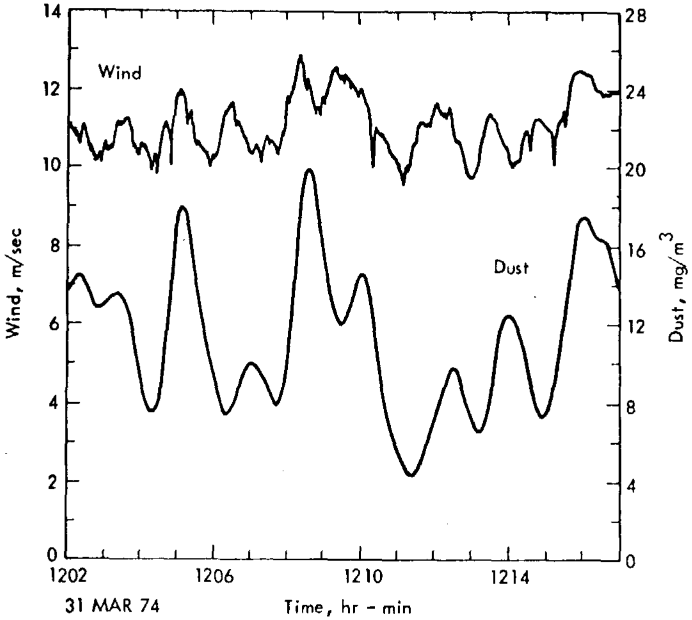

Fig. 1. Detailed information of wind-speed and dust which can be observed in the atmosphere when sensors are appropriately sensitive.

The assumptions for equations (1) and (2) other than already expressed are that (a) steady-state conditions exist for time-averaged quantities, (b) horizontal gradients are insignificant so that all fluxes are vertical, and (c) the equations are applied in a zone known as the constant flux layer beneath the surface boundary layer, which grows nearly linearly with distance downwind from a surface property discontinuity.

MEASUREMENT PRINCIPLES

Relatively new developments in instrumentation have increased the feasibility of making dust-flux measurements by either (1) the eddy-correlation method or (2) the flux-gradient method. The first is technically more difficult. Both methods were simultaneously demonstrated, perhaps for the first time, by Shinn *et al.*, 1976. Gillette *et al.*, 1972; Gillette, 1976; and coworkers have used the second method extensively for several years. The eddy-correlation method requires high-speed data acquisition and/or processing and is at present still in the prototype phase because of the requirement for a speed of response better than one second. The integrating nephelometer used by Shinn (*loc. cit.*) is unsuitable at low mass loadings and is marginal at high winds. Recently, Husar and Macias (1976) developed a diffusion charging, fast-response aerosol detector which has been feasibility demonstrated for particle flux using the eddy-correlation method at Argonne National Laboratory, Wesley *et al.*, 1976. With these new developments, perhaps resources will be applied to meet our needs, which is to have an adequate tool for examining processes that rate limit Pu transport.

The flux-gradient method contains more critical assumptions than the first method and is more bulky and less useful as a probe for contaminated areas of limited extent. It is, however, more adaptable to long-term monitoring. Measurements of the gradient have to be performed in a relatively shallow zone beneath the surface boundary layer. The concentration gradient of dust being rapidly suspended (upward dust flux) has been found to conform to a power-law (Fig. 2). That is

$$\chi = A\, z^{P} \qquad \text{A,P constants} \tag{4}$$

so that $$\frac{d\chi}{dz} = P\,\frac{\chi}{z}\,. \tag{5}$$

In strong wind speed cases, it was estimated (Shinn *et al.*, 1976) that the power $P \approx -0.25$, but much more data should be examined. In measurements of concentration gradients, more than two detectors are required (preferably more than three) and the detectors must be matched in response and calibration. Optical particle detectors are best; gravimetric methods are least satisfactory.

In summary, both of the approaches to measure dust-flux have drawbacks, they are about equal in cost, and perhaps it is more satisfactory to operate both simultaneously.

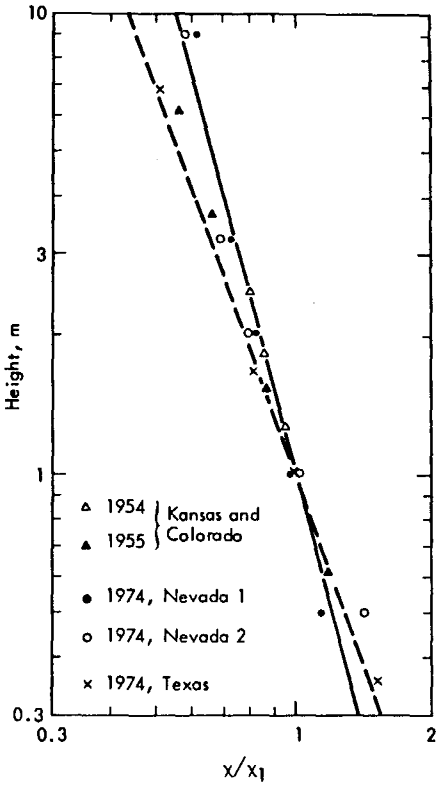

Fig. 2. Typical dust gradients with height during strong suspension events. Solid line is a power law with exponent -0.25; dashed line is power law with exponent -0.35.

PROBLEMS OF Pu TRANSPORT REVEALED IN APPLICATIONS OF THE METHODS

Dust-flux measurements at the Nevada Test Site and elsewhere have shown that even under nearly ideal conditions, there are major difficulties in relating a measured dust-flux to the underlying surface properties. It may be possible to determine the physical factors causing particles to initiate motion, but in order to be practical, these factors should also be parameterized to measurable variables in some general way.

Shinn *et al.*, 1976, found that upward dust-flux (suspension) apparently could be parameterized to "soil erodibility-index" using an empirical expression as follows:

$$F = - F_o \, (U_*/U_o)^{\gamma+1} \qquad (6)$$

where U_o is constant, and F_o and γ are hypothesized to depend on soil erodibility index, perhaps as shown in Fig. 3. Even though this hypothesis is not adequately verified, it is important to note that (1) the soil erodibility factors will strongly control the suspension rate of Pu host particles, and (2) the rate of Pu suspension is proportional to wind speed raised to a power of $\gamma + 1$, which has been observed to exceed a value of 6. This fantastically sensitive response to wind speed is verified in experimental data of Gillette (1976) and coworkers. Without the sensitivity of fast response, dust-flux measurements, these observations could not have been made, and their implications for Pu-transport would have gone unnoticed.

The Nevada Test Site is unique in that the natural suspension rate due to convective winds is very low compared to more erodibile sites in western U.S. After several seasons of dust-flux investigations, we have concluded that the natural desert shrublands and dry lakes are not subject to wind erosion unless they are physically disturbed such as by foot or vehicular traffic. The desert shrublands are covered by a "desert pavement" consisting of small pebbles weakly bound together by a surface crust. Likewise, the dry lakebeds are covered by a crust following each rainstorm. Our investigations are inconclusive about long-term dust-flux at the Nevada Test Site because the magnitude of the flux is so low as to require newer instrumentation than we presently have.

There have arisen, however, several definite questions at the Nevada Test Site which require further research. These are as follows:

1. What is the role of saltation flux (horizontal transport of larger soil particles, bouncing along the surface) on the overall net loss of Pu by vertical dust-flux?
2. What are the magnitudes of the rate of deposition and the rate of suspension occurring simultaneously during wind events to produce a net Pu transport?

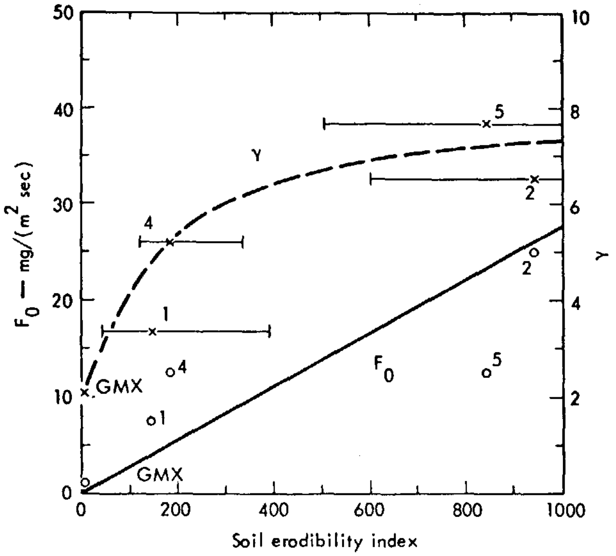

Fig. 3. Relationship of the power γ and the reference flux F_0, to the erodibility of soil surfaces in Texas and Nevada.

3. What are the rates of foliar deposition of Pu on a short-term basis and of what importance is the accumulation of drifted soil around shrubs?

4. What parameters of soil erodibility can be utilized for assessment of long-term Pu flux, especially under conditions of a desert pavement?

5. Are the dust-fluxes heterogeneously distributed because of local variations in erodibility and if so, are there conditions where nonsteady meteorological approaches can be used?

All of the above questions are possible to attack using either one or both of the dust-flux methods discussed here. Our future partipation we hope will address some of these questions.

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PLUTONIUM AND TRACER PARTICLE RESUSPENSION:
AN OVERVIEW OF SELECTED BATTELLE-NORTHWEST EXPERIMENTS

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ABSTRACT

Plutonium resuspension at Rocky Flats was determined in July 1973, by measuring airborne plutonium as a function of sampling height and of wind speed. The maximum airborne plutonium-239 concentration was 3700 aCi/m^3. The maximum plutonium-239 activity per gram of collected airborne soil was 50 pCi/g. Airborne plutonium concentrations were shown to increase as the sixth power of wind speed, but the data were very imprecise.

In measurement of fallout resuspension caused by burning vegetation, the ^{137}Cs concentration in a forest fire smoke plume was 22 times the concentration in ambient air.

Resuspension rates for a man walking across a tracer area on an asphalt surface ranged from 1×10^{-5} to 7×10^{-4} fraction of tracer resuspended/pass. For vehicular traffic driven on a tracer lane, resuspension rates from the asphalt surface ranged from 10^{-4} to 10^{-2} fraction resuspended/pass. When the vehicle was driven on the lane adjacent to the tracer lane, resuspension rates were smaller, ranging from 10^{-5} to 10^{-3} fraction resuspended/pass. Resuspension rates decreased when the tracer had been on the ground for several days. In comparison to the asphalt surface, vehicle-caused tracer resuspension rates from a cheat grass area were lower, ranging from 10^{-5} to 10^{-4} fraction resuspended/pass.

Wind caused tracer resuspension rates were on the order of 10^{-11} fraction resuspended/sec for nonrespirable particles. Resuspension rates for respirable particles increased as the 4.8th power of wind speed, ranging from 10^{-11} to 10^{-7} fraction resuspended/sec. In another study, airborne respirable soil concentrations measured as a function of wind speed increased as the 0.6 to 3.2 power of wind speed.

INTRODUCTION

Radioactive particles deposited on natural or man-made surface can be resuspended by wind and mechanical activity. Wind resuspension may occur over a wide area as well as a local area. In contrast, mechanical activity resuspension is usually more localized and may present an immediate inhalation problem to the worker in a contaminated zone. In both wide-area and local resuspension of radioactive particles, particles are transported downwind and in sufficient concentration could become a potential radiological hazard to man. Sources for resuspended particles include radioactive fallout as well as releases from nuclear facilities. Radioactive fallout is a worldwide phenomenon in which stratospheric debris is deposited on environmental surfaces. Subsequently, deposited fallout particles can be resuspended and transported downwind. At present, the significance of fallout resuspension is unknown. Data are needed to define the relative inhalation hazard between fallout particle resuspension and the direct delivery of stratospheric debris. Currently, fallout resuspension is assumed to contribute little radiological hazard to man. Radioactive particle resuspension is probably more important at nuclear facilities near which the environment has been contaminated with radioactive particles. These particles can be resuspended by both wind stresses and mechanical disturbances and transported downwind. However, resuspension mechanisms are poorly understood and, consequently, resuspension rates and potential airborne inhalation hazards cannot now be adequately predicted.

The need for such predictions are not new: resuspension has been known for many years to be occurring at nuclear sites. Wilson et al. (1961) and Anspaugh et al. (1969) obtained some of the earliest of such data at the Nevada test site. Ground radioactivity contours were determined as a function of time after a test detonation. The contours indicated that deposited radioactive particles must have been resuspended by prevailing winds and that ground surface concentrations decreased with increasing distance from the test center during the initial time period after the test (Anspaugh et al., 1969). Subsequent ground radioactivity contours showed a migration of radioactivity away from the test site, indicating that resuspension had occurred.

More recently, resuspension has been of considerable interest at the Rocky Flats nuclear plant in Colorado where ground surfaces were contaminated with plutonium from leaking storage barrels containing plutonium-contaminated cutting oil (Sehmel and Lloyd, 1976a; Krey et al., 1973; Krey et al., 1976; Volchok et al., 1972; Johnson et al., 1976). After the leakage was discovered, the barrels were removed and corrective actions were taken, but plutonium resuspension from residually contaminated soil surfaces is still occurring.

Although environmental plutonium resuspension is receiving attention, resuspension physics is poorly understood. Resuspension is frequently characterized by a "resuspension factor". The resuspension factor is defined as the ratio of airborne pollutant concentration (amount/m^3) at breathing height divided by the ground surface contamination level (amount/m^2). Thus, the resuspension factor has units of m^{-1}. Reported resuspension factors vary many orders of magnitude with values from 10^{-11} up to 600 m^{-1} (Stewart, 1967; Mishima, 1964; Sehmel and Lloyd, 1976b). Resuspension factor variations have not been adequately explained as a function of experimental conditions.

Resuspension factors from about 10^{-9} to 10^{-5} m^{-1} are often used in hazard evaluations. The resuspension factor is useful since a worker's inhalation hazard is most likely related to the local resuspension caused by his work activities within a contaminated zone; however, resuspension factors are only a very rough estimate of the potential airborne contaminant concentration since resuspension factors cannot be accurately predicted. In addition to local resuspension, airborne contaminated particles can reach workers from upwind contaminated areas. Hence, both local and upwind resuspension should be considered.

The resuspension factor is an index of only the potential inhalation concentration and not the total resuspension release rate from a surface-contaminated area. Resuspension release rates are needed for source terms in calculating total downwind diffusion and transport of resuspended particles. It is only recently that particle resuspension rates have been measured (Sehmel and Lloyd, 1976b).

OBJECTIVES

The objective of this research is to measure particle resuspension rates from environmental surfaces caused by wind and mechanical disturbances. Subsequently, generalized models to predict resuspension rates can be developed and incorporated into downwind transport models (Horst, 1976). The objective of this paper is to summarize reported resuspension rates and parameters that have been determined by Battelle-Northwest between 1971 and early 1976. These include plutonium resuspension measurements at Rocky Flats in July, 1973, resuspended fallout concentrations in the smoke of a forest fire, and measurements using controlled tracer particles.

EXPERIMENTS

The experiments for measuring particle resuspension reported here have been reported in fuller detail in the following references:

- plutonium from contaminated environmental surfaces at Rocky Flats (Sehmel and Lloyd, 1976a),
- fallout particles in forest fire smoke (Sehmel and Orgill, 1976),
- controlled inert tracer particles from selected surfaces on the Hanford reservation (Sehmel and Lloyd, 1976b; Sehmel, 1973a; Sehmel, 1976), and
- soil particles from ground surfaces, also at Hanford (Sehmel, 1975a).

Most of the wind-caused resuspension research concerns resuspension from vegetated areas. Experiments concerning local resuspension caused by mechanical activity include tracer studies of resuspension rates for a man walking across an asphalt strip and for cars and trucks driven on asphalt or cheat grass.

Two different resuspension rates are used. For wind-caused resuspension, resuspension rates are reported as the fraction of particles resuspended/sec. Thus, the total wind-caused resuspension is a product of the surface contamination area, the surface contamination level, the duration of resuspension, and the resuspension rate. For mechanical disturbances by vehicular or pedestrian traffic, resuspension was measured each time a car, 3/4-ton truck, or person passed across the length of a 3-m-wide, tracer-contaminated area. Thus traffic resuspension rates are reported as the fraction of particles resuspended/pass.

PARTICLES

Resuspension was measured for several types of particles. The particle size distribution of plutonium on the soils at Rocky Flats was uncontrolled since the original release was unplanned. A forest spray operation provided an opportunity to measure resuspension of DDT as tracer particles not specifically controlled for size. The controlled, inert tracer particles used were submicrometer $CaMoO_4$ particles and ZnS particles with an 8-μm mass aerodynamic equivalent diameter.

AIR SAMPLERS

Airborne resuspended particles were either sampled with total air samplers or sized while airborne with particle cascade impactors. Particle cascade impactors were used for plutonium and $CaMoO_4$ particles.

The particle cascade impactor(a) for sampling respirable particles was attached to a rotating cowl, allowing simultaneous sampling of larger nonrespirable particles. The cowl-impactor system is shown in Figure 1 (Sehmel, 1973b). Particles entering the 15-cm-dia cylindrical sampler

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(a) Anderson 2000, Inc., Model 65-100 High-Volume Sampler Head, P. O. Box 20769, AMF, Atlanta, Georgia 30302.

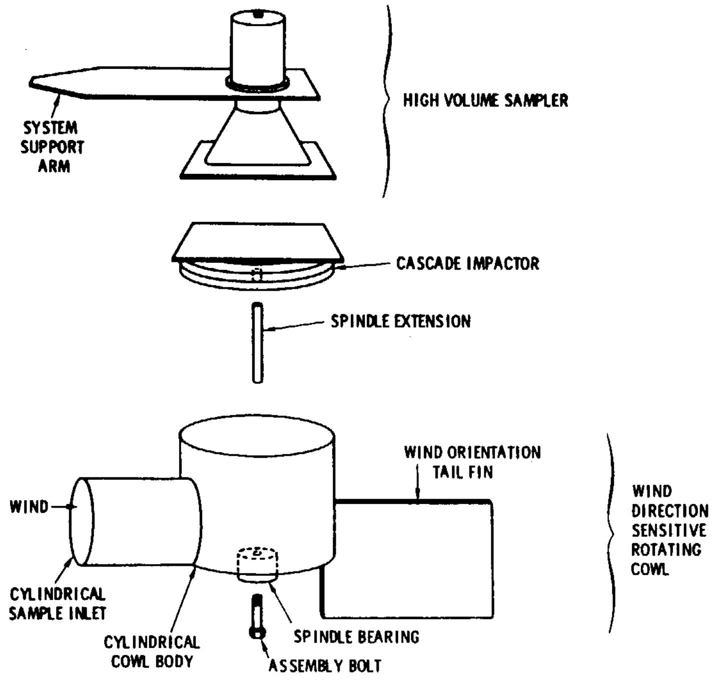

FIGURE 1. Rotating Cowl and Impactor

inlet of the cowl either settled on the cowl floor or were drawn up into the impactor. Particles settling on the cowl floor will be called "non-respirable" in this report. Respirable particles entering the particle cascade impactor were separated into nominal aerodynamic diameter ranges of 7, 3.3, 2.0 and 1.1 μm, which are impactor stage 50% cutoff diameters for unit-density spheres. Smaller particles were collected on an impactor backup filter.

RESULTS AND DISCUSSION

Particle resuspension rates were determined for $CaMoO_4$, ZnS, and DDT tracer particles. In contrast to resuspension rates, only airborne concentrations as a function of wind speed were determined for plutonium resuspension at Rocky Flats, and only total airborne radionuclide concentration was determined for the forest fire smoke. Results for each set of experiments will be discussed separately.

RADIOACTIVE PARTICLES

Airborne plutonium concentrations in July 1973 at Rocky Flats, Colorado were measured as a function of respirable particle diameter, wind speed, and sampling site. Total radionuclide concentration in smoke from a forest fire near Mt. St. Helens, Washington was measured on September 6, 1975.

Plutonium Resuspension at Rocky Flats

Plutonium resuspension at Rocky Flats was investigated experimentally. In early work, an empirical resuspension model was developed by Sehmel and Orgill (1973) based on published weekly plutonium concentrations at Health and Safety Laboratory sampling station S-8 along the site's eastern security fence. These plutonium data were analyzed in terms of the meteorology occurring during sampling times. Collected airborne plutonium was related to hourly average wind speeds and wind directions. Model results indicated that airborne plutonium concentrations increased as the 2.1 power of wind speed. Subsequently, the model was used to predict airborne concentrations for a succeeding time period. These results showed a wide difference between predictions and experimental results. The interpretation of these differences was that the plutonium resuspension source characteristics changed as a function of time (Sehmel and Orgill, 1974).

Battelle-Northwest experimental measurements of plutonium resuspension at Rocky Flats were made in July 1973 (Sehmel and Lloyd, 1975a, 1976a). Airborne plutonium concentrations were measured at three sampling sites east of the plant. The first sampling site was along the eastern security fence.

This site was called sampling site A. Sampling site B was along the eastern cattle fence, and sampling site AB was between sites A and B. The distance from site A to site AB was 392 m, and from site AB to site B was 277 m. Airborne plutonium at these sites was sampled and analyzed as a function of sampling height, particle size, and wind speed. For comparison, a particle cascade impactor sample was simultaneously collected at a background site 13 km away in the mountains.

Fallout levels of plutonium-239 entering the area were estimated from the cascade impactor operated in the mountains. There was no detectable plutonium-239 activity on the 7, 3.3, and 1.1-μm impactor stages, and there was no radiochemical result for the 2-μm stage. The only detectable background plutonium activity was on the backup filter which nominally collects submicrometer particles. Airborne plutonium-239 concentration at the background station was 4 ± 3.5 aCi/m^3, which corresponds to 0.7 ± 0.62 pCi/g of airborne soil on the backup filter. Error limits are the 2σ radiochemical counting limits.

Airborne plutonium-239 concentrations at the three Rocky Flats sampling stations were reported in aCi/m^3 of air and pCi/g of airborne soil. The maximum airborne plutonium-239 concentration was 3700 aCi/m^3. The maximum plutonium-239 concentration on the airborne soil was 50 pCi/g total airborne soil and 69 pCi/g for the respirable fraction of airborne soil collected on the 2-μm particle impactor stage. All airborne plutonium-239 concentrations were significantly less than maximum permissible concentrations of soluble plutonium-239 in air for occupational exposure in a 40-hr work week (2×10^6 aCi/m^3) or nonoccupational exposure in a 168-hr week period (6×10^5 aCi/m^3) (International Commission on Radiological Protection, 1959).

The functional relationship between airborne plutonium resuspension concentrations and wind speed could not be developed as unequivocally as initially anticipated (Sehmel and Lloyd, 1976a). This was due in part to the inadvertent loss of about a fifth of the collected filter samples during radiochemical analysis. Unfortunately, most samples from the higher wind speeds were lost. Nevertheless, some incremental wind speed results were obtained.

Airborne plutonium concentrations were a function of both sampling height and particle diameter. Airborne concentrations in aCi/m^3 are shown for site AB in Figure 2 for each stage of the particle cascade impactors. In contrast to simple modeling concepts, airborne concentrations did not always decrease with an increase in height. There were unexpectedly high plutonium-239 concentrations at this site for several particle diameters and heights.

Plutonium was associated with particles collected on each stage of the particle cascade impactors. Since there was no plutonium in the upper stages of the impactor at the background mountain site, the plutonium-239 found in the upper stages of the impactors at the Rocky Flats sampling sites indicates that resuspension was occurring. Resuspension of submicrometer particles also occurred at Rocky Flats.

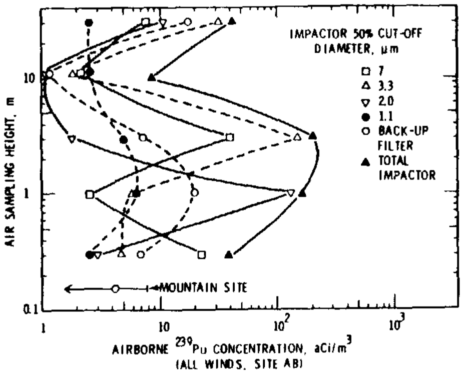

FIGURE 2. Airborne ^{239}Pu Concentration at Site AB as a Function of Impactor Collection Site

The general trend of the complete airborne plutonium-239 concentration data is a decrease in concentration with increasing distance eastward from site A (Sehmel and Lloyd, 1976a). As might be expected, this decrease in concentrations corresponded to increasing distance from the original oil storage area, which was the principal source of ground contamination. However, significant deviations did occur in the concentration profiles of airborne plutonium-239 with both distance and height. These deviations might be attributed to sampling "hot" soil particles which contain relatively more plutonium than average. These increases in average airborne plutonium-239 concentrations were present at both sites AB and B.

As indicated in Figure 3 for site AB, "hot" particles may have been present in the 2.0-µm size range. In this case, the concentration at the 1-m height of site AB is 1 to 2 orders of magnitude greater than at other heights for this site. More important to the "hot" particle concept is the concentration at the 10-m height of B. This concentration of 230 aCi/m^3 was the largest plutonium-239 concentration for 2-µm particles measured at any location. This relatively high concentration was unexpected since this sampling location was the most remote from both the ground and the original oil storage area. This suggests that other relatively "hot" particles could also be escaping from the plant boundaries; however, due caution is indicated in interpreting this "hot" particle concept. The total of 6 dis/min collected on the 2-µm stage or 230 aCi/m^3 is much less than the maximum permissible air concentration of 2 x 10^6 aCi/m^3 (occupational). It is conceivable that the majority of this "hot" plutonium might have been attached to one soil particle.

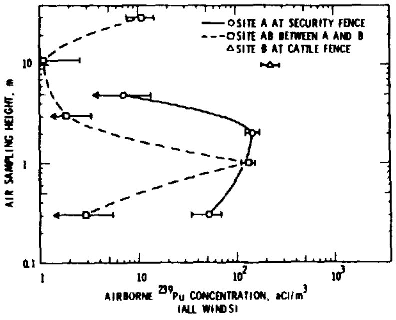

FIGURE 3. Airborne ^{239}Pu Concentrations from Impactor 2.0 µm Stage Collections

Even with the limited plutonium data collected in this experiment, it was evident that airborne plutonium-239 concentrations increased with an increase in wind speed. In Figure 4, total airborne concentrations are shown for air sampled at all wind speeds (average wind speed of 0.9 m/sec), at wind speeds from 4.1 to 6.3 m/sec, and at wind speeds from 6.3 to 9.8 m/sec. Airborne plutonium-239 concentrations at wind speeds from 4.1 to 6.3 m/sec are definitely larger than average airborne concentrations for continuous air sampling. However, the 2σ radiochemical counting statistics error limits are too large to determine the wind speed dependency. Nevertheless, an attempt to approximate airborne plutonium-239 concentrations and consequently the resuspension rate dependency upon wind speed was made for the 7-µm-diameter particles. This was for the 0.3-m height at sampling site AB. For the three data points taken at the 0.3-m height, plutonium-239 concentrations tended to increase as the 5.9th power of wind speed.

This July 1973 plutonium resuspension experiment at Rocky Flats showed resuspension of both plutonium-238 and plutonium-239. However, all airborne plutonium concentrations were significantly below maximum permissible concentrations in air. Since plutonium-239 was collected on each particle cascade impactor stage, the suggestion is that most plutonium was attached to soil particles when the plutonium was resuspended.

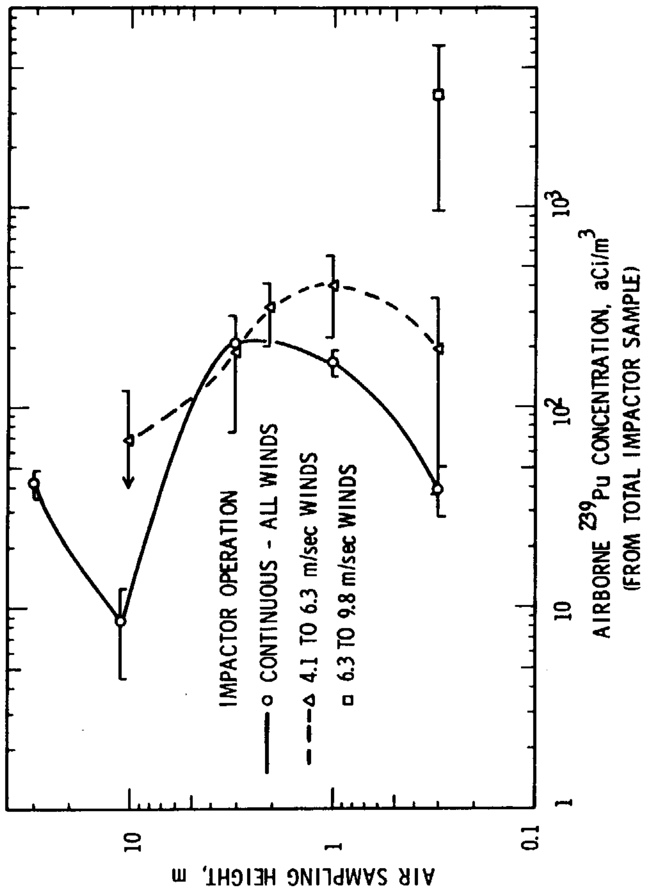

FIGURE 4. Total Airborne ^{239}Pu Concentration at AB Site as a Function of Wind Speed

Fallout Resuspension from Burning Vegetation

Another resuspension stress is the release of radioactive particles by burning contaminated vegetation (Sehmel and Orgill, 1975, 1976). To determine an indication of this release, smoke from a forest fire was sampled with an aircraft-mounted air filter. Air activity concentrations measured on the smoke filter were compared to concentrations measured with a similar filter in background air uncontaminated with smoke. A comparison of the radioactivity in the smoke sample versus the background sample indicated that radioactivity was released by the forest fire. Results are shown in Table 1 for 8 radionuclides. The airborne concentration ratio (smoke/background) for equal volumes of smoke and background air ranged from 1.2 to 22.2. All of these concentration ratios indicate the release of deposited fallout by the forest fire. However, release rates are unknown. Calculations of release rates would require knowledge of the smoke plume's total volume and average smoke concentration as well as the contamination level in the burning forest.

TABLE 1. Radionuclide Air Concentration: Smoke/Background

Isotope	Concentration Ratio
^{7}Be	1.2
^{54}Mn	3.8
^{95}Zr	1.9
^{95}Nb	1.8
^{106}Ru	2.9
^{125}Sb	4.6
^{137}Cs	22.2
^{144}Cs	2.9

TRACERS

Tracer particles placed on selected surfaces were used to measure resuspension rates caused by both mechanical and wind resuspension. Mechanical resuspension was measured for vehicular traffic on asphalt and cheat grass areas and pedestrian traffic on an asphalt area. Wind resuspension was measured as a function of wind speed and also as a function of respirable and nonrespirable particle diameters.

Mechanical Resuspension Rates

Mechanical resuspension includes both vehicular resuspension and pedestrian resuspension.

Vehicular Resuspension

A 3/4-ton truck and a car were driven over ZnS tracer particles placed upon a lane of asphalt road. Resuspended tracer was measured to determine resuspension rates (Sehmel, 1973a). Results are shown in Figure 5 for particle resuspension rates at vehicle speeds of 5, 15, 30, and 50 mph. The resuspension rate is the fraction of particles resuspended from the tracer lane each time the vehicle was driven down the road (fraction resuspended/pass). When a car was driven through the tracer lane, resuspension rates increased with the square of car speed from about 10^{-4} to 10^{-2} fraction resuspended/pass. This means that these resuspension rates were proportional to car-generated turbulence. When the car was driven on the lane adjacent to the tracer lane, resuspension rates were lower for each speed but increased with speed from about 10^{-5} to 10^{-3} fraction resuspended/pass.

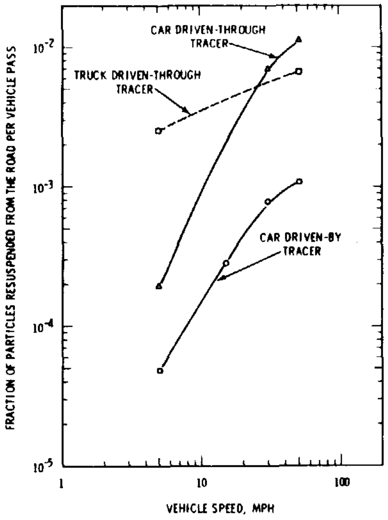

FIGURE 5. Rates of Particle Resuspension Caused by Vehicle Passage Over an Asphalt Road

Resuspension was also measured when a 3/4-ton truck was driven on the tracer lane. Resuspension rates for truck passage increased from about 10^{-3} to 10^{-2} fraction resuspended/pass. Since resuspension rates were higher, truck-generated surface stress turbulence appears to have been much greater than for car-generated turbulence. For vehicle speeds above 30 mph, resuspension rates for car and truck passage are comparable.

Resuspension rates were also a function of the time tracer particles were on the asphalt road. As shown in Figure 6, particle resuspension rates decreased as a function of time. For these data, the tracer had been on the road for four days. Vehicle-generated resuspension rates increased from about 10^{-5} to about 10^{-3} fraction resuspended/pass as vehicle speed increased from 5 to 50 mph. For both vehicles, resuspension was greater when the vehicle was driven through the tracer lane than when driven on the lane adjacent to the tracer lane.

Resuspension caused by truck passage through a cheat grass area (Figure 7) was also measured (Sehmel, 1976). Results are shown in Figure 8 along with resuspension rates from the asphalt road. Truck-caused resuspension in the cheat grass area was always less than that on the asphalt road. This decrease is attributed to the protective action of cheat grass in hindering truck-generated turbulence in reaching the ground and resuspending tracer.

Resuspension from the cheat grass area decreased for truck speeds from 5 to 30 mph. This decrease is attributed to the experimental truck speed sequence. The initial truck speed was 5 mph. Apparently, the relatively larger resuspension rate at 5 mph was caused by the most readily resuspended particles being removed from the cheat grass. In succeeding experiments at increasing truck speeds up to 15 mph, and possibly 30 mph, all readily resuspended tracer was removed from the cheat grass foliage. When truck speed was subsequently increased from 30 to 40 mph, resuspension per pass also increased. Apparently, increased air turbulence at the base of the cheat grass increased resuspension rates.

The results above indicate that particle resuspension rates are a function of both vehicle speed and the relative location of the material being resuspended. Tracer particles at the base of the cheat grass are more protected from resuspension forces and are less readily resuspended than material on the cheat grass upper foliage.

Pedestrian Resuspension

Resuspension caused by a man walking along the ZnS tracer lane of the asphalt road was also measured (Sehmel and Lloyd, 1972). The man walked across the tracer area in a leisurely fashion; the stride and paces per second were not measured. For tracer on a 3-m-wide road lane, the reported resuspension rate was the fraction of particles resuspended each time the person walked

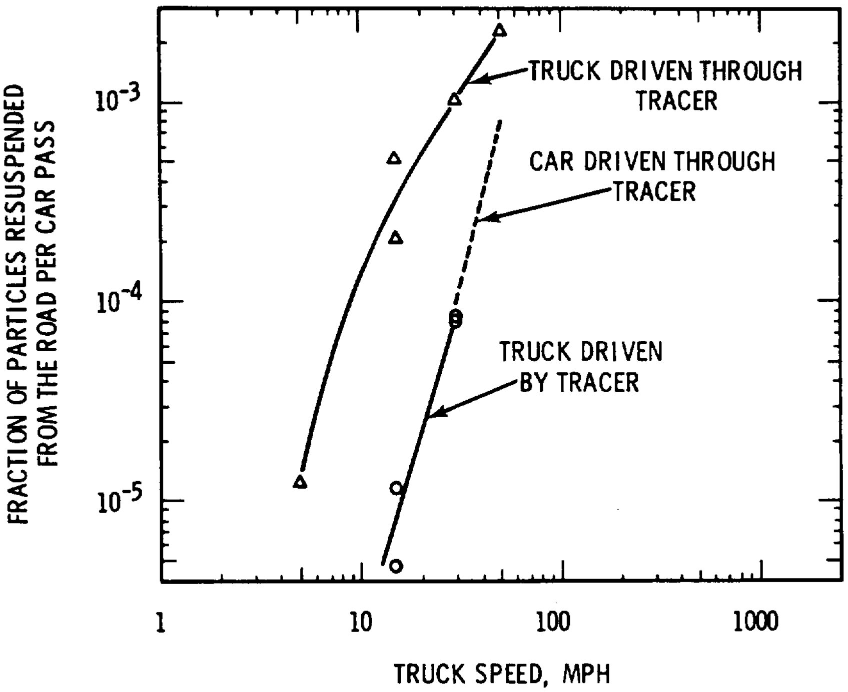

FIGURE 6. Rates of Particle Resuspension Caused by Vehicle Passage Over an Asphalt Road Four Days After Particle Deposition

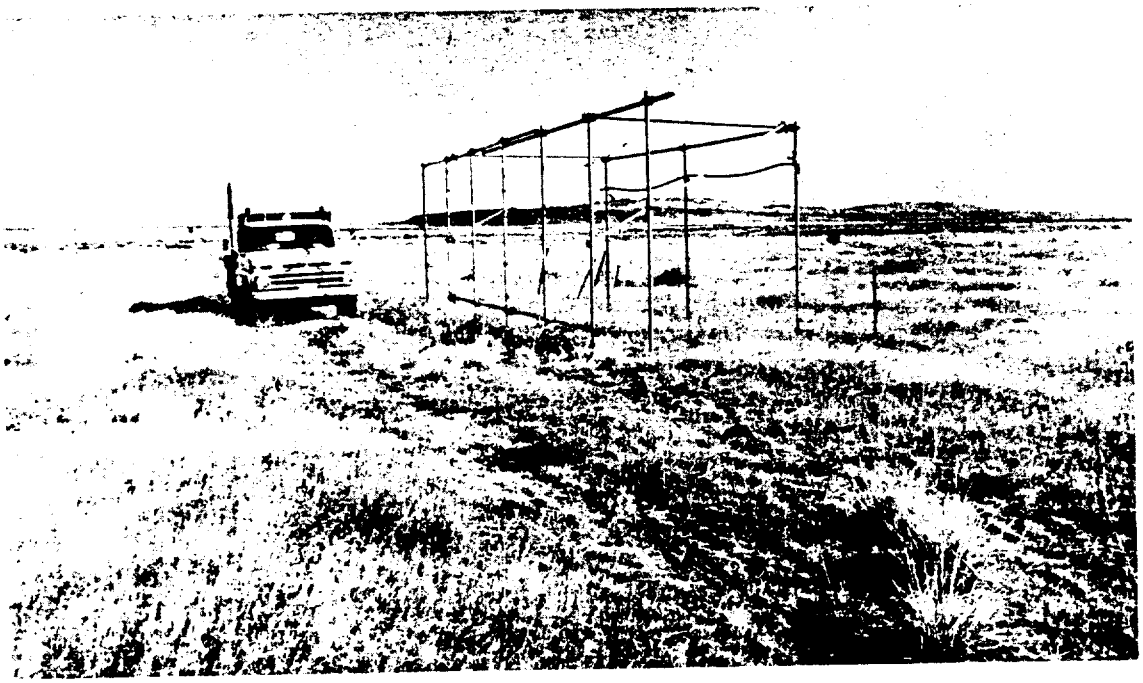

FIGURE 7. Cheat Grass Road Resuspension Site

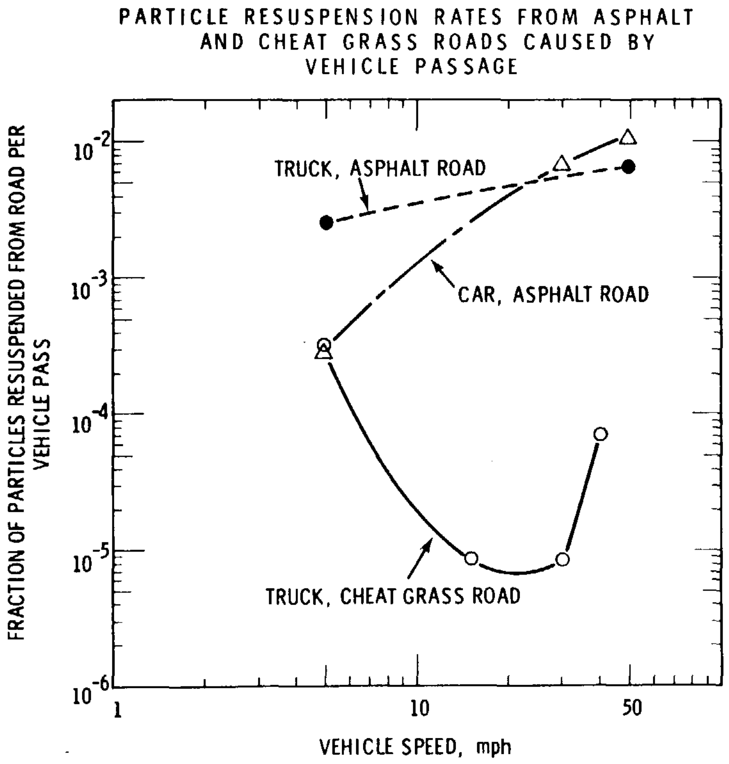

FIGURE 8. Rate of Particle Resuspension Caused by Vehicle Passage Over Asphalt and Cheat Grass Roads

down the length of the tracer lane. With wind speeds of 3 to 4 m/s, pedestrian resuspension rates were from 1 x 10^{-5} to 7 x 10^{-4} fraction resuspended/pass along the tracer lane. This pedestrian-generated turbulence was greater than wind stresses during the experiment.

Wind-Caused Resuspension

Experimental values of wind-caused resuspension rates of tracer particles from environmental surfaces have not been experimentally determined from mass balance techniques other than for the present data (Orgill et al., 1976; Sehmel, 1975b; Sehmel and Lloyd, 1972, 1975b, 1976b, 1976c). Some data were initially obtained using 8-μm mass medium diameter (MMD) ZnS particles and average wind speeds from 1 to 5 m/sec. More extensive data as a function of wind speed were obtained using submicrometer $CaMoO_4$ particles. Average resuspension rates for ZnS particles were measured for resuspension from an asphalt surface (Sehmel and Lloyd, 1972) and a cheat grass surface (Sehmel, 1976). For average wind speeds of 1 to 4 m/sec, wind resuspension rates from an asphalt surface ranged from 5 x 10^{-9} to 6 x 10^{-8} fraction resuspended/sec. For average wind speeds of 1 to 5 m/sec, wind resuspension rates from a cheat grass surface ranged from 5 x 10^{-9} to 6 x 10^{-8} fraction resuspended/sec.

Wind-caused resuspension was measured for submicrometer $CaMoO_4$ particles deposited in a lightly vegetated area on the Hanford reservation shown in Figure 9. Tracer particles were deposited in a circular area of 23-m radius around the centrally located air sampling tower. Resuspended particles were measured at the tower as a function of wind speed increments for respirable particle diameters and for nonrespirable particles at all wind speeds. Respirable particles were collected within particle cascade impactors (Figure 1) while nonrespirable particles were collected by impaction and gravity settling within cowls.

Wind-caused resuspension rates are shown in Figure 10 as a function of wind speed. Resuspension rates ranged from about 10^{-11} to 10^{-7} fraction resuspended/sec. Different functional dependencies of resuspension rates on wind speed can be obtained from these data, depending upon which set of wind speed increments are used. During the January to February period, air sampling was for large wind speed increments, while in subsequent experiments wind speed increments were smaller. The straight lines shown in Figure 10 were drawn through all data points. In these cases, resuspension rates increased with the 1.0 to 4.8 power of wind speed. However, when only data points for smaller wind speed increments are used, wind-caused resuspension rates increased with wind speed to the 4.8 power for 7, 3.3, 2.0, and 1.1-μm-diameter particles as well as for the smaller particles collected on the cascade impactor back-up filter.

For comparable wind speed increments, tracer resuspension rates were nearly independent of the time the tracer was on the ground surface. However, it

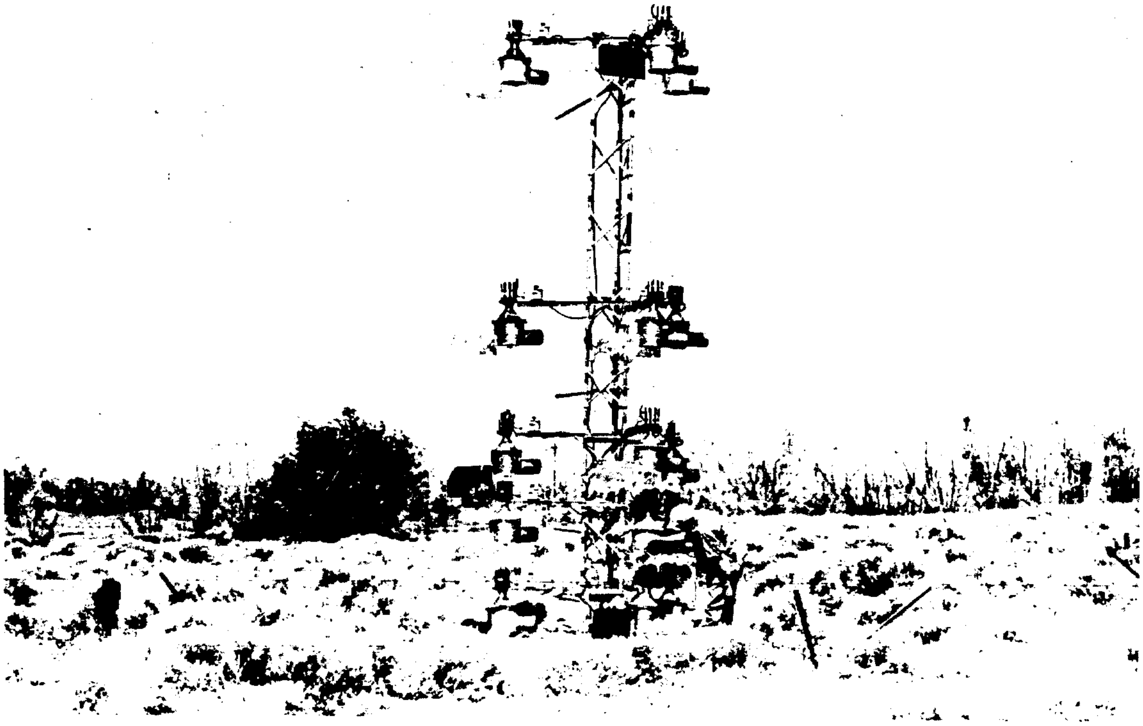

FIGURE 9. Wind-Tracer Resuspension Site on the Hanford Reservation

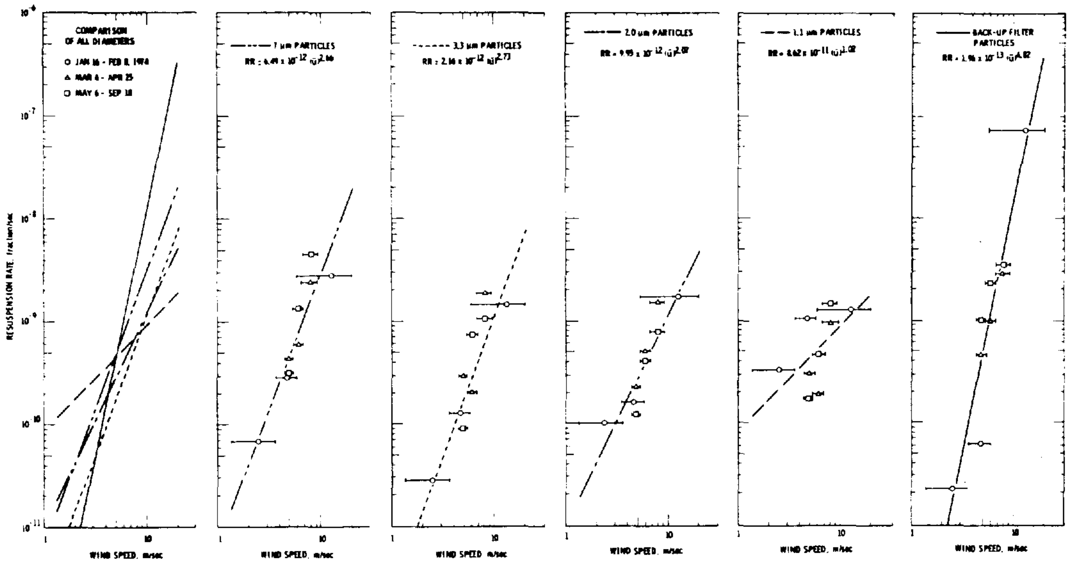

FIGURE 10. Resuspension Rates as a Function of Particle Diameter and Wind Speed Increments

is often assumed in theoretical modeling that particles become less available for resuspension with time. The assumption in these models is that pollutant particles become fixed or attached to soil particles and subsequently "migrate" into the ground surface. This process is called weathering.

The independence of wind-caused resuspension rates with time is a significantly different observation than some others have made. Radioactive resuspension literature indicates that airborne radioactivity concentrations decrease with a weathering half-life of 35 days (Wilson et al., 1961). In contrast, if there is a weathering half-life for the controlled tracer experiments described above, the weathering half-life must be on the order of years. Some differences in reported weathering might be explained by how air samples were collected. In early work, air concentrations were measured continuously. In contrast, in these tracer experiments air concentrations and hence resuspension rates were measured as a function of wind speed. Even these differences in determining weathering half-lives illustrate how poorly weathering is understood.

Average wind-caused tracer resuspension rates are reported for both respirable and nonrespirable particles in Figure 11. In these cases, respirable refers to all particles collected within cascade impactors while nonrespirable refers to particles collected within cowls. Nonrespirable particle resuspension rates were nearly independent of time and were of the order of 10^{-11} fraction resuspended/sec.

Resuspension rates for respirable particles ranged from about 10^{-11} to 10^{-7} fraction resuspended/sec. These resuspension rates did not decrease with time. For the first two sampling periods, resuspension was measured for all wind speeds. In succeeding experiments, resuspension rates were measured only for wind speeds above 1 and above 4 m/sec. The upper or solid line portion of the respirable particle curve corresponds to resuspension rates calculated for the wind sampling periods. These periods correspond to wind speeds above 1 and above 4 m/sec. The lower limit of the respirable particle curve corresponds to resuspension rates calculated by assuming resuspension time corresponding to the total time that cascade impactors were in the field (i.e., time included for winds less than 1 and less than 4 m/sec).

Initial Generalized Wind Resuspension Rate Correlation

Guidelines for estimating resuspension rates are needed based upon existing experimental resuspension rate data. An initial correlation was developed from data reported for uranine particles resuspended from a smooth surface (Sehmel, 1975b), ZnS from an asphalt surface (Sehmel and Lloyd, 1972), submicrometer molybdenum tracer from a vegetated desert soil (Sehmel and Lloyd, 1976b), and for DDT from a forest (Orgill et al., 1976). Each of these surfaces has a much different estimated surface roughness height, z_0, ranging

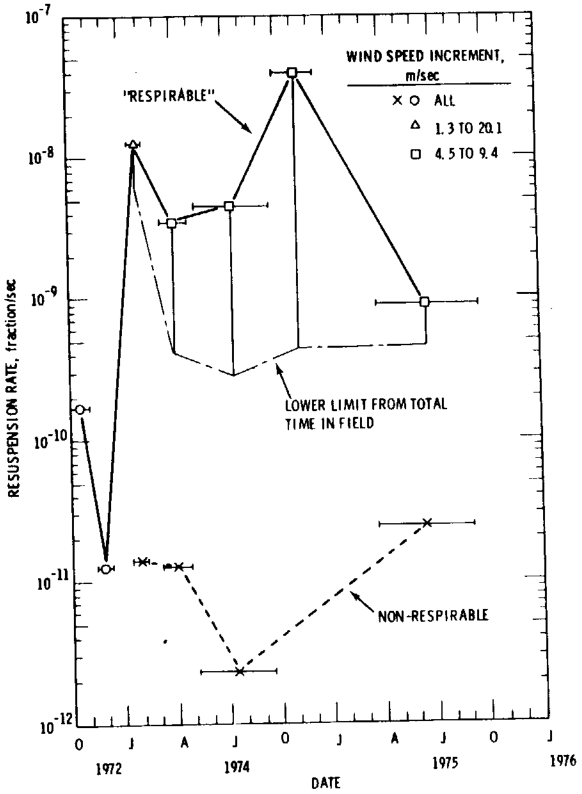

FIGURE 11. "Average" Wind Caused Resuspension Rates (Lightly Vegetated Desert on Hanford Reservation)

from 4 x 10^{-3} cm for the smooth surface to 1 m for a forest. Roughness height is calculated from the log-linear velocity profile and is the height at which the extrapolated velocity profile reaches zero velocity.

Ranges of measured average resuspension rates are correlated as a function of measured or estimated surface roughness heights in Figure 12 (Sehmel, 1975b). Resuspension rates range seven orders of magnitude from 10^{-10} to 10^{-3} fraction resuspended/sec. The practical significance of these numbers can be made apparent by noting that a year is 3.2 x 10^7 sec.

The initial resuspension rate correlation indicates that resuspension rates decrease as surface roughness increases, at least for the three smaller roughness heights. However, measured resuspension rates for DDT sprayed on a forest are two orders of magnitude greater than rates for the desert soil (Orgill et al., 1976). This is an unexpected and unexplained increase in resuspension rates. A possible explanation of the increase might be increased resuspension caused by tree movement in the wind. Also, various other gross differences in controlling variables and experimental factors may have influenced results. Since the data are so extremely limited, this apparent correlation should be used with extreme caution until correlations are developed based upon several physical parameters instead of only z_0. Nevertheless, this correlation does give some justification for estimating resuspension rates until better correlations are developed.

AIRBORNE SOIL CONCENTRATIONS AS A FUNCTION OF WIND SPEED

One resuspension modeling concept is that deposited pollutant particles lose their identity as discrete particles, becoming attached to host soil particles when resuspended (Sehmel, 1975a). If the physics of soil resuspension were understood more adequately, it might be anticipated that the soil particle resuspension models could be used as a simple basis for developing predictive contaminant particle resuspension models. Consequently, soil resuspension was investigated by experimentally determining rates at which airborne dust concentrations in the respirable size range increased with increasing wind speed. Experiments were conducted as a function of wind speed on the Hanford reservation at three sites.

Increases in airborne particle number distributions as a function of wind speed are shown in Figure 13 for particle diameters of 1.1, 1.9, and 3.6 μm. In each case, data points are plotted at the lower value of each experimental wind speed increment. Wind speed increments are indicated by the horizontal lines extending to the right from each data point.

Average airborne particle number concentrations increased rapidly and reproducibly with increases in wind speed. In the left portion of the figure, concentrations increased as the 2.9 power of wind speed for wind speeds between 4.5 and 9.4 m/sec during sampling periods beginning on March 3 and May 6. Concentrations in the summer months from May 6 to September 18, 1974 were greater than for the spring months from March 4 to April 25, 1974, but were less than for late winter months from January 16 to February 8, 1974.

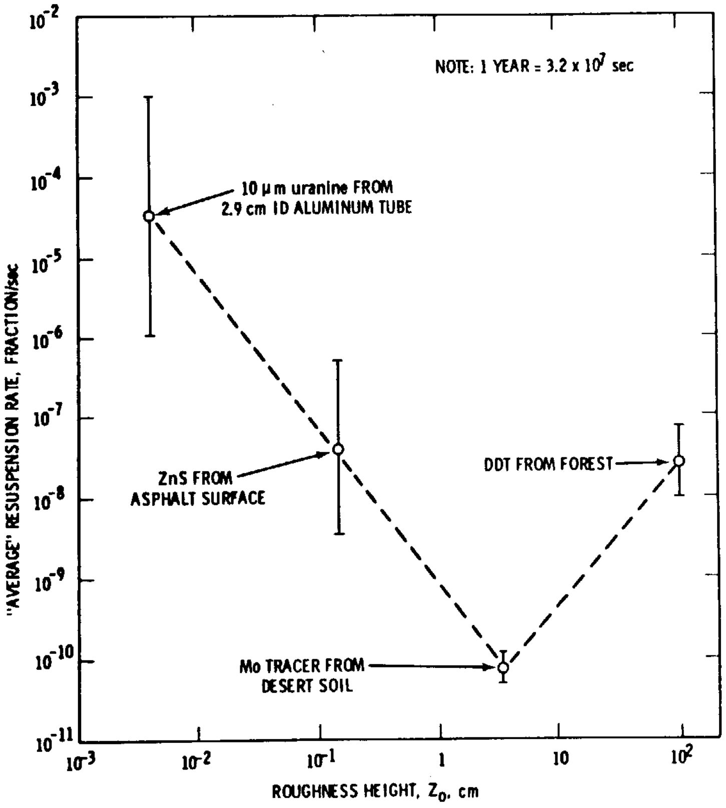

FIGURE 12. Initial Correlation of Wind Caused Resuspension Rates

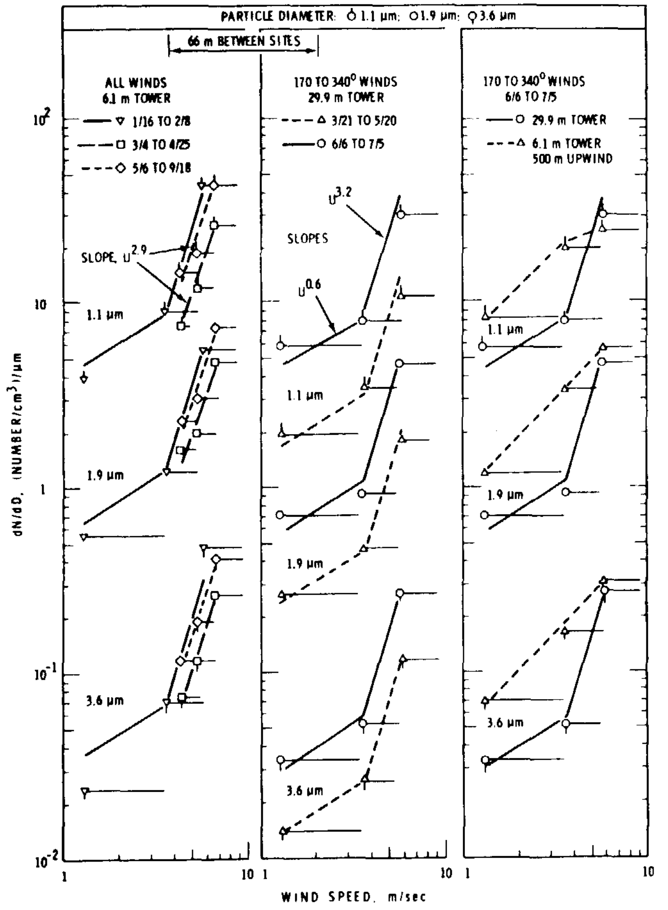

FIGURE 13. Airborne Soil Particle Concentrations as a Function of Particle Diameter, Wind Speed, Sampling Season, and Sampling Site, 1974

The remaining particle number concentration data in the three sections of Figure 13 (including the January 16 data) were determined using larger wind speed increments for air sampling. The solid lines in each of the three portions of the figure were calculated by least squares techniques. These techniques minimized data point deviations around solid lines while retaining a constant slope. Airborne soil concentrations increased with either the 0.6th power (below 4.5 m/sec) or 3.2nd power (above 4.5 m/sec) of wind speed. The differences in exponents might be indicative of some threshold wind speed near 4.5 m/sec above which soil more readily becomes airborne.

Differences in air concentrations were measured as a function of sampling tower separation for the June 6 to July 5, 1974 data shown in the right portion of the figure. In this case, air was sampled simultaneously at a 500-m separation for 170° to 340° winds (winds from the west are 270°). Upwind concentrations were greater except for the 1.1-μm particle data point at a wind speed of 5.8 m/sec. The significant difference is that at the upwind tower airborne concentrations increased with the first power of wind speed while at the downwind tower airborne concentrations increased with wind speed to the 0.6 and 3.2 power. Reasons for the differences between upwind and downwind concentrations can only be hypothesized at present. The differences are probably caused by different wind speeds at the upwind and downwind towers, deposition of particles in the area between towers, and, probably of most significance, the greater area of open soil available for resuspension adjacent to the upwind tower.

Average airborne mass loadings were calculated for each wind speed (total collected within impactors). Dust loading in μg/m^3 are shown as a function of wind speed in Figure 14. Dust loadings ranged from 13 to 360 μg/m^3. As shown for two lines representing sampling times from March 4 to April 25, 1974, and May 6 to September 17, 1974, mass loadings for respirable particles increased with wind speed to the 2.9th power for wind speeds greater than 4.5 m/sec.

CONCLUSIONS

Particle resuspension rates are a function of at least wind speed and mechanical disturbances. Mechanical disturbances such as vehicular traffic or a man walking can cause high local resuspension rates. In comparison, average wind resuspension rates from a local area could be less important per unit area than local mechanical disturbance resuspension. However, wind-caused resuspension rates apply to the entire contaminated area. If one were to compare relative resuspension from wind-caused and mechanical disturbances, one would need to know the total surface contamination area for wind resuspension versus small localized surface contamination levels

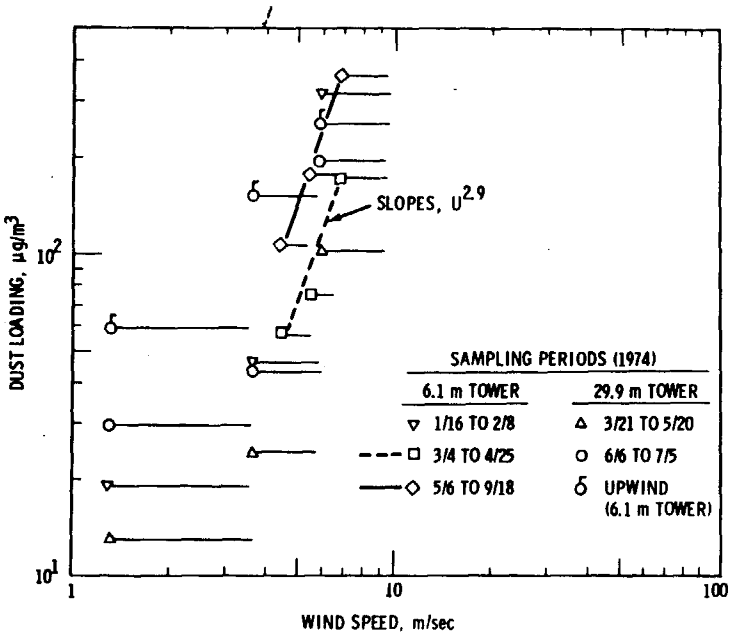

FIGURE 14. Airborne Dust Loading Collected Within Impactors as a Function of Wind Speed

for mechanical disturbance resuspension rates. Both mechanisms, however, do resuspend and transport potentially hazardous respirable particles.

Resuspension rates are needed for inclusion as source terms in atmospheric diffusion and transport equations; however, model predictions are no better than the uncertainty in the source data. In the case of resuspension rates, uncertainties are very large. Much research is yet needed to develop resuspension models to predict particle resuspension rates for any situation.

Wind-caused resuspension rates from a sparsely vegetated area have only been measured with submicrometer tracer particles. The potential effect of different particles diameters on resuspension rates is unknown. It might be hypothesized that similar results would be expected for other submicrometer particles of interest since submicrometer particles are probably attached to host soil particles when the particles are resuspended. If the particles of interest were much larger, it is unknown whether the particles would be resuspended attached to host soil particles or resuspended as discrete particles.

The change in airborne concentration of a pollutant as a function of time is often attributed to a weathering half-life, the fixation of the pollutant particle into the ground surface soil. For radioactivity, the weathering half-life was on the order of 35 days. In contrast, weathering half-lives for respirable tracer particles are now estimated here as being on the order of years. Predictions using weathering half-lives of months versus years could have a significant implication in environmental hazards evaluations. At the present time, credit for decreased airborne radioactivity from resuspension could be attributed to a weathering half-life of months. If a weathering half-life of years were applicable, the potential downwind inhalation hazard from resuspended particles would be significantly increased. Additional experimental data are needed to determine what weathering half-life should be used in hazards evaluations.

Vegetation on a resuspension surface will decrease resuspension rates. This is rather obvious, but can be definitely concluded from decreased tracer particle resuspension rates for vehicles driven on a cheat grass area compared to an asphalt area. Since vegetation does decrease resuspension, it should be retained on all areas of potential surface contamination and existing surface contaminated areas until constructive cleanup operations can be initiated.

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A REVIEW OF RESUSPENSION MODELS

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ABSTRACT

Resuspension is classified, according to the prominent path of exposure and the type of consideration involved in estimating concentrations, as general resuspension, local resuspension, and transfer resuspension. General resuspension can be driven by winds or mechanical disturbances while local resuspension and transfer resuspension are primarily caused by mechanical disturbances. Modeling techniques consist of the resuspension factor, the resuspension rate and mass-loading. For general resuspension the resuspension rate is preferred while the resuspension factor or mass-loading approach can be used for local resuspension. Data for estimation of transfer resuspension are not really adequate to even make a first approximation as to its importance.

Resuspension of contaminants on the ground leading to inhalation is generally assumed to be an important pathway of exposure to people. Methods of calculation for prediction of this pathway (models) are used widely in Environmental Impact Statements, risk assessments, and derivation of standards. In recent years there has been a new surge of interest in this pathway with many experiments and theoretical studies bringing us closer to an understanding of the phenomena involved and providing improved data for input to calculations. There is, however, much to be learned before reasonably accurate predictions for various areas of the country will allow us to eliminate the conservatism built into most predictions of resuspension. In the following material I discuss some of the basic concepts and calculational methods now in use.

I have usually differentiated resuspension into two categories: general resuspension and local resuspension. General resuspension is that which results in concentrations over an area and, in which, the meteorological transport parameters play an important role. This category can be further subdivided by the driving force for the resuspension: wind or mechanical disturbance. Local resuspension occurs as a result of a localized disturbance of the soil causing material to become airborne. The exposure is to only one or a few individuals in the immediate vicinity of the disturbance. In general, meteorological conditions play a minor part. An illustration of local resuspension would be an individual digging in the area and producing a cloud of dust in his breathing zone. An interesting variation, which should be given a different name such as "transfer resuspension," is the movement of contamination from the soil to another object from which an increased probability of inhalation may occur, usually at a later time. An example of this is the individual digging in the ground, contaminating his clothing, with the contamination released by the movement of the fibers in a region close to the nostrils. An extreme example would be pulling a contaminated shirt over one's head.

In the application of resuspension data to various problems, three primary models have been used: the resuspension factor, the mass-loading of the air, and the resuspension rate. These models are discussed briefly before noting the appropriate use.

The resuspension factor is the ratio of the concentration of contaminant in the air to the quantity of contaminant per unit area at the location where the air is sampled. Thus, the value is in length^{-1}, usually m^{-1}. It is the oldest of the descriptive models. Although frequently attributed to Langham, it was used in the late 40's with numerical values obtained from data on dust in the air during demolition operations in the rebuilding of London. Its advantages are its simplicity and the fact that nearly all of the information that has been gathered up to a few years ago has been expressed in this form. Thus, values are available for use. Its disadvantages are 1) the lack of a built-in classification as to the type of terrain and cause of the resuspension, and 2) the denominator expresses only the very local condition while the numerator, for general resuspension, includes the integrated effect from upwind. Thus, if the area upwind contains a "hot spot" the resuspension factor will be high. The lack of classification is well illustrated by Mishima's tabulation (Mishima, 1964) where the values vary by 13 orders of magnitude. However, in addition to the apparent change in resuspension with aging, some of the results represent wind resuspension, others mechanical resuspension upwind and, still others, local resuspension from automotive traffic. For more closely defined activities one should be able to reduce the range of values for these activities by a wide factor.

The mass-loading approach considers the contaminant in the air to be directly associated with the soils in the immediate region. The air concentration is estimated by multiplying the concentration contaminant in the soil by the concentration of dust in the air. Its advantage is, again, the simplicity of application. However, for general resuspension, it suffers from much the same difficulties as the resuspension factor in describing the effect of a source upwind. One problem, which is not well defined, is the potential partitioning of the plutonium in the soil particles. Not all particles have equal probability of resuspension and the particle size distribution in the air is distorted by preferential deposition. These can lead to distortion even if particle size analysis is done. In spite of these difficulties, however, Anspaugh et al. (1975) have shown some interesting correlations with total soil content.

The first documented use of the resuspension rate was in a paper in the second Geneva Conference (Healy and Fuquay, 1958). The resuspension rate describes the fraction of the material on the ground that becomes airborne under the influence of a given action. This provides a "source term" or the quantity entering the air per unit time, usually per second. The dispersion downwind and subsequent deposition can be obtained by use of the meteorological correlations which are used in dispersion calculations. The major advantage of the resuspension rate is that it permits realistic estimates of the contribution from areas upwind. Such estimates are not simple, however, since they involve numerical integration over a predetermined deposition pattern. Disadvantages, aside from complexity, are primarily the lack of accepted values for the resuspension rate in different terrains and the variation of this rate with such variables as wind-speed or soil conditions. Data are being obtained, however, primarily through the work of Anspaugh (Anspaugh and Phelps, 1974) at the Nevada Test Site, Sehmel (Sehmel and Lloyd, 1976) with tracers at Hanford and plutonium in the Rocky Flats area, and Gillette (Gillette and Blifford, 1974; Gillette et al., 1972) in studies of the resuspension of soil in agricultural fields. It is believed that future extension of this work will permit choosing realistic values for application in other areas. It should be noted, however, that this is a far more sophisticated approach than the resuspension factor in that it requires knowledge of the contamination contours, the characteristics of the soils in the area, and the detailed meteorological patterns. Thus, it is doubted that a "handy-dandy" rule of thumb to be applied to only a few measurements will ever be derived from this concept.

Earlier we mentioned the types of resuspension. We will now discuss the application of the basic models to these types. Before that, however, I would like to recall a long series of discussions with Wright Langham concerning the relative merits of the resuspension factor and the resuspension rate. I had always felt the resuspension factor to be too simplistic in its concept and provided unreal answers. Finally, Wright, in his pragmatic manner, replied that he didn't see what the fuss was all about. If the resuspension factor is measured under the worst conditions of soil disturbance, it will give a safe answer. I could not really answer him since, for use in this manner, he was correct.

For general resuspension, which implies the estimate of ambient concentration from material arising from an upwind area, the resuspension rate should be the method of choice. This technique permits the concentration at different positions in and around the contaminated area with non-uniform deposition to be estimated and accounts for different meteorological and soil conditions. It is of interest that a large portion of the values for the resuspension rates now available have been measured for short time periods so that the variations in meteorological conditions during the period of the measurement are minimized. However, in application to radioactive materials, our interest is primarily in the long-term average. Thus, predictive calculations should be done using a wind rose which gives the joint probability of direction, stability, and wind speed. The wind speed is of particular importance in wind resuspension since there now appears to be clear evidence that the pick-up rate (or source term) varies as a multiple power of the wind speed, apparently on the order of the third power in relatively undisturbed areas such as the GMX area in Nevada or Rocky Flats, and as powers up to the ninth, and perhaps higher, in agricultural fields after plowing. I have made a few calculations using the wind rose in Meteorology and Atomic Energy (Slade, 1968) for the concentration down-wind from various sizes of square areas of uniform contamination. These results show that with the contaminated area varying by a factor of 10^4 to 10^5, the concentration close to the edge of the area varies by only a little more than one order of magnitude. At distances great enough that the source may be considered a point source the variation in concentration is about in the same ratio as the source size.

In addition to wind-driven pickup, general resuspension can occur from mechanical disturbance of the area. For this process it appears reasonable to assume that the rate of resuspension is a function of only the type and severity of the disturbance with the dispersion downwind a function of the meteorology. Here, again, the time of averaging is important. However, the time of averaging can also define the shape and size of the source as well as the effective resuspension rate and dispersion. For example, a contaminated dirt road can serve as a source as traffic passes over it. Averaging in time gives this as a line source. If the average traffic is 10 cars per day then the daily resuspension rate is ten times that of one car. A farmer plowing a field will at each instant of time represent a point source. As he passes the length of the field, these point sources will represent a line source averaged over the time of passage. Each line integrated with all others will eventually describe an area source with the resuspension rate equal to the point source rate divided by the time taken to plow the field. In averaging over a one year period, the average resuspension rate will be further decreased by the ratio of the time to plow the field to one year.

Values for the resuspension rate or experiments well enough documented to permit derivations are scarce. Sehmel (1976) has studied the resuspension rate from traffic on asphalt roads and on areas covered with cheat grass. On the asphalt road the values ranged from 10^{-8} to 18 x 10^{-3} sec^{-1} with the rate proportional to the speed of the vehicle. He noted that the depletion of the contaminant with any significant traffic would be rapid. On the cheat grass, no consistent pattern developed since the rate for the first and slowest truck was higher than subsequent ones presumably due to the rapid resuspension of the material deposited on the cheat grass. Several studies of agricultural operations at Savannah River (Milhane, et al. 1976) and Lawrence Livermore Laboratory (Myers et al. 1975) permitted rough estimates of resuspension rates. Thirteen of the values were on the order of 10^{-8} to 10^{-7} sec^{-1} while about five were higher with a maximum of 1 x 10^{-6} sec^{-1}. Calculated estimates, using

integration over areas up to 2000 meters on a side indicated that the mechanical disturbance can be of greater importance than wind resuspension in areas where sizeable disturbances occur, even assuming that the disturbance is carried out only for 40 hours per year.

Local resuspension applies to the concentrations in the breathing zones of individuals producing resuspension by mechanical disturbance. Either the resuspension factor approach or the mass-loading approach appears to be useful and appropriate in this case. The localized air concentration from the small area of the disturbance can be used meaningfully without the uncertainties from upwind areas. However, it is necessary to categorize the types of actions and their durations to match the categories of actions leading to the given resuspension factor. The mass-loading approach can be used along with estimated or measured dust concentrations in the air. Again using the time of 40 hours per year as appropriate to a disturbance with a resuspension factor of 5 x 10^{-7} m^{-1}, the local resuspension may be of greater importance than wind resuspension.

Transfer resuspension could, in theory, be estimated from data on the amount of contamination transferred to clothing or other objects and the subsequent transfer of this contamination to the body. However, the quantity and quality of such data are not adequate to address this problem in any quantitative fashion, particularly when the large number of actions possible are considered.

We have attempted to get some feeling for this by looking at the data on lead in the environment and people, particularly children. Lead is a widespread element used in many ways by man and occurs as a waste in many smelting operations. Lead poisoning is a common ailment. There is a considerable body of information on exposures from dust or soils in the vicinity of smelters or in metropolitan areas where the lead has been released from gasoline fumes.

Unfortunately, it is impossible to obtain a direct correlation between environmental concentrations and intake because lead is absorbed from the GI tract, particularly in children, and one cannot distinguish between inhalation and ingestion. Further, there is no well-established correlation between lead in the blood, the common method of measurement, and intake. There have been several studies in cities which indicated that the ingestion of paint chips could not account for the observed high blood levels. (Lepow et al. 1975; Sayre et al. 1974; Vostal et al. 1974). From measurements of the dirt and lead on children's hands, there was a clear relationship between these values and the household levels. Further, the blood levels did not increase until the child reached the crawling age. It was concluded that the lead in the dust of the home or area was transferred to the child. Several observations indicated that the lead in household dust was in concentrations about ten times those in the soil outside the house. However, this may not be generally true since it will depend upon the source of the lead.

On the other hand, more detailed studies indicate the soils and dust not to be important. For example, Ter Haar (1974) used ^{210}Pb in the environment as a tracer for lead and noted that the ^{210}Pb content of the excreta from children with signs of exposure and those with low exposure was identical. He concluded that dust and air-suspended particles were not the source of lead in these urban children. Barltrop (1974) measured the soil, blood levels of mother and child, and feces from children in two towns with natural lead levels of about 900 μg/g and 400 μg/g. In a further preliminary study he compared two villages where the soils were 10,000 ppm and 500 ppm. He concluded, "The results of our studies to date suggest that local soil lead levels of the order of 10,000 ppm are without major significance and that on present evidence the recent concern with regard to contaminated soils in cities is not well founded."

These results combine the direct ingestion and inhalation pathways. It is, however, encouraging to note that those who have directly studied the relation between quantity in the children and quantity in the soil, indicate that there appears to be no great problem. However, we will only be able to tell this unequivocally with plutonium when greater attention is paid to this problem and appropriate measurements are made.

In the foregoing, I have considered the models in a somewhat idealized fashion in that I have ignored many factors of importance in the actual application. For example, in the general resuspension model, I have ignored the very important factors of soil condition, coverage of the soil by vegetation or rocks, terrain influences on dispersion, and the estimation of deposition. The latter two are problems which relate to the estimation of dispersion and are receiving attention but will result in more complicated analyses. The first two require more studies of resuspension under various conditions and, perhaps, more theoretical modeling of the attachment forces and the influence of soil particle distribution on probability of the particles being removed from the ground. In the local resuspension and transfer resuspension considerably more work is required in defining and measuring appropriate parameters and in making use of opportunities to assess the overall intake from a soil source.

There are two other items in the overall resuspension-inhalation model which could be of some importance in a particular situation. The first is the application of the ICRP Task Group (ICRP, 1966) on lung dynamics model for inhaled material and the second is the importance of sizes of contaminated particles in the soil.

The ICRP Task Group model accounts for the deposition of individual particle sizes (AMAD) in three compartments of the respiratory tract - the nasopharyngeal region, the tracheo-bronchial region, and the pulmonary region. It then describes the movement from these regions to the blood stream and the lymph with different retention times for each compartment depending on the class or solubility of the substance. The later retention and distribution factors have been revised by the ICRP Committee on Plutonium Metabolism (ICRP, 1972).

If one is to use this model it should be applied as given unless specified changes are made. The results of one such application is given in Table 1. where the inhalation to result in 500 mrem/yr to each organ at the end of 70 years continuous exposure is presented. Interestingly enough, the skeleton is the limiting organ in both class Y and class W material with only a factor of about 5 of 6 difference with particle size for particles from 0.1 to 50 μm AMAD. This is due to the increased deposition of larger particles in the upper respiratory tract and the significant rapid absorption prescribed for these regions. Obviously one can prescribe a third class in which the movement from the lung is slower and the absorption from the upper regions smaller so that the lung becomes critical. Regardless of the constants used, it is apparent that one cannot completely ignore those particle sizes above the "respirable" range unless it has been shown that the material in the soil has a solubility such that movement from the upper respiratory tract is negligible. Thus, in any detailed analysis, it is not proper to use the current MPC's in air for soluble material based on only absorption to the blood stream and for lung based only on dose to lung. The true situation is a combination of both.

The particle size question has been intensified by Johnson et al. (1976) who advocate applying the Colorado limit for plutonium in soils only to the soil particles less than 5 μm obtained by completely breaking up the aggregates. In an earlier report (Healy, 1974) I recommended that the limit be applied to the very surface of the soil and to particles less than 100 μm. This was done in an attempt to use data on resuspension rate from one experiment in a more general manner. Now that actual resuspension rate data are available, this assumption is no longer necessary.

One objection to the particle size proposal is given by the interpretation of the ICRP lung model in Table 1. Unless it is shown that the plutonium in the soil is more insoluble than the ICRP assumes, then the contribution of the larger particles from either general resuspension or local resuspension is of importance.

When one selects a particular particle size to estimate a hazard, it is assumed that only this particle size is of importance and all others may be neglected. Let us see the effect of this on a local resuspension situation which occurs for 40 hours per year with a disturbance for which a resuspension factor of 5×10^{-7} m^{-1} is appropriate. We will use d as the depth of soil appropriate to the resuspension factor, ρ as the soil density, and f as the fraction of the particles of interest by weight in the soil. I have used an air limit of 3.5×10^{-14} μCi/ml or 0.035 pCi/m^3 averaged over the year. If we further assume that all of the plutonium in the soil is in the small particles, then the minimum limit for the small particles and the soil considered separately is:

$$\text{pCi/g} = \frac{1500}{\rho f d}$$

Some representative values are given in Table 2.

Table 1. Quantity of Pu Inhaled per Day to Result in 0.5 rems/yr after 70 Years (pCi)

Particle Size-AMAD	Class Y			Class W		
	Skeleton	Liver	Lung	Skeleton	Liver	Lung
µm		(pCi/day)			(pCi/day)	
0.1	0.9	1.5	2.4	0.57	0.94	24
0.2	1.1	1.8	3.0	0.65	1.1	30
0.5	1.5	2.4	4.0	0.70	1.2	40
1	1.7	2.7	4.8	0.67	1.1	41
2	2.0	3.2	6.0	0.61	1.0	60
5	2.7	4.5	10	0.57	1.0	100
10	3.2	5.3	13	0.57	0.94	130
20	4.4	7.2	24	0.58	0.97	240
50	5.0	8.2	40	0.55	0.91	400

Table 2. Soil and Small Particle Limits

Sampling Depth	% Small Particles	Limit for Small Particles	Limit for Total Soil	*Air Conc.
cm		pCi/g	p/Cig	$\mu g/m^3$
1	1	75,000	750	100
	10	7,500	750	1,000
	30	2,500	750	3,000
5	1	15,000	150	500
	10	1,500	150	5,000
	30	500	150	15,000

*During disturbance

As can be seen, the limiting value in the soil changes with the amount of fines while the total soil value remains the same. This is the result of our assumption of a constant resuspension factor under all conditions of the soil.

In fact, the concentration in the air is a result of both the activity on the air-borne soil and the concentration of air-borne soil in the air. Data are not available to indicate whether the dust concentration in the air is a direct function of the fractions of small particles in the soil under different stimuli for resuspension. When one considers the extremes of this case, a clay bank of all small particles or a soil consisting of sand with only a few small particles, the hypothesis appears questionable. However, only detailed experimental data, not assumptions, will clarify the relationship.

Let me finally give a few thoughts on models and modeling. The biggest problem in applying any model, particularly a complex one, is in obtaining estimates of the proper parameters. These cannot rely only on one set of experiments, even if done in an area of interest, but must include consideration of as much of the published data as possible. This leads me to the conclusion that modeling is not a simple mathematical exercise in which others supply numbers for given phases of the problem. The modeler must be knowledgeable about the information he is using and apply it with full knowledge of its strengths and weaknesses. To me, a strict requirement in any modeling exercise should be a justification of any parameter or assumption in sufficient detail so that the reader can understand both the strengths and the weaknesses of each number.

In any modeling exercise, there are many unknowns or poorly-knowns. An important function of the exercise is to define these items and to identify those of critical importance so that they may be investigated. Again the approach should be one of increased knowledge about the subject and not a simple search for a number. Such numbers are normally widely variable and the modeler can select one suitable to his purposes only on basis of complete knowledge of the information available and the field in which he is working.

A final problem in modeling is the tendency to believe the results. Let me end with a quote on this furnished by Stan Auerbach which summarizes what I would say, however in a much improved manner over what I can muster. "The result is a work of art, sometimes good, sometimes bad, but almost always giving us, the creators, a feeling of euphoria. This euphoria often lives on for some time unless we make the mistake of comparing our creation with God's. For, unfortunately, He rarely agrees with us, and He is, by definition, always right." (Passioura, 1973).

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BRIEF SURVEY OF PARTICLE WORK RELEVANT TO THE TEST SITES

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ABSTRACT

This paper presents an overview of the available data on the physical, chemical, and radiochemical properties of fallout particulates, of the methods by which both samples and data have been obtained and their limitations, and of the availability that remains mostly uncorrelated, but from which much information can be gleaned or inferred that is of interest to the NAEG program. Nevertheless, experimental characterization of particulates with special emphasis on transuranics is not only still possible but advisable.

INTRODUCTION

This paper is a condensation of a report to ERDA's Nevada Operations Office containing an overview of the available data on the physical, chemical, and radiochemical properties of fallout particulates, of the methods by which these data have been obtained and their limitations, and of the availability and usefulness of residual samples. This overview is limited to those nuclear device tests that have generated measurable local fallout. Thus, consideration of safety tests, for example, is included, but consideration of free airbursts is limited only to what appears relevant as a result of their place in the height-of-burst spectrum.

The information presented has been gleaned primarily from review of data compilations, from the author's own experience, from a few other documents that were readily available, and from personal contacts. Since 1965, data for some of the pre-1958 shots have been developed primarily by the author and mostly from the residual cloud samples, particularly with regard to particle size distributions and radioactivity, these data having been correlated to a great extent. Other information from shots has been developed by several investigators.

Initiation of this project is particularly timely. Since the effective cessation of nuclear testing resulting in releases of radioactivity into the atmosphere many years ago, the community of individuals who are familiar with debris sampling and analysis, and with the systematics of physical, chemical, and radiochemical properties of the debris has become well-dispersed and has effectively decreased considerably. In addition, the selection and availability of such samples has decreased. If additional work with samples still available is to be accomplished, such work should not be postponed.

DEVICE SHOTS

The nature and properties of the particulates formed as a result of explosions involving nuclear materials depend on the type of device, but mostly on the detonation conditions and the substrate. Thus, one may distinguish tests on the basis of the height of burst (HOB), or better, on the basis of the scaled height of burst (SHOB). Although not entirely correct, one may use a $(\text{yield})^{-1/3}$ - scaling law, so that the scaled height of burst is:

$$(\text{SHOB}) = (\text{HOB})(\text{Yield})^{-1/3}$$

where the height of burst may be expressed either in meters or in feet, and where the yield may be expressed in kilotons or in megatons TNT-equivalent. On the basis of height-of-burst (depth-of-burst) differences, one may distinguish between:

Underground (fully contained) bursts (of no interest, except when early venting occurred);

Cratering (partially contained) bursts, including very shallow bursts; surface bursts (HOB)<5 m, scaled to 1 kt);

Near-surface bursts (5 m <(HOB <130 m, scaled to 1 kt including tower shots and many balloon shots);

Free airbursts (HOB >130 m, scaled to 1 kt; of no interest) including upper atmosphere bursts (no surface material swept into the cloud.)

The substrate or medium also varies considerably. On this basis, one may distinguish between:

Underwater bursts (of no interest);

Water surface bursts (Pacific Proving Grounds--of marginal interest);

Shallow-water surface bursts, on a barge (Pacific Proving Grounds--of interest only for evaluation of Marshall Islands);

Bursts on coral (with or without a steel tower);

Bursts in or on alluvium (NTS, with or without a steel tower);

Bursts on hard rock.

In some cases, the substrate is partially modified by the presence of large quantities of added materials, such as concrete or steel.

The properties of the particulates are also affected by the nature of the shot:

Nonnuclear events involving nuclear materials; actual nuclear detonations involving fission or fusion or both.

Device composition also is a variable. The overall radionuclide spectrum is (neutron spectrum) and the fissionable material used. In addition, however, the specifics of the resulting transuranic distribution depends to some extent on whether Pu was present in the device as fuel, or formed from U through Np by neutron capture.

SAMPLING

Since the validity of an analysis can be no better than the sample analyzed, sampling must be considered in some detail.

A great variety of sampling methods and of samplers has been used, sometimes deployed in specific arrays. What was used depended upon the type of sample desired, but also upon the state of the art. A survey of samplers and sampling methods used over the years is shown in Table 1. All of the samples taken constituted only a very small fraction of the cloud, or of the total amount of radioactive and nonradioactive debris lifted. Fallout samples from Bravo, an approximately 10-Mt ground-surface shot of the 1954 Castle series, may have contained as little as 10^{-12} or less of the amount lifted. Cloud samples collected by aircraft may have contained from 10^{-10} to 10^{-14} of the total cloud. Thus, no sample taken was likely to be completely representative of the cloud or of the fallout. The considerable inhomogeneities that exist in clouds also would generally preclude representative samples from having been obtained.

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*This quotation is from the unclassified abstract of the series.

Table 1. Survey of Samplers and Sampling Methods

- I. Fallout
  - 1. Trays/surface collectors
    - a. Always-open collector (gross or total collector)
    - b. Open-close collector
    - c. Gross-fallout collector
  - 2. Sequential collectors
    - a. Differential fallout collector
    - b. Incremental collector
    - c. Intermittent fallout collector
    - d. Special incremental collector
  - 3. Sticky surfaces
    - a. Tape fallout monitor
  - 4. Aerosol monitors
    - a. Cascade impactor
    - b. Electrostatic precipitator
    - c. Conifuge
- II. Cloud
  - 1. Filter units (presumably isokinetic)
    - a. ACC type V asbestos fiber
    - b. IPC 1478 impregnated cellulose
    - c. Membrane filters
    - d. Glass fiber
  - 2. Sticky wires
  - 3. Cascade impactors
  - 4. Electrostatic precipitators
- III. Miscellaneous
  - 1. Hi-vols
  - 2. Scoops
  - 3. Resuspension samplers
  - 4. Radar (remote measurement)
  - 5. Core drills

Fallout samplers are either passive or active, with the great majority of samples taken in the passive mode. By active sampling, we mean sampling by means of impactors, precipitators, conifuge, etc. The other samplers mentioned are, then, passive.

The always-open collector remains uncovered during an event. This collector may be a tray, or a bottle with a funnel. With this type of collector, the fallout may become considerably diluted with extraneous dust entering the collector from the base surge and as a result of wind action before, during, and after the actual collection period. Furthermore, no information can be obtained (without parallel and independent appropriate radiological monitoring equipment) about the time of first arrival of fallout particles and the time of cessation of fallout.

An improvement is the gross-fallout collector, consisting of a bottle with a funnel equipped with a lid that automatically opens at shot time. This lid is then replaced about 12 hours later. A further improvement is the open-close collector which not only can be automatically opened, but which also can be automatically closed. However, some dilution of fallout particulates still occurs even with this last type of collector.

More detailed information is provided by sequential samplers. Differential, incremental, and intermittent fallout collectors are different types of collectors that are programmed to expose sequentially new collecting surfaces for equal increments of time. After some tests, 20 or more sequential samples have been collected at any one location. The "special incremental collector" has been adapted to shipboard use in the Pacific by the addition of a provision whereby the samples are automatically delivered to a laboratory below deck.

Sometimes the bottom of the fallout trays was coated with a thin layer of grease to prevent the collected particles from being blown away. Other trays were lined with plastic, so that the sample could be removed by simply lifting the plastic sheet out of the tray. Sometimes the trays contained small nylon pebbles to simulate a soil surface.

Whereas these sequential samplers are usually trays of one sort or another, a movable sticky tape has also been used as a sequential sampler. Mostly, however, this type of collector (tape fallout monitor) was used in conjunction with a radiation monitor to corroborate time-of-arrival data from the intermittent fallout collectors or similar devices.

Among the less frequently used active samplers, the "conifuge" needs special mention. This is a particle-sizing device consisting of a pair of rotating concentric cones separated by a narrow annular space. The particles are introduced into this space, and follow trajectories determined by their mass and the centrifugal force resulting from the rotating cones. Thus segregated, the particles deposit on the surface of the outer cone.

Impactors, mostly Casella, some Anderson, were used frequently to obtain a separation of the fallout into size fractions and to obtain mass distribution.

At the Nevada Test Site, these various collectors were placed on the ground or on low platforms. In the Pacific, various seaborne craft were also used for the location of such samplers. Thus, one has used Liberty ships outfitted as instrumented test vessels traversing fallout areas, instrumented landing craft modified to operate in a fallout zone, and unmanned barges, anchored in the path of prevailing winds.

Clouds were almost exclusively sampled by means of filter units mounted in pods attached to aircraft. The samplers, having a fixed geometry, were designed to approach isokinetic conditions as much as possible, but deviations from such conditions occurred as a result of variations in the altitude of sampling (barometric pressure), air velocity (air speed of the sampling aircraft), and ambient temperature. Several designs have been used.

Originally the filters were ACC Type V (Army Chemical Corps), fabricated from an asbestos base. Dissolution of these filters was quite cumbersome, and their impurity content was unsatisfactory. Hence, the transition was made to cellulose-based IPC-1478 filter paper impregnated with "Kronisol" (di-butoxy-ethylphthalate). The filtration and permeability characteristics of this material have been investigated by several researchers (Inst. of Paper Chemistry, 1960; Stafford and Ettinger, 1970a, 1970b, 1971). Exact duplication of field sampling conditions in the laboratory is very difficult, and most studies only approximated field conditions. Diffusion is the primary mechanism of particle collection, particularly for the smaller-size particles. For the larger particles, the relationship between pore size and particle size dominates. Thus, the efficiency is dependent upon the particle size. For particles larger than a few microns, the efficiency approaches 100%, decreases to about 20% at 10^{-2} - 10^{-1} µm, and may then increase again towards smaller particle sizes. As a result, measured size distributions will usually be biased against the smaller sizes.

Sticky wires were an important sampling method used in the Roller Coaster project conducted at the Tonopah Test Range to simulate accidental explosions of nuclear weapons under storage conditions. Arrays of wires coated with a sticky substance were suspended in the anticipated path of the cloud from tethered balloons (Tracerlab, Division of LFE, 1966). Collection occurred by inertial impaction and clearly discriminated against the smaller particles. Nevertheless, this method yielded estimates of the activity distributions in the clouds projected on a vertical plane approximately perpendicular to the wind direction. In addition to the gross activity measurements on the wires, the particles in the samples could also be further analyzed. On the other hand, besides the limitations imposed by the collection efficiency of the wires, the sticky-wire method for collecting cloud samples is limited in altitude and by some lack of mobility of the system resulting in possible poor cloud interception as a result of wind direction changes.

Turning now to the "miscellaneous" category, we mention the following:

High-volume samplers (hi-vols) were used primarily in the peripheral areas of the test site and outside the test site mostly to determine atmospheric levels of radioactivity for monitoring purposes.

Scoops were (and are) used to sample soil for subsequent analyses. To be mentioned here is the "Weasel," occasionally used in the fifties, being a remotely operated M-29 tank equipped with a scoop. Surface samples have also been obtained by coring, to obtain depth profiles of radioactivity.

In recent years, special samplers have been designed and used by Anspaugh *et al.* (1974) to determine resuspension on the Nevada Test Site. These samplers consist of a vertical array of collection tubes positioned at a slant and are used passively.

Finally, radar has been used to obtain cloud profiles, particularly at early times, and to estimate particle size distributions in clouds.

ANALYTICAL DATA

Since the inception of nuclear testing, a large number of properties has been measured often on isolated debris particles, but also on integral samples consisting of debris, nonradioactive admixtures to the clouds such as soil, and contaminants, usually windblown dust. Detailed analyses have been performed on samples from some selected tests, but some data are available from most shots. Limitations in the depth of the analyses were imposed by the large requirements of manpower, equipment, and time. Nevertheless, the detailed work that has been done encompasses a wide range of yields and detonation conditions. Table 2 lists the number of tests for which various data are available in greater or lesser detail.

Particle isolation was accomplished mostly by location of the individual particles by autoradiographic techniques followed by micromanipulation. These particles were usually about 2 µm or more in diameter. However, not all radioactive particles are identified, because autoradiography is limited by some minimum level of activity for each length of exposure. The submicrometer radioactive-particle population is not represented at all, and the remainder of the population is only partially represented.

The properties that have been measured are listed in Table 3. These measurements are further discussed below.

PARTICLE SIZE DISTRIBUTION

Size distributions have been measured primarily to estimate debris transport and for input into fallout models. Together with radioactivity data, they are used to evaluate the hazards to the biosphere (including man) resulting from local, intermediate, and worldwide fallout. They have been determined by means of optical or electron microscopy (or both) of integral samples and of

Table 2. Number of Events of Different Type Analyzed for Different Particle Properties

	Number of Shots by Type				
Property	Tower	Ground Surface	Balloon	Underground	Water Surface
Size	21	8	8	3	5
Shape	10	8	1	2	6
Color	10	8	8	1	6
Density	4	5	1	1	3
Activity location	5	6	0	2	3
Magnetism	4	2	1	0	0
Elemental and chemical composition	7	7	1	2	7
Solubility	9	5	1	2	5
Specific activity	15	7	6	3	7
Activity/size	17	6	2	2	3
Decay	25	7	2	3	8

Note: This table does not imply any extent or detail of the analyses.

Table 3. Properties of Debris That Have Been Measured

A. Particle Assemblies

1. Physical
   - a. Size and mass distributions
     - a1. By direct measurement (microscopy)
     - a2. By size separation: by sieve analysis
       from impactor characteristics
   - b. PuO_2-equivalent size distributions
   - c. Isotopic abundances
2. Chemical
   - a. Elemental and mineral composition
   - b. Solubility
   - c. Biological availability (not specifically addressed in this review)
3. Radioactivity

   Measured on total sample or size fractionated samples
   - a. Gross alpha, beta, gamma/specific activities as f (time)
   - b. Individual fission products/specific abundances
   - c. Uranium and transuranics/specific abundances
   - d. Induced activities

B. Single Particles

1. Physical
   - a. Color
   - b. Shape
   - c. Surface and internal characteristics
   - d. Density
   - e. Size
   - f. Magnetism
   - g. Electrostatic Properties
   - h. Agglomeration
   - i. Isotopic abundance
2. Chemical
   - a. Elemental and mineral composition--internal structure
   - b. Solubility
3. Radioactivity
   - a. Gross alpha, beta, gamma/specific activities as f (time)
   - b. Individual fission products/specific abundances
   - c. Uranium and transuranics/specific abundances
   - d. Distribution of activity

samples that have been size-separated in the laboratory. Mass or volume distributions can be derived from the size distributions but have primarily been determined by the weighing of size fractions obtained by sampling in the field with cascade impactors, or in the laboratory by sieve analysis. For impactor sampling, the applicable size range is determined by the first stage collecting all particles larger than about 10 µm and by the backup filter collecting the particles that are smaller than about 0.5 µm. Sieve analysis is useful for samples consisting mostly of large particles, the smallest sieve opening used being 44 µm. Samples, or sieve fractions, consisting mostly of small (<50 µm) particles may be separated into size fractions by allowing the particles to settle in an appropriate liquid having a small density gradient to eliminate convection currents. The particles that collect at the bottom of the liquid column are removed at predetermined times. As many as 14 size fractions have been thus obtained (Heft and Steele, 1968). High-speed centrifugation has also been used. The size distribution of the fractions are added with the fraction weights as weighting factors. These fraction weights are obtained by filtering the fractions and weighing, or by chemical analysis. Figure 1 shows the size distributions of one such set of size fractions. The overlap of the size ranges may have been caused by such factors as shape and density variations, entrainment of small particles by large particles, and so forth.

Particle size measurements are always made on small aliquots of the samples, usually on filter collections. Sections of the filters were ashed, the ash was dispersed in an appropriate liquid. Microscope slides or electron microscope grids were prepared from the dispersion.

Individual particles were measured in the light microscope by means of a calibrated scale, later also by means of an image-splitting device or a Porton graticle. Whereas in the first two methods, the actual sizes of individual particles were measured, the use of the graticle places the particles in size bins and effectively determines a histogram with logarithmicly equal size intervals. For irregular particles, the definition of size is not always clear, and may differ among investigators. The geometric mean of the largest and smallest diameter has been used to define size. Size has also been defined as the length of the projection of the particle on an (imaginary) horizontal line. Thickness has never been considered.

Clearly, none of these size and mass distribution measurements distinguish between radioactive and nonradioactive particles. In a few cases, e.g., with balloon tests and one test buried at a shallow depth, a distinction was made between fused smooth spherical or nearly spherical particles and irregular particles to obtain separate size distributions (Russell, 1965; Nathans *et al.*, 1970). The spherical particles were assumed to be primary debris particles. They are significant only below about 5 µm. On a few samples from other tests, radioactive and nonradioactive particles were distinguished by autoradiography, and size distributions of these radioactive particles were measured separately. This procedure is, of course, extremely cumbersome.

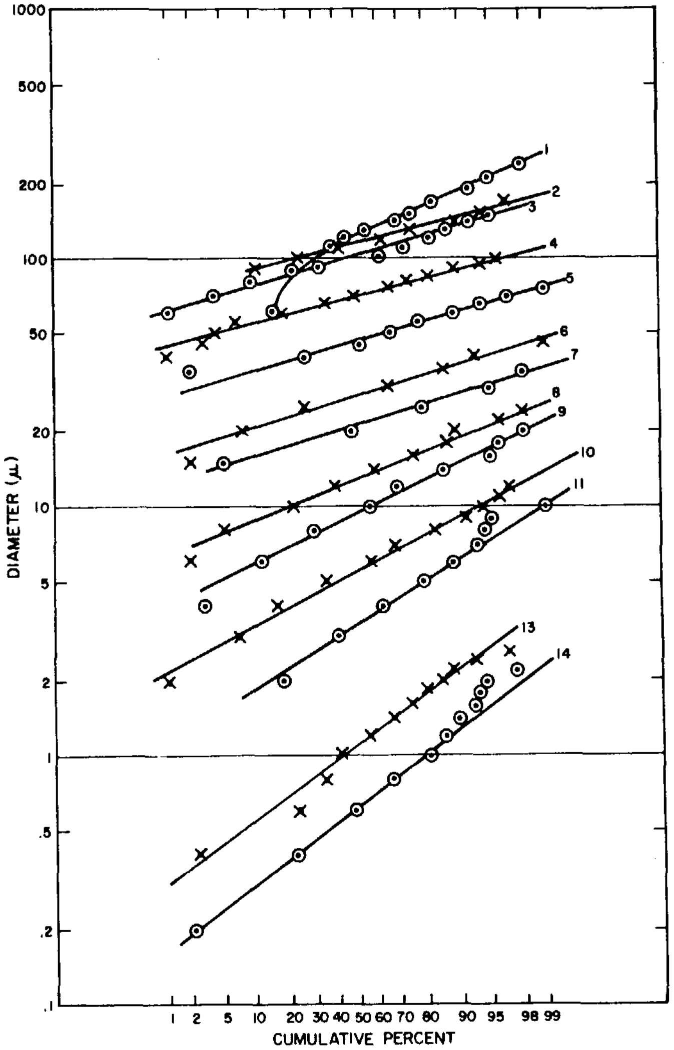

Figure 1. Particle size distribution of size fractions from a Johnie Boy sample

In quite a few cases, the measurements were made on fallout samples collected throughout the close-in fallout field. This may make it possible to derive estimates of the size distributions of debris on the soil anywhere in the fallout field. Such work would be quite complicated, however, in areas where fallout from more than one shot is present.

Most of the size distributions reported before the mid-60s show lognormality. These results are probably correct for the fallout samples. Lognormal size distributions of cloud samples, particularly early samples, are probably biased against the larger-particle end of the distribution. Later work has shown that many cloud samples have a particle size distribution that may be described as lognormal below a few micrometers, but obey approximately a r^{-m}-law, with m having a value between 3 and 4, above the range of log-normality (Russell, 1965; Nathans *et al.*, 1970) (Figure 2).

In a very few cases, where the fallout particulates have a sufficiently high plutonium content, special methods have been employed that take advantage of the special properties of plutonium nuclides (Nathans *et al.*, 1976). By autoradiographic registration of alpha particles ("hollow-star technique"), the plutonium content of each particle is registered as distinct tracks on a photographic emulsion. These tracks form a star and the number of tracks is converted to a "PuO_2-equivalent" particle diameter. This method has been applied to soils from the fallout field of a safety shot (Schulz *et al.*, 1975) and to a few resuspension samples. The lower-size limit depends on the isotopic half-life and on the exposure time, but with reasonable exposure times extends down to below 0.1 μm PuO_2-equivalent diameter for plutonium-239. Particles above 1 or 2 μm may be identified, measured, and isolated for additional measurements. These special methods also make it possible, in many cases, to obtain information from old debris for which normal autoradiographic techniques are no longer applicable.

Density-gradient separations have also been applied in a few cases (Tamura, 1976; Soinski and Nathans, 1976). This method is particularly useful for the gross separation of undiluted debris particles from particles that consist primarily of soil components and is based on the observation that such particles have specific gravities well in excess of 3.0 or thereabout.

OTHER PHYSICAL PROPERTIES

1. Shape. Particle shape was measured to determine if the radioactive debris possesses any special characteristics that distinguish these particles from background, entrained inactive material, and contamination. The radioactive particles were generally identified by autoradiography, occasionally by micromanipulation, and the use of a beta-gamma meter or phosphors. Usually a sufficient number of particles were viewed to encompass the various shapes (and colors) that were present.

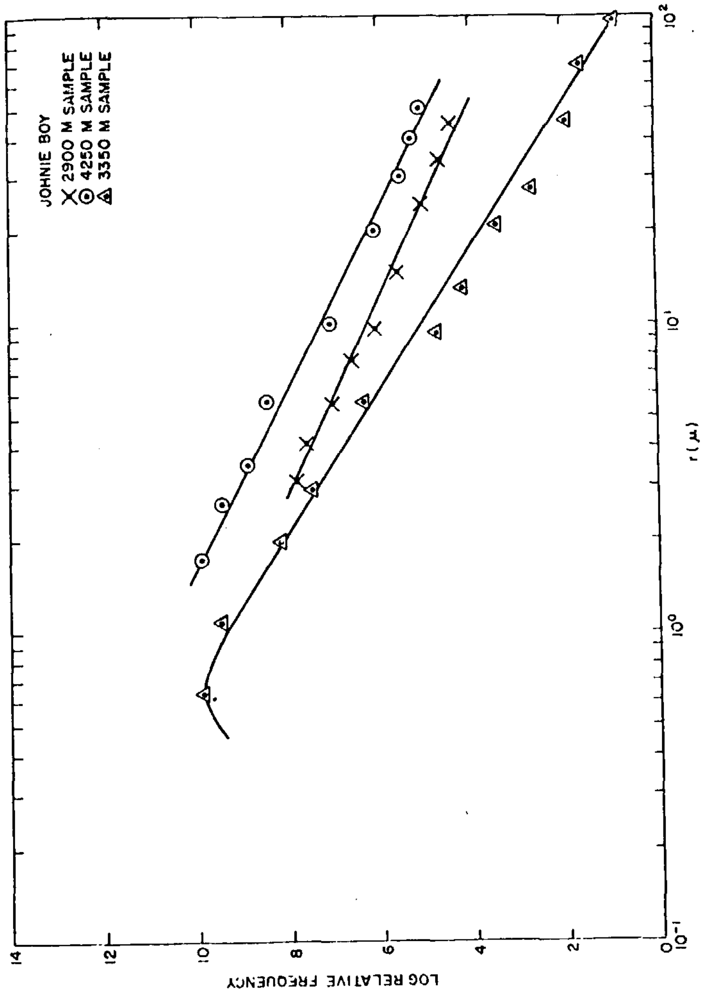

Figure 2. Particle size distributions of three Johnie Boy samples as derived from the particle size distributions of size fractions

The debris particles generated by free airbursts were usually spherical or spheroidal in shape (Mamuro *et al.*, 1962; Bjornerstedt and Edvarson, 1963; Benson and Leventhal, 1964). As a result of fusion at high temperatures, occasionally an irregular particle was seen. More common was the occurrence of accretions, small particles attached to larger ones. The attachment obviously took place when at least one of the partners had solidified, or had become sufficiently viscous to prevent coalescence.

When the scaled height of burst becomes small enough to allow entrainment of ground surface material (between 100 and 125 m), the round radioactive particle population is augmented by radioactive particles that are irregular in shape. These irregular particles may or may not have a smooth surface. The fraction of spherical particles tends to decrease with decreasing scaled height of burst, but does remain significant in the size range below a few microns, as was inferred from microscopic examination of cloud samples from Johnie Boy, a low-yield shot buried just under the surface. Sometimes spherical particles were found to be attached to irregular particles originating from the soil or rock at and around ground zero (GZ). Large (>1 mm), smooth, glassy particles have also been found in fallout from ground-surface shots. It has been speculated that these particles were formed by throw-out of molten material from the crater at early times.

Several classifications of particles have been reported with regard to shape as well as color. There is some subjectivity involved, particularly in providing descriptions and details. In addition, populations of particles of different types usually vary from test to test. This observation also affects the reported classifications.

A general review of shapes of particles from tests through the 1956 Redwing series has been written by Adams *et al.* (1958). His review covers most of the work that has been done on balloon shots, tower shots, and ground- and water-surface bursts. Some additional information has been obtained from tests of these types from the post-Redwing period (Plumbbob, Hardtack, Sunbeam series, cratering shots), but detailed morphological examination of particles had become deemphasized.

It may generally be concluded that fallout particles present on and in soil are not all recognizable. Glassy particles, that have the appearance of having been fused at one time or another, are the most likely candidates for being radioactive fallout particles. Any other particle types cannot be so assigned on the basis of shape alone.

2. Color. For lack of universally used color standards, the colors as reported are somewhat subjective. Generally, however, the colors vary from very dark (black) to very light and transparent, with many gradations within a shot. The predominant color range appears to depend on the soil, and on the presence or absence of a steel tower. Some observers report color in more detail than others (Schuert, 1958). In tower shots, dark colors usually dominate some of the particles from NTS surface shots.

Color is not a good criterion for the identification of debris, alone or in combination with particle shape. Correlation between color and activity is tenuous at best for airburst debris and is expected to be nonexistent for debris from surface and near-surface shots.

3. Surface Characteristics. Surface appearance and surface characteristics also are nonspecific and correlate only partially with radioactivity, depending on the history of the individual particles. Material that has entered the fireball very early and that has become vaporized or that has become liquified into a thin liquid usually formed spherical particles with a smooth surface. Smooth surfaces have also been noted on some large particles from surface shots that appear to have been formed from molten soil or rock ejected from an incipient crater. Smooth surfaces are further found on particles that became only partially fused, but that maintained their approximate dimensional ratios. Other particles never became hot enough for even partial fusion and are unaltered except for an occasional accretion of fused particles.

   More recently, scanning electron microscopy has been applied to the study of surface characteristics. Thus, several types of particles have been identified, for example, in debris recently collected from Area 13 (Schultz *et al.*, 1975; Soinski and Nathans, 1975): (1) exfoliating, but generally spherical; (2) irregular in both shape and morphology; (3) rough interior partially covered by a smooth shell; (4) spheroidal with smooth surfaces. The possibilities of using the scanning electron microscope, particularly for the study of weathering, have not been sufficiently investigated, however.

4. Internal Characteristics. Spherical or other-shaped particles have been ground to hemispheres for physical observation or in preparation for electron microprobe analysis. The internal structure of these particles range from fused solid homogeneous to particles with voids and instertices and inclusions and onion skin. These effects are obviously based on the cooling conditions and the environment in which the cooling occurred.

5. Density. The density of spherical particles may be determined by measuring their rate of all in a liquid and applying Stokes' law with or without Cunningham's slip correction. A microcolumn with observation by means of a microscope is used for particles between 3 and 10 μm (Benson *et al.*, 1967.). Smaller particles become too subject to convection currents, larger particles fall too fast. This method requires careful manipulation of the particles, and losses run between 10 and 30%, dependent on the operator. The density of larger spherical particles may also be determined by a Stokes' fall method, but in a longer column having a larger diameter (Wrigley and Gleit, 1964).

   For nonspherical particles, the aforementioned method fails because the shape factors are generally not sufficiently known. Density gradient columns are used for these particles, but with particle assemblies rather than with individual particles. As many density cuts as desired can be made.

The density of particles from airbursts has been found to be between 3.25 and 5.0 g/cm^3, with most values lying between 3.5 and 4.0 g/cm^3. Particles from surface and near-surface bursts have densities close to those of the soil rocks, with the exception of most of the particles that had been completely fused and that contain primarily device materials. These latter particles have densities greater than 3 g/cm^3. Some high-density particles which have been confirmed as mixtures of uranium and plutonium have been observed in Roller Coaster.

6. Magnetism. Magnetic (ferromagnetic) particles were found in a number of tower and ground-surface shots. This magnetism could usually be traced to significant amounts of iron having been present in the shot environment as additions (tower, e.g.). No quantitative measurements were made. It was noted, however, that dark particles were generally more magnetic than light particles. These particles were also found to be highly radioactive.

7. Electrostatic. Early experiments in operation Fitz William attempted to identify particles by their electrostatic charge resulting from their radioactivity. Electrophoretic separation of particles was done by this laboratory in a liquid with the observation that the particles both positively and negatively charged were found.

8. Isotopic Abundances. Isotopic abundances of the uranium and plutonium isotopes have been extensively measured in both aggregates and individual particles by single- and double-focussing mass spectrometric methods. This has proven to be a powerful tool for identifying the origin within the device of the particle.

9. Chemical Composition. The chemical nature of fallout and cloud particulates is greatly dependent upon the environmental shot conditions.

   The activity in determining this property decreased considerably after the late 1950s for particulates from all but free airbursts. Since then, a number of cloud samples from surface and near-surface bursts were analyzed for major constituents for the purpose of reconstructing a sample weight so that radionuclide specific abundances could be calculated. Compositions thus obtained included the contributions from unaltered, inactive soil particles drawn into the cloud.

   The methods that were employed included X-ray diffraction, emission spectroscopy, wet chemistry, dissolution, and atomic absorption spectroscopy, but also the application of the petrographic microscope to thin sections. The electron microprobe analyzer has been used rarely for particulates from shots yielding significant fallout. More recently, the ion microprobe and scanning electron microscope have been added as an analytical tool for particles.

   Many of these methods were applied to individual particles as well as to representative sample aliquots. The work is generally quite time-consuming and has, therefore, been somewhat limited. Also, fallout from coral shots has been more extensively investigated than fallout from alluvium (NTS) shots.

   X-ray diffraction results showed, as expected, that the fallout samples contained essentially the same minerals as the soil on or over which the devices were detonated. Single selected particles from coral shots showed mostly CaO, $Ca(OH)_2$, $CaCO_3$. Selected particles from tower shots on alluvium showed the presence of magnetite, Fe_3O_4, but were mostly vitreous.

   Spectrochemical analyses were at best semiquantitative. Most analyses were done on selected single particles from a few shots. The results show some minor differences in composition with the substrate (soil with or without a tower). These differences are usually readily explainable.

   Petrographic examination of thin sections from selected particles showed a variety of structures that can readily be explained on the basis of the phenomenology of the events: partial fusion, total fusion, incorporation of fine grains, some chemical alteration (in the case of coral shots), and so forth. Sometimes autoradiography was used to locate radioactive regions within a particle.

   Quantitative chemical analysis was mostly concerned with the iron and lead content of the particles. Other analyses performed were moisture and organic matter. In the case of fallout from coral shots, significant amounts of water were often found in the collectors, and chemical and radiochemical analyses and measurements were performed on all phases. These analyses included some or all of the following: Ca, Na, K, Cl, Mg, Fe, Cu, Al, Si, and U.

   Most electron microprobe analyses were done on particles from airbursts. Few fallout particles were thus analyzed. Large particles were sectioned and scanned over a cross section. A few selected particles from Area 13 were analyzed intact (Schulz *et al.*, 1975). It is noted that by this method only the surface and subsurface regions of particles are analyzed.

10. Solubility. Of particular importance to hazards assessment is the availability of radionuclides in debris and in fallout to the biosphere other than through self-implantation of particles. Thus, radionuclides may become available through solubilization, and experiments with different fallout samples have been conducted to determine the solubility of radionuclides. Effectively, however, these experiments are leaching experiments.

    The value of these experiments is very difficult to assess. The leachability depends on the distribution of the radionuclides in and on the particles, on the properties of the particles, and on the chemical form of the nuclides, therefore often also on the particle size, but in particular, the leachability depends on the leaching liquid. Therefore, regardless which liquid is used in laboratory experiments, often there remains a problem of translating the laboratory results to the "real world."

    In general, there are two primary phenomena to be considered. One phenomenon is the solubilization of radionuclides in the soil, perhaps followed by the movement of solubilized material through the soil. This solubilization would occur in an environment created by rainwater and the interaction of rainwater with the soil. This interaction creates conditions of pH, mineral content, and organic content of the water, some of the organic components possibly having a complexing capability. The second phenomenon is the solubilization of radionuclides as a result of their direct interaction with vegetation or body fluids. This is a situation which is extremely difficult to simulate in the laboratory, whereas simulation of the interaction of fallout with soil and rain can perhaps reasonably be simulated in environmental chambers or otherwise (Larson *et al.*, 1960).

The reported solubility studies show how the various investigators have struggled with these problems. Solubilities in water and in 0.1 N HCl have been used most often as indicators of biological availability (Tamura, 1976), but other liquids have also been used, such as nitric acid, sodium thiosulfate, citric acid-sodium phosphate buffer, seawater, ammonium oxalate, chelating agents (EDTA and others). Various types of experiments also have been conducted: leaching through columns, leaching in a static system, series leaching, leaching of single particles on watch glasses, even a single experiment *in situ* on the Test Site. Variables that have been considered are shot environment, particle size, activity per unit weight, distance from GZ. The measurements that were made were almost exclusively gross activity measurements with a determination of the fraction of the activity that was solubilized. Rarely were analyses performed for individual nuclides such as barium-140 and/or strontium-89,90.

Larson *et al.* (1960) has summarized significant findings from experiments with Nevada Test Site fallout during the decade prior to 1959. Debris from balloon shots appears to be more "soluble" than debris from tower shots; smaller particles are more "soluble" than larger particles. Quantitative numbers on the fraction of activity leached are quite variable.

It would appear that leaching or solubility experiments that provide data only in terms of gross activity are not very useful. The chemical characteristics of various nuclides and the distribution of nuclides in and on the particles will result in different dissolution rates and different solubilities. For example, incidental data obtained at our laboratory with cloud samples from ground-surface bursts showed strontium-90 to be more leachable than promethium-147.

11. Radioactivity. The prime concern for local fallout from shots was originally the external radiological hazard of the fallout field over a period of time after each shot that such hazard may have been significant. Thus, measurements were usually restricted to beta and gamma radiation and most of the data generated before the early sixties are of only limited use for the assessment of hazards that are now present on the test sites, these hazards being associated with the presence of only a very few long-lived nuclides, particularly alpha-emitters. Alpha-emitting daughter products such as americium-241 were not considered.

Data are often reported as specific activities, that is, as counts or disintegrations per minute per unit volume (μm^3) or per unit mass (g).

The measurements were sometimes made on samples as collected (or size-separated) or on single particles. In the latter case, a modal activity or an average activity is calculated or determined graphically from frequency curves. Some standardization was effected by reporting, when possible, the activities at T_o + 25 days (600 hours). This time was chosen to allow the decay of variable quantities of induced activities.

The measurements of gross activities are of some use in one respect; where size fractions or individual particles are involved, some average behavior of activity with particle size is indicated, or a yield-dependence of radionuclide concentrations can, and sometimes had been, inferred. Thus, a large amount of data have been reported and plotted, mostly in the form of frequency plots, showing gross activity as a function of particle size. For one or two tests, fallout particles between 300 and 2,000 μm were separated by morphology into five classes and their activity was measured and plotted as a function of size. Data for individual radionuclides were not reported with very few exceptions, and then mostly in the form of fissions per gram (equivalent to molybdenum-99 concentrations), or as strontium-89, barium-140, or induced activity data.

After the early sixties, a different approach was taken to the analysis of fallout and cloud samples (Nathans, 1970). The isolation and analysis of individual particles from shots of interest was virtually abandoned. The limitations of gross activity measurements was more realized in the experimental programs and individual radionuclide analysis was emphasized more. Thus, a more fundamental approach was being used with the expectation that as a result, particle and radionuclide behavior could be generalized into a predictive system for various types of shots. This approach was justified also by the improvements in computer hardware and software capabilities, allowing much more sophisticated transport and fallout models to be developed. An example is DELFIC (DOD Experimental Land Fallout Interpretive Code (1966)) which considers cloud rise, transport, and settling, under different meteorological conditions including rainout.

As a result, shots of interest such as Johnie Boy, Sedan, and others were analyzed in much more detail than had been done before. In addition, many shots from the fifties were revisited. Thus, cloud samples from virtually all ground-surface shots and from many tower and balloon shots were reanalyzed in considerable detail, fallout samples not having been available. During this work, samples were separated into size fractions. Radionuclide specific abundance was determined as a function of the particle size, but related only to the volume or mass of radioactive and nonradioactive particles together. In the case of these old shots, the data acquisition was obviously limited, because of decay, to only a few nuclides.

AVAILABILITY OF SAMPLES

A large number of contacts were made with people and institutions that have been or are still involved with the analysis or utilization of nuclear debris to ascertain the existence and location of samples. As we expected, the results were disappointing. A rather considerable inventory still exists of samples collected by aircraft at the weapons laboratories. A few selected samples are still available at LFE Environmental. Fallout collections are few and far between. The principal reasons are that many of these fallout collections were used up in their entirety for analysis and that a number of samples was discarded some years ago.

Some fallout samples are now under the control of the Nevada Operations Office. These samples are mostly from Areas 11 and 13 ("Program 57") and have been collected at distances greater than 1 mile from GZ. Some Roller Coaster samples on planchets are available at LFE Environmental. About 200 fallout samples from Shasta, Johnie Boy, Small Boy, and Sedan are located at Camp Parks near Livermore, California. All these samples are well documented with regard to shot location and collection method. This documentation is still in the possession of the original investigators. In addition, a few as yet nondescript samples may be available at Battelle-Northwest in Richland, Washington, and at the Ballistics Research Laboratory in Aberdeen, Maryland.* A complete inventory of all available and relevant samples has not been made.

DISCUSSION

A large body of data has been generated on fallout as well as on clouds from many events of different type that have contaminated the test sites. Projects were conducted independently, each having its own objectives and its own experiments. As a result, there was little correlation between experiments, and very few review articles or reports in which results were put together and critically examined. Even data from different projects relating to single shots often remained uncorrelated.

The correlation of data of similar type from different tests is difficult because sample collection methods, analytical techniques, and data presentations differed. Sometimes sample documentation was inadequate. Cross-calibrations were almost nonexistent. Subjectivity in reporting was sometimes present, for example, in the case of shape and color.

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*The BRL samples have now been transferred to LFE Environmental.

The reasons for data acquisition and the use to which data have been put are quite varied. Emphasis has changed over the years, in part because people were getting smarter, in part because requirements changed.

Data acquisition for the purpose of evaluation of device performance has always occurred. Such data are of limited use for today's needs for fallout characterization. In the early days, understanding particle and fallout formation and the formation of radiological countermeasures were the prime objectives for most of the projects, the former objective having persisted for a long time. This particular objective was also sought in a number of projects involving laboratory simulation of conditions under which fission products would be incorporated in particles.

The definition of the fallout field and its properties was also of interest for certain military applications, and became of interest to the Plowshare program (Peaceful Use of Nuclear Explosives) for what today would be called Environmental Impact Assessment. Data were also generated, particularly during the last ten years or so, to provide inputs to fallout models for model verification and for predictive purposes. Estimates of soil burdens in nuclear clouds also result from these and similar activities.

Some data not separately discussed in this review were generated for the purpose of evaluating effects from accidents; for example, data from the Roller Coaster and Kiwi (nuclear propulsion reactor) tests.

Studies of the modes of entry of fallout or radionuclides into the biosphere have always provided an important impetus to the gathering of a variety of data. Solubility studies were certainly directed towards this purpose. In recent years, fallout-particle characterization studies also have increased for use in such studies. Thus, hazard assessment, either of already contaminated areas or in a predictive mode, has become the prime interest.

Of particular interest at the present time is the characterization of the radioactive fallout particles at the Nevada Test Site, especially with regard to their chemical and radiochemical properties. The plutonium and americium content of the particles, the concentration as a function of the particle size, the distribution within the particles, the chemical form, the "solubility," are all of interest for the estimation of the redistribution of fallout (by wind or mechanical action) and modes of entry into the biosphere. Many of the data, once they are correlated, are directly applicable to the question of fallout characterization. However, plutonium, being an alpha emitter, was more or less ignored during the earlier work. Nevertheless, useful inferences may be made from what has been observed generally with regard to radionuclide fractionation and some actual data that are available.

Making inferences with regard to transuranics concentrations and distributions, although useful, is not wholly satisfactory. Requirements that have been laid down or that may be formulated in the near future necessitate a more definitive characterization of the transuranics-bearing particulates and their temporal and chemical behavior in the environments in which they exist. Such a characterization is by no means a trivial task. Ideally, a physical separation of these particulates from the soil particles is required. This is a task which the passage of time has generally made much more difficult, more time-consuming, and more error-prone than it was during the period shortly after the generation of the fallout. In particular, the standard autoradiographic techniques do not apply anymore: The principal fission-product activities remaining after 10 or 20 years are the strontium-90--yttrium 90 and cesium-137--barium-137 activities which comprise over 90% of the residual activity. A few percent of the remaining activity is caused by promethium-147. These activities are insufficient for adequate registration, especially since fractionation of strontium-90 and cesium-137 is quite severe, such that fallout from surface and near-surface bursts is usually much depleted in these nuclides relative to zirconium-95 or promethium-147, or even transuranics. The fallout itself is, of course, depleted in the smallest particles. In fallout from the more deeply buried cratering shots, the fractionation pattern appears to be reversed, however. This fallout also contains little or no fused material.

A few methods that have been used for miscellaneous purpose are available to provide a concentration, if not a separation, of the desired particulates: density separations with or without size separations, alpha-track and/or fission-track radiography with particle isolation. These methods would be useful to provide at least a partial characterization of the contamination as it exists today. There is some evidence that the transuranics were mostly concentrated in a small fraction of the fallout particulates, and although some changes in the distribution of these nuclides may have taken place as a result of weathering, this conclusion is probably still valid. Thus, a combination of data correlation and well-planned sample analysis will provide answers to most or many of the questions that are now being posed.

CONCLUSIONS

There exists a rather large body of data describing physical, chemical, and radiochemical properties of debris from a number of tests of different types and of interest for the characterization of the contamination of the test sites. Most of these data remain uncorrelated. Although a great deal of data correlation is possible, the effort would be hampered by differences in methods used, in some cases by the subjectivity of the observers/reporters. Furthermore, the objectives of the programs under which data were acquired usually did not bear much relation to each other. After correlations have been made, some inferences can be made that will be very useful for understanding the characteristics of the transuranics-containing particulates. A more definitive experimental characterization of these particulates has become rather difficult because the identification of particles of interest by relatively simple techniques is virtually impossible as a result of radioactive decay and the partial failure by morphological and color criteria. However, the application of special methods that focus on alpha-emitters still makes far-reaching identification possible.

There is a significant lack of useful data on biological availability. The solubility experiments that were conducted should be very critically reviewed, not only with regard to their applicability to biological availability, but also with regard to their validity.

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MEASUREMENTS OF AMERICIUM AND PLUTONIUM IN THE ATMOSPHERE*

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ABSTRACT

Alpha spectrometry and high sensitivity mass spectrometry have provided complementary data which have proven useful in understanding fluctuations in the atmosphere, particularly the relationship between plutonium activity levels and variations in the ^{240}Pu/^{239}Pu isotope ratios. A summary of past plutonium data plus some recent measurements are presented. Previously unpublished ^{241}Am data collected from 1963-1970 are also presented. These data indicate that the ^{241}Am/^{239}Pu atom ratio in the atmosphere has been decreasing since 1967. Explanations for this anomaly are postulated.

INTRODUCTION

Virtually all the transuranic activity on the surface of the earth has come via the atmosphere. Therefore, for any predictive model of plutonium buildup in the biosphere to be accurate, it is critically important to assess the element's magnitude, distribution, physical form, and chemical form in both the stratosphere and troposphere. The primary sources of activity in the past were 325 KCi of ^{239,240}Pu fallout debris from nuclear testing (Hardy *et al.*, 1973) and 1.7 KCi of ^{238}Pu from the burnup of the SNAP-9A capsule in 1964 (Harley, 1964). Accidents involving aircraft carrying nuclear weapons resulted in localized releases 1000 times less (Langham, 1968). The largest accidental releases from land-based facilities have been 50 times smaller still (Hammond, 1971; Krey, 1976).

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*This work was conducted under the auspices of the Air Force Technical Applications Center.

The activity levels of atmospheric plutonium are well documented by collections from rockets, high-altitude balloons, aircraft, and ground-based filtration units. The most extensive survey is contained in the HASL Quarterly Reports in conjunction with Project Airstream (Krey and Toonkel, 1975; Krey, 1967). The $^{240}Pu/^{239}Pu$ isotopic ratios in the atmosphere have also been measured. The most extensive survey of $^{240}Pu/^{239}Pu$ ratios was obtained from the atmospheric sampling and radiochemical analysis program that was conducted from 1959-1970 by the U.S. Department of Defense, Atomic Energy Commission, and the National Oceanic and Atmospheric Administration. Fission products in many of the samples were measured (Anonymous, 1961-1970). The plutonium isotopic ratios were reported in HASL-273 (Anonymous, 1973).

A review of the preceding data shows an excellent correlation with known atmospheric injections (Glasstone, 1964; Jackson, 1976). These data were combined with other measurements and led to the development of various models of atmospheric transport including intrastratospheric transport, effects of seasonal variations, and transfer mechanisms from the stratosphere to the ground (Stewart *et al.*, 1958; Dingle, 1965; Pierson, 1969; Krey and Krajewski, 1970). They also provide a means for predicting mean atmospheric residence times, backcalculating the total atmospheric burden of Pu and other nuclides, and forecasting general global fallout patterns (Brewer, 1949; Rand Corporation, 1953; Haxel and Schumamm, 1955; Dobson, 1956; Libby, 1956; Kuroda, 1958; Machta and List, 1958; Martell, 1959; Machta and List, 1959; Prawitz, 1964; Beck, 1966; Davidson *et al.*, 1966; Feeley *et al.*, 1966; Karol, 1966; Machta, 1966; Bhandari *et al.*, 1966; Swindle and Kuroda, 1969).

The average yearly concentrations of plutonium in the stratosphere and the troposphere of the northern hemisphere are shown in Fig. 1. Yearly stratospheric plutonium levels were calculated from the data reported in HASL-273. Samples collected near ground level in New York (Bennett, 1976) were used to illustrate the trends in tropospheric plutonium. It is interesting to note from Fig. 1 that the ratio of plutonium in the stratosphere to that in the troposphere changes from year to year. This variation is explained by the fact that high-altitude injections must settle to near the tropopause before they are available for transfer to the troposphere.

Today, samples collected at ground level are representative of the isotopic composition of plutonium in both the troposphere and stratosphere. The troposphere has a mean residence time 30 days--a value which is 12-15 times shorter than the mean residence time of the stratosphere (Libby, 1956; Burton and Stewart, 1960; Kuroda *et al.*, 1962; Volchok, 1967; Krey and Krajewski, 1970). Therefore, tropospheric plutonium must be continuously replenished by the transfer of submicron sized fallout particles from the stratosphere (Lockhart *et al.*, 1965; Shleien *et al.*, 1966). Resuspension of previously deposited debris is secondary and extremely negligible source of tropospheric plutonium. Typical values for resuspension are 10^{-9} -10^{-10}m -1 (Volchok, 1971; Anspaugh *et al.*, 1974; Bennett, 1976).

The plutonium associated with fallout is a complex mixture originating from two primary sources. The plutonium may be residual fuel from the nuclear device or it can be made by neutron capture reactions during the explosion. Therefore, the $^{240}Pu/^{239}Pu$ isotope ratios can fluctuate over a large range.

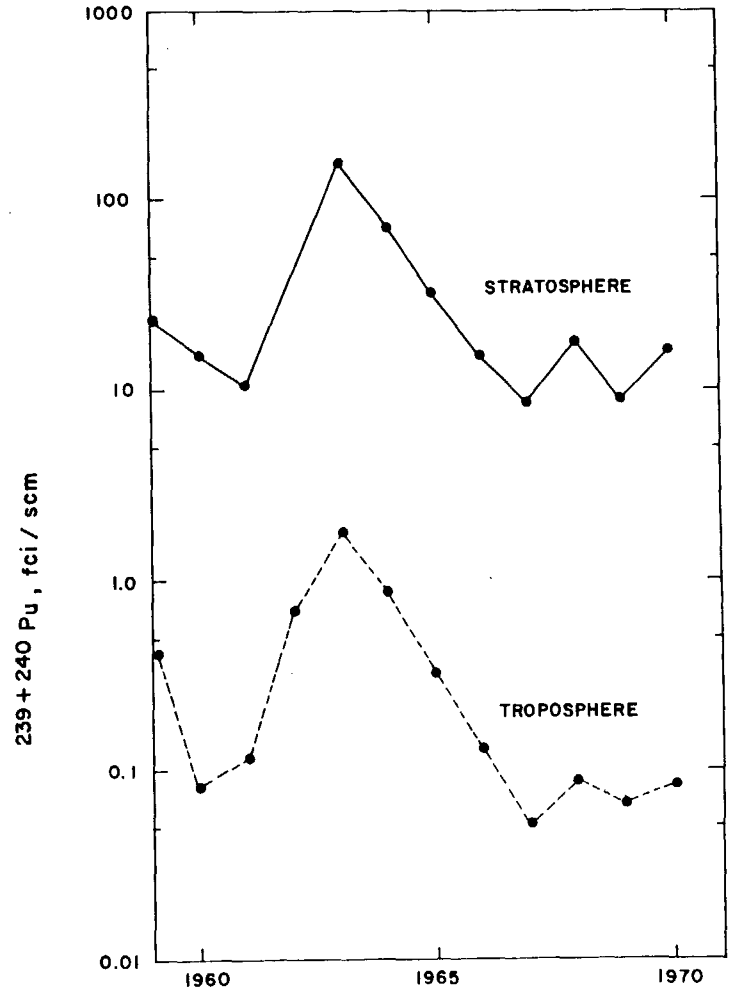

FIG. 1 ATMOSPHERIC PLUTONIUM LEVELS

For example, ^{240}Pu/^{239}Pu ratios measured in environmental samples vary from 0.02 at NTS to 0.40 in the South Pacific Test Area (Efurd *et al.*, 1976). Weapons grade plutonium has ^{240}Pu/^{239}Pu ratio of 0.06 (Krey and Krajewski, 1972).

The ^{240}Pu/^{239}Pu isotope ratios may be used to identify and quantify plutonium resulting from a specific release even in the presence of copious amounts of fallout plutonium (Hardy *et al.*, 1972). This separation procedure is based on the assumption that fallout plutonium has a unique ^{240}Pu/^{239}Pu isotope ratio. Krey *et al.* (1976) derived a mean atmospheric value from the samples reported in HASL-273. They pooled the samples collected before mid-1967 to calculate a ^{240}Pu/^{239}Pu ratio of 0.179 that is in excellent agreement with the mean value of 0.176 ± 0.014 observed in soils containing only fallout plutonium.

The approach of Krey *et al.* (1976) is an excellent method for predicting the buildup and isotopic composition of plutonium in soil samples where the debris from the individual injections have been superimposed to form a composite sample. For soil samples, one may assume 25% of the atmospheric plutonium burden was injected during 1956; most of the remaining plutonium was injected during 1962. The mean residence times of the stratosphere and troposphere are short; therefore, the majority of debris from these atmospheric test series were deposited on the earth's surface by mid-1967. The injections of 1967 and subsequent years have been too small to change the ^{240}Pu/^{239}Pu isotope ratios in the soil. However, one may not assume that there has been a constant ^{240}Pu/^{239}Pu isotope in the atmosphere. The atmosphere is self-cleansing; therefore, its ^{240}Pu/^{239}Pu isotope ratio tends to reflect the more recent injections. Although the injections after mid-1967 did not alter soil concentrations, they were large enough to change the ^{240}Pu/^{239}Pu isotope ratios of the atmosphere.

The variations in the ^{240}Pu/^{239}Pu isotope ratios in the northern and southern hemispheres on a yearly basis as calculated from the data in HASL-273 are shown in Fig. 2. It is interesting to note that the plutonium isotope ratios in both hemispheres track fairly well until 1968. From 1968-1970, the ratios in the southern hemisphere became significantly lower than those in the northern hemisphere. Because of the small number of analyses reported for the southern hemisphere between 1968-1970, the estimations of the average annual ratios should be considered only approximations. However, they do definitely indicate that the atmospheric ^{240}Pu/^{239}Pu isotope ratios have changed with time. The trends in the northern hemisphere are less obvious.

The purpose of this report is to document the plutonium analysis performed by the McClellan Central Laboratory that complement the 1959-1970 study.

In addition, this report documents the americium measurements which were obtained from 1963-1970.

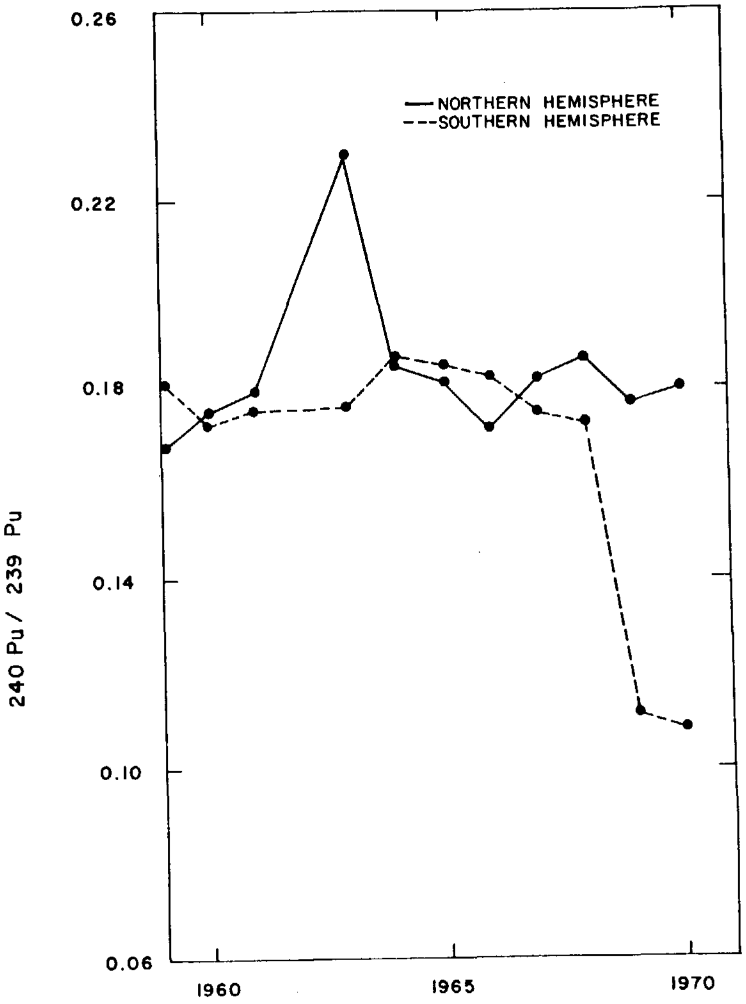

FIG. 2 PLUTONIUM ISOTOPIC RATIOS

MATERIALS AND METHODS

In 1975 and 1976, the McClellan Central Laboratory determined the activity levels and isotopic compositions in plutonium samples from Edwards Air Force Base in California. A second sampling station was located at Ramey Air Force Base in Puerto Rico. The atmospheric plutonium samples were collected on IPC-1478 filter papers (DASA-1168; Rimberg, 1968), which were exposed by high-volume ground-based filtration units. The papers were dissolved in HNO_3, HF, $HClO_4$, and then spiked and equilibrated with ^{242}Pu tracer. Plutonium was extracted by the thenolytrifluoracetone (TTA) method (Cuninghame and Miles, 1956; Coe, 1965) and analyzed on high-sensitivity thermal ionization mass spectrometers.

RESULTS AND DISCUSSION

If samples are collected in regions where resuspension of fallout debris is negligible, the plutonium isotopic ratios measured by ground-based filtration units are representative of both the troposphere and stratosphere. This method of sample collection was used to update the 1959-1970 background study.

The plutonium isotopic ratios and activity levels of the sample obtained by ground filter units at Edwards Air Force Base in California and Ramey Air Force Base in Puerto Rico are shown in Table 1. The locations display daily variations in both the isotopic ratios and activity levels. The average $^{240}Pu/^{239}Pu$ ratio at Edwards Air Force Base was 0.146 ± 0.017. The average $^{240}Pu/^{239}Pu$ ratio at Ramey Air Force Base was 0.159 ± 0.011. Similar results were obtained from samples collected at other locations. These data seem to indicate that the $^{240}Pu/^{239}Pu$ isotope ratios in the northern hemisphere have continued to decrease since the last measurements were made in the 1959-1970 background study. The decrease in $^{240}Pu/^{239}Pu$ isotopic ratios is the result of the atmosphere injections that occurred during and after 1967, the date when atmospheric testing was resumed.

Additional samples were collected at each location and analyzed for plutonium activity levels. The results are reported in Table 2. Edwards Air Force Base had an average plutonium concentration of 0.021 fCi/SCM. Ramey Air Force Base had an average atmospheric concentration of 0.004 fCi/SCM.

The difference in the average concentrations of the two locations may possibly be explained by the fact that the rate of deposition from the stratosphere increases with distance from the equator to a maximum at 40°-50° latitude.

Table 1. Plutonium Isotopic Ratios From Ground-Based Filtration Units

EDWARDS AIR FORCE BASE, CALIFORNIA			
Day (1975)	Volume Sampled (KSCM)	$^{240}Pu/^{239}Pu$	aCi/SCM
189	70.4	0.141 ± .002	5.5
190	12.2	0.154 ± .003	53.1
195	36.7	0.137 ± .001	41.4
197	28.2	0.144 ± .015	17.9
199	24.3	0.139 ± .004	161.7
204	25.2	0.141 ± .011	11.9
206	23.6	0.121 ± .004	15.2
211	24.9	0.102 ± .009	18.2
213	24.3	0.132 ± .004	25.5
273	37.0	0.138 ± .008	6.7
288	73.4	0.138 ± .003	4.3
289	11.8	0.163 ± .008	3.5
303	12.0	0.157 ± .013	2.1
305	26.5	0.139 ± .003	5.3
307	35.3	0.135 ± .006	5.9
317	24.3	0.159 ± .006	9.5
324	25.2	0.113 ± .004	10.2
329	34.8	0.140 ± .003	10.5
336	38.2	0.155 ± .008	4.9
29 (1976)	24.4	0.147 ± .004	9.3
31	25.0	0.176 ± .007	10.1
34	36.3	0.142 ± .005	8.1
37	23.8	0.186 ± .005	5.6
39	37.5	0.158 ± .014	3.9
43	24.5	0.171 ± .012	6.1
45	24.4	0.163 ± .005	16.4
48	44.6	0.152 ± .002	6.0
57	24.8	0.138 ± .002	13.8
64	28.8	0.158 ± .005	7.7
65	24.0	0.152 ± .008	5.5
69	37.1	0.151 ± .009	14.9
72	23.7	0.142 ± .003	29.5
76	37.7	0.110 ± .002	11.3
77	23.7	0.145 ± .003	15.6
84	24.6	0.143 ± .006	16.8
89	35.7	0.142 ± .002	10.6
91	25.5	0.161 ± .004	9.8

Table 1. Continued

RAMEY AIR FORCE BASE, PUERTO RICO			
Day (1975)	Volume Sampled (KSCM)	^{240}Pu/^{239}Pu	aCi/SCM
253	24.5	0.161 ± .007	6.3
258	36.7	0.149 ± .007	2.3
265	35.6	0.162 ± .009	2.2
281	24.4	0.164 ± .007	3.0
283	24.5	0.162 ± .007	4.1
309	24.5	0.144 ± .007	1.7
328	35.9	0.143 ± .009	1.0
332	24.5	0.154 ± .011	1.7
349	36.8	0.179 ± .010	5.7
356	36.8	0.155 ± .008	3.2
358	24.2	0.147 ± .007	1.9
363	61.2	0.161 ± .011	3.9
5 (1976)	39.6	0.151 ± .011	3.7
14	24.4	0.176 ± .007	2.5
28	24.6	0.158 ± .013	2.7
33	48.5	0.153 ± .004	1.9
40	36.6	0.142 ± .003	3.8
49	23.5	0.176 ± .006	7.2
51	24.4	0.180 ± .009	3.5
59	37.1	0.161 ± .007	2.8
83	49.5	0.162 ± .010	3.8

Table 2. Plutonium Activity Levels

EDWARDS AIR FORCE BASE, CALIFORNIA		
Day (1975)	Volume Sampled (KSCM)	Pu aCi/SCM
192	23.9	39.7
202	37.0	39.5
209	36.4	12.9
246	24.3	33.8
248	24.6	8.5
251	24.9	7.2
253	24.6	7.5
255	24.6	35.0
258	24.3	13.3
260	24.6	12.5
262	23.9	3.8
269	36.2	17.0
277	23.9	5.3
302	47.3	12.6
310	24.2	71.1
312	23.6	28.6
315	39.9	9.4
322	35.7	9.4
326	25.2	10.5

Table 2. Continued

RAMEY AIR FORCE BASE, PUERTO RICO		
Day of Year	Volume x 10^3 SCM	Pu aCi/SCM
(1975)		
248	24.5	11.2
251	36.7	8.5
255	24.4	28.2
260	24.6	3.8
262	25.4	9.7
267	24.6	3.3
269	24.4	2.7
272	36.7	2.9
274	24.5	8.7
276	24.4	4.3
279	36.8	6.1
286	36.7	2.2
288	24.4	4.3
290	28.4	1.4
293	32.8	1.7
295	24.4	2.8
297	24.6	2.2
301	48.7	2.1
302	24.4	4.7
307	36.5	1.8
311	36.8	4.6
314	36.7	1.5
316	24.3	2.5
321	36.8	1.9
323	24.2	3.5
325	25.2	1.2
330	24.5	2.9
335	36.5	2.5
339	49.0	2.6
342	36.8	2.9
344	24.6	4.0
351	24.6	4.8
353	24.3	3.8
365	24.5	9.4

Table 2. Continued

RAMEY AIR FORCE BASE, PUERTO RICO		
Day of Year	Volume x 10^3 SCM	Pu aCi/SCM
(1976)		
2	24.4	12.4
5	39.6	3.7
7	26.4	2.7
9	24.2	3.1
12	35.2	3.3
19	36.5	3.2
21	24.7	2.5
23	24.7	3.5
26	36.1	3.0
30	24.2	4.2
37	22.1	3.2
44	24.1	3.6
47	37.9	5.9
54	37.1	3.7

AMERICIUM

Many of the samples collected in the 1959-1970 background survey were also analyzed for their ^{241}Am content. Aircraft sampling was normally conducted in the vicinity of three latitudes: 70°N, 35°N, and 10°N. Stratospheric samples were collected quarterly at altitudes ranging from 40,000 to over 60,000 feet. For brevity, the details of the individual sample collection are omitted in this manuscript. The measurements were performed at McClellan Central Laboratory and Tracer Lab West--now called L.F.E. Environmental Laboratories.

Table 3 shows the results of the study for samples whose measurement uncertainty were less than 25%. It is interesting to note that the concentration of ^{241}Am as well as the ^{241}Am/^{239}Pu ratios vary from sample to sample. The amount of ^{241}Am formed is a function of both the age and irradiation history of the debris.

When sets of ^{241}Am data are averaged, general trends become apparent. Table 4 shows the average annual ^{241}Am and ^{241}Am/^{239}Pu levels obtained by averaging the data reported in Table 3 on a yearly basis. The data seem to indicate that as a first approximation, one may assume that all the atmospheric americium was injected as a point source in 1963. This burden decreased with an apparent half-life of 1.6 years.

It is interesting to note that the ^{241}Am/^{239}Pu atom ratio in the atmosphere increased yearly from 1963-1967. This was the result of intermixing of debris containing different relative amounts of ^{241}Am. To a lesser extent, the ^{241}Am/^{239}Pu ratio was influenced by the ingrowth of ^{241}Am from the decay of ^{241}Pu. From 1967-1970, the americium/plutonium ratio decreased in contradiction with expectations. The reason for this anomaly is that much of the old debris that was injected from 1956-1963, containing relatively large quantities of ^{241}Am, has been removed from the atmosphere. The fresh injections of debris during 1967 and subsequent years containing smaller quantities of ^{241}Am relative to ^{239}Pu were superimposed over the previous inventory. The net effect is an increase in the total atmospheric ^{239}Pu relative to ^{241}Am.

CONCLUSIONS

The ^{240}Pu/^{239}Pu isotope ratios in the atmosphere have changed with time. A possible explanation for this variation is that recent injections of debris play a major role in determining the characteristics of the transuranic elements in the atmosphere.

Measurements of stratospheric ^{241}Am from 1963-1970 seem to indicate that as a first approximation, the ^{241}Am burden of the atmosphere may be considered as a point source injection during 1963. The ^{241}Am concentration of the atmosphere decreased from a high of 5.92 x 10^6 atoms/SCM in 1963 to 0.27 x 10^6 atoms/SCM

Table 3. ^{241}Am Background Summary 1963-1970

LOCATION		70N		35N		10N	
Atoms Exp	Alt (Feet)	-06/SCM	+03/N239Pu	-06/SCM	+03/N239Pu	-06/SCM	+03/N239Pu
1963	40K	2.60	1.34				
	40K	2.29	1.35				
	50K	4.53	1.16				
	50K	4.33	1.16				
	60K	7.02	1.07	6.85	1.03	5.83	1.19
	60K	7.02	0.99	5.92	0.95	6.57	1.29
	60K	7.28	0.50	7.05	0.91	8.69	1.57
	60K	6.23	0.94				
	60K	6.51	0.98				
1964	40K	1.59	0.47				
	40K	2.26	0.25				
	40K	1.18	0.14				
	40K	3.03	1.42				
	40K	3.00	1.31				
	40K	2.77	1.18				
	40K	2.23	1.83				
	40K	1.21	0.18				
	50K	4.10	1.10	1.86	1.22		
	50K	4.16	1.18	1.63	1.06		
	50K	3.77	1.38	0.93	1.36		
	50K	2.47	0.99	0.73			
	50K	1.98	1.47	1.00	1.64		
	50K	2.20	1.50	0.77	1.68		
	50K	0.32	0.19	0.71	1.53		
	50K	0.30	1.03	0.40	0.95		
	60K	2.52	0.15	3.11	0.18	4.13	1.69
	60K	1.13	1.29	0.70	1.35	4.22	1.92
	60K	2.60	2.14	0.35	1.26	3.77	1.70
	60K	2.92	1.33	0.35	1.24	3.33	1.78
	60K	2.60	1.56	3.08	1.15	3.34	1.33
	60K	2.17	1.43	6.71	1.58	4.53	1.40

Table 3. Continued

LOCATION Atoms Exp	Alt (Feet)	70N		35N		10N	
		-06/SCM	+03/$N^{239}Pu$	-06/SCM	+03/$N^{239}Pu$	-06/SCM	+03/$N^{239}Pu$
	60K	0.29	1.03	0.36	0.95		
	60K	2.60	1.07	4.93	1.06		
	60K	4.47	1.07	4.92	1.06		
	60K	2.61	1.07	4.92	1.06		
	60K	4.47	1.38	4.25	1.33		
	60K	4.22	1.31	2.83	1.28		
	60K	4.47	1.38	4.25	1.33		
	60K	4.21	1.31	2.83	1.28		
	60K	3.37	1.20	3.20	1.00		
	60K	3.12	1.24	3.51	1.55		
	60K	2.63	1.35	3.06	1.28		
1965	40K	1.15	1.96	4.93	2.20	8.52	2.16
	40K	2.86	2.76				
	40K	1.81	2.06				
	40K	1.60	1.83				
	40K	0.95	2.56				
	40K	0.73	2.11				
	40K	0.95	2.56				
	40K	0.73	2.11				
	40K	0.95	2.56				
	40K	0.73	2.11				
	40K	0.39	2.70				
	40K	0.14	0.86				
	40K	1.34	1.82				
	50K	2.83	1.73	3.06	1.85	6.20	2.91
	50K	2.68	1.72	3.90	2.03	4.47	1.74
	50K	0.32	5.03	0.33	1.98		
	50K	2.70	1.99	1.52	2.37		
	50K	0.16	2.17	1.36	2.16		
	50K	1.98	2.31	3.59	3.59		

Table 3. Continued

LOCATION Atoms Exp	Alt (Feet)	70N		35N		10N	
		-06/SCM	+03/$N^{239}Pu$	-06/SCM	+03/$N^{239}Pu$	-06/SCM	+03/$N^{239}Pu$
	50K	1.44	2.01	0.19	1.86		
	60K	1.42	2.97	1.30	2.87	1.57	3.23
	60K	1.23	2.53	0.96	1.82	1.71	2.67
	60K	1.66	3.30	1.75	2.31	2.76	2.53
	60K	0.99	1.91	1.73	1.92	2.32	2.06
	60K	2.97	2.16	1.85	2.60	2.09	3.16
	60K	2.43	1.60	1.69	2.26	1.43	2.41
	60K	1.66	2.06	2.18	2.20	0.31	1.97
	60K	2.50	2.97	1.73	1.85	1.87	3.54
	60K	1.83	1.93	1.72	1.92	2.32	2.06
	60K	1.66	3.30	1.73	2.31	2.76	2.53
	60K	0.99	1.91	1.72	1.92	1.54	2.21
	60K	1.42	2.97	1.30	2.87	1.58	3.23
	60K	1.23	2.53	0.96	1.82	1.71	2.67
	60K	1.67	1.78	2.77	1.95		
	60K	0.93	1.60	2.30	1.89		
	60K	1.90	1.53	1.87	3.54		
	60K	3.63	1.79	1.54	2.21		
	60K			2.25	2.45		
	60K			2.60	2.66		
1966	40K	0.99	2.05	0.31	2.41		
	50K	1.13	2.49	0.36	2.56		
	50K	1.71	2.22	1.18	2.24		
	50K	0.06	2.35	0.22	2.83		
	60K	2.13	2.43	1.68	2.54	0.45	2.44
	60K	1.21	2.52	1.39	2.72	1.01	2.83
	60K			1.24	2.39	1.03	2.86
	60K					0.90	2.91

Table 3. Continued

LOCATION Atoms Exp Alt (Feet)		70N -06/SCM	 +03/N^{239}Pu	35N -06/SCM	 +03/N^{239}Pu	10N -06/SCM	 +03/N^{239}Pu
1967	40K	0.44	3.08	0.17	2.56		
	40K	0.25	2.47	0.07	3.38		
	40K	0.28	2.97				
	50K	1.16	2.80	0.43	2.37		
	50K	0.59	2.06	0.36	2.76		
	50K	0.56	2.53				
	60K	0.89	2.79	1.10	3.12	0.50	2.55
	60K	0.66	2.58	0.93	2.69	1.03	2.77
	60K	0.86	3.19		3.01	1.08	3.06
	60K	0.30	3.44	0.38	0.82	0.73	2.98
	60K			0.87	1.53		
1968	40K	0.39	0.97	0.19	1.45		
	40K	0.34	1.18				
	40K	0.40	0.85				
	50K	0.52	1.25	0.24	1.01		
	50K	0.78	0.96	0.25	1.05		
	50K	0.73	0.68	0.13	1.35		
	60K	0.85	0.78	0.87	1.87	0.51	1.92
	60K	0.35	0.81	0.64	0.76	0.46	1.69
	60K	0.40	0.85	0.76	0.80	0.45	1.56
	60K	0.31	1.71	0.91	0.93		
	60K	0.20	1.05	0.67	1.36		
	60K	0.17	1.63	0.62	1.60		
	60K			0.49	1.60		
1969	40K	0.26	1.47	0.03	4.51		
	40K	0.09	0.41				
	40K	0.20	0.95				
	50K	0.27	0.86				
	50K	0.28	1.06				

Table 3. Continued

LOCATION Atoms Exp Alt (Feet)		70N -06/SCM	 +03/N^{239}Pu	35N -06/SCM	 +03/N^{239}Pu	10N -06/SCM	 +03/N^{239}Pu
	50K	0.80	3.76	0.33	1.04	0.22	1.62
	60K	0.30	1.44	0.35	0.97	0.46	2.00
	60K	0.71	1.61	0.25	0.74	0.33	1.59
	60K	0.37	2.00	0.39	1.13	0.56	1.36
				0.26	0.73	0.30	1.12
				0.34	0.10		
				0.22	0.80		
1970	40K	0.31	0.77	0.18	0.98	0.01	2.02
	40K	0.25	0.88				
	50K	0.43	0.41	0.20	0.48		
	50K	0.50	0.35	0.04	0.35		
	60K	0.27	0.39	0.46	0.64	0.21	1.00
	60K	0.24	0.52	0.43	0.48		
	60K	0.19	0.49	0.51	0.48		
	60K	0.19	0.65	0.23	0.31		

Table 4. Average Annual Americium Values

Date	^{241}Am x 10^{-6} atoms/SCM	^{241}Am/^{239}Pu atom ratio
1963	5.92	0.0011
1964	2.68	0.0012
1965	1.91	0.0023
1966	1.00	0.0025
1967	0.62	0.0027
1968	0.49	0.0012
1969	0.33	0.0014
1970	0.27	0.0007

in 1967 with an apparent half-life of 1.6 years. This indicates that ingrowth from decay of ^{241}Pu is a negligible source of ^{241}Am in the atmosphere. From 1967-1970, the ^{241}Am/^{239}Pu atom ratio decreased because the injections of plutonium contained relatively smaller quantities of americium.

ACKNOWLEDGMENTS

Thanks are given to SSgt Thomas W. Nibarger and Howard V. Hamilton of the Chemistry Section of McClellan Central Laboratory for separating and purifying the plutonium samples reported in this study. Special thanks are given to Mrs. Marcene Beckinger for her help in the preparation of this manuscript.

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DEVELOPMENT OF SPECIALIZED INSTRUMENTS AND TECHNIQUES FOR RESUSPENSION STUDIES

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(Ed. Note: Previously published in
NAEG Report NVO-142, pp. 221-233.)

ABSTRACT

An intensive study on the resuspension of plutonium at the Nevada Test Site has been initiated. The main thrust of the study is to develop a mathematical model for describing the concentrations of plutonium in air as a function of the source and driving forces. It is intended that the model be a basis for assessing potential health hazards and developing cleanup recommendations. In addition, the study is providing an opportunity to develop a generalized resuspension model, applicable in general for establishing environmental and biological assessment of nuclear projects involving plutonium.

Apparatus and experimental techniques for studying the dynamics of plutonium and soil particle behavior have been developed. This has included the development of ultrahigh-volume air samples (1500 m^3/hr) which allow collection of adequate samples of plutonium at worldwide air concentration levels in two hours of sampling time. We have also developed samplers for measuring the fraction of particles moving in creep and saltation. The Air Resources Laboratory, National Oceanic and Atmospheric Administration, has established an elaborate meteorological data-gathering system for measuring micrometeorological parameters, during sampling periods at the Nevada Test Site. Advanced optical methods for *in situ* sizing and counting particles have also been employed. This technique allows observations of particle size and concentration of a few-minute time scale, such that rapid changes in meteorological parameters may be more directly correlated with the concentration of airborne particles and their size distributions.

The most intensive field program to date has been in the GMX area. Data have also been collected in Area 13 and Mercury. Ultrahigh-volume air samplers and high-volume cascade impactors were used to measure the concentration of plutonium in air and aerodynamic particle fractions of airborne soil and plutonium. Analysis of air samples collected by REECo from February, 1971, to July, 1972, shows that the GMX site, which was contaminated 17 years ago, still represents a significant resuspension source. However, the average air concentration of

resuspension source. However, the average air concentration of resuspended ^{239}Pu outside the exclusion area is only a small fraction of the presently accepted maximum permissible concentration for occupational exposure. It was also concluded that the air concentrations of ^{239}Pu in Mercury may be influenced by the local NTS sources.

Measurements using cascade impactor studies indicate that there is no difference in the distribution of activity with particle size for the three species ^{238}Pu, ^{239,240}Pu, and ^{241}Am. The total mass, however, was found to be distributed distinctly differently with a smaller median aerodynamic diameter and a larger geometric mean. The data also showed that the fraction of the resuspended plutonium aerosol at GMX which would be expected to undergo pulmonary depositions to be approximately 0.2 based upon the ICRP Task Group on Lung Dynamics model.

Experimental results have also shown that there is no obvious correlation of specific Pu activity with particle size. An average specific activity of 890 dpm/g, or about one-third of that found in the soil in close proximity to the cascade impactors, was measured. Particle data from the cascade impactors showed that the ratio of ^{239,240}Pu to ^{241}Am activity is the same as reported for soil in the vicinity of the cascade impactors. Preliminary results from the ultrahigh-volume air sampler runs indicate a gross correlation between many of the wind speed-related parameters and the concentration of resuspended plutonium.

Experience and data gathered at the GMX site were used to derive simple predictive models for air concentrations of plutonium due to resuspension on the Eniwetok Atoll.

INTRODUCTION

The Resuspension Element of the Nevada Applied Ecology Group (NAEG) was formed to undertake the task of studying the movement of plutonium at the Nevada Test Site (NTS) by wind-driven forces and assessing the potential biological hazards associated with airborne plutonium particles. The element also was to provide input to the Nevada Operations Office of the Atomic Energy Commission regarding cleanup of plutonium-contaminated areas. This involvement gave the Lawrence Livermore Laboratory (LLL) Bio-Medical Division an opportunity to study fundamental processes involved in the resuspension of plutonium particles from a soil surface, and to develop a time and spatial dependent mathematical model describing average concentration of airborne plutonium as a function of the source (geometrical configuration, soil surface characteristics) and driving forces. It is intended that the model will also provide the necessary input to models of lung dynamics of plutonium behavior in the human respiratory tract. The NAEG Resuspension Element's plan for meeting the above objectives is diagramed in Fig. 1.

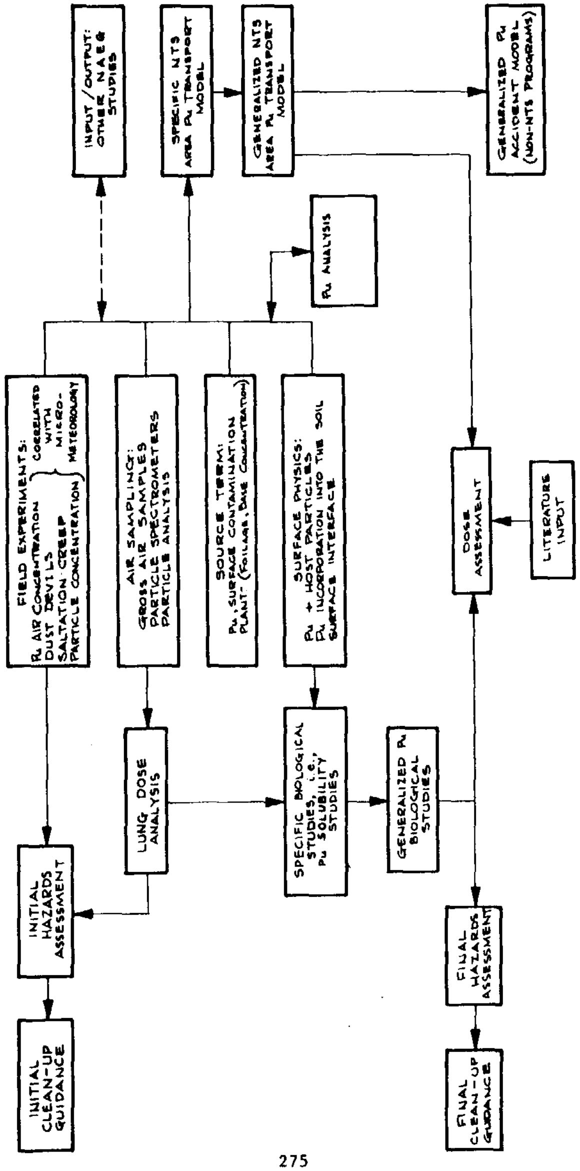

FIG. 1. THE INTERRELATIONSHIP BETWEEN THE FIELD EXPERIMENT (DATA GATHERING) AND OBJECTIVES OF NAEG.

Prior to LLL involvement in the NAEG program, Livermore had carried out resuspension studies related to nuclear cratering experiments and accidental releases from nuclear device tests. One of these studies was in conjunction with the Schooner cratering experiment (Anspaugh *et al.*, 1969; Anspaugh *et al.*, 1971; and Anspaugh *et al.*, in press).

Radioactivity measurements were made on an arc approximately 50 miles from the Schooner surface ground zero (SGZ). Some of the highest readings were obtained in the vicinity of the Queen City Summit outside the northern boundaries of NTS. Based upon conjecture that the high readings were caused by unique interactions between wind flow and topography at the Queen City Summit region which had existed during the Schooner cloud passage, it was hypothesized that similar conditions could have existed during atmospheric device testings in previous years at NTS which could have resulted in unusually high plutonium depositions at the same site.

Consequently, soil samples were analyzed for plutonium and data were obtained in support of the hypothesis. Plutonium levels in soil at Queen City Summit were found to be 100 times over the average background levels in U.S. soil. Subsequent measurements were made by the Environmental Protection Agency's National Environmental Research Laboratory, Las Vegas (formerly the U.S. Public Health Service), which confirmed the LLL's earlier findings. These findings added impetus to carry out resuspension studies at NTS.

Subsequent to starting the plutonium resuspension field studies, the opportunity arose to conduct a long-term experiment using the radioactivity deposited on the soil surface during the Baneberry accidental venting (Anspaugh *et al.*, in press). The incident occurred in December, 1970. The study was carried out over approximately a one-year period. The study was directed at observing the resuspension of radioactivity as a function of time.

Following the Baneberry study, efforts were directed toward designing a series of experiments at plutonium-contaminated areas on NTS. A joint proposal from LLL and NOAA/ARL outlining the experimental plan was submitted for AEC review. Early in 1972, research, development, and procurement were started on special apparatus needed to carry out the experiment. This included developing ultrahigh-volume air samplers, in-situ particle spectrometers, light-scattering-type particle counters, and establishing a micrometeorology field laboratory. A plutonium-contaminated site was selected and surveyed for layout of the experiment.

RESUSPENSION EXPERIMENT: GMX-AREA 5

The GMX Site, in Area 5, was selected for conducting the first plutonium resuspension experiment. This area had previously been used for carrying

out a series of experiments involving plutonium and high explosives. At no time were nuclear detonations involved. These plutonium-high explosive tests started in late 1954 and ended in early 1956. Thus, as of this date, the mean life of the plutonium source at GMX is 18 years. Following the 1954-1956 series of tests, the highly contaminated area was fenced off and remained essentially undisturbed.

Various groups within the NAEG program have gathered data which support the resuspension studies. These data include the areal distribution of plutonium, the distribution and concentration of plutonium within the soil, and a population census of vegetation covering the GMX study area.

The initial resuspension experiment was designed with the goal of developing a model to predict air concentrations of plutonium, given the source characteristics and relevant meteorological parameters. The objective was then to conduct a series of measurements within and downwind of the plutonium source. This was to be done during a period of time when the winds were consistent. Examination of several years of wind data showed that this was possible for a 2-4 hr period. The experiment was laid out as shown in Fig. 2. The line showing the sampling stations is located at 020° true, which corresponds to the mean direction of winds over 10 mph from March 24 to August 30, 1971, at the GMX site.

Micrometeorological measurements were directed at characterizing the driving forces; i.e., winds, turbulence, and related parameters that are considered to be most likely related to moving or picking up particles from the desert pavement. These measurements are discussed in detail by Kennedy and Booth (this report). The main micrometeorology station was established 1200 ft downwind from GZ, at R-1, as shown in Fig. 2. Air-sampling stations were established at GZ, 1200 ft, 2400 ft, and 4800 ft from GZ. In addition to the downwind sampling stations, control stations were established upwind from GZ, at Mercury, and in Livermore, California. These background stations utilized the identical type of ultrahigh-volume air sampler.

During a field experiment, which usually lasted from 1-4 hr, the various micrometeorological parameters were recorded and the ultrahigh-volume air samplers were operated. All the samplers were synchronized to start and stop together. Concurrent with these measurements, particle spectrum analyses at various heights above the ground were made using the Climet particle analyzer (Koval, this report). Also overlapping in time with these measurements, background air samples were collected 900 ft upwind from GZ; at Mercury, Nevada; and in Livermore, California.

Saltation/creep samplers have been installed for collecting particles that roll or bounce on the soil surface (Reichman, this report). These samplers as yet have not been integrated into the field experiments. It is planned to finish evaluation studies in 1974 and use the instruments shortly afterwards.

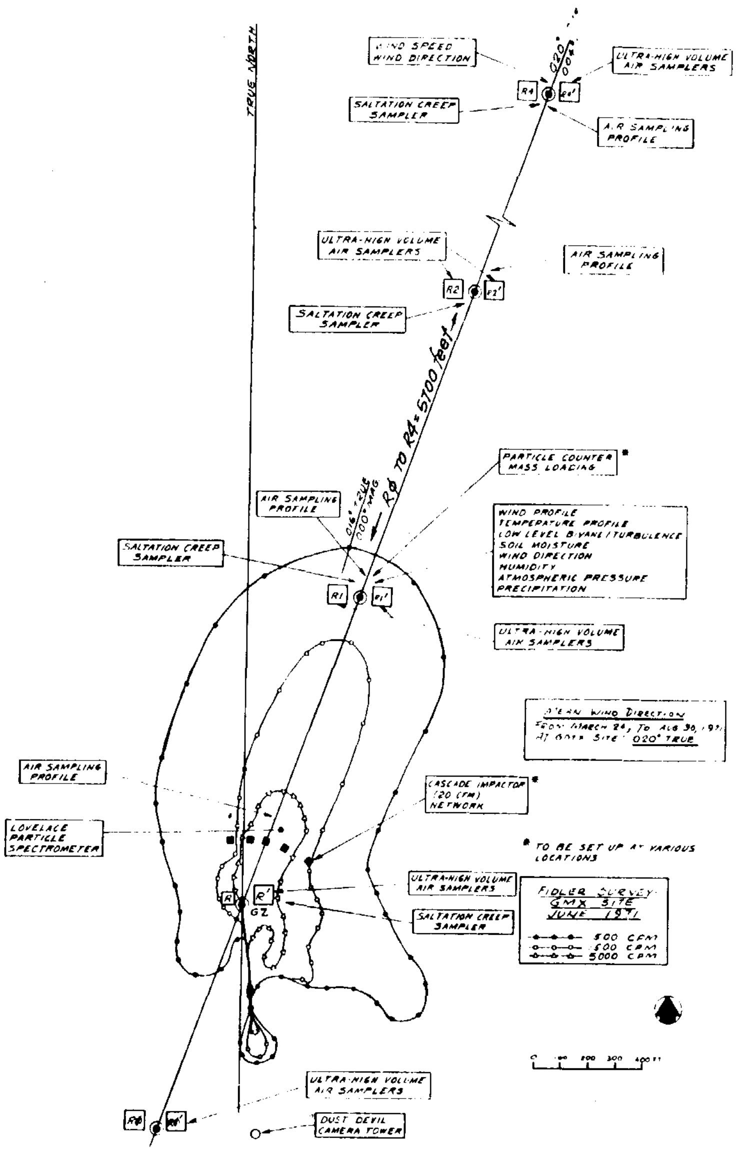

FIG. 2. LAYOUT OF THE RESUSPENSION EXPERIMENT AT GMX, AREA 5.

Cascade impactors were located on an arc 250 ft from GZ, and were run continuously for approximately 30-day periods. These instruments gave results on particle size fractions for plutonium and total airborne particulates. Results are given by Anspaugh and Phelps (this report).

A time-lapse camera on an 80-ft tower was located upwind from GZ such that the entire GMX experimental site was in its field of view. The camera has been operated during the daylight hours with a frame exposure every 5 sec. During the times of high dust devil activities (June-September), the camera was used to record dust devils traversing the GMX experimental site. This was done to qualitatively assess the impact of dust devils on transporting surface material into the atmosphere. The data which were available for the GMX experiment are presented in detail by Anspaugh and Phelps (this report). A preliminary analysis of the air concentration data collected by REECo prior to the initiation of this study is also presented by Anspaugh and Phelps (this report).

DEVELOPMENT OF SPECIALIZED INSTRUMENTS AND TECHNIQUES

A variety of specialized instruments had to be designed and fabricated before the plutonium redistribution experiments could be conducted at NTS. One of the major undertakings was the development of an air sampler capable of collecting adequate amounts of airborne plutonium particulates during a 2-4 hr sampling period. This time period corresponded to the time "window" during which consistent meteorology could be expected. Calculations indicated that even if the most sensitive radiochemical procedures were used in analyzing the air filters for plutonium, an air sampler with a through-put at 1500-2000 m^3/hr would be required. Samplers with these required flow rates were not available; therefore, development of an ultrahigh-volume air sampler meeting the desired specifications proceeded. Tests on the prototype sampler were very encouraging, indicating that even at global background levels, surface air could be sampled for a time period of 1 hr, and accurate determinations of plutonium air concentrations could be made. Fourteen ultrahigh-volume air samplers were eventually fabricated. They are deployed in Area 13, GMX, and Mercury. One unit is in operation at Livermore.

Micrometeorological forces that may cause resuspension of particles from the soil surface have significant transient behavior over very short periods of time, probably less than a second to a few minutes. The airborne particles resulting from resuspension normally are collected by use of a filter media -- airpump combination. The collected particles are then analyzed for plutonium, and the concentration of radioactivity in air is determined from the ratio of plutonium to the amount of air sampled. Therefore, since sampling must be done over a minimum of 1 hr, which corresponds to the least time possible with the ultrahigh-volume air sampler previously mentioned, mechanisms related to pickup of particles from the soil surface cannot be studied in real time and with high resolution.

To overcome these limitations, two light-scattering instruments were adapted to measure the mass loading of particles in air and the number distribution as a function of particle size over very short periods of time. Both instruments give data in real time, such that it is possible to correlate micrometeorological events of short duration with the concentration of airborne particles. These instruments examine the particle spectrum in the region of 0.1 to 10 μm, and do not differentiate between radioactive and nonradioactive particles. Nevertheless, valuable data have been obtained on the mass loading of particles in air as a function of the forces causing pickup from the soil surface.

The first instrument employed was a light-scattering-type particle analyzer. This instrument, when used with a pulse-height analyzer, will give particle size histograms over a range of 0.5 to 10.5 μm. Sufficient data can be collected in periods of 2 min or less. Particle sizes as measured by this technique are related to their physical diameters. If the physical properties of the particles are known, aerodynamic or "effective" particle sizes can be determined. This is necessary for modeling of particle deposition in the human respiratory tract. Details of the instrument and its application to the GMX experiment are covered by Koval (this report). Another light-scattering device currently employed is the nephelometer. It has extremely fast response (< 1 sec); however, it does not give particle size distribution data, only relative particle concentration. It is an exceptional instrument for correlating very short-time duration meteorological events with mass loading of particles in the air.

A recently introduced high-volume (20 cfm) cascade impactor was adapted for use in the GMX experiment. This instrument has five stages which correspond to 1.1, 1.1-2.0, 2.0-3.3, 3.3-7.0, and greater than 7.0 μm. The filter papers may be analyzed for total plutonium per stage and total mass for all particles on each stage. Thus, the particles picked up from the soil surface may be characterized according to their aerodynamic sizes, and the relationship between plutonium and total mass may be established. Further refinements are being made on this instrument which will reduce reentertainment of particles and improve its particle intake characteristics.

Particles that roll or bounce along the soil surface (creep/saltation) may be responsible for the most significant net movement of surface mass compared with other processes. Existing instruments that had been used in agricultural erosion studies and for measuring the movement of sand in windstorms were evaluated. A series of laboratory studies were conducted at LLL on borrowed or copied instruments. Out of this work, a multistage sampler was developed, capable of collecting the particle fraction moving by creep and particles moving in saltation at various levels above the surface. Details on the design and preliminary results are covered by Reichman (this report).

Initial dust devil observations were made with a time-lapse camera adapted for use on an 80-ft tower. The camera was programmed to expose one frame every five sec. Experimentation with optimum camera location and lens selection resulted in ability to view the entire GMX experimental site. This system was developed as a survey tool in order to get an approximate idea of the dust devil population. A visual count in the GMX and surrounding area during a 1-hr period in July, 1973, showed a dust devil population of approximately one dust devil formation per minute, with an average lifetime of about 2 min.

An elaborate meteorological data-gathering system, capable of measuring wind and temperature profiles, fluctuations of the horizontal and vertical wind structure, soil moisture, soil temperature, humidity, rainfall, and other miscellaneous meterological measurements, was set up. This was under the direction of the Air Resources Laboratory-Las Vegas, and is covered in detail by Kennedy and Booth (this report).

FUTURE STUDIES

A number of problems dealing with understanding redistribution of radioactivity at NTS still remain. These problems are related to possible long-term health and safety issues, future land uses of NTS, and other AEC nuclear programs. So far, LLL studies have involved the movement of plutonium at the GMX site, and redistribution of gamma emitters released from the Schooner cratering event and the Baneberry device test. To date, the GMX studies have focused on plutonium particles carried into suspension by the force of the wind. Emphasis has been on collecting samples of airborne plutonium particles and measuring a number of micrometeorological parameters, such that the relationship between air concentrations of plutonium and the important driving forces may be understood, and a mathematical model constructed. The wind forces studied so far have been diurnal in nature, and occasionally those associated with dust storms.

Dust Devils

There is, however, another driving force: dust devils, which might be a significant mechanism under certain circumstances for transporting surface radioactive material into the atmosphere.

Because of the high wind speeds generated close to the ground and the large vertical structure, dust devils are capable of removing and transporting vertically a significant portion of loose surface material. Dust-devil measurements by Sinclair (1966, 1973) indicate that on the average, a typical dust-devil source region of 285 km^2 provides a total mean seasonal vertical dust transport of over 7×10^3 tons of desert sand. This is one to two orders of magnitude larger, for the same surface area, than pre-dust storm conditions (Gillette *et al.*, 1972) and smaller by approximately the same amount within the most intense dust storms. Consequently, the vertical and horizontal transport of surface material by dust devils may in itself be an important component in the overall resuspension budget.

However, of more importance is the question of transport and ultimate fate of radioactive materials by dust devils and, in general, by atmospheric convective elements that originate in the surface boundary layer. LLL plans to make direct quantitative measurements of the amount, the distance, and the direction of dust transported from a known dust devil source region at NTS. The analysis of these data will provide estimates of the possible radioactive substances such as plutonium by dust devils.

Variation in Site Characteristics

The availability of plutonium for resuspension is related to a significant degree to the soil-plant surface characteristics of the plutonium-contaminated area. This is particularly true for aged sources that are found at NTS. A number of plutonium-contaminated sites exist at NTS. Each of the sites have more-or-less different and unique characteristics that influence the availability and movement of plutonium. The properties of importance are the initial physical-chemical form of the source, source age, source strength (surface related), source area and shape, physical-chemical makeup of the soil, soil surface structure, plant coverage, animal population, climatology, and land utilization. These properties are to be intensively studied.

An example of an area not yet studied is the Tonopah Test Range. This area is suitable for cattle grazing. A native species of grass, not found in the GMX area, is in abundance. Also, the physical "desert pavement" characteristics are significantly different than those so far studied. The Tonopah area affords an opportunity to study a plutonium-contaminated area with surface characteristics not heretofore studied.

Man-Made Effects on Redistribution

Displacement of surface soil by mechanical means can increase the rate of redistribution. The impact from future uses of NTS is of particular interest. In addition to continued device testing, future use may include radioactive fuel processing, radioactive waste storage, solar energy research, and agricultural uses. Coupled with these uses are a variety of ways that sufficient mechanical disturbances to contaminated sites can result. These include construction of facilities and roads, vehicular traffic over desert pavement, and grazing by cattle. A number of the areas contaminated from past device testing and nuclear cratering events should be studied. A natural follow-up experiment in the GMX area would be to introduce a variety of man-made disturbances to the soil surface and quantitate the change in resuspension of plutonium.

Source Term Analysis and Redistribution of Radioactivity at NTS

An evaluation of the total NTS contribution of dose to man is limited by the lack of accurate mapping of the resuspendable radioactive source material distributed throughout the site. However, elements within the NAEG program are compiling information on several radionuclides and their distribution in soil, which, if extended to all radionuclides and areas, could be the basis for input to such a mapping.

Assuming that a data bank on radionuclide distribution at NTS were established, the other necessary inputs would be a comprehensive modeling of the meso-scale meteorology related to wind flow at NTS, and an understanding of resuspension for the variety of source characteristics present throughout the site.

This study would provide information on the relationship between source and the dose to man from existing radioactivity and possible future uses.

ACKNOWLEDGMENTS

The authors wish to thank the numerous persons who have contributed to this study. Members of the Lawrence Livermore Laboratory who constructed and fielded the experimental apparatus, at times under very trying conditions, included J. Taylor, C. Fry, F. Green, V. Fowler, and B. Tuckey. R. Kauder of the Air Resources Laboratory-Las Vegas was a principal participant in the meteorological measurements and data handling efforts. We also wish to thank I. Aoki and A. Bicker of the Reynolds Electrical and Engineering Co., Inc., for supplying air-sampling data for the GMX area, and R. Cooper, who was extremely helpful and resourceful in maintaining the field apparatus. Several members of the U.S. Atomic Energy Commission, Nevada Operations Office, staff have contributed their support. They include P. B. Dunaway, M. G. White, R. Lease, and H. Kayuha. Finally, a special thanks to S. Lewis for typing the manuscript, D. Schilf for typing the numerous reports, and to A. Boyce, who did the illustrations.

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PLUTONIUM CONTAMINATION OF VEGETATION IN DUSTY FIELD ENVIRONMENTS

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ABSTRACT

Transport of plutonium in food chains of grazing animals and mankind by vegetation carriers becomes an important avenue of contamination in dusty field environments. Findings indicate that most of the activity present in vegetation of such areas at the Nevada Test Site (NTS) is superficial contamination resulting from the attachment of particles to foliage surfaces during resuspension. We suspect, however, that the root uptake pathway eventually will become more significant as the result of natural concentration and recycling processes at work in the field within the plant root zone.

INTRODUCTION

Our assignment for this symposium is the consideration of the problem of plutonium contamination of vegetation in dusty field environments. In accepting this challenge, we acknowledge several timely reviews and discussions concerning various aspects of transuranic element behavior in ecosystems which have appeared in recent literature (Bernhardt and Eadie 1976; Brown 1976; Carfagno and Westendorf 1973; Dahlman et al. 1976; Francis 1973; Hakonson 1975; Hanson 1975; Healy 1974; Mullen and Mosley 1976; Price 1973; Romney and Davis 1972; Stannard 1973). We shall address the problem of vegetation-carrier transport of plutonium in relation to certain radionuclide cycling processes at work in the desert environment as illustrated in Fig. 1. The pathways of ingestion and inhalation are included in this illustration to give symmetry to the overall problem, but discussion of them is assigned to other participants in this symposium.

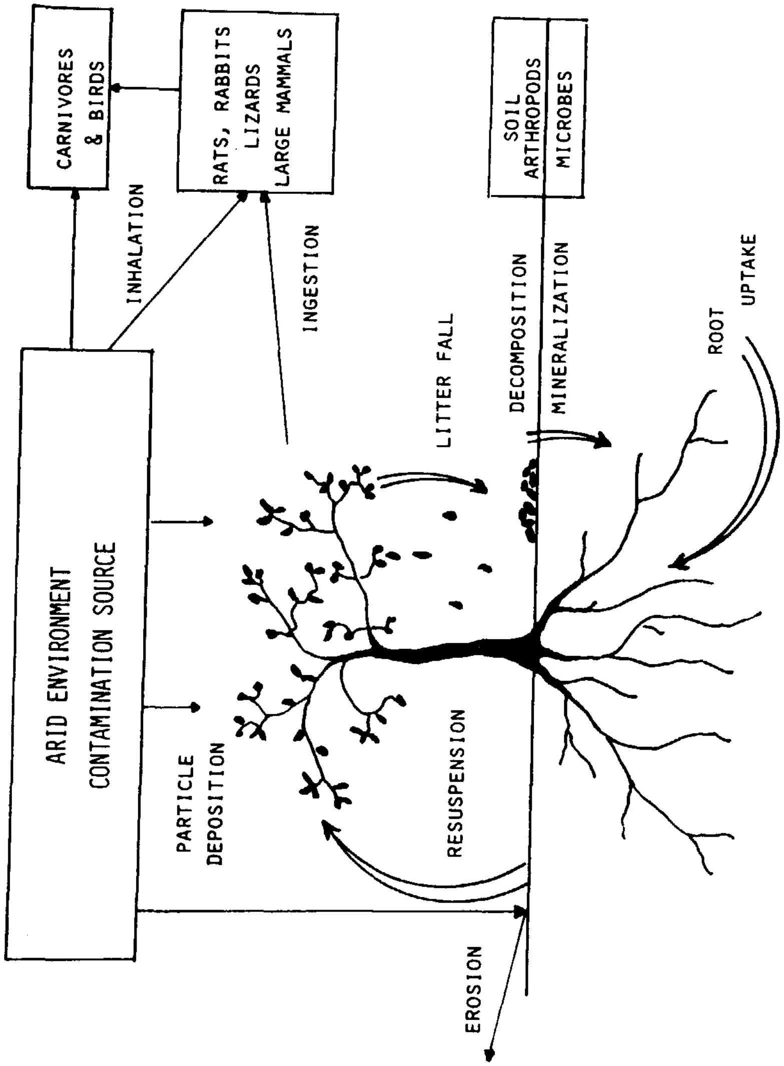

Fig. 1. Simplified illustration of pathways of natural cycling and concentration processes at work in the desert ecosystem.

AIRBORNE FALLOUT DEPOSITION

Earlier studies of fallout from nuclear weapon tests disclosed the biological significance of superficial contamination on plant surfaces resulting from radioactive airborne particle deposition (Fowler (ed) 1965; Larson *et al*. 1966; Romney *et al*. 1963; Russell (ed) 1966). Radioactive materials may reach the tissue of plants in two principal ways: First, airborne materials may be deposited upon the above-ground parts of plants and either adhere to their surfaces or be absorbed. Second, materials which have entered the soil may be absorbed by roots along with the nutrients on which plants depend for growth (Russell (ed) 1966). The significance of the root uptake-pathway is governed largely by the biological availability of each given radionuclide during its interactions in soil. Thus, if radioactive materials are insoluble they will most likely contaminate plant tissues only superficially (Romney *et al*. 1963; Martin 1965).

Early studies done by the UCLA groups on vegetation in fallout contaminated areas at the Nevada Test Site indicated that several natural processes influenced the fate and persistence of fallout debris both from nuclear and non-nuclear contamination events. The period during and shortly after particulate deposition was characterized by conditions of instability largely controlled by wind activity. Rainfall, snow, and periods of calm air movement near ground level hastened later development of a quasi-stable condition wherein particulate movement occurred primarily through processes governing resuspension. Vegetation contamination could be prevented or markedly reduced by protective covering during the unstable and quasi-stable periods after fallout had been deposited (Rhoads *et al*. 1971; Romney *et al*. 1971). Measurements of fallout particles on soil and plant material sampled within downwind fallout patterns showed that a partitioning into different sized particles normally occurred during initial fallout deposition. Thus, the mean particle size generally decreased at greater distances downwind from ground zero (Larson *et al*. 1966).

More recent studies by the NAEG in aged fallout areas at NTS and Tonopah Test Range (TTR), where plutonium was dispersed by chemical explosives, give evidence that this partitioning and patterning has continued to be reflected in the superficial contamination of the indigenous vegetation (Romney *et al*. 1974, 1975, 1976a, 1976b). The vegetation-to-soil inventory ratios determined in the various activity strata within several different fallout patterns seem to show that a greater proportion of the deposited $^{239-240}$Pu source material has moved onto vegetation at greater distances away from ground zero. Examples are given in Table 1. Inasmuch as the activity entrapped on plant foliage primarily represents material in the resuspendable particle size range, the amount of contamination on foliage is less in proportion to the total amount of fallout activity deposited on soil at points nearer to ground zero compared to points farther away. Autoradiographs of annual plant leaf tissues collected near ground zero at Area 13 in 1976 showed that the activity present on vegetation was still in discrete particles of suspendable size range nearly 20 years after fallout had occurred (Wallace, unpublished data).

Table 1. Estimated Inventory of $^{239-240}Pu$ for Vegetation in Activity Strata of Aged Fallout Areas at NTS (Romney et al. 1976). Higher numbered activity strata are nearer ground zero.

Activity strata	n	Mean ± S.E. (n Ci/g dry)	Mean[a] ± S.E.[b] (n Ci/m^2)	Inventory ± S.E. (millicuries)	Percent	Veg. invent. / Soil invent. ± S.E.[c]
NTS AREA 11, SITE A (1956)						
1	12	.0015 ± .00057	.76 ± .35	.095 ± .045	95	.0029 ± .0018
2	18	.00064± .000099	.33 ± .11	.0026 ± .00082	3	.0050 ± .0020
3	6	.0010 ± .00038	.54 ± .24	.00026 ± .00011	0.3	.00031 ± .00033
Total	36			.098 ± .045	100.3	.0028 ± .0017
NTS AREA 11, SITE B (1956)						
3	11	.10 ± .024	60 ± 40	.58 ± .14	24	.0020 ± .00085
2	14	.19 ± .053	110 ± 31	.78 ± .23	33	.00049 ± .00019
4	19	.57 ± .087	330 ± 52	1.0 ± .16	43	.00023 ± .000055
Total	44			2.4 ± .31	100	.00039 ± .000058
NTS AREA 11, SITE C (1956)						
2	12	.14 ± .026	61 ± 12	1.2 ± .23	27	.0018 ± .0014
3	14	.36 ± .054	160 ± 25	1.0 ± .16	24	.0018 ± .00052
4	17	1.1 ± .41	490 ± 180	1.9 ± .72	44	.00035 ± .00013
5	5	1.2 ± .38	530 ± 170	.21 ± .069	5	.000088 ± .000049
Total	48			4.3 ± .78	100	.00048 ± .000089
NTS AREA 11, SITE D (1956)						
2	12	.17 ± .033	72 ± 14	2.5 ± .48	35	.0016 ± .00034
3	13	.24 ± .079	99 ± 33	1.4 ± .46	19	.00045 ± .00025
4	20	.72 ± .14	300 ± 58	1.7 ± .32	23	.00031 ± .00011
5	11	1.3 ± .21	550 ± 89	1.7 ± .28	23	.00020 ± .000088
Total	56			7.3 ± .79	100	.00039 ± .000084

[a] Mean (n Ci/m^2) $\cong$ (mean biomass in g/m^2) x mean $^{239-240}Pu$ concentration in n Ci/g

[b] S.E. $\cong$ [(mean g/m^2)^2 x Var (mean n Ci/g) + (mean n Ci/g)^2 x Var (mean g/m^2) - Var (mean n Ci/g) x Var (mean g/m^2)]$^{1/2}$

[c] S.E. $\cong R\left[\frac{\sigma^2_x}{x^2} + \frac{\sigma^2_y}{y^2} - \frac{2r\sigma_x\sigma_y}{xy}\right]^{1/2}$ where R = x/y; r = estimated correlation between x, y

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Studies of airborne particulates around a contaminated area at the Rocky Flats Plant yielded an average resuspension factor of 10^{-9} m^{-1} during an 8-month sampling period. This factor was near 10^{-6} m^{-1} for particulate material collected upon sticky paper exposed to suspendable fine soil particles from the soil surface. Size distribution studies of all suspended particles containing plutonium indicated a geometric mean diameter of about 10 µm (Volchok 1971, 1972). Results from cyclone and elutricator samples indicated median diameters of about 5 µm. Additional measurements by Sehmel and Orgill (1973) and Sehmel and Lloyd (1974, 1975) in the Rocky Flat area gave resuspension factors ranging from 10^{-9} to 10^{-5} m^{-1}. The Rocky Flats area is subject to winds which are occasionally very strong and gusty. Environmental surviellance data strongly indicate that the movement of contaminated soil particles by wind was a major force causing the original dispersion of plutonium from a barrel storage area (Krey and Hardy 1970; Whicker et al. 1973). The initial source of most of the outlying contamination at Rocky Flats was from leakage of cutting oil containing plutonium. Subsequent studies by Nathans and Holland (1971) of the transformation from this source to suspendable material led to the concept that the resuspendable "hot" particles are agglomerates of small plutonium-containing particles and larger soil grains. Environmental studies at Rocky Flats by the group at Colorado State University have shown some movement of plutonium into several ecosystem compartments within study plots. Data indicate some 96 to 98 percent of the plutonium in the ecosystem is associated with the 0-3 cm depth of soil. About 1 to 3 percent appears to be associated with root and litter samples, while the amounts in standing vegetation and small mammals are generally less than 1 percent. Findings indicate that most of the plutonium present in vegetation samples is superficial contamination resulting from the attachment of particles to biological surfaces. There is also some indication that grass species have higher concentration of plutonium than other species, again probably as the result of the greater surface area per unit mass in the grasses (Whicker 1973; Little 1976).

Healy (1974) completed a noble task of synthesizing into a significant treatise some of the early air sampling data obtained during the safety shots at NTS, where plutonium was dispersed by chemical explosives. Data are given and literature cited which show resuspension factors ranging from 10^{-10} to 10^{-4} m^{-1} for arid or semiarid environments. The higher values represent more unstable, outdoor conditions. Langham (1969) used a value of 10^{-6} m^{-1} in assessing limits for a weapons accident of the type simulated by the safety tests in which plutonium was dispersed at NTS. A continuing study of resuspension at NTS test areas is underway by elements of the Nevada Applied Ecology Group (Anspaugh et al. 1974, 1974a, 1974b, 1974c, 1975). Resuspension factor values calculated from recent experiments now fall within the range of 10^{-11} to 10^{-9} m^{-1} compared to values of 10^{-10} to 10^{-6} m^{-1} measured about 20 years ago at some NTS sites. Most of the total mass of suspendable material is found between diameters of 0.7 µm and 15 µm (Bretthauer et al. 1974; Shinn and Anspaugh 1975). Studies by Tamura (1974, 1975, 1976), have shown that from 50 to 75 percent of the plutonium in soil samples collected from an aged fallout area at NTS is predominantly associated with the coarse silt (20-53 μm) fraction. Ten percent or less usually is associated with the soil fraction less than 5 μm diameter. Highest activities also are associated with heavy mineral fractions (> 2.9 g/cm^3), suggesting the presence of oxides.

Fallout particles intercepted by plant foliage are principally of sizes smaller than 44 μm diameter (Romney et al. 1963). The foliage of vegetation at NTS selectively traps these small-sized particles in the matted hairs and crevices and on resinous glands of the leaf surfaces. The capacity for retaining fallout particles largely depends upon the mechanical-trapping characteristics of plant surfaces. We believe that most of the plutonium contamination now found on vegetation sampled from these aged plutonium fallout areas at NTS is superficial contamination from particulate material. This mechanism of contamination is presently the most important route through which plutonium enters the food chain of grazing animals. However, the relative importance of superficial contamination compared to the root uptake route may diminish with passing time for reasons to be discussed shortly. Fortunately the resuspension factors and vegetation biomass are not great at these sites. The undisturbed desert soil remains quite stable partly because most of the suspendable surface material was removed by wind and water erosion ages ago. Resuspension problems now arise particularly when the stabilized soil surface crust is mechanically disturbed. That is one of the reasons that recommendations have been made to avoid disturbing these aged fallout areas (Wallace and Romney 1975; Rhoads 1976). The purpose for this caution is evident from the data in Table 2, which summarizes values for the estimated inventory of plutonium for vegetation in these fallout areas. Only small amounts of the total quantity of plutonium originally deposited presently appear to move to the standing vegetation from fenced-in fallout contaminated soils which are now undisturbed, except for natural wind conditions. Advantage can be gained from keeping these sites as little disturbed as possible. This includes surface erosion by water (Hakonson et al. 1976; Hakonson and Nyhan 1976).

Table 2. Summary of Estimated Inventory of $^{239-240}Pu$ for Vegetation in Aged Fallout Areas (Romney et al. 1976)

Fallout Area	n	Inventory ± S.E.[a] (millicuries)	Vegetation invent. / Soil inventory ± S.E.[a]
NTS Area 5	113	.86 ± .14	.00034 ± .000046
NTS Area 11A	36	.098 ± .045	.0028 ± .0017
NTS Area 11B	44	2.4 ± .31	.00039 ± .000058
NTS Area 11C	48	4.3 ± .78	.00048 ± .000089
NTS Area 11D	56	7.3 ± .79	.00039 ± .000084
NTS Area 13	141	26.4 ± 3.5	.00060 ± .000076
TTR DT	48	.62 ± .19	.00012 ± .000036
TTR CS1	53	.67 ± .34	.00013 ± .000078
TTR CS2	63	4.3 ± .63	.00015 ± .000030
TTR CS3	41	4.6 ± 1.2	.00016 ± .000037

Time plutonium was dispersed by chemical explosive: NTS Area 5, 1954-1955; NTS Area 11, 1956; NTS Area 13, 1957; TTR Areas, 1963.

[a] See note c Table 1.

RECYCLING OF PLUTONIUM TO VEGETATION

Most of the standing biomass of vegetation in the aged fallout areas at NTS is contributed by deciduous shrubs (2,000 to 6,000 kg/hectare) which normally yield about 10 percent of their total weight as new annual foliage. The production of grasses, forbs and annual plant species is spasmodic from year to year, depending upon rainfall and climatic conditions. Seldom, however, does the productivity of these annual species exceed 1 percent of the standing shrub biomass. As the result, only from 200 to 600 kg/hectare of new plant foliage is potentially available to undergo the processes of litter fall, decomposition and mineralization. Most of the fallen litter is moved about by wind action to lodge underneath sheltering shrub clumps where much of the initial breakdown is carried out by consumer organisms. Very little is known about the impact of soil arthropods and micro-organisms on plutonium in these areas at NTS, but deductions from well known effects of such organisms on inorganic nutrient elements during mineralization processes would indicate that they should help increase the biological availability (solubility) of plutonium with passing time. The relationships of microbial processes to the fate of transuranic elements in soil is discussed by Wildung et al. (1977, this volume).

First glance at the data in Table 2 gives the impression of an insignificant amount of plutonium in vegetation of the fallout area. However, when one considers that this contaminated plant foliage goes through annual cycles of litter fall and decomposition and mineralization in concentrated areas underneath shrubs, and that the plutonium will persist for tens of thousands of years, one must recognize the possibility of an increasing significance this cycling process will have on increasing root uptake with passing time. The significance of this concentration process should be seen first with ^{241}Am because of its greater biological availability (Romney et al. 1976b). Studies on the difference in edaphic properties underneath shrub clumps and in adjacent bare soil areas at NTS show that highly significant concentration processes have been at work increasing the levels of plant nutrients and organic matter in the root zone underneath shrubs (Romney et al. 1973a, 1973b). Hanson (1975) recently discussed what appears to be differential biological availability and concentration between ^{238}Pu and ^{239}Pu in soils, vegetation and animal components of the Trinity Site ecosystem. He cited the data of Hakonson and Johnson (1973) showing that $^{238}Pu/^{239}Pu$ ratios increased from 0.05 (soils) to 0.10 (plants) to 1.0 (mammals), respectively.

RESUSPENSION IN CONTAMINATED AREAS

Once radioactive material has been deposited upon soil, the main concern is to control subsequent resuspension, especially in dry, dusty areas. Whether or not the contaminating radionuclide remains in an original particulate form or undergoes chemical and physical transformations which result in its being carried by soil particles may have no particular biological consequence. Either form deposited upon the surfaces of plants becomes a source of contamination in the diet of grazing animals. The mechanisms of resuspension and its consequences are discussed by other participants in this symposium. The subject of transuranic resuspension and the need for standards for controlling health effects from plutonium in soils is discussed in a recent treatise by Healy (1974).

Optimum conditions for resuspension usually are found in arid, dusty environments, but such movement of contaminated particulate material is not necessarily limited to those areas. In follow-up studies after the accident at Palomares, Spain, Iranzo (1968) called attention to the entrapment of plutonium by external foliage of agricultural crops harvested from contaminated soil. About one half of the contaminant could be removed by washing the foliage of tomato plants compared to from 73 to 95 per cent removed by washing fruit. Deposition on the surfaces of leaves and stems has been identified as the principal mechanism of plutonium contamination of vegetation collected from sampling locations around and adjacent to the Savannah River Plant which is situated in a humid area (McLendon et al. 1976). Some of this contamination could arise either from direct fallout deposited downwind of the source stack or from subsequent resuspension of contaminated soil. A study to measure resuspension during field preparation and planting of winter wheat at the SRP site was reported by Milham et al. (1976). Small, but detectable amounts of airborne plutonium averaged 210 fCi/m^3 at 7.6 m and 10 fCi/m^3 at 30.5 m distances downwind from the edge of the field under cultivation. The air at the tractor operator's face level contained 49 fCi/m^3. The average concentration of plutonium in the 0-5 cm layer of soil was 3100 fCi/g; estimated resuspension factors were of the order of 10^{-8} m^{-1}.

Work by the Los Alamos group at the Trinity site in New Mexico showed that the horizontal distribution of plutonium in soil continued to be largely determined by the original fallout deposition pattern. The Trinity soil samples contained elevated amounts of plutonium in coarser size fractions (> 105 μm) near ground zero and relatively larger amounts in finer fractions at increasing distances from ground zero (Hakonson and Nyhan 1976). Contamination of grasses within the fallout pattern, as a function of distance downwind from ground zero, generally followed the pattern observed in the 0-5 cm soil core fraction. The plutonium concentrations in grasses, lichens and mosses were consistently elevated above the levels observed in forb, shrub and tree samples. This was considered to reflect higher entrapment efficiency for grasses because of greater tissue surface areas (Hakonson and Johnson 1973; Hakonson and Nyhan 1976).

ROOT UPTAKE PATHWAY

Most soil-plant studies focusing upon plutonium movement through plant roots indicate relatively low uptake through this pathway. Concentration ratios for plants grown in potted soil ranged from 10^{-8} to 10^{-3} (Plant Panel 1975). However, in view of the long half-life of plutonium, one is plagued with nagging questions concerning the extent to which the transuranics will become more biologically available through this pathway with passing time. Some evidence already has appeared indicating an increased relative availability (or mobility) of ^{238}Pu and ^{241}Am in given situations (Essington et al. 1976; Hakonson and Johnson 1973; Romney et al. 1975).

It is difficult to differentiate between the amounts of plutonium in vegetation attributable to root uptake and superficial contamination. Attempts, therefore, have been made to accomplish this by pot uptake experiments conducted under air-filtered, glasshouse conditions. Table 3 summarizes some results obtained from soils collected from the aged fallout areas at NTS. Use is made of the Pu/Am ratio in soil and plant samples and also the concentration ratio (CR) for comparing data representing field and glasshouse conditions. The term, CR, is a simple concentration ratio calculated from the activity/g. plant divided by activity/g. soil. For the field data the soil activity is that contained in the 0-5 cm surface layer; the glasshouse soil activity is based on thoroughly blended, potted soil. We are not, therefore, comparing the same conditions in the two systems because the roots of field-grown plants should feed much deeper in the soil; certainly below the top 5 cm layer. What makes this field data most useful in Table 3 is that the 0-5 cm soil Pu/Am ratio more nearly reflects that ratio for the resuspendable material deposited on the leaf surfaces. Considerable difference exists between the plutonium contents in the different soils tested. We should also point out that the field data are overall ratios and CR values obtained from many samples analyzed from the field (n > 30) while the glasshouse values are means of six replicates. The important findings from these data are the similarities of the Pu/Am ratios for field soil and vegetation samples which we believe shows principally a superficial source of contamination on vegetation under field conditions. In addition, the calculated CR values for plutonium in the field ranged from 10^{-3} to 10^{0} compared to 10^{-6} to 10^{-3} obtained from our pot uptake tests through the root uptake pathway. The Pu/Am ratios in pot-grown vegetation are much lower than those in the potted soil, indicating uptake of ^{241}Am in much greater amounts than $^{239-240}$Pu. Other data from these tests (Romney et al. 1976b) have shown that synthetic chelating compounds and acidulation agents added to soil significantly (P .05) increase root uptake of these transuranics.

Table 3. Summary of $^{239-240}$Pu and ^{241}Am Ratios and Pu CR Values for Vegetation and Soil Under Field Conditions Compared to Root Uptake Experiment Under Glasshouse Conditions

Soil Source	Field Conditions			Glasshouse Conditions		
	Soil Pu/Am	Plant Pu/Am	Pu CR**	Soil Pu/Am	Plant Pu/Am	Pu CR*
NTS Area 11B	7.7 ± 0.14	8.5 ± 1.0	1.3E-2 to 1.6E-1	6.5 ± 0.17	0.83 ± 0.11	1.5E-4
NTS Area 11C	6.0 ± 0.08	5.2 ± 0.1	4.5E-2 to 3.4E-1	5.3 ± 0.17	0.35 ± 0.13	1.8E-4
NTS Area 11D	5.8 ± 0.15	4.1 ± 0.18	2.7E-2 to 1.7E-1	5.2 ± 0.12	0.34 ± 0.12	1.1E-3
NTS Area 13	9.4 ± 0.15	7.9 ± 0.2	7.8E-2 to 4.4E-1	5.6 ± 0.27	0.19 ± 0.03	1.1E-4
TTR DT	23.5 ± 0.73	15.8 ± 1.4	1.1E-2 to 9.4E-2	21 ± 0.67	0.97 ± 0.72	2.6E-4
TTR CS1	26.0 ± 0.76	16.2 ± 0.52	7.4E-3 to 4.2E-2	19 ± 0.33	0.57 ± 0.36	4.3E-4
TTR CS2	22.2 ± 0.41	11.6 ± 0.64	1.4E-2 to 9.2E-2	21 ± 1.9	6.3 ± 1.3	7.6E-4
TTR CS3	22.0 ± 0.28	17.0 ± 0.93	3.9E-2 to 5.3E-2	19 ± 0.67	1.9 ± 0.26	5.5E-4

* Data given are for soybean foliage. Seed CR values ranged from 10^{-6} to 10^{-4}. Data are summarized from Romney et al. (1975, 1976).

** Range of mean CR values calculated for activity strata in fallout area. CR values for individual collection sites ranged from 10^{-3} to 10^{0}.

CONCLUSIONS

Two principal incorporation mechanisms are involved in the vegetation-carrier transport of plutonium in the diet of grazing animals and mankind: (1) superficial entrapment of particulate material with possibilities of foliar absorption of soluble contaminant, and (2) root uptake of the contaminant entering soil. It is important to reduce this transport of plutonium as much as possible. Findings from studies in dusty field environments at NTS indicate that superficial contamination is presently the most important route. However, certain natural cycling and concentration processes are underway which, in terms of the long half-life of plutonium, should gradually increase the importance of the root uptake pathway. We suspect that even now the root uptake pathway has increasing importance relative to superficial contamination in humid ecosystems, except at sites subject to local fallout material from such sources as emission stacks of processing facilities. The aged plutonium fallout areas at NTS will continue to be of value as sites in which to make periodic assessments to detect changes in the cycling and concentration of plutonium in various components of the ecosystem. Because of its greater solubility, ^{241}Am at these sites may show an earlier indication of such changes. We believe it always will be important to avoid disturbances that will increase resuspension and erosion in plutonium contaminated sites at NTS and elsewhere. In their present natural states we believe that the plutonium contaminated sites at NTS and TTR present no radiological hazard to grazing animals and mankind so long as residence within the fenced exclusion areas is prohibited.

ACKNOWLEDGMENTS

Work was conducted under Contract E(04-1) GEN-12 between the United States Energy Research and Development Administration and the University of California. Supplemental support was provided by the Nevada Applied Ecology Group, Bioenvironmental Science Division, USERDA Nevada Operations Office. We are indebted to members of the NAEG management and sample collection, preparation and radiochemical analysis teams for their devoted efforts in support of this work.

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FOLIAR AND ROOT PATHWAYS OF PLUTONIUM CONTAMINATION OF VEGETATION

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ABSTRACT

Plutonium enters the terrestrial environment from testing of nuclear weapons and from nuclear technologies. Assimilation of Pu by vegetation depends on source term characteristics and mode of exposure to different Pu sources in the environment. For Pu released to the atmosphere from nuclear facilities, direct deposition of airborne particles on foliage accounts for most of the incipient contamination of vegetation. Assimilation of Pu via the roots generally yields Pu concentrations in edible tissues that are lower by at least an order of magnitude; however, lifetime exposure of humans will depend increasingly on the entry of Pu into foods by the root pathway. These concepts are confirmed from information obtained from contaminated environments at Savannah River and Oak Ridge. Radiologic doses calculated for ingestion of Pu-contaminated vegetables show that direct deposition of airborne Pu on surfaces yields doses that are a factor of ten higher than those for Pu assimilated into the plants by roots.

INTRODUCTION

Environmental plutonium (Pu) originates from two sources: fallout from atmospheric nuclear testing and emissions from nuclear fuel cycle facilities. Fallout from nuclear testing will likely decrease unless extensive weapons testing in the atmosphere is resumed, but future emissions from the nuclear fuel cycle may increase as a result of Pu recycling. The potential radiological toxicity and long half-life of Pu dictate that its behavior in the environment be thoroughly understood.

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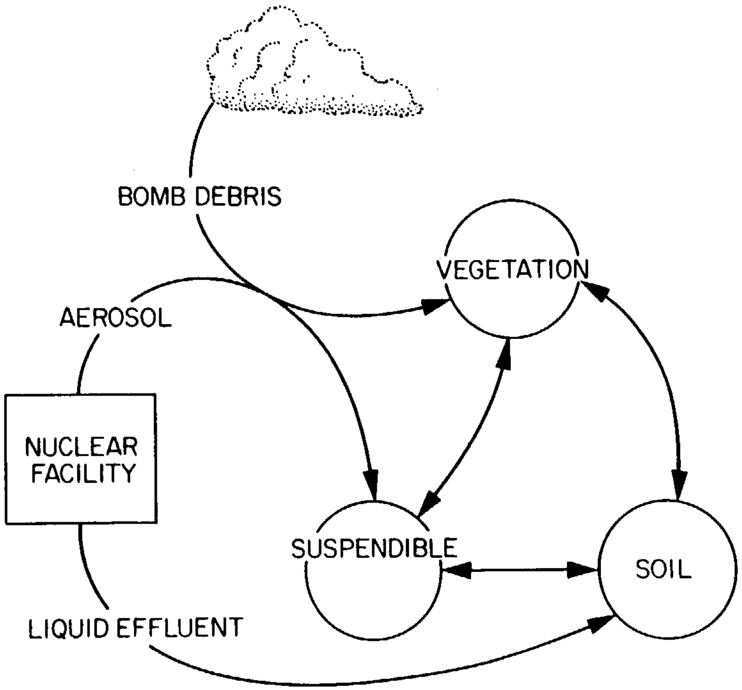
Much research on Pu behavior in terrestrial environments has been conducted in arid regions contaminated by plutonium from weapons testing. Comparatively little research has been done on the behavior of Pu in environments of the southeastern United States that are characterized by higher precipitation, acid soils, dense vegetation cover on mineral soil, a myriad of ecological niches, and complex trophic and food-chain relationships, all of which may influence the cycling of the trace element, Pu. Limited information is available on the effect of biota and organic matter on the biogeochemical behavior of Pu in ecosystems and the importance of biotransfer (i.e., root assimilation, absorption from GI tract). These biotransfers can ultimately impact on human exposure to Pu via the ingestion pathway. The purpose of this paper is to present comparative information on the contamination of vegetation either on surfaces or by assimilation into tissues, in two different environments of southeastern United States. At Savannah River, South Carolina, vegetation was exposed to both atmospheric and soil sources of Pu while at Oak Ridge, Tennessee, the source of the Pu was soil that had been contaminated 30 years ago during Manhattan Project Operations.

PRINCIPAL MODES OF INCORPORATION OF PLUTONIUM BY VEGETATION

Atmospheric emission of Pu from nuclear facilities is expected to be a future mode of Pu input to terrestrial environments; although contamination from liquid releases cannot be excluded because liquid effluents have historically deposited Pu in terrestrial environments. For Pu originating from either fallout or nuclear facilities, the main pathways for incorporation into vegetation are illustrated in Fig. 1.

The aerial pathway involves direct deposition of particleborne Pu on vegetation or deposition on the soil surface. Direct deposition leads to levels of contamination that are incipiently high but of short duration. Rapid decontamination occurs with weathering half-times of 15 to 30 days for deposits of fallout particles on foliar surfaces (Dahlman *et al.*, 1975). Similar rates of removal are projected for particleborne Pu. , Contamination of foliar surfaces also results from suspendible particles being transported from the soil surface to aerial portions of vegetation. Suspendibles may be transported to the entire plant canopy as windblown dust or to basal plant parts as mud splashed by rain.

Following deposition of atmospheric or liquid forms of Pu in the soil matrix, a fraction becomes available for assimilation by plants via the root pathway. Uptake of Pu by roots and translocation throughout the plant places the element in intimate association with food substances consumed by man over long-term intervals. As a result of Pu uptake by roots and direct assimilation into tissues, the biochemical associations may alter its chemical properties. Just as biogeochemistry affects the uptake of Pu by plants, interactions between Pu and plant biochemicals may influence the long-term metabolism of Pu by humans. Consequently, it is advisable to distinguish between Pu contamination on foliar surfaces, a physical phenomenon, and Pu assimilated via the root pathway, a process involving biophysical and biochemical mechanisms.



PATHWAYS OF PLUTONIUM MOVEMENT TO VEGETATION

FIGURE I.

Vegetation is continuously contaminated as a result of chronic low-level release of Pu to the atmosphere at Savannah River, and direct deposition of Pu on vegetation by the aerial route probably is the most important pathway (Milham *et al.*, 1976). By comparison, at Oak Ridge, Pu was deposited on a floodplain in 1944 and plant uptake from the contaminated soil by the root pathway is considered to be the major pathway. With different source characteristics for Pu in terrestrial environments, the relative importance of aerial vs. root pathways of Pu incorporation by vegetation can be evaluated.

PLUTONIUM IN SAVANNAH RIVER AND OAK RIDGE ENVIRONMENTS

Concentrations and characteristics of Pu in Savannah River and Oak Ridge environments are presented in Table 1. An important distinction between the sites is that the annual atmospheric input of Pu to the study area at Savannah River is more than 3 orders of magnitude greater than the input to the floodplain site at Oak Ridge (footnote c, Table 1). The maximum concentration of Pu in soil at Savannah River (3 mCi km^{-2}) is located within 1 km of the processing plant, while the concentration of Pu in offsite soils beyond 20 km from the processing plant was less than 2 mCi km^{-2} (McLendon *et al.*, 1976). The offsite soils are approximately equivalent to the average accumulation of Pu from global fallout in soil of middle latitudes (Hardy *et al.*, 1973). Near the processing plant, the ambient average air concentration was 1.7 fCi m^{-3} which increased by 1 to 2 orders of magnitude when dust was created by agricultural activities (Milham *et al.*, 1976). Selective analysis of suspendible particles from the soil surface (approximately the 1 mm zone) shows that this fraction contains one to two orders of magnitude higher concentration of Pu than the 0 to 15 cm increment of the soil profile (McLendon *et al.*, 1976; Milham *et al.*, 1976).

Current release of Pu from Oak Ridge facilities to the atmosphere is negligible (UCCND, 1975, 1976). Incremental contribution of Pu to the local terrestrial environment is not distinguishable quantitatively or isotopically from Pu that originates from global fallout. However, Pu released from Oak Ridge in liquid effluents has contaminated aquatic and terrestrial environments along White Oak Creek.

A floodplain contaminated in 1944 contains from 25 to 150 pCi g^{-1} of Pu mixed throughout the top 15 cm of the soil profile. The deposited Pu has been subjected to soil and ecological processes for over 30 years. Vegetation growing at the floodplain site is exposed only to soil Pu. The element is not detectable in daily air samples unless airborne dust is generated by disturbances such as soil cultivation, a practice not routinely carried out in contaminated environments.

Isotopic ratios of Pu can help differentiate between several potential sources causing environmental contamination. Thus, contributions of Pu from local sources and deposition from fallout can be distinguished, provided isotopic ratios of source terms are different. For example, the α % ^{238}Pu for airborne

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TABLE 1

PLUTONIUM IN ENVIRONMENTS OF NUCLEAR FACILITIES AT SAVANNAH RIVER AND OAK RIDGE

Environmental Matrix	Savannah River[a,b,c]		Oak Ridge[c]	
	3 mCi km^{-2} isopleth	Offsite	Plant Area	Floodplain
Deposition (pCi m^{-2} wk^{-1})	4.3 (58)[d]	0.057 (12)	6 x 10^{-5}[e]	
Soil Concentration (pCi g ^{1})				
Suspendible	1.08 (43)	0.006 (16)	NM[f]	NM
Soil (0 - 15 cm)	0.09 (14)	0.006 (8)	0.03[g]	25 to 150 (5)
Air Concentration (fCi m^{-3})				
Ambient[h]	1.7 (15)	0.035 (17)	0.016[g]	ND[i,j]
During Soil Cultivation	210 (17) 10 (35) [k]	NM	NM	4.1 (5)

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[a] The 3 mCi km^{-2} isopleth is within 1 km of the SRP source; the offsite region is beyond 20 km of the source.

[b] Data on Pu released to atmosphere and onsite airborne concentrations from Milham *et al.*, 1976; soil concentrations of Pu are from McLendon *et al.*, 1976; Offsite deposition and airborne concentrations are from Anonymous, 1975; Onsite deposition of Pu is unpublished data.

[c] Pu releases to the atmopshere in 1974 were 9 mCi yr^{-1} and 0.004 mCi yr^{-1} for Savannah River and Oak Ridge respectively.

[d] Values in parentheses are α % ^{238}Pu; α % ^{238}Pu = [^{238}Pu]/[Total Pu] x 100.

[e] Calculated for plant area and floodplain from data and model given in Environmental Monitoring Report, UCC ND 302, pp. 17 and 24.

[f] Not measured.

[g] Soil value is an average for 0-1 cm soil depth computed from 1974, 1975 data from Health Physics monitoring stations 33, 34, and 38 (Environmental Monitoring Reports UCC ND 302 and Y/UB-4) and from site survey report (Oakes and Shank, 1977); Air value based on composite of samples from ORNL air monitoring stations (Oakes and Shank, 1977).

TABLE 1
(Continued)

[h] Values determined for breathing zone using high-volume air samplers (1-2 m^3 min^{-1}).

[i] Not detectable in 346 m^3 air sample; minimum detectable level is 0.1 pCi per sample.

[j] Measurements for airborne Pu at ORNL floodplain were made at 1 m from edge of garden plot; Pu values in samples collected 10 cm from soil surface were 0.14 and 26.3 fCi m^{-3} during ambient and cultivation conditions respectively.

[k] High value determined 7.6 m from edge of field; low value 30.5 m from edge.

Pu from the processing plant at Savannah River is 58, compared with an α % ^{238}Pu of 4 for Pu from global fallout (Hardy *et al.*, 1973). The α % ^{238}Pu of Pu in environmental materials collected from the 3 mCi km^{-2} isopleth confirms that the Pu comes from the processing plant. While all Pu in Savannah River environments shows enrichment in ^{238}Pu relative to global fallout, the effect is greatest for fresh deposition and the suspendible fraction on the soil surface. In the Savannah River case, the ^{238}Pu/^{239}Pu activity ratio can serve as a useful identifier of the source of Pu and its movement in the terrestrial environment. This technique is not applicable at Oak Ridge because there are no reported differences between the α % ^{238}Pu of global fallout, local sources, and indigenous deposits.

PLUTONIUM CONTAMINATION OF TERRESTRIAL VEGETATION BY THE AERIAL PATHWAY

Vegetation in the Savannah River study area was contaminated by direct deposition on foliage by a small quantity of Pu chronically released from processing stacks to the atmosphere. For example, within the 3 mCi km^{-2} isopleth, 99% of the airborne Pu originates from stack effluents, and Pu in vegetation is attributed to direct deposition on foliage because the ^{238}Pu/total Pu activity ratio in vegetation (average of 62, Table 2) is closer to the ratio of airborne Pu (58) than to the ratio of soil Pu (14). Suspendible particles on the soil surface having a ratio of 43 may also contribute to surface contamination of vegetation. Since the leaves were not cleaned prior to processing, surface contamination was not eliminated as a possible cause of Pu in vegetation.

Results of Table 2 provide evidence that direct deposition, a physical phenomenon, is responsible for the Pu in the vegetation of the Savannah River environs. For a metabolic pathway, root uptake, to be responsible for the observed isotopic ratios in vegetation, plants would have to assimilate more ^{238}Pu than ^{239}Pu from soil. There is no evidence that plants can distinguish between Pu isotopes and selectively assimilate them nor are there even scientific bases or hypotheses of differential metabolism provided the chemical compounds of the isotopes are similar.

The concentration ratio (CR = [Pu]veg/[Pu]soil) is frequently used to express plant-incorporated Pu relative to soil concentration, and a common application of the CR is to compare results obtained from diverse experiments or field observations. Observed CRs for the Savannah River environment range from 0.075 to 1.5 (Table 2). These CR values are several orders of magnitude greater than values determined in the absence of contamination of vegetation by the aerial pathway (Schulz *et al.*, 1976; Dahlman *et al.*, 1976). Whenever soil is not the primary source of plant-incorporated Pu, there will be limitations in the conventional use of the concentration ratio. It would be advisable to consider a different expression relating to Pu in vegetation to Pu in a source when soil is not the main contributor.

TABLE 2

PLUTONIUM CONCENTRATION AND CONCENTRATION RATIOS (CR) OF VEGETATION IN THE ENVIRONS OF SAVANNAH RIVER

Vegetation	Location[a]	Pu Concentration (pCi g^{-1})	α % ^{238}Pu	CR[b]
Annual-Biennial	A	1.78	62	1.5×10^{0}
Perennial	A	0.836	63	7.2×10^{-1}
Briar	A	0.217	69	1.9×10^{-1}
Wheat Straw	A	0.088	58	7.5×10^{-2}
Plum	B	0.084	75	2.1×10^{-1}
Honeysuckle[c]	C	0.010	50	1.1×10^{-1}
Camphorweed[c]	C	0.013	48	1.4×10^{-1}

[a] Concentrations of Pu in soil were 1.16 pCi g^{-1}, 0.40 pCi g^{-1}, and 0.09 pCi g^{-1} for locations A, B, and C respectively.

[b] Concentration ratio is $[Pu]_{veg}/[Pu]_{soil}$.

[c] Data from McLendon *et al.*, 1976.

Plant habit may affect interception and retention of Pu according to data of Table 2. At site A, containing the highest soil Pu, plants exhibiting the annual-biennial habit (Solidago, Aster, and Gnaphalium) contained the most Pu followed by perennial species (briars and grasses) and wheat, which contained the least Pu. The comparison of Pu concentration of the annual-biennial and perennial habit at site A was confounded by the large quantity of dead material that was included in the perennial samples which could have diluted the concentration. The comparison is more appropriate at site C with camphorweed (annual-biennial) and honeysuckle (perennial). At this site, the concentrations were essentially identical. The low Pu concentration and CR of wheat indicate the effect of the short time of exposure to the airborne Pu. Concentration ratios for native vegetation at site A ranged from 0.2 to 1.5. With an α % ^{238}Pu value of about 60 in the native vegetation, the Pu was attributed to deposition of airborne Pu on leaf surfaces.

At sites B and C, which contained successively lower soil concentrations of Pu, CRs were about 0.1. The high α % ^{238}Pu value suggests airborne Pu as the source.

Relative contribution of airborne Pu by direct deposition vs. surface contamination by suspended particles of soilborne Pu was estimated from serial wheat samples (Table 3). Early in the season (March), when leaves are in close proximity to the soil surface, the isotopic ratio of $^{238}Pu/^{239}Pu$ was similar to that of the suspendible fraction (38 vs. 43). Later in the season (April, June), when leaves have extended 50 to 100 cm above the soil surface, the α % ^{238}Pu value of approximately 60 reflects increased deposition of airborne Pu. Plutonium concentration and CR decreased from March to April. During this period of rapid plant growth, the values decreased because of biomass accumulation occurring simultaneously with a constant Pu deposition rate. The Pu concentration and CR increased from April to June because of lower biomass accumulation while Pu deposition continued. The changes in the Pu content and α % ^{238}Pu over the three-month period indicate the importance of duration of exposure of the foliage to an airborne source.

INCORPORATION OF PLUTONIUM BY VEGETATION VIA THE ROOT PATHWAY

Root uptake is an important route for Pu incorporation into vegetation once the element becomes part of the soil matrix. This pathway is particularly important over extended time intervals in humid and heavily vegetated environments where contamination of leaf surfaces by suspended soil particles is not a major contributor of Pu to vegetation. The contaminated floodplain site at Oak Ridge is representative of such conditions, and experimental results from field studies describe root uptake of Pu by native species and vegetable crops (Table 4).

TABLE 3

PLUTONIUM CONTENT, α % ^{238}PU, AND CONCENTRATION RATIOS OF WHEAT STRAW AT VARIOUS TIMES DURING THE GROWING SEASON

Sampling Month	Pu (pCi g^{-1})	α % ^{238}Pu	CR
March	.05	38	2.3×10^{-2}
April	.02	61	9.3×10^{-3}
June	.09	58	4.2×10^{-2}

TABLE 4

PLUTONIUM CONCENTRATION AND CONCENTRATION RATIOS OF VEGETATION FROM THE ORNL FLOODPLAIN

Vegetation	Pu Concentration[a] (pCi g^{-1})	CR[b]
Native Plants		
Tree	2×10^{-3}	6×10^{-5}
Shrub	1×10^{-2}	2×10^{-4}
Herbaceous	5×10^{-2}	2×10^{-3}
Vegetable Crops[c]		
Foliage		
Bushbean	1×10^{-1}	2×10^{-3}
Soybean	1×10^{-1}	2×10^{-3}
Tomato	3×10^{-1}	5×10^{-3}
Fruit		
Snapbean	6×10^{-3}	1×10^{-4}
Shelled bean	9×10^{-3}	1×10^{-4}
Tomato	5×10^{-3}	1×10^{-4}
Soybean	4×10^{-2}	7×10^{-4}
Soybean (minus pod)	3×10^{-3}	5×10^{-5}

[a] Values are means of 3 to 24 replications; standard errors typically range from 20% to 50% of mean estimates (Dahlman *et al.*, 1976).

[b] $CR = [Pu]_{veg}/[Pu]_{soil}$.

[c] CR values of cultivated crops are based on soil concentration of 63 ± 0.4 (S.E.) pCi g^{-1}.

A small fraction of Pu is assimilated from soil by native and cultured plants. Concentrations of ^{238}Pu* ranged from 0.002 to 0.05 pCi g^{-1} in native vegetation. Concentration ratios based on soil containing 25 to 150 pCi g^{-1} of Pu ranged from 10^{-3} to 10^{-5} for herbaceous and woody species. Native species included both annuals and perennials. Plutonium was assimilated by vegetable crops planted in a field plot on the floodplain where the soil Pu contained 63 ± 0.4 (S.E.) pCi g^{-1}. Concentrations of ^{239}Pu in foliage of three different species ranged from 0.1 to 0.3 pCi g^{-1}. Concentrations of Pu in edible tissue (0.003 to 0.04 pCi g^{-1}) of the vegetable crops were lower by approximately 1 to 2 orders of magnitude. Most CR values for vegetable foliage were approximately 10^{-3}, while the CR values for fruits were on the order of 10^{-4} or less.

Both Pu concentrations and CR values of native and cultivated species exhibit high variability. Coefficients of variation (CV) are greater than 50% and frequently exceed 100% (Dahlman *et al.*, 1976). Differences in Pu among species within foliage and fruit data sets were not great. Therefore, individual observations were pooled within the respective data sets, and a "t" test showed that the Pu concentration of foliage (N=35) was significantly different ($p \leq 0.01$, df 64, F=5.15) from that of fruits (N=31).

Before Pu in vegetation can be attributed to uptake by the root pathway, it is necessary to demonstrate that results are not an artifact of surface contamination. From four different kinds of information, it appears that surface contamination is not the source of Pu in vegetation at the ORNL floodplain site. (1) Deposition of Pu from either global fallout or from local sources is insufficient to yield the concentrations observed in native species or cultivars from the floodplain (7 x 10^{-4} pCi g^{-1} from combined deposition** vs. 1 x 10^{-1} pCi g^{-1} observed. (2) Vegetation samples were washed and rinsed in a sonic bath before they were processed for analysis. Microscopic examination of washed leaves showed no soil particles or debris on the surfaces following the cleaning treatment. (3) Different ratios of U, Th, and Pu in soil and in vegetation suggest that surface contamination is not the main source of Pu observed in plant tissue (Table 5). The ratios of these elements would be the same in both soil and vegetation if surface contamination were the source of the elements. Element ratios are commonly used by geochemists in differentiating the environmental behavior of elements. When the ratios differ between soil and vegetation, as observed in this case, the evidence suggests a metabolic mechanism rather than surface contamination. (4) Ambient airborne Pu is not detectable in the canopy of the species sampled.

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*Includes ^{240}Pu.

**Accumulation of Pu in vegetation (P) from fallout and local Oak Ridge sources is calculated from $P = \frac{A \times V \times t \times r \times 10^{-3}\ \text{pCi} \times \text{fCi}^{-1}}{M} = 7 \times 10^{-4}\ \text{pCi g}^{-1}$; where A is air concentration (0.016 fCi m^{-3}, Table 1), V is deposition velocity (0.01 m sec^{-1}), t is time interval of accumulation (150 days or 13 x 10^6 sec), r is fraction of Pu deposited on vegetation that is retained at end of growing season (0.1), and M is density of foliage (300 g m^{-2}).

TABLE 5

RATIOS[a] OF INDIGENOUS AND ANTHROPOGENIC ACTINIDES IN SOIL AND VEGETATION OF THE OAK RIDGE FLOODPLAIN

	U	Th	Pu
Soil	1.0	2.1	16.3
Vegetation			
Bushbean	1.0	0.50	3.3
Soybean	1.0	0.66	3.1
Millet	1.0	0.20	0.15

[a] Ratios in terms of μg g^{-1}, μg g^{-1} and dpm for U, Th, and Pu respectively normalized to 1.0 for U (Bondietti and Sweeton, 1977).

RELATIVE RADIOLOGICAL HAZARDS IN CONTAMINATED ENVIRONMENTS

Observations from dry, windy environments suggest that surface contamination of plant leaves accounts for much of the Pu of vegetation from Pu-contaminated areas (Romney *et al.*, 1976; Hakonson and Bostick, 1976; Little, 1976). Likewise, deposition of airborne Pu released from nuclear facilities can be a major mechanism for Pu entry into vegetation (McLendon *et al.*, 1976). Frequently under such conditions, the Pu content of vegetation is considered an artifact of surface contamination and Pu uptake by the root pathway is deemed insignificant. However, both root and aerial pathways can control entry of Pu into vegetation according to the results of Tables 2 and 4. The aerial pathway controls Pu levels of plants when foliage is exposed to airborne Pu; whereas the root pathway is important over long time intervals after cessation of emissions to the atmosphere and after Pu is incorporated in the soil matrix.

Vegetation contaminated by airborne Pu exhibits an incipient concentration 1 to 3 orders of magnitude greater than observed from assimilation by the root pathway. For a typical Pu concentration of 0.3 pCi g^{-1} in vegetables, total body and bone doses were calculated assuming that an individual obtains a dietary intake of 250 g day^{-1} from the contaminated environment, and that assimilation of Pu from the GI tract is $3 \times 10^{-3}\%$ (ICRP, 1959). From chronic ingestion of the surface contaminated vegetation, the 50-year dose commitment would be 20 and 0.5 mrem for bone and total body (Table 6). When Pu is incorporated into vegetation by the root pathway, calculated doses are substantially less, simply because of the lower Pu content of ingested material.

Although radiologic impact on humans depends on source characteristics and mode of exposure, potential and comparative hazards can be evaluated for contaminated environments represented by the Savannah River and Oak Ridge examples. It should be recalled that Pu in air at the Savannah River site is approximately 2 orders of magnitude greater than at Oak Ridge, while Pu of the floodplain soil is about 2 orders of magnitude greater at the Oak Ridge site than at Savannah River (Table 1). Furthermore, dose from ingestion of foods containing surface contaminated Pu can be reduced by normal food processing procedures. For example, by threshing wheat grain from contaminated chaff, the Pu content was lowered by 98%. Accordingly, the 20 and 0.5 mrem bone and whole body doses would be reduced to 0.4 and 0.01 mrem respectively.

Although the reference environments at Savannah River and Oak Ridge presently are excluded from public access, the calculated doses would not necessarily prohibit human occupancy of both areas if ingestion is the critical pathway. The 50-yèar dose commitments are less than the 100 mrem yr^{-1} average that individuals receive from natural sources of radiation (NAS/NRC, 1972), and the doses do not exceed the 25 mrem limit proposed for a uranium fuel cycle (USEPA, 1975). Of course, population doses would also depend on exposures from other components of the fuel cycle. On the assumption that people lived in an environment where measurable airborne Pu deposited on vegetation, the resultant hazard from ingestion would be less than direct human exposure by inhalation, particularly if surface contamination of edible foods can be removed by routine

TABLE 6

CALCULATED TOTAL BODY AND BONE DOSES FROM INGESTION OF PLUTONIUM ASSOCIATED WITH PLANTS GROWING IN CONTAMINATED ENVIRONMENTS

Mode of Contamination	Pu in Soil and Vegetables (pCi g^{-1})		Dose[a] (mrem yr^{-1})	
	Soil	Vegetable	Total Body	Bone
Deposition of Airborne Pu on Plants	0.1-1.0	.3[b]	.49	19.9
Uptake of Pu by Plants	25-150	.06[c]	.11	4.2

[a] Dose calculations based on ingestion of 250 g day^{-1}, 365 day yr^{-1}; conversion factors of 7.87×10^{-1} and 1.91×10^{-2} give ^{239}Pu dose (rem μCi^{-1} ingested) for bone and total body respectively; ^{238}Pu dose conversion factors were 6.85×10^{-1} and 1.72×10^{-2} for bone and total body respectively (Killough and McKay, 1976).

[b] Isotopic composition is 60% ^{238}Pu, 40% ^{239}Pu. Concentration is an average of values given in Tables 2 and 3.

[c] Isotopic composition is 15% ^{238}Pu, 95% ^{239}Pu. Concentration is average of values in Table 4.

processing techniques. The 50-year dose commitment resulting from chronic inhalation of air containing 1.7 fCi m^{-3} of Pu (Table 1) would be approximately 80 and 2 mrem yr^{-1} to bone and whole body respectively. Calculated dose from inhalation would be a factor of 200 greater than that from ingestion of processed contaminated food. Dose from inhalation would exceed the proposed limits for a uranium fuel cycle (25 mrem, USEPA, 1975); consequently human occupancy of the contaminated area would not be permitted.

SUMMARY

As a result of chronic low-level release of Pu to the atmosphere at Savannah River, direct deposition of Pu on vegetation is the most important pathway of contaminating vegetation. By comparison, Pu uptake by roots from contaminated soil is the major pathway at an Oak Ridge site. The direct deposition pathway leads to higher Pu concentration in vegetation. Calculated doses to humans consuming contaminated vegetation are less than those estimated from natural sources and do not exceed proposed limits.

ACKNOWLEDGMENTS

The authors gratefully acknowledge the support of the Transuranic Study Group at the Savannah River Plant and the Analytical Chemistry Division at Oak Ridge National Laboratory. At Savannah River, this includes J. E. Pinder, D. Pain, and D. C. Adriano of the Savannah River Ecology Laboratory; A. L. Boni, J. C. Corey, J. H. Horton, and R. C. Milham of the Savannah River Laboratory; and H. R. McLendon of the Savannah River Plant. At Oak Ridge, analytical assistance was provided by T. Scott, N. Teasley, and J. L. Thompson. Thoughtful discussion and critical reading of the manuscript by E. A. Bondietti, C. T. Garten, and T. W. Oakes are appreciated. Manuscript typing by M. Stooksbury and Jennifer Merritt is gratefully appreciated.

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ROOT UPTAKE OF TRANSURANIC ELEMENTS

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ABSTRACT

The uptake of elements by plant roots is one of the important pathways of entry of many elements into the food chain of man. Data are cited showing plutonium concentration ratios, plant/soil, ranging from 10^{-10} to 10^{-3}. Concentration ratios for americium range from 10^{-7} to 10^{+1}. Limited experiments with curium and neptunium indicate that root uptake of curium is similar to that of americium and that plant uptake of neptunium is substantially larger than that of curium and americium. The extreme ranges of concentration ratios cited for plutonium and americium are due to a number of causes. Experimental conditions such as very intensive cropping will lead to abnormally high concentration ratios. In some experiments, addition of chelating agents markedly increased plant root uptake of transuranic elements. Particle size and composition of the source material influenced uptake of the transuranics by plants. Translocation within the plant, and soil factors such as pH and organic matter content, all effect concentration ratios.

INTRODUCTION

The purpose of this paper is to discuss the magnitude of (and some factors determining) plant root uptake of several transuranic elements. To date, a number of reviews have appeared in the literature on this subject. These reviews include those of Bernhardt and Eadie (1976); Brown (1976); Dahlman et al. (1976); Francis (1973); Price (1973). Since a number of reviews exist, and three of them are as recent as 1976, it is felt that it would be a duplication of effort to make an in-depth review of the published literature. Rather, this shall be an examination of some factors affecting plant uptake of transuranics along with a look at recent information on this subject.

EXPERIMENTAL CONDITIONS

Probably one of the more significant factors affecting reported plant root uptake of plutonium has been the highly artificial conditions under which many of the experiments have been carried out. The uptake-translocation experiments have frequently involved highly unusual conditions such as one plant per 1 to 10 grams of soil along with growth and uptake periods of several days to several weeks using seedlings. Such experiments subject the soil to intensive extraction of nutrients and all other elements, including the transuranics. In addition, such intensive short term cropping precludes growing the crops to maturity, thus excluding significant plant parts such as grain, etc. Generally, at the time such work was carried out, there were very good reasons for performing the experiments in the manner chosen. Caution must be taken in application of data obtained from these experiments to more usual soil-plant systems. It should not be implied that such data are applicable to agricultural systems. For example, a review of the literature by Lipton and Goldin (1976) led them to conclude that "plant concentration factors for plutonium are in the order of 10^{-4}." However, most of the articles referred to were reports of work where very high and unusual soil to plant ratios were employed. It will be shown later in this discussion that the concentration ratios are not in the order of 10^{-4}, but range over many orders of magnitude. It is most important that extrapolations not be made from specialized plant root uptake experiments to field conditions which govern introduction of plutonium and other transuranics into food chains via this pathway.

MAGNITUDE OF ROOT UPTAKE AND TRANSLOCATION

A Workshop on Environmental Research for Transuranic Elements was held at Seattle, Washington, in 1975. A group charged to discuss plant uptake of transuranium elements was designated the Plant Panel and will be referenced in this paper as "Plant Panel (1976)." The Panel listed data (Table 1) on plant uptake where plants were grown in containers and efforts were made to exclude foliar contamination. The concentration ratios (plutonium in plant/plutonium in soil) ranged from 10^{-8} to 10^{-3}. In addition to the plutonium concentration ratios, the Panel also summarized data on americium concentration ratios (Table 2). Here the range of concentration ratios was even larger than that of the plutonium CR's. The range of concentration ratios reported was 10^{-7} to 10^{+1}. These experiments were also carried out in containers and attempts were made to exclude foliar contamination. The most striking feature of both the plutonium and americium concentration ratios was the enormous range reported; that is, for root uptake and translocation, and excluding foliar contamination, the range was 5 orders of magnitude for plutonium and 8 orders of magnitude for the americium uptake.

Table 1. Plutonium concentration ratios for plants grown in potted soil (from Plant Panel, 1976).

Plant	Range of CR's	Conditions	Rapporteur
Ladino clover	10^{-5} to 10^{-4}	In 120 kg of NTS soil; 70 nCi g^{-1} soil; CR's increased by seven times in 5 years.	Romney
Alfalfa	10^{-5} to 10^{-4}	In 3 kg of NTS soil; 0.6 nCi g^{-1} soil; highest CR's involve chelate treatment.	
Barley			
Straw	10^{-5} to 10^{-3}		
Grain	10^{-6} to 10^{-3}		
Soybean		In 3 kg of NTS soil; 5 nCi g^{-1} soil; highest CR's involve chelate treatment.	
Forage	10^{-4} to 10^{-3}		
Bean	10^{-6} to 10^{-4}		
Barley			Schulz
Grain	10^{-7}	NTS soil; high-fired Pu oxide; 10 to 50 nCi g^{-1} soil	
Leaf	10^{-5}		
Wheat			
Grain	10^{-8} to 10^{-6}	Pu-chloride and Pu-nitrate (^{239}Pu at 0.5 μCi g^{-1})	
Leaf	10^{-6} to 10^{-3}		
Barley			
Leaf and stem	10^{-5} to 10^{-3}	Pu-nitrate (^{238}Pu and ^{239}Pu) 10 μCi g^{-1}.	Wildung
Grain	10^{-7}		

Table 2. Americium concentration ratios for plants grown in potted soils (from Plant Panel, 1976).

Plant	Range of CR's Soil	+ Lime	+ DTPA	+ Lime and DTPA	Conditions	Rapporteur
Pot culture						
Corn					10 nCi g^{-1}; chelate applied to soil.	Wallace
Soil pH = 6	10^{-2}		10^{-1}			
Soil pH = 8.5	10^{-1}		10^{0}			
Barley						
Soil pH = 7.5	10^{-1}		10^{0}			
Soil pH = 6	10^{-1}		10^{0}			
Bush bean						
Soil pH = 7	10^{-1}		10^{1}			
Bush bean					2 nCi g^{-1}; chelate added to Am stock	Adriano
Sand texture	10^{-1}	10^{-1}	10^{0}	10^{+1}		
Clay texture	10^{-1}	10^{-1}	10^{-1}	10^{+1}		
Corn						
Sand texture	10^{-1}	10^{-1}	10^{-1}	10^{0}		
Clay texture	10^{-1}	10^{-2}	10^{-1}	10^{0}		
Barley						Schulz
Leaf	10^{-4}				Contaminated soil from NTS areas 11, 13; 3 kg pot $^{-1}$.	
Grain	10^{-5}					
Wheat grain	10^{-7} to 10^{-5}					
Alfalfa	10^{-4} to 10^{-3}				Am-sol'n added to soils; 3 kg pot $^{-1}$.	
Barley					In 3 kg NTS soil; 0.6 nCi g^{-1} soil; highest CR's involve chelate treatment.	Romney
Straw	10^{-5} to 10^{-3}					
Grain	10^{-5} to 10^{-3}					
Soybean						
Forage	10^{-3} to 10^{-1}				In 3 kg NTS soil; 5 nCi g^{-1} soil; highest CR's involved chelate treatment.	
Bean	10^{-4} to 10^{-2}					

Upon inspection of the rather limited comparative data presented, it appeared to the Panel that americium is taken up by plants about 100 to 1000 times more readily than is plutonium. Also, the data indicate that the presence of complexing agents such as chelates could well influence plant root uptake and translocation of both plutonium and americium.

In addition to gross uptake and translocation of plutonium, the relative concentration in fruit versus vegetation was examined by the Panel (Table 3). From the various data presented, it appeared that the fruits tended to be much lower in plutonium than the foliage, with the range of relative concentration of plutonium in the fruit being 0.5 to 0.001 of that of the plant foliage.

Table 3. Difference factors of Pu concentration ratio for leaf-stem versus seed-fruit (from Plant Panel, 1976).

Plant	Seed-fruit CR's relative to leaf-stem	Condition	Rapporteur
Soybean	0.01 to 0.001 lower	Pot culture	Romney
Barley	0.01 lower	Pot culture	Schulz
Barley	0.02 to 0.01 lower		
Barley	0.01 to 0.001 lower	Pot culture	Wildung
Soybean			
Peas			
Barley	0.01 lower	Field lysimeter	Cline
Cheatgrass			
Bush bean	0.1 lower	Garden plot	Dahlman
Millet	0.5 lower		
Wheat	0.1 lower	Field crop	Adriano

INFLUENCE OF SOURCE CHARACTERISTICS

A group of investigators (Adams *et al.*, 1975) studied uptake of plutonium from $^{238}PuO_2$ microspheres. The microspheres were 100 μm particles, which are in the fine sand range and have relatively little surface area. The plutonium concentration ratios found in this study were in the order of 10^{-9} to 10^{-7}. These concentration ratios were based on plant ash weight and therefore would be about a factor of 10 lower if based on the more usual dry plant weight. This group of investigators also found that plutonium uptake was about 3 orders of magnitude higher where the soil was spiked with plutonium nitrate solution prior to conducting the uptake experiments. Now, if the addition of the plutonium-nitrate solutions to non-radioactive soils resulted in 0.1 μm plutonium-bearing particles in the soil, the surface areas of the particles so formed would be larger by 3 orders of magnitude. That is, if the diameter of the particle is reduced 1000 fold, from 100 μm to 0.1 μm, the surface area per gram would go up by a factor of 1000. From this consideration it is certain that the particle size of the transuranic present in soil will influence availability.

In addition to particle size, the particle composition of the source material can be expected to have an influence on availability of the transuranic in question. Upon studying barley uptake of plutonium and americium of soil from Area 13 of the Nevada Test Site, Schulz et al. (1976b) found that americium was taken up and translocated about 8 times more readily than plutonium. Now, this is much less of a difference than the 100 to 1000 fold greater availability of americium, compared to plutonium, reported by the Plant Panel (1976). The analytical data on particles recovered from Area 13 indicate that the particles are primarily plutonium-uranium oxides. It would appear likely that availability of plutonium to plants will be related to the chemical characteristics of the plutonium-uranium particle. Americium-241 will be produced by decay of ^{241}Pu present in the particle and will therefore reside throughout these particles. To a large extent, then, it could be expected that the availability of americium to plants will therefore be governed by the chemical and physical characteristics of these plutonium-uranium oxide particles. It should be noted, however, in other recent work much lower americium/plutonium CR's than those reported by the Plant Panel (1976) have been reported by Schulz et al. (1976a).

SOIL CHARACTERISTICS AND CHELATING AGENTS

A number of investigators have studied the effect of synthetic chelating agents on plant uptake of transuranics. Wallace (1974) found that DTPA greatly increased the uptake of ^{241}Am from soils under all conditions studied. Lipton and Goldin (1976) found that chelates caused increases in plant uptake of ^{239}Pu from sand cultures in the order of 10^3. Romney et al. (1970) found that plant uptake of ^{239}Pu from contaminated Nevada Test Site soil was enhanced by the chelating agent DTPA. In other experiments by Romney et al. (1976) chelating agents were generally effective in increasing plant uptake of either plutonium or americium.

On the basis of evidence cited, it is clearly established that synthetic chelates can enhance plant uptake of transuranics. This could well be important where chelating agents are used in nuclear fuel reprocessing and in agriculture, etc. However, the influence of natural chelates on transuranic uptake is not so clear. Studies on the effect of the role of natural soil humates in making metals more readily available to plants go back to at least 1932, and their "carrier" effects have been established for a long time. Soil humates are formed by decomposition of organic matter by microorganisms. What might be the effect of organic matter on plant uptake of transuranics?

Work at Battelle Northwest Laboratories (Garland et al., 1974) showed that incubation of soil with carbon and hydrogen resulted in increased plant uptake of plutonium in one soil and in the other soil, it did not.

At UCLA, a 5-year cropping experiment was carried out (Romney et al., 1970) In this experiment, the concentration or CR of the clover crops increased each year the experiment was continued. Now, was the increased uptake of plutonium due to possible organic complexation of plutonium by soil organic matter, or was it simply due to increased root concentration in the containers as the plants grew older, or neither? In another experiment at UCLA (Romney et al., 1976), organic matter--in the form of alfalfa meal--was added to contaminated NTS soil. The results from this experiment were somewhat erratic and the added organic matter had no statistically significant influence on root uptake of the radionuclides. In some cases the plant uptake of plutonium and americium appeared to be reduced by the addition of organic matter. This is certainly a possiblity as "free" or "soluble" plutonium and americium could have been temporarily tied up in a large microorganism population generated by the organic matter additions. Such an event would be expected to be transitory, however. In this same series of experiments the investigators studied the effect of added nitrogen and sulfur on uptake of plutonium and americium and again no clear-cut effect was observed.

It certainly is reasonable to expect some soil properties such as nutrient status, organic matter content, pH, cation exchange capacity, etc. to exert an influence on the plant uptake of transuranics. However, as just mentioned, the experimentation carried out at UCLA did not show any significant response to fertilizers or added organic matter.

A number of investigators have studied the effect of soil pH, and in some cases there does not seem to be an increase in uptake from soils at lower pH's. Again, the work is not conclusive.

At Battelle Northwest Laboratories some experiments have been carried out which suggest that soil organic matter may inhibit plant uptake of plutonium (Cataldo et al., 1975). It is indeed possible that organic matter in a soil could make transuranics less available. We could have a mechanism where the chelating agent could be a soil humate that is not available for root uptake. Transuranics bound by the humate would then be unavailable for plant uptake. It is also possible that lower concentration ratios could be due simply to the higher exchange capacity of soil with higher organic content.

Another factor that seems to affect the relative availability of plutonium to plants is the concentration of the element in soil (Wildung and Garland, 1974). In this experiment both the Neubauer method and a split-root technique were used to study plant uptake of plutonium. The results of this experiment showed a marked increase in the concentration ratio as the plutonium concentration in soil decreased.

Up to now, plant uptake of only two of the transuranium elements, plutonium and americium, have been discussed. Price (1972) carried out an experiment where plant uptake of two additonal transuranics was measured. In this experiment the plutonium concentration ratio was about 10^{-5} and the americium uptake was about 2 orders of magnitude larger. Curium uptake was quite similar to that of americium. The uptake of neptunium was substantially larger than that of curium and americium.

SUMMARY

Data has been cited giving plutonium concentration ratios of 10^{-10} to 10^{-3} and americium CR's of 10^{-7} to 10^{+1}. Some sense can be made of these extremely large ranges. Certainly, particle size and composition of the transuranics can have a very large effect on these ratios. This can be perhaps a factor of 10^3. Plant species and the plant part can account for another 10^3. It is likely that soil factors such as pH, organic matter, etc. can account for at least 10^2. Chelation accounts for at least 10^2 in some of the results reported. It also seems likely that experimental conditions could account for several orders of magnitude in the concentration ratios.

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RETENTION, ABSORPTION, AND TRANSLOCATION OF FOLIAR CONTAMINANTS

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ABSTRACT

The interaction of airborne pollutants with foliage of terrestrial plants has been investigated from many aspects including interception, retention and absorption. Although interception parameters for both gaseous and particulate pollutants have been effectively modeled, the behavior and fate of pollutants following foliar interception are unresolved. This is especially true for particulates. Particles having 10 to 200 μm diameters exhibit retention half-times of 10 to 24 days. However, direct and indirect data suggest that submicronic particles are more effectively retained on plant foliage than are larger particles analogous to close in fallout. Studies are presented to describe the retention behavior of submicronic size particles deposited onto foliage of bushbean and sugar beet plants. Retention efficiency was evaluated using a simulated rainfall. These studies showed submicronic particles to be increasingly less available for leaching with increasing residence time on the leaf, with > 90% of the foliar plutonium deposits being firmly held to the leaf surface. Retention mechanisms are discussed based on leaf morphology and leaching regimes employed. The absorption of foliar plutonium and its subsequent translocation to seed and root tissues was dependent on a number of parameters including chemical form and the presence or absence of a solution vector.

INTRODUCTION

Terrestrial plants represent an effective sink for atmospheric pollutants, both gaseous and particulate. Frey *et al.*, (1964) estimate that of the total land area of the earth, approximately 35% is covered by forest vegetation, 30% by brushlands, tundra and deserts, and 25% is employed for agricultural purposes. Although this provides a large and effective surface area for the interception of airborne pollutants, many questions remain unresolved, especially with respect to particle behavior and fate on interaction with plant canopies. Atmospheric transport and resuspension studies have adequately described the modes of transport of particles, their atmospheric residence time, their behavior with respect to particle size, and their sedimentation and deposition rates (Slinn, 1976). Much of the information available concerning the interception of particulates by plant canopies results from nuclear-related fallout and resuspension studies. These have generally been concerned with the extent of foliar contamination resulting from dry deposition and/or wet deposition, with subsequent calculation of retention times based on soluble or readily leachable components found in fallout particles such as ^{131}I, ^{137}Cs, and ^{90}Sr. Resuspension data from the NTS provides a basis for much of the information we have concerning the interception of plutonium contaminated soil particles and their retention.

The behavior of the transuranic elements in the environment and their potential for transfer in the food chain has been the subject of extensive study over the past 25 years. Although there is a general understanding of many problems concerning atmospheric transport, and the behavior of plutonium in specific ecosystems (NTS, Rocky Flats, etc.), little is known of the controlling mechanisms' influence on the bioavailability of plutonium and the other transuranics, and their subsequent transfers along the food web to man. With the current stratospheric depletion of fallout plutonium (Bennett, 1976), the inhalation route to man is greatly reduced. This would then suggest that the major sources of transuranic elements would be from resuspension of fallout contaminated soils on a global basis, resuspension from highly contaminated local sources, accident situations, and low-level releases from nuclear facilities.

A frequent practice in present radiological safety estimations is to discount foliar sorption and emphasize the soil to root pathway for food chain entry of transuranic and other radioelements (Vaughan *et al.*, 1976). Typical dose assessment codes assume a rapidly declining exponential loss of material from leaves (Soldat, 1971). This is certainly not a general situation. It does not apply to the behavior of plutonium aerosols to be described herein, and probably applies only to very large particles and to certain gaseous radioelements like iodine (Markee, 1971). The relatively greater importance of the foliar entry pathway compared to root absorption for worldwide fallout was, of course, recognized long ago (Chamberlain, 1970; Russell, 1965). In later studies, for particles probably of wind resuspended origin, it has been shown that 87% of the ^{90}Sr, 81% of the ^{137}Cs, and 73% of the ^{144}Ce in forage plants was derived from foliar contamination (Romney *et al.*, 1973). This was done by comparing plants grown with and without plastic enclosures at the NTS site. Currently, the relatively greater importance of foliar to root pathways to the plant has been incorporated in the LMFBR Final Environmental Statement (USAEC, 1974). Despite the inconsistencies in many other dose assessment codes, risk tends to be minimized because of extremely conservative limits specified at points where radioelements enter the human body. However, as a matter of systematic practice, an improved quantitative understanding of the basic environmental processes is required. This becomes especially important in situations: 1) where new technology may lead to different physical (size) and chemical characteristics of the source term for release, especially in nuclear fuel reprocessing plants; and 2) where comparatively large increases in the handling of radioelements are projected for the future (ERDA, 1975).

The following discussion will briefly review what is known concerning the absorption and adsorption potential of foliar surfaces, and attempt to describe the fate of transuranic particulates on interception by plant foliage, by extrapolation from the behavior of other particulates and the limited information on plutonium.

CANOPY INTERCEPTION

In the HERMES Model (Soldat, 1971) air to plant correlations of fallout Sr, Cs and I_2 have led to an average estimator, 0.25, representing the interception fraction, i.e., the fraction of each month's surface deposition from air and from sprinkler irrigation that is initially retained by agricultural vegetation, before leaching by rainfall. Such a value represents an operational definition, in practice subject to marked variation depending on specific circumstances of exposure.

In field observations, another way of considering canopy interception has been to calculate the interception coefficient, f. This coefficient (sometimes erroneously termed "interception fraction") was found to vary 1000-fold in various field situations (Bloom *et al.*, 1974).

$$f = d/dt\ (C_v) \div C_a\ V_d, \qquad cm^2/g$$

where $d/dt\ (C_v)$ = instantaneous rate of radioactivity deposition on plant leaf, pCi/g dry/sec

C_a = air concentration, pCi/cm^3

V_d = deposition velocity, cm/sec

A range this wide for estimates of initial deposition clearly establishes that aerosol polydispersity and other unassayed variables control foliar deposition and retention, as measured operationally. Probably chief among these variables is the particle size distribution, since depositon velocities can be estimated with greater accuracy for known particle sizes (USAEC, 1974). Values determined for air concentration and instantaneous deposition rates, in past field observations, have generally represented an unknown distribution of particles. Leaf type and degree of mineralization may also affect these estimates, but they are likely to be the only minor variables.

BOUNDARY-LAYER PARAMETERS AFFECTING DEPOSITION

Although particle deposition is an aerodynamic problem with a basis in the physical sciences, in real-world situations, the interaction of particles with a dynamic surface such as a plant canopy "requires a modification of predictive formulae which are mostly just empirical fits to data" (Slinn, 1975). Several of the factors which become difficult to quantitate due to their ever-changing nature in the boundary layer include turbulence, eddy effects and effects on particle momentum under these conditions (Droppo, 1976). An additional factor affecting particle deposition includes convective currents established by heat and water fluxes originating at foliar surfaces. This would again lead to a necessary modification of predictive models for the deposition process.

Under aerodynamically controlled conditions, however, measured deposition rates on plant leaves agree closely with the values predicted theoretically from measured aerosol parameters (Craig *et al.*, 1976), suggesting that either aerodynamic perturbations under field conditions or subsequent retention characteristics are the primary variables of concern. As regards retention, areas requiring attention include particle bounce and resuspension from the foliar surface as a function of absolute particle size and entrapment of particles at the surface. Although there is little information on this problem, the microtopography of the leaf surface (roughness) and its chemical characteristics with respect to wetness, stickiness and charge (Holloway, 1971) must have a profound effect on the efficiency of retention at the time of impaction. These effects may be more closely related to other physicochemical properties of the particle than to its simple solution solubility.

THE PROBLEM OF RETENTION OF PARTICULATES ON FOLIAR SURFACES

The retention of particles on foliar surfaces is dependent on many parameters associated with the foliar surface and physical aspects of the particle itself. Leaf factors which can affect the efficiency of particle entrapment include components of the leaf which affect roughness (Holloway, 1971), namely, venation, surface features of epidermal cells, nature of the cuticle surface, nature and frequency of trichomes and the microstructure of surface wax. Each of the microtopographical features of the leaf may contribute to the entrainment and retention of particulates. Other factors affecting retention include surface stickiness (presence of organic and inorganic secretion), leaf wetness and charge attraction between particulate and surface waxes and components. In addition, retention is very dependent on particle size, particle density, wind speed within the boundary layer, and solubility when considering a particular element comprising a particle.

Available information on foliar retention is sometimes disconcerting and contradictory when trying to reconcile the retention and behavior of relatively insoluble particulates with early fallout data on soluble or volatile fission products. Early fallout work, with respect to fission products, has been reviewed by Chamberlain (1970) and Russell (1965). In general, these reviews indicate a retention half-time for soluble fission products of 10-14 days, with losses resulting from reentrainment of carrier particles and sloughing of surface wax (Moorby and Squire, 1963), and loss through rainfall (Middleton, 1959). Except for radioiodine (Markee, 1971), such a short retention halftime is probably characteristic only of large aerosol particles, as illustrated below.

Several studies have approached the problem of particulate retention using a simulated quartz fallout containing adsorbed fission products.

Witherspoon and Taylor (1969) found that 88-177 μm diameter (MMD)[1] particles were more effectively entrained by pine foliage than oak over a 33-day period. Although wind resuspension accounted for a 90% reduction in number of particulates in oak leaves after only one hour, as compared with a 10% reduction in pine, the first rainfall (t_o + 1 day) accounted for a 15% reduction of particle activity remaining at one hour. A similar study by Witherspoon and Taylor (1970) presented data for the retention of 44-88 and 88-175 μm (MMD)[1] particles by various agricultural plants. These studies indicate that average wind speeds of 0.5 mph over the initial 12-hour period following contamination are more effective in removal of smaller particles, while average wind speeds of 1.1 mph over a 12-36 hour period resulted in a higher loss of larger particles. Subsequent to t_o + 6 days, varying amounts of rainfall occurred; these resulted in a marked reduction in retention for both particle size ranges. The resuspension behavior of these particles is in keeping with theoretical and empirical measurements on the inertial forces within the boundary layer required to resuspend spores (Aylor, 1975, 1976; and Aylor and Parlange, 1975). Subsequent studies of Witherspoon and Taylor (1971) with 1-44 μm (MMD)[1] simulated fallout particles reported longer weathering half-lives for 1-44 μm particles than those reported earlier with 44-88 and 88-175 μm particles. Loss rates were also less affected by time or rainfall after a residence time of seven days. This would suggest that particle size does in fact play an important role in the extent of foliar retention.

Although these studies aid in our understanding of the interception and retention of larger particles, analogous to close in fallout, questions arise as to the behavior of submicron fallout particles. Both Iranzo (1968) and Romney *et al.* (1975) have reported that Pu-containing material resuspended in contaminated field situations is difficult to remove from contaminated foliage with as much as 50% being tenaciously held on foliar surfaces. This would suggest that there are factors affecting retention other than the passive association of particles with relatively flat foliar surfaces with only inertial forces influencing their removal or resuspension as with larger particles (> 10 μm).

Wedding *et al.* (1975) have shown that deposition of 6.77 ± 0.02 μm (AMAD) uranium particles is related to surface leaf roughness. By analogy, the leaf roughness factors affecting deposition should also affect retention. The effect of wind and rainfall of foliarly deposited $PbCl_2$ particles (1-3 μm, MMD) was evaluated by Carlson *et al.* (1975). These studies showed Pb particulates to remain fixed to leaves under controlled conditions for up to four weeks following fumigation; reentrainment wind speeds up to 6.7 m sec^{-1} were ineffective in removal of surface deposits. Losses due to simulated rainfall were proportional to amount of rainfall; mists were more effective than droplets in removing foliarly deposited lead, with only 15 and 5% of the foliar deposits, respectively, being leachable.

Limited data are available on the retention of plutonium on foliar surfaces; these result from laboratory studies employing a low windspeed exposure chamber for contamination of plant canopies (Cataldo *et al.*, 1976). Figure 1 describes the leachability of two forms of ^{238}Pu dioxide

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[1] MMD assumed, particles physically measured.

as a function of residence time on foliage of bushbean, using a simulated rainfall of 0.4 cm in 7 minutes. The particles deposited onto the foliage had aerodynamic sizes (AMAD ± GSD) of 1.274 μm ± 1.63 and 0.734 μm ± 2.16 for freshly prepared and water-aged oxides, respectively. The count modes for the log normal distributions were 0.142 and 0.019 μm, respectively. These latter values represent the particle diameter (absolute size) with the highest frequency within the family of particles. The percentage of plutonium retained on foliage following mild leaching ranged from 92-99%. These data are qualitatively similar to those obtained for submicronic Pb particles (Carlson *et al.*, 1975), and contrary to data obtained with larger simulated fallout particles (Witherspoon and Taylor, 1969, 1970, 1971). Both the fresh and hydrated oxide exhibit a reduced leachability with increased residence time on the leaf. The retention mechanism may be related to physical entrapment of the submicron particles in small fissures on the leaf surface or to charge adsorption. The inability to readily remove plutonium from foliar surfaces has been noted by several authors (Hanson, 1975; Romney *et al.*, 1975; and Iranzo, 1968); however, the mechanisms controlling retention are unclear.

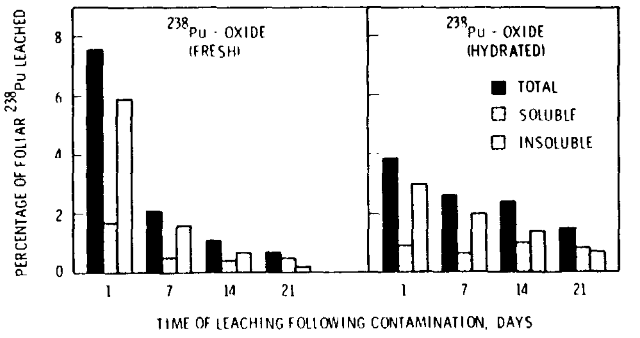

Fig. 1. Leachability of plutonium from bushbean foliage. Sets of four plants each were leached at 1, 7, 14, or 21 days following contamination. x ± SEM (n=4).

Little (1973) employed weakly acidic solutions to study the physical processes of ion exchange involved in the retention of heavy metals such as Pb on foliage. A comparison of the leachability of foliar plutonium using a synthetic rainwater with and without 0.1% HNO_3 is shown in Table 1. Leaching with acidic solution results in a moderate increase in insoluble plutonium leached from leaves contaminated with fresh PuO_2, while there is a substantial increase exhibited with leaves contamined with the hydrated oxide. The large increase seen in the soluble components may result from a solubilization of noncrystalline plutonium on the surface of the particles. The increased leachability of the hydrated oxide, as compared with the fresh oxide may be related to the larger surface area available for reaction (0.019 μm as compared to 0.142 μm diameter of the fresh oxide).

Even though much of the foliar deposited plutonium is unavailable for leaching with weakly ionic, pH 5.8 solution, the increased removal of both soluble and insoluble components with acidic solutions may indicate that a portion of submicron particulates intercepted by foliage may be held on the leaf surface by charge phenomena and by physical entrapment, and not necessarily buried in waxy plates. A comparison of the leaching behavior of plutonium for two-plant species with varying surface roughness is shown in Table 2 (Cataldo, unpublished). Plants were leached with 800 ml of solution, with leachate being collected in 50 ml fractions. Since total plutonium in the leachate decreased logarithmically, with plutonium activity in the last few leachate fractions approaching background levels, reported retention values represent Pu not readily leachable.

Scanning electron micrographs of the leaf blades show bushbean leaves to have moderate to low surface relief, while sugar beet possesses leaves which are relatively flat. Difference in surface microtopography between these two species related primarily to patterns of wax deposition and the presence of trichomes in bushbean. In bushbean, surface wax is laid down in such a way as to form high longitudinally oriented ridges with deep crevasses, with the surface of the waxy plates being relatively smooth. By comparison, the surface wax of sugar beets form relatively shallow irregular convolutions, with the surface of the wax deposits being rougher than in the case of bushbean. The trichomes of bushbean are ∿ 150 μm high and are spaced ∿ 190 μm apart, providing additional surface relief. It is this microtopgraphy, and its effect on particle entrapment and leaf surface wettability which may provide a basis for understanding the processes involved in particle retention.

In general, the leaching data for sugar beet and bushbean suggests that retention of particles on foliar surface is a function of both surface roughness and particle size. In the case of the larger fresh oxide particles (count mode ∿ 0.142 μm), substantially more of the Pu is leachable from smooth leaf surfaces under both leaching conditions. This may, in fact, be the result of physical entrapment of particles in comparatively deep fissures or crevices contributing to surface roughness in bushbean, especially if it is assumed that a particle must be suspended in a water droplet to be removed from the leaf surface. Similarly, the effect of acid leachate may be in alleviating the attractive forces holding particles

Table 1. Effect of Acid Solution on Leachability of Foliar Plutonium at Seven Days Following Exposure of Bushbean Plants to Fresh and Hydrated Plutonium Dioxide [1,2]

Compound	Leached Component	Leaching Solutions Synthetic Rainwater	 0.1% HNO_3 in Synthetic Rainwater
		% leached[3]	
^{238}Pu-Dioxide	Soluble	0.5 ± 0.1	1.0 ± 0.1
	Insoluble	1.6 ± 1.0	2.5 ± 0.6
	Total	2.1 ± 1.1	3.5 ± 0.7
^{238}Pu-Dioxide (Hydrated)	Soluble	0.6 ± 0.1	4.5 ± 0.2
	Insoluble	2.0 ± 1.3	5.3 ± 0.5
	Total	2.5 ± 1.4	9.8 ± 0.2

1. Plants leached with 200 ml of solution (equivalent to a 7 min rainfall of 0.4cm)
2. Plant foliage exposed to polydispersed aerosols. Freshly prepared oxide had an AMAD of 1.27 µm and GSD of 1.63, while the hydrated oxide had an AMAD of 0.73 µm and GSD of 2.16. Count modes for the aerosols were 0.140 and 0.018 µm for the fresh and hydrated oxides, respectively.
3. Leachability expressed as µCi leachate/ (µCi leached + µCi remaining on leaves) x 100; 4 replicates $\bar{x}$ ± SEM

Table 2. Effect of Continuous Leaching Regimes on Removal of Plutonium Particles From Leaves of Bushbean and Sugar Beet[1,2]

Plant Species	Leaf Roughness	Plutonium Form	Plutonium Retained on Leaves After Leaching[3] Synthetic Rainwater	 Synthetic Rainwater + 0.1% HNO_3
			%	
Phaesolus Vulgaris (Bushbean)	Moderate	Fresh $^{238}PuO_2$	97.6 ± 0.9	97.0 ± 0.4
		Hydrated $^{238}PuO_2$	95.5 ± 1.2	71.6 ± 6.7
Beta Vulgaris (Sugar Beet)	Smooth	Fresh $^{238}PuO_2$	82.0 ± 4.9	64.7 ± 9.8
		Hydrated $^{238}PuO_2$	95.7 ± 1.1	83.0 ± 3.5

1. Plants leached with 800 ml of solution (equivalent to a 28 min rainfall of 1.7 cm), 7 days following exposure.
2. Plant foliage exposed to polydispersed aerosols. Particle size data for bushbean reported in Table 1. For sugar beet, fresh oxide had an AMAD of 1.59 µm and GSD of 1.76, while the hydrated oxide had an AMAD of 0.75 µm and GSD of 1.84. Count mode for the aerosols were 0.130 and 0.048 µm for the fresh and hydrated oxides, respectively.
3. 4 replicate samples, $\bar{x}$ ± SEM

to leaf surfaces, particularly in the case of sugar beet. The retention behavior of the smaller hydrated oxide particles (count mode ∿ 0.019 μm) is slightly different than for the fresh oxide. The synthetic rain water was about as effective in removal of particles from both bushbean and sugar beet, while the acid leach was slightly more effective with bushbean.

Although the gross surface structure of bushbean and sugar beet leaves is obviously different, the microtopography of the surface itself may not be as different with respect to retention of very small particles (0.02 μm). This, in fact, may explain similarities in the retention behavior of plutonium deposited onto foliage of sugar beet and bushbean. Obviously, with limited data, it is impossible to generalize as to mechanisms controlling the fate of particles associated with foliar surfaces. However, in the case of the small hydrated oxide particles, leachability and retention is not only dependent on particle size with respect to leaf topography and physical attraction such as charge, but also the ability of water droplet to contact the particle, thus wettability and contact angle become important (Gregory, 1971), along with a host of environmental factors (Hull *et al.*, 1975).

Aside from our lack of understanding of mechanism, it is important to note that the behavior of small particles, such as plutonium on leaf surfaces with respect to retention half-time, differ markedly from that commonly reported for fission products and larger particles (> 10 μm). The tenacity of plutonium attachment observed by Romney *et al.*, (1975) and Iranzo (1968) tend to reinforce the above laboratory studies with plutonium.

AVAILABILITY OF FOLIAR DEPOSITS FOR UPTAKE AND TRANSPORT TO OTHER PLANT TISSUES

Foliar structures are a source of many organic and inorganic substances which either migrate to the surface by diffusion and mass flow or are actively exuded by secretory structures. This provides a chemical environment on the leaf surface enabling readily hydrolyzable species to be complexed or chemically stabilized and therefore more available for foliar absorption. Since foliar surfaces also represent an efficient absorptive structure (Wittwer *et al.*, 1963; Franke, 1967 and 1971), foliar applicaton of micro-nutrients to correct nutrient deficiencies is found to be effective, especially in those situations where a specific nutrient tends to be immobile and not as available for plant uptake on soil amendment (Krantz *et al.*, 1962; Franke, 1967). The actual mechanisms involved in foliar absorption are not totally understood. Available information would indicate that although the cuticle of the leaf is hydrophobic in nature, penetration is facilitated via intermolecular spaces (Fisher and Boyer, 1972), modification in cutin composition at anticlinal epidermal walls and the presence of ectodesmata (Franke, 1967, 1971), and trichomes (Benzing and Burt, 1970). The role of stomates as a route of foliar penetration under normal conditions is in question and is currently held to be of negligible importance (Greene and Bukovac, 1974).

The relative importance of foliar absorption as compared to root absorption as a route of entry into plant tissues depends on several factors. For soluble species which remain relatively available in soil solution, root absorption processes are as, or more effective than, foliar absorption processes. This does not imply that foliar surfaces are not effective sites of absorption. Elements reported to be absorbed and transported from foliar surfaces include inorganic and organic N, Rb, K, Na, Cs, P, Cl, S, Zn, Cu, B, Mn, Fe, and Mo (Wittwer *et al.*, 1963). In the case of specific nutrient deficiencies, foliar applicaton is the method of choice (Bradford, 1966; Labanauskas, 1966). This is especially true for those nutrilites which tend to readily hydrolyze in soil solution or are rapidly adsorbed to soil particles, thereby limiting availability for root adsorption.

The fate, with respect to foliar absorption, of relatively insoluble elements such as plutonium making up or carried on discrete particles is in some way analogous to the behavior of micronutrients such as Fe, which tend to form relatively insoluble products in aqueous environments. If we can assume that small particles containing plutonium (<1.0 μm) can be retained in foliar surfaces over an extended period of time, the question arises as to the absorptive capacity of foliar surfaces for available plutonium. Since absorption of a particular element is a function of the concentration of the available or "soluble" component, an extended residence time on plant foliage may provide time necessary for "soluble" components to be chemically modified and/or absorbed by internal tissues. This may represent a more efficient route of entry, as compared to the same finite amount of plutonium deposited into soil where insolubilization and adsorption to soil particles are more likely to occur, thus reducing ever further the concentration available for root absorption.

Absorption data from laboratory studies with bushbean plants contaminated with aerosolized plutonium are given in Tables 3 and 4 (Cataldo, unpublished); the protocol for this study has been previously reported by Cataldo *et al.* (1976). The objective of this study was to evaluate the extent of absorption and translocation of foliarly applied plutonium based on chemical form supplied, and the presence or absence of a solution vector (simulated rainfall). All plants were exposed to plutonium at 20 days from planting (preflowering) and held for an additional four weeks to allow time for both absorption of plutonium and seed filling. Seed and root tissues were analyzed to determine the extent of translocation of plutonium; these were protected from aerosol contamination. The pots containing soil and root were double-bagged with polyethylene and sealed at the lower stem; the seed tissue was contained in pods formed after exposure. Transport ratios (TR values) for root and seed tissues from plants not subjected to leaching (solution vector) ranged from $<4.5\times10^{-6}$ to 3.3×10^{-5}. Applicaton of a simulated rainfall to provide a solution vector for diffusion and absorption of soluble components on the leaf surface resulted in an increased uptake and transport of Pu to seed and root tissues for all compounds of Pu studied. Apparent differences between the various Pu forms may result from the relative size of the "soluble" fraction. The fresh plutonium dioxide was truly particulate at the time of contamination, the aged oxide

Table 3. Extent of Translocation of ^{238}Pu from Contaminated Foliage of Bushbean to Seed Tissues in the Absence and Presence of a Solution Vector[1]

Time of Leaching (Days Following Contamination)	Stage Of Development	Plutonium Chemical Form[2]			
		Pu Oxide (Fresh)	Pu Oxide (Hydrated)[3]	Pu Citrate	Pu Nitrate
		TR[4]			
Control--No Leach		$<4.5\times10^{-6}$	1.1×10^{-5}	5.4×10^{-6}	6.8×10^{-6}
Day 1	Preflowering	2.6×10^{-5}	4.1×10^{-5}	8.4×10^{-5}	2.6×10^{-4}
Day 7	Flowering, Seed Development	1.7×10^{-5}	1.8×10^{-5}	1.8×10^{-4}	1.4×10^{-3}
Day 14	Seed Filling	1.8×10^{-5}	4.4×10^{-5}	3.5×10^{-5}	1.1×10^{-5}
Day 21	Seed Filling Completed	$<5.0\times10^{-6}$	4.3×10^{-5}	4.7×10^{-5}	4.0×10^{-6}

1. Plants leached with 200 ml of solution, equivalent to a 7 min rainfall of 0.4 cm.

2. All compounds supplied from solutions at pH5.8 to 7.0; aerosol characteristics for the fresh and hydrated oxides given in Table 1; aerosols of Pu-citrate had an AMAD of 1.61 μm and a GSD of 1.86, count mode was 0.200 μm; Pu-nitrate had an AMAD of 2.29 μm and a GSD of 1.91, count mode was 0.152 μm.

3. Aged in H_2O at pH 7.0 for 10 months

4. Transport Ratio = pCi/gm Seed Tissue/pCi/gm Contaminated Leaf Tissue; average of four replicate samples

Table 4. Extent of Translocation of ^{233}Pu From Contaminated Foliage of Bushbean to Root Tissues in the Absence and Presence of a Solution Vector[1]

Time of Leaching (Days Following Contamination)	Stage Of Development	Plutonium Chemical Form[2]			
		Pu Oxide (Fresh)	Pu Oxide (Hydrated)[3]	Pu Citrate	Pu Nitrate
		Tr[4]			
Control--No Leach		$<6.6\times10^{-6}$	$<4.8\times10^{-5}$	7.3×10^{-6}	3.3×10^{-5}
Day 1	Preflowering	2.2×10^{-5}	1.6×10^{-5}	4.7×10^{-5}	1.1×10^{-4}
Day 7	Flowering, Seed Development	1.4×10^{-5}	2.2×10^{-5}	5.7×10^{-5}	4.6×10^{-4}
Day 14	Seed Filling	1.1×10^{-5}	4.3×10^{-5}	7.1×10^{-5}	1.4×10^{-5}
Day 21	Seed Filling Completed	1.6×10^{-5}	1.9×10^{-5}	1.7×10^{-4}	3.3×10^{-5}

1. Plants leached with 200 ml of solution, equivalent to a 7 min rainfall of 0.4 cm.

2. All compounds supplied from solutions at pH 5.8 to 7.0; aerosol characteristics given in Table 3.

3. Aged in H_2O at pH 7.0 for 10 months

4. Transport Ratio = pCi/gm Root Tissue/pCi/gm Contaminated Leaf Tissue; average of four replicate samples

consisted of particles with a fractured crystal lattice, the citrate represents a relatively stable soluble complex, and the nitrate an unstable complex which rapidly hydrolizes on dilution to form colloidal hydroxides. The order of bioavailability with respect to transport to seed and root was plutonium nitrate (hydroxide) > Pu citrate > aged oxide > fresh oxide.

An interesting aspect of these data is that maximum TR values are obtained when the simulated rainfall occurs at day 7 or 14, the time of rapid seed development. This is of interest from the standpoint of plutonium mobility within the plant and its chemical form. It is generally accepted that the movement of materials out of mature leaves requires that they be transported in the phloem. Entry of molecules into this transport conduit is metabolically regulated with the loading process being highly specific for individual organic metabolites and inorganic elements (Crafts and Crisp, 1971). There is growing evidence that many inorganic nutrilites, especially multivalent cations, are transported as organic complexes in both the xylem (Tiffin, 1971; 1967; Bradfield, 1976) and phloem (van Goor and Wiersma, 1976). By analogy to the behavior of nutrilites, plutonium must be transported out of the contaminated leaves via the phloem. Similarly, it is unlikely that inorganic plutonium could remain soluble at the pH of phloem cell sap (Ph 7.2-8.5, Ziegler, 1975). Therefore, the possibility exists that the mobile plutonium which was deposited in seed and root tissues may be complexed with phloem mobile organic species. This would serve to explain the apparent increase in TR values seen during the time of seed development. During this period, there is a significant change in both the composition and quantity of specific metabolites being produced by leaves and being exported to metabolic sinks such as seeds and roots. This may increase the potential for soluble species of plutonium to become complexed with organic metabolites and be subsequently exported to metabolic sinks. Although this is a tentative judgment and subject to substantive studies, this interpretation serves to explain the observed results based on known metabolic aspects of plant function.

CONCLUSIONS

The ability of terrestrial plants to accumulate potentially hazardous elements from soils via root absorption and their relative importance in the food web to man, has prompted numerous studies over the past 25 years. The vast majority of these investigations have been concerned with soil-plant transfer routes, since the soil represents a major repository for pollutants released to the environment and because the plant root is an efficient solute absorbing structure. Until recently, the foliar portions of plants were considered to play a minor, transient role at best, with respect to dose assessment problems.

Our current understanding of the aerodynamic behavior of particles and anticipated reductions in particle size distributions of materials like plutonium through an expanded nuclear energy program, suggests that a reevaluation of the role of plant foliage in particle interception and absorption of materials contained on airborne particulates is in order.

This need is supported both by early investigations and studies currently underway. Early studies of worldwide fallout, and current work on wind resuspended contaminated soils, indicate that foliar retention and foliar absorption may be as important and in some cases exceed the performance of roots with respect to food chain transport problems. As regards leaching of foliar deposits, a critical evaluation of past literature suggests that aerosol polydispersity and large particle size (e.g., 45 μm, MMD) may explain the comparatively large degree of leaching or "weathering" reported for the latter experiments. This view is reinforced by data reported for well characterized particles of Pb and Pu in laboratory studies and field observations for fallout Pu. These latter investigations indicate a sizeable fraction (>80%) of submicronic particles deposited onto foliage to be tenaciously held on leaf surfaces under varied conditions (e.g., simulated rainfall, wind). Aside from the potential health implications associated with increased foliar retention, the problem of foliar absorption must be considered. In the reported studies, a substantial fraction of the foliar plutonium deposits were transported to seed and roots. Transport ratios were affected by both the presence of a solution vector (simulated rainfall) and the timing of its application with respect to stage of plant development.

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CHRONIC RADIATION INDUCED CHROMOSOMAL ABERRATIONS IN NATIVE SHRUBS AT NEVADA TEST SITE

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ABSTRACT

Radiation effects at the morphological and phenological level within vegetation at NTS have been looked for almost from the start of nuclear testing in Nevada, and have, within the last decade been described both for areas contaminated with nuclear debris and for the vicinities of experimental radiation sources. Radiation effects at the chromosomal level of plant organization in radiation contaminated natural environments apparently have neither been sought nor observed. *Artemisia spinescens* shrubs from Site D, Area 11, Plutonium Valley NTS which have been irradiated since 1956 with uncertain doses estimated to range from 35 R to 140 R for a ten year period proved to have chromosomes quite suitable for microscopic examination. Meiotic pollen mother cells from *A. spinescens* showed, among seven classes of chromosome aberrants, 14.2% aberrants chromosomes compared to 3.8% from plants about one mile south outside the Plutonium Valley enclosure where background radiation levels occurred. These values were not statistically significant at conventionally accepted levels of significance; however when the highest percentage occurrence in each population was omitted, the difference was significant at the 95% level. For comparison two other shrub species from UCLA's Rock Valley experimentally irradiated plot, also chronically irradiated at 2.5 to 4.8 R/day since 1964 to doses very much higher than those at Site D were also investigated. Both species showed chromosome aberrations and in one species, *Krameria parvifolia*, the difference in occurrence of chromosome aberrants was different statistically for the higher radiation doses. The same kinds of chromosome aberrants were noted for all species studies and these were the kinds of aberrants noted as resulting from experimental radiation doses in the laboratory.

INTRODUCTION

Since the morphological effects of ionizing radiation such as reduced vegetative growth, flowering, seed set and germination, death of shoot apices, and anomalous flowers and leaves are caused by alterations in genetic material, it seems worthwhile to study radiation effects at the chromosomal level in the field despite the variables in natural habitats. There have been many investigations of radiation-induced chromosomal aberrations in plants in the laboratory (Sax, 1941, 1950; Giles and Conger, 1950; Sparrow, 1951), but there appear to be few references concerning low-level radiation effects over long time periods in the field. The work on chronic low-level effects has been primarily concerned with morphological damage encountered in areas of natural radiation (Osburn, 1961; Mericle and Mericle, 1965). Bailey and Luquire (1967), however, studied chromosome morphology in *Tradescantia* populations from naturally-irradiated areas and found no significant differences attributable to radiation.

We have not been able to find any reports of cytological effects from long-term chronic radiation from nuclear debris in the field. In addition to our investigation of this area where morphological changes have not been observed, we also report on two species from the Rock Valley experimentally-irradiated area of Nevada Test Site (NTS) where morphological effects have been noted on one of the species reported on here, i.e., *Krameria parvifolia* (Vollmer and Bamberg, 1975) to be discussed subsequently. The search for morphological and phenological effects resulting from radiation within areas contaminated with radioactive materials at NTS have been carried out almost from the beginning of testing of nuclear devices there. To date, evidence of radiation damage to vegetation at NTS has been found in the vicinities of nuclear cratering experiments, or in the fallout patterns of accidental venting of radioactive debris. Some effects on vegetation have also been noted adjacent to large gamma radiation sources set up in controlled experiments within NTS (Kaaz, et al., 1971; Rhoads, et al., 1969; Vollmer and Bamberg, 1975).

Radioactively-contaminated areas where morphological or phenological effects have not been observed are also of interest from another point of view, particularly where, as in this case, there have been low-level doses for one to two decades. In some of these areas, the accumulated doses approach levels associated elsewhere with morphological effects. Site D, Area 11, NTS, one of our study sites is such an area. This site is contaminated by plutonium spread primarily by a non-nuclear high explosive, although there was possibly a small component of nuclear fission debris accompanying the initial distribution of plutonium.

Plant species with relatively small numbers of large chromosomes are most suitable for cytological investigations. Of the nine shrub and seven annual species collected and examined, only three shrubs proved to be satisfactory for this kind of chromosome study; these included two Asteraceae: *Machaeranthera tortifolia* (Gray) Cronq. and Keck., (n=6), and *Artemisia spinescens* D.C. Eat. (n=9). The third belongs to the Krameriaceae, *Krameria parvifolia*, Benth., (n=6).

Visual cytological methods require that cells to be examined must be collected when the chromosomes are condensed and individually distinct. The meiotic microsporocyte division fulfills this requirement and provides maximum aberration information. Only in this division can cells heterozygous for inversions and translocations be detected with conventional staining techniques, in addition to the dicentrics, fragments, and ring chromosomes visible in mitotic cells.

Although there was the initial intent to study annual plant species, for the last three years there have not been sufficient annuals, due to unfavorable growing conditions, to fulfill this objective.

In Area 11, NTS, which was to have been the site of this annual species investigation, there was one shrub species with flowers which occurred with a distribution adequate to allow collection of flower buds in areas for which there are radiation background measurements providing long-term dose estimates (Rhoads and Franks, 1975) and in areas which have "normal" background levels of radiation. This was *Artemisia spinescens*. The two other shrub species were collected in Rock Valley, NTS, where phenological changes have been noted in certain species subsequent to long-term chronic exposure to gamma radiation (Kaaz, Wallace, and Romney, 1971; Vollmer and Bamberg, 1975).

The Rock Valley irradiated study site has been described in detail elsewhere (French, et al., 1974). In brief, the area has been irradiated since 1964 by a differentially shielded ^{137}Cs source at the center of a 340 m diameter circular plot. Phenological studies of two shrub species within the area have been reported: *Ephedra nevadensis* had fruit and stem production reduced after five years (Kaaz, et al., 1971). Reduction in fruit and leaf production, among other effects were noted in *Krameria parvifolia* (Vollmer and Bamberg, 1975) one of the subjects of this study. Although the Rock Valley facility cannot be considered as a low-level radiation area, as far as plants are concerned, it is a long-term chronic irradiation facility which serves in part as a basis for understanding the results of the area of our primary concern, Area 11.

MATERIALS AND METHODS

Flower buds were collected in April 1975 and from 31 March to 19 May 1976. The buds were kept in a fixative of 95% ethanol, chloroform and acetic acid (6:3:1) for 24 hours, then transferred to 70% ethanol for refrigerated storage until microscope slide preparation began.

Twelve shrubs of *Artemisia spinescens* of a total population of about 25 shrubs at Site D, Area 11, and six of 22 shrubs sampled from about one mile south, just outside the enclosure gate provided anthers with pollen mother cells at the proper stages for chromosome analysis. Several other species were taken in the Rock Valley experimental area. Only two, *Machaeranthera tortifolia* and *Krameria parvifolia*, provided cells in the proper stages for analysis. Fourteen shrubs of *M. tortifolia* and 11 of *K. parvifolia*, from two radiation levels, a higher and a lower were useable. The radiation exposure rates are indicated on tables in Results. We were not successful in collecting buds of these species in areas with normal radiation backgrounds.

Anthers were squashed directly on slides and cells were examined using Beeks' (1955) acetocarmine squash technique. Vials with buds collected in the field were coded when collected and this code number was carried over to the microscope slide. These were not decoded until after scoring, i.e., scoring of the chromosome conditions was done blindly. Scoring was done in the following manner. The slides were systematically scanned under 200X magnification with a Zeiss phase contrast microscope. Detailed analysis of selected cells was done at 100X under an oil immersion lens. Scoring consisted of counting aberrant cells in several categories relative to those cells appearing to be normal by using a hand-tally instrument to keep running counts.

In both *A. spinescens* and *M. tortifolia* late prophase I to metaphase I cells were scored separately from anaphase I to telophase I because certain aberrations are only detectable in one or the other of these categories. *K. parvifolia* was handled differently because only two of the ten plants examined were suitable for scoring in the late prophase I to metaphase I category. Most of the chromosomes in these stages were indistinguishable because they were clumped together. However, many cells in later stages were present and anaphase I to telophase I was scored separately from anaphase II to telophase II in the *K. parvifolia* plants examined. About 150 cells were scored for each category per plant when suitable material was available.

Photographs were made of chromosomes using a 4 by 5 inch Polaroid-camera attachment providing both negative and positive images. Photographs reproduced herewith are from enlargements made from the Polaroid negatives.

The radiation doses to vegetation at Site D, Area 11 were discussed in some detail earlier (Rhoads and Franks, 1975). Dose measurements from thermoluminescent dosimeters (TLDs) in the field for 69 days extrapolated to a ten-year period provided estimated doses from 35 R to 140 R/10 years, at a height of 25 cm above the soil surface. These estimates at or near sampled shrubs are shown in Table 1. Since most of the shrubs, if not all, have been there since the initial contamination of the area in 1956, the doses will be uncertain for a shrub or a portion of a particular shrub. These uncertainties will be discussed in some detail in a later section. In addition there is an uncertainty about a possible dose contribution from fission products which may or may not have accompanied the initial distribution of plutonium. Neither was the dose from americium production from decay of plutonium calculated in the dose. Because of these factors our dose estimates are probably low. The area can be characterized, however, as an area with a chronic dose from low-energy gamma emitters over a relatively long period of time. The beta component is thought to be small, but until there is a characterization of the radionuclides present this component of the radiation field cannot be evaluated.

There are still other uncertainties about the radiation history of this area which prevent assigning dose estimates to vegetation of the area with confidence. Although the area is at present accurately described as a chronic, low-level radiation area, there may have been, during the period of above ground tests at NTS fallout patterns which covered the area, since it lies to the east of Yucca Flat. This fact also casts some doubt on the radiation doses to those shrubs used as controls in this experiment which now occur in an area with what is considered a normal radiation background. In any event these latter shrubs are not subject to long-term chronic radiation.

Table 1. Pollen mother cell chromosomal aberrations in *Artemisia spinescens*

SHRUBS FROM SITE D, AREA 11, NTS (IRRADIATED)

Sample Number	Prophase I – Metaphase I								Anaphase I – Telophase I				
	8II,2I[1]	7II,4I[2]	9II,0,I[3]	Multivalents	Fragments	Total Aberrations	"Normal"	Total	Dicentrics	Total Aberrations	"Normal"	Total	Dose R/10 Year[4]
27	1	0	0	0	0	1	27	28	0	0	12	12	90
31	5	0	0	0	1	6	144	150	2	2	213	215	42
33	1	0	0	1	0	2	9	11	0	0	0	0	85
39	4	0	0	0	0	4	148	152	4	4	13	17	85
46	1	0	0	147	0	148	19	167	0	0	0	0	85
47	2	0	0	0	0	2	148	150	0	0	0	0	132
50	10	1	0	0	1	12	138	150	26	26	51	77	90
119	7	0	3	0	2	12	139	151	1	1	7	8	85
121	0	0	0	0	1	1	33	34	0	0	0	0	
135	7	0	0	0	0	7	143	150	0	0	10	10	
159	2	1	0	0	0	3	58	61	0	0	0	0	106
167	9	0	3	1	2	15	133	148	8	8	92	100	69
Totals	49	2	6	149	7	213	1139	1352	41	41	398	439	
Percents	3.6	0.1	0.4	11.0	0.5		84.2	99.9	9.3		90.7	100.0	

Percent Aberrants, Prophase I – Metaphase I (15.7 Total Aberrations)

Percent Aberrants, Anaphase I – Telophase I (9.3 Total Aberrations)

Percent Aberrants, Prophase I – Metaphase Plus Anaphase I – Telophase I = 254/1791 = 14.2%[5]

SHRUBS FROM OUTSIDE IRRADIATED AREA

Sample Number	8II,2I[1]	7II,4I[2]	9II,0,I[3]	Multivalents	Fragments	Total Aberrations	"Normal"	Total	Dicentrics	Total Aberrations	"Normal"	Total	Dose R/10 Year[4]
19	3	0	0	0	2	5	145	150	2	2	1	3	
44	2	0	0	4	1	7	57	64	2	2	12	14	
61	10	0	0	0	4	14	136	150	3	3	197	200	
77	0	0	0	0	0	0	31	31	1	1	50	51	
110	0	0	1	0	0	1	149	150	0	0	0	62	
160	1	0	0	0	0	1	64	65	0	0	0	---	
Totals	16	0	1	4	7	28	582	610	8	8	323	331	
Percents	2.6	0	0.2	0.7	1.1		95.4	100.0	2.4		97.6	100.0	

Percent Aberrants, Prophase I – Metaphase I (4.6 Total Aberrations)

Percent Aberrants, Anaphase I – Telophase I (2.4 Total Aberrations)

Percent Aberrants, Prophase – Metaphase Plus Anaphase I – Telophase I = 36/941 = 3.8%[5]

[1] Eight bivalents and two univalents.
[2] Seven bivalents and one quadrivalent.
[3] Nine bivalents and one fragment or ring.
[4] Samples 121 and 135 were collected in the vicinity of the other samples but were not close to a point of known dose.
[5] Values are not statistically different at conventional levels based on Mann Whitney test.

RESULTS

Flower buds from *Artemisia spinescens* with meiotic cells were collected on 10 April 1975 and 1 and 15 April 1976. The nine pairs of near equal-sized metacentric chromosomes of *A. spinescens* are large and provide an excellent subject for cytological analysis (Fig. 1). During interphase, a variable number of different-sized extranuclear bodies are present (Fig. 2). Their appearance and disappearance seems to coincide with that of the nucleoli.

Fig. 1. The normal condition of nine large metacentric bivalents in *Artemisia spinescens*.

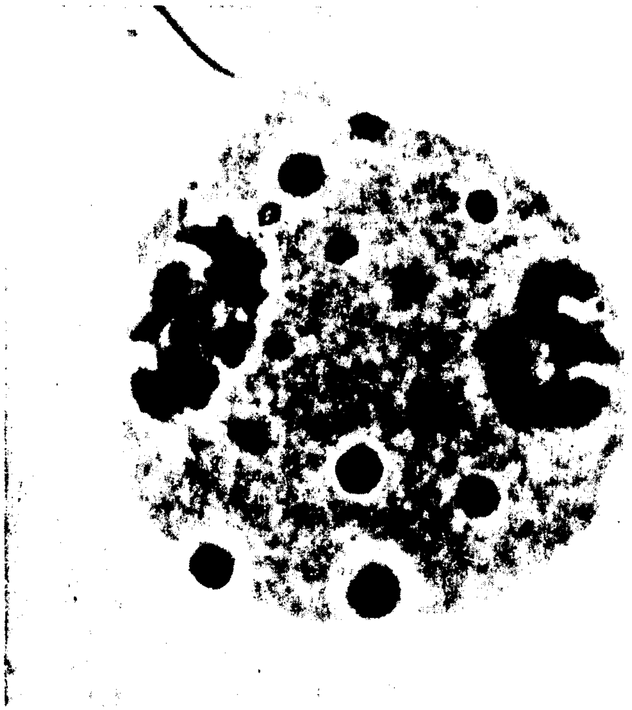

Fig. 2. Small spheres are the extranucleolar bodies of *Artemisia spinescens*.

Cells from 12 shrubs of *A. spinescens* were collected in the contaminated area, Site D, Plutonium Valley, Area 11, NTS, and from six shrubs about 1 km south where radiation levels were background, i.e., the control area. Of the 2,732 cells analyzed, 1,791 came from plants collected in the contaminated area and 941 came from plants in the control area. In the contaminated area 14.2% of the cells had abnormal configurations, and in the control area 3.8% were abnormal (Table 1). In Table 1 and in succeeding tables we have called "Normal" all cells which did not possess obviously aberrant configurations. Several categories of aberrants were noted. These are: eight bivalents and two univalents, multivalents of three, four, and six, fragments, dicentrics, and ring chromosomes. A large portion of the aberrations from the contaminated area came from a single plant (sample #46) in which 85% of the late prophase-metaphase I cells had seven bivalents and one quadrivalent (Fig. 3) indicating that it is heterozygous for a reciprocal translocation. Many cells from both areas had eight bivalents and two univalents (Fig. 4), and a few cells from both areas had nine bivalents and a ring chromosome or a univalent (Fig. 5).

Because the aberrant cells noted in sample #46 are so much more frequent than in the rest of the population we have recalculated the values in Table 1 both including and omitting the data from that shrub. When this data is omitted the percentage of aberrant cells drops from 14.2% to 6.6%, compared with 3.8% for non-irradiated shrubs.

Fig. 3. Seven bivalents and ring quadrivalent (note arrow) from *Artemisia spinescens* sample #46 which is heterozygous for a translocation.

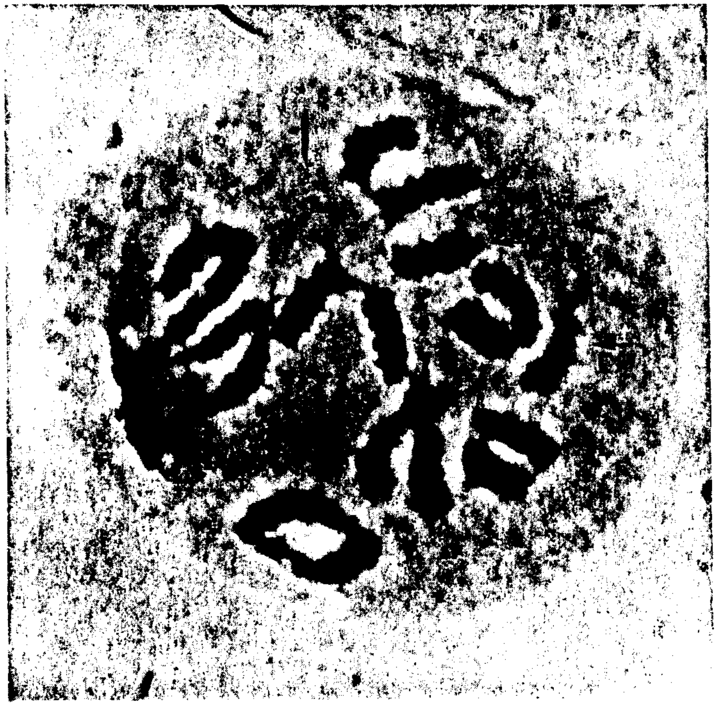

Fig. 4. The frequent condition of eight bivalents and two univalents (note arrows) in *Artemisia spinescens*.

A Mann-Whitney statistical test (Conover, 1971) for differences between the irradiated and non-irradiated populations utilizing all data does not indicate a difference at the usually accepted levels of statistical significance. (Test statistic = 51; w.95 = 57 for a two tailed test. Null hypothesis that the average proportion is same in both populations cannot, therefore be rejected.) If however, the highest value in each population is omitted from the calculations, the difference is significant at the 95% level. (Test statistic = 43; w.95 for a two tailed test is 42. The null hypothesis that there is no difference between the average proportions in both populations can, therefore, be rejected.)

Fig. 5. The arrow points to a ring chromatid in *Artemisia spinescens*.

The species, *Machaeranthera tortifolia* came from the experimentally irradiated plot in Rock Valley, NTS. This species is characterized by six chromosome pairs consisting of three large and three slightly smaller pairs (Fig. 6). A large number of extranuclear bodies occur in telophase I and telophase II (Fig. 7), although they are not as large or prominent as those of *Artemisia spinescens*. Flower buds collected 14 and 29 April 1976 contained meiotic cells at the proper stage for analysis. Chromosomal aberrations detected included five bivalents and two univalents, acentric fragments, and dicentrics (Figs. 8, 9, 10, and 11; Table 2). No multivalents were observed. Of the 2,714 cells analyzed, 1,767 came from nine plants with doses ranging from 2.5 to 4.5 R/day, and 947 cells were from five plants outside the immediate irradiated area where the dose rate was less than 1.0 R/day. One of this latter group came from an area remote to the radiation source where the radiation level was background. It has been included with the lower level for analysis, however. A large part of the aberrant cells from the high dose area came from a single plant which contained 64% of the anaphase cells showing a dicentric and/or a fragment (Fig. 11) showing that the plant was heterozygous for a paracentric inversion.

Fig. 6. The normal condition of six large bivalents in *Machaeranthera tortifolia*.

The Mann-Whitney test does not indicate significant differences between these populations.

The third species *Krameria parvifolia* which came also from the Rock Valley site, has six large chromosomes (Fig. 12). Flower buds with meiotic cells were collected on 19 and 20 May 1976.

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The species, *Machaeranthera tortifolia* came from the experimentally irradiated plot in Rock Valley, NTS. This species is characterized by six chromosome pairs consisting of three large and three slightly smaller pairs (Fig. 6). A large number of extranuclear bodies occur in telophase I and telophase II (Fig. 7), although they are not as large or prominent as those of *Artemisia spinescens*. Flower buds collected 14 and 29 April 1976 contained meiotic cells at the proper stage for analysis. Chromosomal aberrations detected included five bivalents and two univalents, acentric fragments, and dicentrics (Figs. 8, 9, 10, and 11; Table 2). No multivalents were observed. Of the 2,714 cells analyzed, 1,767 came from nine plants with doses ranging from 2.5 to 4.5 R/day, and 947 cells were from five plants outside the immediate irradiated area where the dose rate was less than 1.0 R/day. One of this latter group came from an area remote to the radiation source where the radiation level was background. It has been included with the lower level for analysis, however. A large part of the aberrant cells from the high dose area came from a single plant which contained 64% of the anaphase cells showing a dicentric and/or a fragment (Fig. 11) showing that the plant was heterozygous for a paracentric inversion.

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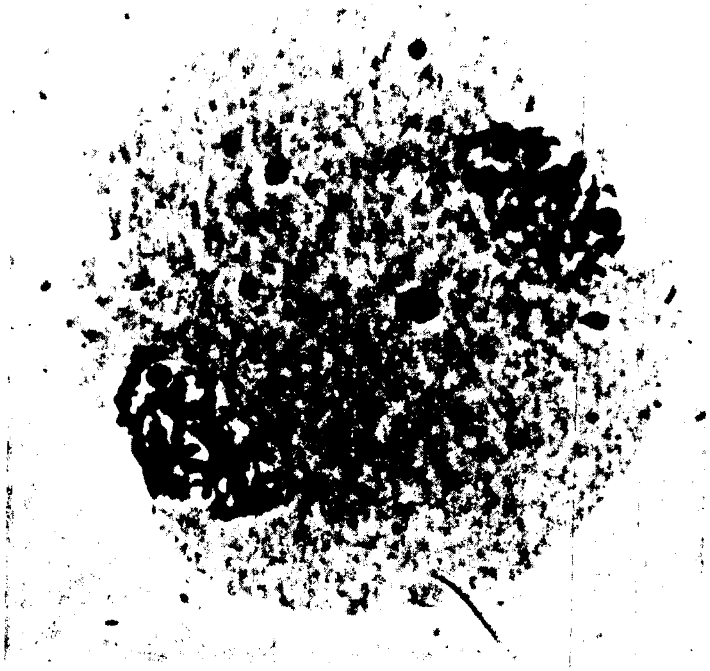

Fig. 7. Small spheres are extranuclear bodies in *Machaeranthera tortifolia*.

Aberrations observed on the two radiation doses include dicentrics and fragments in anaphase I – telophase I and anaphase II – telophase II (Fig. 13). Of the 2,839 cells scored, 1,866 were from seven plants from the enclosure near the radiation source with doses ranging from 3.2 R/day to 4.8 R/day; and 973 were collected from four plants outside the immediate range of the radiation source with a dose of less than 1.0 R/day. The anaphase I – telophase I cells with the higher dose had 9.1% aberrations, as compared with 1.0% from the cells from the lower dose area. For anaphase II – telophase II 10.4% of the cells from the higher dose area were aberrant compared to 2.8% of those in the lower dose area (Table 3).

Fig. 8. The infrequent condition of five bivalents and two univalents (note arrows) in *Machaeranthera tortifolia*.

The Mann-Whitney test indicated a difference in the average proportions of aberrants between the two populations at the 99% confidence level. (Test statistic = 28; w.005 for a two-tailed test = 27; the null hypothesis that the populations are the same could therefore be rejected.)

Because of the small number of cells found in metaphase I we have not tabulated these data; however, one plant from the higher dose area showed 94.9% aberrations and one plant from the lower dose area had only 1.6% aberrant cells. The high value for these metaphase I aberrations was primarily due to a large number of cells with four bivalents and one quadrivalent, suggesting that the plant was heterozygous for a translocation (Fig. 14).

Fig. 9. A fragment (note arrow) in *Machaeranthera tortifolia*.

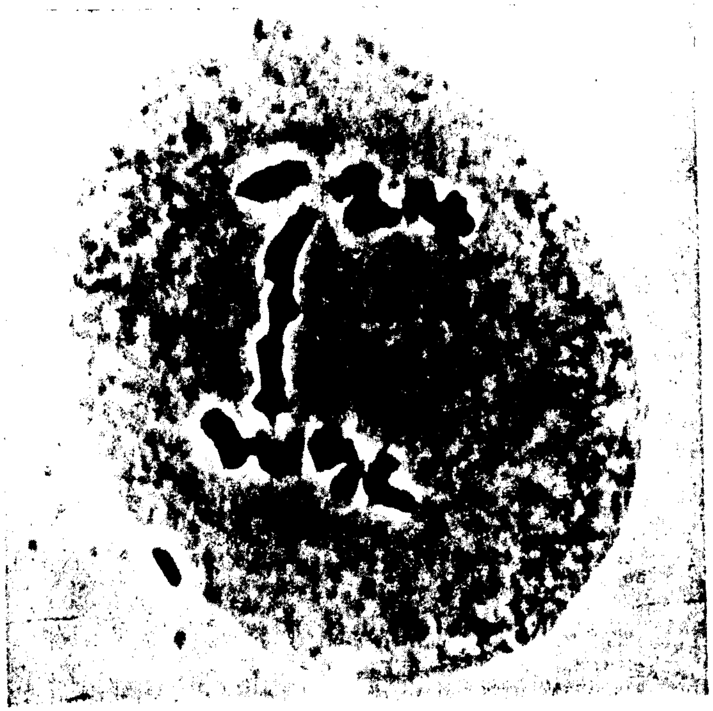

Fig. 10. A dicentric cell in *Machaeranthera tortifolia*.

Fig. 11. Dicentric and fragment from *Machaeranthera tortifolia* sample #68 which is heterozygous for a paracentric inversion.

Table 2. Pollen mother cell chromosomal aberrations in *Machaeranthera tortifolia* from Rock Valley Experimental Radiation Field

NEAR THE SOURCE – DOSE RATE 2.5 TO 4.5 R/DAY

Sample Number	Prophase I – Metaphase I					Anaphase I – Telophase I				
	Fragments	5II,2I[1]	Total Aberrations	"Normal"	Total	Fragments	Dicentrics	Total Aberrations	"Normal"	Total
68	1	1	2	149	151	0	93	93	52	145
132	0	1	1	149	150	0	6	6	37	43
133	0	0	0	157	157	0	0	0	39	39
134	0	1	1	111	112	0	0	0	0	0
151	7	1	8	68	76	0	5	5	28	33
157	0	0	0	151	151	0	9	9	77	86
208	0	0	0	150	150	2	4	6	71	77
210	0	1	1	155	156	1	0	1	27	28
211	0	2	2	149	151	0	4	4	58	62
Totals	8	7	15	1239	1254	3	121	124	389	513
Percents	0.6	0.6		98.8	100.0	0.6	23.6		75.8	100.0

Percent Aberrants Prophase I – Metaphase I (1.2 Total Aberrations)

Percent Aberrants Anaphase I – Telophase I (24.2 Total Aberrations)

Percent Aberrants Prophase – Metaphase Plus Anaphase I – Telophase I = 139/1767 = 7.9%[2]

OUTSIDE IMMEDIATE SOURCE AREA – DOSE RATE LESS THAN 1.0 R/DAY

114	0	0	0	156	156	2	8	10	98	103
118	3	0	3	153	156	0	9	9	4	13
122	1	0	1	149	150	0	3	3	30	33
145	0	1	1	59	60	0	3	3	34	37
219	0	0	0	153	153	1	7	8	73	81
Totals	4	1	5	670	675	3	30	33	239	272
Percents	0.6	0.1		99.2	99.9	1.1	11.0		87.9	100.0

Percent Aberrants Prophase I – Metaphase I (0.7 Total Aberrations)

Percent Aberrants Anaphase I – Telophase I (12.1 Total Aberrations)

Percent Aberrants Prophase – Metaphase Plus Anaphase I – Telophase I = 38/940 = 4.0%[2]

[1] Five bivalents plus two univalents.

[2] These values are not statistically different based on Mann Whitney test.

Fig. 12. The normal condition of six large bivalents in *Krameria parvifolia*.

Fig. 13. Dicentric and fragment in *Krameria parvifolia* telophase II.

DISCUSSION AND CONCLUSIONS

There is a large literature, too large for citing here, showing that ionizing radiation produces detectable changes in chromosomes. The following discussion therefore assumes that ionizing radiation causes chromosomal aberrations and attempts to address the possibility that the low-level chronic radiation doses at Site D, Area 11 may have produced an increase in chromosome aberrations in *Artemisia spinescens* there.

The occurrence of chromosome aberrancies in *A. spinescens* in the chronic, low level radiation exposure of Site D, Area 11, NTS, is well substantiated. The fact that the percentages of aberrancies do not meet generally accepted levels of statistical significance for a conclusive difference between the Site D population and our control population raises several questions in interpreting these data. The first question concerns the uncertainty of dose, already mentioned, for those shrubs in Site D. In light of what appears to be a relatively high aberrancy rate in those shrubs where background radiation is "normal," the question of dose must also be considered for those shrubs selected as controls. Since we were not able to obtain, within the time for this work, similar cytological material from regions outside areas of possible radiation doses from nuclear tests in the last 20 years, we have no basis for knowing what the normal frequency of chromosomal aberrancies may be. In addition, there appears to be nothing in the literature concerning the frequency of chromosomal aberrations in plants in nature. Giles (1940) studied species of *Tradescantia,* a herbaceous genus, in the laboratory which "have shown a comparatively high frequency of spontaneous chromosome alterations ..." A more sensitive hybrid used by him had a frequency of 0.3% and the mean for all *Tradescantia* was 0.11%. Steffensen (1964) also reported on the spontaneous rate of chromosomal aberrancies in *Tradescantia.* He found a 1.3% rate of occurrence (13 aberrants in 997 cells) and that the radiation dose which doubled this rate was only 0.75 R. (His were acute doses and the effectiveness of acute doses in causing radiation damage at the phenological level and presumably at the chromosomal level is known to be higher than for chronic doses [Sparrow and Puglielli, 1969]). Nichols, 1941 studied spontaneous chromosome aberrancies in onion seedlings where the frequency was variable and high, however "No chromosome aberrations were found in the primary root tip divisions in onion bulbs ..." In short, there appears no firm basis for evaluating what the spontaneous aberrations rate in *Artemisia* might be unless one is willing to compare a woody shrub with herbaceous species in the laboratory. Steffensen's radiation dose for doubling the rate of occurrence of chromosome aberrants, 0.75 R, even though it is an acute dose, is of interest, however, since it suggests that chromosomes are sensitivie to low radiation dose exposures.

It is also of interest that woody species, generally, are more sensitive to radiation damage than herbaceous species by a factor of about two (Sparrow and Sparrow, 1965).

The large variability in the frequency of occurrence of chromosome aberrancies in both the Site D population and the control population also raises some questions as to precisely what the variability in doses may have been to the tissues we have collected for examination, a variability beyond that contributed by the uncertainty of dose exposures to a particular shrub at a particular location with the irregular and uncertain distribution of Pu contaminating the

areas around shrubs already mentioned. (It should be pointed out that the possibility radiation exposure from external radioactive contamination of the cells themselves is quite unlikely because of the use of cells from within anthers taken from unopened flower buds. Such cells had never been exposed to the open environment.) If for example a shrub produced branches from its base or from underground, the branches' exposure to ionizing radiation will have been only for the time it has existed above the surface; and this will determine the dose it will have received. It has, therefore, to be recognized that we are dealing with large variations in radiation doses even from one part of a shrub to another part. From this it follows that a large variability is a likely characteristic of the occurrence of any aspects of radiation effects under conditions of long-term low-level chronic radiation, particularly where the term of the radiation exceeds the life span of the shrub itself, or parts of it. Under these conditions, even if the radiation field were extremely uniform, the exposure doses to particular samples taken from vegetation within such a field would undoubtedly be quite variable; hence, variability in any manifestation of radiation effects probably ought to be expected. This kind of variability, of course, does not lend itself to statistical treatment except where very large samples can be taken; and the cytological examination of large samples demands a large expenditure of time and money. It seems probable it is these variabilities that do not allow statistical support for a conclusion that chronic low-level exposures produced increased chromosomal aberrancies. We feel, however, the evidence firmly supports the hypothesis that such has occurred at Site D.

The hypothesis that chromosomal aberrancies are indeed caused by ionizing radiation in the field even where morphological changes are not apparent is supported by the occurrence of similar aberrancies in the two other shrubs, *Machaeranthera* and *Krameria* from the Rock Valley, NTS experimentally-irradiated shrub populations. It should be noted that both these shrubs received radiation exposures much higher than the Site D shrubs. Again, however, it can be anticipated that the radiation doses for particular samples taken from the field will be quite variable for different reasons to those for the Site D plants. The Rock Valley samples were irradiated from a central source, a high energy gamma emitter in a stationary position 15 meters above the surface (French, et al., 1974). From this point, exposure doses to shrubs at the ground surface will be varied by shadow effects imposed by other shrubs between the sampled shrubs and the central source. This is particularly the case of lower-growing shrubs like *Krameria* or *Machaeranthera.* Although the dose rates in the air above a particular shrub can be predicted and measured with relatively great accuracy and will have a uniformity varied primarily by atmospheric conditions, the ground level doses will no doubt vary widely and will depend not only on the atmospheric conditions but on the mass of vegetation interposed between the sampled shrub, or parts of it, and the radiation source.

The variability in occurrence of aberrancies among our shrub species is likely to be different for still other reasons, in part at least, beyond arguments presented so far. The difference in variability between *Machaeranthera* and *Krameria*, as possibly reflected in the statistical analysis, may be attributable to the phenology of the two shrubs. *Machaeranthera*, a suffruticose perennial, develops flower heads from tissue which arises, apparently, annually from a low woody base which is itself of varied longevity, and may arise from underground. *Krameria*, on the other hand, produces branches, also from underground, but much less frequently on which flower are borne for several successive years.

Fig. 14. Four bivalents and ring quadrivalent (note arrow) in *Krameria parvifolia* plant heterozygous for a translocation.

Table 3. *Krameria parvifolia* pollen mother cells with chromosomal aberrations from Rock Valley Experimental Radiation Field

NEAR SOURCE 3.2 TO 4.8 R/DAY

Sample Number	Anaphase I – Telophase I						Anaphase II – Telophase II					
	Fragments	Dicentrics	Both	Total Aberrations	"Normal"	Total A-T I	Fragments	Dicentrics	Both	Total Aberrations	"Normal"	Total A-T II
222	10	3	2	15	126	141	6	3	4	13	76	89
223	13	6	1	20	137	157	9	11	2	22	61	83
224	13	1	2	16	115	131	6	1	2	9	141	150
229	4	0	3	7	145	152	10	4	4	18	133	151
230	12	2	0	14	142	156	9	3	1	13	139	152
232	9	1	0	10	140	150	2	1	0	3	48	51
233	7	3	3	13	140	153	4	3	1	8	142	150
Totals	68	16	11	95	945	1040	46	26	14	86	740	826
Percents	6.5	1.5	1.1		90.9	100.0	5.6	3.1	1.7		89.6	100.0
Percent Aberrants, Anaphase I – Telophase I (9.1 Total Aberrations)							Percent Aberrants, Anaphase II – Telophase II (10.4 Total Aberrations)					
Percent Aberrant, Anaphase I – Telophase I + Anaphase II – Telophase II = 181/1866 = 9.7%[1]												

OUTSIDE IMMEDIATE SOURCE AREA – LESS THAN 1.0 R/DAY

236	1	1	0	2	124	125	3	0	2	5	155	160
242	0	0	0	0	150	150	2	0	0	2	76	78
255	2	0	0	2	150	152	1	1	2	4	146	150
256	2	1	0	3	147	150	0	0	0	0	8	8
Totals	5	1	0	6	571	577	6	1	4		385	396
Percents	0.9	0.2	0		99.0	100.0	1.5	0.3	1.0		97.2	100.0
Percent Aberrants, Anaphase I – Telophase I (1.0 Total Aberrants)							Percent Aberrants, Anaphase II – Telophase II (2.8 Total Aberrations)					
Percent Aberrant, Anaphase I – Telophase I + Anaphase II – Telophase II = 17/973 = 1.75%[1]												

[1] These values are statistically different based on Mann Whitney test.

The flower producing tissue of a branch of *Krameria* is therefore more likely to have been exposed more uniformly in time than a branch with a flower head in *Machaeranthera*, and quite possibly with larger doses for a position equidistant from the radiation source for the following reason. *Krameria* is more likely to form its own clumps, independent of nearby species compared to *Machaeranthera*, which often grows under, or within, shrubs with heavier woody stems such as *Lycium* and *Ephedra*. This particularly contributes to the uncertainty of dose to any particular tissue within *Machaeranthera* shrubs. We note in passing that differences in the rate of occurrence of chromosome aberrations at two dose levels for *Krameria* is significantly greater at higher doses than at lower doses by conventionally accepted statistics.

There are still other conditions which support the hypothesis that chromosomal aberrations should be expected even in the low-level radiation areas:

1) High interphase chromosome volumes. Precisely the characteristics which render a plant suitable for cytological analysis, i.e., a small number of large chromosomes with a high interphase chromosome volume, characterizes the more radiosensitive species (Sparrow, 1962; Sparrow, et al., 1965). All three species tested here have relatively large chromosomes and should therefore be among the more radiosensitive. Wallace and Romney, 1972 repored that *Machaeranthera tortifolia* and *Atriplex spinescens* were the most sensitive, based on phenological or morphological changes, of 31 NTS species studied using gamma radiation in the laboratory. We have already noted that *Krameria parvifolia* in Rock Valley was one of the shrubs responding to increasing exposure rates with reduction in shrub size, more dead stem material, and less foliage on live stems (Vollmer and Bamberg, 1975).

2) *Artemisia*, particularly, may be more sensitive to radiation effects for yet another reason. The meiotic division was found to be the most radiosensitive stage in the plant's life cycle (Sparrow, 1951); and a plant which has a long meiotic period would probably be still more sensitive. Such is likely the case for *Artemisia* which breaks dormancy very early in the spring (it is among the first to do so); and because of the cool conditions the meiotic cycle is likely to be longer than for other, later-flowering, species. Thus there appears to be a longer time than for other shrubs during which *Artemisia* may be radiation sensitive.

3) Environmental stresses are known to intensify radiation damage, stresses such as heat, cold, and drought (McCormick and Platt, 1962; Woodwell, 1962) to which the NTS species are all subjected.

A few words should be said about the detectability of chromosome aberrations. There are no doubt many aberrations that will remain undetected because they occur in cells whose chromosomes are insufficiently spread out for cytological analysis. Some aberrations cannot be detected. These include pericentric inversions, homozygous inversions and translocations, heterozygous paracentric inversions in which crossover did not occur within the inverted loop, and inversions with two strand double cross-over within the inversion loop. Also fragments may be hidden in a group of migrating chromosomes, dicentrics may break early in anaphase, and multivalents are sometimes difficult to distinguish from superimposed bivalents. For all these reasons, the percentages of aberrations detected probably do not present more than an uncertain fraction of the total. Detectable aberrancies are, however, generally acceptable as comparative basis for analyzing populations.

Unlike carefully controlled laboratory studies of radiation induced chromosome aberrations, it is not possible to know in which stage of the cell cycle or in what part of the plant life cycle the actual radiation "hits" occurred. Sparrow, et al., (1961) noted that in chronic irradiation effects, most observed damage is a result of the previous one or two cell divisions leading to lethal conditions produced by fragments and dicentrics. Aberrations such as heterozygous inversions and translocations, however, are not lethal in somatic tissue because the complete complement of genetic material is maintained, although in a different order. These structural rearrangements sometimes produce morphological or physiological changes by altering linkage groups (Swanson, et al., 1967). We noted the occurrence of a high frequency of aberrants indiciating a heterozygous inversion in *Machaeranthera* (Table 2, Shrub Sample #68), and a a similar high frequency indicating a translocation in *Artemisia* (Table 1, Shrub Sample #46).

If indeed there are radiation induced chromosomal aberrations in Site D, Area 11, according to our hypothesis there is some data which allows some predictions as to long-term ecological effects. Chromosomal aberrations can lead to a general decline in productivity and eventual disappearance of radiosensitive species in an ecosystem (Garrett, 1967; Woodwell and Sparrow, 1965; Woodwell, 1962; Woodwell and Whittaker, 1968). However, radiation has long been utilized as a method of producing beneficial mutations in crop plants (Smith, 1958; William and Scully, 1961); and it is possible that genetic changes could occur as a result of radiation which might establish individuals or species better adapted to their environment. These adaptations could include radioresistance due to polyploidy, smaller chromosomes, asexual reproduction, and faster mitosis (Sparrow and Evans, 1961; Evans and Sparrow, 1961). With such mutations the resistant species might expand and become dominant.

Finally, whether one accepts the hypothesis that there are more than normal chromosome aberrancies in Site D, Area 11, or rejects it depends on interpretation of statistical criteria. The argument has been presented that large variability should be expected on biological and environmental conditions, and that producing conclusively adequate statistical data requires very large samples which verge on unfeasibility because of the complexity of cytological procedures.

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TRANSURANIC ELEMENTS IN TERRESTRIAL ANIMALS AND THE ENVIRONMENT: AN INTRODUCTION

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ABSTRACT

This discussion provides background information to the session on the "Transuranic Elements in Terrestrial Animals." Briefly outlined are some of the historical events leading to the introduction and dispersion of the transuranic elements into the biosphere, to the establishment of the Nevada Applied Ecology Group (NAEG), and to the studies conducted by the Environmental Monitoring and Support Laboratory (EMSL-LV) and the University of Nevada-Las Vegas involving the transuranics distributed by the "safety shots" and the nuclear weapons testing program at the Nevada Test Site and the Tonopah Test Range. These studies are described in relation to the overall objectives of the NAEG program. Other potential sources of the transuranic radionuclides are also discussed.

SESSION INTRODUCTION

Historically, the nuclear weapons test in Alamagordo, New Mexico, in July of 1945 resulted in the first significant injection of transuranic elements into the biosphere. The first thermonuclear device detonated in 1952 also injected large amounts of radioactive debris into the stratosphere (Thomas and Perkins, 1974). The higher energy yield of thermonuclear devices led to the increased production of transuranic elements and their distribution in the biosphere in significant amounts as a result of U.S., Russian, British, French, and Chinese nuclear weapons testing programs carried out in subsequent years.

The burnup of a SNAP reactor in 1964 during atmospheric reentry has also contributed to transuranic materials being introduced into the biosphere. Other incidents such as those at Palomares, Spain, and at Thule, Greenland, had the potential of adding substantially to the worldwide inventory. Furthermore, transuranium nuclides are presently being generated on a long-term basis as a result of the production of fuels for nuclear power reactors.

The historical background related to the establishment of the Nevada Applied Ecology Group (NAEG) is described in the preface of the first NAEG report entitled "Dynamics of Plutonium in Desert Environments" NVO-142 edited by Dunaway and White (1974). This report states that the Planning Directive NVO-76 established the NAEG under the Office of Effects Evaluation, whose responsibility includes the determination and prediction of effects of radioactivity on biota, particularly with respect to food chains and other environmental factors affecting man. The stated purpose of the NAEG is "...to coordinate the ecological, radiation monitoring and other environmental programs necessary to support continued nuclear testing activities and to provide the mechanism to effectively comply with the requirements of the National Environmental Policy Act of 1969..."

An early concern of the former AEC, currently the Energy Research and Development Administration (ERDA), and the U.S. military organizations responsible for nuclear weapons was for the safety of these devices with respect to storage, handling, transport and accidents. This, in turn, led to a number of experiments designed to test for "safety against fission reaction." Basically, these experiments consisted of detonating a chemical explosive in close proximity to an assembly of plutonium and/or uranium and were appropriately called "safety shots." These tests have resulted in the local distribution of plutonium and/or uranium at specific sites located at such areas as the Nevada Test Site (NTS), Tonopah Test Range (TTR) and the Pacific Proving Grounds. The wisdom of carrying out these tests was demonstrated by accidents near Palomares, Spain, Thule Air Force Base, Greenland, and elsewhere.

To date, intensive studies on the transuranium nuclides carried out at specific "safety shot" areas at the NTS and at the TTR have been a principal function of the NAEG since its inception. Much of the historical background for these studies is described in the preface to NVO-76 mentioned previously (Dunaway and White, 1974). However, during the past year or so, increasing attention has been directed toward extending these studies to include areas in the immediate vicinity of old nuclear test events which were associated with Plowshare cratering tests, surface shots or accidental ventings.

As a result, both large and small animal studies have been incorporated into this program in the form of field and laboratory studies. The large animal studies are carried out by the Environmental Monitoring and Support Laboratory of the Environmental Protection Agency at Las Vegas (EMSL-LV). These studies are concerned primarily with the transfer of plutonium and other transuranium nuclides to man via the food chain, with special emphasis on milk and meat. The principal animals used in these studies include dairy cattle, dairy goats, and beef cattle (specifically the NTS range herd). The objectives of the laboratory experiments conducted at EMSL-LV include determining: (a) the transport of plutonium and other transuranic elements from orally administered doses to milk and tissues (or meat products) and the excretion rates of these materials in urine and feces; (b) the transfer of transuranics to juvenile animals via *in vivo* labeled milk; (c) transfer of ingested plutonium and americium to edible tissues and eggs of domestic fowl. (d) solubility studies of known chemical forms of transuranic elements, as well as field-deposited materials on forage, in an artificial rumen-simulated gastrointestinal system which relate directly to the solubility and subsequent absorption of radionuclides in the intact animal. In general, the field studies conducted by EMSL-LV involve: (a) beef cattle placed to forage on contaminated rangeland along with fistulated steers used as sample collectors for forage, and (b) the collection of indigenous wildlife such as deer, coyotes and jackrabbits for the analysis of transuranic nuclides in their tissues.

The fistulated steers which are used as vegetation sample collectors have a surgically placed "window" to the rumen from which ingested samples can be retrieved in order to determine the preference grazing cattle have for local vegetation. In addition, such rumen samples may also be analyzed to determine the qualitative and quantitative makeup of the radionuclides associated with the ingested forage. Selected animals from the range herd are slaughtered on a semiannual schedule and their tissues are examined for abnormal pathology and are analyzed for specific radionuclides (Smith, 1975).

In situ ecological studies of small indigenous vertebrates have been carried out by Drs. Bradley and Moor of the University of Nevada-Las Vegas (Bradley and Moor, 1975). These studies include ecology, population, and reproduction studies of small mammals, birds and reptiles in contaminated areas of the NTS and the TTR. Tissue samples from these animals are analyzed for radionuclide content in order to relate these to existing contaminants in the environment, especially plutonium and americium.

Thus it can be seen that the large and small animal programs indeed develop required mission information for ecological radiation monitoring in both large and small animals. The domestic animals are of economic importance and are a direct link in the food chain to man, and the small indigenous vertebrates are a part of local ecosystems. Much of the derived information is used as input for modeling the potential impact of the transuranium radionuclides on man through food chain transport and on local ecosystems.

In addition to the nuclear testing programs, transuranic nuclides are also produced in the generation of electrical energy via fission reactors and it has been estimated that by the end of the century approximately one quarter of a million tons of radioactive wastes will have been generated, including approximately 2 x 10^9 curies of the transuranium elements. Thomas and Perkins (1974) have presented an excellent review of this material; for example, they show that the activities of ^{241}Pu/^{241}Am and of $^{242/244}$Cm in nuclear wastes range from two to three orders of magnitude greater than ^{239}Pu. However, they also mention that it is obvious that the ratios of transplutonium elements to ^{239}Pu in nuclear fuels themselves will be much less than those in the nuclear wastes. During transport or reprocessing, these wastes could be accidentally released to the environment in a variety of chemical or physical forms. The information gained from the study of transuranics in the environment and food chain of man could be of great benefit in such an event. It is toward this end that we have also had a certain amount of DBER support, through the NAEG, to study the transport of curium and neptunium in the cow-milk/meat link of the food chain and the experimental portion of two curium studies and one neptunium study have been completed. However, the analysis of the data from these studies is not as yet complete, but will probably be presented at the next NAEG annual meeting.

As a part of an EPA-funded program, our Laboratory has recently published a literature review on "The Availability, Uptake and Translocation of Plutonium Within Biological Systems" by Mullen and Mosley (1976).

This introduction serves to introduce the next four speakers who will cover their studies in detail. Drs. Bradley and Moor will cover the small indigenous vertebrates, Dr. Sutton will discuss the transport to milk, Dr. Smith will review the grazing studies, and Mr. Barth will discuss the application and significance of the artificial rumen and simulated gastrointestinal systems.

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PLUTONIUM AND OTHER TRANSURANICS
IN SMALL VERTEBRATES: A REVIEW

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ABSTRACT

The published data relevant to transuranics in small vertebrates inhabiting terrestrial environments is reviewed. Experimental results indicate that atomic size and valence state affect rates of absorption, transportation, and excretion of transuranics in living systems. Whereas there is a marked tendency for transuranics to hydrolyze to insoluble colloidal products at physiological pH, complexing agents and chelation enhance solubility and transportability. The natural modes of uptake of transuranics by vertebrates include absorption from the gut, the intact or damaged skin, and inhalation. Absorption from the gut into the bloodstream is very low. Potential hazards may exist if complexing or chelating agents are present, if absorption is continuous, or if exposure involves young animals. The intact skin provides an effective barrier to absorption of transuranics. Relatively high levels of absorption may occur when transuranics are administered subcutaneously or intramuscularly, particularly with increased acidity and solubility of the compounds. Inhalation is probably the most hazardous natural route of uptake. Insoluble transuranic compounds are retained in the lung and soluble compounds are transported rapidly via the blood to bone, liver, and other organs. Deposition of plutonium in mammalian gonads resulting in a decrease in spermatogenesis, ovarian damage, and reduced fecundity is documented. The current knowledge of the behavior of transuranics in terrestrial environments is limited. Plutonium and americium uptake by small mammals has been documented. High variability in animal tissues makes analysis difficult. In addition, there is evidence that uptake may be species related. Whereas resuspension of sedimentation (inhalation) may be the most important route of uptake, high levels of plutonium and americium in the gut of small mammals indicate that chronic ingestion may be an important hazard. Plutonium body burdens were related to depressed leukocyte count on a statistical basis. Reduced rodent populations in areas of high plutonium concentrations illustrate the problems of evaluating uptake by mobile animals.

INTRODUCTION

The general concensus from the open literature is that, potentially, the most biologically hazardous materials known to man are radionuclides belonging to the actinide series. The transuranics (neptuniun, plutonium, americium, and curium) may represent the most significant hazards in our environment because of their increasing production in reactor operations and generally long half-lives (McKay, 1961). However, few data have been published on the behavior of transuranics in terrestrial environments and these are mostly limited to plutonium. The purpose of this presentation is not to review the exhaustive literature on laboratory studies of transuranics which have been mainly plutonium studies, but to assemble the published data relevant to transuranics in native small vertebrates inhabiting somewhat natural terrestrial environments.

CHEMICAL PROPERTIES OF BIOLOGICAL IMPORTANCE

In the actinide series, there is a gradual contraction in atomic and ionic radii from thorium to lawrencium. In addition, the ionic radius is smaller for higher valence states of each element. These size variations are important in determining relative complex-forming tendencies and ion-exchange properties (Cleveland, 1967). In general, this decrease may be accompained by an increase in solubility of oxide, in the stability of chelates, and a decrease in basicity. In addition, multiple valency states of many of the actinides are especially important in determining complex-forming tendencies. These cations are readily hydrated at physiological pH giving rise to insoluble, colloidal, and probably polymeric products. Less readily hydrated cations may exist as complexes which are more soluble and more monomeric, hence more transportable in living systems. In addition, isotopes of high specific activity (low mass) are more readily transported indicating mass is related to the formation of colloidal or polymeric material. Atomic size and valence state, then, affect rates of absorption, tranportation, and excretion of actinides in living systems. The trivalent oxidation state is dominant for americium and higher actinides. The most stable oxidation states for transuranics neptunium and plutonium are 5 and 4, respectively (ICRP, 1972). Durbin (1975) reported that tetravalent Pu, the most important oxidation state in biological systems, is the most likely to polymerize forming radiocolloids under physiological conditions. For a thorough review of chemistry of actinides, see Katz and Seaborg (1957), Cleveland (1967), and Taylor (1973).

LABORATORY STUDIES

The biology of transuranics was the subject of several symposia (Hanford, 1962, 1972; IAEA, San Francisco, 1975). A good review of the metabolism of some actinides can be found in ICRP (1972) and Durbin (1973). Whereas actinides have a number of valence states and tend to hydrolyze to insoluble complexes in physiological systems, it has been demonstrated that if they enter the body as stable compounds with a chelating agent, their biological behavior may be considerably modified. It should be emphasized that introduction into biological systems during experimentation has been highly variable as to mode of introduction, chemical form, and dosage, and the results often do not readily lend themselves to interpretation or generalization. The general consensus, however, is that it is extremely unlikely that uncomplexed transuranic ions can exist to any significant extent in vertebrate systems.

The physiological behavior of the transuranics appears to be related to mode of uptake and is valence dependent. Durbin (1962, 1975) and Nenot *et al.* (1972) discussed transuranics in mammalian tissues in relation to chemical properties and route of uptake. A decrease in ionic radius was negatively correlated with skeleton burden and positively correlated with liver burden. Schubert *et al.* (1961) and Lindenbaum and Rosenthal (1972) have pointed out that colloidal solutions are taken up preferentially by organs containing reticuloendothelial cells, primarily liver, but also spleen and bone marrow. More monomeric solutions are largely retained in the skeleton. Taylor (1972) discussed interactions of transuranium elements and the components of cells and tissues. He emphasized the importance of proteins as complexing molecules and the lysosomal association of some transuranics in cellular metabolism.

Of these transuranics, plutonium has received the most attention and has been studied intensively in laboratory animals. Whereas much fewer data are available for other transuranics, the available data suggest that generalizations about the behavior of plutonium may be carefully extended to these other transuranics.

From the results of numerous studies summarized in Wick (1967), ICRP (1972), Hodge *et al.* (1973), and several symposia (Hanford, 1962, 1972; LASL, 1974), the metabolic distribution of plutonium is determined by the chemical form, route of uptake, age, sex, physiological condition, and species of animal. The natural modes of entry include absorption from the gastrointestinal (GI) tract, from the intact or damaged skin, and inhalation.

The results of many laboratory studies indicate that actinides are not absorbed from the digestive tract in significant quantities. This was expected because the high pH in the duodenum of mammals favors hydrolysis and the formation of colloidal and polymeric material. Hamilton (1947, 1948) reported that less than 0.05% of some lanthanides and actinides were absorbed from the GI tract of rats. In later studies with rats, Katz *et al.* (1955) and Weeks *et al.* (1956) estimated that from 0.002 to 0.05% Pu was absorbed from the GI tract. In their study, skeletal deposition was approximately 90% of absorbed dose and no significant losses from the skeleton were reported during a 250 day observation period. Increased absorption of actinides has been reported when administered with chelating agents or citrate complexes. The percentage of intragastrically administered Pu absorbed in the GI tract of rats increased 700 fold when administered with DTPA plus citrate (1.7% of administered dose) when compared to absorption of Pu nitrate (0.0011%) (Baxter and Sullivan, 1972). Citrate and DTPA, when administered separately, were slightly less effective in increasing absorption, 0.15 and 0.79% of absorbed dose, respectively, although Pu retention was highest with citrate (0.091%). Ballou *et al.* (1962) also reported that neptunium citrate was more readily absorbed than Np nitrate and that Np absorption in the gut was higher than Pu by an order of magnitude. Scott *et al.* (1949), using $^{241}AmCl_3$ and $^{242}CmCl_3$, estimated < 0.01 and 0.05%, respectively, were absorbed in the GI tract. Comparable values were reported using $^{249}BkCl_3$ and $^{253}BkCl_3$ (Hungate *et al.*, 1972). Elevated GI absorption of Pu in young animals has been emphasized by several investigators. Buldakov *et al.* (1970) reported high absorption and skeletal retention in young dogs. Ballou (1958) and Baxter and Sullivan (1972) estimated that Pu absorption in young rats was from 2 to 100 times greater than for adults. Moskalev *et al.* (1969) reported on the effects of continuously feeding ^{239}Pu citrate to rats from age 3 months till death. The skeleton burden was always 2 or 3 times the liver burden with the result that 5 of 68 rats developed osteosarcomas with an estimated average bone dose of 57 rads.

It is apparent from abundant literature that absorption of actinides from the GI tract into the bloodstream is almost insignificant under most conditions. Potential hazards may exist, however, if actinides are present with complexing or chelating agents, if absorption is continuous, or if young animals are exposed.

Available literature indicates that the epidermis of small mammals is an effective barrier to the absorption of transuranics, particularly the insoluble compounds most likely to be found in the environment. Increased absorption has been reported with increased acidity and solubility of plutonium compounds (Oakley and Thompson, 1955; Buldakov *et al.*, 1970; and others). Relatively high levels of absorption and retention have been reported with dermal, subcutaneous, and intramuscular administrations (ICRP, 1972; Durbin, 1973). Jee (1972) has suggested that subcutaneous and intramuscular injection, which may be similar to puncture wounds sustained under natural conditions, along with inhalation represent plutonium uptake routes most likely to result in significant skeleton burdens. Langham *et al.*, 1962, reported no systematic absorption of plutonium oxide when it was applied to abraded skin of rabbits. When a solution of $PuCl_3$ in dimethyl sulfoxide was placed on the naked sole of a rat's foot, 0.26% of the applied dose was detected in the circulating blood within 5 minutes (Watters and Johnson, 1970). Khodyreva (1968) applied plutonium nitrate (pH_5) to the skin of rabbits and estimated 0.15% was absorbed and retained after 14 days. Hungate *et al.* (1972) found that 0.03% per cm^2 of ^{253}einsteinium was absorbed when 0.01 N nitric acid was applied to naked rat skin. A maximum of 1.2% of plutonium metal implanted subcutaneously in rabbits and mice was absorbed and distributed throughout the body during lifetime (Lisko and Kisieleski, 1953). In their studies, the liver/skeleton uptake ratios were dissimilar between the two species, probably reflecting the rapid liver clearance rate reported in numerous studies of the rat. The average rat skeleton contained almost 10 times more plutonium than the average rabbit skeleton. Osteogenic sarcoma of the spine was reported from the rat with the highest burden. The results of their data indicate the desirability of obtaining more information on plutonium, and other transuranics, uptake in numerous species of animals. Weeks and Oakley (1953, 1954) reported plutonium absorption as high as 2% with 10 N nitric acid and 0.3% with 0.1 N nitric acid after 5 days. Oakley and Thompson (1955) reported increases in absorption with an increase in depth of administration. Using Pu $(NO_3)_4$ with 0.1 N nitric acid, an increase in percentage of administered dose absorbed from 0.33% in intact skin to 2.7% from subcutaneous injection with a corresponding increase in liver deposition was reported. Cable *et al.* (1962) estimated up to 12% retention in the lymph nodes and 4.8% in bones after 7 days when Pu $(NO_3)_4$ was injected intradermally. Their data indicated that percent absorption may be inversely related to dosage as Thompson (1967) later suggested. Scott *et al.* (1948) and Taylor (1969) have reported absorption values of 65 and 51% of PuO_2Cl_2 and plutonium nitrate, respectively; 256 days after administration of PuO_2Cl_2 47% was retained in the skeleton.

Whereas rather low levels of uptake of transuranics by means of ingestion or parenteral absorption into vertebrate systems may be important particularly in chronic situations, laboratory experiments indicate that inhalation is the most hazardous natural route of entry. Inhaled insoluble Pu produced lung tumors (Moskalev, 1972; Bair and Thompson, 1974). Significant increases in lung tumors of rats were reported with accumulated doses of 50 rads using inhaled ^{239}Pu citrate (Moskalev, 1972) and 9-32 rads with soluble ^{238}Pu (Sanders, 1973). Several works provide summaries of inhalation studies relating to the retention, translocation, and biological effects of Pu and in some cases other transuranics (Abrams *et al.*, 1947; Bair and Willard, 1963; Morrow, 1966; Buldakov, 1970; Sanders, 1973; ICRP, 1972; Bair *et al.*, 1973 and others). The results of these studies indicate that absorption from the lung varies from less than 1% to as much as 10 to 15%. Absorption, fate, and effects of inhaled transuranics depend on such factors as solubility, chemical form, respiratory rate, and particle size. In addition, Bair *et al.* (1966) and Buldakov *et al.* (1970) have pointed out important differences between acute versus chronic exposures.

Insoluble Pu compounds ($^{239}Pu\ O_2$) are retained in the lung and associated lymph nodes and are translocated to the bone and liver very slowly, where they are retained; hence, resulting pathologies are somewhat confined to these tissues. Soluble Pu compounds are translocated relatively rapidly via the blood to bone and liver and, to a lesser extent, lymph nodes, kidney, spleen, and gonads. Translocation rates and retention times differ between some species as rats loose Pu from liver much more rapidly than dogs. Stannard and Morrow (1956) demonstrated the importance of respiratory rate and volume to Pu deposition. Bair and Willard (1963) indicated that deposition and retention of Pu was greatest when inhaled as large particles of 0.6 μ CMD. Inhaled Pu O_2 and more soluble Pu compounds result in lymphopenia. Bair *et al.* (1966) reported all but two dogs with less than 0.003-0.0002 μCi/g Pu O_2 in lung showed persistent lymphopenia and leukopenia throughout their life span. Bair *et al.* (1973) reported there is a possible relationship between Pu-induced lymphopenia, lymp node pathology, decreased immunological capability (Dilley, 1970), and the pathogenesis of Pu-induced pulmonary neoplasia.

Whereas the greatest risk from internally deposited transuranic elements has classically involved the lung, liver, and bone (Thompson *et al.*, 1972,) deposition of Pu and other actinides in mammalian gonads has been documented. Richmond and Thomas (1975) in their review reported the fraction of administered burden of Pu for rats and mice gonads to range from 4.3 x 10^{-5} to 9.8 x 10^{-4}. These values are comparable to values reported for soft tissues other than lung, liver, and bone when differences in sex, mode of introduction, and form of the elements administered are considered. Smith *et al.* (1976) reported comparable levels of Pu in male and female gonads when ^{237}Pu and ^{239}Pu citrate were injected interperitoneally. Because testes are heavier than ovaries, ovaries had a greater Pu burden than testes on a weight basis. Excretion rate in females was also higher than males by a factor of 2.7 within the first 24 hours post-injection. The uptake of Pu by the placenta and fetus has been demonstrated (Finkel, 1947; Sikov and Mahlum, 1968; Moskalev *et al.*, 1969; Ovcharenko, 1972). The percentage of stillborn litters, as well as the number of stillborn young in viable litters, increased following intravenous administration of 0.016 µCi/g Pu (Finkel, 1947). In addition, a lower incidence of conception was seen. These findings are supported by the work of Sikov and Mahlum (1968). Ovarian damage in mice has been reported with large doses of 30 µCi/kg Pu (Bloom, 1948). Testes mass was reduced 58% 18 weeks after an intravenous injection of 10 µCi/kg ^{239}Pu citrate (Beechey *et al.*, 1975). This reduction was attributed to germ cell death corresponding to a 51% reduction in epididymal sperm count. In addition, the authors reported a 5% increase in spermhead abnormalities. Green *et al.* (1975, 1976) reported nonhomogenous distribution of Pu in testes of mice. Plutonium was concentrated in the interstitial tissues outside the seminiferous tubules resulting in an increased dose to stem cells when compared to the average dose calculated for whole testes. These data indicate a high potential risk to genetic material which may result in depression of spermatogenesis, ovarian damage, and reduced fecundity.

FIELD STUDIES

The dynamics of transuranic elements in natural environments have been reviewed by Romney and Davis (1972), Price (1973), and Bennett (1974) and was the subject of two major recent symposia (Hanford, 1972; and IAEA Symposium in San Francisco, 1972). Romney *et al.* (1970) collected small animals over a ten-year period from contaminated sites at NTS. Tissue samples from Kangaroo rats (*Dipodomys merriami* and *D. microps*) were polled to provide sufficient material for plutonium analysis. Their results were reported in DPM per sample; hence, intertissue comparisons are difficult, and their work cannot be directly compared to later investigations. They concluded that the relatively high levels of Pu found in the GI tract, which was probably due to ingestion of soil particles, indicated an additional pathway of uptake besides inhalation. The importance of ingestion as a primary route of entry of residual Pu was further emphasized by Romney and Davis (1972). Paglia (1968) reported on hematopathologic surveys of Kangaroo rats in Pu contaminated and uncontaminated study areas of NTS. Blood cell depressions were noted in rats from some contaminated areas, but a physiological significance remains to be determined. Kangaroo rats from areas of highest Pu contamination (720 mg/m^2) showed the highest Pu body burdens and greatest blood cell depressions. Hakonson and Johnson (1973) in their survey of the Trinity Site 27 years after the event found considerable variability in levels of Pu in rodent tissues. Concentration of Pu in rodent lungs was generally higher than for other tissues and was similar for that determined for soils and grass in relation to distance from ground zero. They concluded that the relatively high concentrations in the lungs suggest resuspension as the most important mechanism in biological redistribution of plutonium. In a similar investigation in liquid waste disposal areas of LASL, Hakonson *et al.* (1973) collected rodents for plutonium analysis. Plutonium concentrations in animal tissues at one sample station had as much as three orders of magnitude variation, with lung and pelt samples having highest mean Pu concentrations. Based on these data, they suggested that resuspension of sedimentation was main route of Pu uptake by animals. In addition, a rather low discrimination rate was reported. Plutonium concentrations of liver and carcass were from 10^{-1} to 10^{-2} the concentration of hide and lungs. McClendon *et al.* (1975) sampled cotton rats (*Sigmodon hispidis*) in the vicinity of the Savannah River Plant, Aiken, South Carolina. Based on 8 adult rat samples, concentation ratios of Pu in animals to Pu in soil and vegetation were from 10^{-2} to 10^{-1}, respectively. Similar levels of plutonium were found in juvenile and immature rats. Little (1976) noted that, in general, data from rodents collected at the Rocky Flats facility agreed with earlier ecological investigations with two noteworthy exceptions. The small mammal internal and external tissues were at about the same contamination level (10^{-3} nCi/g dry), and bone tissue was the lowest by 1-2 orders of magnitude. He suggested that contamination of small mammals was related to soil Pu, although clear relationships between Pu concentrations in small mammal tissue and soil were not clear. He concluded that a biologically available Pu compound or particle size distribution accounted for the similar internal and external tissue burdens. Plutonium uptake was attributed to a combination of inhalation and ingestion. The result was a high degree of variability in tissue burdens.

Plutonium concentrations in small mammals of NTS (Moor and Bradley, 1974; Moor *et al.*, 1976) reflect somewhat different results than those from other research sites. Whereas data from Hakonson and Johnson (1973) and particularly Little (1976) indicate that contamination levels of internal and external tissues were similar, internal tissue (carcasses) of small mammals from NTS were from one to three orders of magnitude lower than external tissues. In addition, concentration factors, or the ratio of Pu in tissue to Pu in soil and vegetation, were from one to two orders of magnitude lower. Residual Pu contamination at the NTS area, however, was from one to two orders of magnitude higher than Rocky Flats and five to six higher than Savannah River or Trinity Sites (Tables 1 and 2).

In way of summarizing the results of our study in Area 11-C, NTS:

1. Whereas there is a high degree of variability in tissue burdens, Pu and Am levels in small mammals appear to be related to residual Pu levels of soil and vegetation. Tissue burdens of small mammals from a High Activity study plot were significantly higher ($P<.01$) than from a Low Activity study plot (Table 2).

Table 1. Summary of Environmental Studies Dealing With Plutonium Uptake in Small Mammal Populations.

Sample	$\bar{X}$ Pu nCi/g dry wt.	$\bar{X}$ Pu Tissue/Soil Ratio	$\bar{X}$ Pu Tissue/Vegetation Ratio	Reference
Soil	4.6×10^{-5}			McLendon *et al.*, 1976 Savannah River
Vegetation	7.1×10^{-6}			
Small Mammals				
Carcass	4.6×10^{-7}	1.4×10^{-2}	1.4×10^{-1}	
Soil	3.1×10^{-4}			Hakonson and Johnson, 1973 Trinity Site
Vegetation	4.6×10^{-6}			
Small Mammals				
Hide	1.2×10^{-5}			
Carcass	3.0×10^{-5}	9.7×10^{-2}	6.5×10^{0}	
Soil	8.6×10^{-1}			Little, 1976 Rocky Flats
Vegetation	2.9×10^{-2}			
Small Mammals				
GI Tract	7.0×10^{-3}			
Hide	1.5×10^{-3}			
Carcass	8.6×10^{-3}	1.0×10^{-2}	2.5×10^{-1}	
Soil	4.1×10^{-2}			Little, 1976 Rocky Flats
Vegetation	5.6×10^{-4}			
Small Mammals				
GI Tract	2.2×10^{-3}			
Hide	2.6×10^{-3}			
Carcass	7.3×10^{-3}	1.8×10^{-1}	1.3×10^{-1}	

Table 2. Plutonium Concentrations in Resident Small Mammals of NAEG Intensive Study Area 11-C, NTS (Moor *et al.*, 1976).

Sample	$\bar{X}$ Pu nCi/g dry wt.	$\bar{X}$ Pu Tissue/Soil Ratio	$\bar{X}$ Pu Tissue/Vegetation Ratio	Pu/Am $\bar{X}$ ± SE
High Activity Strata*				
Soil	$7.2 \times 10^{+1}$			6.0 ± 0.3
Vegetation	1.4×10^{0}			5.2 ± 0.3
Small Mammals				
Hide	1.8×10^{0}			6.7 ± 1.3
GI Tract	6.7×10^{-1}			6.6 ± 0.1
Carcass	4.9×10^{-2}	6.8×10^{-4}	3.5×10^{-2}	9.2 ± 1.6
Low Activity Strata**				
Soil	1.5×10^{0}			6.0 ± 0.3
Vegetation†	5.1×10^{-1}			5.2 ± 0.2
Small Mammals				
Hide	9.5×10^{-2}			6.3 ± 0.6
GI Tract	1.1×10^{-1}			6.2 ± 0.3
Carcass	2.9×10^{-3}	1.9×10^{-3}	5.7×10^{-3}	7.2 ± 1.2
Perognathus longimembris				
Hide	9.4×10^{0}			3.4 ± 0.7
GI Tract	6.0×10^{-1}			6.2 ± 0.2
Carcass	1.5×10^{-1}	2.1×10^{-3}	1.1×10^{-1}	6.5 ± 0.4
Dipodomys spp.				
Hide	3.2×10^{0}			5.1 ± 1.0
GI Tract	1.0×10^{0}			6.7 ± 0.2
Carcass	3.3×10^{-3}	4.6×10^{-5}	2.4×10^{-3}	5.8 ± 0.7

*>25,000 CPM ^{241}Am based on FIDLER Surveys.

**<25,000 CPM ^{241}Am based on FIDLER Surveys.

†Values calculated from data presented by Romney *et al.*, 1975.

2. Pu uptake evidenced by tissue burdens and concentration ratios may be related to the species of animal considered. In Table 2, *P. longimembris* had significantly higher ($P<.01$) Pu tissue burdens and concentration ratios than *D. microps* even though both are granivores. Data from animals of different trophic categories are presently being analyzed (Table 3).

3. Pu/Am ratios are highly variable in pelts and carcasses of small mammals and in some species are significantly different ($P<.05$) than ratios reported from soil and vegetation samples from their home ranges. Pu/Am ratios in the GI tract, however, showed relatively low variability and were consistent with ratios of soil and vegetation. This may imply that inhalation is the main route of Pu entry, as has been suggested by most other investigations, or it may imply that Pu and/or Am is preferentially absorbed in the GI tract. The much higher variability in Pu/Am ratios in the pelt is consistent with the former hypothesis.

HEMATOLOGICAL STUDIES

Laboratory studies on the effects of Pu on the hemopoietic systems of mammals are numerous and are summarized in Wick (1967) and Durbin (1972, 1973). Finkel and Biskis (1962) reported a lower than normal hemaglobin value in moribund mice after injection of 15.6 μc/kg or more and a depression of peripheral leukocytes with 3.1 μc/kg or more. Sanderson and Jackson (1972) found a similar lowering of white blood cells and lymphocytes in rats with a intra-abdominal injection of 2.5-3.0 Ci Pu. Bone marrow aplasia was observed in rats and hamsters dying within 30 days after injections of 50 μCi/kg or more of Cm and Am. The hematological effects of ^{241}Am were similar to Pu in growing rats (Langham and Carter, 1951) and ^{247}Cf and ^{252}Cf were suggested to be at least equal in toxicity to Pu in beagles (Dougherty, 1972).

To determine if Pu body burdens were related to blood cell reductions in rodents of Area 11, NTS, correlation coefficients were determined for blood cell counts and Pu body burdens. Significant depressions in some leukocyte counts were correlated with elevated Pu carcass burdens.

In Figure 1, total leukocyte counts were plotted against Pu carcass burdens of *D. merriami* and a least squares fit for the power curve plotted. Total leukocyte counts were negatively correlated ($P<.01$) to Pu carcass burdens. In Figure 2, in a similar manner, relative lymphocytes were negatively correlated ($P<.01$) with Pu carcass burdens in *P. longimembris*.

Whereas these results are preliminary, and based on small sample sizes, levels of Pu in carcasses of some rodent species in Area 11, NTS, appear to be related to depressed leukocyte counts on a statistical basis. There is no evidence at this time that these depressions are of a deleterious nature. Additional data from examination of animals with high Pu body burdens are needed. We plan to evaluate blood cell formation in the bone marrow of these animals. By comparing blood cell counts in bone marrow with peripheral blood cell counts, one can

Table 3. ^{239}Pu and Pu/Am Ratios in Granivores and Insectivore-Omnivores From Area 11-C, NTS (values in nCi/g ash) (Moor *et al.*, 1976).

	Low Activity Stratum*				High Activity Stratum**			
	N	^{239}Pu $\bar{X}$ SE	N	Pu/Am $\bar{X}$ SE	N	^{239}Pu $\bar{X}$ SE	N	Pu/Am $\bar{X}$ SE
GRANIVORE								
Pelt	26	0.714 ± 0.267	20	5.90 ± 0.59	16	22.792 ± 9.320	16	5.92 ± 1.78
GI Tract	26	0.155 ± 0.039	26	6.07 ± 0.39	16	3.344 ± 0.950	16	6.59 ± 0.15
Carcass	26	0.008 ± 0.005	20	5.58 ± 0.84	16	0.263 ± 0.137	16	7.17 ± 0.67
INSECTIVORE-OMNIVORE								
Pelt/Skin	11	1.316 ± 0.499	8	7.18 ± 1.75	11	8.911 ± 2.948	10	7.82 ± 1.93
GI Tract	12	1.159 ± 0.530	8	6.75 ± 0.34	11	9.891 ± 4.217	9	6.67 ± 0.26
Carcass	12	0.022 ± 0.012	9	10.84 ± 3.12	11	0.148 ± 0.302	11	12.17 ± 3.67
COMBINED VERTEBRATES								
Pelt/Skin	37	0.893 ± 0.240	28	6.27 ± 0.64	27	17.136 ± 5.732	26	6.66 ± 1.31
GI Tract	38	0.472 ± 0.182	34	6.23 ± 0.31	27	6.0111± 1.869	25	6.59 ± 0.13
Carcass	38	0.013 ± 0.005	29	7.21 ± 1.18	21	0.216 ± 0.083	21	9.20 ± 1.58
SOIL†	23			6.2 ± 0.39	24			6.0 ± 0.30
VEGETATION†	26	2.0 ± 0.40		5.3 ± 0.20	22	8.3 ± 0.30		5.0 ± 0.25

*<25,000 CPM ^{241}Am based on FIDLER Surveys by REECo personnel.
**>25,000 CPM ^{241}Am based on FIDLER Surveys by REECo personnel.
†Data from Romney *et al.*, 1975.

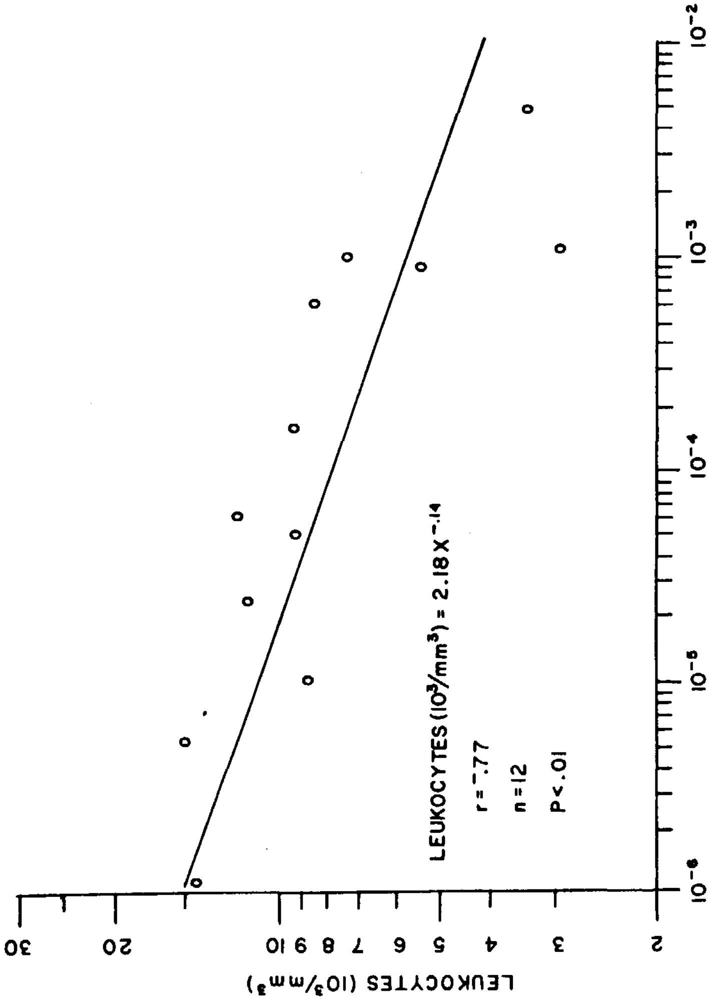

Figure 1. Total leukocyte counts in relation to ^{239}Pu carcass burdens in *D. merriami* from Area 11-C, NTS (Moor *et al.*, 1976).

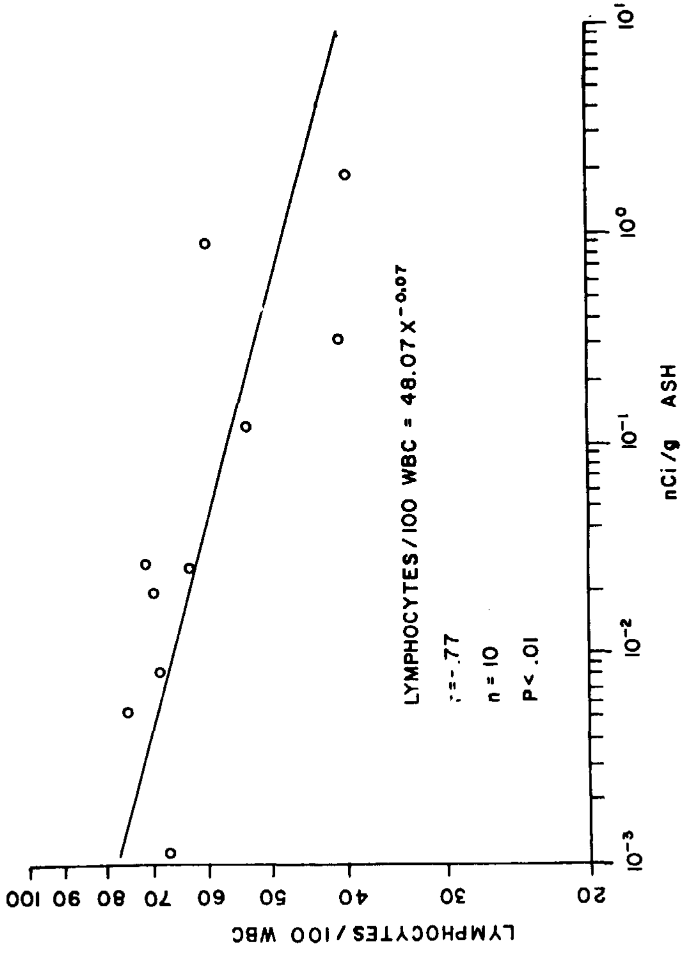

Figure 2. Relative lymphocyte counts in relation to ^{239}Pu carcass burdens in *P. longimembris* from Area 11-C, NTS (Moor *et al.*, 1976).

determine if depressed peripheral counts are caused by a block in the synthesis of certain cells or a reduction in the number of circulating cells.

RESIDENT RODENT POPULATIONS OF AREA 11-C

One particularly difficult problem in evaluating tissue burdens of small vertebrates occupying different trophic levels and inhabiting defined activity strata is the apparent reduced resident rodent populations in the vicinity of GZ.

Table 4 presents data on total number of captures and number of resident rodents encountered in the High and Low activity strata of the inner grid during eighteen months of study in Area 11-C. If we assume random distribution of animals in the grid, an assumption which is probably met only under ideal conditions, an estimate of the number of animals captured in the High Activity Stratum can be made by multiplying the total number of captures (447) by the proportion of the area encompassed by the High Activity Stratum (23.5%). One hundred and five captures are expected using these calculations which are not significantly different than the number actually captured in this stratum (103). All species shown in Table 4 were captured in both High and Low Activity strata. The number of resident animals expected in the High Activity Stratum can be estimated in the same manner. There were eighty-two resident animals in the grid of which nineteen were expected from the High Activity Stratum. Only nine resident animals, however, were recorded from this stratum, a significantly ($P<.01$) lower number than expected. Of the six resident species, only four were found in the High Activity Stratum. In addition, of these resident animals, only one species, *D. merriami*, was represented by more than two individuals. These data show a reduced species richness, species diversity (H') and population density of resident rodents in the High Activity Stratum near GZ and illustrate the problems of collecting adequate samples of most species of rodents.

We do not have an explanation for the reduced number of residents in the High Activity Stratum. Beatley (1976) reported that *D. merriami* replaced *D. microps* as a dominant rodent in some disturbed areas of NTS. Whereas the low number of *D. microps* may be explained by habitat alteration around GZ, it does not explain the absence of resident *A. leucurus* or *O. torridus*. Whereas a reduced number of some species of rodents may be expected when native habitat is altered, experience by the investigators in the Mohave desert suggests that many rodents, particularly *A. leucurus*, are likely to be more numerous in disturbed habitats with sufficient vegetation cover. We do not believe, therefore, that the depauperate rodent fauna of Area 11-C, GZ, can be explained satisfactorily strictly on the basis of mechanical disturbance, as the area is not denuded of vegetation. We are not suggesting at this time that this phenomenon is related directly to Pu or Am levels; however, it is a hypothesis worthy of further investigation.

Table 4. A Comparison of Trapping Success and Residency of Rodents in Relation to Activity Strata of Area 11-C, NTS (Moor *et al.*, 1976).

Species	Number of Captures			Number of Residents		
	Low Activity Stratum	High Activity* Stratum		Low Activity Stratum	High Activity* Stratum	
		Expected	Observed		Expected	Observed
Ammospermophilus leucurus	51	14.1	9	14	3.3	0
Dipodomys merriami	60	21.2	30	6	2.6	5
Dipodomys microps	107	29.1	17	25	6.3	2
Perognathus longimembris	60	18.8	20	12	3.1	1
Neotoma lepida	2	0.9	2	0	0	0
Onychomys torridus	34	10.1	9	12	2.8	0
Peromyscus spp.	30	10.8	16	4	1.2	1
Totals	344	105.0	103	73	19.3	9**
Number of Species	7	7	7	6	6	4
Species Diversity (H')				2.37	2.43	1.66

*Represents 23.5% of trapping grid (>25,000 CPM ^{241}Am).

**Significantly different than expected ($P<.01$); Chi squared.

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REVIEW OF GRAZING STUDIES
ON PLUTONIUM-CONTAMINATED RANGELANDS

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ABSTRACT

Literature is cited that has provided data on tissue actinide concentrations in grazing animals when the actinide dosages resulted from artificial administration or from periodic exposure. Only one long-term study is known where a reproducing beef herd was restricted to a plutonium-contaminated environment. Highlights of this study that are reviewed and discussed include: relationship of ingesta concentrations to food habits; tissue concentration related to length of exposure and level of exposure; and the concentration range in various tissues. Emphasis is given to the gonadal concentration which is approximately 25 times that of muscle and blood. Future study plans are also discussed.

INTRODUCTION

In recent years, there have been several reports published that listed data on actinide levels in the tissues of grazing or browsing animals that periodically ranged over plutonium-contaminated areas or were exposed to worldwide fallout (Fountain, 1961; Brechbill, 1969; Hakonson *et al.*, 1973; Smith and Giles, 1974; Smith and Giles, 1975; Smith and Black, 1975; Smith *et al.*, 1976a; Smith *et al.*, 1976b; and Brown *et al.*, 1976). Other studies have been conducted in which actinides were administered either orally, intravenously, or by inhalation (McClellan *et al.*, 1962; Moskalev *et al.*, 1969; Stanley *et al.*, 1974; Stanley *et al.*, 1975; and Sutton, 1976). However, only one study has been conducted in which reproducing beef cattle were restricted for an extended period of time to a plutonium-contaminated range whose native vegetation supplied all subsistence.

This study, conducted under the auspices of the Nevada Applied Ecology Group, took place within the fenced compounds of the Project 57 site in Area 13 of the Nevada Test Site and extended from May 1973 to April 1976. During this time, tissue samples were collected from 20 cattle, 4 fetuses, 2 goats, and selected wildlife. In addition to the tissue samples collected from the sacrificed animals, ingesta samples were collected from the rumen-fistulated steers (steers with a capped tube inserted into a permanent surgical opening into the stomach) which periodically grazed the most highly contaminated areas.

Details on the experimental design and the analytical data from the beef tissues and rumen ingesta samples collected prior to 1976 have been discussed previously (Smith, 1974; Smith, 1975 ; Smith 1976a; Smith *et al.*, 1976c; and Smith, 1976b). As shown in Table 1, the last cattle grazing the Area 13 range were sacrificed, necropsied, and sampled in January and March of 1976. However, data from these last samples are not yet available, so the following discussion will consist largely of a review of data previously available.

RESULTS AND DISCUSSION

Relationships were noted between the seasonal variations observed in the botanical makeup and the actinide content of rumen ingesta. Grasses were preferred in the late spring and early summer months, while shrubs made up the bulk of the diet during the rest of the year. The plutonium activities measured in rumen contents were higher when *Eurotia lanata* was the preferred shrub species. It is thought that this observed increase in actinide concentration with a concurrent decrease in solubility is related to the retention of soil particles by the involucres of *Eurotia lanata* plants which are in a reproductive phase during the fall.

Results to date from *in vitro* rumen digestion studies indicate increased solubility of the plutonium and americium-241 found in rumen contents from animals grazing the outer compound versus that found in the ingesta of animals from the inner compound (Barth, 1975; Barth, 1976).

No significant lesions relating to plutonium exposure have thus far been found upon necropsy or histopathological examination. Reproduction has been normal. The natural mortality rate was low as only one aged cow died during the 3-year study period. Cause of this death was not determined as the carcass was destroyed by scavengers. Eight study goats were lost to coyote predators.

The highest actinide concentrations in tissues were reported from a cow which grazed the inner compound (the area of highest contamination). The next highest concentrations were found in an animal that had grazed the longest period of time (636 days) in the outer compound.

In general, the plutonium-239 activities measured in tissues on a wet weight basis approximated a log normal distribution and fell within the following categories:

<1	pCi/kg	- muscle, blood serum, blood cells, whole fetus (less than 5 mos)
<5	pCi/kg	- kidney
<10	pCi/kg	- bone, gonads
<20	pCi/kg	- liver
<100	pCi/kg	- lungs
<500	pCi/kg	- tracheobronchial lymph nodes

Those cattle samples that were environmentally contaminated were distributed as follows:

Animal No.	Date of Birth or Entry into Study Area	Sex	Wt. (kg)	Location	Date of Sampling	Duration in Study Area	Age at Sampling	Breed	Remarks
1	May 2, 1973	F	252	Outer	07/09/74	431 days	12 yrs.*	Angus-Hereford	30-day fetus--entire fetus submitted as sample.
2	May 2, 1973	F	409	Inner	10/25/73	176 days	10 yrs.*	Angus-Hereford cross	
3	May 2, 1973	F	432	Outer	10/25/73	176 days	10-11 yrs.*	Angus-Hereford cross	8-month fetus--tissues sampled.
4	May 2, 1973	F	300	Outer	07/09/74	431 days	11 yrs.*	Hereford	Squamous cell carcinoma.
5	May 2, 1973	F	298	Outer	01/29/75	636 days	8-9 yrs.*	Angus-Hereford cross	
6	May 2, 1973	F	325	Outer	07/09/74	431 days	12 yrs.*	Angus-Hereford cross	45-day fetus--entire fetus submitted as sample. Diaphragmatic hernia.
7	May 2, 1973	F	Unk	Inner	04/17/75	730 days*		Angus-Hereford cross	Died in compound early April 1975.
8	May 2, 1973	F	328	Outer	10/25/73	176 days	10 yrs.*	Angus-Hereford cross	
9	May 2, 1973	F	382	Outer	03/31/76	1,064 days	10 yrs.*	Angus	8-month fetus--tissues sampled.
10	Born April 26, 1973. Entered inner compound May 2, 1973.	F	285	Inner	01/28/76	1,001 days	3 yrs.	Angus-Hereford cross	Lived in inner compound entire life. Dam #7.
11	October 20, 1973	F	32	Inner	10/25/73	5 days	5 days	Angus-Hereford	Dam was #2.
12	May 15, 1973	F	93	Outer	10/25/73	163 days	5.5 mos.	Hereford-Angus	Brachycephalic dwarf. Dam #8.
13	August 3, 1973	M	250	Outer	01/29/75	544 days	1.5 yrs.	Angus-Hereford	Dam #5.
14	Born October 7, 1973.	F	405	Outer	01/28/76	843 days	2.25 yrs.	Angus	Dam #6.
15	July 2, 1973	M	311	Outer	01/29/75	576 days	1.5 yrs.	Angus-Hereford	Dam #4.
16	Born August 26, 1973.	M	409	Outer	03/31/76	948 days	2.5 yrs.	Angus	Dam #9.
17						NUMBER NOT USED			
18	Born June 16, 1975	F	184	Inner	01/28/76	226 days	7.5 mos.	Hereford	Dam #10.
19	Born June 20, 1975	M	173	Outer	01/28/76	222 days	7.5 mos.	Angus	Dam #14.
20	Born November 10, 1973.	M	302	Outer	03/31/76	872 days	2.25 yrs.	Angus-Hereford	Dam #1.
30	Unknown	F	205*	Outer	01/16/76		Yearling	Hereford-Charolais cross	Wandered into outer compound in December 1975. Wandered animal found and sampled on Bald Mountain.

*Estimated.

Table 1. Sampling Statistics - Area 13 Beef Animals

<500 pCi/kg - reticulum sediment
<1,000 pCi/kg - rumen fluid
<5,000 pCi/kg - rumen vegetation
<20,000 pCi/kg - skin and hair

As shown in Figure 1 (from Smith *et al.*, 1976c), the plutonium-239/plutonium-238 ratios in the tissues of sacrificed cattle were generally less than the ratio of 42 observed in ingesta from fistulated steers grazing the inner compound of Area 13, which tends to support the hypothesis that plutonium-238 is more readily assimilated. A plutonium-239 retention factor of 0.00056% for bone + muscle + liver was calculated on the basis of ingesta activities (Smith *et al.*, 1976c). This factor was verified by two different statistical approaches by other investigators (Gilbert *et al.*, 1976).

It was found that actinides crossed the placental barrier and were deposited in fetal tissue at nearly the same levels as found in the dam's corresponding tissues.

Actinide concentrations in the gonads (both male and female) were greater than that found in most other soft tissues, e.g., the median plutonium-239 value in the gonads was approximately 25 times greater than that of muscle (Smith, 1976b). As shown in Figures 2 and 3, similar relationships were found in tissues from cattle grazing near the Rocky Flats plant in Colorado (Smith and Black, 1975). While it must be remembered that the data base from both of these herds is small, the data do tend to support the work of others who have noted higher gonadal concentrations in other species (Takizawa, 1973; Krey and Bogen, 1962; Morrow *et al.*, 1967). Other workers have noted insignificant gonadal concentration (Richmond and Thomas, 1974). Perhaps a more definite trend may be apparent when the analytical data from gonads of the 1976 Area 13 cattle become available.

As shown in Table 2, other animals sampled from the study area included two goats, three foxes, one coyote, and a jackrabbit. Data from the tissues of these animals indicate that levels in most of the tissues fell within the same ranges as from the cattle. However, concentrations were higher in the muscle of the goat and in the muscle, liver, and kidney tissues of the predators.

The collection of soil sediments from the ingesta of both fistulated steers and from a sacrificed cow indicated that less than 0.5 kg of soil was ingested daily.

The last animals grazing the Area 13 range were sacrificed, necropsied, and sampled in January and March of 1976. When available, the data from these animals should allow a more definitive comparison of levels found in nearly identical animals (two cow-calf pairs) who spent their entire lives within two contaminated environments of different magnitude. Of intense interest also are data from the gonads of animals (male and female) bred and born within the study area. Additional data will be available on the placenta transfer of actinides. Tissues from the last of the original cows will furnish information as to whether the actinide tissue levels have reached equilibrium within the 3-year grazing period.

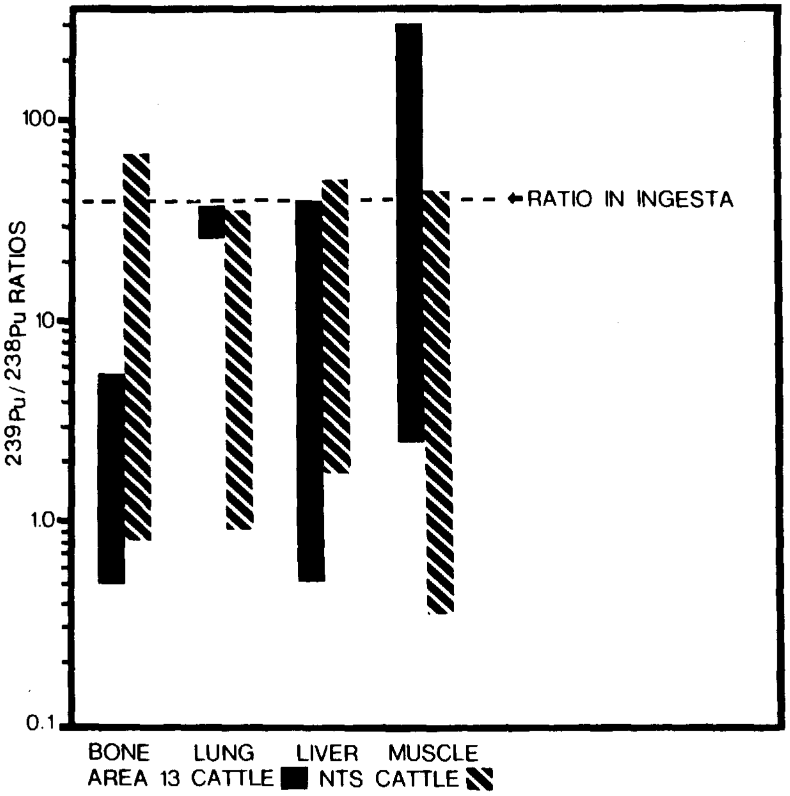

Fig. 1. Ranges of $^{239}Pu/^{238}Pu$ ratio in tissue from Area 13 vs. NTS cattle.

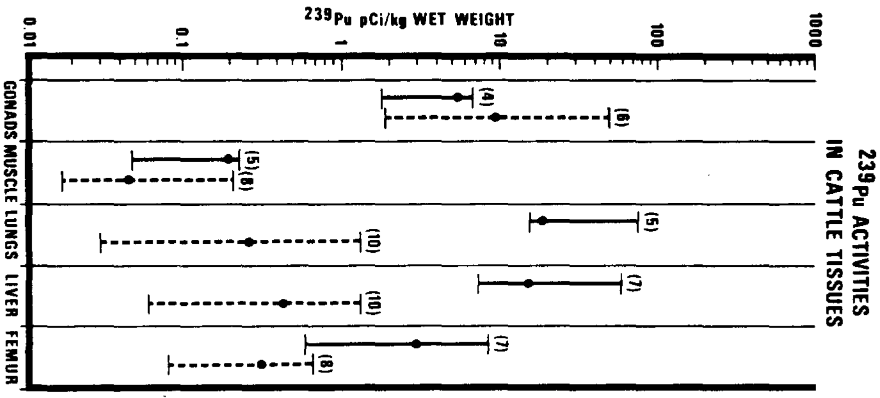

Fig 2. ^{239}Pu activities in cattle tissues.

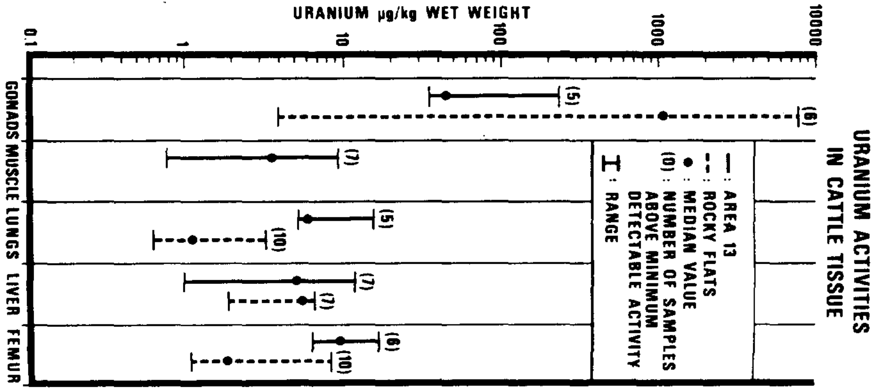

Fig. 3. Uranium activities in cattle tissue.

Animal No.	Date of Birth or Entry into Study Area	* Sex	Wt. (kg)	Location	Date of Sampling	Duration in Study Area	Age at Sampling	Breed	Remarks
Goat #1	May 6, 1973	F	27	Inner	08/07/73	93 days	1 yr.	Mixed	
Goat #2	May 6, 1973	F	32	Inner	10/25/73	172 days	3 yrs.	Mixed	
Coyote 1	Unknown	M	12	Area 13	10/09/74	Unknown	Adult		Suspected goat killer confirmed by stomach contents.
Fox 1	Unknown	F	1.8	Area 13	05/20/74	Unknown	Adult		South side of compound.
Fox 2	Unknown	M	2	Area 13	05/20/74	Unknown	Adult		South side of compound.
Fox 3	Unknown	M	1	Area 13	05/20/74	Unknown	6 mos.		South side of compound.
Rabbit #1	Unknown	M	1	Area 13	05/20/74	Unknown	Adult	Jackrabbit	Hunter kill NW corner of outer compound.

Table 2. Sampling Statistics - Other Area 13 Animals

FUTURE PLANS

Recommendations for future studies on nuclear sites are as follows:

1. Pick two nuclear sites with different plant communities (one similar to Area 13) to determine (a) if uptake is affected by type of preferred forage, and (b) if actinides produced in nuclear tests are similar metabolically to those produced by the safety tests. Use rumen-fistulated steers to determine food habits.

2. Use two cattle populations--one being similar to the Area 13 cattle, which would be sacrificed on the same time schedule as used for the Area 13 study, the other would be a sufficient number of young animals to allow paired annual sacrifices so length of time for equilibrium could be determined.

3. The goat portion of the study should be repeated. Predator control would be essential. A combination of fences, noisemakers, and other control measures may suffice.

The need for other studies will probably become apparent after all data are received and analyzed.

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APPLICATION OF THE ARTIFICIAL RUMEN AND SIMULATED BOVINE GASTROINTESTINAL FLUIDS PROCEDURE IN THE STUDY OF THE BIOAVAILABILITY OF TRANSURANICS

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ABSTRACT

An artificial rumen and simulated abomasal and intestinal fluids procedure was used to study the alimentary availability of plutonium-238. When plutonium-238 was administered as plutonium nitrate, 10.1% remained soluble following the artificial rumen incubation period and 15.3% following the abomasal period; 30.1% and 32.7% remained soluble when the fluid was held at pH 4 and 5, respectively, during the duodenal phase. The solubility increased to 60.1% following the addition of bile and enzymes with adjustment of the pH to 6 to simulate the jejunum. The increase in plutonium solubility in the simulated jejunal fluid was found to be due to the presence of bile. Plutonium administered as a citrate-buffered plutonium solution was 9.0% soluble following the rumen incubation period, 13.1% following the abomasal period, and 22.5% and 24.8% when held at pH 4 and 5, respectively, in the duodenal phase. The solubility increased to 59.6% following the addition of bile and enzymes with adjustment of the pH to 6. Plutonium administered as 0.06-μm plutonium dioxide spheres was 1.5% soluble following the rumen incubation period, 2.3% following the abomasal period, and 3.6% and 3.9% when held at pH 4 and 5, respectively, in the duodenal phase. Solubility increased to 7.4% following the addition of bile and enzymes with adjustment of the pH to 6.

Rumen contents of cattle grazing on plutonium-contaminated desert vegetation at the Nevada Test Site, Area 13, were collected quarterly and incubated in simulated bovine gastrointestinal fluids to study the alimentary availability of field-deposited plutonium-238, plutonium-239, and americium-241. Results to date indicate that the highest concentrations of plutonium and americium in the rumen contents occurred during the late summer or fall. This coincides with the reproductive or "woolly" phase of *Eurotia lanata* (winterfat), which made up a considerable portion of the vegetation eaten. The solubilities of plutonium and americium were highly variable according to the season and to the predominant vegetation ingested. In most cases plutonium-238 was more soluble than plutonium-239. The solubility of americium-241 was similar to that of plutonium-239.

The application of *in vitro* solubility to predict tissue retention and milk secretion is discussed.

INTRODUCTION

The artificial rumen and simulated abomasal and intestinal fluids procedure was developed and used in conjunction with ruminant metabolism trials as an *in vitro* screening device to evaluate radionuclide binding materials for their possible use as feed additives to reduce the levels of radioactivity in milk and meat (Barth and Bruckner, 1969a,b; Barth *et al.*, 1969). The radionuclides of interest were mainly radiostrontium and radiocesium. The effects of these binding materials on the availability of essential minerals were also studied. At this Laboratory, this procedure has been used to study the solubility and factors affecting the availability of purified forms, field-released forms, and fallout forms of iodine-131 and other radionuclides.

A study by Barth and Mullen (1972) strongly indicated a correlation between radionuclide solubility in the *in vitro* procedure discussed in this paper and radionuclide availability for absorption in ruminants. The artificial rumen and simulated abomasal and intestinal fluids procedure can be used to predict, with reasonable accuracy, the tissue retention and secretion of various chemical and physical forms of radionuclides deposited on edible vegetation. However, the solubility determined *in vitro*, and either the tissue retention or secretion determined *in vivo*, of a purified form of the radionuclide must be known.

The artificial rumen and simulated abomasal and intestinal fluids procedure is currently being used in the Nevada Applied Ecology Group (NAEG) plutonium program to study the solubility and behavior of purified and field forms of transuranics in the ruminant digestive tract. In the case of purified or laboratory forms, information is obtained concerning the effects of ruminal-gastrointestinal conditions such as pH of the medium, enzymes, and bile. The solubility of a purified form, in conjunction with metabolism trial data, may be used to predict milk secretion and tissue retention of a field or fallout form.

Data concerning the effects of season, type of vegetation, and grazing conditions on the biological availability of selected transuranics present are obtained from trials using field or fallout forms. Studies utilizing the artificial rumen and simulated abomasal and intestinal fluids procedure in the NAEG plutonium program are described and the application of the available data is discussed in this paper.

BRIEF GENERAL PROCEDURE

Rumen juice was collected from a fistulated steer, added to a basal medium containing nutrients and saturated with carbon dioxide; the pH was adjusted to 6.5 with sodium carbonate. Erlenmeyer digestion flasks were inoculated with the rumen juice preparation and a radioactive tracer was added. Incubation was allowed to proceed with carbonation for about 24 hours at 39.5°C. The artificial rumen was converted to simulated abomasal fluid by the addition of hydrochloric acid and pepsin, and adjustment of the pH to 3. Abomasal incubation was allowed to proceed for 3 hours.

The simulated abomasum was then converted to simulate the intestine. The duodenum was simulated by adjustment of the pH to 4 and then 5. Simulated duodenal fluid was held at each pH only long enough to remove samples. The early jejunum was simulated by the addition of bile, pancreatin, trypsin, and erypsin, and adjustment of the pH to 6. Incubation was allowed to proceed for 2 hours. The lower small intestine was simulated by adjusting the pH to 7.5, followed by a 2-hour incubation period.

Samples were removed from the digestion flasks during all phases of the digestion period and centrifuged, and the supernatant fluid was collected for the analyses of soluble transuranics.

This procedure is modified as necessary to meet the objectives of various NAEG program studies. A more complete description of the general laboratory procedure is given by Barth and Mullen (1972).

RESULTS AND DISCUSSION

Since these studies are in progress, all interpretations are subject to reconsideration as additional data become available.

Purified Plutonium Studies

In these trials the solubilities of plutonium-238 nitrate, plutonium-238 citrate, and plutonium-238 dioxide were studied in an artificial rumen and simulated abomasal and intestinal fluids procedure (Barth and Mullen, 1974). The *in vitro* procedure used was similar to that described under the general procedure.

When plutonium was administered as plutonium nitrate solution to rumen juice, 13.8% remained soluble shortly after administration (about 15 minutes), 10.1% remained soluble following the artificial rumen incubation period, 15.3% following the abomasal period, and 30.1% and 32.7% when held at pH 4 and 5, respectively, in the duodenal phase. The solubility increased to 60.1% following the addition of bile and enzymes and adjustment of the pH to 6.

When plutonium was administered as plutonium citrate (citrate buffered plutonium solution), 20.6% remained soluble shortly after administration, while 9% remained soluble following the artificial rumen incubation period, 13.1% following the abomasal period, and 22.5% and 24.8% when held at pH 4 and 5 respectively, in the duodenal phase. This increased to 59.6% following the addition of bile and enzymes and adjustment of the pH to 6.

When plutonium was administered as plutonium dioxide spheres having a count median diameter of 0.06 μm, 5.2% became soluble shortly after administration, 1.5% was soluble following the artificial rumen incubation period, 2.3% following the abomasal period, and 3.5% and 3.9% when held at pH 4 and 5, respectively, in the duodenal phase. This increased to 7.4% following the addition of bile and enzymes and the adjustment of the pH to 6.

The sharp rise in soluble plutonium observed for all forms following the addition of bile and enzymes and adjustment of the pH to 6 was found to be due to the presence of bile rather than to enzymes or change in pH.

An example is shown in Table 1 of the application of the *in vitro* solubility of a purified form of a radionuclide in conjunction with metabolism trial results to predict tissue retention of another form of this radionuclide. In this case a comparison is made between the predicted liver and bone retention of plutonium-238 dioxide, calculated from its *in vitro* solubility and relationship to the *in vitro* solubility and actual tissue retention of plutonium-238 citrate, and the actual tissue retention of plutonium-238 dioxide determined by a metabolism trial. The actual tissue retention data are from Stanley *et al.* (1975).

The *in vitro* solubility during the duodenal phase was used to predict plutonium tissue retention since this phase is presently believed to be the most important aspect of plutonium absorption. The values for pH 4 and pH 5 were combined and averaged for both plutonium-238 citrate and plutonium-238 dioxide. Then, a ratio of the average solubility of plutonium-238 citrate to plutonium-238 dioxide was calculated (plutonium-238 citrate/plutonium-238 dioxide = 6.39). The predicted liver retention of plutonium-238 from plutonium-238 dioxide was calculated by dividing the actual retention of plutonium-238 from plutonium-238 citrate by the *in vitro* solubility ratio ($3.5 \times 10^{-3}\%/6.39 = 5.5 \times 10^{-4}\%$).

The plutonium-238 citrate and plutonium-238 dioxide metabolism trials were not done in an identical manner. In the case of plutonium-238 citrate the cows were sacrificed 93 days following a single dose, while in the case of plutonium-238 dioxide the cows were sacrificed 73 days following the last multiple dose (Stanley *et al.*, 1975). However, the predicted retention values of plutonium-238, from plutonium-238 dioxide, by liver and bone are in reasonable agreement with the actual retention values.

This prediction procedure is based on the assumption that once the plutonium becomes soluble in ruminants, the absorption of soluble plutonium by the animal and subsequent deposition and retention by tissues will be similar from both forms of plutonium administered. Table 2 illustrates this. The portion of the dose administered which is actually soluble in the duodenum and therefore available for absorption, as calculated from *in vitro* solubility, is referred to as the "effective dose." The percentages

TABLE 1. PREDICTED BOVINE TISSUE RETENTION COMPARED TO ACTUAL RETENTION OF AN ORAL DOSE OF PLUTONIUM-238 DIOXIDE

In Vitro Solubility Ratio* Plutonium-238 Citrate / Plutonium-238 Dioxide	Tissue	Actual Retention of Plutonium-238 Citrate** (%)	Predicted Retention of Plutonium-238 Dioxide (%)	Actual Retention of Plutonium-238 Dioxide** (%)
6.39	Liver	3.5×10^{-3}	5.5×10^{-4}	6.0×10^{-4}
	Bone	2.4×10^{-2}	3.8×10^{-3}	3.2×10^{-3}

*Explained in text

**Stanley *et al.* (1975)

TABLE 2. COMPARISON OF THE THEORETICALLY SOLUBLE PLUTONIUM-238 FROM PLUTONIUM-238 CITRATE AND PLUTONIUM-238 DIOXIDE RETAINED BY TISSUES

Plutonium-238 Form	In Vitro Solubility* (%)	Dose Administered to Cow** (mCi)	Effective Dose to Cow (mCi)	Liver Retention+ (mCi)	Effective Dose Retained by Liver (%)	Bone Retention+ (mCi)	Effective Dose Retained By Bone (%)
Citrate	23.6	3.0	0.708	10.5×10^{-5}	14.8×10^{-3}	0.72×10^{-3}	10.2×10^{-2}
Dioxide	3.7	19.0	0.703	11.4×10^{-5}	16.2×10^{-3}	0.61×10^{-3}	8.7×10^{-2}

*Explained in text.

**Stanley *et al.* (1975)

+Calculated from data of Stanley *et al.* (1975)

of the effective dose of plutonium-238 retained by the liver and bone are in close agreement for both forms of plutonium-238.

An alternate procedure for the prediction of tissue retention is to calculate the product of the known percentage of the effective dose retained by the tissue times the *in vitro* solubility of the unknown (liver: $14.8 \times 10^{-3}\%$ times $3.7\% = 5.5 \times 10^{-4}\%$; bone: $10.2 \times 10^{-2}\%$ times $3.7\% = 3.8 \times 10^{-3}\%$), (Table 2).

These prediction procedures are valid only when tracer or micro levels of a radionuclide are administered or ingested and the tissue uptake or milk secretion is proportional to the effective dose present.

Field-Ingested Transuranics Study

In these trials the solubility of plutonium-238, -239, and americium-241 from the rumen contents of cattle grazing on plutonium contaminated desert vegetation was determined in *in vitro* bovine gastrointestinal fluids. This study, which is still in progress, was initiated in November 1973 (Barth, 1975; Barth, in press). Americium-241 was first included during the November 5, 1974, trial.

Rumen-fistulated cattle were allowed to graze periodically in the inner enclosure and nonfistulated cattle were allowed to graze continually in the outer enclosure of Area 13 of the Nevada Test Site (Smith, 1975). The inner enclosure directly surrounds the ground zero of a high-explosive detonation of an atomic device during a safety test. The outer enclosure is less heavily contaminated and most of this area is located a greater distance downwind from ground zero than the inner enclosure.

Samples of whole rumen contents were collected from the fistulated cattle following a 48-hour grazing period. Samples were collected from the nonfistulated cattle at the time of slaughter.

Samples of whole rumen contents were added directly to simulated abomasal fluid. The abomasal and intestinal digestion procedures were similar to those described earlier. The entire contents of each flask were separated into solid and liquid fractions by preliminary filtration and centrifugation. The entire solid and liquid fractions were analyzed for plutonium-238, plutonium-239, and americium-241.

The behavior of field-ingested plutonium in these fluids was, for the most part, qualitatively similar to the behavior of the purified forms of plutonium-238 used in the studies described earlier by Barth and Mullen (1974). Generally, there was a rise in plutonium-238, -239, and americium-241 solubility when the pH was increased to 4.5 in order to simulate the duodenum. This initial increase in solubility was followed by an additional increase in plutonium solubility during the jejunal incubation period following the addition of bile and enzymes and adjustment of the pH to 6. In the case of americium-241, this additional increase was not consistent. The quantitative behavior of field-ingested plutonium differed considerably from that of purified plutonium-238 dioxide reported by Barth and Mullen (1974).

For brevity, the *in vitro* solubility data for field-ingested plutonium-238, -239, and americium-241 are presented for only the duodenal phase in Table 3, since this phase is currently believed to be the most important in regard to plutonium absorption (excerpted from Barth, 1975 and Barth, in press). In the simulated duodenal fluid, plutonium-238 was consistently more soluble than plutonium-239. In the other simulated fluids (data not shown), this was usually the case. The solubility of americium-241 in these fluids was found to be similar to that of plutonium-239.

In making comparisons of radionuclide concentrations in rumen contents between trials, only very large differences are meaningful since the animals may graze at will throughout the enclosures. Also, types of plants ingested and the resuspension of radionuclides may vary. However, in the available data, a trend concerning peak activity of plutonium in the rumen contents appears to be developing. Results to date (Table 3) indicate that the highest concentrations of plutonium in the rumen contents of cattle grazing at Area 13 have occurred during the late summer or fall, followed by reduced concentrations during the winter or early summer. An exception to this occured during the May 29, 1975, trial.

Analyses of the vegetal composition of the rumen contents indicated that *Eurotia lanata* (winterfat) made up a considerable portion of the vegetation ingested, and was frequently the predominant species. Observations were not made of the field conditions of *Eurotia lanata* during these trials. However, visual observations were made of the field conditions of *Eurotia lanata* growing in Area 13 during the summer and fall of 1975 and the winter of 1975/1976 (data not included in this report). The reproductive phase of *Eurotia lanata* characterized by the long-haired fruiting involucres, appeared during October 1975. During January 1976, some fruiting involucres remained. The onset of the reproductive phase will vary with the climatic conditions. This suggests that the involucres could greatly enhance the entrapment of resuspended particles, resulting in higher concentrations of plutonium-238, -239, and americium-241 in the rumen contents.

During the May 17, 1974, and the July 10, 1974, trials, a large increase in plutonium solubility, accompanied by a marked reduction in plutonium concentration of the rumen contents, was observed concurrently with a reduction in the intake of *Eurotia lanata* and an increase in the intake of *Oryzopsis hymenoides* (Indian rice grass) or *Sitanion jubatum* (squirrel tail grass).

Barth and Mullen (1974) reported average minimum and maximum solubilities of 1.5% and 8.4% for plutonium-238 dioxide in an artificial rumen and simulated abomasal and intestinal fluids procedure. However, during the May 17, 1974, and July 10, 1974, trials, the solubility of plutonium reached 90% and above. Also, during the January 29, 1975, trial, plutonium-238 -239, and americium-241 solubilities of over 62% were observed during the lower intestine digestion phase (data not shown). This suggests the possibility that during these specific trials most of the ingested plutonium was in a form other than plutonium dioxide and that most of this plutonium represented the low level internally incorporated in plant tissue.

TABLE 3. SOLUBILITY OF FIELD INGESTED PLUTONIUM AND AMERICIUM-241 IN SIMULATED BOVINE DUODENAL FLUID - pH 4.5*

Date	Concentration In Rumen Contents, pCi/kg			Percent Soluble		
	^{238}Pu	^{239}Pu	^{241}Am	^{238}Pu	^{239}Pu	^{241}Am
Nov. 6, 1973	236	8539	-	0.49	0.29	-
Feb. 19, 1974	158	3869	-	13.5	0.76	-
May 17, 1974	14	24	-	95.1	44.5	-
July 10, 1974**	244	443	-	96.6	90.0	-
Aug. 8, 1974	1742	60994	-	0.96	0.14	-
Nov. 5, 1974	729	28024	5553	1.3	0.24	0.43
Jan. 19, 1975	62	2240	356	2.0	0.69	1.1
Jan. 21, 1975	144	6738	943	†	0.15	0.44
Jan. 29, 1975**	37	1448	213	3.2	0.64	0.75
Mar. 12, 1975	416	11378	1629	†	0.88	1.4
May 29, 1975	589	20202	2814	0.22	0.13	0.10

*Excerpted from Barth (1975) and Barth (in press).

**Outer enclosure.

†Below ^{238}Pu detection limit.

During the July 10, 1974, and January 29, 1975, trials, the rumen contents were collected at the time of slaughter from nonfistulated cattle grazing in the outer enclosure. Factors which were present in the outer enclosure, and which may have contributed to the marked increase in plutonium-238, -239 and americium-241 solubility, were the greater distance of the grazing area from ground zero and a higher grazing intensity. Romney *et al.* (1975) stated that, as a general rule, the mean fallout particle size decreases with increasing distance downwind from the point of detonation. Hence, a reduced mean particle size is expected in the outer enclosure which would, in turn, result in an increase in plutonium and americium dissolution.

During this study, the outer enclosure was more intensely grazed than the inner enclosure. This allowed for greater regrowth of the edible vegetation in the inner enclosure, while in the outer enclosure it was continually eaten back. It appeared by visual observation that there were fewer of the fruiting involucres on the *Eurotia lanata* in the outer enclosure. This suggests a reduced plant surface entrapment of resuspended particulate material, thereby increasing the proportion of plutonium-238 -239, and americium-241 presumably internally incorporated into plant tissue. Under these experimental conditions the effects of distance from ground zero and grazing intensity cannot be differentiated.

Examples are shown in Table 4 of the applications of *in vitro* data in conjunction with metabolism study data to predict the tissue retention and milk secretion of field-deposited plutonium. Ratios of plutonium solubility in the simulated duodenal fluid of plutonium-238 dioxide to field-ingested plutonium-238 were calculated for each rumen contents collection (Nov. 6, 1973: 3.7%/0.49% = 7.55). The predicted tissue retention or milk secretion was calculated by dividing the actual tissue retention or milk secretion of plutonium-238 from plutonium-238 dioxide, as determined in the metabolism study of Stanley *et al.* (1974 and 1975), by the *in vitro* solubility ratio (liver: $6 \times 10^{-4}\%/7.55 = 0.79 \times 10^{-4}\%$; milk: $2 \times 10^{-5}\%/7.55 = 0.26 \times 10^{-5}\%$).

In order to predict the metabolic behavior of the field-ingested plutonium, the soluble fraction must be absorbed, retained, and secreted in a manner similar to the effective dose administered during the metabolism study (Stanley *et al.*, 1974 and 1975) on which the prediction is based.

As previously mentioned, during the May 17, 1974, and July 10, 1974, trials plutonium solubilities reached 90% and above, which is a marked departure from that of other field-ingested plutonium trials. In addition, Barth and Mullen (1974) reported average simulated duodenal fluid solubilities of 31.4% 23.6%, and 3.7% for plutonium-238 nitrate, plutonium-238 citrate, and plutonium-238 dioxide, respectively. This suggests the possibility that the chemical nature of most of the plutonium ingested during these two field-ingested plutonium trials was dissimilar to that of the purified plutonium-238 forms employed in the *in vitro* and metabolism studies.

Since the field and soluble forms of plutonium ingested during the May 17, 1974, and July 10, 1974, trials are unknown at present, it is not certain whether metabolism trial results of plutonium-238 dioxide are applicable in these

TABLE 4. EXAMPLES OF PREDICTED BOVINE TISSUE RETENTION AND MILK SECRETION OF FIELD-INGESTED PLUTONIUM-238

Collection Date	In Vitro Solubility Ratio	Liver* (% of Dose)	Bone* (% of Dose)	Milk** (% of Dose)
Nov. 6, 1973	7.55	0.79×10^{-4}	0.43×10^{-3}	0.26×10^{-5}
July 10, 1974	0.038	1.58×10^{-2}†	8.42×10^{-2}†	5.26×10^{-4}†
Aug. 8, 1974	3.85	1.56×10^{-4}	0.83×10^{-3}	0.52×10^{-5}
Jan. 19, 1975	1.85	3.24×10^{-4}	1.73×10^{-3}	1.08×10^{-5}

*Based on plutonium-238 dioxide metabolism data of Stanley *et al.* (1975). (Nineteen day multiple dose, 73 days post treatment).

**Based on plutonium-238 dioxide metabolism data of Stanley *et al.* (1974). (Collection period: through nineteen day multiple dose, 7 days post treatment).

†Predictions for July 10, 1974 are valid only if the soluble portion of the field-ingested plutonium-238 is metabolized in a manner similar to that of the effective dose administered during the plutonium-238 metabolism study (Stanley *et al.* 1974 and 1975). See discussion in text.

specific cases. The soluble plutonium during the May 17, 1974, and July 10, 1974, trials could possibly have a considerably greater efficiency of absorption than the soluble plutonium from the dioxide form, in which case the predicted tissue retentions and milk secretion shown in Table 4 for July 10, 1974, would be minimal.

Predictions of tissue retention and milk secretion were not calculated for field-ingested plutonium-239 because the *in vitro* purified plutonium studies and the metabolism studies employed plutonium-238 only and there appear to be possible differences in the availability of plutonium-238 and plutonium-239 due to factors other than particulate solubility. Hanson (1974) states that another physical process that appears to emerge from several studies is the changing availability of plutonium-238. This is indicated by a changing plutonium-238/plutonium-239 ratio in soils, vegetation, and animal components of the Trinity-site ecosystem from 0.05 (in soils) to 0.10 (in plants) to 1.0 (in mammals), respectively, as we go up the food chain. Both *in vitro* and *in vivo* studies are necessary to compare the *in vitro* solubility of purified forms of plutonium-238 dioxide and plutonium-239 dioxide, and to compare the metabolism of the effective dose of each.

So that americium-241 tissue retention and milk secretion predictions can be made from the available data from the field-ingestion study, both *in vitro* and metabolism trials of purified americium-241 are necessary.

An examination of the data presented in Table 3 indicates a very wide variation throughout the year in both plutonium and americium ingested and their availability for absorption. In evaluating soil-plant-animal relationships at Area 13, the data from any single trial or season, alone, could be very misleading. Variations between years have not been assessed.

An important characteristic of the field-ingested transuranics study is the specificity of the data in regard to the field conditions under which the rumen contents were collected. Some of these varying field conditions are season, predominant species of vegetation ingested, stage of growth of vegetation ingested, and distance of grazing area from the point of detonation.

In the case of Area 13, NTS, *Eurotia lanata* frequently made up a considerable portion of the vegetation ingested. There were drastic seasonal variations in the concentrations of plutonium in the rumen contents coinciding with the reproductive or "woolly" stage of *Eurotia lanata* in which its particulate collection efficiency is presumed to be high.

Due to the specificity of the data, results obtained from these studies in Area 13 may be applied to other areas only with reservations. The field conditions mentioned above are factors which should be taken into consideration.

The results indicate the importance of conducting trials during field studies throughout the year so that all field conditions affecting radionuclide collection efficiency of the vegetation and radionuclide availability for absorption by livestock will be covered.

FUTURE PLANS

A study is necessary to compare the *in vitro* solubility of purified forms of plutonium-238 dioxide and plutonium-239 dioxide. Other studies are planned to determine the *in vitro* behavior of americium-241 and curium-243.

Future soil microorganisms-artificial rumen joint studies have been designed to compare the biological availability of biologically incorporated plutonium with nonbiological plutonium, and to determine the effects of soil microbiological activity on the solubility of plutonium-238, plutonium-239, and americium-241 present in contaminated soil.

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TRANSPORT OF PLUTONIUM VIA FOOD PRODUCTS OF ANIMAL ORIGIN

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ABSTRACT

Various plutonium studies have been conducted using domestic animals in an attempt to determine what fraction of a quantified exposure would reach the edible animal products. Following either intravenous or oral plutonium doses to selected livestock species, milk, liver, eggs and skeletal muscle have been considered as critical deposition sites or materials almost irrespective of resulting nuclide concentrations. Milk and eggs received primary attention as their production represents an efficient way of converting dietary crude protein and energy into edible substances. Potential problems concerning the biological availability of plutonium from *in vivo* labeled food materials and the hazards associated with recycling carcass residues have been considered.

INTRODUCTION

It is anticipated that with the increased use of nuclear power systems plutonium will be produced in greater quantities and may present a major health and ecosystem hazard. The long physical and biological half-life and high relative toxicity have dictated that considerable effort be devoted to quantifying plutonium transport through the various trophic levels.

A relatively small number of plant and animal species contribute the majority of food products for the human population. In studies designed to assess potential nuclide transport via the ingestion of contaminated food, research efforts are directed toward those species which are of the greatest nutritive benefit to man. Dairy cows, chickens, beef cattle, goats and pigs have all been used as research animals at the Environmental Monitoring and Support Laboratory-Las Vegas (EMSL-LV) in order to investigate the biological transport of various radionuclides. Domestic animals are currently being utilized at EMSL-LV in a series of continuing experiments to assess the transport characteristics of transuranic elements.

Plutonium studies outlined in this paper are somewhat different from many transuranic metabolism projects in that, almost irrespective of nuclide concentration, the most critical materials are milk, liver, eggs and skeletal muscle. Milk and eggs are especially important materials since their production represents an efficient way of converting dietary crude protein and energy into edible products. Conversion efficiencies are probably of even more significance for future generations which may not have the same agricultural operations that Americans enjoy today.

Once nuclide transport via edible animal products has been confirmed, some further and perhaps more subtle problems become evident. Two such problems are recycling and biological availability. In beef cattle, approximately 44 percent of the live animal weight is not edible, but substances such as bone meal and blood are occasionally used as a protein concentrate for animal feed. Another recycling problem is introduced through the practice of using poultry and cattle manures in some ruminant diets. Recycling problems may be of marginal importance today since grain is usually used as a feed supplement, but they will probably become more important in the future.

The second major question concerns the biological availability of plutonium once it has been incorporated into an edible animal product. Gastrointestinal uptake of plutonium received considerable attention in several early works. In a metabolism study on rats, Scott *et al.*, (1948) observed that the average value for gastrointestinal absorption of plutonium using three different valence states was approximately 7.0×10^{-3} percent. Comparative studies on the intestinal uptake of plutonium nitrate revealed no significant differences in total absorption between the rat and pig (Weeks *et al.*, 1956). These results were quantitatively similar to those obtained in a rather extensive report by Katz *et al.*, (1955) which, following a chronic oral plutonium treatment to rats, presented the mean gastrointestinal absorption and retention value at 3.0×10^{-3} percent of the administered dose. However, animal age, nutritional status and the relative nuclide availability from *in vivo* plutonium-labeled food have, in some instances, affected the amount of plutonium absorbed.

Ballou (1958) reported that plutonium absorption in day-old rats was 85 times that in the adults and that the amount absorbed dropped abruptly in 21-day-old rats to near adult levels. While investigating the effects of plutonium on mice treated *in utero*, Finkel (1947) discussed the relative concentrations transported across the placenta or through the milk following parturition. Further studies (Finkel and Kisieleski, 1976) have suggested that the gastrointestinal absorption of plutonium was increased (3.2 percent of dose retained in 18-day-old rats) when milk labeled *in vivo* with plutonium was administered. Although a fourfold increase in plutonium uptake was noted in iron-deficient mice (Ragan, 1975), iron deficiency would probably not be prevalent at the domestic animal level. Iron requirements of the laying hen, for example, are large in proportion to maintenance requirements, but most domestic animals receive rations of high nutritive value. However, varying degrees of iron deficiency and irregular dietary regimes do exist in the human population and the relative hazards of plutonium-labeled food might be affected by an individual's nutritional status.

This paper will provide a brief overview of some recent plutonium studies using domestic animals and will attempt to direct attention toward selected areas of the food-production cycle where further investigations might prove beneficial. The early experiments of McClellan *et al.*, (1962) and Sansom (1964) have been referenced in previous reports and a comprehensive review on plutonium in biological systems, ranging from large biomes to molecular transformations within individual cells, has already been prepared by Mullen and Mosley (1976).

METHODS AND MATERIALS

Plutonium metabolism experiments at this Laboratory have used Leghorn hens, Toggenberg goats and Holstein cows as research animals. Hereford cattle are used in a separate effort for grazing studies conducted on a contaminated range at the Nevada Test Site. The largest number of individual metabolism studies used dairy cows. While no metabolism studies were done using beef cattle, it should be remembered that a significant percentage of beef comes from cows and bulls discarded from dairy herds. Furthermore, among dairy breeds, Holsteins produce the best carcass for beef purposes.

A fundamental objective of these metabolism studies was to determine what fraction of a quantified exposure would reach the edible animal products. Specific collection and analytical procedures as well as more detailed experimental objectives have been discussed previously (Potter *et al.*, 1971; Stanley *et al.*, 1974; Sutton *et al.*, 1976b; Mullen *et al.*, 1976). Basically samples collected during the studies were analyzed for plutonium-238, based on the 17 keV x-ray from the plutonium isotope. The samples were counted using a phoswich detector containing a thin NaI scintillator backed by a thick CsI scintillator. Overall measurement error was assessed by considering potential uncertainties in the sampling and analytical scheme (Sutton *et al.*, 1976b; Mullen *et al.*, 1976). Those samples found to contain low plutonium concentrations were subsequently prepared for analysis by other laboratories which employed more sensitive assay techniques (ashing, extraction, electroplating and counting with an alpha spectrometer).

RESULTS AND DISCUSSION

Summary results from previous studies are presented for dairy cows (Tables I and II), goats (Table III) and chickens (Table V). As noted in Table I, total plutonium transport to bovine milk following oral exposure was not great and, on a percentage-of-oral-dose basis, was observed to be 2×10^{-4} and 2×10^{-5} following citrate-buffered plutonium nitrate and plutonium dioxide treatments, respectively (Stanley *et al.*, 1974). In some cases, total muscle activity closely approximated the total liver activity but was, of course, based on a much greater mass of tissue.

TABLE I

TRANSPORT OF PLUTONIUM-238 TO EDIBLE BOVINE PRODUCTS FOLLOWING INGESTION OF EITHER CITRATE-BUFFERED PLUTONIUM NITRATE OR PLUTONIUM DIOXIDE

Plutonium Dose	No. of Animals	% of Dose Secreted in Milk (Cumulative)	% of Dose/ℓ of Milk at Peak Concentration	Time of Sacrifice	% of Dose in Liver	% of Dose/g of Liver	% of Dose in Muscle (Skeletal)	% of Dose/g of Muscle	Approximate % of Dose Retained in Carcass Residue (Without Muscle and Liver)
Citrate Buffered Plutonium 3mCi/animal acute dose	4	2.0×10^{-4}	4.9×10^{-6}	2 animals sacrificed 93 days post-treatment	3.5×10^{-3}	3.7×10^{-7}	2.0×10^{-3}	6.7×10^{-9}	2.6×10^{-2}
Plutonium Dioxide 1mCi/animal /day for 19 consecutive days	4	2.0×10^{-5}	2.7×10^{-7}	2 animals sacrificed 42 days post-treatment	5.3×10^{-4}	5.3×10^{-8}	8.7×10^{-5}	3.3×10^{-10}	1.4×10^{-3}
				2 animals sacrificed 73 days post-treatment	6.2×10^{-4}	5.8×10^{-8}	1.3×10^{-4}	3.4×10^{-10}	1.6×10^{-3}

TABLE II

TRANSPORT OF PLUTONIUM-238 TO EDIBLE BOVINE PRODUCTS FOLLOWING A SINGLE INTRAVENOUS INJECTION OF APPROXIMATELY 16 mCi CITRATE-BUFFERED PLUTONIUM NITRATE

Time of Sacrifice	% of Dose Secreted in Milk (Cumulative)	% of Dose/ℓ of Milk at Peak Concentration	% of Dose in Liver	% of Dose/g of Liver	% of Dose in Muscle	% of Dose/g of Muscle	Approximate % of Dose Retained in Carcass Residue (Without Muscle and Liver)
6 days post-injection (2 cows)	1.9	3.5×10^{-2}	33	2.5×10^{-3}	5.9	2.6×10^{-5}	55
13 days post-injection (2 cows)	1.7	2.5×10^{-2}	33	2.4×10^{-3}	3.3	1.0×10^{-5}	52

TABLE III

PLUTONIUM TRANSPORT TO MILK IN DAIRY GOATS THAT RECEIVED 75 µCi OF CITRATE-BUFFERED PLUTONIUM NITRATE PER ANIMAL PER DAY FOR THREE CONSECUTIVE DAYS

Goat Number	1	2	3
Plutonium Isotope	^{238}Pu	^{238}Pu	^{239}Pu
Total I.V. Dose (µCi) (citrate buffered)	225	225	225
Average Daily Milk Production (kg)			
8-day average	4.6	3.0	1.3
30-day average	-	2.7	2.6
Percentage of Total Dose Secreted in Milk			
128 h post	1.0	1.6	1.0
672 h post	-	2.0	1.1
Excretion Half-life of Plutonium in Milk (h)	-	9.28 ± 1.85 108 ± 13.3 >656	12.6 ± 1.57 84.2 ± 9.34 >700

Intravenous doses of citrate buffered plutonium nitrate have been given to cows (Sutton *et al.*, 1976b) and goats (Stanley and Mullen, 1971; Sutton *et al.*, 1976a). Approximately 1.5 percent of the intravenous dose was recovered in cow's milk during the first 120 hours after injection. This plutonium transfer to bovine milk was somewhat greater than expected. Percentages of dose recovered in the liver and muscle of the four injected cows are shown in Table II. When goats were used as experimental animals, approximately 1 percent of an acute citrate-buffered plutonium dose was recovered in the milk during the first 18 days after injection. Percentages of total dose secreted in goats' milk following multiple plutonium injections are presented in Table III. While many factors were different in the respective projects, Table IV presents a brief comparison of the above mentioned studies. The information is outlined as a percentage of the administered dose transferred to milk.

The biological transport of plutonium, under somewhat natural grazing conditions, has been discussed by Smith *et al.*, (1976). This report reviewed a continuing project on nuclide uptake by Hereford beef cattle grazing on a plutonium-contaminated range at the Nevada Test Site. The plutonium-239 burden in liver and muscle of a 409-kg cow, sacrificed 177 days after being introduced to the contaminated range, was 2.4×10^{-4} and 3.2×10^{-5} percent of the estimated total dose. However, when the oral dose was based on plutonium concentrations in only the liquid fraction of rumen ingesta, the percentages calculated for liver and muscle retention were of course higher (1.5×10^{-3} and 2.0×10^{-4}, respectively). Differences between total dose and available dose in ruminants have also been discussed by Barth and Mullen (1974) and Barth (1977 - this publication).

Citrate buffered plutonium and relatively insoluble particulate plutonium dioxide have been administered orally to respective groups of laying hens (Table V). This work was reported by Mullen *et al.*, (1976). Yolk was the only egg fraction in which plutonium activity was detected, with peak concentrations being reached 12 days after the initial ingestion. When plutonium citrate doses were stopped, yolk concentrations of plutonium decreased rapidly with an initial half-time of approximately 2 days as compared to a half-time of 4.4 days after the termination of plutonium dioxide ingestion.

Figure 1 outlines some potential transport and recycling pathways for plutonium in food products. Plutonium's long half-life produces most of the problem. These pathways, some of which currently represent a very minor contribution, could become more important when long-range hazard predictions are formulated. The main point of emphasis in this figure is the need to examine the biological availability of *in vivo* labeled food products. Liver, meat, milk and eggs that have been contaminated *in vivo* should be fed to suitable animals (adult and juvenile) so as to insure that the low degree of plutonium absorption does not change under these somewhat more natural exposure conditions. The pig, because of certain anatomical and physiological digestive similarities to man, might be a good experimental animal for such studies.

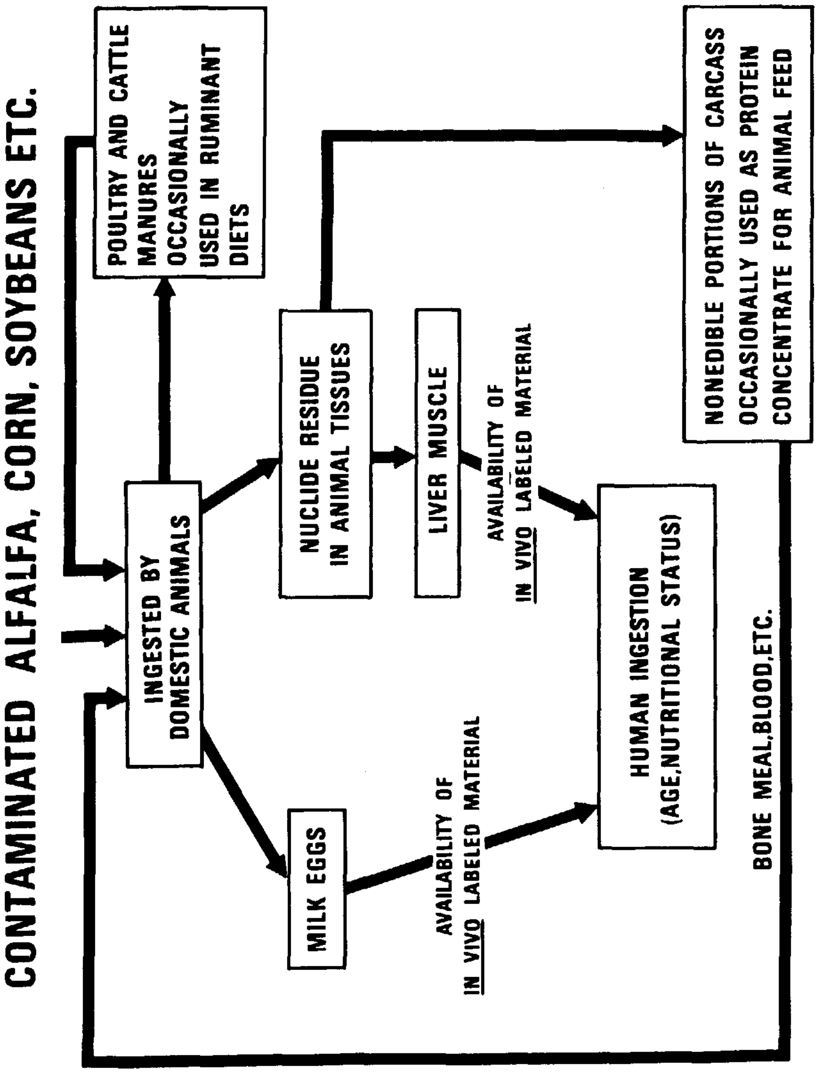

Figure 1 - Outline of some potential transport and recycling pathways for transuranic elements in food products. These pathways, some of which currently represent a very minor contribution, could become more important when long range hazard predictions are formulated.

TABLE IV

BRIEF COMPARISON OF SELECTED STUDIES SHOWING THE PERCENTAGE OF EITHER INTRAVENOUS OR ORAL PLUTONIUM-238 DOSES TRANSFERRED TO MILK IN DAIRY ANIMALS

Animal	Number of Animals	Type of Dose	Duration of Collection(h)	Total % of Dose Transported to Milk
Goat	1	I.V. citrate (acute)	432	1.0
Goat	2	I.V. citrate (multiple)	128	1.3
Cow	1	I.V. citrate (acute)	72	1.3
Cow	4	I.V. citrate (acute)	120	1.5
Cow	4	oral citrate (acute)	720	2.0×10^{-4}
Cow	4	oral dioxide (multiple)	624	2.0×10^{-5}

Some experiments on the transport of plutonium in dairy products have already been conducted by this Laboratory. While it has been noted that transfer of plutonium to milk is relatively small, the subsequent production of cheese concentrated the available plutonium (Miller *et al.*, 1972). In further work, the biological availability of plutonium from milk labeled *in vivo* was investigated. Juvenile goats and calves were given multiple oral doses of either *in vivo* plutonium labeled milk collected from intravenously dosed lactating adults, or *in vitro* plutonium labeled milk adjusted to the same plutonium concentration as the *in vivo* labeled milk. Analyses have not been completed on the calf tissues, but in the preliminary study using goats, no clearly discernible differences in plutonium retention were noted between the two treatment groups.

TABLE V

TRANSPORT OF ORALLY-INGESTED PLUTONIUM TO EGGS, LIVER AND SKELETAL MUSCLE IN CHICKENS

Number of Chickens	Sacrifice Time (days post-treatment)	Plutonium Dose	% of Daily Dose in Eggs at Peak Conc.	% of Daily Dose/g of Egg Yolk at Peak Conc.	% of Total Dose in Liver	% of Total Dose/g of Liver	% of Total Dose/g of Muscle
10	12	1 µCi Pu citrate per chicken per day for 2 weeks	4.0×10^{-2}	2.2×10^{-3}	7.3×10^{-4}	2.0×10^{-5}	4.7×10^{-7}
10	30				1.1×10^{-4}	2.5×10^{-6}	9.7×10^{-7}
10	12	1 µCi Pu dioxide per chicken per day for 2 weeks	1.8×10^{-4}	1.0×10^{-5}	5.0×10^{-5}	1.2×10^{-6}	1.1×10^{-7}
10	30				8.0×10^{-6}	1.8×10^{-7}	7.8×10^{-8}

Confusion sometimes arises when such terms as biological availability are employed and in many cases there is no easily referenced definition. In pharmacological studies, biological availability ultimately describes the extent to which a chemical, at effective concentrations, reaches appropriate receptor sites. In the context of plutonium absorption from *in vivo* contaminated food, biological availability concerns relative uptake and its expression would primarily depend on the quantity of plutonium recovered in the tissues at time of sacrifice. Therefore, since availability is a relative concept, it is important to use at least two, possibly three, groups of experimental animals. One group of animals would receive the *in vivo* plutonium labeled material (milk, liver, eggs, etc.) and another group might receive *in vitro* plutonium labeled material at approximately the same dosing concentration. However, a third group of animals that received plutonium nitrate via capsule, at the same total dose, would also be beneficial as a modified control group and comparisons could readily be made with previously reported gastrointestinal uptake values.

If studies are conducted to evaluate the effects of nutritional status on plutonium uptake, adaptive homeostatic mechanisms must be considered. The close association of plutonium and iron metabolism has already been mentioned and it should be remembered that a change in the percentage of intestinal absorption is one way animals maintain homeostatic control over iron (Miller, 1975). Some investigations on iron absorption are concentrating on an intracellular protein, the synthesis of which increases under conditions of iron deficiency (Forth and Rummel, 1973).

The potential problem of plutonium recycling is also noted in Figure 1. Slaughterhouse wastes, such as blood and bone meal, and animal wastes, such as cattle and poultry manures, can be recycled and included in the ruminant diet. Under certain conditions this process might reintroduce contaminated material into a rather short transport system. While grain is most frequently fed as a supplement, the above mentioned waste products have already been used on a limited scale. In the case of poultry feeding, grains contribute readily digestible starches while such substances as soybean meal and animal by-products (meat and bone meal) have been added to enhance protein quality. In assessing potential problems due to recycled feed materials, it should be remembered that the specific ration components (given to most domestic animals) are affected by various economic factors. Due to changing feed and produce prices, alterations in feed composition are continually made to achieve the most desirable, and ultimately the most profitable ration.

FUTURE PLANS

I. Metabolism of Americium in Dairy Animals.

Both americium and plutonium are deposited extensively in the skeleton and liver but differences, primarily in liver concentration, have been noted. Furthermore, americium appears to be more rapidly removed than plutonium from the plasma following intravenous injection.

A series of investigations will be conducted to determine the biological transport of americium-241 in ruminants. Emphasis will be placed on nuclide transfer to those animal products sold for human consumption.

Phase I will use three (or more) goats and Phase II will use four cows as experimental animals. Single oral or citrate-buffered intravenous nuclide doses will be given to each animal. Both phases will be conducted at the Nevada Test Site Farm and the collection procedures will be similar. Milk, blood, urine and fecal samples will be taken for analysis over a six day period after dosing. Pretreatment samples, to include those used as counting standards, will also be collected for analysis. Approximately seven days after dosing, the animals will be sacrificed, at which time tissue samples will be taken to establish the distribution pattern for americium-241.

Gamma counting is considered the most feasible analytical technique for use in conjunction with these experiments. Americium concentrations in the sample material will therefore be determined by counting the 60 keV gamma-ray of americium-241. Selected samples will also be prepared for radiochemical analyses.

II. Biological Transfer of Actinides via *In Vivo* Labeled Food

While there are currently no specific plans to continue studies on the gastrointestinal uptake of nuclides from *in vivo* labeled food products, the authors believe that such studies would be of considerable benefit. Furthermore, studies which also include pigs and chickens should continue to be part of the overall program because of the significant contribution these species make to food production.

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Transuranics Statistics and Modeling

TRANSURANIC SPECIATION IN THE ENVIRONMENT

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ABSTRACT

The speciation of transuranium elements in the environment was discussed with particular emphasis on Pu. The thermodynamic approach to evaluating Pu speciation was reviewed and examples presented to exemplify the complexities of Pu chemistry in aqueous solutions. An example of the actual evaluation of Pu oxidation states in natural water was presented. The results, which indicated that Pu(III) and/or Pu(IV) dominated the soluble Pu fraction, were compared to a simple model involving the hydrolytic species of Pu(IV). Literature data were also evaluated with respect to the Pu(IV) model. In all cases, the soluble concentrations of Pu in natural waters were in reasonable agreement with the model. Dissolution studies with PuO_2 were reviewed and data presented to demonstrate that oxidized Pu (Pu(V) and/or Pu(VI)) species may be responsible for the relatively high Pu concentrations reported. The mechanism of oxidation appeared to be related to radiolytic effects. The plant uptake of transuranium elements was discussed and the influence of actinide oxidation state illustrated. Plutonium in one soil-plant uptake experiment resembled Th more than U in plant availability.

INTRODUCTION

The environmental behavior of the transuranium elements is complex because of both source term differences and chemical differences between each element. An understanding of the characteristic chemical states of the actinides is important in evaluating their present behavior and in projecting their biogeochemistries.

The oxidation state chemistry of the transuranium elements is unique in that the elements, when present in a given oxidation state, behave alike chemically. The individual oxidation states, however, display different chemistries. In effect, it is oxidation state, rather than the element *per se* which governs chemical behavior. While this seems obvious, it is often overlooked when isotopes, usually with different specific activities, are compared. Thus differences which are noted in the behavior of isotopes may often actually be related to specific activity or oxidation state differences. It is particularly important with Pu and possibly Np that the oxidation states present be defined as chemical form differences may influence biological or environmental interpretations (Dahlman *et al.*, 1976).

The most stable oxidation states of the transuranium elements are probably Np(V), Pu(IV), Am(III), Cm(III). While Np, Pu, and Am can exhibit at least four oxidation states in aqueous solutions (III, IV, V, VI), only Np and Pu are likely to exist in more than one oxidation state environmentally, although Am(IV) might be stable at high pH's and in oxidizing environments (Pourbaix, 1966).

Three general topics will be addressed in this paper. They encompass the potential oxidation states of Pu in the environment as predicted by thermodynamic procedures; what oxidation states of Pu have been evaluated in natural or laboratory solutions which have environmental relevance; and the effect that oxidation state has on plant availability of the actinide elements.

Eh-pH DIAGRAMS AND MINERAL STABILITY CALCULATIONS

Stability field diagrams relating the species of Pu which dominate solid and solution phases under varing Eh-pH conditions have been constructed by Kraus (1949), Pourbaix (1966), Andelman and Rozzell (1970), and Polzer (1971), using thermodynamic constants for the reactions of interest. An evaluation of the phase relationships between Pu species will not be attempted here. Rather, those studies will be briefly reviewed as will a particular example. The reader is referred to Garrels and Christ (1965) or Pourbaix (1966) for more information on the construction of these types of diagrams.

Kraus (1949) presented an early assessment of the effect of pH on the stability of the various Pu species. The stability of Pu(V) at weakly acidic to neutral pH's, and the instability of Pu(III) to oxidation by O_2 were among the major points made. While this early paper was oriented towards Pu chemistry at macroconcentrations, it is of interest historically.

Polzer (1971) presented an extensive examination of the stability of Pu oxidation states under environmental pH's. Both solid and aqueous phase speciation was considered. Figure 1 is taken from that analysis. Because of different sign conventions, the Eh values in the figure are opposite in sign from the usual presentation of environmental-type Eh-pH diagrams (Garrels and Christ, 1966). It will be noted that in the pH range of 4 to 9, and the Eh range of +0.6 to -0.3 volts (-0.6 to +0.3 volts in the figure), soluble Pu^{3+} and PuO_2^+ species dominate. Because of its low solubility, Pu(IV) was not considered. The line between the domains of Pu^{3+} and PuO_2^+ represents the point of equal concentrations of the two species. Polzer (1971) pointed out that this analysis (Figure 1) considered only uncomplexed species (except with OH^-).

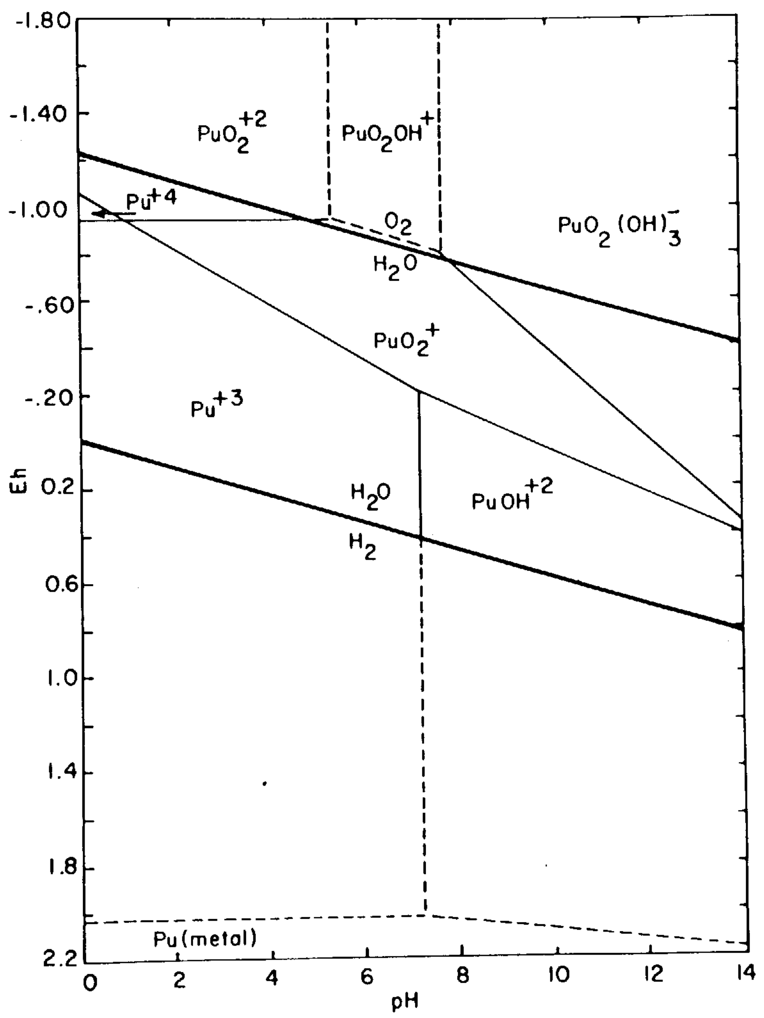

Fig. 1. Stability relations of dissolved plutonium species in water at 25°C and one atmosphere total pressure (Polzer, 1971).

Silver (1971) presented an interesting approach to evaluating the speciation of Pu in natural waters. The stability of PuO_2^+ at environmental pH's was recognized and discussed. The major difficulty in appreciating Silver's approach, however, lies in his use of "alpha factors" to account for changes in ion activities due to complexation. For example, millimolar Pu(IV) concentrations are predicted in certain circumstances where complexation is active. How realistic are such concentrations is a question which remains to be answered.

Andelman and Rozzel (1970) examined the stability relationships of Pu in aqueous solutions. Pentavalent Pu apparently was not considered because of its tendency to disproportionate; however, Kraus (1949), Silver (1971), and Polzer (1971) recognized that this reaction was pH and concentration dependent. Consequently, Pu^{3+} was determined to be the dominant species if complexation by $CO_3^=$ was not considered, but masked by $PuCO_3^{2+}$ if carbonate was included. Schell and Watters (1975), in their review, used the solubility diagram derived by Andelman and Rozzell (1970) to demonstrate the complexity of Pu species in natural waters. It was pointed out by Andelman and Rozzell, however, that the effect of redox potential was <u>not</u> considered for that diagram.

Rai and Serne (1977) took a slightly different approach in evaluating Pu speciation. In addition to hydrolytic species, Rai and Serne considered other inorganic ligands (F, HPO_4^-, SO_4^-, Cl^-, $CO_3^=$) in their paper. Their analysis was aimed at predicting soil solution activities of soluble Pu species and solid phase compounds under varying redox and pH regimes. It was concluded that under reducing conditions ($pO_{2(g)}$ = 80 atm.)* and pH 8, Pu^{3+} and its complexes dominate. In oxidizing environments, $PuO_2CO_3OH^-$ is the predominant soluble species at pH 8.0. This approach is illustrated in Figure 2, which was taken from their analysis of the activity of various Pu ions in solutions in equilibrium with $PuO_2(s)$. The assumed conditions were pO_2 = 16 atm.; pCO_2 = 3.52 atm.; pCl^- = pSO_4^{2-} = 2.5; pF^- = 3.5; and $pH_2pO_4^-$ = 5.0. As Figure 2 illustrates, their analysis was a comprehensive attempt to integrate the chemistry of Pu with the characteristics of natural solutions. The dominant solid phase under those conditions, according to their analysis, was $PuO_2(s)$.

An example of an Eh-pH approach is discussed at this point to both illustrate the basic concept and to demonstrate the type of complexities which do not appear in an analysis of Pu alone. The oxidation of Pu(III) to Pu(IV) at environmental pH's might be written as follows:

$$Pu^{3+}_{(aq)} + 4\ H_2O = Pu(OH)_{4\,(s)} + 4\ H^+ + e^- \qquad (1)$$

where aqueous Pu^{3+} is in equilibrium with solid phase $Pu(OH)_4$. The Nernst equation of reaction (1) is:

$$Eh = E^o + \frac{0.059}{n} \log \frac{(Pu(OH)_4)\ (H^+)^4}{(Pu^{3+})} \qquad (2)$$

where E^o, the formal potential in volts, and 0.059 are constants; n is the change in electrons in the reaction; and ( ) represents ion activity. The activity of a pure solid is taken as unity, thus equation 2 reduces to

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*$pO_{2\,(g)}$ = - log of the O_2 pressure.

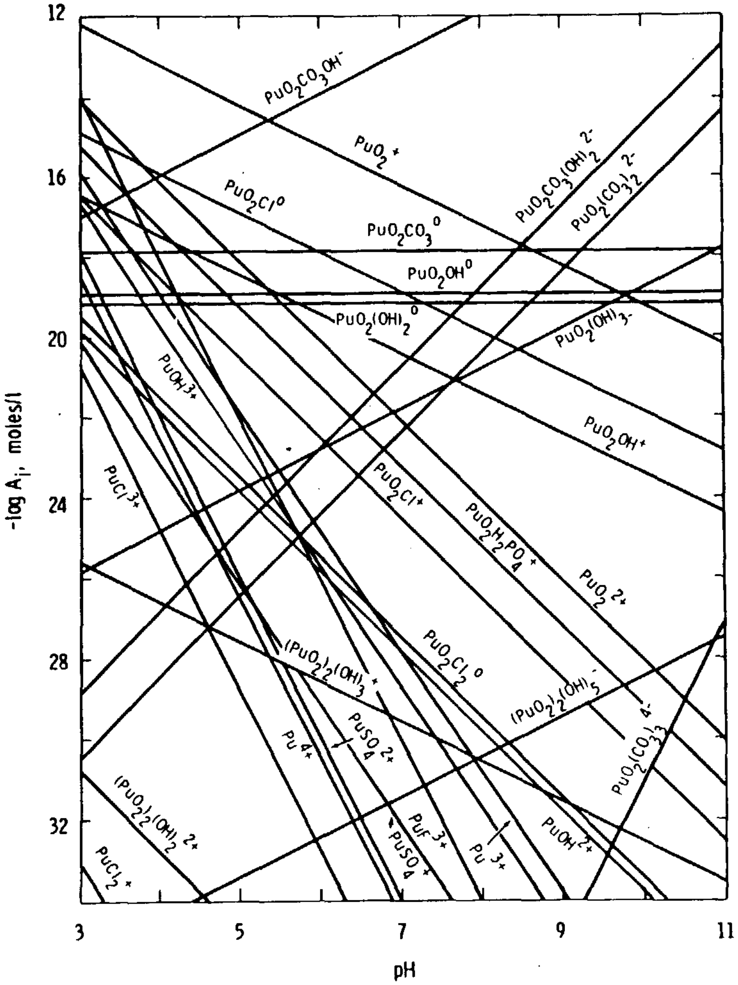

Fig. 2. The activity of various Pu ions in the soil solution in equilibrium with $PuO_{2\,(s)}$ under oxidizing conditions (Rai and Serne, 1977).

$$\mathrm{Eh} = E^{o} + 0.059 \log \frac{(H^{+})^{4}}{(Pu^{3+})} \tag{3}$$

or

$$\mathrm{Eh} = 1.182 - 0.236\ \mathrm{pH} - 0.059 \log (Pu^{3+}) \tag{4}$$

Equation (4) is taken from Pourbaix (1966), who used $E^{o} = 1.182$ volts for $Pu(OH)_4$ as the solid phase. Using the assumption that ion activity approximates ion concentration, equation 4 can be used to establish the Eh-pH line at which a given concentration of Pu^{3+} will be in equilibrium with solid-phase $Pu(OH)_4$. Equation 3 and 4 predict that as pH increases (Eh constant), the equilibrium $[Pu^{3+}]$ will decrease. Conversely, when pH is constant, lowering Eh increases the maximum concentration of Pu(III) stable in the presence of $Pu(OH)_4$.

Figure 3 is an Eh-pH diagram comparing the Pu^{3+} - $Pu(OH)_4$ and Fe^{2+} - $Fe(OH)_3$ redox system. The diagram was constructed using applicable equations (Pourbaix, 1966). The Fe^{2+} - $Fe(OH)_3$ line was calculated at 10^{-4} $\underline{M}$ (Fe^{2+}) using the equation:

$$\mathrm{Eh} = 1.057 - 0.1773\ \mathrm{pH} - 0.0581 \log (Fe^{2+}) \tag{5}$$

The Pu^{3+} - $Pu(OH)_4$ lines at 10^{-4} and 10^{-8} (Pu^{3+}) were calculated using equation 4 above. This approach suggests that under environmental conditions where Fe(III) is reduced to Fe(II), Pu(IV) may be susceptible to reduction to Pu(III). This reduction would occur more readily under acidic conditions. The relatively greater abundance of Fe over Pu in the environment also suggests that the redox reactions of Fe may have an influence on Pu chemistry. This illustration was specifically used to point out that in the environment, even if equilibrium was presumed to be established, other substances like Fe can become important relative to Pu. Thus at dilute Pu concentrations, environmental Fe^{2+} concentrations could be sufficiently in excess of Pu to influence Pu chemistry in a manner not obvious when Pu calculations are considered alone.

Ferrous iron is, of course, a reductant towards Pu^{4+} in acidic solutions (Connick, 1954) as Figure 1 shows (i.e., at pH 1, Fe^{2+} is a stronger reductant than Pu^{3+}).

Besides Fe^{2+}, other reductants are present in the environment. Bondietti *et al.* (1976) discussed the influence of organic substances of Pu oxidation states and suggested that Pu(VI) or Pu(V) appear unstable in the presence of humic substances and polysaccharides.

The use of stability field diagrams and mineral stability calculations has one very important limitation which must be kept in mind. The results of such analyses are valid only for the conditions considered. Reactions which are not considered may also occur, adding additional complexities to the real world behavior of Pu in soils, sediments, or natural waters. Stability field diagrams also do not consider kinetic parameters of the given reactions.

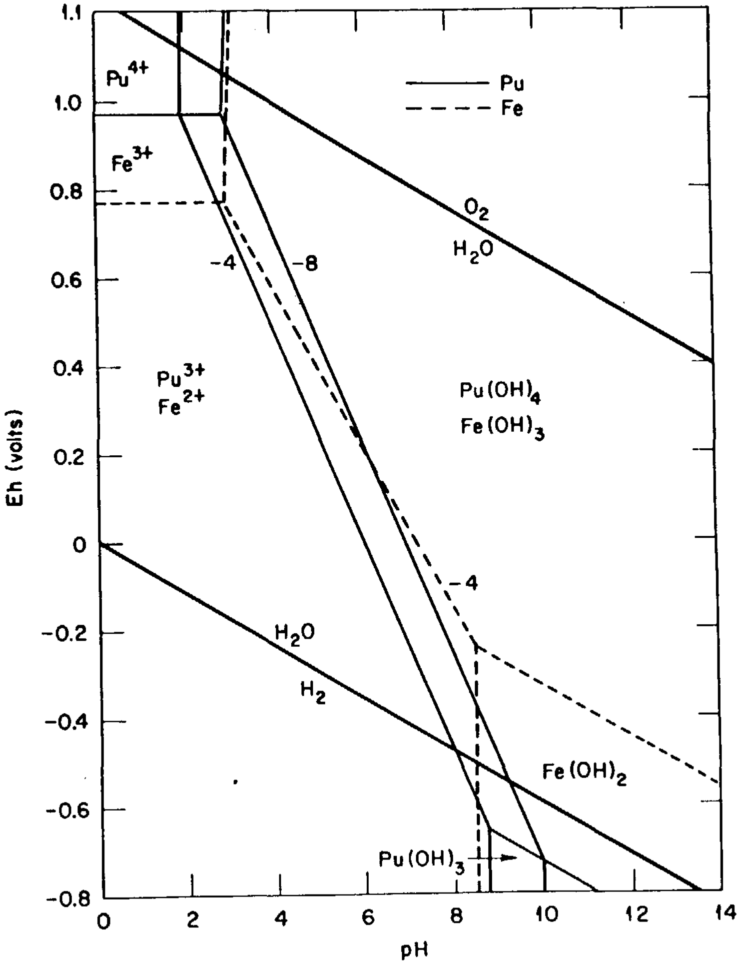

Fig. 3. Comparison of the stability relationships for soluble Pu^{3+}(10^{-4} and 10^{-8} $\underline{M}$) and Fe^{2+}(10^{-4} $\underline{M}$) in equilibrium with $Pu(OH)_4$ and $Fe(OH)_3$, respectively. One atmosphere total pressure and 25°C. Plutonium (V) and (VI) states not included in analysis.

However, the thermochemical approach provides the necessary boundary parameters upon which experimental efforts can be based. Thus, while it is difficult to imagine natural environments in which Pu species are at equilibrium, the delineation of speciation domains through such calculations provides valuable information not readily obtained experimentally.

CARBONATE COMPLEXES

Both Polzer (1971) and Andelman and Rozzell (1970) considered the possibility of carbonate complexation of Pu(IV). The only Pu(IV) carbonate complex which has been reported is $PuCO_3^{2+}$, having a stability constant of 10^{47} (Gelman *et al.*, 1962). The validity of this very high constant has been questioned, however (Cleveland, 1970); thus, the use of this constant by Polzer (1971) and Andelman and Rozzell (1970) may be questionable. Rai and Serne (1976) chose not to include it in their analysis.

Substantial research on Th(IV) and U(IV) indicates that the carbonate complexes of tetravalent actinides are important only at high carbonate concentrations. Thus $U(CO_3)_5^{6}$ is stable only over a narrow range of pH's (McClaine *et al.*, 1956). Desorption studies using 0.025 M HCO_3^- showed that U(VI) bound to proteins was more effectively removed than U(IV) (Dounce and Flagg, 1949). The same authors also reported oxidation-reduction potential studies which indicated that HCO_3^- was a better complexer for U(VI) than U(IV). A similar case is apparent for Pu. Moore and Dam (cited in Connick, 1954) estimated the Pu(IV) - Pu(VI) potential in 45% K_2CO_3 to be near -0.2 volt, indicating strong complexation of Pu(VI).

The potential in 1 M OH^- is about -0.4 volt (Connick, 1954), which may indicate that carbonate interacts with Pu(VI) more strongly than PU(IV) when in competition with OH^-. Under environmental conditions, the greater tendency of tetravalent actinides to hydrolyze may thus make carbonate complexation more important for the less hydrolytic hexavalent state. This effect is recognized in the geochemistry of Th(IV) and U(VI); carbonates playing a role in the mobility of U(VI) but not Th(IV) (e.g., ^{230}Th produced from radiodecay of ^{234}U is rapidly removed from seawater). Bondietti *et al.* (1976) observed a greater tendency for PU(VI) to remain in solution than Pu(IV) in dilute bicarbonate solutions.

The available information suggests that the tendency of carbonates, at typical environmental concentrations, to complex with Pu(IV) in an environmentally important manner be viewed with some reservations. While carbonates do complex Pu(IV), the CO_3^{2-}/OH^- ratio appears critical.

FIELD OBSERVATIONS ON Pu SPECIATION

To evaluate the oxidation states of Pu in environmental solutions, a method which selectively separates one or more oxidation states is necessary. Because of the dilute concentrations of Pu which are found in near-neutral solutions, the method(s) would have to rely on the coprecipitation of Pu (or any other actinide) with a carrier. Bismuth phosphate, for example, is an excellent carrier of tri- and tetravalent actinides. The $BiPO_4$ precipitation of PU(III) + Pu(IV) has been used for nuclear fuel reprocessing and to determine Pu in water samples (Kooi *et al.*, 1958). Bismuth phosphate precipitates do not carry significant amounts of the penta- and/or hexavalent species of Pu, Np, and U. Therefore this approach was taken to determine the oxidation state of Pu in water and will be discussed briefly.

The samples of natural water assayed for Pu oxidation states were taken from White Oak Lake (WOL), a freshwater impoundment on the Oak Ridge National Laboratory reservation. The analytical methods employed for the natural water samples were essentially as outlined by Scott and Reynolds (1975), except that treatment with $NaNO_2$ at 70° C. was omitted. The water samples were collected and filtered (Whatman 50, then Millipore 0.45 μm). Eighteen liters were acidified, ^{236}Pu tracer added (for recovery efficiency), and the solution made 0.1 M in NH_2OH and 10^{-3} M in $FeCl_2$. This sample was used to determine total Pu (the NH_2OH and Fe(II) reduce Pu(VI) and Pu(V)). An additional 18 liters of filtered water were tagged 1 hour before acidification with a mixture (27% Pu(IV), 73% Pu(V) + Pu(VI)) of ^{242}Pu. The $BiPO_4$ procedure (without valence adjustment) was used to selectively determine Pu(III) + PU(IV).

Table 1 summarizes the results on Pu behavior in filtered WOL water. When determined, the Pu appeared anionic rather than cationic. That is, it was quantitatively retained by Dowex-1 anion exchange resin but not by Dowex-50 cation resign (NA^+ form). One sample (September, 1975) also showed that the Pu which passed the 0.45 μm filter also passed a 10,000 MW Amicon membrane filter, suggesting low molecular size. Measurements of redox potential and pH immediately after sample collection and at the time of analysis showed only slight changes (the redox potentials (Eh) ranged from 0.32 to 0.42 volts; pH from 7.9 to 8.9). The oxidation states of the indigenous Pu appeared to be largely Pu(III) or Pu(IV), in that there was no difference found between the amount of Pu carried by the $BiPO_4$ precipitates (reduced and nonreduced). The recovery of ^{242}Pu was 25% of the total added, which was about what was expected if only Pu(III) and Pu(IV) was carried. Thus, while added ^{242}Pu was fractionated based on known oxidation state differences, the indigenous Pu was not. The results suggest that for these water samples, the dominant oxidation states present were (III) or (IV). For calculation purposes, the $^{239,240}Pu$ activity was used to obtain the molar concentrations of Pu, assuming that ^{239}Pu was dominant.

The exact nature of the solution-phase Pu species which exist in natural waters are not known. To the extent that species characteristics in WOL water are understood, i.e., low molecular weight, Pu(III) or (IV), and anionic, it is of interest to compare the observed characteristics of Pu in filtered water

TABLE 1. CHARACTERISTICS OF Pu IN THE < 0.45 um FRACTION OF WHITE OAK LAKE WATER

Study and Date	Moles/Liter* Pu, x 10^{15}	Comments
Solution Characteristics		
Oct. 1974	0.8	100% anionic
Sept. 1975	2.1	100% anionic, $\leq$ 10,000 MW
Valence State Characterization		
April 1976		
Total	1.1 ± 0.1	$BiPO_4$ (reduced)
Pu (III) + (IV)	1.1 ± 0.1	$BiPO_4$
Oct. 1976		
Total	1.5 ± 0.2	$BiPO_4$ (reduced)
Pu (III) + (IV)	1.6 ± 0.2	$BiPO_4$

*10^{-15} $\underline{M}$ ^{239}Pu = 14.7 fCi/l.

with a simple model for Pu speciation in solution. This model would involve the most stable oxidation state, (IV), and OH^- ion.

Baes and Mesmer (1976) have evaluated the hydrolytic properties of Pu(IV) and calculated hydrolysis quotients for the stepwise hydrolysis of Pu^{4+}. These hydrolytic stability constants have been used to calculate the concentrations of five Pu(IV) species which could theoretically exist in equilibrium with crystalline PuO_2. The results of these calculations are presented in Figure 4. It can be noted that as the solution pH increases, the successive addition of OH^- to Pu^{4+} results in a neutral species, $Pu(OH)_4^0$, dominating the soluble species from about pH 4.2 to 5.5. By analogy to U(IV), Baes and Mesmer calculated the theoretical stability constant for a $Pu(OH)_5$ species. This hypothetical species dominates the Pu species present in solution above pH 5.5. At pH 8, about the pH of WOL water, the calculated equilibrium concentation of this species would be p13.5. The observed concentrations of the negatively-charged, Pu(III) or Pu(IV) species in WOL was ∿ p15. Thus, the observed concentrations of Pu did not exceed this simple model for Pu(IV) speciation in water.

In light of the above observations, a number of reported concentrations of Pu in filtered natural water from different environments have been compared to this estimated PuO_2 solubility. Table 2 summarizes the references used for this evaluation. Generally, these investigators reported Pu results in radioactivity units (i.e., pCi/l). The reported activity values for $^{239,240}Pu$ have been converted to moles/l ^{239}Pu.

Where water pH's were not given, pH 8 was assumed. The maximum values reported by each investigator and the $Pu(OH)_5^-$ line are plotted in Figure 5. The radiation concentration guide (RCG) value* for soluble ^{239}Pu in unrestricted areas is also plotted.

Fitting observed data to a theoretical solubility limit was not done to suggest that crystalline PuO_2 controls the soluble Pu in natural solutions. It is generally the case that observed concentrations of trace metals in natural solutions (for example, seawater) are not controlled by the solubility product of the least soluble salt (Turekian, 1969). For an element like Pu, it is not likely that solubility will control solution concentrations because of the trace levels released to the environment. However, to the extent that an upper limit on solution concentrations can be defined, it is important to evaluate what concentrations of Pu could be expected in contaminated sites. If reported values of soluble Pu remain at or below a predicted value, confidence in our understanding of the solution behavior of Pu is enhanced.

It is important to note that only Pu(IV) was considered in the data-fitting model. The previous discussion on theoretical speciation indicated that Pu(III) could be important. However, those analyses considered Pu(IV) to be so insoluble that it could be neglected relative to the solubility of Pu(III). We have chosen to consider only Pu(IV) because it may be the most stable oxidation state, although not necessarily the most soluble. Plutonium (V) and (VI) were not considered solely because they do not appear to dominate the soluble Pu in WOL.

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*USAEC Rules and Regulations, Title 10, Part 20, Appendix B, Table II, Dec. 10, 1969.

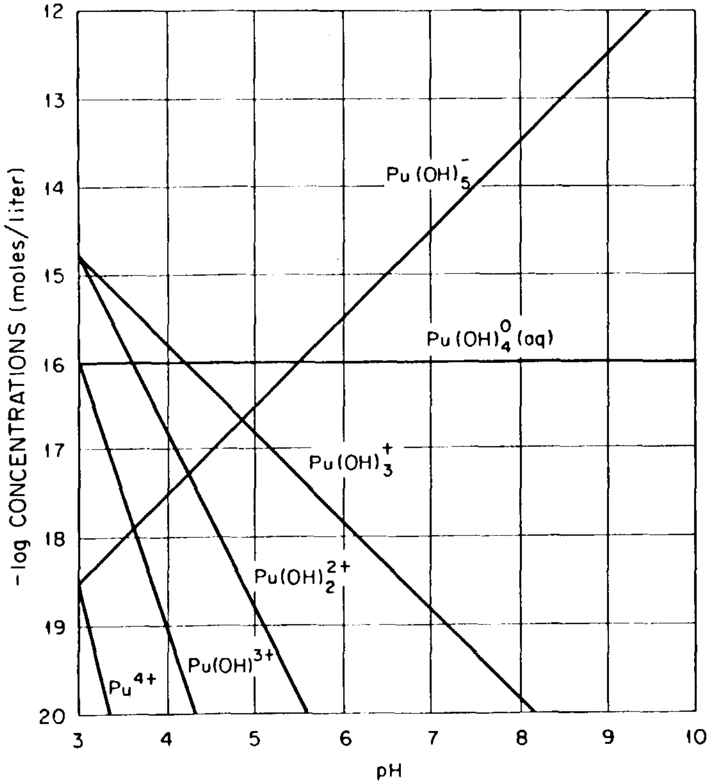

Fig. 4. Estimated concentrations of hydrolyzed Pu(IV) species in solutions saturated with crystalline PuO_2 (Baes and Mesmer, 1976).

TABLE 2. REFERENCES AND STUDY SITES FOR DATA PLOTTED IN FIGURE 5. SITES WERE CHOSEN TO REPRESENT A VARIETY OF LOCATIONS.

Figure Code	Study Site	Investigator/Source
1.	Oak Ridge, Tn. (WOL)	Bondietti, 1976
2.	Miami River (Ohio)	Bartelt *et al* 1974
3.	U-Pond, Hanford (Wa.)	Emery *et al* 1976
4.	Maxey Flats (Ky.)	Unpublished*
5.	Maxey Flats (Ky.)	Unpublished*
6.	Seawater	IAEA**
7.	Windscale (U.K.)	Heatherington *et al* 1976
8.	Lake Michigan	Wahlgren *et al* 1976
9.	Bombay Harbor (India)	Pillai and Mathew, 1976
10.	Pacific Ocean	Miyake *et al* 1976
11.	Enewetak Atoll	Noshkin *et al* 1976
12.	Los Alamos	Hakonson *et al* 1976
13.	ORNL (Burial Ground)	Bondietti, 1976
14.	Rocky Flats (Colo.)	Johnson, 1974

*Trench water samples, commercial radioactive waste burial ground. S. A. Reynolds, ORNL, personal communication.

**IAEA intercalibration sample (IAEA-SW-I-3) taken nearshore to an unspecified nuclear facility.

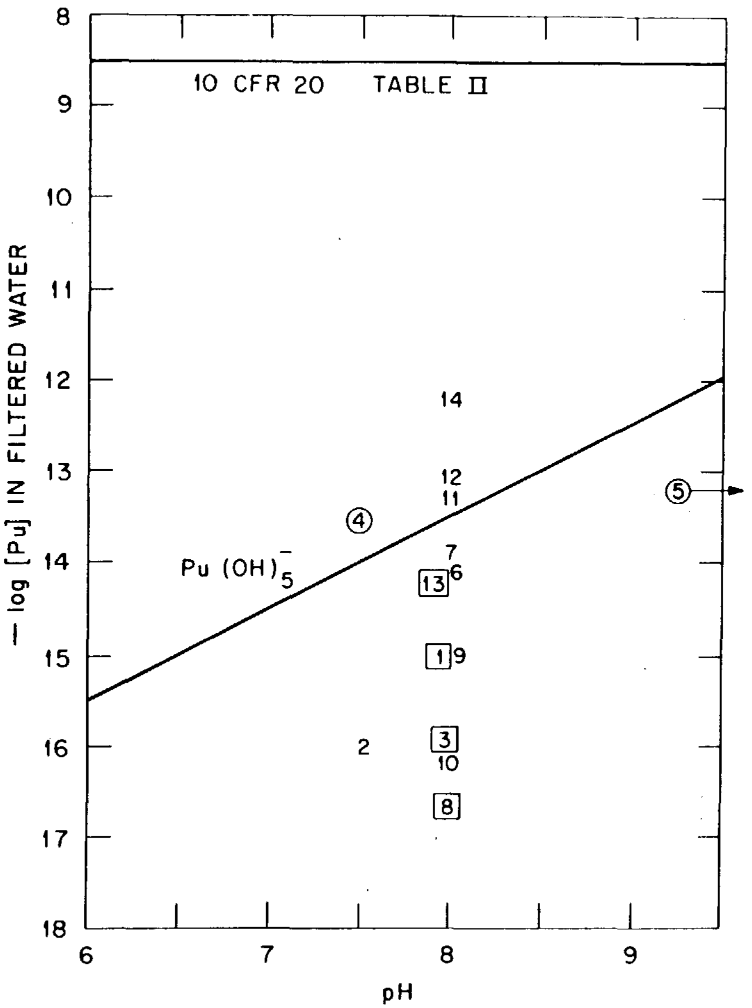

Fig. 5. Comparison of reported concentrations of plutonium in filtered water with the dominant specie in Fig. 4. See Table 2 for number key. Boxed numbers indicate speciation was studied; circles denote commercial radwaste burial ground.

The previous discussion on the nature of solution-phase Pu species which could be in equilibrium with PuO_2 indicated that if Pu(IV) species were the only soluble species in solution, the amount of monomeric Pu which would be found in solutions contacting crystalline PuO_2 would be extremely small (provided no complexes were present). However, when PuO_2 is placed in near-neutral solutions, Pu appears in the aqueous phase in concentrations for exceeding that predicted by simple chemical solubility. Furthermore, $^{238}PuO_2$ appears to "dissolve" faster than $^{239}PuO_2$ (Patterson *et al.* 1974). Adams *et al.* (1975) reported "soluble" concentrations of $^{239}PuO_2$ microsphere dissolution studies which were near 10^{-11} $\underline{M}$. Bondietti and Reynolds (1976) and Dahlman *et al.* (1976) reported soluble Pu species near 10^{-10} to 10^{-11} $\underline{M}$ Pu in $^{239}PuO_2$ dissolution experiments, with the oxidation states of Pu in solution determined. Smith *et al.* (1972) reported ultrafiltration data for PuO_2 suspended in water. From their report, ^{238}Pu concentrations of 10^{-8} $\underline{M}$ can be estimated for the Pu species which passed an ultrafilter with 26 A pores.

The "dissolution" of PuO_2 can be summarized with two general statements: first, the observed concentrations of "soluble" Pu can exceed that expected if chemical solubility alone (i.e., the dissociation of Pu(IV) monomeric species from a solid) controls the solution phase concentrations; and secondly, the specific activity of the incorporated Pu isotope appears to affect the rate of dissolution.

The abnormal solubility of PuO_2 in water has led to several hypotheses. The higher radiation density of $^{238}PuO_2$ has been implicated as the reason for its faster apparent dissolution rate (Patterson *et al.*, 1974; Fleischer, 1975). These authors, among others, suggested that fragmentation of the oxide lattice could release PuO_2 of colloidal dimensions; consequently, the solution phase would contain Pu in forms other than simple ions. This phenomenon (aggregate recoil) results from alpha emission recoils which have sufficient kinetic energy to easily break chemical bonds (Fleischer, 1975). Fleischer (1975) proposed a model which suggested that the differences in "solubility" between the 238 and 239 isotopes of Pu when present as the oxide was solely due to this aggregate recoil effect. Recoils which occur at the surface of the oxide could account for ∿ 10^4 ejected atoms of Pu per effective recoil. Fleischer compared his model to experimental data on Pu solubilization and found good agreement between observed dissolution rates and theoretical emissions of aggregates of PuO_2.

Patterson *et al.* (1974) also suggested that the radiolysis of water at the oxide surface could produce species which might reduce Pu(IV) to Pu(III), increasing the solubility of Pu. A major product of α-radiolysis of water is H_2O_2, and in acid solutions reduction of higher oxidation states to Pu(III) is well known. However, Kraus (1949) pointed out that hydrogen peroxide would oxidize Pu(III), but not reduce Pu(IV), at near-neutral pH's. The action of peroxide on Pu in acidic solutions is very complex, and the results of Pu oxidation state changes observed for acid solutions (the commonly reported situation) may not be relevant at environmental pH's.

Bondietti and Reynolds (1976) reported that substantial amounts of an oxidized Pu species (Pu(V) or Pu(VI)) occurred in neutral solutions contracting high-fired PuO_2. The PuO_2 used for those studies was in the microspheres, having a diameter range of 149-174 μm, a specific surface area of 0.012 m^2/g, and made by the "sol-gel" process. The microspheres had been calcined at 1150° C. For the experiments described here, 2-3 mg amounts of the microspheres were used (weighing accuracy was 0.01 mg). Sodium bicarbonate (1 m $\underline{M}$) and organic solutes were equilibrated with the microspheres for varying time periods.

When bicarbonate was used, two types of Pu were found in the aqueous phase. The initial period of water contact resulted in the solution phase Pu being dominated by a "refractory" Pu species. This Pu species displayed the properties of Pu polymer (Bondietti and Reynolds, 1976) because it failed to extract as an ionic species into organic solvents and sedimented in the ultracentrifuge. While this PuO_2-like species dominated the initial equilibration period (several weeks), the presence of an oxidized Pu species (V or VI) was also observed. The rate of appearance of this oxidized Pu was nearly constant over periods of up to 127 days. The net result of the appearance of these two Pu forms was to have an initially high and erratic rate, which appeared to be due to the refractory species, and a subsequent slower rate which was entirely contributed by the oxidized species.

The initial contribution of this refractory Pu resulted in initially higher "dissolution" rates which were very erratic; this observation also pervades the literature. Thus, Patterson *et al.*, 1974, in their review and experiments, reported that the initial dissolution rates of PuO_2 (238 and 239 isotopes) are higher and more erratic than the subsequent, longer-term rate which was relatively constant. Those investigators attributed the initial, rapid rate as possibly due to the initial reactions of the oxide with the solution. We would propose that an alternative but not unrelated mechanism could involve recoil aggregates; that is, the decay events which occurred postcalcination would cause a buildup of recoil-derived fragments at the microsphere surface. When placed in water, these are released to the aqueous phase as colloidal material. In our case, we have observed that the Pu in this initial release behaves as PuO_2 and also sediments in the ultracentrifuge, while the oxidized Pu species do not.

The oxidation of Pu appears to be due to radiolysis. Hydrogen peroxide will oxidize tracer amounts of Pu(IV) to Pu(V) or (VI) in alkaline solutions (Connick, 1954). We have observed, for example, the oxidation of Pu(IV) by peroxide under the following initial conditions: 10^{-11} $\underline{M}$ Pu(IV), 10^{-6} $\underline{M}$ H_2O_2, and 10^{-3} $\underline{M}$ $NaHCO_3$. Figure 6 illustrates that in the case of the microspheres, oxidation apparently occurs near the oxide surface. The appearance of $^{239}Pu^{(o)}$, as shown in the figure, increased with time, starting at about 1.5 x 10^{-11} g dissolved ^{239}Pu at 1 week, and increasing to about 12.9 x 10^{-11} g dissolved Pu at 10 weeks. In addition to the Pu originating from the oxide, the aqueous phase (20 ml) initially contained about 66 x 10^{-11} g of ^{242}Pu. The ^{242}Pu was added initially in the tetravalent state. Throughout the 10-week period, only about 3% of the total added ^{242}Pu appeared in the oxidized state, even though the mass of ^{242}Pu was comparable to the ^{239}Pu mass appearing in solution.

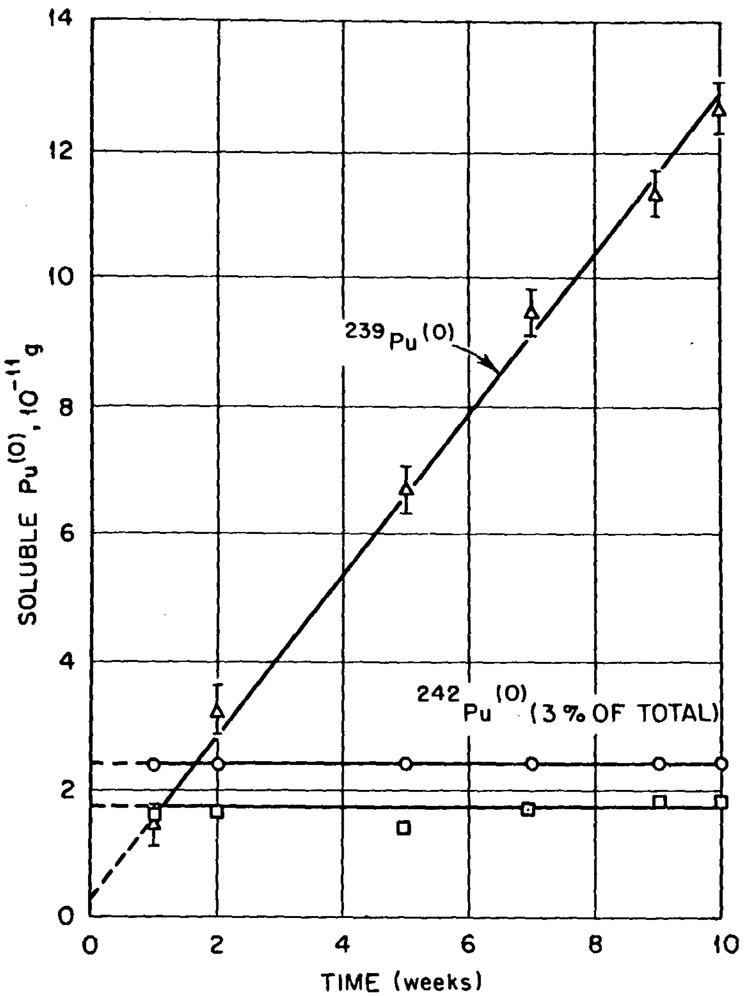

Fig. 6. Appearance of oxidized plutonium in 10^{-3} $\underline{M}$ sodium bicarbonate solution contacting 1.77 mg of PuO_2. The ^{239}Pu originated from the PuO_2, while ^{242}Pu had been added separately.

The two data lines for ^{242}Pu represent experiments with and without PuO_2. The 3% oxidized Pu probably formed during addition to the bicarbonate solution. In any event, ^{242}Pu in the bulk solution was not being oxidized, but Pu originating from the microspheres was. This suggests that impurities in the solution or atmospheric oxygen are not involved but that the oxidation occurred prior to or during transfer from the solid to solution. We might expect that the intense radiation field at the surface of the oxide is involved.

It appears that for large particles of PuO_2, where the radiation field near the oxide surface can be substantial (about 10^5 rad/hour at 20 μm for the microspheres used in the above work), oxidation of Pu to the more soluble penta- or hexavalent state results in concentrations of Pu which thus exceed solubility limits of the tetravalent state. For submicron particles of PuO_2, the importance of this radiolysis effect would be reduced since the number of emergent alpha particles from the interior of the oxide would be less, and consequently, radiolysis would be less intense.

Apart from radiolysis-induced oxidation, however, the actual dissociation of Pu from the oxide matrix may be physical. Fleischer's model (1975) explained the appearance of Pu solely as a physical effect. Thus, aggregates of Pu could be displaced from the oxide surface by recoil phenomenon; oxidation may or may not be related to this effect in that these fragments enter a zone of intense radiolysis. Thus, the actual "dissolution" may not be related to oxidation--only the subsequent solubility of the oxidized Pu results in it being observed in solution. Table 3 summarizes observations on the long-term dissolution of PuO_2 microspheres in inorganic and organic solutes. Bicarbonate, acetate, citrate, and ethylenediaminetetracetic acid (EDTA) were employed as solutes. The rates of Pu appearance in the aqueous phase represent only nonrefractory Pu. The refractory Pu observed during the initial phases of oxide-solution contact are not included; thus, the table reflects only the long-term, rather constant rate. One of the interesting results is that EDTA and citrate slightly accelerate the dissolution rate over acetate or inorganic solutes, and plutonium (IV) is the oxidation state of the solution phase species. This is due either to a failure of oxidation to occur in the presence of complexers of Pu(IV) or to reduction by the organics (Bondietti *et al.*, 1976). Overall, the similar rates suggest that the oxide is extremely insoluble and that complexers like EDTA do not substantially change the dissolution rates. This observation has been made by Rabbe *et al.* (1973).

The basic evidence suggests that the dissolution of PuO_2 is very slow, on the order of hundreds of years for submicron particles (Fleischer, 1975). Due to radiolysis effects, oxidized species can occur in some cases but their environmental stability is probably low (Bondietti *et al.*, 1976; Dahlman *et al.*, 1976). The radiolysis-induced oxidation may be important in waste management-related problems, but probably not for submicron, dispersed, environmental-type PuO_2. However, the effect of oxidation may be important in laboratory studies of the type described here. Obviously, understanding the speciation of Pu in experimental solutions is essential for interpretation of the results.

TABLE 3. DISSOLUTION RATES OF PuO_2 MICROSPHERES IN INORGANIC AND ORGANIC SOLUTES

Solute	Days	Dissolution Rate 10^{-10} g/cm^2-day	$Pu^{(o)}$* %
$NaHCO_3$	33-91	0.27**	95+
Acetic Acid			
pH 2	760	1.9	32
pH 4	760	0.23	20
pH 6	680	0.05	92
Citrate (pH 6)	715	2.7	3
EDTA (pH 6)	715	5.5	1

*Fraction of non-refractory Pu which was in the Pu (V) or (VI) state.

**Average of 3 separate samples in $NaHCO_3$.

BIOLOGICAL AVAILABILITY

While the primary purpose of this paper is not to review biological uptake of transuranium elements, it is appropriate to examine those experiments or field observations where the reported data indicate differences in biological uptake which can be ascribed to chemical state. In particular, the dominant oxidation state of Pu might be deduced from availability to plants in cases where the state in the soil is not known. Jacobson and Overstreet (1948) conducted clay adsorption and plant uptake experiments with different oxidation states of Pu. Plutonium which was added initially as Pu(VI) was assimilated by barley plants to a greater degree than Pu added in the tri- or tetravalent states. Price (1973) compared the plant uptake of Np(V), Pu(IV), Am(III) and Cm(III) and observed that Np was assimilated the greatest. Americium and Cm were taken up about equally, as might be expected since they are both trivalent and chemically very similar, while Pu was assimilated the least. Cummings and Bankert (1971) compared Pr, Ce, and Pu and found that two lanthanides, which are characteristically trivalent, were about ten times more available to oats than Pu. The Pu oxidation state was not given; however, the lower uptake of Pu would suggest the tetravalent state dominated since the trivalent state of the actinides and lanthanides are similar chemically. Adams *et al.* (1975) compared the uptake of Pu, U, and Am and observed the same effect; Am was taken up more readily than Pu, while U was similar to Am.

Differences in plant availability of the different oxidation state species (MO_2^+, MO_2^{2+}, M^{3+}, M^{4+}) may be related to their relative reactivity with soil components or their relative insolubilities. Plant uptake appears to be consistent with the relative tendencies of these oxidation states to hydrolyze (Dahlman *et al.*, 1976).

Table 4 presents the relative plant uptake of Th, Pu, and U from soil contaminated in 1944 by trace Pu. A description of this site was discussed in this symposium by Dahlman and McLeod. Representative concentration ratios (C.R.) are presented for snap beans, millet, soybeans, and tomatoes. These CR's are based on the 8 M acid extractable soil concentrations of the three elements. This extraction removes all of the soil Pu; consequently, natural U and Th were compared using the same extraction conditions. As can be observed, the relative uptake of Pu and Th are similar, indicating that plant roots appear to be mobilizing both elements in a similar manner. Uranium is more readily assimilated, which is consistent with the observed biogeochemical behavior of U and Th; U being more available than Th. This provides indirect evidence that Pu may behave like Th, a concept consistent with the thesis that Pu should be tetravalent in aerobic environments (Bondietti *et al.*, 1976).

Table 5 presents additional extraction data for the three elements from this soil. When 1 M HNO_3 or 10% Na_2CO_3-5% $NaHCO_3$ solutions were used as extractants, the ratios of Th to Pu were similar to the 8 M acid values. While weak nitric acid or carbonate did not substantially differ in their extraction of U (73 and 71%, respectively), dilute nitric acid was not effective in extracting Pu or Th and carbonate was only intermediate between the strong (8 M) and weak (1 M) acid. This behavior is probably due to the fact that in 1 M acid, Pu and

TABLE 4. COMPARATIVE PLANT UPTAKE OF ACTINIDES FROM ORNL FLOODPLAIN

Plant	Concentration Ratio*, **		
	^{238}U	^{232}Th	^{239}Pu
Snap Bean	0.01	0.002	0.002
Soybean	0.01	0.004	0.003
Millet	0.01	0.0001	0.0001
Tomato	0.02	0.007	0.006

*Typical values; C.R. = [plant]/[soil].

**Soil concentrations based on 8 M HNO_3 extractable.

TABLE 5. EXTRACTION OF U, Th, AND Pu FROM FLOODPLAIN SOIL USING MILD EXTRACTANTS

	U	Th	Pu
Extractant	%*		
1 M HNO_3	73	7.9	7.7
10% Na_2CO_3 - 5% $NaHCO_3$	71	45	54

* % of 8 M HNO_3 extractable U (8.18 μg/g), Th (16.8 μg/g) and Pu (144 dpm/g).

Th are still cationic, while in 8 M HNO_3 or in the strong carbonate solutions, both cations would form negatively-charged complexes which do not readily adsorb. The extraction conditions can change the Pu oxidation state present and therefore, these results only support the plant uptake data which indicated that Pu and Th are likely present in similar forms.

While neptunium has not been studied extensively, plant uptake (Price 1973) and soil adsorption (Routson *et al.*, 1975) studies have indicated higher mobility relative to the rest of the actinides. Bondietti (1976) noted evidence for reduction of Np(V) to Np(IV) in an acid soil. Thus, while Np may be very available to plants if it remains oxidized (Np(V)), if Fe^{2+} or other reductants are present, Np(IV) may be stabilized to some extent. Tetravalent Np would be expected to behave like Pu(IV) (low plant uptake) and attention should be paid to instances where Np uptake is similar to Pu since this may indicate that Np(IV) is being stabilized.

SUMMARY

The question of which Pu oxidation states dominate in the environment has been discussed, using thermodynamic approaches and environmental measurements to illustrate the complexity of the problem. When thermochemical calculations are considered, trivalent Pu appears to be a likely soluble state, particularly under reducing conditions. Penta- and hexavalent states have also been calculated to be important. The oxidation states of Pu in White Oak Lake water appear to be III or IV. While Pu(III) might be present (based on thermodynamic considerations), the observed concentrations are consistent with the presence of Pu(IV). Observed concentrations of soluble Pu in various natural waters do not remotely approach current permissible limits.

Concentrations of soluble Pu exceeding predicted solubilities have been observed in PuO_2 dissolution studies. This phenomenon appears related to complex oxidation state changes as the result of radiolysis; however, the absolute dissolution rate of PuO_2 is very small and some evidence is available to raise the question of the relative importance of chemical solubility vs. physical fragmentation as the important "dissolution" mechanism.

The relative availability of transuranium elements to plants appears related to the oxidation state present in the soil. From the limited data available, the pentavalent oxidation state appears to be more available than the other valences. Plutonium uptake by plants appears to be less than reported for Am, Cm, or U. In one case, the behavior of Pu and Th appeared similar. This would suggest that plant assimilation of these elements should follow the valence order $5 > 6 \approx 3 > 4$. This order is the inverse of the relative tendency of adsorption to soil.

The information presented exemplifies the complexities of evaluating Pu behavior in natural solutions and identifies how further research should be structured to answer important questions of environmental mobility and biogeochemistry of

Pu and the other actinides. It is apparent that Pu is extremely insoluble and thus does not appear to enter food chains readily. No evidence exists to suggest that aging in the environment will change this fact.

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DISTRIBUTION PATTERNS AND TRANSPORT OF PLUTONIUM IN FRESHWATER ENVIRONMENTS WITH EMPHASIS ON PRIMARY PRODUCERS

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ABSTRACT

The major repository for transuranic elements entering aquatic ecosystems is the bed sediment. Observed K_d values for plutonium across a wide spectrum of aquatic systems are surprisingly uniform (on the order of 10^5). Plutonium arrives at the bed sediment as a result of association with and subsequent settling of suspended particulate matter. Consequently, the major role of phytoplankton in plutonium kinetics in aquatic systems has been postulated to be one of plutonium removal from the water column. The very high affinity of plutonium for particulate matter in aquatic systems makes it difficult to use the traditional expression of Concentration Factor (CF) as a measure of the tendency of biota to accumulate this element. A proposed term, Trophic Transfer Factor (TTF), relates concentrations observed in biota to that of the sediment. The underlying assumption is that, due to the high K_d's, accumulation in tissues of organisms at higher trophic levels will be dominated by gut absorption rather than by direct uptake from water. Plutonium in most freshwater systems is transported predominately in association with suspended particulate matter. In some organically rich systems, a fraction of the hydrologically mobile plutonium pool may be complexed or otherwise associated with naturally occurring organic acids. Although this association may result in increased environmental mobility, it does not appear to result in increased availability to aquatic biota.

INTRODUCTION

With the exception of a few site-specific studies, historical information on the behavior of transuranics in aquatic ecosystems is related predominantly to nonpoint-source inputs of high-fired oxides to these systems from the atmosphere. Available information indicates that the concentration of plutonium in tissues of resident biota of these systems is very low. Low concentrations in biota may be related to two factors: 1) solubilization of high-fired oxide is extremely low in natural systems; and 2) apparent trophic transfer factors are expected to be in the range of 10^{-3} to 10^{-6}, based primarily on results of mammalian gavage studies (Weeks et al., 1956; Katz and Weeks, 1954; Buldakov et al. 1967). Higher reported uptake values appear to be related to the presence of gut loading and from surface contamination by sedimentary materials. This explanation seems plausible when one considers the kinetics of transuranic elements in aquatic systems.

The objectives of this paper are to:

1. review the distribution and transport of plutonium in aquatic systems and place the role of primary producers in transport processes in the proper perspective; and

2. suggest terminology for expressing plutonium concentration in aquatic biota that more clearly reflects the functional relationships that exist between abiotic and biotic components of aquatic ecosystems than the term CF (concentration factor) presently in use.

DISTRIBUTION PATTERNS

The major repository of transuranic elements entering aquatic systems is the bed sediment. Observed K_d values for plutonium across a wide spectrum of aquatic systems (both freshwater and marine) are surprisingly uniform and on the order of 10^5 (Table 1). A significant portion of Pu is thought to arrive at the bed sediment surface as a result of association with, and subsequent settling of, suspended particulate matter (Wahlgren et al., 1976). (Pillai and Mathew, 1976). The organic (phytoplankton):inorganic ratio of suspended particulate matter varies, depending on characteristics of the system. For example, the ratio will be lower in flowing water systems and open oceans than in lakes and estuaries. The plutonium associated with inorganic suspended particulate matter may become incorporated in the bed sediment with very low hydrologic mobility. Conversely, the fraction of plutonium reaching the bed sediment associated with the organic suspended particulate matter may then be subjected to long-term biologically and/or chemically mediated transformations resulting in its association with components of sediment that exhibit greater hydrologic mobility, (i.e., chelated, associated with organic matter, complexed with inorganic substances, or soluble). However, the biological availability of plutonium from these two components of bed sediment may not differ significantly. The strength of the association between plutonium and mineral and/or organic components of the sediment may be comparable.

Table I. Distribution Coefficients for Plutonium Isotopes in Freshwater and Marine Systems

K_d	Isotope	Environment*	Reference
9.0×10^4	237	L,F	Trabalka & Eyman, 1976
$1.3\text{-}9.4 \times 10^4$	237	L,M	Duursma & Parsi, 1974
$1.2\text{-}7.9 \times 10^4$	239	L,F	Trabalka & Frank, 1976
$4.8 \times 10^4 - 1.3 \times 10^5$	239	M	Pillai & Mathew, 1975
$\sim 5 \times 10^4$	239	F	Bowen, 1974
3×10^5	239	F	Wahlgren, et al., 1975

*F = Freshwater M = Marine L = Laboratory

MOVEMENT IN SYSTEMS

The major role of phytoplankton in plutonium kinetics in aquatic systems has been postulated to be one of removal of a significant fraction of plutonium from the water column (Wahlgren et al., 1976; Hetherington, 1976). However, collection techniques are such that phytoplankton cannot readily be separated from inorganic suspended particulate matter. The reported plutonium Concentration Factor (CF) values for algae (Figure 1) may, in fact, be high due to the inclusion of inorganic suspended particulate matter which would have a CF value of 10^5. The correlation between percent silicon content and plutonium concentrations in phytoplankton samples (Yaguchi et al., 1974) was attributed to the predominance of diatom frustules in the samples analyzed. Wahlgren et al. (1976) reported a correlation between percent ash weight and plutonium concentrations. They also concluded that plutonium was associated with diatom frustules in the plankton samples. However, in neither case was the contribution of associated inorganic suspended particulate matter to the observed plutonium concentrations qualified. Wahlgren et al. (1976) reported a distribution coefficient for suspended sediment materials of $\sim 3 \times 10^5$. The inclusion of a small amount of those materials in the ash residue of phytoplankton samples may offer a plausible alternative interpretation of the observed correlations.

In any event, that portion of the plutonium observed in phytoplankton appears to be surface-associated. This is the case for giant brown algae studied by Hodge et al. (1974). It is doubtful that active absorption of a type that would lead to significant biomagnification could be possible. This is borne out by available environmental data comparing CF values for algae with those for organisms at higher trophic levels (Figure 1). One would certainly expect, as suggested by Edgington et al. (1976), that surface-to-volume ratios may have some predictive value in determining expected plutonium concentrations of organisms at lower trophic levels, assuming that cross-contamination with sedimentary particulates is eliminated. The physical transport of plutonium in aquatic systems is predominately a result of its association with abiotic components of the system, (Hetherington, 1976). Nevertheless, even though biological transport accounts for a very small fraction of the plutonium pool, it must be given consideration because the potential exists for transfer to man via this ingestion pathway.

FOOD CHAIN TRANSPORT IN AQUATIC SYSTEMS

The very high affinity of plutonium for particulate matter in aquatic ecosystems (distribution coefficient $\sim 10^5$) makes it difficult to use the traditional expression of Concentration Factor (CF) as a measure of the tendency of biota to accumulate this element in tissue. Rather, we believe the observed concentrations of plutonium in aquatic biota should be related to the primary abiotic source in the system, sediment (both suspended and bottom). In order to express this relationship, the term Trophic Transfer Factor (TTF) has been used by various researchers (Lipke,

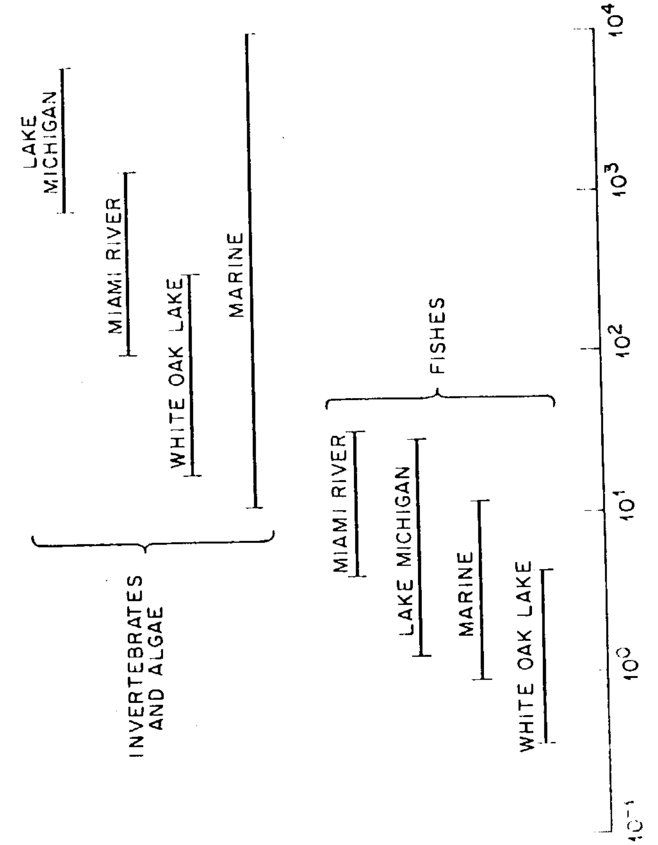

Figure 1. Concentration Factors (CF) for Pu in Various Freshwater and Marine Environments. CF is defined as [Pu] in Organisms (wet weight)/[Pu] in water.

1971; Trabalka and Eyman, 1976; Elwood et al., 1976). This concept substitutes the concentrations of an element in sediment and suspended particulate matter for the concentration in water normally used in the calculation of a CF. The underlying expectation is that, due to the high K_d's, observed element accumulation in tissues of higher trophic levels will be dominated by gut absorption rather than by direct uptake from water. Again, we stress that external contamination with sedimentary particulate matter and gut loading are not considered to represent true uptake and should be considered separately. An example of the utility of the approach can be seen in Figure 2 where TTF is used rather than CF to express transfer of plutonium from abiotic to biotic components in various systems. This figure clearly shows that plutonium is discriminated against in food chains of aquatic systems. TTF values for fishes of ∿ 10^{-3} to 10^{-6} are comparable to those observed in mammalian gavage studies (Baxter and Sullivan, 1972; Carritt et al., 1947; Weeks et al., 1956). This term, then, serves as a realistic measure of plutonium discrimination in food chains. If our hypothesis is correct, (i.e., sediment cross-contamination has contributed a major fraction of observed plutonium concentrations in phytoplankton materials in previous studies) the maximum possible TTF value should be on the order of one. In fact values in Figure 2 are significantly less than one, as expected.

Some of the variation in TTF values observed can be explained by the relative trophic position of the organisms analyzed. The number of intervening food chain transfers between the organism analyzed and the abiotic source of plutonium should be inversely related to the observed TTF value. This is demonstrated in Figure 2 where organisms at lower trophic levels have higher TTF values than fishes. Therefore, in assessing potential transfer of plutonium to man from aquatic ecosystems, it is important to concentrate on those food sources most closely linked to sediment as a measure of maximum plutonium in human food. These would represent a short, single trophic transfer food chain as opposed to the traditional concept of the grazer food chains. Examples of important groups include bottom-feeding fishes, shell fish, and rooted macrophytes such as rice. A major dietary component of a large segment of the world population is rice. Although we could find no data on accumulation of plutonium in rice, this information is important since it is representative of a single trophic transfer from sediment and/or water to man. Noshkin (1972) pointed out that marine organisms associated with the sediment-water interface, (i.e., benthic invertebrates) contain one hundred times higher plutonium burdens than marine free swimming vertebrates.

Benthic invertebrates and rooted macrophytes (*Ludwigia, Elodea,* and *Sagittoria*) from White Oak Lake contained comparable concentrations of $^{239-240}$Pu which were one to two orders of magnitude greater than fish from the lake. Data on fishes from White Oak Lake show a trend of decreased concentrations of $^{239-240}$Pu at higher trophic levels (Table 2). Comparable concentrations in plutonium were observed in the three species which feed on bottom dwelling invertebrates (bluegill and goldfish) or on suspended particulate matter (shad). Stomach content analysis of these three species showed significant amounts of clay material to be

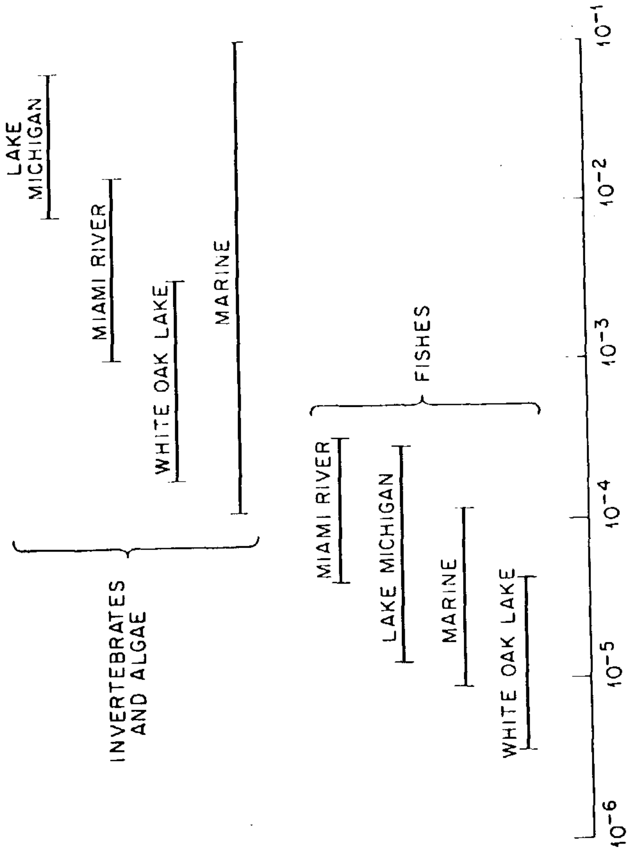

Figure 2. Trophic Transfer Factors (TTF) for Fresh Water and Marine Environments. TTF is defined as [Pu] in Organisms (wet weight)/[Pu] in Sediment or Suspended Particulate Matter (wet weight).

Table II. Concentrations of $^{239,240}Pu$ and Concentration Factors for Fishes from White·Oak Lake

Species	$^{239,240}Pu$ Content				Concentration Factor[c,d]	
	Carcass[a]		G.I. Tract			
	pCi/g	S.E.[b]	pCi/g	S.E.	C.F.	S.E.
Largemouth bass	2×10^{-4}	6×10^{-5}	6×10^{-3}	4×10^{-4}	0.4	0.2
Bluegill	1×10^{-3}	8×10^{-4}	4×10^{-2}	5×10^{-5}	3	2
Goldfish	1×10^{-3}	1×10^{-3}	8×10^{-2}	3×10^{-2}	3	3
Shad	2×10^{-3}	3×10^{-5}	4×10^{-1}	7×10^{-2}	4	0.1

[a] Total fish minus G.I. Tract

[b] Standard Error

[c] Concentration Factor (CF) is defined [Pu] in organisms (wet weight)/[Pu] in water. Water concentration of Pu used in calculation of CF values = 4×10^{-4} pCi/g.

[d] C. F. values for carcass.

present. The largemouth bass, conversely, had no clay material in the stomach contents and the CF value was approximately one order of magnitude lower (0.4 vs 4) than for the other two fish species (Table 2).

Future research on pathways of plutonium to man from aquatic ecosystems should concentrate on those food chains which have the smallest number of trophic transfers between abiotic sources in the system and man. Additionally, emphasis on the relationship between the chemical characteristics of plutonium in abiotic components of the system and availability for short food chain transport to man should be stressed.

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PLUTONIUM-237: AN IMPORTANT RESEARCH TOOL FOR STUDIES OF ENVIRONMENTAL PLUTONIUM KINETICS

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ABSTRACT

The production and use of the photon-emitting isotope plutonium-237 in investigations of the uptake, retention, and distribution of monomeric plutonium (IV) both in an aquatic vertebrate, the channel catfish *Ictalurus punctatus*, and in a littoral aquatic micro-ecosystem, are presented. The rationale for use of plutonium-237 in environmental studies is discussed.

Chelation can either enhance or reduce the uptake of ingested plutonium relative to plutonium hydroxide (monomer) in channel catfish. The highest observed retention (whole body) at 63 days was 3.8% of ingested dose for plutonium-237 citrate, while retention of the fulvate was 0.6%. Reduced uptake of the fulvate complex is due either to its high molecular weight (> 10,000) or to its stability in metabolic systems. Increased uptake of plutonium-237 citrate is attributable to instability of the complex in metabolic systems.

Tissue distribution studies revealed that relatively little ($\leq$ 10%) of intracardially injected plutonium citrate was excreted. Blood clearance rates were similar to those found in small mammals, with the plutonium being primarily associated with the plasma protein transferrin. The fractional body burdens in bone, liver, and kidney 17 days after injection were 31%, 24%, and 9% of the injection dose, respectively. High kidney burdens relativy to mammals are expected, since the kidney functions as the major site of homopoiesis in teleosts. Absence of significant excretion indicates that a short half-life component of elimination following gut clearance in gavage studies is due to plutonium labeling of the gut.

A distribution coefficient of 9×10^4 was observed for sediment in a year-old aquatic microcosm spiked with plutonium-237 nitrate. A materials balance at 90 days post-spike provided the following estimates: 0.001% in water, 0.04% in biota, and over 99.9% in sediments. Concentrations in whole animals including fish were surprisingly uniform (within a factor of 10, 1.2-9.9% of mean sediment concentration). This was related to gut loading of sediments and/or surface contamination. The uptake by rooted macrophytes not exposed to surface contamination was quite small: ≤ 0.03 to 0.1%.

Sorption of plutonium to plant surfaces, on gut walls, and on exoskeletons appears to dominate in submerged components of aquatic systems. Results of Pu behavior are discussed in terms of ecological significance and potential for transfer to man.

INTRODUCTION

There are three important reasons why we have used the plutonium-237 isotope so extensively in our laboratory work involving aquatic organisms.

Firstly, plutonium-237 is predominantly a photon emitter-100 Kev-complex x-rays accompany 45% of all decays while α-emissions occur in only 3 out of every 100,000 transitions (Lederer et al.,1967). Thus, one is able to do repeat counting of individually-tagged organisms in an uptake experiment. Since destructive analytical techniques are not required, one can follow uptake and elimination patterns in some organisms over the entire lifetime. Even where destructive sampling is required (as in tissue distribution studies, for example), analytical procedures are both simple and inexpensive. One is also able to reduce the sample size required for a given end point since individual variability as a function of time is determined in uptake and elimination work.

Secondly, plutonium-237 has a high specific activity - approximately 1×10^4 Ci/g in our product. Thus, µCi concentrations of plutonium-237 contain the same concentrations of plutonium atoms as pCi levels of plutonium-239. Therefore, we are able to work with reasonably high activity levels of plutonium-237 to reduce radioanalytical error and at the same time maintain plutonium atom concentrations which closely mimic a "real-world" situation. This is an important requirement in terms of the environmental chemistry of the element.

Thirdly, plutonium-237 has a relatively short physical half-life of 45.6 days; and, as noted previously, negligible α-activity. Thus, the investigator can avoid the enormous costs associated with use of double-containment facilities required for reasonable quantities of plutonium-239 or plutonium-238. One has essentially the same freedom in experimental design as if the radionuclides were Zn-65, or Cs-137, or Ce-144, for example.

PRODUCTION AND CHARACTERISTICS

Plutonium-237 is produced by helium ion bombardment of ^{235}U. Trace quantities of plutonium-236 provided the only significant contamination of the material we received. Table 1 illustrates the physical characteristics of plutonium-237 for comparison with α-emitting isotopes (Eyman et al.,1976). The characteristics for plutonium-246 are included since it represents an alternative photon emitter for use in plutonium research. Its disadvantages are its very short physical half-life and low specific activity (as a consequence of contamination with its parent isotope, ^{244}Pu). Although we did undertake some early work involving plutonium-246, we would not recommend it for future aquatic environmental studies because its specific activity is unrealistically low.

TABLE 1. COMPARATIVE PHYSICAL CHARACTERISTICS OF GAMMA EMITTING AND ALPHA EMITTING ISOTOPES OF PLUTONIUM

ISOTOPE	HALF-LIFE	PHOTON ENERGY (MEV)	SPECIFIC ACTIVITY (CI/G)
^{237}Pu	45.6 D	.100 COMPLEX	1.1×10^4
^{246}Pu	10.85 D	.225	0.27^A
^{238}Pu	86 Y	α	17.48
^{239}Pu	2.44×10^4 Y	α	0.06
^{244}Pu	8.3×10^7	α	1.77×10^{-5}

^{A}LOW SPECIFIC ACTIVITY DUE TO CONTAMINATION WITH ^{244}Pu.

EXPERIMENTAL METHODS AND RESULTS

Three specific experiments are described below which illustrate the usefulness of plutonium-237 as an environmental research tool.

Gavage Study

We conducted an experiment whereby channel catfish received a dose of plutonium-237 incorporated in a diatomaceous earth slurry and administered by intragastric injection (gavage) (Eyman et al.,1976). The purpose of this experiment was to examine the effect of chemical form on the gastrointestinal uptake of plutonium by a fresh-water fish. The choice of fish species was determined by three factors: (1) Its importance as a food species for man; (2) its propensity to ingest significant quantities of sediment along with its diet; and (3) its prevalence in the southeastern U.S. where significant facilities associated with the Breeder Reactor technology are to be located. The choice of chemical forms to be incorporated in the diatomaceous earth slurry was such that we had a representative sedimentary form (fulvate) and two reference forms (citrate and hydroxide) for comparison both with mammalian results and with our previous data. Doses contained approximately 20 nCi of plutonium-237/fish (represents a mass equivalent to approximately 1 pCi of plutonium-239). The average weight of specimens tagged was 57 g. Fish were whole-body counted up to 90 days at time intervals which gradually increased from 1 day to 30 days. Retention of plutonium was expressed as a percentage of the initially measured whole-body burden.

Elimination coefficients were calculated by separating different components of the whole-body retention curve (Richmond, 1958). The initial rapid loss component (K_{b1}) represents that portion of the ingested dose that passed through the animal and was not assimilated. The half-time for gut clearance ranged from 0.51 to 1.17 days (Fig. 1). Elimination of the fraction of the ingested dose that was absorbed and/or associated with the gut wall was separated into two components. The calculated elimination coefficients for the second component were comparable for Pu-fulvate and Pu-hydroxide while citrate (0.0238 day^{-1}) was significantly different ($p < 0.05$) from the other two forms (Table 2). The biological half-lives of this component for the three treatments ranged from 13.4 to 30.2 days. Extrapolation of the second component to day zero was used to estimate the percent uptake of the ingested dose. Uptake ranged from 1.62% for Pu-fulvate to 10.5% for Pu-citrate (Table 2). Calculated uptake of Pu-fulvate was significantly lower ($p < 0.05$) than either of the remaining treatments. Between 50 and 70 days post-ingestion, elimination was dominated by a third, long-term component with a calculated half-life of 534 days. The observed retention of ingested dose on day 90 was 3.8% and 0.5%, respectively, for Pu-citrate and Pu-fulvate. These values are significantly greater than observed in studies with mammals (0.09-0.30%, Pu(IV) citrate; 0.001-0.01%, Pu(IV) nitrate) (Baxter and Sullivan, 1972; Carritt et al.,1947; Weeks et al.,1956).

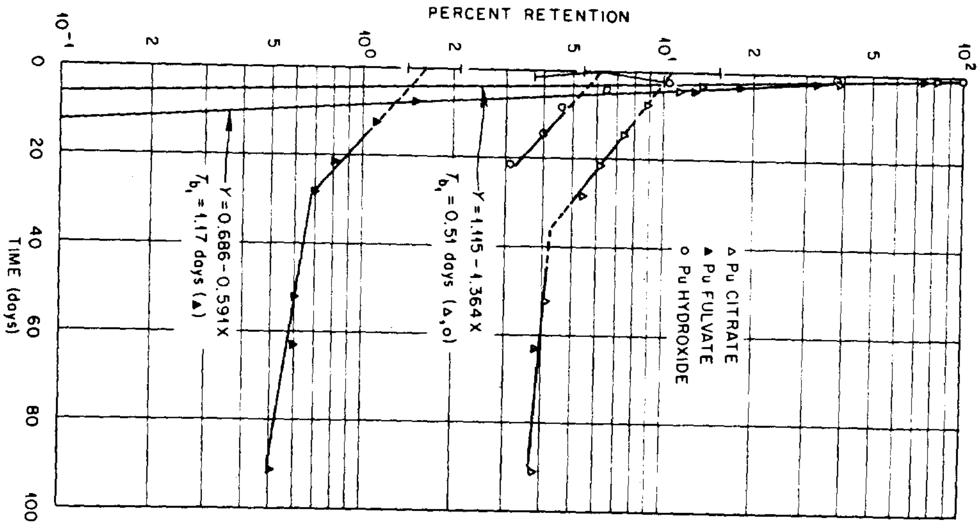

Figure 1. Percent Retention of Ingested ^{237}Pu in the Citrate, Fulvate, and Hydroxide Forms by Channel Catfish.

Table 2. **Biological half-life (T_b), elimination rate (K_b) and percent uptake of ingested ^{237}Pu in various chemical forms by channel catfish (*Ictalurus punctatus*)**

Values are mean ± 1 S.E. and coefficient of determination (r^2)

Chemical Form	N	K_{b_2} (days^{-1})	$T_{b_2}^a$	r^2	% Uptake	K_{b_3} (days^{-1})	Range of K_{b_3}	T_{b_3}
^{237}Pu-citrate	6	−0.024 ± 0.005	30.2 ± 2.8	0.99	10.5 ± 2.57	−0.0013	0–0.0053	534
^{237}Pu-fulvate	5	−0.036 ± 0.016	24.3 ± 4.5	0.97	1.62 ± 0.18			
^{237}Pu-hydroxide	5	−0.036 ± 0.018	13.4 ± 1.7	0.99	6.17 ± 1.39			

$^a T_b$ = days

Additional studies on tissue distribution have shown that after 30 days ∿ 50% of the body burden is associated with the eviscerated whole fish while the remainder is located in and/or on the gastrointestinal tract. Of the fraction in the carcass approximately two-thirds is associated with non-edible material (bone, etc.). Therefore, ∿ 17% of the carcass burden (0.6%-0.1% of ingested dose for Pu-citrate and Pu-fulvate, respectively) resides in edible tissue and may be available for transfer to man from this source.

From these results we conclude that chelation can either enhance or reduce uptake of plutonium in the channel catfish. Plutonium citrate penetrates the gut membrane due to a net negative charge of the complex (Cleveland, 1970). Reduced uptake of Pu-fulvate may be attributable to its stability in the presence of digestive systems and high molecular weight (> 10,000).

Theoretically, soluble plutonium, bound to dissolved humic derivatives (yellow acids), may be hydrologically mobile. However, if such complexes are stable in biological systems (microflora, gastrointestinal tract), then hydrologic mobility will not necessarily enhance the availability of plutonium for entry into biological systems. An increased emphasis on the speciation of Pu which accidentally may be released to surface waters is needed to provide information on its aqueous behavior and to evaluate the availability of the various chemical and physical forms for biological intake.

Intracardial Injection Study

In a companion experiment, we administered plutonium-237 citrate to yearling channel catfish (mean weight 78 g) via intracardial injection (Eyman et al.,1976). Because plutonium is associated primarily with sediments in aquatic systems (distribution coefficient, K_d, approximately n x 10^4), we had conducted experiments to determine the uptake of plutonium by sediment-feeding organisms. We had expected that the second component of elimination in gavage studies would have a very long biological half-life measured in years rather than days. In fact the third component of elimination did fulfill this expectation. We questioned whether the relatively short second elimination component represented tissue absorption from the gut and subsequent excretion from a highly labile pool, or rather the labeling of cells in the gut wall and sloughing of cellular materials as part of the process of gut renewal. This next experiment was designed to answer the questions raised in our gavage studies and also to provide needed information on the tissue distribution of plutonium in an important aquatic food organism of man.

Each fish was given an injection of 7.5 nCi ^{237}Pu citrate in a 0.85% saline solution (0.5% citrate). Animals were kept in 400-liter "living streams" at 25°C as in gavage studies. Fish were whole body counted until sacrifice at 1, 2, 3, 10, and 17 days post-injection. Blood samples were taken prior to sacrifice (heparinized syringes). Plasma proteins were separated by polyacrylamide gel-electrophoresis and the gels were sectioned for counting. Individual tissues were dissected for radioassay and a materials balance (fractional body burden determination) on injected plutonium was performed.

Plutonium-237 distribution and translocation in channel catfish following intracardial injection are basically similar to patterns in mammals after intravenous administration. The fractional body burdens in bone, liver and kidney 17 days after injection were 31%, 24%, and 9% of the injected dose, respectively. High kidney burdens relative to mammals are expected, since the kidney functions as the major site of hemopoeisis in teleosts. Blood clearance rates were similar to those found in small mammals (Fig. 2), with the plutonium being primarily associated with the plasma proteins. Approximately 70% of the plasma-associated plutonium was bound to the iron-transport β-globulin, transferrin. Addition of excess iron to the plasma samples released 42% of the protein bound plutonium in 2 hr. Note also that concentrations of plutonium-237 (expressed as % dose/gram) were highest in kidney and liver. Excretion over the 17-day period was ≤ 10%. The absence of significant excretion indicates that a short half-life component of elimination following gut clearance in gavage studies is due to plutonium labeling of the gut.

Microcosm Study

In another experiment, we spiked a year-old balanced aquatic microcosm including fish (Fig. 3) with 11 μCi of plutonium-237 nitrate (Trabalka and Eyman, in press). At 90 days post-spike, the microcosm components were intensively sampled. The microcosm contained 12 l of water and 6 l of sediments in a large, shallow plastic photographic tray (0.2 m^2 surface) kept in an environmental chamber. We felt a study of plutonium distribution in a microcosm represented the next logical step beyond single organism experiments. The microcosm appears to be a useful tool in ecological studies of highly toxic materials such as plutonium. Aquatic microcosms over a wide range of size and complexity have been used as model ecosystems to study the fate of radionuclides in aquatic ecosystems (Whittaker, 1961; Duke et al.,1969; Wilhm, 1970; Cross et al., 1971; Cushing and Watson, 1973; Trabalka, 1971; Short et al.,1973). Due to the limited availability of plutonium-237, it is not feasible to use the isotope in field studies. Thus, the microcosm experiment allows us to obtain some limited answers to questions which we would pose in field studies. A most important question is whether there are mediating factors at the system level which alter the potential availability to organisms in the system.

A distribution coefficient of 9 x 10^4 was observed for sediment. A materials balance at 90 days post-spike provided the following estimates: 0.001% in water, 0.04% in biota, and over 99.9% in sediments. Concentrations in whole animal including fish were surprising uniform (within a factor of 10, ranging from 1.2-9.9% of mean sediment concentration).

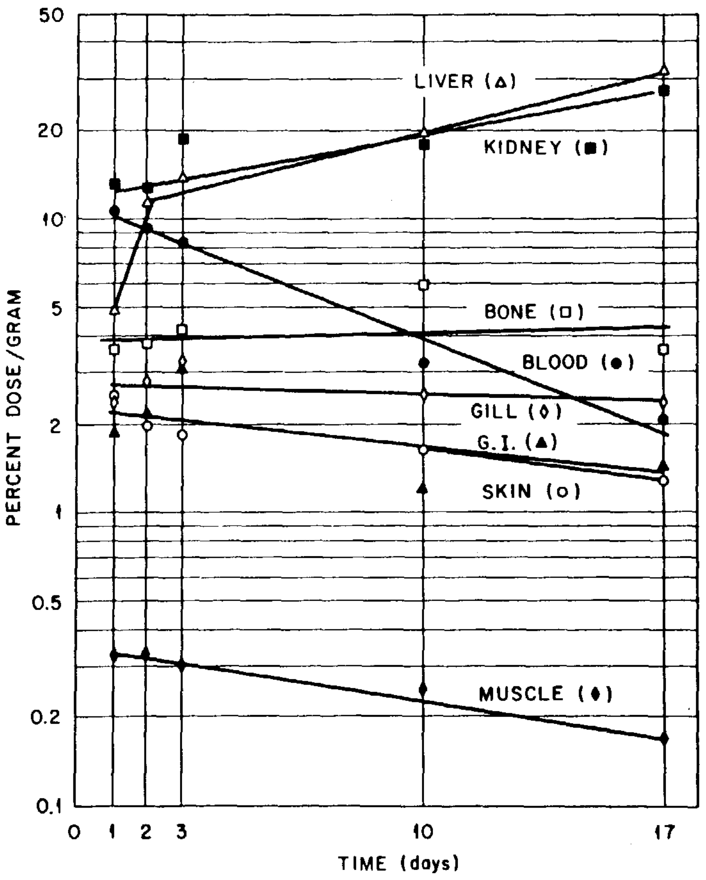

Figure 2. Tissue and Organ Concentrations of ^{237}Pu Administered to Channel Catfish as the Citrate via Intracardial Injection.

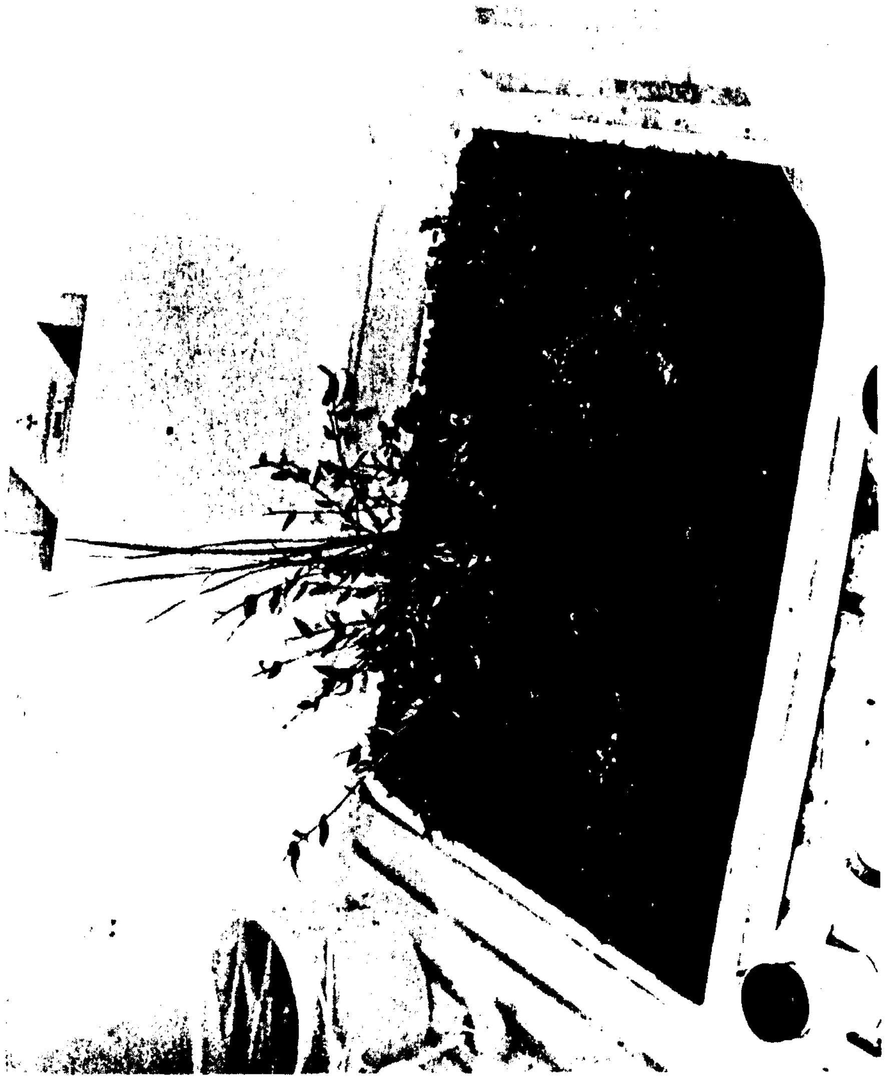

Figure 3. A Littoral Freshwater Microcosm Contaminated with ^{237}Pu

A Trophic Transfer Factor equals the concentration in an organism expressed as a percent of the mean sediment concentration. This was related to gut loading of sediments and/or surface contamination (Table 3). In general, snails exhibited highest concentrations in shells and lost 50% of their plutonium burden when held for 48 hours. The importance of the gut contribution is best shown in the goldfish data. Plutonium concentrations are nearly an order of magnitude less when the gut is isolated. The uptake by rooted macrophytes not exposed to surface contamination was quite small: ≤ 0.03 to 0.1%. The comparison between emergent plants sampled above the waterline, and submerged plants was very instructive in this regard. Concentrations were many orders of magnitude higher in submerged plants; sorption to surfaces is strongly indicated.

CONCLUSIONS

We believe our results have shown the utility of ^{237}Pu as a laboratory tool to shed light on some of the fundamental processes occurring in natural aquatic systems. It is equally obvious that it cannot be used to answer questions relative to long-term time-dependent phenomena. Specific plutonium-contaminated aquatic ecosystems must be preserved as research sites to answer those important questions. Environmental studies carried out in a natural ecosystem will ultimately be required to validate results of laboratory studies.

Our results suggest that surface phenomena at plant surfaces, on gut walls, and on exoskeletons appear to dominate in submerged components of aquatic systems. This is a partial explanation for the higher plutonium levels found in biota from aquatic versus terrestrial environments as reported in reviews on the subject. We also note that trophic transfer factors for sediment-associated animals in the microcosm study are comparable to the percentage uptake of plutonium in our gavage work with channel catfish.

Our results also suggest that surface phenomena also may predominate in terrestrial systems where significant surface contamination occurs either by atmospheric processes or as a result of periodic immersion of floodplains, for example. A major question not addressed by our studies to date would appear to be whether biochemical transformation to a more biologically available form of plutonium could occur over a longer time interval.

ACKNOWLEDGMENTS

We would like to thank J. W. Gooch and C. P. Allen for technical support in gavage studies. We would particularly like to single out M. L. Frank for work involving plasma protein separation and identification. Finally, H. L. Bergman provided significant input regarding physiological behavior of blood-transport proteins and assistance in tissue preparation for radioassay.

Common name	Genus	Body Part	Sample size	Trophic transfer factors
		Emergent plants		
Grass	*Panicum*	Spikes	11	≤0.11
		Stem and leaves	11	≤0.045
Cattail	*Typha*	Stem and leaves	1	≤0.029
		Stem and leaves	1	≤0.040
Watercress	*Nasturtium*	Stem and leaves	7	≤0.089
		Submerged plants		
Algae	*Oedogonium*	Clumps	7	9.1
Moss	*Hygrohypnum*	Branches	60	27.0
Stonewort	*Chara*	Branches	75	0.19 ± 0.078^{a}
Pondweed	*Potamogeton*	Stem and leaves	30	2.5
		Invertebrates		
Snails	*Physa*	Whole body	30	2.4 ± 0.46
		Whole body^{b}	30	1.5 ± 0.29
		Shells^{b}	30	1.9 ± 0.59
		Carcasses^{b}	30	1.2 ± 0.50
	Gyraulus	Whole body^{b}	63	6.4 ± 1.4
	Goniobasis	Whole body	3	7.4
		Whole body	3	9.9
		Whole body^{b}	5	5.0
		Whole body^{b}	16	5.1
		Shells^{b}	16	5.8
		Carcasses^{b}	16	3.5
Amphipod	*Hyalella*	Whole body	130	3.6 ± 0.97
		Vertebrate		
Goldfish	*Carassius*	Carcass-gut	4	0.47^{c} (0.16, ≤0.54, ≤0.60, 1.4)
		Gut	4	30 (18-91)
		Whole body	4	2.3 (1.2-8.1)

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[a] 95% C.I. (counting error). Not shown if less than 10% of mean.

[b] Animals held for 48 hr in uncontaminated spring water before sacrifice.

[c] Geometric means and ranges. Mean for carcasses based on statistically significant values only.

Table 3. Trophic Transfer Factors for Plutonium-237 in Biota of an Aquatic Microcosm.

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DISTRIBUTION OF ^{238}Pu AND ^{239,240}Pu IN AQUATIC MACROPHYTES FROM A MIDWESTERN WATERSHED

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ABSTRACT

Aquatic macrophytes were collected in the Great Miami River, Ohio, above and below Miamisburg and in the canal and ponds, near the Mound Laboratory, which contain sediments of a high activity (~10^3—10^6 times) relative to the river sediments. Macrophytes collected in the river below Miamisburg have higher activities of ^{238}Pu than those collected from above the city. Macrophytes from the canal and ponds contain high specific activities of ^{238}Pu and ^{239,240}Pu with the exception of cattails grown in the ponds.

Concentration factors are reported and discussed with reference to possible modes of plutonium accumulation and distribution within the plants.

INTRODUCTION

In following the biological pathways of plutonium in aquatic ecosystems, it is necessary to examine the primary producers. The concentrations at this level indicate the trophic availability of plutonium to higher levels in food chains. It has been shown that aquatic plants accumulate more plutonium than do successive higher trophic levels (Emery and Klopfer, 1976; Wayman _et al_., 1974; Yaguchi _et al_., 1973a).

The Great Miami River watershed was chosen as the site of plutonium studies because in addition to atmospheric fallout containing the plutonium isotopes which are primarily ^{239}Pu and ^{240}Pu, ^{238}Pu is released to the river in low level radioactive waste effluent from Mound Laboratory. Located adjacent to the river and Mound Laboratory are two small ponds and a canal which contain elevated levels of ^{238}Pu in the water and sediment as a result of a past incident at the Mound Laboratory (Rogers, 1975). Preliminary examinations of the plutonium activities in the aquatic biota from the river, ponds and canal have been reported (Wayman et al., 1974; 1975). This paper studies in more detail the plutonium activities in aquatic plants from these sites with emphasis on macrophytes. Possible modes of plutonium accumulation and distribution within certain plants will be discussed. There is a brief comparison of plutonium activities in the same species of Cladophora present in two adjacent dissimilar sites, the Great Miami River and the adjacent canal.

MATERIALS AND METHODS

The aquatic plants investigated in this report are the green alga, Cladophora, and the macrophytes, Lemna (duckweed), Typha (cattail), Potamogeton (pondweed), Myriophyllum (water milfoil). Lemna is a small floating plant consisting of fronds and roots which hang down from the underside of the leaves. Suspended in the water, the roots extract nutrients and are never in contact with consolidated sediment. The plants prefer hard water and grow abundantly to form interwoven masses in still waters of lakes, ponds and quiet areas of large rivers (Fassett, 1957). Lemna was found flourishing in the canal and small patches were observed in the ponds; however, it was not found in any quantity in the river. Also growing in the canal and ponds was the emergent macrophyte, Typha, which has complex rhizoid root systems extending away from the primary plant and penetrating deeply into the sediment. Some waterfowl and mammals are known to feed on these emergent plants (Reid, 1961).

The only aquatic macrophytes found in the Great Miami River were Potamogeton and Myriophyllum. These plants are submergent species which are rooted into the river bed. Both species have long stems with fine feathery leaves. The plants prefer flowing water which allows the stems and leaves to be continually extended in the water column for maximum gas exchange and nutrient uptake. The submergents are food for waterfowl, mammals and fish and provide substrata for periphyton.

All plants were collected by hand and rinsed to remove excess sediment and detritus. Roots were detached from Typha (except one sample), Potamogeton and Myriophyllum. Samples were immediately frozen and stored until the time of analysis. Just prior to analysis samples were thawed and wet weights determined, the excess water having been eliminated in the field. Next, the samples were dried to a constant weight at 105°C and ashed at 500°C. Radiochemical analysis was performed on aliquots of ashed samples following the procedures described by Nelson et al. (1973).

RESULTS AND DISCUSSION

The concentration of plutonium in sediment from the Great Miami River, Mad River (a tributary of the Great Miami River located upstream of

Miamisburg), ponds and canal are listed in Tables 1 and 2. Mean concentrations of plutonium in the water corresponding to the above sites have also been determined (Table 3).

The activity of ^{238}Pu in sediment and water collected downstream of the Mound Laboratory is one to three orders of magnitude higher than it is in upstream background samples, while the water and sediment in the canal and ponds have three to six orders of magnitude greater ^{238}Pu activities than background. Aquatic plants (Tables 4 and 6) whether rooted in sediment, attached to substrata or free-floating generally exhibit high plutonium activity in plant tissues when there are high concentrations in the water and sediment.

There are two to three orders of magnitude higher ^{238}Pu activities in aquatic plants from sites below the Mound Laboratory effluent pipe than in those collected upstream. Plants from Chautauqua site (1.6 miles below pipe) have two to ten times more ^{238}Pu than samples collected at Franklin (6 miles below pipe). ^{238}Pu activity in sediment, water and biota diminishes with distance from the source, while the ^{239,240}Pu activity in aquatic plants from upstream and downstream of Mound Laboratory shows no apparent trend.

Concentration factors for aquatic plants were determined for plutonium activities in both sediment, $C.F._{sed}$, and water, $C.F._{H_2O}$. All $C.F._{sed}$ for aquatic plants are low (Tables 5 and 6). The lowest values were calculated for aquatic plants from the ponds and canal. The $C.F._{H_2O}$ for pond and canal plants are also lower than the values for river plants (Tables 5 and 6).

In the river, Cladophora accumulates 5—10 times more plutonium than the macrophytes, Potamogeton and Myriophyllum. In Lake Michigan, Cladophora contains four times more plutonium than Potamogeton (Yaguchi et al., 1973b). The concentration factors ($C.F._{H_2O}$) for ^{239,240}Pu in the river for Cladophora are in general agreement with values for Lake Michigan (Yaguchi et al., 1973a). The $C.F._{H_2O}$ for algae in the river is an order of magnitude higher than the $C.F._{H_2O}$ for algae in the canal (Table 5). This may indicate that the Cladophora growing in elevated plutonium levels of the canal water have all the surface sites occupied by ^{238}Pu and therefore the apparent $C.F._{H_2O}$ is lower than in the river where active sites still exist. High saturation favors surface adsorption of plutonium rather than active uptake. For example, brown algae are known to concentrate activity on their outer surfaces (Hodge et al., 1973). The ^{238}Pu $C.F._{H_2O}$ for Cladophora in the canal are comparable to the values for Cladophora in the Hanford waste pond which has a similar concentration of ^{238}Pu in water (Emery and Klopfer, 1976).

The activities of ^{238}Pu in the aquatic plants from the canal and ponds (Table 6) are much higher than the activities in plants from the river due to elevated

Table 1. Activity (fCi/g ash)$^{(a)}$ of ^{238}Pu and ^{239,240}Pu in the sediments of the Great Miami River above and below Miamisburg, Ohio.

Sample	Ash Wt / Dry Wt	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu
Rip Rap Bridge	0.90	0.6 ± 0.2	6.6 ± 0.6	0.09
Rip Rap Bridge	0.99	0.5 ± 0.2	1.0 ± 0.2	0.50
Mad River— Stanley Avenue	0.91	2.0 ± 0.4	6.5 ± 0.6	0.31
Mad River— Stanley Avenue	0.93	0.9 ± 0.2	7.1 ± 0.6	0.13
Mad River— Medway Road	0.92	2.0 ± 0.4	6.0 ± 0.7	0.33
0.8 km S. Chautauqua Bridge	0.88	760 ± 40	<7	>109
Franklin	0.99	80 ± 10	<6	>13

(a) The ± value is 1 σ counting error. When the counting error is >100%, the concentration is recorded as <2 σ.

Table 2. Activity (pCi/g ash) of ^{238}Pu and ^{239,240}Pu in the sediments of the canal and ponds near Mound Laboratory.

Sample	Ash Wt / Dry Wt	^{238}Pu	^{239,240}Pu
North Pond			
A	0.93	0.19 ± 0.02	≤ 0.002
B	0.94	0.15 ± 0.02	≤ 0.002
C	0.93	0.24 ± 0.02	≤ 0.006
Sediment Core Canal	0.93	480 ± 7	--
North Canal			
A	0.95	92.0 ± 2.7	--
B	0.96	91.0 ± 2.8	--
C	0.96	89.5 ± 2.8	--

Table 3. Mean concentrations (fCi/l) of plutonium in water at various sites in the Great Miami River watershed, Ohio.

Sites	^{239,240}Pu	^{238}Pu
Mad River–Stanley Avenue	0.35	1.0
Rip Rap Bridge	0.35	0.35
0.8 km S. Chautauqua Bridge	0.35	5.0
Franklin	0.35	2.5
North Canal	ND	1000
North Pond	ND	350
South Pond	ND	1000

ND = Not detectable.

Table 4. Activities (pCi/kg dry wt)(a) of ^{238}Pu and ^{239,240}Pu in aquatic plants from the Great Miami River above and below Miamisburg, Ohio.

Sample	^{238}Pu	^{239,240}Pu	% Dry/Wet
Cladophora sp.			
Rip Rap Bridge (8)(b)	0.39 ± 0.25—6.3 ± 1.2	1.22 ± 0.21—10.1 ± 2.1	9.8—22.7
0.8 km S. Chautauqua Bridge (3)	438 ± 10—2,400 ± 30	6.7 ± 1.7—23.3 ± 9.5	18.0—24.7
Franklin (3)	208 ± 6—830 ± 17	3.2 ± 1.5—16 ± 2	17.0—19.4
Potamogeton sp.			
Mad River—Stanley Avenue (3)	<0.26—0.69 ± 0.18	0.87 ± 0.17—1.6 ± 0.2	8.8—9.6
0.8 km S. Chautauqua Bridge (2)	353 ± 4, 387 ± 5	2.1 ± 0.4, 2.6 ± 0.4	8.0, 8.6
Franklin (3)	28.8 ± 1.1—95.9 ± 4.5	0.50 ± 0.16—2.7 ± 0.8	2.1—9.0
Myriophyllum sp.			
Franklin (2)	34.3 ± 0.9, 58 ± 3	0.50 ± 0.12, 1.33 ± 0.53	7.0, 7.3

(a) The ± value is 1 σ counting error. When the counting error is >100%, the concentration is recorded as <2 σ.

(b) Number in parentheses indicates the quantity of samples.

Table 5. ^{238}Pu and ^{239,240}Pu concentration factors for aquatic plants in the Great Miami River above and below Miamisburg, Ohio.

Sample	C.F. $_{H_2O}$(a)		C.F.$_{sed}$(b)	
	^{238}Pu	^{239,240}Pu	^{238}Pu	^{239,240}Pu
<u>Cladophora</u> sp.				
Rip Rap Bridge (8)(c)	220—3500	340—28800	0.58—9.4	0.16—1.4
0.8 km S. Chautauqua Bridge (3)	15800—103000	4100—16400	0.65—3.6	>1.1—>3.8
Franklin (3)	14200—59300	1800—7800	2.6—10.3	0.54—2.7
<u>Potamogeton</u> sp.				
Mad River—Stanley Avenue (3)	>30—70	230—430	>0.17—0.46	0.12—0.23
0.8 km S. Chautauqua Bridge (2)	5870, 6430	500, 620	0.41, 0.45	0.26, 0.32
Franklin (3)	700—3500	>90—690	0.36—1.2	0.08—0.45
<u>Myriophyllum</u> sp.				
Franklin (2)	1000, 1600	100, 270	0.42, 0.73	0.08, 0.22

(a) C.F.$_{sed}$ calculated as dry weights of samples and sediments.

(b) C.F.$_{H_2O}$ calculated as wet weight of sample.

(c) The number in parentheses indicates the quantity of samples.

Table 6. Activity (pCi/kg dry wt)(a) of ^{238}Pu and ^{239,240}Pu in plants from the ponds and canal near Mound Laboratory.

Sample	^{238}Pu	^{239,240}Pu	% Dry/Wet	^{238}Pu	
				C.F.$_{H_2O}$(b)	C.F.$_{sed}$(c)
<u>Cattails</u>					
North Pond					
whole (minus root)	11.7 ± 0.8	<0.33	16.2	5.4	0.06
North Canal					
whole (minus root) (2)(d)	14.3 ± 0.9, 59 ± 3	<0.7, 2.1 ± 0.4	12.6, 19.8	2.8, 7.4	$(0.14, 0.62) \times 10^{-3}$
North Pond					
spikes	5.4 ± 0.5	<0.35	19.5	3.0	0.03
stems	8.5 ± 0.7	0.33 ± 0.20	18.1	4.4	0.04
North Canal					
spikes	41.7 ± 2.5	not detectable	33.6	14.0	0.08×10^{-3}
stems	380 ± 20	not detectable	25.3	96.0	0.73×10^{-3}
roots with sediment	$5 \times 10^5 \pm 2 \times 10^3$	2400 ± 200	32.3	1.7×10^5	~1
South Canal					
tips	6.7 ± 0.4	0.41 ± 0.20	20.4		
stems	30.7 ± 1.3	<0.2	16.8		
<u>Grass</u>					
North Canal					
plants minus roots (2)	47.6 ± 2.8, 65.2 ± 3.6	0.39 ± 0.29, not detectable	10.5, 11.2		$(0.09, 0.12) \times 10^{-3}$
<u>Duckweed</u>					
North Canal (5)	8,300 ± 100 to 52,500 ± 600	35 ± 8—152 ± 30	4.5—10.2	600—2900	
South Pond	5,000 ± 200	9 ± 9	7.7	400	
<u>Cladophora</u>					
North Canal	11,000 ± 300	30 ± 15	8.1	910	0.02
South Canal	97,500 ± 900	190 ± 40	11.4		

(a) The ± value is 1 σ counting error. When the counting error is >100%, the concentration is recorded as <2 σ.

(b) Calculated on wet weights on sample.

(c) Calculated on dry weights of both sample and sediment.

(d) The number in parentheses indicates the quantity of samples.

levels in sediment and water (Table 2). Cladophora and Lemna contain approximately the same level of ^{238}Pu, which is the highest seen for all biota from the watershed. Lemna sampled from South Pond is lower in ^{238}Pu activity than that collected from the canal which reflects the differences in activities between the two bodies of water (Table 3). It is suspected that the roots of Lemna which are suspended in the water column would be a route for plutonium uptake as it is for nutrients. However, a more plausible explanation at this time is again surface adsorption due to the high surface to volume ratio of the small plants.

Grass is included in Table 6 with aquatic plants because it was growing in the sediment of a dry canal bed. Analysis of the plants minus roots gives an indication of plutonium translocation within the plant. Comparing the levels of plutonium in the canal sediment (Table 2), with the activity in the grass ($C.F._{sed}$), it appears that little transfer has occurred from the sediment to the stems. Grass has higher ^{238}Pu activity than the emergent, Typha, but the same $C.F._{sed}$. Typha also rooted in the canal sediment, shows little active uptake above the roots. However, it appears that the stems have more ^{238}Pu activity than the spikes or flowering parts (Table 6). The $C.F._{H_2O}$ for ^{238}Pu in Typha from the canal is one to two orders of magnitude lower than the $C.F._{H_2O}$ for the Typha from the Hanford waste pond (Emery and Klopfer, 1976).

SUMMARY AND CONCLUSIONS

The aquatic plants from the Great Miami River have higher levels of ^{238}Pu downstream of Mound Laboratory than upstream. Cladophora concentrates plutonium more than the macrophytes, Potamogeton and Myriophyllum, which exhibit similar but lower total plutonium activities.

Cladophora and macrophytes, Lemna and Typha, from the canal and ponds have elevated levels of ^{238}Pu which is attributed to the higher levels of ^{238}Pu in the sediments and water of the canal and ponds. Cladophora from the canal has higher total ^{238}Pu activity than Cladophora from the adjacent river; however, the canal algae have lower concentration factors. This may be indicating a saturation of surface sorption sites as opposed to active uptake by the plants.

Typha and grass (grown in a dry canal bed) demonstrate that there is very little translocation of ^{238}Pu above the roots. Some differentiation, however, exists in Typha with stems having slightly higher ^{238}Pu activities than spikes.

Primary producers in an aquatic ecosystem are capable of scavenging and accumulating plutonium from the surrounding water and sediments to levels which are the highest observed in all biota. Not only is this an important consideration in the study within aquatic food chains, but it may be significant in providing an additional and possibly more direct route to man via the consumption of waterfowl.

ACKNOWLEDGMENTS

The authors wish to thank W. H. Westendorf, D. G. Carfagno and R. Robinson of Mound Laboratory for their cooperation.

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^{238}Pu AND ^{239,240}Pu DISTRIBUTION IN FISH AND INVERTEBRATES FROM THE GREAT MIAMI RIVER, OHIO

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ABSTRACT

Samples of minnows, shad, goldfish, carp, crayfish, and clams were taken from the Great Miami River in the vicinity of Miamisburg, Ohio. Activities of ^{238}Pu and ^{239,240}Pu are reported for these organisms. In addition, distributions were determined for plutonium between the gastrointestinal tract and the remainder of the organism in carp and between the shell and soft parts of crayfish. The results are discussed with regard to the spatial distribution of the organisms around Miamisburg and the tissue distribution within the animals.

INTRODUCTION

Because of public concern over the toxicity of plutonium and the possibility of biomagnification of radionuclides in food chains, we are studying the distribution of ^{238}Pu and ^{239,240}Pu in the fish and invertebrates of the Great Miami River watershed. These studies are intended to determine if riverine organisms have the same kind of food chain discrimination against plutonium from fallout or industrial wastes as organisms from oceans, lakes and ponds.

The Great Miami River basin in southwestern Ohio represents a moderate size watershed in a temperate climate. Land use if predominantly agricultural, except for a heavily industrialized corridor along the river between Dayton and Cincinnati. The Great Miami River is approximately 260 km long from its source in Indian Lake to its confluence with the Ohio River west of Cincinnati.

Three primary isotopes of plutonium are present in the river basin. The major isotopes in fallout and fairly evenly distributed throughout the watershed are ^{239}Pu and ^{240}Pu, which are analytically indistinguishable. Some

^{238}Pu (approximately 5% of the ^{239,240}Pu) is present in fallout and can be analytically distinguished from ^{239,240}Pu. Elevated concentrations of the ^{238}Pu isotope are found near Mound Laboratory, a plutonium fabrication facility located in Miamisburg, Ohio.

Mound Laboratory releases ^{238}Pu to the Great Miami River in three ways. A 114,000-liter industrial wastewater tank normally containing 50—200 μCi of ^{238}Pu is periodically discharged directly into the river (Carfagno, pers. comm.). About 100 μCi per year is released from two stacks and settles out within 5 to 8 kilometers of the laboratory (Muller et al., 1974; 1975). much of this ^{238}Pu then enters the river during rainfall and subsequent erosion and runoff (Bartelt et al., 1974). Finally, there is a continual movement of ^{238}Pu enriched sediment from a series of ditches, canals, ponds, and storm sewers which empty into the river. Most of the plutonium in this enriched sediment is a result of a spill in 1969 on the laboratory grounds (Rogers, 1975). Although some of the enriched sediment has presumably been carried into the river by now, the remainder constitutes a source of ^{238}Pu particularly important during rainstorms when sediment loss is accelerated. Because the plutonium from Mound Laboratory is the ^{238}Pu isotope and fallout plutonium is primarily ^{239,240}Pu, ratios of ^{238}Pu to ^{239,240}Pu are useful in indicating the source of plutonium in a sample.

MATERIALS AND METHODS

Samples were collected from sites upstream and downstream of Mound Laboratory (Fig. 1). Two sites, Rip Rap Bridge on the Great Miami River and Stanley Avenue Bridge on the Mad River in Dayton, are 20—30 km upstream of Mound Laboratory and considered background sampling sites. Fish were collected using gill nets, seines, and a portable electrofishing unit. Several large carp were obtained as the result of a boom shocking survey for Dayton Power and Light Company conducted by De Pauw University, Green Castle, Indiana. Crayfish were collected by hand or by seining. All samples were frozen in the field and stored in freezers until analysis. The samples were thawed, and wet weights were determined. They were then dried to a constant weight at 105°C and ashed at 500°C. The ash was homogenized and subsamples were spiked with ^{242}Pu as a yield monitor. The samples were digested and analyzed for plutonium according to the procedure of Nelson et al. (1973).

RESULTS AND DISCUSSION

Results of the analyses for the fish samples are summarized in Tables 1 and 2. These samples were whole fish unless otherwise stated. No goldfish were collected at any background sampling site. The carp sample collected in Dayton is upstream of Mound Laboratory's effluent discharge point and

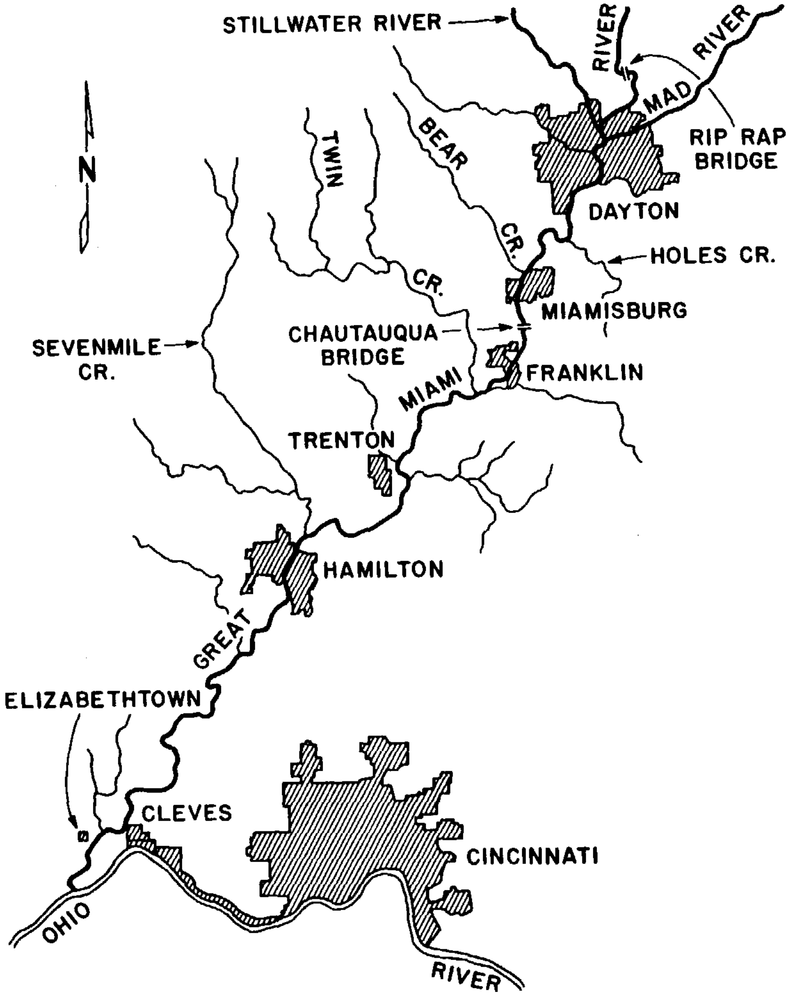

Figure 1. Great Miami River sampling sites.

Table 1. ^{238}Pu and ^{239,240}Pu activities (fCi/g dry wt)$^{(a)}$ in fish.

Sample	Number of Organisms Analyzed	Average Total Length (cm)	% Dry Wt/Wet Wt	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu
Minnows						
Rip Rap Bridge	315 g dry wt	(b)	23.2	0.47 ± 0.07	0.20 ± 0.04	2.41 ± 4.6
Mad River-Stanley Ave.	515 g dry wt		22.8	0.37 ± 0.04	0.53 ± 0.05	0.71 ± 0.10
0.8 km S. Chautauqua Bridge - July 1974	288 g dry wt		25.4	2.89 ± 0.16	<0.051	<56.2
- Aug 1974	300 g dry wt		25.5	22.2 ± 0.6	0.75 ± 0.11	29.6 ± 4.6
Shad						
Rip Rap Bridge	9	21	24.4	0.04 ± 0.02	0.60 ± 0.05	0.08 ± 0.03
Mad River-Stanley Ave.	14	24	26.1	0.03 ± 0.02	0.71 ± 0.05	0.03 ± 0.02
0.8 km S. Chautauqua Bridge	7	19	19.8	54.1 ± 0.9	0.68 ± 0.10	82 ± 12
Goldfish						
0.8 km S. Chautauqua Bridge (3)$^{(c)}$	5—17	15	19.1—20.1	11.4 ± 0.5 —18.9 ± 0.7	0.17 ± 0.05—1.61 ± 0.20	11.8 ± 1.5— 94 ± 27
Franklin (3)	12—41	14	19.0—20.5	2.21 ± 0.08—10.9 ± 0.2	0.41 ± 0.02—0.13 ± 0.02	50 ± 15—122 ± 71
Trenton	14	17	25.3	2.16 ± 0.05	0.17 ± 0.02	13.1 ± 1.2
Carp$^{(d)}$						
Dayton	3	(b)	32.1	0.029 ± 0.004	0.007 ± 0.002	4.09 ± 1.09
Power Plant (3)	1—4	49	15.6—18.5	0.13 ± 0.02—0.90 ± 0.34	<0.02—0.13 ± 0.09	1.9 ± 0.4— >9.0
0.8 km S. Chautauqua Bridge (6)	1—18	22	25.6—28.8	0.63 ± 0.06—1.36 ± 0.09	0.02 ± 0.02—0.09 ± 0.03	7.1 ± 2.5— 72 ± 52
Franklin (4)	11—28	20	25.4—26.3	0.54 ± 0.04—2.22 ± 0.11	0.04 ± 0.02—0.07 ± 0.03	8.8 ± 3.7— 56 ± 28

(a) The ± value is 1 σ counting error. When the counting error is >100%, the concentration is reported as <2 σ.

(b) Not measured.

(c) Number of samples.

(d) Organisms minus GI's.

Table 2. Distribution of ^{238}Pu and ^{239,240}Pu (fCi/g dry wt)$^{(a)}$ between carp bodies and gastrointestinal tracts.

Site	Number of Organisms Analyzed	Average Total Length (cm)	Bodies			Gastrointestinal Tracts		
			^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{238}Pu	^{239,240}Pu	^{238}Pu/^{233,240}Pu
Dayton								
17 Nov 1974	3	(b)	0.029 ± 0.004	0.007 ± 0.002	4.1 ± 1.1	0.22 ± 0.06	0.04 ± 0.02	5 ± 3
Power Plant								
15 Nov 1974	4	45	0.90 ± 0.34	0.13 ± 0.09	7 ± 6	0.27 ± 0.08	ND$^{(c)}$	--
15 Nov 1974	1	48	0.13 ± 0.02	<0.02	>9	0.90 ± 0.26	ND$^{(c)}$	--
15 Nov 1974	1	54	0.23 ± 0.03	0.12 ± 0.02	1.9 ± 0.4	0.46 ± 0.12	0.82 ± 0.14	0.56 ± 0.17
0.8 km S. Chautauqua								
22 July 1974	9	22	1.25 ± 0.07	0.02 ± 0.01	72 ± 52	72 ± 2	0.86 ± 0.19	81 ± 18
23 July 1974	18	21	0.63 ± 0.06	0.03 ± 0.02	21 ± 13	75 ± 2	2.64 ± 0.34	28 ± 4
23 July 1974	17	23	0.88 ± 0.07	0.02 ± 0.02	46 ± 36	82 ± 2	0.65 ± 0.16	125 ± 30
25 July 1974	7	19	0.68 ± 0.07	0.03 ± 0.02	20 ± 11	57 ± 2	0.84 ± 0.29	68 ± 24
13 Aug 1974	1	54	0.64 ± 0.07	0.09 ± 0.03	7 ± 2	36 ± 2	ND$^{(c)}$	--
13—15 Aug 1974	5	(b)	1.36 ± 0.09	<0.04	>39	187 ± 5	0.48 ± 0.29	382 ± 226
Franklin								
24 July 1974	28	20	0.67 ± 0.07	ND$^{(c)}$	--	67 ± 1	0.83 ± 0.12	79 ± 12
24 July 1974	13	20	0.54 ± 0.04	0.05 ± 0.02	11 ± 3	72 ± 2	1.16 ± 0.26	62 ± 14
24 July 1974	24	21	0.58 ± 0.07	0.07 ± 0.03	9 ± 4	76 ± 2	0.89 ± 0.22	87 ± 21
14 Aug 1974	11	20	2.22 ± 0.11	0.04 ± 0.02	56 ± 28	92 ± 3	2.43 ± 0.47	38 ± 7

(a) The ± value is 1 σ counting error. When the counting error is >100%, the concentration is reported as <2 σ.

(b) Not measured.

(c) Not detectable or ≤ background.

shows slightly elevated levels of ^{238}Pu, possibly due to contamination during analysis. The Power Plant station is downstream of Mound Laboratory within 0.5 km of our Chautauqua sampling site. The concentrations of ^{239,240}Pu in different species from all sites are quite similar and fall within the range of values reported from other studies of freshwater and marine fish (Waller et al., 1973; Bowen, 1974; Livingston and Bowen, 1975; Noshkin, 1972; Miettinen, 1976). The elevated concentrations of ^{238}Pu below Mound also show little variance between different species. Since shad are typically filter feeders, carp and goldfish bottom feeders, and minnows selective feeders (Lagler, 1952), feeding preferences appear to have little effect on plutonium concentrations. However, this apparent consistancy may be a function of the small sample size, as more detailed studies by other workers show food and habitat effects on plutonium concentrations in fish (Waller et al., 1973; Noshkin, 1972; Emery et al., 1974).

Carp with gastrointestinal tracts removed show only a slight increase in ^{238}Pu downstream from Mound while activities in gut tissue and contents are more variable and much higher (Table 2). Analysis of stomach contents of carp from the Great Miami River indicate plant and algal detritus along with varying amounts of sediment comprise the major proportion of gut material. Since algal and sediment activities (Wayman et al., 1974; Volchok, 1975) are much higher below Mound Laboratory, the variability in ^{238}Pu concentrations generally indicate the volume of material and relative porportion of high activity sediment in the gut at capture. It is apparent from the data in Table 2 that little of the plutonium taken into the gut transfers across intestinal walls into body tissues; most of it is eliminated in the feces (Emery et al., 1974). This lack of transfer across the gut wall is confirmed in other plutonium distribution studies (Bowen, 1974; Adams and Fowler, 1974). The ^{238}Pu concentrations in gastrointestinal tracts for carp collected at the Power Plant site are much lower than those from other downstream sites. These fish were captured in deep water with a boomshocker and were much larger than those collected by other methods at different sampling sites. Because the Power Plant site is less than 2 km below Mound Laboratory, it is possible that either these fish moved into the area from upstream shortly before capture, or the difference in size and feeding habits caused less sediment to be present in their gut. Either possibility would explain the relatively low ^{238}Pu activities of their gastrointestinal tract and contents.

Table 3 presents data for miscellaneous aquatic organisms. Again, ^{239,240}Pu concentrations are fairly similar upstream and downstream from Mound Laboratory. The clam shells have not been analyzed, but the soft parts have very low levels of plutonium. The ^{239,240}Pu concentrations in crayfish reported here are similar to published results for crayfish from Lake Michigan (Yaguchi et al., 1973). The concentration of ^{238}Pu in a whole frog collected at Floodgate, downstream from Mound Laboratory, is in the range of values found in carp bodies collected downstream. As expected, crayfish show a

Table 3. ^{238}Pu and ^{239,240}Pu activities (fCi/g dry wt)[a] in miscellaneous aquatic organisms.

Sample	% Dry Wt / Wet Wt	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu
Clams				
Rip Rap Bridge (soft parts)	12.8	0.14 ± 0.03	0.04 ± 0.04	3.5 ± 1.0
Crayfish				
Rip Rap Bridge	27.5	<0.37	0.53 ± 0.28	>0.2
0.8 km S. Chautauqua (3)[b]	23.0—24.5	25.5 ± 1.5—37.2 ± 1.7	0.35 ± 0.20—0.44 ± 0.20	59 ± 31—106 ± 65
Franklin	24.6	35.2 ± 1.5	0.12 ± 0.12	288 ± 288
Frog				
Floodgate	24.6	1.07 ± 0.08	0.04 ± 0.02	26.8 ± 0.5

[a] The ± value is 1 σ counting error. When the counting error is >100%, the concentration is reported as <2 σ.

[b] Number of samples.

substantial increase in ^{238}Pu below Mound Laboratory.

Tables 4 and 5 present data from analyses of over 800 crayfish collected at our Chautauqua sampling site downstream of Mound Laboratory. These samples were collected as part of an experiment using a fluorescent dye, Rhodamine WT, to trace Mound Laboratory's industrial waste effluent (Sprugel et al., 1975). The dye concentration in the Great Miami River was monitored using a fluorometer and thus the ^{238}Pu effluent pulse was followed downstream from its release point. Samples were collected before the pulse arrived (A); on the upswing (B); during the peak (C); during the downswing in dye concentration (D); and after the pulse had passed the site (E). Crayfish were analyzed whole or as shell and tissue portions. The shell portion included the thoracic carapace, chelae, pleopods, periopods, abdominal segments, uropod and telson. All shell portions were scraped clean of membranes, viscera, and tissue. Sample D was subdivided into four classes (Table 5) by length measured from the tip of the telson to the point of the rostrum. An additional larger size class (>94 mm) was found only in the A collection and it is also included in Table 5.

The ^{238}Pu concentration in whole crayfish increases somewhat during the pulse (Table 4), apparently due to uptake by soft tissue. In all cases, concentrations of ^{238}Pu are much higher in soft tissue than in the shell agreeing with the findings of Nelson and Noshkin (1973). They reported that highest plutonium concentrations in clams and lobsters were found in the viscera. Results from other studies, however, have shown shell or skeletal portions of animals to have higher levels of plutonium than soft tissue (Noshkin, 1972; Aarkrog, 1971; Ward, 1966). No discernable pattern is present for ^{238}Pu activity in the shell.

The data also show a concentration of ^{239,240}Pu, again most apparent in soft tissues, above levels determined in other samples collected at the same site (Table 3) at different times. We can offer no explanation for this discrepancy in our data.

If one examines the length distribution data (Table 5) from sample D the ^{238}Pu concentrations appear to be dependent on size, possibly related to surface area of the shell and gills; i.e., smaller organisms have a greater surface area per unit weight. The >94-mm size class from sample A, however, does not fit the pattern suggesting some other factor to be more important than surface area or perhaps indicating some sampling problem. Because sample A was the only one to have this largest size class, and it was not subdivided into the smaller size classes comparable to sample D, we can offer no sure explanation.

CONCLUSIONS

1. Fish collected below the effluent discharge pipe show a substantial

Table 4. Distribution of ^{238}Pu and ^{239,240}Pu (fCi/g dry wt)[a] in crayfish.

Sample	Whole			Shell			Tissue		
	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu
A "before"	82 ± 6	7.31 ± 1.76	11	37.0 ± 1.9	0.37 ± 0.26	100	190 ± 11	5.0 ± 2.0	38
B "upswing"	104 ± 6	1.61 ± 0.90	64	25.8 ± 1.5	0.6 ± 0.32	43	196 ± 12	<5.9	>33
C "peak"	118 ± 8	23.2 ± 3.9	5	37.3 ± 1.9	2.27 ± 0.81	16.4	145 ± 9	4.5 ± 2.4	32
E "after"	141 ± 7	11.8 ± 2.0	11	21.6 ± 1.3	0.37 ± 0.17	58.4	244 ± 12	2.8 ± 1.6	85

[a] The ± value is 1 σ counting error. When the counting error is >100%, the concentration is reported as <2 σ.

Table 5. Distribution of ^{238}Pu and ^{239,240}Pu (fCi/g dry wt)[a] between shell and tissue of crayfish as a function of individual length.

Sample	Shell			Tissue		
	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu	^{238}Pu	^{239,240}Pu	^{238}Pu/^{239,240}Pu
D 0–49 mm	39.6 ± 2.8	3.41 ± 0.90	11.6	260 ± 10	9.7 ± 2.4	27
D 50–64 mm	33.8 ± 5.4	4.40 ± 0.90	7.7	204 ± 8	12.9 ± 1.9	16
D 65–79 mm	12.1 ± 1.1	0.72 ± 0.32	16.9	106 ± 8	<13.0	>8
D 80–94 mm	13.9 ± 2.5	2.28 ± 2.28	6.1	61 ± 4	3.2 ± 0.9	19
A >94 mm	24.2 ± 3.1	<4.0	>6.1	159 ± 11	<7.72	>21

[a] The ± value is 1 σ counting error. When the counting error is >100%, the concentration is reported as <2 σ.

increase in ^{238}Pu activity, apparently independent of their feeding behavior.

2. The activities in carp gastrointestinal tracts are much higher than in carp bodies presumably due to sediment found in the gut.

3. It appears that there is little transfer of plutonium across intestinal walls of carp into body tissues.

4. Crayfish collected downstream of Mound Laboratory show elevated levels of ^{238}Pu particularly in soft tissues as opposed to shell.

5. Activities of ^{238}Pu in crayfish tend to increase during an effluent pulse.

6. A size-dependent relationship may exist in crayfish with larger size classes having lower activities per unit weight than smaller size classes possibly due to the difference in relative surface area of the shell and gills.

ACKNOWLEDGMENTS

The authors wish to thank W. H. Westendorf, D. G. Carfagno and R. Robinson of Mound Laboratory for their cooperation; and J. R. Gammon and C. Yoder of De Pauw University for providing us with several fish samples. In addition, we wish to thank Argonne's Center for Educational Affairs for supporting S. E. Groves under the Undergraduate Research Participation Program.

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EFFECT OF NATURALLY OCCURRING ORGANICS ON PLUTONIUM-237 UPTAKE BY ALGAE AND BACTERIA

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ABSTRACT

Naturally occurring organics were concentrated from Skinface Pond, near Aiken, South Carolina and separated into four nominal diameter size fractions (F I > 0.0183; 0.0183 > F II > 0.0032; 0.0032 > F III > 0.009; F IV < 0.009 µm) by membrane ultrafiltration. Each fraction was introduced into *Scenedesmus obliquus* and *Aeromonas hydrophila* cultures at concentrations equal to those found in nature to determine their effects on ^{237}Pu uptake. Plutonium-237 uptake was determined in log phase cultures after 6 hr incubations. The initial plutonium concentration in each flask was 1.1×10^{-4} µCi/ml $^{237}Pu^{+4}(NO_3)_4$. Fractions I and II significantly reduced ^{237}Pu uptake by *S. obliquus*, while F IV increased uptake and F III had no effect. Plutonium-237 uptake by *A. hydrophila* was not significantly different in the presence of F I, F II or F III than tryptic broth medium alone, while F IV significantly increased ^{237}Pu uptake.

INTRODUCTION

While plutonium presently exists at very low concentrations in the biosphere, trophic biomagnification and possible localized contaminations may result in increased plutonium concentrations in organisms of higher trophic levels. Algae and bacteria form the base of aquatic food webs and concentrate plutonium greatly over water concentrations (Noshkin, 1972; Folsom *et al.*, 1975; Gromov and Spitsyn, 1974a, 1974b; Giesy and Paine, 1977a). Cycling processes and biological uptake of Pu must be understood prior to environmental releases so that rational assessments of its hazards can be determined. Accumulation of plutonium by algae and bacteria is dependent upon the aqueous forms of plutonium (Andelman and Rozzell, 1970; Noshkin, 1972). While much is known about plutonium separations chemistry, little is known about the environmental chemistry and speciation of plutonium (Noshkin, 1972).

Francis (1973) stated that chelation with naturally occurring organic soil components may be important in the movement of transuranics in natural food chains leading to man. Szalay (1964) suggested that humic acids be used in the disposal of radioactive wastes because these organic compounds immobilize radionuclides. Pillai and Mathew (1976) studied the effects of humics on Pu solubilization in sea water but emphasized that further information is needed on the chemical behavior and influence of humics on Pu behavior in water. Routson *et al*. (1976) reported that Pu^{+4} tends to form complexes with many organic ligans such as those in soil. While Pu-humic interactions have been investigated (Bondietti, *et al*., 1975; Routson, *et al*., 1976), no studies have considered the effect of humics and fulvics on the availability of plutonium to aquatic biological systems.

Recalcitrant, naturally occurring organic compounds, variously known as humics, fulvics, and tannics are of worldwide distribution in soil and aquatic systems. Because of their ubiquity, these large polyphenolic compounds are involved in the chemical processes of nearly all surface waters (Schnitzer and Khan, 1972). Organic ligans, which can form complexes, with metals are important in determining the form, movement, and availability of trace metals in natural waters (Rashid and Leonard, 1973; Andelman, 1974; Pittwell, 1974; Jackson, 1975; Giesy, 1976). The distribution of organic carbon between various nominal diameter fractions varies spatially and temperally. Because of this variability in nominal diameter of organics an understanding of the effects of the various sized fractions must be obtained instead of total organic ligand.

The surface waters of the Southeast are soft and contain high concentrations of refractory organic compounds. This coupled with the large number of present and projected nuclear power plants and nuclear fuel production and reprocessing facilities for the region makes crucial the understanding of Pu cycling and fluxing processes in aquatic ecosystems. The objective of this study was to determine the effects of various sized organics on Pu uptake by an algal and bacterial species.

MATERIALS AND METHODS

Axenic cultures of *Scenedesmus obliquus* (Türp) Kütz (strain #1592) were obtained from the Indiana University Culture Collection. *Aeromonas hydrophila* (strain #7966) was obtained from the American Type Culture Collection. Stock, axemic *S. obliquus* cultures were maintained in 200 ml Algal Assay Procedure (AAP) medium aerated with sterile air (Anon, 1971). Stock algal cultures were checked periodically for bacteria contamination by plating on peptose agar, incubating in tryptic soy broth (TSB) and microscopic examination. Stock and experimental cultures were incubated at 24 ± 2°C under 4035 lux illumination from balanced spectrum "Growlux" fluorescent bulbs on a 16 hr light-8 hr dark regime. Algal inocula were taken from 10 day old *S. obliquus* cultures with cell densities of 4.2×10^5 cells $\cdot$ ml^{-1} and relative growth rate (K' 0.68 day^{-1}). Initial *S. obliquus* cell densities were concentrated by centrifugation to between 1 and 2×10^5 cells $\cdot$ ml^{-1} in experimental flasks. Stock *A. hydrophila* cultures were maintained in AAP-tryptic soy broth (TSB) medium (Difco). *A. hydrophila* inocula were drawn from 24 hr cultures with cell density of 4.5×10^6 cells $\cdot$ ml^{-1} (K' = 3.6 day^{-1}) to make an initial cell density of 5×10^5 cells $\cdot$ ml^{-1} in experimental flasks. *S. obliquus* and *A. hydrophila* were incubated with $^{237}Pu^{+4}$ for 6 hr on a rotary shaker at 200 rpm. Algal and bacterial cell densities were determined, using calibrated phytoplankton (0.1 ml) and Petroff-Hausser counting chambers respectively.

Plutonium-237 was obtained from Oak Ridge National Laboratory and assayed by the Savannah River Laboratory. Plutonium was prepared as $^{237}Pu^{+4}$ (Table 1). Plutonium stocks (1.1×10^{-2} μCi $\cdot$ ml) were stored in 0.5 *N* HNO_3 in polyethylene.

Water samples were collected from Skinface Pond, Aiken, Co., South Carolina. Particulates of nominal diameter ≥ 0.15 μm were removed and the remaining dissolved and colloidal constituents fractionated and concentrated by membrane ultrafiltration (Giesy and Briese, 1977; Giesy and Paine, 1977b).

Uptake experiments were conducted in 100 ml medium in 300 ml Erlenmeyer flasks. One milliliter of $^{237}Pu^{+4}$ stock was added to AAP or AAP ± 0.5% TBS (bacterial uptake) and adjusted to pH 4.5 with 1.0 *N* NaOH using a microburette. The volume of NaOH required to adjust the experimental solutions containing organics and $^{237}Pu^{+4}$ spike had been previously determined. Organics were present in experimental media at the concentration at which they were found in nature. The resulting experimental media contained 1.1×10^{-4} μCi $^{237}Pu^{+4}$ at pH 4.5. Solutions were mixed and allowed to stand 5 min before algal or bacterial inoculations.

Algae and bacteria were separated from experimental media by centrifugation into a phthalate separator (Giesy and Paine, 1977b). Control cultures, containing all components of the medium but no algal or bacterial cells, were handled in the same manner as experimental cultures to evaluate contamination in the separation procedure or Pu polymerization. Reagent blanks, controls and experimentals were analyzed for ^{237}Pu using a GeLi detector interfaced to an INOTEC 4000 channel pulse height analyzer.

The experimental design was a randomized block design. Since all replicates could not be centrifuged concurrently, experimental units were blocked orthogonally over time. Significance of treatment effects were tested using standard analysis of variance techniques and means separated with Student-Newman Keubs multiple range test. Significance was tested for at the $\alpha = 0.05$ level.

Table I. Flow chart of $^{237}Pu^{+4}$ preparation procedure.

I. REMOVAL OF ORGANIC MATTER

- A. Place Pu solution in HNO_3 solution in beaker.
- B. Evaporate to dryness. Do not bake residue.
- C. Dissolve residue in 2 ml con. HNO_3. Evaporate to dryness. Do not bake residue.
- D. Repeat Step I C.
- E. Add 2 ml 30% H_2O_2 to residue. Evaporate to dryness. Do not bake residue.
- F. Repeat Step I E.
- G. Add 2 ml con. HNO_3. Evaporate to dryness. Do not bake residue.
- H. Repeat Step I G.

II. ADJUST ALL Pu TO Pu^{+4} OXIDATION STATE

- A. Add 10 ml 1N HNO_3 to residue.
- B. Add 1 ml 1M $NaNO_3$. Heat 30 min. (light boil, 400°C) for 30 min. with stirring. Cool.

III. OXIDIZE Pu^{+4} TO Pu^{+5}

- A. Add 0.5 M $KMnO_4$ dropwise with stirring until pink color persists. Add 0.2 ml 0.5 M $KMnO_4$.
- B. Heat for 30 min. (250°C). Cool to 25°C.

IV. REDUCE Pu^{+5} TO Pu^{+3}

- A. Add 1 M $Fe(NH_2SO_3)_2$ dropwise with stirring until solution is clear. Add 0.25 ml 1 M $Fe(NH_2SO_3)_2$.
- B. Stir 30 min. Do not heat.

V. OXIDIZE Pu^{+3} TO Pu^{+4}

- A. Add 4 ml 1 M $NaNO_2$.
- B. Heat for 30 min.
- C. Wash into 25 ml 8 N HNO_3.

VI. ADSORB Pu^{+4} ONTO ANION EXCHANGE COLUMN

- A. Add 8 ml (HNO_3) to solution
- B. Pass solution through column (5 ml volumes) Dowex$^{(R)}$ - 1 x 4, 20-50 mesh resin.
- C. Collect effluent. Wash beaker with 2 ml 8 N HNO_3. Pass washes through column.
- D. Repeat VI C.

VII. ELUTE Pu^{+4} FROM ANION EXCHANGE COLUMN

- A. Wash column with 100 ml 0.5 N HNO_3. Collect effluent as Pu^{+4}.

RESULTS

Total organic carbon (TOC) in Skinface Pond was 31.1 mg/l (Table 2), which is approximately four times the world average for rivers (Garrels and MacKenzie, 1971) which is typical of southeastern aquatic systems (Beck et al., 1974). More than 80% of the TOC was comprised of F III and F IV (Table 2). Fraction I and F II both significantly reduced ^{237}Pu uptake by S. obliquus (Table 3). Fraction III, which contained 36% of the TOC had no significant effect on ^{237}Pu uptake by S. obliquus while F IV significantly increased ^{237}Pu uptake.

A similar trend was observed for ^{237}Pu uptake by A. hydrophila (Table 4). Both F I and F II reduced ^{237}Pu uptake below that in TSB medium alone. As with S. obliquus F III did not significantly affect ^{237}Pu uptake by A. hydrophila, while F IV significantly increased ^{237}Pu uptake.

No polymerization was observed in these experiments. Less than 5% of the plutonium was lost by plating to glass flasks with plating losses less in flasks containing organics.

DISCUSSION

Uptake is herein defined as the association of ^{237}Pu with algal or bacterial cells. Uptake does not imply an active mechanism and no attempt was made to determine if ^{237}Pu was associated with the surface or interior of cells.

Oxidation state may be important in determining the physical and chemical reactions which Pu will undergo in the environment (Rouston et al., 1976). Wahlgren et al. (1976) reported Pu^{+4} to be the predominant oxidation state in Lake Michigan water. For this reason a rigorous procedure was used to assure that all of the ^{237}Pu was in the IV oxidation state at the beginning of the uptake studies. The environmental chemistry of Pu is also pH dependant (Rouston et al., 1976), as is the complexation chemistry of humic acids (Stevenson and Ardakani, 1972). For this reason the pH of experimental media was adjusted to 4.5, which is typical of many southeastern surface waters such as Skinface Pond. While Pu^{+4} self polymerization has been observed, this phenomenon was probably not a major Pu transformation in this study. Polymerization is directly proportional to Pu concentration and inversely proportional to pH (Rouston et al., 1976).

Many investigators have studied the effects of humic and fulvic compounds on trace metal uptake by phytoplankton (Goldberg, 1952; Shapiro, 1957; Giesy, 1976). Several investigators have reported increased trace metal uptake by phytoplankton due to metal organic interactions (Burk, et al., 1932; Provasoli and Pinter, 1959; Johnson, 1964), while others have reported reduced uptake due to humic acids (Provasoli et al., 1957; Provasoli, 1963; Prakash and Rashid, 1968; Siegal, 1971; Giesy, 1976). Shapiro (1966) found that higher

Table 2. Organic carbon distribution in ultrafilter fractionation ranges of Skinface Pond water.

Ultra-filter	Fraction	Nominal Molecular Weight	Organic carbon (mg/l)	Organic carbon (%)
XM-300	FI* > 0.0183μm	FI > 300,000	2.2	7
PM-10	0.0183 > FII > 0.0032μm	300,000 > FII > 10,000	3.8	12
UM-05	0.0032 > FIII > 0.0009μm	10,000 > FIII > 500	11.1	36
UM-05	FIV > 0.0009μm	FIV > 500	14.0	45
TOTAL			31.1	100

*f = Organic fraction isolated

Table 3. Effects of four organic fractions on S. obliquus cellular ^{237}Pu concentration after 6 hr.

Treatment	Final cell density (cell x $10^5 \cdot ml^{-1}$)	$^{237}Pu^{+4}$ Removed From Medium	
		Removed/Cell (10^{-5} pCi $^{237}Pu \cdot cell^{-1}$)	% Total Removed
AAP	$1.1 \pm 0.18^{A*}$	32 ± 6^{B}	29
AAP + F I	1.6 ± 0.36^{A}	6 ± 2^{C}	9
AAP + F II	1.3 ± 0.22^{A}	12 ± 4^{C}	14
AAP + F III	1.3 ± 0.30^{A}	29 ± 6^{B}	34
AAP + F IV	1.3 ± 0.28^{A}	52 ± 7	62

*N + 4, $\alpha = 0.05$, confidence intervals = $\pm 2 S_{\bar{x}}$. Means which are not significantly different from one another are denoted by A, B or C.

Table 4. Effects of 4 organic fractions on A. hydrophila cellular ^{237}Pu concentration after 6 hr.

Treatment	Final cell density (cell x $10^7 \cdot ml^{-1}$)	$^{237}Pu^{+4}$ Removed From Medium	
		Removed/Cell 10^{-8} pCi $^{237}Pu^{+4} \cdot cell^{-1}$	% Total Removed
TSB	4.3 ± 2^{A*}	14 ± 6^C	5.4
TSB + F I	5.1 ± 2.2^A	7 ± 2^B	3.2
TSB + F II	3.6 ± 1.4^A	10 ± 2^B	3.2
TSB + F III	3.1 ± 1.2^A	15 ± 2^C	4.2
TSB + F IV	5.6 ± 2.1^A	51 ± 9	26.0

*N = 4, $\alpha = 0.05$, confidence intervals = ± 2 $S_{\bar{x}}$. Means not significantly different from one another are denoted by A, B or C.

molecular weight fractions of naturally occurring colored organic acids are responsible for most of the chelating capacity in surface waters. Stevenson and Ardakani (1972) reported that metals bound to the larger fractions were less available to plants and less mobile in soil solution. In this study the larger diameter organics reduced ^{237}Pu uptake, presumably by chelating an otherwise sequestering Pu. The smaller diameter organics greatly increased Pu uptake. This may be due to decreased Pu precipitation and adsorption to glass. Organic matter in soil solutions decreased Pu availability to plants (Schulz et al., 1976). Pillai et al. (1976) found humics maintained more Pu in solution than sea water alone but precipitated removing Pu over a long period of time. Smaller diameter organics may also facilitate Pu uptake by crossing the cell membrane as a Pu-organic complex or by juxtapositioning Pu on the cell. Metal organic complexes may readily penetrate cell membranes, thus chelating agents may transport metals into cells in quantities greater than would normally occur (Doyle et al., 1974).

Low molecular weight humic acid fractions and fulvic acids may penetrate the cell membranes of phytoplankton (Prakash et al., 1973) but the higher molecular weight humic acids (F I and F II) probably do not. Prát et al. (1961) and Prát and Pospisil (1959) report that humic acids cannot penetrate plant cell membranes. Humic substances that penetrate cell membranes are inhibitory to all cellular processes (Prát, 1968). The important point is that all naturally occurring organics do not increase or decrease Pu uptake but some increase Pu uptake while others cause a decrease. Waris (1953) suggested that humic substances may directly affect the cytoplasmic membrane. Chaminade (1956) reported humic acids stimulate growth of violet epidermal cells by allowing mineral transport across the cytoplasmic membrane, while Saunders (1957) stated that humic acids may stimulate cell membranes of phytoplankton, thereby allowing an influx of bound metals.

ACKNOWLEDGMENTS

This research was supported by Contract E(38-1)-819 between the University of Georgia and the U. S. ERDA. R. M. Wallace, and E. L. Wilhite helped with Pu^{+4} preparation. R. Geiger and L. A. Briese aided in data collection and J. C. Corey helped with all phases of the study.

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Analysis Methodology for Transuranics

HISTORICAL REVIEW OF TRANSURANIC ANALYTICAL METHODOLOGY

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ABSTRACT

The methods used for the analysis of the transuranic elements neptunium, plutonium, americium, and curium are reviewed. Particular attention is given to those procedures suitable for low-level environmental and biological assessment. The preparation of such samples, derived from a variety of sources and incorporated in different masses and volumes of soils, water, vegetation, and biological matrices, is described. A comprehensive discussion of matrix dissolution and equilibration techniques indicates that complete dissolution employing tracer techniques is to be preferred. Leaching of large soil samples is satisfactory for worldwide fallout. Individual and sequential separation and purification procedures, including precipitation, extraction, and ion exchange methods, are described. Sample preparation techniques for radiometric assay, such as stippling, evaporation, and plating, are reviewed. For both tracer and nuclide assay, electroplating provides the thin samples suitable for alpha pulse height analysis. Past and current counting methods for the transuranic alpha, beta, and gamma emitters are reviewed.

INTRODUCTION

Current interest in the transuranics (TU), particularly Np, Pu, Am, and Cm, stems from problems of potential or actual dissemination in the environment of reactor waste, fallout, and the progress on development of the breeder reactor. It also stems from such studies as the assessment of these elements at Enewetak and Bikini, studies of plutonium sites by the Nevada Applied Ecology Group, and a variety of plant and animal uptake studies. Recently, ^{241}Am, because of its growth from ^{241}Pu, has become a factor in the environment and interest in this isotope increased. Neptunium and curium as well as Am are expected to accumulate in radioactive waste from reprocessing plants and are also of interest.

Although the history of the transuranic elements began a short time ago, with the advent of the nuclear age, the analytical methods employed to separate and

and purify these elements are, in most instances, based on fundamental classical chemistry.

These methods involve precipitation, coprecipitation, extraction, various forms of ion exchange, and are based on the classic oxidation-reduction properties of the actinides and their similarity to the lanthanide series. The +3 state is characteristic of all the actinide elements, uranium to americium, and they are capable of being oxidized all the way to +6; in this respect, the actinides differ from the rare earths. From uranium to americium, the stability of the +6 state decreases regularly and there is a corresponding increase in the stability of the +3 state. At americium, the +3 state becomes the predominant one, and the only state known to exist for curium in aqueous solutions is the +3.

The early analytical methods that were developed were used to separate and purify Pu from alpha emitters and beta-emitting fission products in fresh fission solutions. Subsequently, it became necessary to separate plutonium in trace quantities from a variety of environmentally derived matrices. Plutonium and the other transuranics were not conveniently in solution but in a variety of chemical and physical forms--some as colloids, some as complexes, and others in the form of highly refractory oxides fused with macro amounts of aluminum, iron, silica, etc. Further, in addition to removal of the artificial radionuclides, methods had to be developed to also remove the natural elements. The radiochemical procedures which were developed, therefore, employ the basic chemical techniques, but differ in a very special manner, due to the necessity of separation from environmental matrices. Usually, concentration may be required from large volumes and isolation from macro quantities of stable elements which differ from matrix to matrix and sample to sample. As a result, the procedures employed must be tailored to the specific matrix. Over the years, procedures have become simplified, consisting primarily of a concentration step, decontamination steps involving coprecipitation and ion exchange, and a final isolation for counting. Considerations in the evolution of concentration techniques are losses due to foaming, spattering, entrainment, formation of insoluble oxides, polymerization, etc. A successful procedure produces good yields, clean plates, high purity, with economy of materials and time and use of the least labor intensive methods. In the course of these operations, due to the large volumes and masses of the samples, management of the fumes and waste liquids produced must be considered. Lastly, for the more sensitive chemists, some of the odors produced are not exactly "Chanel No. 5."

Much of the early plutonium chemistry, in 1943, at the University of Chicago Metallurgical Laboratory, was performed with less than 1,000 dpm of plutonium. Since this was all that was available, recovery techniques were devised for recycling the plutonium. Techniques employed then to isolate from a variety of experimental solutions were redox, coprecipitation, and hexone extractions, with great success. Later, in connection with nuclear weapons tests in the late 1940s, the transuranics, primarily neptunium and plutonium in the form of particulates, were recovered from inorganic and organic filter media. There were also many bioassay samples to be analyzed. These were invariably tracer-free. In the early 1950s, plant uptake of plutonium studies were also performed, Jacobson and Overstreet (1948), again tracer-free Animal studies at Lovelace and UC Davis and the TG-57 (1957 Safety Tests at NTS) and Roller Coaster, Major and Wessman (1964), plutonium safety tests made necessary the requirement for procedural development on large biological samples. HASL studies of worldwide fallout and the data produced on atmospheric and surface distribution of the actinides led to the development of standarized procedures, Harley (1957, 1972). Laboratories contributing to the development and application of transuranic procedures are the USERDA primes, such as LASL, LLL, Battelle, etc.; DOD units such as AFTAC, WPAFB, the now defunct NRDL, and LFE (Tracerlab).

The procedures discussed herein apply to inorganic matrices such as soils; aqueous samples such as water, sea water, and liquid bioassay samples; and organics such as vegetation, animal, and filters. In addition to single element procedures, some discussion of sequential procedures is included.

It is interesting to note that current work reported here also includes instrumental techniques which, in some instances, relieves the need for radiochemistry. It is also ironic that after years of development of tracer techniques, because of the desire to measure all transuranic nuclides in waste, there is now a need for reliable and accurate tracer-free techniques.

DISSOLUTION AND CONCENTRATION TECHNIQUES

The transuranics in environmental samples are usually low in activity and as a result, in order to obtain usable data, samples large in mass or volume must be taken. For example, the currently most abundant transuranic in soil is plutonium, with a typical average concentration of 0.02 dpm/g (0-20 cm), Hardy *et al.* (1972). Therefore, in order to recover the nuclide of interest, it is necessary to reduce the volume or mass to manageable amounts and assure its solubilization. Some samples such as soil require prior sieving and grinding. Organic matter must be decomposed. At an early stage, tracer must be added and equilibrated and lastly the final solution conditioned for the initial radiochemical separation and purification steps. Since these elements are not normally volatile, precautions in these procedures are primarily mechanical and involve proper mixing to get a uniform sample and adjustment of temperatures and heating cycles to prevent mechanical losses. In the aqueous phases, acidification is required to prevent polymerization or radiocolloid formation and losses due to plating out prior to equilibration. The methods employed for volume or mass reduction in organic, inorganic, and aqueous samples are shown in Figure 1 and typical concentration steps are shown in Figure 2.

SOILS

Soil varies widely in physical and inorganic chemical composition and in the amount of organic material. A similar statement can be made about the form of

FIGURE 1 TRANSURANIUM PREPARATION, DISSOLUTION AND CONCENTRATION TECHNIQUES

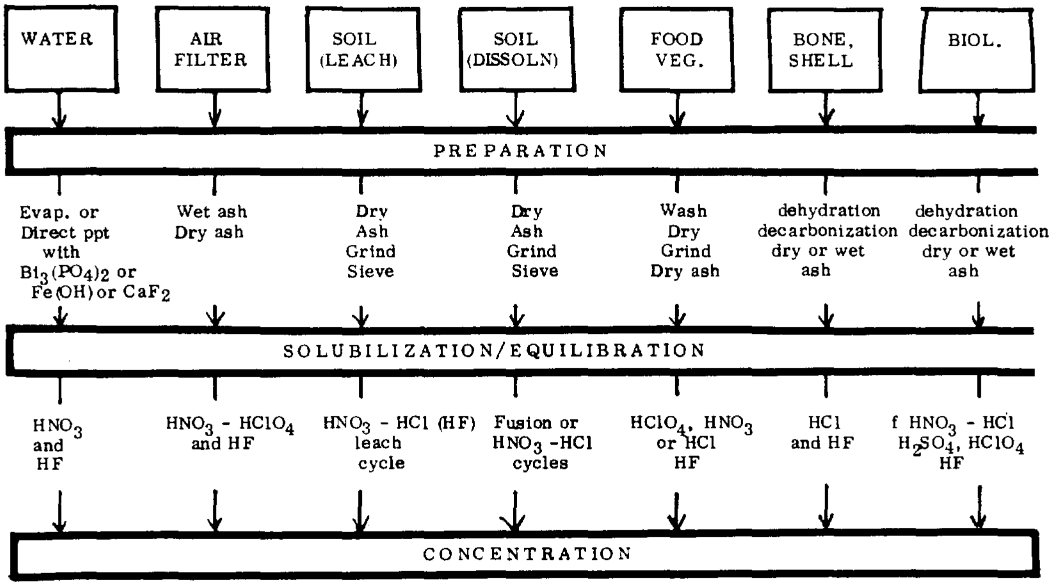
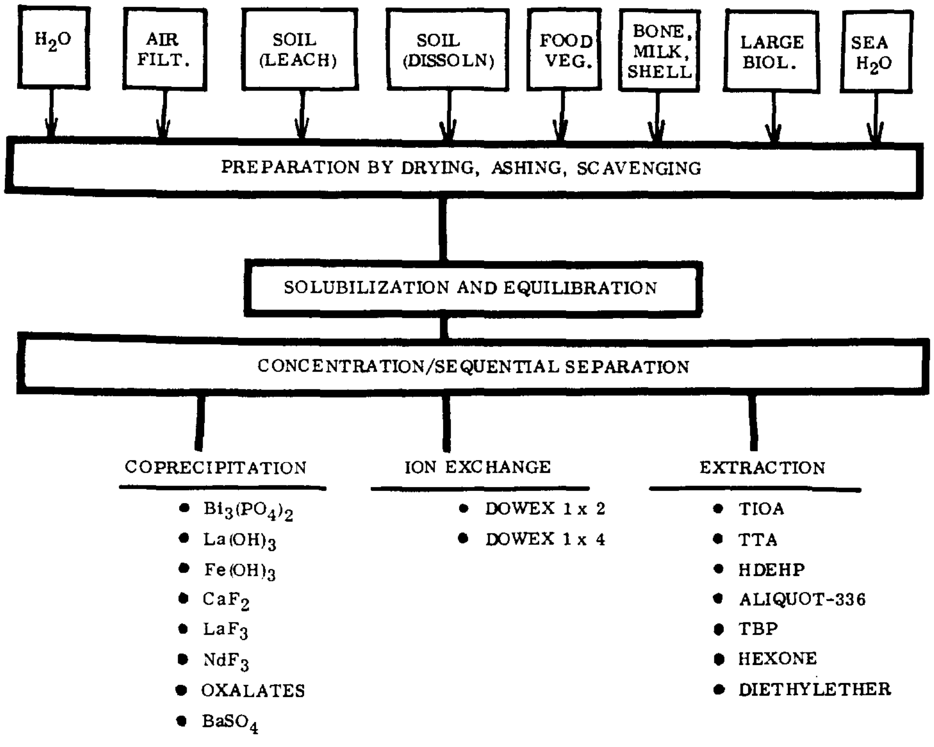


FIGURE 2 TRANSURANIUM PREPARATION AND SEPARATION



the actinides in the soil samples. Many individual procedures have been developed for Np, Pu, Am, and Cm in soil matrices and have been reported in the literature. Procedures have been also reported for separation of these elements in groups of two or more. The evolution of these procedures has been based on the insurance that the actinides have been completely solubilized and equilibrated with tracer, when added. The method of assay also has influenced the concentration technique. Virtually all methods involve drying the soil, charring the organic and humus, and grinding and sieving the sample. However, in the HASL procedure, Harley (1972), the organic is ground up with the sample and destroyed by HNO_3-HCl acid when the Pu is leached. Bernhardt (1976) evaluated sample collection and analyses techniques for environmental Pu.

The greatest controversy has been connected with the leach method versus complete dissolution. For large (1 kg) samples, the acid leach has proven to be the only practical procedure. Leaching methods have been successful with Pu activities, particularly when they are not refractory and loosely bound to the matrix. Geiger (1959) discussed the relative ease of $Al(NO_3)_3$-HNO_3 leaching of Pu from soils exposed to reactor effluents. Bond and Clark (1958) discuss efficiency of leaching Rainier and Tamalpais nuclear debris with groundwater, from the Nevada Test Site, as a function of leaching temperature. Thomas (1957) reports on leaching with HNO_3 and HF in the analysis of Bravo shot soils. Olafson *et al.* (1957) wet ashed New Mexico soils with HNO_3-$HClO_4$-HF and isolated the Pu on a LaF_3 carrier. Everett and Drake (1960) have leached fused Nevada soil samples, covering a wide range of activity, with HNO_3 and reported good yields on the leach solution, but measurements on the residue were not indicated. More recently, Bishop *et al.* (1971) tested 50-g acid leaching and Sill's fusion methods for ^{238}Pu in soil using tracer techniques with comparable results. They used an HNO_3-HF leach and concentrated with TIOA-xylene. Corley *et al.* (1971) successfully leached ^{239}Pu from Hanford surface soil (10-g) using an HCl, HNO_3 cycle and anion exchange and TTA extraction and evaporated the organic phase directly onto a Pt disc. Sill (1975) performed experiments on soils heated to different temperatures and then leached with various combinations of acids. His plutonium activity balances showed the fate of sample and tracer Pu for each and the fact that HF had a large effect on obtaining the true Pu concentration in the soil. The most referred to leaching techniques are those of Talvitie (1971) for 1-g quantities and of Chu (1971) for bulk samples. The HASL acid leach has been adapted to 1-kg and 3-kg soils by Major *et al.* (1972) and is sometimes complicated by colloid formation. Poet and Martell (1972) used an acid leach but did not use HF for Am and Pu. Fowler and Essington (1974) report on a method for plutonium in soil, the HASL-LASL leach method. This formed the basis for the procedure described in AEC Regulatory Guide 4.5, Anonymous (1974a), which was stated to be "acceptable to the Regulatory staff for sampling and analysis of plutonium in soil with the sensitivity and accuracy needed to adequately monitor plutonium in soil in the environs of fuel processing and fuel fabrication facilities." The procedures involved HNO_3-HF leaching, a leach with HCl, and complexing with H_3BO_3. A precipitation of $Fe(OH)_3$ is made with NaOH, carrying down undissolved residues which are then treated with HNO_3 and H_3BO_3 before any undissolved residue is discarded and the Pu purified by usual anion exchange techniques. Hayes *et al.* (1975) simply leached estuarine soils with HCl and concentrated with TIOA in toluene. Bently *et al.* (1971) used acid leach on 50-g soils followed by HDEHP extraction and a LaF_3 precipitation and reported 90% yields. Noshkin and Bowen (1973) leached ocean sediments with HNO_3. Koide *et al.* (1975) applied the acid leach techniques of Talvitie and Wong to both atmospheric dusts and marine sediments.

Complete dissolution techniques are satisfactory for limited sample sizes. Crouch and Cook (1956) reported on fusion of irradiated rocks, soil, and organic matter, and report losses due to fusion insolubles in samples over 250 mg and spattering difficulties. Merritt (1958) fused Chalk River soils with alkalai hydroxides and carbonates but reported interferences from large amounts of alkalai. Sill (1961), and Sill and Williams (1971) have extensively studied the fusion of refractory silicas and later Sill and co-workers (1974) extended the method. For soils and air dusts, after a preliminary HF treatment, sequential potassium fluoride and potassium pyrosulfate fusions are performed. Bretthauer and Hahn (1975) proposed the fusion method as a reference method for Pu in soil. Butler *et al.* (1971) use a similar technique on 5-g samples but extracts with TIOA and then a LaF_3 precipitation.

Complete acid dissolutions of soil samples are suitable for samples up to 100 g, at which point the procedure becomes lengthy and uneconomical. Wet ashing, involving complete acid dissolution, has been found to be the method of choice by Major *et al.* (1965a, 1971) and has been applied to soil samples up to 100 g. HNO_3-$HClO_4$ was used to destroy the organic matter followed by HNO_3-HF treatment. Residues were separated, dissolved, and recombined and concentrated by coprecipitation with $Fe(OH)_3$.

The isolation of the other actinides in soil does not differ materially from that of Pu except in the concentration steps. Major *et al.* (1971) used, for the Nevada soils, a series of HF, HNO_3-HCl steps and H_3BO_3 to decompose the silicon fluorides, and subsequently isolate by anion exchange and HDEHP extraction. Nevissi *et al.* (1975) reported on complete acid dissolution techniques similar to the above for both Pu and Am in Bikini soils. They fused any residues with a $NaOH/Na_2CO_3$ mixture and extracted with TIOA. Edgington *et al.* (1975) applied the methods of Talvitie and Wong to the determination of Pu and Am in Lake Michigan sediments and extracted the Am with Aliquat 336S. Kleinberg (1967), in the LASL collected radiochemical procedures, reports on individual and sequential procedures in Nevada soil debris. For Am and Cm samples up to a few grams, Smith (1967) found LaF_3 precipitation after dissolution was satisfactory for 0.1 to 0.3 g soil, but for larger samples, HDEHP extractions were necessary. Barnes (1967) used anion exchange and elution with dilute thiocyanate for Cm. Smith (1967) used Zr and Sr hold back for Np and a LaF_3 precipitation. Wolfsberg and Daniels (1967) concentrated the actinides from 250-g dissolved samples, first with TBP and then with HDEHP. Sill *et al.* (1974) applied his fusion method followed by extraction with Aliquat 336 separating out first Am, Cm, and then Np and Pu.

VEGETATION

The dissolution and concentration of vegetation samples presents far less problems than soil. Most studies of the actinides in vegetation have been for the purpose of the determination of their partition between soil and plant. Fallout under natural and greenhouse conditions and uptake of solutions of the actinides has been studied. Due to the extremely small uptake, approximately 10^{-4} to 10^{-8}, large volumes of plant material are required. The usual procedure in working with vegetation involves washing, drying, grinding, and dry ashing, after which the residues are treated according to many of the techniques described in soil. Jacobson and Overstreet (1948) studied uptake of Pu in plants using tracer-free techniques and dry ashing. Olafson (1945) processed plant samples by wet ashing using tracer-free methods. Neubould and Mercer (1962) studied the uptake of ^{239}Pu by ryegrass in which the plant samples were wet ashed in HNO_3 and $HClO_4$ and any silica residues were washed. Concentration was by coprecipitation as the fluoride with neodymium carrier. Talvitie (1971) ashed 1-g samples of vegetation and filters in a muffle furnace, pulverized the ash, added ^{236}Pu tracer, and dissolved in HCl. Price (1972) studies the uptake of ^{237}Np, ^{239}Pu, ^{241}Am, and ^{244}Cm by tumbleweed and cheatgrass. He pre-ashed the plant tissue with H_2SO_4 before dry ashing and then pulverized the sulfated ash. Major *et al.* (1974) carbonized up to 500 g of dry vegetation at 250° C then ashed at 600° C. An acid dissolution, including the use of HF, was performed on the ash to solubilize actinide-containing residues.

FILTERS

Filters employed for the collection of atmospheric dust are either inorganic, such as glass fiber and ACC Type V asbestos paper, or organic, such as plastic or natural fibers. In some cases, the matrix mass is small, but for composited filter samples of 10^5 cubic meters of air or ultrahigh-volume air samplers with large sized (1 m^2 IPC filter), the inorganic sample mass may be several grams. The inorganic filters are amenable to HF and acid dissolutions. Early procedures, Beaufait (1952), were developed for the effective dissolution of the Type V filters employed in sampling close-in and long-range fallout. Since these procedures were lengthy, organic filters having the desirable filtration and particle retention properties such as IPC and polyfiber were developed. Hunt *et al.* (1968) used an HNO_3, $HClO_4$, and HF procedure for Am and Cm, subsequently precipitating on $La(OH)_3$. Major and Wessman (1964), in Operation Roller Coaster, used acid dissolution on Casella and Andersen impactor discs and a f-HNO_3 and $HClO_4$ boil down on sticky film samples from the Nevada Safety Tests. Talvitie (1971) ashed and dissolved in 1 N HCl. Khandekar (1976) obtained high-altitude dust samples by swiping aircraft with cotton wool soaked with white petrol, and acid leaching with better than 50% yield of Np, Pu, Am, and Cm. For microsorban (polystyrene) filters, up to 1 m^2, Tracerlab (LFE) (1965) pyrolysed the sample up to a maximum of 525° C until black carbon is decomposed; the residue is treated by the soil HF dissolution procedure.

BIOLOGICAL

Biological samples, which include a variety of tissues, bone, urine, and feces, in most cases require high sample mass to obtain the necessary sensitivity for the actinides. Dehydration and decarbonization of the samples are the initial necessary steps. Special problems are fatty tissues and calcium and phosphates in bone. After such preparatory steps are complete, the procedures are those indicated for inorganic ash. Major *et al.* (1965) wet ashed animal samples up to 3 lb. from the Roller Coaster tests, with f-HNO_3, H_2SO_4, $HClO_4$. Greater than 3 lb. samples employed a Hg catalyst, K_2SO_4, an antifoam agent, and H_2SO_4. Pu was separated from the salts with a cupferron-$CHCl_3$ extraction. Bone was dry ashed, dissolved in HCl, and the Pu extracted. Metabolic samples were dried, boiled to low volume with f-HNO_3, ignited, and then extracted, as with the small animal samples. Sansoni and Kracke (1971) used an iron catalyzed H_2O_2 procedure for rapidly wet ashing organic matrices. For Pu and Am, Major *et al.* (1975) cut up bovine bone and meat into small sections, added H_2O_2 and slowly ashed at 200° C then at 520° C in a muffle furnace to a white ash, and dissolved in HCl. Any residue was dissolved in HNO_3, HF, H_3BO_3, added, combined, and HF and Y added. The Pu and Am are carried on the CaF_2-YF_3 precipitate. Crawley and Goddard (1976) studied ^{241}Am and ^{244}Cm in rats, drying, ashing at 500° C, and dissolving in HNO_3 with ultimate recovery of 55-65%. Shipman and Weiss (1960) isolated Pu in urine by cocrystallization with potassium rhodizonate. Bokowski (1964) determined Am in urine in the presence of Pu, coprecipitating the Pu with $BiPO_4$, wet ashing, and counting the LaF_3 slurry.

Butler (1965) and Major (1968) determined ^{241}Am in urine using ^{244}Cm tracer by wet ashing with HNO_3 and $HClO_4$ and concentrating by an HDEHP extraction and coprecipitation with $Fe(OH)_3$. Lee *et al.* (1975) used HNO_3, HF to rapidly dissolve soft tissue samples up to 1 Kg. Undissolved fat found to be free of activity is filtered off. The method is particularly suited to the analysis of samples from injection or uptake by animals.

AQUEOUS

Techniques suitable for aqueous volume reduction, depending on salt content, are evaporation, coprecipitation, extraction, and ion exchange. Scheidhauer *et al.* (1953) separated Pu from 3-liter water samples quantitatively by chemisorption on solid CaF_2. Seawater samples suitable for Pu and Am measurement require at least 60 liters for Pu and 100 for Am, according to Murray and Kautsky (1975). Pillai *et al.* (1964) reduced the plutonium and then coprecipitated with $Bi_3(PO_4)_2$ from 45 liters of seawater. Bowen *et al.* (1971) acidified the seawater, added tracer and H_2O_2, and carried the plutonium on $Fe(OH)_3$. Noshkin (1972) lists numerous analytical references. Miyake and Sugimura (1975) used the $Fe(OH)_3$ procedure to determine the plutonium content in Pacific Ocean waters. Murray and Kautsky (1975), after an $Fe(OH)_3$ step, used an HDEHP extraction to separate Pu from Am. Livingston *et al.* (1975) have published a review of analytical procedures for the transuranium elements in seawater and marine sediments.

SEQUENTIAL SEPARATION PROCEDURES

Often it is required to analyze more than one radionuclide from a single sample. In order to achieve maximum results from scarce sample material and optimize analytical sensitivity as well as distribute expensive sample preparation costs over several elements, a sequential separation procedure is used. A typical multipurpose 26-element sequential separation scheme, suitable for many radioactive elements including Pu, Np, and U but with no provision for Am or Cm, is that of Crouch and Cook (1956). That portion applicable to Pu, Np, and U is shown in Figure 3. It is quite similar to later procedures and the authors pointed out that certain parts of the procedure required experienced and special manipulation for successful recoveries. The most common transuranic sequential separation procedures in use have been for Pu and Am. A survey of Pu and Am procedures is given by Wessman *et al.* (1974). However, even more extensive sequential separation schemes have been employed successfully at this laboratory for many years on up to 22 elements, although they were limited to certain sample types. Hunt *et al.* (1968) reported on a scheme for analyzing ^{241}Am, ^{242}Cm, and ^{147}Pm in fallout with applicability to other sample lines but the method had low tolerance for rare earths. Nash (1964) had a procedure for the separation of over 30 elements using a Dowex-50 cation exchange column and ammonium alpha-hydroxyisobutyrate at various concentrations and pH. The procedures by Wong *et al.* (1970), Figure 4, shows the sequential steps used for isolating Pu and Am from large seawater samples. Since there are a wide number of such sequential procedures, further discussion is limited to the transuranium elements. One alternative is to dissolve the sample prior to adding carriers and/or tracers and use appropriate sized aliquant portions for each element of interest.

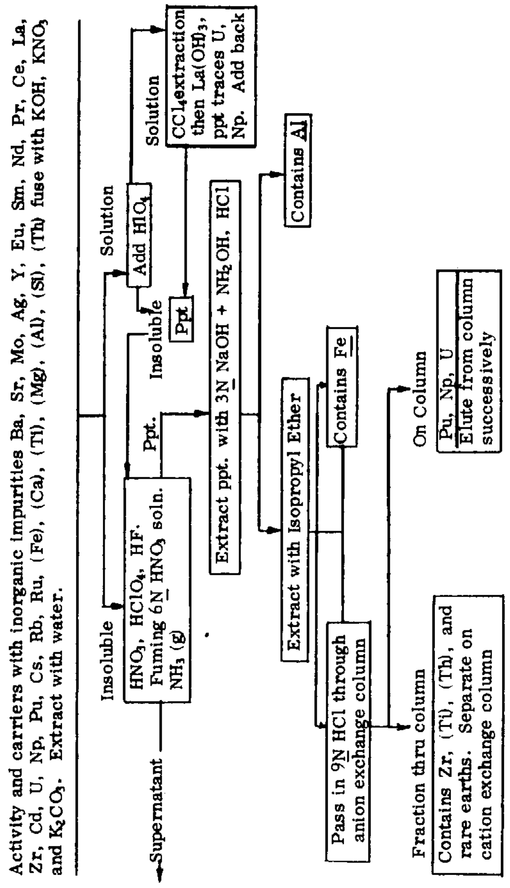
Ion exchange separations have especially been of great value in the separation and identification of the transcurium elements. Thompson *et al.* (1954) employed Dowex-50 columns plus eluants of ammonium lactate and ammonium citrate. Choppin *et al.* (1956) employed ammonium alpha-hydroxyisobutyrate as a superior eluant for Am, Cm, Bk, Cf, E, Fm, and Mv mixtures from Dowex-50 columns. Such separations might be considered the ideal because a number of elements are efficiently separated in one continuous operation. However, usually the sample matrix elements will preclude the use of the classical procedures which also involve the use of rather expensive columns.

Buckley (1969) compiled a manual which contains several procedures for separating various combinations of the actinides, tracer-free, by a combination of precipitation and ion exchange steps, followed by gross alpha counting or alpha specroscopy for identification.

Sill *et al.* (1974) report a very comprehensive method for simultaneous determination of the alpha-emitting nuclides, Ra through Cf, in soil. After a fusion, the elements are precipitated on $BaSO_4$. An extraction of the elements, Th through Pu, into Aliquat-336 is made from an acidic aluminum nitrate solution which is then treated to sequentially strip Th through Pu. Americium, Cm, and Cf are extracted into Aliquat-336 from acid deficient aluminum nitrate and

FIGURE 3 CROUCH AND COOK'S SEPARATION SCHEME AS APPLICABLE TO TRANSURANUMS

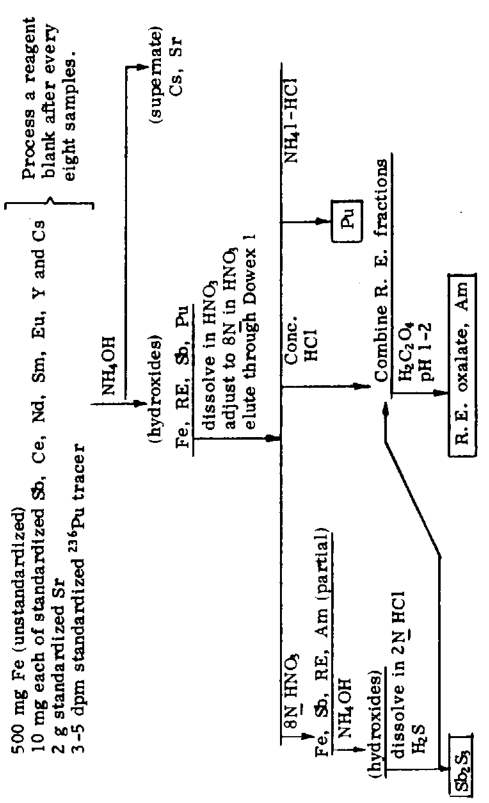
Ref: JINC 2 p 224 (1956)



Activity and carriers with inorganic impurities Ba, Sr, Mo, Ag, Y, Eu, Sm, Nd, Pr, Ce, La, Zr, Cd, U, Np, Pu, Cs, Rb, Ru, (Fe), (Ca), (Tl), (Mg), (Al), (Si), (Th) fuse with KOH, KNO_3 and K_2CO_3. Extract with water.

FIGURE 4 WONG, NOSHKIN AND BOWEN'S PROCEDURE AS APPLICABLE TO THE ANALYSIS OF Pu AND Am* IN SEAWATER SAMPLES

Ref: Methods for Marine Radioactivity Studies, IAEA, Vienna (1970), p 119.



*Private communication with K. M. Wong.

back-extracted with 8 M HNO_3. In the case of soils, Sill recommends further separation of the tervalent actinide group from the lanthanides according to the method of Filer (1974). The latter procedure combines extraction chromatography with bis(2-ethylhexyl)-phosphoric acid (HDEHP) in the presence of diethylenetriaminetetraaccetic acid and the use of a column of Teflon-6 powder. Sill further recommends, when necessary, Moore and Jurriaanse's (1967) method to separate ^{242}Cm from ^{252}Cf whose alpha energies are so identical, alpha spectrometry cannot resolve them.

Wessman *et al.* (1971) reported on a method for trace plutonium analysis in a large variety of sample matrices and this system formed the basis for the determination of the other transuraniums, as later reported (Wessman *et al.*, 1977). A schematic diagram of the procedure is given in Figure 5. The first steps of the procedure, which are adequate to solubulize and equilibrate Pu, are also adequate for any other transuraniums present. Yield tracers are ^{233}Pa (^{239}Np) for ^{237}Np, ^{236}Pu for ^{238}Pu, ^{239,240}Pu, and ^{243}Am for the combined group consisting of ^{241}Am, ^{242}Cm, and ^{243,244}Cm. An anion exchange column (Dowex 1-X4) is used to absorb Np (IV) and Pu (IV) from 8 N HNO_3 while Am (III) and Cm (III) pass through the column. Plutonium is separated from Np on a subsequent Dowex 1-X4 ion exchange column. The column is washed with 8 N HNO_3 and HCl. Plutonium is eluted with NH_4I-HCl and Np with 2 N HCl. The Am, Cm-containing eluant is further purified using steps which have been pretested to assure that Am is not fractionated from Cm. Figure 6 shows an elution curve obtained in testing the final cation exchange column step in the procedure. In actual analyses, the fractionation of Am and Cm is usually within 2 to 3% of each other.

NEPTUNIUM PURIFICATION

Magnusson and LaChapelle (1949) discussed the procedure devised by Seaborg and Wahl (1948) to isolate weighable quantities of ^{237}Np for further study. The classical reactions were used, mostly lanthanum fluoride precipitations to carry Np (IV) or Np (III). Impurities, such as Pu and fission products, were precipitated with LaF_3 in the presence of Np (VI).

Burney and Harbour's 1974-monograph on the Radiochemistry of Neptunium has a very complete discussion on the more recent aspects of Np chemistry including solvent extraction and ion exchange. They point out that Np is easily converted in aqueous solution to oxidation states that exhibit markedly different behavior in coprecipitation, and therefore separation from other elements can be obtained. They further note that the ion exchange, extraction chromatography, and solvent extraction separation are used almost exclusively because more complete separation is attained more rapidly. Burney and Harbour presented a collection of 25 procedures. Fifteen are for a combination of the different separation steps on highly radioactive samples, three are for environmental and biological samples, and seven are for quantitative elemental determinations. Two of the procedures for highly radioactive solutions use either ^{237}Np or ^{239}Np as tracers. None of the ^{237}Np environmental or biological procedures given use

FIGURE 5 SEQUENTIAL SEPARATION OF Np AND Pu FROM Am AND Cm FOR SOIL AND BONE SAMPLES

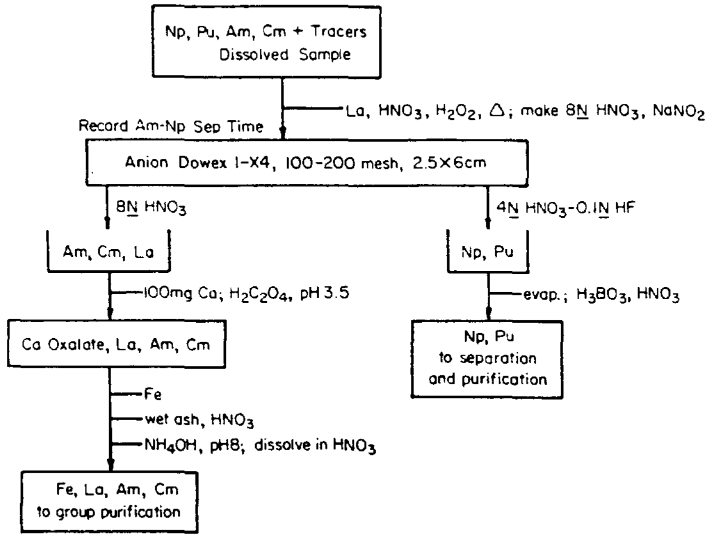
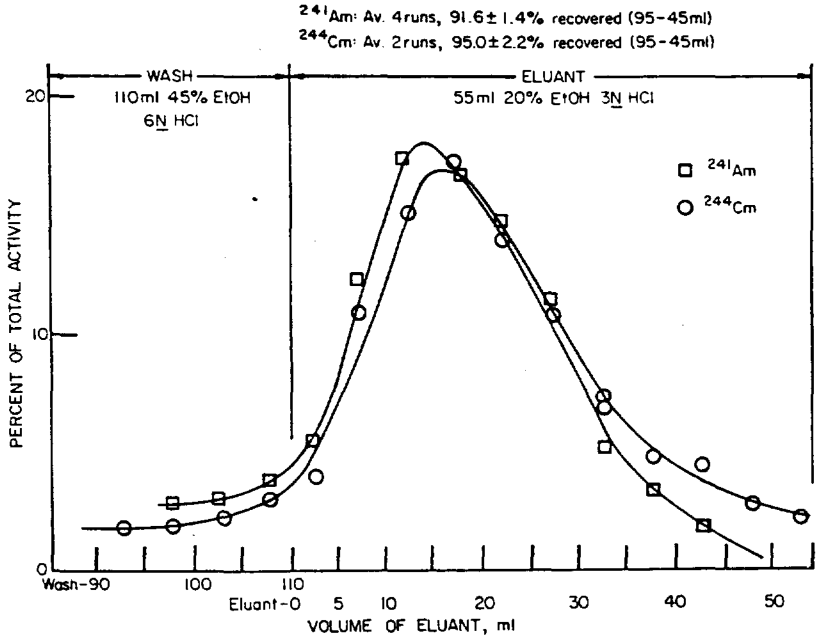


FIGURE 6 ELUTION OF ^{241}Am AND ^{244}Cm FROM A 1.0 CM DIAM., 7.5 CM LONG COLUMN OF DOWEX 50 AS A FUNCTION OF ELUANT VOLUME



tracer. However, the isotope dilution aspect of ^{237}Np analysis is fairly well detailed in the publication.

Wessman *et al.* (1975) presented two purification/separation procedures, each of which utilized a sequential precipitation of $Fe(OH)_3$, LaF_3 $La(OH)_3$; Dowex-1 anion exchange columns and extraction with either tri-isooctylamine/xylene or thenoyltrifluoracetone/toluene extraction. Good yields and good separation from the actinides and fission products and activation products were obtained.

For environmental samples of approximately 10-g ash, Wessman *et al.* (1977) report a rather simple ion exchange separation sufficient to obtain purification from the other transuraniums. Beta-emitting ^{239}Np tracer is used for yielding. An 8 N nitric acid solution of the dissolved samples and tracers is treated to obtain Np (IV), Pu (IV), and U (IV), which are absorbed onto an anion exchange resin column of Dowex 1 x 4; Am and Cm and other impurities pass through. The Np, Pu, group is then eluted with 4 N HNO_3-0.1 N HF and reabsorbed on similar but shorter columns. The column is washed with 8 N HNO_3 and HCl, then Pu is eluted with HCl-NH_4I. The column is washed with HCl and then Np is eluted with 2 N HCl. Both Pu and Np are then ready for plating.

PLUTONIUM PURIFICATION

Chemical considerations for Pu analysis are very similar to those for Np. There are a wide number of procedures. Katz and Seaborg (1957) classify the methods roughly as precipitation, solvent extraction, and ion exchange processes. They further point out that all of these methods depend upon the fact that Pu (like U and Np) has a number of stable oxidation states and that the chemical properties of the element in one state may differ to a very considerable extent from those of the same element in a different oxidation state.

Coleman's 1965-monograph on the Radiochemistry of Plutonium summarizes numerous purification steps for Pu along with comparable information on other elements. Coleman also screened "54 papers with procedures which were written in enough detail to do justice to the name." He included 19 radiochemical procedures and five special procedures in his collection. Only three are environmental procedures: one is for soil, vegetation, and water (TBP extraction); the second is for water, using chemisorption on CaF_2 followed by TTA extraction; and the third is the isolation of Pu in water on $BiPO_4$ precipitates plus co-extraction with ferric cupferride. Seven of the procedures were for Pu in urine using ferric cupferride extraction; PrF_3 precipitation and TTA extraction; LaF_3 and TTA extraction; $BiPO_4$ precipitation, LaF_3 precipitation and extraction into HDEHP; alkaline earth phosphate precipitation and anion exchange and extraction of primary amine from H_2SO_4 solution.

It is unfortunate that, at that late date, only five of the procedures dealt with isotope dilution analysis and none of the environmental urine analysis procedures were specifically designed for isotope dilution by alpha spectrometer analysis. Also, the entire subject of isotope dilution analysis using alpha spectrometry was rather lightly covered and the value of this powerful tool in radiochemical analysis might not be apparent to the novice reader.

Major *et al.* (1965) had reported on a method for determination of plutonium by tracer techniques in large biological samples. A cupferron-chloroform extraction was performed followed by anion exchange purification. The tracer was ^{236}Pu and good yields, typically 55%, were obtained with tissue samples up to 5 kg in size.

Coleman (1965) commented that the anion exchange method is so simple and effective that it had become one of the standard laboratory methods for the separation of Pu. He noted that many examples existed of the use of anion exchange resins for Pu separations in HCl and HNO_3 solutions. Even so, experience has shown that variations in the technique, sample matrix elements, the presence of other alpha emitters, and materials used markedly effect the recovery of the Pu and its radiochemical purity.

A rather general approach on samples with ash or mineral content of 1 gram or greater is to isolate the Pu onto Dowex 1 x 4 from 8 N HNO_3 solution. This may be done by pouring the solution of Pu (IV) through the column or the slurry-column technique of Kressin and Waterbury (1962). In the latter technique, about half the resin is added directly to the solution for bulk absorption and then combined, whereupon the resin is added back to the column. Washes and elutions are performed as designated by the procedure being used. Many interfering ions are removed from the Pu by this column and the Pu is then eluted with 0.4 N HNO_3-0.01 N HF. The same absorption and elution sequence is repeated with a smaller amount of resin and a smaller column, and the Pu is electroplated for counting. Examples of this type of procedure are given in the HASL Manual, Anonymous (1976, 1976a), for 50-gram tissue samples and leaches of 100-gram soils.

An alternate procedure, as given by Wessman *et al.* (1971), is to do the first column purification as in the HASL method. However, after loading the nitric acid solution directly onto the column, it is washed with hydrochloric acid solutions and the Pu is eluted with HCl-NH_4I. There are no significant advantages in this method over the HASL methods cited above; primarily, it is just another approach. For vegetation samples, Major *et al.* (1974) report using a single column of Dowex 1 x 4 and absorbing Pu from HCl solution. The column is then washed with 6 N HNO_3 to remove interfering elements and then converted back to HCl form in order to elute Pu with HCl-NH_4I.

Of the 41 papers given at the 1975-USERDA-IAEA Symposium on Transuranium Nuclides in the Environment (Anonymous, 1976b), 11 papers listed straight ion exchange procedures. Six other methods listed were TIOA extraction, TTA extraction and ion exchange, phosphate precipitation plus anion exchange and $BaSO_4$ precipitation. The methodology of analysis in the remaining papers was either not readily deduced, or was not germane to the subject. In the field of autopsy tissue analysis, Campbell *et al.* (1973) use anion exchange and Thomas *et al.* (1975) use a phosphate coprecipitation followed by anion exchange. Larson and Oldham (1974) have reported on a system using HBr and anion exchange for rapid selective Pu separation.

AM, CM PURIFICATION

The chemical separation methods for Am and Cm used by Thompson *et al.* (1949) in the Manhattan Project to produce sufficient quantities of Am and Cm for studies of these elements might be considered the classical steps. These are fluorides, oxalates, hydroxides, phosphates, iodates, fluosilicates, oxidation reduction reactions, cation exchange with citrate eluant, and a few solvent extraction methods. Many of the above methods, i.e., oxalates, iodates, fluorides, and fluosilicates, were reported to be greatly influenced by interfering elements and precipitation conditions, and temperatures. The commentary provided therein points to many problems which are encountered in using such procedures in Am-Cm analysis, wherein La and Y are employed to carry Am and Cm but they later have to be separated to obtain weighless deposits of Am and Cm for alpha measurements.

Penneman and Keenan's 1960-monograph on the Radiochemistry of Americium and Curium contains an abbreviated summary of the purification steps for Am and Cm. A collection of 15 detailed radiochemical procedures is given. None of them are for environmental procedures, and only one procedure provides for tracer yielding, either ^{243}Am or ^{244}Cm tracer.

For repetitive analyses on a given sample type and matrix, good procedures and good operator competence is usually achieved. For different sample types, the problems are increased, with probably the greatest difficulty being experienced with large (> 10 g) soil samples of varying elemental content. The difficulty is further increased when the practical limitation of minimum cost is also imposed, since this could influence the methods chosen and development of a fail-safe method for a given problem.

Over the years, the basis for Am and Cm analyses has changed. Through the 1950s and the 1960s, the primary analytical requirements existed within nuclear materials production and weapons testing programs. One continuing type of analyses in all of these programs was the need for bioassay of nuclear workers. Low-Beer (1973) surveyed a large number of laboratories in order to compile a review on bioassay procedures. She reports indications that at that time, 1973, the transplutonium elements command very little attention in routine bioassay programs. The procedures reviewed by Low-Beer are either gross alpha methods or specific tracer-free methods. In the gross alpha method, if activity is observed, further specific analyses were to be performed to identify and quantify them. These procedures were said to give high yields 80 to 95% and there is no mention of the use of any tracer in the specific methods.

The present day approach seems to be to always use isotope dilution techniques with tracer. An advantage in the use of the tracer is that the isotope sought is positively identified and the chemical processing losses are accounted for, providing high accuracy and sensitivity. In a tracer-free analysis, in case zero activity is detected, there is no way to establish that the chemical processing yield was indeed not zero due to unexpected sample interferences or some processing anomaly.

A disadvantage of the use of tracers is that precautions must be taken to balance sample activity and tracer activity within certain limits, which are often set by a particular case. Also there is the chance that the tracer being used will be present in the sample. For instance, tabulations of the TU content of spent reactor fuels by Pigford and Ang (1975) and the EPA (Anonymous, 1974) show spent reactor fuels will contain most of the long-lived TU nuclides, including ^{242m}Am, ^{242}Cm, and ^{245}Cm. If environmental samples also have similar compositions, the use of isotope dilution techniques to analyze for ^{241}Am and ^{244}Cm, the usual isotopes of prime interest, must be done with care. Approximate information on the TU's expected in such samples will aid this evaluation. In the worst case, one single sequential analysis, using tracers, may not give optimum accuracy and the analysis will have to be done in parts; but in some cases, the more expensive method of additions will have to be used.

PREPARATION FOR COUNTING

Sample preparation, after purification, depends on the counting technique which is to be employed and the energy and type of radiation measured. A consideration of the radiations of the transuranics from spent fuel and tracers shows that the preponderance require alpha measurements but beta and gamma methods are sometimes applicable. Most samples are now measured by alpha spectroscopy in Frisch grid or surface barrier detectors. Sometimes beta counting, liquid scintillation counting, gamma spectroscopy, or mass spectrometry supplements or replaces alpha spectrometry. The choice of the method is also based upon the sensitivity required and the need for discrimination from other actinides. The capabilities and limitations of the methods must be kept in mind as many different mixtures of the transuranic elements may be encountered.

The alpha-emitting actinides are prepared for counting by several techniques. Due to the short range of the alphas, then, preferably weightless, samples are required for 2 π APHA measurements. Examples of the techniques that have been used are stippling of LaF_3, Bokowski (1964), or PrF_3, Perkins (1961); discussions of the transfer of aqueous and organic solutions for thin films, Hufford and Scott (1949), stippling of $Fe(OH)_3$, Kooi and Hollstein (1962), or the cupferride extract, Brooks (1950), and ignition to the oxide and electroplating, Mitchell (1960). The discs or trays are mostly of stainless steel or platinum although tantalum, Shipman and Weiss (1960), has been used and evaporation onto a watchglass reported. Evaporation under an infrared lamp is the usual technique. Care is taken to spread the precipitate uniformly. Trays as large as 3" have been used so as to obtain thin cakes. A drop of organic film, such as collodion or zapon, is added and the metal disc flamed. Hufford and Scott (1949) review in detail the techniques for preparing thin films such as evaporation from a variety of media, TEG and slurry spreading, electrodeposition, and sublimations.

A variety of scintillation techniques have been applied to actinide measurements. Liquid scintillation methods for alpha counting of ^{239}Pu have been reported by Fontaine and Baude-Malfosse (1961) and for ^{241}Am by Cline (1967). Szidon *et al.* (1968) compared LSC methods with other Pu standarization techniques.

Bogen and Welford (1971) obtained good results using LSC for total beta and alpha assay. Ludwick (1960) applied LSC to beta counting of ^{241}Pu. Price (1972) reports that he obtained 100% counting efficiency for ^{239}Pu, ^{241}Am, ^{244}Cm and 92% for ^{237}Np using a TTWC liquid scintillating gel. McDowell *et al.* (1974) have reported on the use of a combined solvent extraction-liquid scintillation method for Pu and U determination in environmental samples. Alpha counting efficiencies of 100% and an energy resolution of 200 to 300% keV are quoted.

Scintillations produced by Pu alpha particles on a ZnS screen, coupled to a photomultiplier, are reported by Hallden and Harley (1960), Kooi and Hollstein (1962). An extensively applied method for ^{241}Am is to detect the 60-KeV gamma in a NaI(Tl) scintillation well counter as described by Bubernak *et al.* (1956), Cline (1967), Filer (1974), and Wallace (1972). Numerous other detector and sample counting geometries have been discussed, including a thin NaI(Tl) wafer detector and plastic jar containing dissolved soil solutions, 10 g per 100 g of solution, as reported by Major *et al.* (1974), on the use of a solid state planar germanium detector for 10-g soil samples as given by Essington *et al.* (1976). Heimbuch and Gee (1962) developed a scintillating ion exchange resin which they applied to the assay of plutonium. Gee *et al.* (1967) used Alpha-a-Cal, an acetate base backed with a 2-3 mg/cm^2 ZnS, to enchance registration of Am alpha particles on film with several orders of magnitude increase in sensitivity.

Schwendiman and Healy (1958), Campbell and Moss (1963), and Everett *et al.* (1964) all describe the use of nuclear track plates for high sensitivity measurements of Pu in urine.

Isotope dilution surface ionization mass spectrometry has been applied to environmental samples as low as 0.006 dpm by Dupzyk *et al.* (1972) measuring ^{239}Pu, ^{240}Pu, and ^{241}Pu with ^{242}Pu tracer. Hoffman *et al.* (1971) used a similar technique to detect ^{244}Pu in nature. Noshkin *et al.* (1974) used ^{235}Np tracer and mass spectrometry to measure ^{237}Np in sediment samples.

Exposure to neutrons in a reactor and the search for activation or fission products or tracks have also been used, Larson and Oldham (1974a), to provide highly sensitive analysis of the actinides. Some of the early instrumental techniques are those of Jacobson and Overstreet (1948) in which he used a linear amplifer system to assay ^{239}Pu. Rhodes (1957) studied the absorption of Pu in soil with a parallel plate alpha counter. Buenger *et al.* (1962) detected Pu alphas by a 2 π proportional counter. Sill and Williams (1971) used surface barrier detectors for direct alpha spectrometry of environmental samples. Melgard *et al.* (1968) preferred Frisch grid chambers to surface barrier detectors because of buildup of daughter product contamination from samples was not permanent and uniformity of plates was not required. Development of the multichannel analyzers and the associated data acquisition systems has made APHA by either surface barrier or Frisch grid chambers the preferred method for environmental alpha counting.

SUMMARY

There are numerous procedures for the analysis of transuraniums (TU) in environmental samples, and in environmental type samples from laboratory or field studies. There are many variations on reported methods and so many ways to achieve the final result accurately that none can be singled out as the preferred method.

It is known, also, that in actual laboratory operation, there are deviations from published procedures due to sample variations and procedure improvements and publication lag. Also, the accuracy and precision of the final data is known to be much dependent upon individual experience, competence, and quality control.

Possibly, the driving force for continual changes, besides scientific inquiry, is to minimize analysis costs. By their very nature, sensitive TU analysis costs have always been expensive. A comparison of available 1976-cost data to that of 1963 Roller Coaster Project costs was made. Analysis for ^{239}Pu in 10 g of soil is now being performed with lower labor cost, and slightly reduced material costs. The net overall cost is slightly lower. For small filter samples, labor is lower, material higher, and the net change is to a slightly higher cost. Both cost figures show significant procedure and operational cost reductions have been well ahead of the 13-year span of inflation. Cost figures at other facilities are not available but they would also be influenced by changes in overall technology at that facility.

Possibly, a laboratory performing gross analyses in 1963 would have experienced a significant rise in costs in converting to isotope dilution analyses, exclusive of equipment costs.

Presently there is a wealth of procedures available for Pu and Am, although not for Np and Cm. There is progress in sensitivity, accuracy, and cost reduction. Relative to the use of TU analyses as a tool in various studies and as a regulatory requirement, it is expected there will continue to be a need for procedure improvement studies.

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Transuranics Statistics and Modeling

SOME DESIGN ASPECTS OF
TRANSURANIC FIELD STUDIES

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ABSTRACT

In this paper, we discuss some design aspects of transuranic field studies. Some of the principal steps in the design of such studies are given and illustrated using examples. This is followed by a review of sampling designs that have been used at nuclear detonation and safety-shot sites on the Nevada Test Site and elsewhere for estimating spatial pattern and total amounts in soil. Some design aspects of ecosystem-type transuranic studies for estimating total amounts as well as movement of transuranics between ecosystem components are also discussed.

Acceptance sampling using either attributes or measurements is considered as a possible approach for deciding whether to clean up a contaminated site. Three general guidelines for the design of efficient transuranic studies are presented.

INTRODUCTION

The design of a transuranic field study depends on a number of factors including objectives, the quantity, kind, and properties of the radionuclides present (related to the source of contamination), the spatial distribution of the contamination over the study site (trends, hot spots, etc.), and a variety of practical considerations involving budget limitations, size of the study site, time allowed for the study, etc. As a consequence, field sampling designs are necessarily site specific. However, there are general guidelines that can be used to help design efficient transuranic field studies. These guidelines are based on statistical methodology developed for sample surveys in other areas of application and on experience in designing transuranic studies.

The design of any transuranic study should begin with careful planning well in advance of the actual collection of field samples. The first step is to carefully specify objectives. The second step is to define the "population" to which inferences are intended to apply. When these essential steps have been accomplished, decisions concerning the kind of data to collect, the precision in estimates needed to meet objectives, and the actual sampling locations can be made.

No attempt is made here to provide a complete review of the literature regarding the design of transuranic field studies. However, the references included should illustrate various designs that have been used in past studies. Our discussion of possible approaches to deciding whether cleanup is needed should be considered only as a guide to what might be done in actual practice. To our knowledge, no cleanup operation has yet been conducted using the probability approach described here. Practical problems in the field will require modification of our suggestions.

PRINCIPAL STEPS IN DESIGNING TRANSURANIC FIELD STUDIES

Certain factors should be considered in designing transuranic field studies. The description of these which follows has been adapted from Cochran (1963, pp. 5-8).

Objectives

Identification of objectives is necessary in order to choose the most appropriate field sampling design (see Eberhardt, 1976, for discussion). Since most studies have multiple objectives, a compromise design may be necessary. Some common objectives are:

1. Geographical distribution (spatial pattern) of the contamination over the study site (horizontal and with depth).
2. Inventory (total amount) of contamination present in soil (worldwide fallout or local contamination).
3. Amount of radionuclides present in ecosystem components (soil, vegetation, air, invertebrates, vertebrates).
4. Changes in concentration in ecosystem components over time.
5. Rate of movement of radionuclides from one component to another (soil to vegetation, sediments to water, etc.).
6. Resuspension by wind or from the activities of man and animals.
7. Determination of whether a cleanup operation is necessary or to verify that cleanup has been successful.
8. Evaluation of the hazard to man from a given contamination (this may include all of the above objectives).

Population to Be Sampled

Cochran (1963) defines "population" to be the aggregate from which the sample is chosen. It is important to distinguish between the "target population" and the "sampled population." In transuranic field studies, the target population is that part of the environment about which information is wanted. The sampled population refers to the population from which samples are actually collected. Inferences always pertain to the sampled population. This is an important point since the sampled population may not correspond to the target population. As an example, consider sampling the sediments of a pond known to be contaminated by plutonium. The objective might be to estimate the average Pu concentration in these sediments. The target population could be the sediment layer in all areas of the pond, whereas the sampled population might be restricted to certain portions of the pond due to practical reasons. Since samples are drawn from only the "sampled population," conclusions from these samples may not apply to the sediments in those areas of the pond that could not be sampled.

As another example, consider sampling an area contaminated locally by a nuclear detonation or safety-shot test. The "target population" for soil at these sites is usually surface soil (to some specified depth) over the entire site regardless of terrain, rock content of the soil, vegetation cover, etc. Efforts should be made to insure that the "sampled population" is identical to the "target population" so that no portion of the study area is systematically excluded from sampling. For some kinds of field studies, this goal may be impossible to achieve. For example, suppose that we wish to estimate the total deposition of transuranics from worldwide fallout since the start of atmospheric testing in 1945. The target population could be considered to be the surface area of the entire world, whereas the sampled population is restricted to land sites that have acted as fallout collectors since 1945. Those parts of the target population not included in the sampled population are bodies of water, inaccessible land areas such as mountains, locations where transuranics might have accumulated or washed away, or undisturbed sites that could not be sampled due to practical problems, or deliberate exclusion.

Data to Be Collected

Careful consideration must be given to insure that all data required to meet the study objectives will be collected. This step requires a clear understanding of objectives so that all essential information is obtained. It may also be necessary to collect certain data in order to properly interpret other data. For example, if *in situ* (field) measurement instruments are used, it may be necessary to collect additional soil samples to calibrate the *in situ* readings with wet chemistry soil concentrations. These special studies may need to be done before the main study begins.

Precision Desired

Any estimated quantity is subject to error since only a small proportion of potential environmental samples can be collected and analyzed. This error can be reduced in a number of ways such as by taking more samples, using more efficient designs, and statistical analyses, or using more accurate instrumentation and wet chemistry techniques. Since these techniques usually require additional time and money, the administrator or person who is actually going to use the data should decide on the precision required. Furthermore, this should be decided before the study begins. In transuranic field studies, the precision needed to meet objectives may be difficult to define, as, for example, when a decision regarding cleanup of a contaminated area must be made. Decisions based on highly accurate and precise estimates of contamination levels are obviously desirable. However, stringent requirements on precision may increase sampling and analysis costs considerably.

Choice of Measurement Instrumentation and Laboratory Procedures

A variety of measurement methods have been used in transuranic field studies depending on objectives, budget considerations, variability over space and time, and other factors. Some examples are field *in situ* instrument surveys, aerial surveys, and wet chemistry or Ge(Li) scan analyses on environmental samples. The choice of instrumentation will also affect our perception of reality. For example, *in situ* field gamma counters scan a large amount of soil relative to a 10-gram aliquot being scanned in the laboratory. Hence, the variability between adjacent *in situ* readings may be considerably less than that between two 10-gram aliquots from the two scanned areas due to an averaging effect in the field. Each method gives us a different view or perspective on the contamination as it exists in the field. Care should be taken so that the most appropriate instrumentation is used to fit the objectives of the study. For example, an *in situ* measurement may be appropriate for establishing general levels of contamination in soil, but inappropriate for detailed study of the relationship between soil and vegetation concentrations.

Division of Population Into Sampling Units

Before sampling begins, the population should be divided into sampling units that do not overlap and which cover the population to be sampled. The samples actually collected are drawn from this collection of sampling units. As a simple example, the ground area of a study site might be gridded (conceptually or on paper) and certain of these grid cells chosen at random from which the vegetation is collected and analyzed for Pu. If changes over time are expected to occur, as, for example, with resuspension measurements, then the sampling unit might be a unit of space over a specified time period. The totality of sampling units is called a "frame." Conceptualizing the environment as composed of distict units that fill up the environment but do not overlap can be helpful in the design of field studies because it tends to reduce a complex situation into more manageable distinct sampling units.

Selection of Samples

There are many ways of selecting particular sampling units for collection. These include simple random, stratified random, or systematic designs, as, for example, on a grid. The method of choice depends in part on the patterns and trends that are likely to be present over space (or time). For example, if soil is uniformly contaminated in all parts of the study site, then the variance of the estimated average concentration is likely to be similar for any of the above-mentioned plans (assuming equal sampling effort). However, if trends in concentration do exist, then either the stratified random or possibly a systematic design is likely to result in a smaller variance estimate. The choice between stratified random and a systematic grid system depends on the correlation structure between samples' various distances apart. If a considerable amount of data is available, perhaps from a preliminary study, it may be possible to estimate this correlation structure in order to make an objective choice between sampling plans. A knowledge of spatial correlation can also be used to obtain better estimates of averages or totals using an estimation technique termed Kriging (Davis, 1973, pp. 381-390). This technique is also mentioned below in connection with sampling for cleanup.

REVIEW OF TRANSURANIC SAMPLING DESIGNS

This review is divided into two parts; studies at nuclear detonation or safety-shot sites where the spatial distribution of the contaminants is of considerable importance, and ecosystem-type studies where movement of transuranics between ecosystem components is of primary interest. These two parts are not mutually exclusive, however, since ecosystem studies are usually an important part of studies at nuclear (fission) or safety-shot sites.

Local Contamination at Nuclear Detonation or Safety-Shot Sites

Radionuclide contamination at safety-shot and nuclear detonation sites is generally highest near ground zero (GZ) with the pattern of contamination determined in part by wind speed and direction at the time of detonation, and whether the assemblies of Pu were covered by a steel plate, buried in the soil, etc. Considerable information may be available from *in situ* instrument surveys conducted following the shot or prior to the main soil-sampling effort. These advance surveys may serve to define the general level or spatial distribution of radionuclide concentrations over the area. At safety-shot sites (studied by the Nevada Applied Ecology Group (NAEG): see Dunaway and White, 1974; White and Dunaway, 1975, 1976), the FIDLER instrument was used to measure ^{241}Am, which at these sites gives a general idea of the ^{239,240}Pu present in surface soil.

Aerial surveys have also been used to estimate spatial distribution. Examples here are the gamma surveys for ^{60}Co, ^{137}Cs, and ^{241}Am over the islands of the Enewetak Atoll (Stuart and Meibaum, 1973), gamma surveys for ^{137}Cs and ^{241}Am over the Hanford Reservation in 1973 (Bruns, 1976), and an aerial radiological survey of the Nevada Test Site (NTS) (EGG, 1972). (See Burson, 1974, for recent research progress in airborne surveys.) In situations where ^{241}Am and ^{239,240}Pu concentrations in the same aliquot are related, surveys for ^{241}Am hold promise for estimating the spatial pattern of Pu in soil.

Survey information may in some cases be useful for obtaining at least a rough estimate of the Pu inventory. To do so requires estimation of a "calibration" relationship between the *in situ* or aerial survey readings and Pu concentrations obtained in soil samples. Gilbert and Eberhardt (1976) evaluated the suitability of estimating Pu in soil aliquots taken from safety-shot sites using

FIDLER data obtained at random soil sample locations immediately prior to taking the soil sample. They found the method (termed "double sampling") to be applicable for higher levels of contamination where the correlation between FIDLER readings and Pu wet-chemistry concentrations was sufficiently high and the cost ratio of FIDLER to wet-chemistry analyses sufficiently low.

Once surveys have estimated the spatial distribution pattern, additional survey readings and/or soil samples are sometimes taken along a transect running from GZ down the "hot line" or center of the fallout pattern. Closer spacing between sampling may be used near GZ in order to estimate more accurately the rapid changes in concentration levels likely to occur in that area. This was the general approach used at the NAEG safety-shot sites, the Plutonium Inventory and Distribution Program at NTS (Church *et al.*, 1974; Brady and Church, 1975), and at the Trinity site in New Mexico (Hakonson and Johnson, 1973). The purpose of such transect samples is usually to gather information to facilitate the design of more intensive studies. Soil samples collected along a single transect are usually too few and widely spaced to be useful for estimating inventory.

If the objectives of the study require additional information on soil concentration, there are several design options open to the investigator depending on objectives. If the *in situ* or aerial surveys have satisfied the requirements for information on the spatial distribution of contamination, then soil samples may be collected primarily to estimate inventory. In this case, the survey information may be useful for stratifying the study site within which soil samples are collected. The idea of stratification is to enclose areas of different concentration levels into separate strata (subareas) so that the variability between samples within strata is as small as possible relative to the variation between stratum means. If the sample locations within strata are chosen at random, this design is termed stratified random sampling (Cochran, 1963). Stratified random sampling was used at the NAEG safety-shot sites (Gilbert and Eberhardt, 1974; Gilbert *et al.*, 1976a) and tended to result in estimates of inventory with smaller standard errors than if stratification had not been used (Gilbert *et al.*, 1975).

If *in situ* or aerial measurements are not sufficiently well correlated with soil concentrations, or if such surveys have not or cannot be done, the design of the intensive soil sampling plan may need to fulfill the two objectives of spatial distribution and inventory. One approach is to arrange a rather fine grid over maps of the study area and to choose at random a number of squares defined by the grid. This approach was used for sampling soil on most islands in the 1972 Enewetak Radiological Survey (Lynch and Gudikson, 1973). The selection process can be performed using a random number table such as produced by the Rand Corporation (1955), or by using a good pseudo-random number generator on a computer. The locations actually sampled would be the centers of the randomly chosen squares.

An alternative design would be to first stratify the area based on ^{241}Am Ge(Li) scans on soil samples collected on a grid. Once the strata are determined, a grid within each stratum (grid size need not be the same for all strata) could be established. Then two or more locations could be chosen at random within each grid square in each stratum.

The above discussion has concerned contamination from local sources. In this situation, it is usually best to avoid sampling at only "undisturbed" locations, since these locations may not be representative of the area (Eberhardt, 1976). However, when estimating total deposition from worldwide fallout, it is common to sample only undisturbed sites (Hardy and Krey, 1971). The distinction is whether interest centers on estimating the existing spatial distribution and inventory or on estimating the total amount deposited over time. Undisturbed sampling locations have also been used to estimate the spread of contamination from the Rocky Flats plant (Krey and Hardy, 1970). The issue involved here is important for other study objectives, also. Consider a study for evaluating the need to clean up an area. In terms of potential health risk, it may be more important to determine where most of the contaminant is presently located than to know only the total amount initially deposited.

Sampling to Assess Relationships

After information has been obtained about the spatial distribution of transuranics, there may be interest in estimating the amounts of contamination in various ecosystem components and the movement rate of transuranics between components. The design of these studies depends to a large extent on the particular ecosystem being studied. The design of such studies should be based on whatever information is available or can be obtained on where the bulk of the contamination is located, and how this contamination is likely to move through the environment.

Three ecosystem-type studies may be briefly mentioned. These include NAEG studies in Nevada at 10 safety-shot sites referenced above. Soil and associated vegetation samples were collected at all 10 sites according to a stratified random sampling plan. Small mammals were also collected at some sites, and in Area 13 (Project 57), cattle were grazed in the Pu-contaminated areas to obtain information on uptake and tissue distribution of Pu due to grazing a contaminated area (Smith *et al.*, 1976). An important objective of these NAEG studies is to evaluate the potential hazard to man from this contamination. A provisional Pu transport and dose estimation model has been developed for this purpose by Martin and Bloom (1976). Gilbert *et al.* (1976) gave an initial synthesis of the Area 13 Pu data for soil, vegetation, small mammals, cattle, and a "hypothetical man." Other transuranic ecosystem studies include those at the Savannah River Plant and in some of the canyons at Los Alamos. At Savannah River (McLendon *et al.*, 1976), samples of soil, a resuspendible fraction of soil, vegetation, grasshoppers, and cotton rats were collected and their Pu (^{238}Pu, ^{239,240}Pu) concentrations compared in order to study the transport of Pu in a humid climate. The primary objective of the Los Alamos studies (Hakonson *et al.*, 1976) was to estimate the ^{238}Pu, ^{239,240}Pu, and ^{137}Cs levels in canyon ecosystem components (stream sediments, vegetation, small mammals) as a function of distance below waste discharge areas.

The design of such studies is complicated due to incomplete knowledge of how transuranics move through the environment, and of the spatial patterns and correlations over time and space. Under the assumption that samples collected near each other are more alike than those from further apart, the collection of adjacent soil and vegetation samples is often practiced. However, Gilbert *et al.* (1975) illustrate that in desert environments, the correlation between

Pu concentrations in such "paired" samples is often low. In a desert environment where resuspension plays an important role in determining vegetation concentrations, differences among species have also been found (Romney *et al.*, 1975). This suggests the need in such an area to avoid lumping species together when computing average concentrations or correlation coefficients. The design of small mammal studies is complicated by a lack of close correlation (at least in desert environments) between concentrations in animal tissues and those in surrounding soil. Designs for these kinds of studies must consider those variables that might account for the observed variation. Thus, it might be possible to stratify over time, space, species, depth of soil, weather conditions, etc., in order to estimate the components of variance.

SAMPLING FOR CLEANUP

There have been a number of instances where cleanup operations or decontamination procedures have been undertaken to remove or stabilize radionuclide contamination in soil. Wallace and Romney (1975) discuss the procedures and experience gained at a number of locations and give an extensive reference list. A companion reference is Rhoads (1976), who gives a position paper on treatment of certain Pu-contaminated areas on the Nevada Test Site. Our concern here is with the elements of a sampling program that might be applicable to the question of whether a cleanup operation is required.

Two sampling approaches under the general title of "acceptance sampling" are presented below as possible procedures for deciding whether cleanup is required. Only the bare essentials are given here. Setting up these plans would require attention to many design details. The two approaches are called acceptance sampling by (1) attributes and (2) measurement. We begin with acceptance sampling by attributes.

A criterion for deciding when cleanup is required might include an upper limit on soil concentrations that should not be exceeded by most samples. The proportion of soil samples collected that exceed this limit can be used to decide whether cleanup is necessary or if a cleanup operation has been successful. The basic idea is to specify (1) an activity level, L, for soil; (2) a proportion, p_1, of samples with activities greater than L that is acceptable, i.e., for which cleanup is not required; (3) a proportion, p_2 of samples with activities greater than L that is not acceptable, i.e., for which cleanup is required; (4) the allowable risk, α, of wrongly concluding that cleanup is necessary; and (5) the risk, β, of wrongly concluding that cleanup is not necessary. Once these quantities have been specified, it is possible to determine (1) the number of samples, n, required in order to meet the α and β specifications, and (2) a rejection number, r, such that cleanup is required if r or more of the n samples have activities greater than L. This approach assumes a willingness to tolerate a certain proportion, p_1, of samples with activities greater than L without cleaning up the area. Of course, p_1 can be specified to be as close to zero as we please.

The risk, β, would, presumably, be specified near zero since the consequences of not cleaning up a contaminated area might result in undue risk to the inhabitants of the area. The "power" of the design is $1-\beta$, i.e., $1-\beta$ is the probability that the area is cleaned up when it should be cleaned up (when the proportion of soil in the area with activities greater than L is p_2). The number of samples, n, and the rejection number, r, can be determined for given values of p_1, p_2, α, and β using tables prepared by Burstein (1971). The above approach is known as "acceptance sampling by attributes," the elements of which are discussed elsewhere, e.g., Freeman *et al.* (1948) and Burr (1976).

We note that attribute sampling is ordinarily used in situations where the "attribute" can be measured accurately for each element examined and decisions about a given population (often a quantity of manufactured product) are to be made on the basis of the sampled elements. Hence, we are neglecting "counter error" here and assuming decisions are to be made on the basis of whether or not sample elements from a given area (e.g., soil aliquots) indicate that a proportion of such elements are above some set limit.

Turning now to acceptance sampling by measurements, the cleanup decision is based on average soil concentrations rather than on the proportion of samples with concentrations exceeding L. Using the average approach, it is necessary to specify both an acceptable and unacceptable average soil concentration denoted by μ_1 and μ_2, respectively, as well as the α and β risks defined above. It is also necessary to specify a value for the anticipated standard deviation (σ) of the concentration values. This average concentration approach requires that the sample mean, ($\bar{x}$), be normally distributed, which may be approximately true even if the individual sample concentrations are skewed (commonly the case in transuranic studies). Nevertheless, it may be preferable to transform the data to logarithms so that the test is made on the basis of the average logarithm of concentrations. This approach requires that μ_1, μ_2, and σ also be in log units. Once μ_1, μ_2, σ, α, and β are specified, it is relatively simple to determine the number of samples, n, to collect and the critical value, K (Burr, 1976, pp. 325-328). Letting $\bar{x}$ be the average concentration of the n collected soil samples, the decision is made to clean up the sampled area if $\bar{x} \geq K$. No cleanup action is taken if $\bar{x} < K$. See Burr (1976, pp. 332-336) for the usual case when σ is unknown.

The choice between the attribute and average concentration approach to deciding the cleanup question hinges on the normality assumption. Quoting from Burr (1976, p. 324), "Unless we can build up evidence on the distribution of measurements, we had better stay with the method of attributes." However, the attribute approach may call for substantial numbers of samples (Burr, 1976, p. 347). If the average concentration approach is used, and if transuranic concentrations in adjacent soil samples are not independent, then a useful estimate of the average concentration $\bar{x}$ might be obtained using an estimation procedure called Kriging (Davis, 1973, pp. 381-390; Delfiner and Delhomme, 1975). Kriging makes use of the correlation structure (if it exists) to obtain an optimum weighted average. We are presently engaged in research to determine the kinds of correlation structures that might exist for transuranics in soil. A disadvantage of the attribute approach is that the additional information available concerning a correlation structure is not used in the decision-making process.

Sampling for cleanup would most likely be done in a sequential manner in order to keep costs to a minimum. The basic idea would be to take a limited number of samples and make a decision whether to clean up or to withhold judgment until more samples are collected. The general approach as it is applied in industry is discussed by Burr (1976, Chapter 12). The use of acceptance sampling using either attributes or average concentrations requires that samples be collected at random within relatively homogeneous areas. The procedure outlined above should thus be applied separately within each such area. The assumption of random sampling within areas is important in order to preserve the chosen α and β risks. Consequently, the use of alternative sampling plans such as sampling on a systematic grid would need to be carefully evaluated before being used in a field study.

Once the initial cleanup operation has been completed, additional samples will be required to assure that cleanup to the level desired has been accomplished. The design of this phase of the sampling program can also be based on acceptance sampling, although the details may change from those described above. If a final demonstration (certification) is required and the attribute approach is adopted, then Table 12 in Burstein (1971) can be used to determine the number of samples required to be $100(1-\beta)\%$ sure that the true proportion of samples with concentrations greater than the cleanup level, L, is less than p_2. This approach is based on the assumption that the cleanup operation has been successful so that all samples will have concentrations less than L. Sampling to insure that cleanup criteria have been met should also be done independently for homogeneous areas.

The design of a cleanup sampling plan may involve a combination of *in situ* measurements and Pu concentrations in soil samples. If so, these two measurement systems must be well calibrated so that their results can be related. The double sampling approach investigated by Gilbert and Eberhardt (1976) at NTS safety-shot sites offers an approach to estimate the optimum allocation of effort between the *in situ* measurements and soil samples.

SAMPLING DESIGN GUIDELINES

Based on our experience in helping design some of the NAEG soil and vegetation studies at safety-shot sites, we would like to suggest several guidelines relative to the design and statistical analysis of environmental radionuclide studies. These guidelines are directed towards estimating the total amount and spatial distribution of transuranics, but the design principles are also applicable to ecosystem-type studies.

Guideline 1: Break Study Area Into Homogeneous Subareas

On the basis of field surveys, information from similar studies, topography of the land, vegetation patterns, or any other relevant information, attempt to divide the total study area into subregions (commonly termed "blocks" or "strata") such that the variability in radionuclide concentrations within subregions is less than that expected for the entire study site. If this stratification is successful, the end result should be a more precise estimate of inventory for the whole site. In addition, of course, estimates of inventory will be obtained for each stratum separately, which are useful for relating trends in soil concentrations with concentrations in other ecosystem components.

The success achieved in increasing the precision of the inventory estimate for the entire study site depends on how effective the stratification has been in dividing the area into relatively homogeneous blocks or strata (Gilbert *et al.*, 1975). An inappropriate choice of strata will yield little if any benefit in increased precision. It has been our experience that it may not be possible to construct a satisfactory statification plan without conducting field surveys and/or collecting samples prior to the main study. This leads us to the next guideline.

Guideline 2: Consider Pilot Studies

If relevant information on the variability and spatial pattern of the radionuclide is not available, a pilot study may yield information that will aid in the design of the main study. It will, hopefully, give indications of trends and levels of variability necessary for defining strata. The kind of information obtained in the pilot study should be appropriate for estimating the number of samples needed in each of the strata to achieve the maximum possible increase in precision of the estimate of inventory or to achieve a specified precision needed to meet the objectives of the study. For example, FIDLER survey readings for ^{241}Am would not in themselves be adequate for estimating the variability of ^{241}Am concentrations in 10-gram aliquots of surface soil. A FIDLER survey in conjunction with soil samples collected at a number of FIDLER locations would, in general, be a better approach. Then it might be possible to relate the variability and level of FIDLER readings to those for soil sample concentrations in order to approximate the optimum number of samples for each strata. Of course, the total funds available for the study put an upper limit on the total number of samples that can be collected and analyzed. The number of samples allocated to the various strata could also involve costs of collecting samples if this cost varies substantially between strata. Gilbert *et al.* (1976a) discuss how the allocation was accomplished for the Area 13 (Project 57) safety-test site on NTS.

Guideline 3: Use Random Sampling Within Strata or Random Sampling in Conjunction With Systematic Sampling

Careful consideration needs to be given to the method used to determine the actual sampling locations. Statisticians tend on theoretical grounds to favor random over systematic sampling. The theory of statistics that permits the formulation of probability statements and inferences about the universe from which sample data are taken is based on the notion of drawing samples at random. However, the greater ease with which systematic samples on a grid, say, can be collected under field conditions, and the need in many cases to insure that all portions of the study site are sampled are reasons for considering systematic sampling.

Gilbert *et al.* (1975) and Gilbert (1976) found that the use of random sampling within strata tended to leave gaps in the pattern of sample location points which contributed to biases observed in estimated Pu contours obtained using computer algorithms. This problem might have been reduced had a systematic (grid) sampling plan been used within each stratum to insure more uniform coverage. The grid spacing could have varied between strata in accordance with the allocation of samples obtained using the ideas discussed above under Guideline 2.

An alternative sampling plan for these safety-test sites would make use of both systematic and random sampling. One approach would be to grid off each stratum (different grid sizes being allowed for the various strata) and to choose two or more samples at random within each grid square. The grid size could be chosen to insure that the maximum possible distance between samples is acceptably short, while the number of samples within each grid square could be chosen on the basis of the variability expected between samples. This approach attempts to use the best features of both systematic and random sampling.

SUMMARY

The design of a transuranic field study must take into account site specific characteristics of the contamination such as deposition patterns, and the quantity, kind, and properties of the transuranics present. However, general design guidelines can be formulated to aid in the design of these studies. Principal planning steps include (1) a clear statement of objectives, (2) appropriate definition of that part of the environment about which information is desired (the "target population") and that part from which samples are actually taken and to which inferences apply (the "sampled population"), (3) determining the kinds of data and the degree of precision required to meet study objectives, (4) appropriate choice of measurement instrumentation and laboratory procedures, (5) division of the sampled population into sampling units, and (6) design or selection procedure used to select sampling units for analysis.

The design of studies at sites contaminated by local sources of contamination may include surveys using ground or aerial *in situ* radiation detection devices. These can help define the spatial pattern of contamination and, in some situations, be used in conjunction with wet chemistry analysis of soil samples to estimate total amounts or average concentrations using double sampling. Stratified random sampling has been shown to be an efficient design for totals or averages at safety-shot sites. Selecting soil samples from undisturbed locations is an acceptable procedure when estimating total accumulated deposition from worldwide fallout, or from local sources such as stack emissions where the size of Pu particles is in the micron range. However, undisturbed sites are, in general, not appropriate at safety-shot or nuclear detonation sites where patterns of deposition may be complex and/or when interest centers on determining the present deposition pattern.

Acceptance sampling by attributes or measurements are two possible approaches for deciding whether cleanup of an area is necessary. When using the attribute approach, the decision is made on the basis of the proportion of sample concentrations exceeding some present level. No assumption of normality is needed, but the number of samples required may be greater than when using the measurement approach. The measurement approach uses average concentrations to make a decision and depends on the assumption of normality for its validity. Sequential sampling plans seem to offer the most promise for cleanup studies. Three general guidelines are suggested relative to the design of transuranic field studies: (1) divide the study area into homogeneous subareas (strata) if possible, (2) conduct pilot studies if necessary to obtain data for planning the main sampling effort, and (3) within each stratum use random, systematic, or some combination of random and systematic sampling unless study objectives specifically require that only "undisturbed" locations are appropriate.

ACKNOWLEDGMENTS

We would like to thank Pam Doctor for reading the manuscript and offering suggestions for improvements. This paper was funded by the Nevada Applied Ecology Group, USERDA, Nevada Operations Office and by the Division of Biomedical and Environmental Research, USERDA, Washington, D.C., under Contract EY-76-C-06-1830.

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SOME NEW STATISTICAL CONCEPTS FOR QUALITY CONTROL

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ABSTRACT

A statistical technique is presented whereby radioisotope quality control can be substantially improved. The basis of this technique is an examination of the statistical distribution of the data.

INTRODUCTION

The rule generally used in most quality control work for setting acceptance limits is to arbitrarily accept some multiple of the standard deviation and use this along with the mean value to define acceptance or control limits. Currently there is significant discussion about what multiple of the standard deviation should be used and why one should choose any specified value. What is usually overlooked in these discussions is that this commonly used rule contains the inherent assumption that the data are normally distributed. The most frequently used multiple is three standard deviations which, if the assumption of normality is correct, implies a 1/4 percent probability that a good data value will be judged to be out of control.

We believe that the current discussion, which concerns what this probability value should be, is of minor consequence. What is of consequence is that the data satisfy the assumption of having a normal distribution. If this assumption is not reasonable for a data set, then a rule using a multiple of the standard deviation yields statistically invalid results, and some type of more sophisticated statistical algorithm must be developed.

DISCUSSION

We examined our own radioisotope quality control data to test the assumption of normality and we found that most of our data does not follow a normal distribution, even when the common transformations of data, such as the logarithm and square root, are used. Our data seem to be best described statistically as a heterogeneous normal distribution, also known as a mixture of normal distributions.

The following example of cesium-134 is typical of our data. I will present the example in the sequence of the data analysis steps. The first step is to plot the cumulative distribution of the data on normal probability paper. The plot of the example data is shown in Figure 1. It is obvious that the data do not fall on one straight line, the requisite for a single normal distribution to describe the data. Note that the slopes of the top and bottom segments are the same, thus the possibility that they are two pieces of one distribution. The center portion of the curve could be composed of one- or two-line segments. These questions of the components of the curve must be resolved by statistical test. The several possible structures were programmed as statistical functions and compared using the maximum likelihood ratio test. In more detail, these steps are as follows.

We start with a statistical model of the data distribution. The model for a mixture of three normal distributions is shown in Figure 2. The parameters denoted as "p" determine the proportion of the data "explained" by each of the component normal distributions. Since the total of all proportions must equal 100 percent of the data, two of these proportions are sufficient to define all three components. The extensions of this formula to two components, or more than three, should be obvious. A three-component model has eight parameters to be determined from the data. A not so obvious extension is to restrict mean values or standard deviations to be equal. For example, in Figure 2 we might replace the u_3 with with a u_2 so that u_2 appears twice in the formula giving a seven-parameter model. In about one-fifth of our data sets, though not in the example presented here, this restriction to equal mean values was statistically the best model.

The next step is to combine the model with the observed data using the likelihood function. This step is explained in detail in most elementary mathematical statistics textbooks.

After we have the likelihood function, we wish to choose those parameter values (in our case the "p's," "u's," and "σ's") that maximize the likelihood function; this is the statistical principle called maximum likelihood estimation. The discipline of computer sciences has recently provided some generalized functional maximization programs which can act

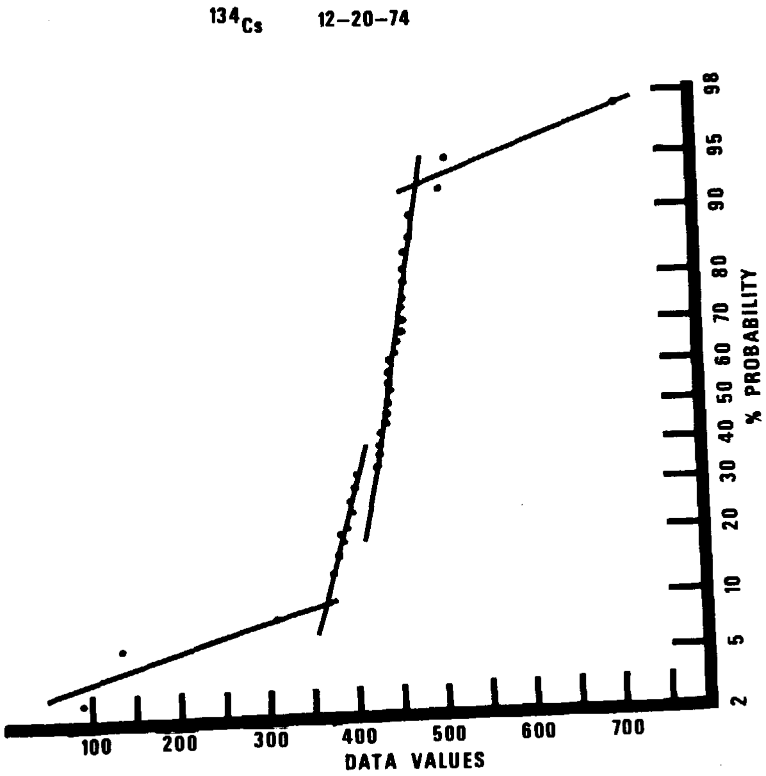

Figure 1. Cumulative Normal Probability Plot

$$f(x)=p_1*N(u_1,\sigma_1)+p_2*N(u_2,\sigma_2)+(1-p_1-p_2)N(u_3,\sigma_3)$$

$$\text{where}\quad N(u,\sigma)=\frac{1}{\sigma\sqrt{2\pi}}e^{-\frac{1}{2}\left(\frac{x-u}{\sigma}\right)^2}$$

Figure 2. Mathematical Expression of a Mixture of Normal Distributions

directly upon the likelihood function to find its maximum and the associated parameter values. The particular maximization technique used on this data was the "Simplex" algorithm; there are currently available a number of other good algorithms.

The computations outlined above are performed for a sequence of successively more complex statistical models of mixtures of normal distributions, starting with a single normal distribution, then a mixture of two normal distributions with the same mean, then two normal distributions with different means, and so on. The choice of the most complicated model to try is made from the cumulative distribution plot, such as shown in Figure 1. In our data a maximum of four distributions was used. These models were compared using the likelihood ratio test; the details of this test are also found in elementary mathematical statistics texts. Note that, with this test, we cannot decide if any one model is a good fit to the data or not, we can only decide the relative merits of the models used. The successively more complex models were pairwise compared using the likelihood ratio principle until no statistically significant improvement in the fit to the data was found.

A model consisting of a mixture of three normal distributions was found to best represent our example data; these distributions are plotted in the black curves on Figure 3. The broken line curve was calculated from the sample mean and standard deviation of all the data assuming a single normal distribution. The relative size of the black curves, the area under these curves, is drawn here to represent the proportions of the data in each of the component distributions. The broken line curve is simply drawn a convenient size. Specifically this data set is best modeled by 21 percent of the data having a mean of 390 and a standard deviation of 195 (the wide curve at the bottom), 22 percent of the data having a mean of 394 and a standard deviation of 11 (the lower middle curve), and 57 percent of the data having a mean of 454 and a standard deviation of 13 (the tallest curve).

Some additional information is of interest. The total sample size was 37. The "known value" of the material sent to the laboratories was 452, essentially the same as the 454 mean of the tallest curve, thus we suspect that this component curve represents the "good" laboratories. The average of all the data, represented by the broken line curve, was 440, which, if used to characterize the data set, suggests a bias from the known value--a bias that disappears if we use the component curves. Consider the range of concentrations defining the 95-percent area of the "good" or tallest curve. Eleven percent of the wide distribution is within this good range. Since the wide group represents 21 percent of the data, about 2 percent (21%*11%) of the data values would be misclassified as good when in fact the values are from poorly performing laboratories that just happened to hit it right this time. Less than one percent of the lower middle curve is actually within this 95 percent interval of the upper middle curve. One could, of course, repeat this exercise with any acceptance criteria one wished. We suspect the lower middle group represents a group of eight laboratories with good

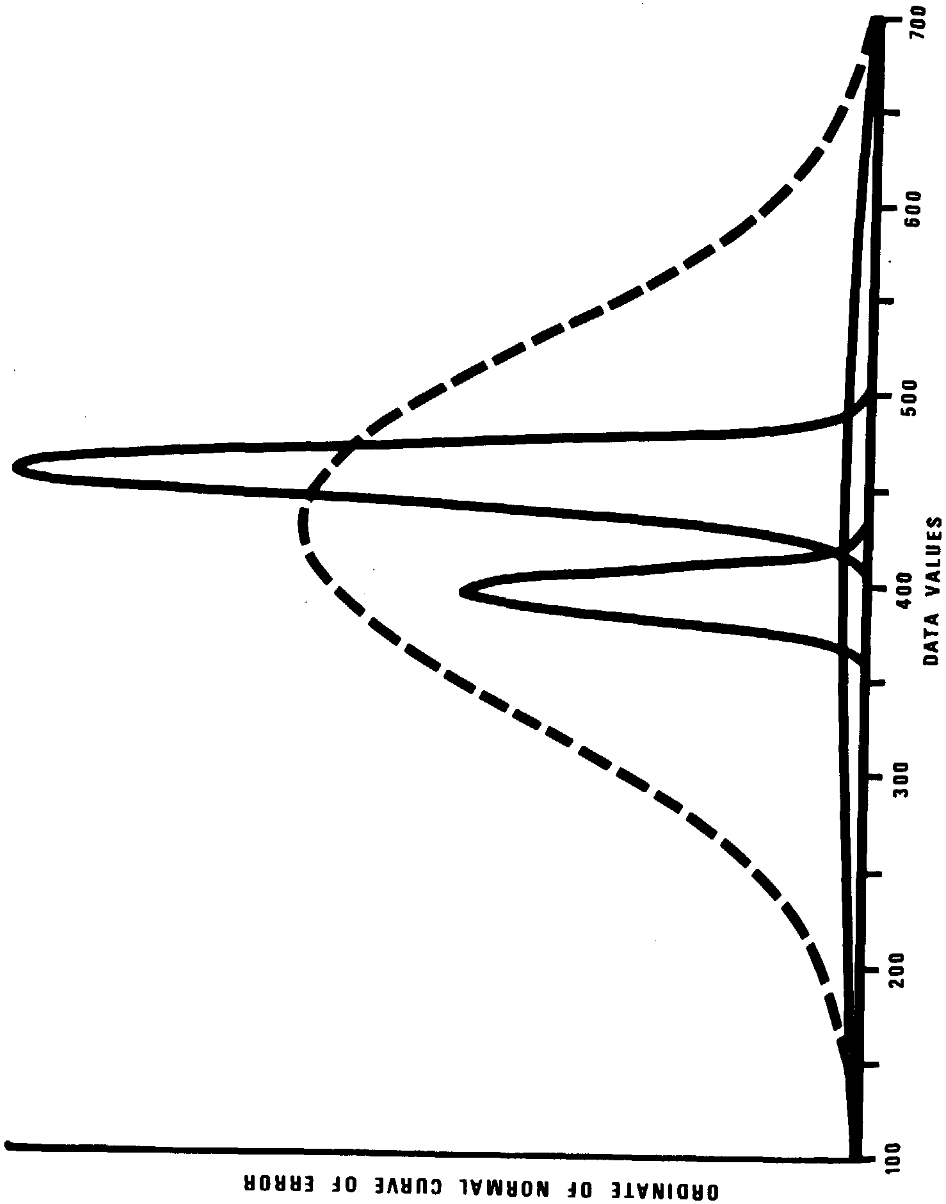

Figure 3. Estimated Population of Sampled Data

quality control but a bad standard because the standard deviations of the two middle groups are not significantly different statistically. We have not had the opportunity to investigate this type of possible cause and by now, since the data are 2 years old, the standards and calibrations used on these data may not be recoverable. Had the possibility of this situation been suspected in advance of the actual cross-check test, sufficient information could have been collected to investigate this and other possible causes.

It is obvious that if any reasonable quality control standard were applied to the total data set of this example, represented by the broken line curve in Figure 3, the result would be an unreasonable set of acceptance limits as well as a loss of information contained by the data. Perhaps our current uneasiness with acceptance rules is due to our intuitive recognition of this unreasonableness. We err in placing the blame on the acceptance rule rather than on the type of data we work with.

CONCLUSION

This is just one of about 60 quality control data sets involving 10 different radioisotopes that have so far been examined in our laboratory, all but one showed a statistically significant (maximum likelihood ratio test) mixture of distributions. Similar statistical work has also found such mixtures in thermoluminescent dosimeter data, plutonium in soil data, and air pollutant data. The plutonium in soil mixture-of-distributions has been identified as caused by two distinctive sizes of plutonium particles by using autoradiographs of soil samples; about 85 percent of the plutonium in the soils found at the Nevada Test Site is in the form of an extremely fine dust and 15 percent is particles large enough to be counted under a dissecting microscope. This has been called the "hot particle problem." Mixtures of distributions are a well known and accepted phenomena in medical statistics where the components distributions are typically identified as sick and well patients, male and female, and so on.

Our quality control data differ from the hot particle problem and the medical examples cited above in the reasons for the structure in the data. The structures in the hot particle situation and in the medical examples are due to a true structure in the media sampled. In our quality control data, aliquots of a single homogeneous laboratory-prepared medium are sent to the participants, and the structure is introduced during the sample analysis procedure. There is nothing in the present statistical analysis to distinguish the various causes of a heterogeneous data structure. The statistical analysis is the same whatever the cause. The cause can only be defined by follow-up investigations.

When several measurements of different variables are available on each of the experimental subjects, this mixture of distributions situation becomes the well established discriminant analysis and statistical classification problem. In our case of finding a mixture of distributions in quality control data, the next statistical step is to go back to the laboratories involved. and collect data on their operating procedures and to collect quantitative data on as many variables as possible. A classical discriminant analysis on these additional data would then tell us which variables or factors are important in distinguishing "good" from "bad" laboratories and what values of those variables are characteristic of a good laboratory. This information can then be used to help turn the "bad" laboratories into "good" laboratories--that's the real purpose of quality control.

RATIO ESTIMATION TECHNIQUES IN THE ANALYSIS OF ENVIRONMENTAL TRANSURANIC DATA

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ABSTRACT

Ratios play a prominent role in the analysis of environmental transuranic data. The mathematical assumptions underlying the use of ratios are presented. The properties of seven different estimators of an average ratio and their standard errors are discussed with respect to their applicability to transuranic field studies. The behavior of these estimators, their standard errors, and confidence intervals are compared for three representative sets of data from safety-shot sites at the Nevada Test Site and Tonapah Test Range. Guidelines are given for deciding if an average ratio is appropriate. Recommendations for avoiding an inappropriate estimator are discussed.

INTRODUCTION

The purpose of this paper is to discuss the properties of ratios as they apply to environmental transuranic studies. We first review some ways ratios are used in transuranic field studies and some types of statistical analyses currently being applied to ratios. We also discuss several methods for computing an average ratio giving the mathematical and statistical assumptions underlying each method and the factors to consider in choosing a method for a particular set of data. These methods are illustrated using data from Nevada Applied Ecology Group (NAEG) studies conducted at safety-shot sites at the Nevada Test Site (NTS) and Tonopah Test Range (TTR). Finally, some recommendations are listed for (1) deciding if an average ratio is a suitable quantity to summarize a set of data, and (if (1) is true) then (2) choosing the most appropriate estimator of the average ratio.

USE OF RATIOS IN TRANSURANIC FIELD STUDIES

Ratios play a prominent role in environmental transuranic studies. A major application is to ecosystem models where a ratio is used to express the relationship between transuranic concentrations in various ecosystem components. Two examples are found in the Proceedings of the Workshop on Environmental Research for Transuranic Elements (1976, pp. 23-24): the concentration ratio (CR), defined as

$$\frac{\text{activity per weight of plant part}}{\text{activity per weight of substrate or reference material}},$$

and the inventory ratio (IR), defined as

$$\frac{\text{activity per unit area in product}}{\text{activity per unit area in source}}.$$

Other uses of ratios in environmental transuranic studies include detecting whether differential movement between two elements, say Pu and Am, is occurring in the soil profile, and as a means of detecting local contamination by some transuranic element by noting a change in its ratio to another element (or isotope) from their background level ratio.

This paper deals mainly with the use of average ratios. It is important to realize, however, that there are situations where an average ratio is not appropriate, e.g. when there is a systematic change in the ratio over space or time. An illustration of this is the detection of local contamination mentioned above. Suppose we have the situation in Figure 1: a ^{239}Pu source and sampling sites located downwind from it. Rather than calculate an average ratio of ^{239}Pu to ^{238}Pu over all sites, it would seem more appropriate to know how the ratios change with increasing distance from ground zero.

Ratio data have been presented in a variety of ways in the environmental transuranic literature. A simple list of ratios or average ratios from different sites is sometimes given. This is appropriate when the data come from different study sites and there is no justification for pooling the data or for performing any further statistical treatment. Ratios have also been presented in histogram form. This was done, for example, by Emery et al. (1976), to compare the distribution of ratios in various ecosystem components in their study of U-pond at Hanford. One can also plot the numerator versus the denominator or the logarithms of the numerator versus logarithms of the denominator and view the estimation of a ratio as a regression through the origin. Alternatively, ratio data are often presented as an average ratio, sometimes with an estimated standard error, but often without any indication as to how the average or its standard error were calculated.

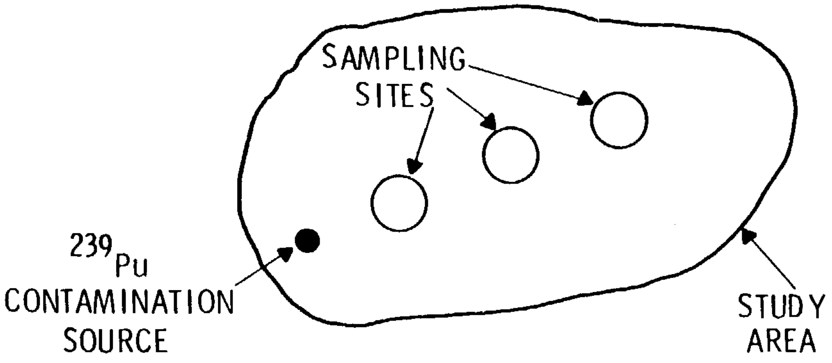

Figure 1. Change in Ratio ^{239}Pu/^{238}Pu Over Space

There are certain problems associated with estimating an average ratio. The statistical properties of ratios have not been fully developed and the properties of average ratios that are known do not appear to be widely understood. It is known, e.g. that the estimated average ratio is often statistically biased and in most cases the variance of the estimate is only an approximation (Cochran, 1963, p. 160-163). The problem is further compounded by the many ways an average ratio can be estimated.

METHODS OF COMPUTING AN AVERAGE RATIO

The mathematical assumption underlying the calculation of an average ratio between two quantities Y and X is that the true ratio is constant at each value of X and that if X = 0, then Y = 0. This implies that a plot of Y against X is a straight line through the origin (illustrated by line A, Figure 2). If this is true then the ratio Y/X which is the slope of the line (in this case 0.5), is constant for all values of X. If the relationship between Y and X is a straight line, but not through the origin (line B, Figure 2), then the ratio Y/X changes for different values of X (from 1.5 to .7) although the slope of the line is the same (0.5) as that of line A.

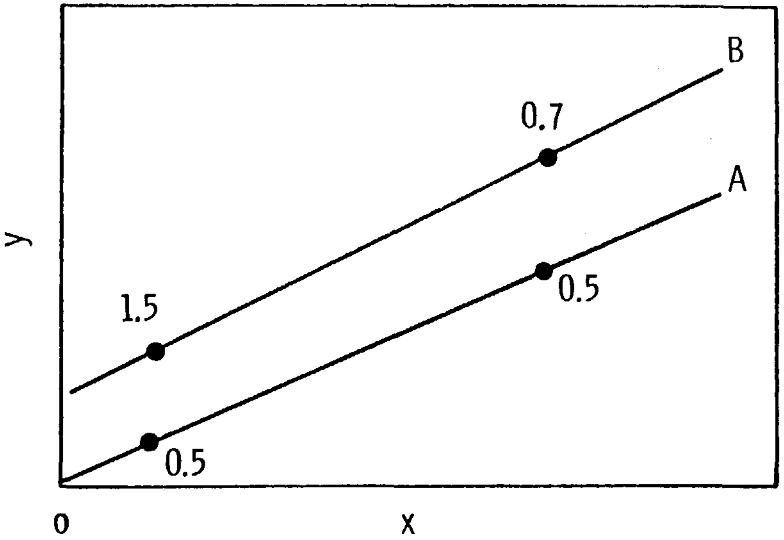

FIGURE 2. Relationship of Ratios to Regression

In transuranic field studies both the numerator and denominator are often highly variable. However, the classical approach to ratio estimation is one of regression through the origin applied to an experimental situation which assumes that the numerator has variability but that the denominator, which is under the control of the experimenter, is constant except for possible measurement errors. In this latter situation the optimal estimator of the ratio depends on the relationship of the variance of Y to the value of X at which it is measured.

Consider first the case where the denominator has very little variability relative to the numerator so that the classical approach is justified. We assume the regression goes through the origin so that the slope and the average ratio are synonymous. Suppose the variance of Y is the same for all values of X (Figure 3). This relationship is illustrated by dotted lines representing roughly ± one standard deviation from the regression line. In this situation the slope of the regression line (estimate of the average ratio) is given by

$$R_1 = \sum_{i=1}^{n} X_iY_i \Big/ \sum_{i=1}^{n} X_i^2 \ ,$$

where n is the number of data pairs (Y_i,X_i). Its variance is given by

$$V(R_1) = \left\{ \sum_{i=1}^{n} Y_i^2 - \left(\sum_{i=1}^{n} X_iY_i \right)^2 \Big/ \sum_{i=1}^{n} X_i^2 \right\} \Big/ (n-1) \sum_{i=1}^{n} X_i^2 \ .$$

If the distribution of Y about the regression line is normal for a given X, then the 100(1-α) percent confidence interval for R_1 is estimated by

$$R_1 \pm t_\alpha \sqrt{V(R_1)} \ ,$$

where t_α is the value of the t distribution with (n-1) degrees of freedom and confidence probability 1-α. Commonly chosen values of α are .05 or .01 giving 95% or 99% confidence intervals, respectively.

If the variance of Y increases proportional to X as X increases (illustrated by dotted lines in Figure 4), then the optimal estimator of the average ratio (slope) is $R_2 = \overline{Y}/\overline{X}$, where

$$\overline{Y} = \frac{1}{n} \sum_{i=1}^{n} Y_i \quad \text{and} \quad \overline{X} = \frac{1}{n} \sum_{i=1}^{n} X_i$$

The variance and confidence interval are

$$V(R_2) = \left\{ \sum_{i=1}^{n} \left(\frac{Y_i^{\,2}}{X_i} \right) - \left(\sum_{i=1}^{n} Y_i \right)^2 \bigg/ \sum_{i=1}^{n} X_i \right\} \bigg/ (n-1) \sum_{i=1}^{n} X_i^{\,2} \qquad (1)$$

and

$$R_2 \pm t_\alpha \sqrt{V(R_2)},$$

respectively, where t_α is defined above.

A third situation (Figure 5) is when the standard deviation of Y is proportional to X. The optimal estimator of the slope of the regression line (average ratio) is then

$$R_3 = \frac{1}{n} \sum_{i=1}^{n} \frac{Y_i}{X_i}$$

Its variance is estimated by

$$V(R_3) = \left\{ \sum_{i=1}^{n} \left(\frac{Y_i}{X_i} \right)^2 - \frac{1}{n} \left(\sum_{i=1}^{n} \frac{Y_i}{X_i} \right)^2 \right\} \bigg/ n(n-1),$$

and its confidence interval by

$$R_3 \pm t_\alpha \sqrt{V(R_3)}.$$

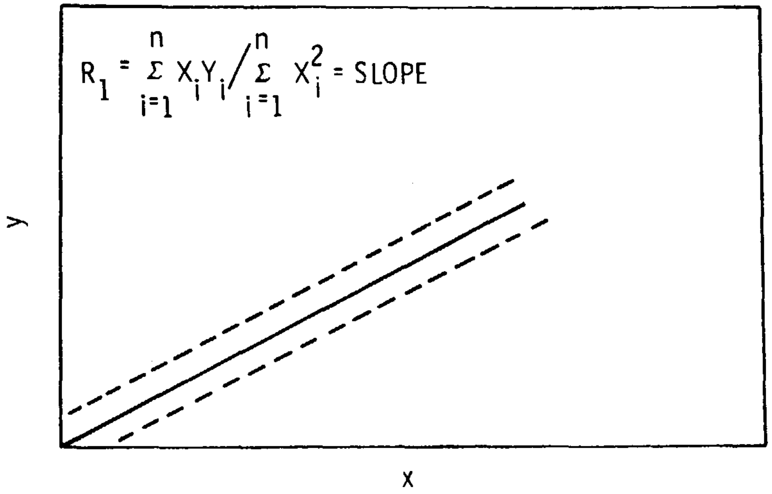

FIGURE 3. Ratio Estimation When Var(Y) is Independent of X.

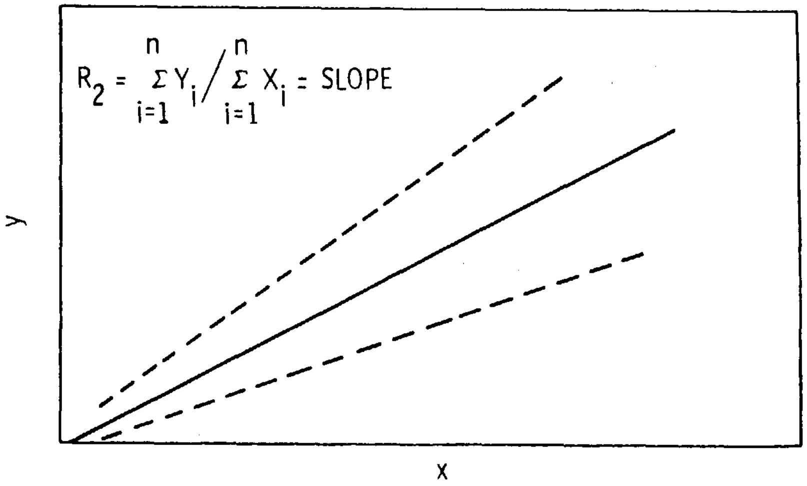

FIGURE 4. Ratio Estimation When Var(Y) is Proportional to X .

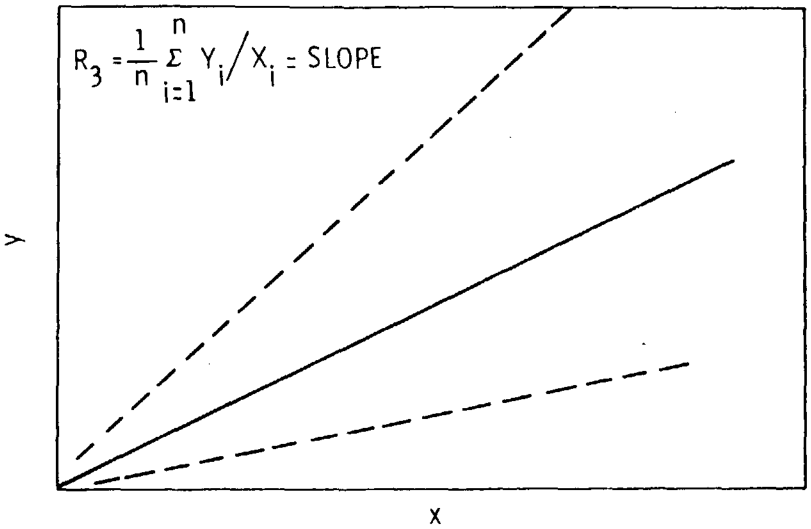

FIGURE 5. Ratio Estimation When Var(Y) is Proportional to X^2.

Recall that these estimators are based on the assumption that X is known with little error. In that case, each estimate (R_1, R_2, R_3) is statistically unbiased and the choice between them depends on whether the variance of Y is constant, proportional to X, or proportional to X^2 (Snedecor and Cochran, 1967 pp. 166-170). But what if X is not "constant", the usual case in transuranic field studies? For example, Y and X might be $^{239-240}$Pu and ^{241}Am concentrations, respectively, in the same aliquot of soil. Cochran (1963, p. 161) shows that the ratio estimate $R_2 = \bar{Y}/\bar{X}$, even though Y and X are both random variables, is an unbiased estimate of the ratio of the true means if the regression of Y on X is a straight line through the origin. Hence, regardless of whether X is constant or variable, if the data forms a straight line through the origin, $\bar{Y}/\bar{X}$ is an unbiased estimate of the ratio of true means. However, when X is a variable, the variance of R_2 is only approximate and equation (1) may not be appropriate for estimating Var(R_2). Cochran's (1963, p. 31) approximate variance for R_2 is

$$V'(R_2) = \sum_{i=1}^{n} (Y_i - R_2X_i)^2/n(n-1)\bar{X}^2 \quad . \qquad (2)$$

The approximate confidence interval for R_2 (assuming R_2 is normally distributed) is

$$R_2 \pm z_\alpha\sqrt{V'(R_2)} \quad , \qquad (3)$$

where z_α is the value of the standard normal deviate corresponding to the appropriate probability α. The assumption of normality is reasonably good if the coefficients of variation of both $\bar{Y}$ and $\bar{X}$, $s_Y/(\sqrt{n}\,\bar{Y})$ and $s_X/(\sqrt{n}\,\bar{X})$, respectively, are less than 0.1 and the number of observations is greater than 30 (Cochran, 1963, p. 164). If these conditions do not apply, $V'(R_2)$ (equation 2) will tend to be too small. Cochran (1963, p. 164) gives an alternative method for computing confidence limits of R_2 that should be more accurate than the above method (equation 3), but is also more difficult to compute. When the sample size is small, the t distribution rather than the standard normal is recommended for calculating the confidence interval.

When the straight line is restricted to pass through the origin, as in our case to estimate an average ratio, and both X and Y are normally distributed, then the maximum likelihood estimate of the slope (common ratio) is also $R_2 = \bar{Y}/\bar{X}$ (Creasy, 1956). The variance of the observations from the regression line (line with slope R_2) was approximated by Creasy (1956) as

$$s^2_{Y \cdot X} = \frac{\sum_{i=1}^{n} (Y_i-\bar{Y})^2 - \left(\sum_{i=1}^{n} Y_i \Big/ \sum_{i=1}^{n} X_i\right) \sum_{i=1}^{n} (X_i-\bar{X})\, Y_i}{n-1} \quad . \qquad (4)$$

The variance of R_2 for this case has not been completely derived, but Ricker (1973) recommends

$$V''(R_2) = s^2_{Y \cdot X} \Big/ \sum_{i=1}^{n} (X_i-\bar{X})^2 \qquad (5)$$

(where $s^2_{Y \cdot X}$ is computed using equation 4) as being as good an approximation as the computationally involved method proposed by Creasy. The confidence interval is then given by

$$R_2 \pm t_\alpha\sqrt{V''(R_2)}$$

The effects on the variance of R_2 when Y and/or X are not normally distributed are not fully understood, except for a few exceptions where some exact results are known (Mielke and Flueck, 1976). One of these exceptions that is potentially applicable to radionuclide data is where Y and X are distributed as bivariate lognormal variables. With Y and X both lognormally distributed, the ratio Y/X is also lognormally distributed (Aitchison and Brown, 1969, p. 11) so that the quantity

$$R_4 = \exp\left[\frac{1}{n} \sum_{i=1}^{n} \log_e \frac{Y_i}{X_i}\right]$$

is an estimate of the <u>median</u> ratio in the original (untransformed) scale. An approximate confidence interval on the true median ratio is given by the antilogarithms of the lower and upper confidence limits U_L and U_H for

$$\frac{1}{n} \sum_{i=1}^{n} \log_e \frac{Y_i}{X_i} .$$

U_L and U_H are easily obtained since

$$\frac{1}{n} \sum_{i=1}^{n} \log_e \frac{Y_i}{X_i}$$

is normally distributed when Y_i/X_i is lognormal. Thus, an approximate confidence interval for R_4 is given by the limits

$$R_4 - \exp\left\{U_L\right\} \text{ and } R_4 + \exp\left\{U_H\right\} . \tag{6}$$

Another estimate of the average ratio is the median of the observed ratios denoted here as R_5. R_5 is nonparametric in the sense that no assumptions on the form of the distributions of Y, X, or Y/X are made. As an example of the computation of R_5 suppose we have a sample of 5 observations ranked from smallest to largest, say 18, 19, 23, 28 and 35. The sample median (R_5) is the middle observation, or in this case 23. It estimates the median value that would be obtained if all possible observations had been taken. An advantage of R_5 is that confidence limits are easily calculated from tables of the binomial distribution, as illustrated e.g. by Snedecor and Cochran (1967, p. 124) or Conover (1971, p. 111).

At this point, we can now make a few general recommendations on how to proceed when estimating an average ratio. First, view the problem as a regression of Y on X and plot Y versus X. If a straight line relationship through the origin appears reasonable, then the average ratio given by $\overline{Y}/\overline{X}$ should be statistically unbiased. If the relationship is a straight line, but not through the origin, then the ratio is not constant for all X. In this case an average ratio does not adequately summarize the data and it is better to estimate the regression of Y on X with a non-zero intercept.

We note that the bias discussed in this paper refers to only statistical bias, i.e. the bias in the estimate arising due to the data not completely satisfying the assumptions underlying the statistical treatment of the data. We ignore here any systematic biases that may be introduced due to such things as inappropriate field sampling or sample preparation techniques and analytical bias in the laboratory.

NUMERICAL EXAMPLES

We have chosen three sets of data from NAEG studies at safety-shot sites on NTS and TTR to illustrate and compare the various methods of calculating estimates of an average ratio.

The first set of data consists of ^{241}AM and $^{239-240}$Pu concentrations on the same aliquot from a soil sample collected at 15 random locations from stratum 3 of the Double Track site, TTR (Gilbert, <u>et al</u>, 1975). The ratio of interest here is Pu to Am. A plot of the data is given in Figure 6, where the points are labeled by the value of their ratio.

A straight line through the origin seems to fit the data very well. The ratios of the 15 data pairs are ordered from smallest to largest at the top of the figure. The ratio estimates, their standard errors and 95% confidence intervals (CI) are given in Table 1. The first method is R_1 where X is considered to be a constant and Var(Y) is assumed <u>constant</u> for all X. Methods 2, 3 and 4 all use the same estimate $R_2 = \overline{Y}/\overline{X}$ but with three different estimates of variance, $V(R_2)$, $V'(R_2)$, and $V''(R_2)$, respectively. Method 2 is based on the assumption that X has no variability while methods 3 and 4 assume X is a variable. Method 2 assumes Var(Y) is proportional to X. Method 5 is R_3, the average of the individual ratios. It assumes that X is not a variable and that Var(Y) is proportional to X^2.

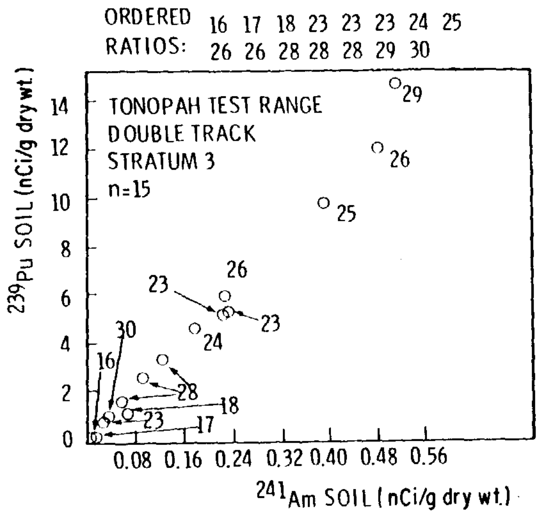

FIGURE 6. Example of Linear Regression Through the Origin.

The confidence limits for all five of these methods are based on the normal distribution i.e. that the estimated average ratio is normally distributed. In addition, Method 4 assumes that both X and Y are themselves normally distributed. Method 6 (R_4) is the estimated median of the ratios based on the assumption that the ratios are lognormally distributed. Method 7 (R_5), the median of the observed ratios, is independent of any distribution assumptions.

The range of the estimates in Table 1 is from 23.82 for the lognormal estimate, R_4, to 26.12 for R_1. R_4 falls into each confidence interval except for those of R_1 and Method 3. R_3 and R_5 are outside the confidence interval for R_1. The confidence intervals for the estimates based on a distributional form (Methods 1 to 6) are much smaller (2.34 to 5.69) than the one for the sample median (9.77). This is due to the fact that the observations are fairly evenly spaced. Since the data fit a straight line through the origin, all of the methods gives about the same estimate of the average ratio regardless of the distribution involved.

TABLE 1. Estimates of the Average Pu to Am Ratio in Soil, Double Track Site, Stratum 3, TTR.

Method	Estimator	N	Estimated Average Ratio	Standard Error	95% CI	Length of CI
1	$R_1 = \sum_{i=1}^{n} X_i Y_i \Big/ \sum_{i=1}^{n} X_i^2$	15	26.12	0.546	(24.95, 27.29)	2.34
2	$R_2 = \overline{Y}/\overline{X}$ with $V(R_2)$	15	25.58	1.34	(22.74, 28.43)	5.69
3	$R_2 = \overline{Y}/\overline{X}$ with $V'(R_2)$	15	25.58	0.763	(23.94, 27.22)	3.24
4	$R_2 = \overline{Y}/\overline{X}$ with $V''(R_2)$	15	25.58	1.12	(23.22, 27.95)	4.74
5	$R_3 = \frac{1}{n} \sum_{i=1}^{n} Y_i/X_i$	15	24.23	1.12	(21.84, 26.61)	4.77
6	$R_4 = \exp\left\{\frac{1}{n} \sum_{i=1}^{n} \log_e (Y_i/X_i)\right\}$	15	23.82	—	(21.39, 26.53)	5.14
7	R_5 = sample median	15	24.71	—	(18.32, 28.10)	9.77

The second set of data consists of ^{235}U concentrations in soil and associated vegetation from 17 random locations in Area 11, A Site, NTS. Gilbert and Eberhardt (1974) report that the ratio of vegetation mean to soil mean tends to decrease toward ground zero. However, the data from the three activity level strata are combined here to illustrate the effect on ratio estimators when a constant ratio is not present. The vegetation concentrations are plotted versus the corresponding soil concentrations in Figure 7. The individual ratios (at the top of Figure 7) are skewed with the bulk of the data lying below 0.10 with two moderately large values (0.22 and 0.25) and two exceedingly large values (2.0 and 3.0). The various ratio estimates, their standard errors, and the 95 percent confidence limits are given in Table 2.

The most striking aspect of this data is the larger mean ratio R_3 as compared to the other estimates (0.371 compared to 0.032, 0.045, 0.076, and 0.102). This is due to the two very large individual ratios and it illustrates the effect of using an arithmetic mean to estimate a "typical value" when data are highly skewed.

The other estimates are not as similar as those for the Pu/Am data. Note, e.g. that the confidence interval for R_1 does not overlap those for R_3, R_4 and R_5. The widest confidence interval for $R_2 = \bar{Y}/\bar{X}$, (0.005, 0.085), using $V(R_2)$ does not contain the lognormal estimate ($R_4 = 0.102$), while the smallest interval (0.026, 0.065) using $V'(R_2)$ does not contain the sample median ratio ($R_5 = 0.076$). The estimated standard error for R_2 using $V''(R_2)$ was impossible to calculate because of a negative variance. This set of data illustrates the diversity of average ratio estimates which is possible when the ratio does not remain constant throughout the data set, and when the distribution of the observations is skewed.

The third set of data consists of ^{238}U concentrations in soil and associated vegetation for the same 17 locations in Area 11, A Site, as the ^{235}U concentrations (Gilbert et al., 1975). The plot of the data is given in Figure 8 where similar to the ^{235}U data, there seems to be no linear relationship between concentrations of associated soil and vegetation samples. In addition, the ratios are skewed in a similar manner to the ^{235}U data.

Comparing the ratio estimates in Table 3, we again see, due to the skewness of the data, that the mean of the ratios ($R_3 = 0.0689$) is outside the 95% confidence limits of each of the other estimates. The other estimates lie in the confidence intervals of all methods with the exception of the sample median which lies outside the confidence interval for Method 3, which is extremely narrow.

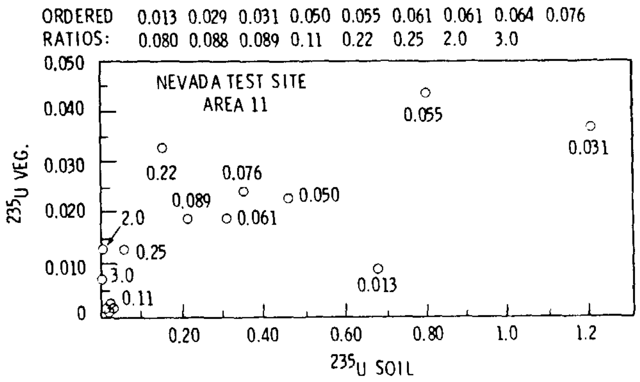

FIGURE 7. Vegetation vs Soil, ^{235}U (ug/g Dry)

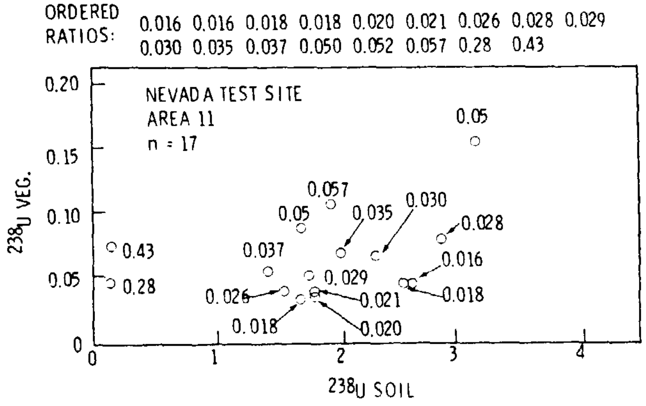

FIGURE 8. Vegetation vs Soil, ^{238}U (ug/g Dry)

Table 2. Estimates of Average Vegetation to Soil Ratios of ^{235}U Area 11, A Site, Strata Combined.

Method	Estimator	N	Estimated Average Ratio	Standard Error	95% CI	Length of CI
1	$R_1 = \sum_{i=1}^{n} X_iY_i \Big/ \sum_{i=1}^{n} X_i^2$	17	0.032	0.003	(0.025, 0.038)	0.013
2	$R_2 = \overline{Y}/\overline{X}$ with $V(R_2)$	17	0.045	0.019	(0.005, 0.085)	0.080
3	$R_2 = \overline{Y}/\overline{X}$ with $V'(R_2)$	17	0.045	0.0092	(0.026, 0.065)	0.026
4	$R_2 = \overline{Y}/\overline{X}$ with $V''(R_2)$	17	0.045	unable to calculate due to negative variance		
5	$R_3 = \frac{1}{n}\sum_{i=1}^{n} Y_i/X_i$	17	0.371	0.201	(0.055, 0.796)	0.852
6	$R_4 = \exp\left\{\frac{1}{n}\sum_{i=1}^{n}\log_e(Y_i/X_i)\right\}$	17	0.102	——	(0.050, 0.208)	0.158
7	R_5 = sample median	17	0.076	——	(0.045, 0.109)	0.064

Table 3. Estimates of Average Vegetation to Soil Ratios of ^{238}U, Area 11, A Site, Strata Combined.

Method	Estimator	N	Estimated Average Ratio	Standard Error	95% CI	Length of CI
1	$R_1 = \sum_{i=1}^{n} X_iY_i \Big/ \sum_{i=1}^{n} X_i^2$	17	0.0307	0.0041	(0.0220, 0.0394)	0.017
2	$R_2 = \overline{Y}/\overline{X}$ with $V(R_2)$	17	0.0336	0.009	(0.0142, 0.0530)	0.039
3	$R_2 = \overline{Y}/\overline{X}$ with $V'(R_2)$	17	0.0336	0.00015	(0.0333, 0.0339)	0.0006
4	$R_2 = \overline{Y}/\overline{X}$ with $V''(R_2)$	17	0.0336	0.003	(0.0278, 0.0394)	0.012
5	$R_3 = \frac{1}{n}\sum_{i=1}^{n} Y_i/X_i$	17	0.0689	0.0274	(0.0107, 0.1270)	0.116
6	$R_4 = \exp\left\{\frac{1}{n}\sum_{i=1}^{n}\log_e(Y_i/X_i)\right\}$	17	0.0374	——	(0.0231, 0.0604)	0.037
7	R_5 = sample median	17	0.0293	——	(0.0185, 0.0501)	0.032

The surprising thing about this data set is how closely the ratio estimates do agree as compared to the ^{235}U data, despite the apparent similarity in structure of the two data sets.

CONCLUSIONS

We have discussed the assumptions underlying some ratio estimates, their standard errors, and confidence intervals. Their behavior was compared for three sets of data from NAEG studies. As a consequence of this, there are several points that should be considered before calculating an average ratio. These are

1. Plot the data for each variable against a spatial or time coordinate to see if there is a pattern. Calculating an overall ratio when there is a trend with location or time may obscure an important relationship. The ^{235}U and ^{238}U data sets are possible examples.
2. Plot the data (Y versus X) to see if a linear relationship through the origin seems reasonable. If not, an average ratio may not be the appropriate quantity to describe the phenomena.
3. Order the ratios to get an indication of their distribution, particularly with respect to skewness. If the distribution of the ratios is skewed,

$$\frac{1}{n}\sum_{i=1}^{n} Y_i/X_i$$

   will be unduly influenced by extraordinarily large ratios Y_i/X_i.
4. The sample median of the ratios is a "safe" estimate of the average ratio in the sense that no assumption on the type of distribution is required. However, the length of the confidence interval may be large.

ACKNOWLEDGMENT

This paper was funded in part by the Nevada Applied Ecology Group (NAEG), ERDA/NV, and in part by DBER, ERDA/HQ under Contract No. EY-76-C-06-1830.

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NEVADA APPLIED ECOLOGY GROUP

MODEL FOR ESTIMATING PLUTONIUM TRANSPORT AND DOSE TO MAN

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ABSTRACT

A Standard Man is assumed to live in and obtain most of his food from a plutonium-contaminated area at the Nevada Test Site (NTS). A plutonium transport model, based on the results of other Nevada Applied Ecology Group plutonium studies, is used to estimate potential chronic rates of ^{239}Pu inhalation and ingestion as functions of the average concentration of ^{239}Pu (C_s = pCi/g) in the surface soil (0 to 5 cm depth) of the reference area. Assuming the transport system to be in steady state, the estimated inhalation and ingestion rates (pCi/day) are 0.002 C_s and 0.2 C_s, respectively.

A dose estimation model based on parameters values recommended in publications of the International Commission on Radiological Protection is used to estimate organ burdens, accumulated doses, and dose commitments as functions of exposure time. Estimated doses (rems) due to chronic inhalation and ingestion of ^{239}Pu for 50 years at the rates indicated above are: thoracic lymph nodes, 0.610 C_s; lungs, 0.026 C_s; bone, 0.014 C_s; liver, 0.009 C_s; kidney, 0.003 C_s; total body, 0.0007 C_s; and gastrointestinal tract (lower large intestine), 0.0002 C_s. Inhalation accounts for 100 percent of the estimated dose to lungs and thoracic lymph nodes and for about 95 percent of the estimated dose to bone, liver, kidney, and total body. Ingestion accounts for > 99 percent of the dose to the gastrointestinal tract.

According to ICRP recommendations for individual members of the public, the dose rate to the lungs after 50 years' exposure should not exceed 1.5 rems/year. The plutonium transport and dose estimation model described in the report indicate that the average concentration of ^{239}Pu in the surface (0 to 5 cm) soils of contaminated areas at NTS which could result in a maximum dose rate of 1.5 rems/year to the lungs is approximately 2.8 nCi/g or about 140 μCi/m^2 for soils weighing 1.0 g/cm^3.

INTRODUCTION

An important goal of the Nevada Applied Ecology Group (NAEG) Plutonium Program is to evaluate the potential radiological hazard to man due to the presence of plutonium in various nuclear safety test areas at the Nevada Test Site (NTS). As the contaminated areas of interest are uninhabited, we have based our analysis on the assumption that a Standard Man resides in and obtains most of his food from a plutonium-contaminated area at NTS.

In this report, we utilize information provided by other NAEG studies to develop a plutonium transport model which attempts to characterize the general behavior of plutonium in a typical NTS ecosystem and provides a basis for estimating potential rates of plutonium ingestion and inhalation by the hypothetical Standard Man.

These estimates of inhalation and ingestion rates provide the input for a dose estimation model which is used to calculate potential organ burdens, cumulative organ doses, and dose commitments due to chronic inhalation and ingestion of ^{239}Pu. Though several are considered, the preferred dose estimation model is based entirely on the recommendations and publications of the International Commission on Radiological Protection (ICRP).

Finally, a procedure is described whereby the combined results of the transport model and the dose estimation model may be applied to the practical problem of deciding whether and to what extent environmental decontamination might be required to limit or reduce potential health hazards due to plutonium.

A preliminary model of potential plutonium transport from the environment to man was introduced during the planning stage of the NAEG plutonium program to insure consideration of laboratory and field studies which would provide the data and parameter estimates required for implementation of a more detailed transport and dose estimation model to be developed later in the program. Some of the parameters sought at the outset have proved to be elusive or impossible to measure accurately and, consequently, the proposed dynamic model has not been fully implemented. This report represents our best effort to judge and interpret the information currently available and to select the best available methods for estimating potential intake rates and doses. The design of the transport and dose estimation models plus the assumptions and parameter values selected for their implementation comprise what we believe to be a reasonable and conservative working hypothesis which provides a method for evaluating the potential health hazards associated with plutonium-contaminated areas at the NTS. As a working hypothesis, it is subject to continuing reappraisal, and the results or conclusions derived from it are subject to unavoidable uncertainties. To a considerable extent, however, these uncertainties are compensated for by conservative assumptions which tend to result in overestimates of potential intake rates, organ burdens, and doses rather than underestimates.

PLUTONIUM TRANSPORT TO MAN

Preliminary Model

Figure 1 is a diagram of the potential transport pathways considered in the preliminary planning model. The large square represents an arbitrary boundary of a contaminated area. Boxes represent the principal ecosystem compartments of interest, and arrows represent net transport via the pathways indicated. Arrows which cross the arbitrary boundary represent net transport out of the system.

The distribution of Pu in the contaminated areas of principal interest at the NTS has been described by Gilbert *et al.* (1975). Present levels of soil contamination in the areas of interest range from about 1.0 μCi/m^2 to >6,000 μCi/m^2. Because these levels of soil contamination resulted from nuclear safety tests carried out from 1954 through 1963, and because current fallout rates are insignificant compared with existing levels of contamination, Figure 1 shows no current Pu input to the system.

Under these conditions, the Pu concentration in soil is the principal factor forcing the transport system. Air is contaminated by resuspension of Pu-bearing soil particles. Vegetation is contaminated internally by root uptake from soil and externally by deposition of resuspended particles. Pu input to herbivores is due to ingestion of soil and vegetation and to inhalation. Pu could reach man by inhalation of contaminated air, by accidental ingestion of contaminated soil, and by ingestion of milk or meat (skeletal muscle or internal organs) from animals raised in the contaminated area. Drinking water for herbivores and man is assumed to come from deep wells or from sources outside the contaminated area and to contribute nothing to Pu intakes by herbivores or by man.

If it could be assumed that (1) the major ecosystem compartments and important transport pathways are as indicated in Figure 1, (2) the plutonium in each compartment is well mixed with the other contents of the compartment, and (3) the net rate of transfer from one compartment to another can be expressed as the product of a transfer coefficient and the quantity of plutonium in the transmitting compartment, then the intercompartmental flux of plutonium could be represented by a system of linear, first order, ordinary differential equations, the general formula for which is

$$\frac{dY_j}{dt} = \sum_{\substack{i=1\\ i\neq j}}^{n} \lambda_{ij} Y_i - Y_j \sum_{\substack{i=1\\ i\neq j}}^{n} \lambda_{ji} \qquad (1)$$

$$j = 1,2,3\ldots n$$

where j is the compartment of reference and all other compartments are designated i,

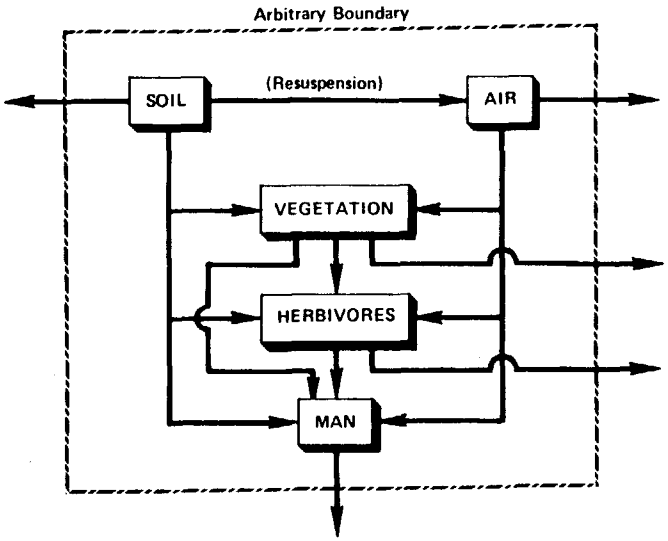


FIGURE 1. PRINCIPAL PATHWAYS OF PLUTONIUM TRANSPORT TO MAN

Y_j is the amount (pCi) of Pu in compartment j any time t (days),

and λ_{ij} and λ_{ji} are transfer coefficients (day^{-1}) for flows into and out of compartment j.

The positive expression on the right side of Equation (1) represents the flow rate into compartment j while the negative expression represents the rate of flow out of j. The amount present in a given compartment at a given time, $Y_j(t)$, is therefore dependent on the rates of input and output.

In a general way, Figure 1 and Equation (1) identify the principal kinds of information needed to estimate the transport of plutonium to man. The compartments of Figure 1 indicate the principal ecosystem components, and the arrows indicate the pathways of transport from environment to man via inhalation and ingestion. Equation (1) suggests that intercompartmental flow rates might be expressed as the product of a transfer coefficient and the quantity of plutonium in the transmitting compartment. It was recognized, however, that some parts of the transport system (Figure 1) might not behave in accordance with the first-order kinetics model suggested by Equation (1). Consequently, the objectives of the NAEG plutonium studies were stated in broader terms. The general objectives related to estimation of potential human ingestion and inhalation rates were simply (1) to determine plutonium concentrations in ecosystem components and (2) to quantify the rates of plutonium transfer among ecosystem components.

SOIL

Plutonium Concentration in Soil

Various soil surveys have been conducted to delineate highly contaminated areas at NTS, to determine the horizontal and vertical distribution of plutonium in contaminated soils, and for various other purposes (see several papers in Dunaway and White, 1974; White and Dunaway, 1975, 1976, 1977). An inventory of $^{239,240}Pu$ in the surface soils (0-5 cm depth) of NAEG study areas is given by Gilbert *et al.* (1975, p. 379) in terms of $\mu Ci/m^2$. As mentioned earlier, soil is the principal reservoir for plutonium at NTS and soil concentration (pCi/g) is the factor which drives or forces the transport system. In developing equations to estimate potential plutonium inhalation and ingestion rates for the hypothetical Standard Man, we shall relate the concentrations in air and foods to the average concentration in soil. Average soil concentrations were computed for each study area based on the inventory data provided by Gilbert *et al.* (1975, p. 379), and these are given in Table 1. In order to check the consistency of the algorithm in data used for conversion, strata averages were also computed and compared with data reported by Gilbert *et al.* (1975). These strata averages are also shown in Table 1. The algorithm for converting the inventory data to average soil concentrations was:

TABLE 1. Average Concentrations of $^{239-240}$Pu in Surface Soils (0-5 cm Depth) of NAEG Study Areas*

Study Area	Strata	Soil (pCi/g)	Study Area	Strata	Soil (pCi/g)
Area	1	34	Clean	1	257
13	2	103	Slate	2	1,286
	3	411	3	3	4,688
	4	952		4	8,196
	5	1,847		Avg.	485
	6	14,245	Area	2	724
	Avg.	213	11-B	3	5,323
				4	33,224
GMX	1	52		Avg.	7,462
5	2	725	Area	2	786
	3	4,555	11-C	3	2,016
	4	8,596		4	31,977
	5	79		5	138,568
	Avg.	175		Avg.	6,921
Double	1	123	Area	2	895
Track	2	6,422	11-D	3	4,385
	3	3,440		4	19,802
	4	50,459		5	53,018
	Avg.	305		Avg.	6,318
Clean	1	339			
Slate	2	1,405			
1	3	2,314			
	4	2,637			
	Avg.	511			
Clean	1	87			
Slate	2	1,559	Total Study Area -		11,379,100 m^2
2	3	5,721	>1000 pCi/g		393,700 m^2
	4	5,605	>2000 pCi/g		201,000 m^2
	Avg.	789			

* Based on Gilbert et al. (1975, p. 379), Table 2, and Equation (2).

$$C_{si} = 2 \times 10^{-5} I_i/(A_i \rho) \tag{2}$$

where, C_{si} is the average concentration of plutonium in the soil of stratum i (pCi/g),
I_i is the inventory (total amount) of plutonium in the soil of stratum i (pCi),
A_i is the area of stratum i (m^2),
2×10^{-5} is the reciprocal of the volume of soil in 1 m^2 x 5 cm deep (50,000 cm^3/m^2 = 10^4 cm^2/m^2 x 5 cm), and
ρ is the average bulk density of the soil in the study area of reference (g/cm^3).

Estimates of I_i and A_i are given by Gilbert *et al.* (1975, p. 379). Estimates of soil bulk densities are given in Table 2 for each study area. These data, not previously published, were also provided by Dr. Gilbert (personal communication). The strata averages in Table 1 are reasonably consistent with the data reported by Gilbert *et al.* (1975).

Losses From Soil Compartment

As suggested by Figure 1, plutonium may be transferred from the soil compartment to compartments representing other ecosystem components. It may also be removed from the soil of a given area by water or wind erosion. Percolation into the profile could remove plutonium from the surface where it is most susceptible to resuspension, and could, if the soil were plowed and rainfall plentiful, transport some plutonium below the root zones of crop plants.

Owing to the extreme variability of plutonium concentrations in soil samples taken from the same general area and to the arbitrary nature of soil compartment boundaries, it would be difficult to design field studies to estimate the overall rate of plutonium loss from the soil compartment. In fact, no such studies have been undertaken in the field or in the laboratory and we have no basis for assuming that the average concentrations of plutonium in the soils of contaminated areas will decrease significantly in the next 100 years or so. Consequently, the soil concentrations given in Table 1 will be treated as constants for the areas indicated, i.e., the soil compartment is assumed to be a continuous and constant source for plutonium transfer to other compartments. In the absence of any evidence that the rate of plutonium loss is, in fact, significantly greater than the rate of loss due to radioactive decay, the equation for the soil compartment is:

$$C_s = C_s(0)\exp(-\lambda_A t) \tag{3}$$

where, Cs is the average concentration of plutonium in the surface soil of a contaminated area at time t (pCi/g),
$C_s(0)$ is the initial concentration as given in Table 1,
λ_A is the radioactive decay rate of ^{239}Pu (7.7829 x 10^{-8} day^{-1}),
and t is time in days.

TABLE 2. Mean Bulk Density (g/cm^3) of Surface Soils (0-5 cm Depth) of NAEG Study Areas*

Study Area	Number of Samples	Bulk Density (g/cm^3)	
		Mean**	Standard Deviation
Area 13	169	1.14	0.017
GMX 5	115	1.18	0.013
Double Track	90	1.09	0.032
Clean Slate 1	89	0.918	0.016
Clean Slate 2	94	0.926	0.010
Clean Slate 3	100	0.908	0.011
Area 11-B	50	0.835	0.015
Area 11-C	48	0.866	0.033
Area 11-D	54	1.010	0.019

* R. O. Gilbert (personal communication).

** The relatively low bulk densities of these surfaces soils may be related to the vesicular structure of the soil layers immediately below desert pavement.

AIR

As indicated in Figure 1, plutonium contained in surface soils may be resuspended and transported to vegetation via external deposition or to herbivores and man via inhalation and some of it could be carried by wind and redeposited beyond the arbitrary boundary. In the absence of data to the contrary, we have assumed that deposition and resuspension processes in contaminated areas at NTS are in approximate steady state although data presented by Anspaugh and Phelps (1974, pp. 292-294) suggest that resuspension may exceed deposition at least to a small degree.

Deposition Velocity

The rate at which resuspended plutonium is deposited on soil could be estimated as the product of a deposition velocity (cm/day) and concentration in air (μCi/cm^3) to yield a rate that has dimensions of μCi/cm^2 day. Deposition velocities are functions of meteorological factors and the aerodynamic properties of plutonium-bearing soil particles and soil surfaces.

Deposition velocities measured under field conditions have been reported by Van der Hoven (1968); Sehmel, Sutter, and Dana (1973); and Healy (1974). Measurements under controlled conditions in a wind tunnel have been reported by Sehmel *et al.* (1973) and Sehmel (1973, 1975). These data indicate that the deposition velocity increases with increasing air velocity, increases with increasing particle size for sizes greater than about 1 μm, increases with decreasing particle size for sizes less than 0.01 μm, exhibits a minimum somewhere in the range of 0.01 to 1 μm, and is strongly influenced by the type of surface roughness. The wind tunnel data of Sehmel *et al.* (1973) for grass surfaces indicate the deposition velocity is approximately proportioned to both air velocity and particle size in the range of 2 to 13 m/sec and 1 to 100 μm. These grass data appear to correspond closely to field conditions provided a proper value is assigned to surface roughness.

Tamura (1976) has reported that more than 65 percent of the plutonium in soil samples from Area 13 is associated with soil particles in the range of 20 to 53 μm. Using the grass data of Sehmel *et al.* (1973) at 2.2 m/sec, the corresponding range of deposition velocities is from 3 to 20 cm/sec. Particles on the order of 20 to 50 μm could play an important role with respect to external contamination of vegetation, but particles this large are of little concern with respect to inhalation. As respirable particles are generally <10 μm, the corresponding deposition velocities suggested by the grass data would be <1 cm/sec.

Deposition Models

Both Healy (1974) and Sehmel (1975) present results of models used to predict deposition velocities. Healy's results indicate deposition velocity is proportioned to air velocity and is strongly dependent on atmospheric stability. Sehmel's results indicate deposition velocity increases as a nonlinear function of air velocity, exhibits a minimum value as a function of particle size, and is not strongly dependent on atmospheric stability. Both sets of results indicate a strong dependence on surface roughness. In order to apply either model to field conditions, it is necessary to estimate or measure the surface roughness and velocity profile.

For most applications to NTS, the grass data of Sehmel *et al.* (1973) appear to be the best analog. The trend of these data, in the range of 2 to 13 m/sec air velocity and 1 to 100 μm particle diameter, is approximately

$$V_d/U = 3\times10^{-4}\ d_p \qquad (4)$$

where

V_d is the deposition velocity (cm/sec),

U is the wind velocity (cm/sec),

d_p is the particle diameter, (μm).

For 10 μm particles, Equation (4) yields values similar to Healy's results for neutral atmospheric stability.

Resuspension Factor

The resuspension of plutonium from soil is often expressed as the ratio of air concentration ($\mu Ci/m^3$) to surface soil concentration ($\mu Ci/m^2$). Many such measurements have been made at NTS (Mork, 1970; Anspaugh and Phelps, 1974) and in the vicinity of Rocky Flats, Colorado (Volchok, 1971). The measured magnitudes of this ratio range generally from 10^{-5} to 10^{-11} m^{-1}. To estimate "acceptable soil concentrations," Anspaugh (1974) used a value of 10^{-9} m^{-1} for NTS. These ratios are, to say the least, extremely variable with respect to time and environmental factors such as wind speed and direction, rainfall, and disturbances affecting aerodynamic properties of soil surfaces. Other factors affecting this ratio are the aerodynamic properties of plutonium-bearing particles and their susceptibility to saltation and resuspension. There is evidence that the ratio tends to decrease with time after fallout-contamination of soil (Anspaugh, Phelps, Kennedy, and Booth, 1973; Anspaugh, 1974; Kathren, 1968). Anspaugh *et al.* (1975) have proposed a model in which the air/soil ratio decreases as a function of time from a maximum of 10^{-4} to a minimum of 10^{-9} m^{-1}, i.e.,

$$C_a/C_{ss} = 10^{-4}\ \exp(-\lambda t) + 10^{-9}$$

where C_a is the air concentration ($\mu Ci/m^3$),
C_{ss} is the soil surface concentration ($\mu Ci/m^2$),
and $\lambda = \ln(2)/0.15$.

This model is consistent with data collected over the years at NTS.

Resuspension Models

There have been many attempts to develop mathematical models to simulate resuspension (Amato, 1971; Mills and Olson, 1973; Killough and McKay, 1976). Most of these are based on models of wind erosion developed by Bagnold (1960) and, as a function of wind speed, take the form

$$C_a = K\ (U - U_T)^3\ C_{ss}/U \qquad (5)$$

where

U_T is a threshold wind speed, (m/sec)

and

K is a constant (sec/m^3).

Others (Sehmel and Orgill, 1973, and Shinn and Anspaugh, 1975) have used a power-law expression of the form

$$C_a = K\ U^n \qquad (6)$$

where

K and n are empirical constants derived from the data.

Sehmel and Orgill (1973) found $n = 2.1$ when they fit Volchok's (1971) data for plutonium resuspension at Rocky Flats to Equation (6). Shinn and Anspaugh also found $n = 2.1$ for dust flux at NTS. We approximately fit Sehmel's (1975) data for calcium molybdate resuspension at Hanford and also arrived at a value of about 2.1. The only contrary data are Shinn and Anspaugh's results for a plowed field in Texas which yielded $n = 6.4$.

The empirical value of about 2 for n when derived for different tracers, different soils, and different climates (provided the soil is undisturbed) tends to provide indirect confirmation for the theoretically derived form of Equation (5). However, both K and U_T in Equation (5) are functions of particle size, soil moisture content, surface roughness, relative humidity, and the time period over which the wind speed is averaged. Some attempts have been made to theoretically include many of these factors (especially particle size), but the theory does not seem to describe adequately the variations in the data. Thus, K and U_T must be treated as empirical constants for the present. Consequently, there is no practical benefit in using Equation (5) in preference for the simpler Equation (6). However, one must still have at least one experimental measurement of resuspension and wind speed in order to set the value of K in Equation (6) for the particular area.

Mass Loading

In the absence of data to implement Equation (6) for a given area, Anspaugh (1974) suggests that a mass loading factor (L_a) of 100 μg(soil)/m^3(air) be used for predictive purposes. If one assumes that the radioactivity of one square meter is associated with 50 kg of soil (5 cm depth x 10^4 cm^2/m^2 x $10^{-3}kg/cm^3$), a mass loading factor of 100 $\mu g/m^3$ is equivalent to a resuspension factor of 2 x 10^{-9} m^{-1}. The theoretical basis for the mass loading approach is described by Anspaugh (1974). Anspaugh *et al.* (1975) provide comparisons showing that predicted air concentrations based on L_a = 100 $\mu g/m^3$ are in good agreement with measured air concentrations.

We shall use the suggested mass loading factor to represent average conditions at NTS, but it must be noted that higher than average wind velocities (Shinn and Anspaugh, 1975) or mechanical disturbances such as plowing (Milham *et al.*, 1976) could cause the mass loading factor to be temporarily much higher than 100 $\mu g/m^3$.

VEGETATION

General Hypothesis

As shown in Figure 1, vegetation may be contaminated externally by deposition of resuspended material or internally by uptake from soil, or by both processes simultaneously. Other mechanisms of external and internal contamination have been identified or postulated, but direct deposition from air and root uptake appear to be the processes most important to consider in attempting to develop a general model.

Externally deposited material may be removed from plant surfaces by weathering, i.e., the mechanical action of wind and rain, and it may be diluted by plant growth. Internally deposited material may also be diluted by growth but not by weathering. Processes which remove biomass from vegetation (e.g., grazing, cropping, root decay, dehiscence of aboveground parts, etc.) also remove plutonium. If they exceed growth rates, these processes may reduce the total amount of plutonium in the vegetation compartment of an ecosystem. Different plant species may vary widely with respect to their ability to retain externally deposited plutonium or to assimilate plutonium from foliar deposits or soil, and translocation within the plant may result in large differences regarding plutonium concentrations in different plant organs. In the present discussion, we do not attempt to distinguish one plant species from another. We assume that plutonium is uniformly distributed in edible plant materials, and that processes which remove biomass from the vegetation compartment have no effect on the concentration of plutonium in the remaining biomass.

Differential equations expressing the principal processes described above can be written as follows:

$$dy_{ve}/dt = k_{av}C_a - (\lambda_w + \lambda_g + \lambda_A)y_{ve} \qquad (7)$$

$$dy_{vi}/dt = k_{sv}C_s - (\lambda_g + \lambda_A)y_{vi} \qquad (8)$$

where: y_{ve} is the concentration in vegetation of externally deposity Pu(pCi/g),
k_{av} is an air → vegetation deposition rate coefficient ($m^3/g\cdot day$),
C_a is the concentration of Pu in air (pCi/m^3),
λ_w is a weathering rate coefficient (day^{-1}),
λ_g is a vegetation growth rate coefficient (day^{-1}),
λ_A is the radioactive decay rate coefficient for ^{239}Pu (day^{-1}),
y_{vi} is the concentration in vegetation of internally deposited Pu (pCi/g),
k_{sv} is a soil → vegetation uptake rate coefficient (day^{-1}), and
C_s is the concentration of Pu in soil (pCi/g).

Equations (7) and (8) represent the external and internal components of plutonium in vegetation. The former is due to foliar deposition, the latter to root uptake. It is assumed that plutonium taken up via roots can be translocated to stems and leaves, but this rate is difficult to estimate. Consumers of vegetation are connected to both compartments simultaneously, and this is the same as summing the two components. Assimilation of externally deposited materials and their translocation to other parts of the plant has been demonstrated experimentally for various kinds of substances applied externally to foliage; but, in the case of plutonium (which is most probably deposited on foliage in the form of insoluble particles) foliar assimilation is assumed to be zero. A recent study (Cataldo, Klepper, and Craig, 1976) has demonstrated that translocation of foliarly deposited plutonium to roots and seeds can occur. However, the accumulation ratios observed in the absence of a solution vector (simulated rainfall) were on the order of 10^{-6} for both fresh and aged PuO_2, i.e., the observed concentrations in leaf tissue were about 200,000 to 500,000 times higher than the observed concentrations in seed and root tissue, respectively.

While foliar deposition and root uptake of plutonium have been studied separately in a variety of experiments, there is no reliable method for distinguishing between the two components in a plant sample if both are present. If plants are grown in contaminated soil or culture media and only the aerial parts are assayed, one may assume that the activity detected was internally deposited. If the aerial parts of plants are collected from a recently contaminated area which had no plutonium in the soil prior to the contaminating event, one may assume that all or nearly all the activity detected was externally deposited. If, on the other hand, plant samples are taken from an area which was contaminated years ago, it is likely that most of the plutonium contained therein is due to external contamination by resuspended soil particles and that only a small fraction is due to internal contamination by root uptake.

Recent evidence (Romney *et al.*, 1975; Wildung and Garland, 1974) suggests the possibility that (a) the biological availability of plutonium in contaminated soil may increase with time after environmental release so that successive crops of annuals would take up successively greater amounts of plutonium, (b) perennial plants may accumulate plutonium to a much greater extent than previously indicated by short-term uptake experiments, and (c) plant uptake may increase with decreasing plutonium concentration in the soil. Either or all of these factors could have the effect of making biological transport of plutonium to man progressively more important relative to physical transport and inhalation. These considerations are interesting because most assessments of the potential hazards of environmental plutonium attributed little importance to biological transport and ingestion compared to physical transport and inhalation.

Foliar Deposition

Estimation of deposition velocity, V_d, i.e., the ratio of surface deposition rate to air concentration, was discussed earlier as applied to soil surfaces.

Deposition velocities have also been determined experimentally for different kinds of plants, and several kinds of vegetation, with respect to various aerosols and particulates. In the present study, however, it was more convenient to base estimates of air to plant deposition rates on the product of air concentration, soil/air deposition velocity, and a plant (or vegetation) interception factor. The plant interception factor is defined as the amount initially deposited per gram (dry weight) of plant material divided by the amount initially deposited per unit area of soil surface.

Much of the available information concerning the interception of airborne radionuclides by plants has come from studies made in the fallout fields produced by nuclear test detonations at NTS. Romney *et al.* (1963) summarized early studies in the vicinity of the Nevada Test Site. They found that levels of fallout deposition on plants varied with respect to: (a) downwind distance from the detonation point, (b) lateral distance away from the midline of the fallout field, (c) a variety of morphological features associated with different plant species, and (d) the level of fallout deposition on soil surfaces near the contaminated plants. While there was no constant relationship between fallout concentrations on plants (dpm/g) and on soils (dpm/m^2), there was a good correlation between radioactivity in plant samples and in the fraction of fallout samples with particle sizes less than 44 μm. Apparently, the larger particles were deposited close to the detonation point and were not as readily intercepted by plants as were the smaller particles which were deposited farther downwind; or, if they were intercepted, they were apparently more easily removed by weathering processes. In the laboratory, Romney *et al.* (1963) found that 50 to 90 percent of the gross radioactivity on fallout-contaminated plants could be removed by washing with water or a wetting agent such as versene. These and similar studies demonstrated the possibility of predicting fallout interception by plants, but they did not provide quantitative methods for doing so.

Miller and Lee (1966) carried out extensive studies of fallout interception by plants. The plants were cultivated in gardens near San Jose, Costa Rica, and the fallout was provided by continuing eruptions of Irazú, a nearby volcano. Miller and Lee also developed a comprehensive theoretical model of fallout interception by plants. The model assumes different sets of constants for different fallout particle size classes, different morphological characteristics of foliage, and different meteorological conditions. Unfortunately, their model is practically unworkable, in spite of its elegance, because it requires the use of constants and other parameter values which are rarely if ever available for predictive purposes.

The values of the interception factor determined experimentally by Miller and Lee varied only slightly with respect to the different species of cultivated plants they studied, and the only meteorological condition consistently correlated with large differences in measured values of the interception factor was relative humidity. The particles intercepted by plants were essentially the same sizes as those deposited on adjacent soil surfaces. In both cases, the mass median diameters for the volcanic dust deposited as fallout were generally between 50 and 100 μm. The weighted averages of interception factors for all the plant types tested (mostly garden vegetables) were: 95.7 ± 66.9 cm^2/g for damp exposure conditions (relative humidity greater than 90 percent) and 47.4 ± 29.7 cm^2/g for dry exposure conditions.

Interception factors based on nuclear testing experience are about one or two orders of magnitude lower. For detonations involving the incorporation of large quantities of soil material in the initial cloud, estimates range from 1.9 to 11.1 and have a mean of 3.7 cm^2/g. For detonations involving the incorporation of little or no soil material in the initial cloud, the estimated values are about an order of magnitude lower.

Miller and Lee noted that the foliar samples obtained at the weapons test experiments were apparently subjected to an unknown degree of weathering before they were taken, while the primary samples in [our studies] were collected at the end of a 12- to 24-hour period of exposure to more or less continuous fallout from Irazú, and the weight of dust deposited on leaves was often greater than the dry weight of the leaves. Heavy dust deposits such as these are easily dislodged by the slightest mechanical disturbance, and moderate rains were observed to remove more than 90 percent of the material deposited.

Martin (1965) studied the interception and retention of ^{89}Sr and ^{131}I by desert shrubs (primarily *Atriplex confertifolia* and *Artemisia tridentata*). His estimates of the plant interception factor were based on concentrations of ^{89}Sr and ^{131}I in plant samples collected 5 days after fallout deposition and estimates of the theoretical deposition rates for these two radionuclides on unobstructed soil surfaces in the same locations ranging from about 10 to about 100 miles downwind from the detonation point. Estimates for different study areas range from 1.49 to 11.05 cm^2/g, with the higher values occurring in the more distant areas. The overall mean for ^{89}Sr was 4.09 cm^2/g, and the overall mean for ^{131}I was 4.00 cm^2/g: approximately an order of magnitude lower than Miller and Lee's average value for dry deposition conditions. Although the discrepancy may appear to be a large one, it is easily accounted for by the effects of weathering during the 5 days that elapsed between the time of initial fallout deposition and Martin's first collection of plant samples.

Weathering Rate

To estimate the effective rates of ^{89}Sr and ^{131}I loss from fallout-contaminated plants, Martin (1964) collected additional sets of plant samples at intervals of 10, 15, 30, and 60 days after fallout. Factoring out the loss of ^{131}I due to vaporization and the losses of both radionuclides due to radioactive decay, the data indicated that the weathering (environmental) half-time for these two radionuclides increased with respect to time after fallout. During the interval from 5 to 15 days after fallout, the average weathering half-time was about 20 days. The value obtained for the interval from 15 to 30 days after fallout was about 30 days; and it increased, for the interval from 30 to 60 days postdetonation, to about 130 days. The D+5-day concentrations can be compared with assumed D+0 concentrations which would reconcile the difference between Martin's estimate of the plant interception factor and Miller and Lee's estimate of the foliage contamination factor. This procedure suggests an average weathering half-time of approximately 1.4 days during the interval from 0 to 5 days after fallout.

These observations lead us to the hypothesis that the decay-corrected concentration of a radionuclide in fallout-contaminated plant material is a very rapidly declining exponential function of time at times soon after the contaminating event but approaches a lower asymptote. As this hypothesis appears to be correct, the effective rate at which a radionuclide is removed from surfaces following external deposition cannot be expressed precisely by a single coefficient because the weathering half-time increases as a function of time after contamination. If the initial deposition is a heavy one, a significant fraction of it (perhaps as much as 90 percent) may be removed by weathering in a matter of hours, or a few days at most. A portion of what remains after this initial period of fast weathering (something in the range of 10 to 60 percent) is so tightly trapped that it cannot be removed even by vigorous washing (Romney *et al.*, 1963). Presumably, this nonremovable fraction is composed predominately of particles which are small and mechanically trapped on plant surfaces.

Plant Growth Rates

As indicated earlier, the growth of new plant tissue may dilute both the external and the internal concentrations of plutonium or other transuranium elements in plant materials. As different plant parts may grow at different rates, it is obvious that the growth rate of interest with respect to external contamination is the growth rate of leaves (and other edible parts formed above ground). If we assume that internal plutonium due to root uptake is uniformly distributed to all parts of the plant, the growth rate of interest with respect to dilution of root uptake is the overall growth rate, i.e., the growth rate of leaves plus the growth rate of all other plant parts.

Plant growth is not a continuous process, nor is it the same for all species in a given area or for all the parts of a given plant. In the temperate zone, at least, plant growth is confined to the warm season, and the rate of growth is not uniform throughout the growing season because different plant organs develop at different times. Ignoring the morphogenic aspects of plant growth (i.e., the differentiation and development of structure), growth is most simply conceived as an increase in biomass (i.e., dry weight of tissue per unit area). For annuals, the biomass at the beginning of the growing season consists of seeds; for herbaceous perennials in which the aboveground parts die back during the winter, it consists mostly of roots and other belowground parts; for woody perennials, it consists of roots and stems (mostly dead tissue) plus, in the case of evergreens, leaves, and vestiges of fruits produced the previous growing seasons. In addition to these seasonal and species variations, it is reasonable to suppose that the roots, stems, leaves, fruits, and other organs of a given species grow at different times and at different rates and that additional variations can be expected in response to environmental factors such as temperature, soil moisture, and availability of nutrients.

To attempt a mathematical description of vegetation growth which includes all factors mentioned above (and others not mentioned above) would be a monumental undertaking. What is needed at present is a simple expression of growth rate, as a continuous function, which will provide a reasonable but conservative estimate of the potential, overall concentration of plutonium in plant materials which have been contaminated externally by airborne deposits and/or internally by root uptake from soil.

To obtain a rough estimate of growth rate, we can define λ_g as follows:

$$\lambda_g = \ln (1 + P_n/B_o)/365 \qquad (9)$$

where: λ_g is the growth rate coefficient averaged over the year (day^{-1}),
P_n is the net gain in biomass during a growing season (g/m^2),
and B_o is the biomass at the beginning of the growing season (g/m^2).

Odum (1971) has estimated that the average gross primary productivity (GPP) of deserts and tundras is about 200 kcal/m^2 per year. As the fraction of GPP (0.2) used up in respiration does not appear as new tissue, the dilution growth rate is proportional to 0.8 GPP = 160 kcal/m^2 per year. At 4.5 kcal/g dry weight (Odum, 1971), this amounts to a net gain of $P_n = 36$ g/m^2, approximately. The biomass of desert vegetation varies from place to place. The mean biomass for Area 13 is $B_o = 289$ g/m^2 (Wallace and Romney, 1972). Substituting these values of P_n and B_o in Equation (9), $\lambda_g = 3 \times 10^{-4}$ day^{-1}. Assuming internally deposited Pu to be uniformly distributed above- and belowground, this would be the value to use in Equation (8). Assuming two-thirds of P_n to be above ground and one-third below ground, the dilution growth rate for the external (aboveground) component, Equation (7), would be $\lambda_g = 2 \times 10^{-4}$ day^{-1}.

Root Uptake and Plant/Soil Concentration Factor

In order for plutonium to enter plants via root uptake, it must first reach the roots. Plowing, of course, accomplishes this "transport" quite rapidly by mixing the soil, but the downward movement of plutonium in an undisturbed soil profile is such a slow process that much of the plutonium deposited on the surface may stay near the surface for many years. To circumvent the variability inherent in these and other soil processes affecting the behavior of plutonium in soils (factors reviewed by Price, 1973a; Francis, 1973), we have made the simplifying and conservative assumption that plutonium deposited on soil is diluted by only the top 5 cm of soil and that root uptake is related to the resulting concentration in surface soil, i.e., the probable concentration of plutonium in the root-zone soil is deliberately overestimated.

Most of the available data (Price, 1973a; Francis, 1973) on plutonium uptake by plants has been derived from short-term greenhouse experiments. Typical values thus derived for the plant/soil concentration factor range from 10^{-3} to 10^{-6}. Uptake has been shown to be enhanced by reduction of pH or addition of chelating agents. There is some evidence that plutonium uptake by plants may increase with time (Romney *et al.*, 1970) and that the mobility of plutonium, i.e., its ability to move in the soil and its availability for root uptake, may increase with time as a result of chelation due to bacterial decay of soil organic matter.

The evidence that the concentration factor may increase exponentially as a function of time is scanty but deserves consideration because of its implications. Romney *et al.* (1970) grew ladino clover (*Trifolium repens L.*) on a plutonium-contaminated soil under greenhouse conditions and cropped it repeatedly over a period of 5 years. The resulting estimates of C_{FV} = dpm/g (plants, dry weight) per dpm/g (soil, dry weight) were as follows:

Year	C_{FV}
1	1.91×10^{-5}
2	4.14×10^{-5}
3	4.38×10^{-5}
4	7.10×10^{-5}
5	13.95×10^{-5}.

A least squares fit of these data to an exponential function yields the following expression:

$$C_{FV} = 1.31 \times 10^{-5} \exp (0.452\ t),$$

where t is time in years and 0.452 is the apparent growth rate coefficient, i.e., the data indicate that C_{FV} would be expected to double in about 1.5 years. By extrapolation to 20 years, the concentration factor would be 0.11, a value within the range of field observations for perennials (see below) but misleading nonetheless. Romney *et al.* (1970) attribute the "apparent" increase of C_{FV} to root growth during the time the experiment was being conducted and not to any change in the biological availability of the plutonium contained in the contaminated soil.

In fact, the plant/soil ratios observed under field conditions are generally too high to be explained by root uptake. Romney *et al.* (1975) collected soil and plant samples from contaminated areas at NTS. The mean plant/soil ratios for different groups of paired samples ranged from 0.004 to 0.44 and were inversely related to the mean soil contamination of each group which ranged from 5.9×10^{-5} to 0.12 µCi/g. The weighted mean ratio for 506 paired samples was 0.096 ± 0.004. The plants in this study were desert shrubs growing in areas which were contaminated with plutonium as a result of nuclear safety tests conducted from 1953 to 1964. Ratios obtained by growing plants in these same soils were on the order of 10^{-3} to 10^{-4} which indicates that no more than 1 percent of the Pu in plant samples from contaminated areas at NTS is likely to be due to root uptake.

Environmental monitoring data from Hanford (Bramson and Corley, 1973; Nees and Corley, 1974; Fix, 1975) indicate ratios in the range of 0.05 to 1.0 for soil concentrations in the range of 2×10^{-9} to 4×10^{-8} µCi/g. Similar data from Savannah River (McLendon *et al.*, 1976) indicate ratios from 0.009 to 0.97 for

soil concentrations in the range of 1.3×10^{-9} to 1.6×10^{-7} µCi/g of ^{239}Pu and 2×10^{-10} to 4.6×10^{-8} µCi/g of ^{238}Pu. The higher plant/soil ratios are usually found at lower soil concentrations.

In general, plant/soil ratios for ^{239}Pu which are based on plant and soil samples collected under field conditions range from 10^{0} to 10^{-3}, while ratios based on laboratory studies which preclude external contamination range from 10^{-3} to 10^{-6}. Considering the situation at NTS, we believe it is reasonable to assume that approximately 99 percent of the plutonium associated with the vegetation compartment is due to external contamination and that no more than 1 percent is due to root uptake.

Data on the plant/soil ratio for other transuranium elements are very limited. Romney *et al.* (1975) measures ^{241}Am concentrations of the vegetation from the areas at and near the Nevada Test Site. Grouped according to species and location, the mean plutonium/americium ratio in vegetation ranged from 2.0 to 28.3 with typical values being about 10. Similar groups of soil (Gilbert *et al.*, 1975) ranged from 5.3 to 26 with typical values also being about 10. These analyses indicate that the long-term plant/soil ratio for americium is not significantly different from plutonium. The data on the short-term plant/soil ratio indicate significant differences which may be related to the solubility of the element. Price (1973b) measured the uptake of ^{237}Np, ^{239}Pu, ^{241}Am, and ^{244}Cm by tumbleweed and cheatgrass from various solutions applied to the soil. The americium uptake was about 2 to 30 times greater than plutonium, curium was about 2 to 40 times greater, and neptunium was about 100 to 1,000 times greater. Bennett (1976) summarized much of the short-term data and concluded that americium and curium uptakes were about 10 to 30 times greater than plutonium.

Variation of Plant/Soil Ratio

Data presented by Romney *et al.* (1975) demonstrate that the mean concentrations of plutonium in soils and plants decreases with increasing distance from ground zero locations, while the vegetation/soil ratios within sampling strata show a tendency to increase. Tamura (1976) provides a graph of soil activity versus distance from ground zero and fits the data to a power curve of the form $y = ax^{-b}$ where y is soil activity and x is distance from ground zero. We have no comparable curve for vegetation, but we assume it would be of the same form. Based on this assumption, the vegetation/soil ratio could be expressed as follows:

$$y_v = a_v x^{-b_v},\ C_s = a_s x^{-b_s}, \qquad (10)$$

$$y_v/C_s = (a_v/a_s)x^{(b_s-b_v)}$$

If $b_s > b_v$, the vegetation/soil ratio will increase with increasing distance from ground zero and decreasing soil concentration.

To obtain an expression for plutonium concentration in vegetation as a function of soil concentration, a regression analysis was performed (Gilbert, personal communication) to fit the available data (636 paired samples of vegetation and soil) to the following equation:

$$\ln(y_v) = (\ln a) + b \ln(C_s),$$

or

$$V = A + bS$$

where

$$V = (\ln y_v),\ S = \ln(C_s),\ A = (\ln a)$$

$$b = \sqrt{\frac{\sum V^2 - \frac{1}{n}(\sum V)^2}{\sum S^2 - \frac{1}{n}(\sum S)^2}}$$

$$A = \bar{V} - b\bar{S},$$

where $\bar{V}$ is the mean of V and $\bar{S}$ is the mean of S.

This method of calculating b is indicated because measurements of both y_v and C_s are subject to error (see Ricker, 1973).

The results of this analysis are summarized as follows:

Parameter	Standard Deviation
$\bar{V} = -3.4961$	2.3096
$\bar{S} = -0.9322$	3.0382
$b = 0.7602$	0.0221
$A = -2.7871$	0.0458
$r = 0.8084$ (correlation coefficient)	
$n = 636$	

Taking, antilogs, the mean vegetation/soil ratio is:

$$C_{FV} = y_v/C_s = 0.0303/0.3937 = 0.0770$$
$$(\text{nCi/g})/(\text{nCi/g}),$$

which is somewhat lower than the value (0.096) obtained from the grouped data presented by Romney *et al.* (1975).

The equations for vegetation concentration (nCi/g) as a function of soil concentration (nCi/g) is:

$$y_v = 0.0620\, C_s^{0.76} \tag{11}$$

The corresponding equation for the vegetation/soil ratio is:

$$C_{FV} = y_v/C_s = 0.062\ C_s^{-0.24} \tag{12}$$

Thus, for areas in which soil concentrations are 10, 1.0, and 0.1 nCi/g, the predicted vegetation/soil ratios would be 0.036, 0.062, and 0.107, respectively.

Equations (11) and (12) demonstrate the dependence of vegetation concentration on soil concentration and the fact that the vegetation/soil ratio tends to increase as soil concentration increases. The use of either equation for predictive purposes may be limited by the extreme range of soil and vegetation values and/or by various site specific factors which are not considered in the regression analysis. It might be better to apply the regression analysis to sampling strata means as shown in Table 3.

Except for Strata 3 and 4 in Area 13, the measured and predicted values given in Table 3 seem to agree quite well, but the equations for Area 13 and GMX-5 (footnote to Table 3) predict higher values (especially at higher soil concentrations) than would be obtained from Equations (11) and (12), which are based on samples from all study areas.

Discussion

Equations (11) and (12) shed some light on how vegetation/soil ratios may be expected to vary with respect to soil concentration, but they do not explain why. To approach this and related questions, we refer back to Equations (7) and (8), the proposed differential equations for the external and internal components of plant contamination. For the time being, at least, we can dismiss Equation (8) from further consideration because the greenhouse studies have shown that root uptake cannot account for more than a small fraction of the vegetation/soil ratios observed at NTS.

Equation (7) for external contamination has the following solution:

$$y_{ve} = \frac{k_{av}C_a}{\lambda_w + \lambda_g + \lambda_A}\left[1-\exp(-(\lambda_w + \lambda_g + \lambda_A)t)\right] \tag{13}$$

where the parameters are defined following Equation (7), and

$$k_{av} = V_d F_v \tag{14}$$

$$C_a = L_a C_s \tag{15}$$

where, V_d is the deposition velocity on soil (cm/day),
F_v is a vegetation interception factor (cm^2/g (vegetation)),
and L_a is a mass loading factor (g(soil)/cm^3(air)).

Table 3. Mean Concentrations (nCi/g) of $^{239-240}Pu$ in Soil and Vegetation and Vegetation/Soil Ratios by Strata in Area GMX-5 and Area 13

Strata	Soil (Means)[a]	Vegetation (Means)		Vegetation/Soil	
		Measured[a]	Predicted[b]	Measured[a]	Predicted[b]
Area GMX-5					
5	0.084	0.0083	0.0099	0.13	0.12
1	0.059	0.0092	0.0075	0.16	0.13
2	0.73	0.064	0.055	0.075	0.075
3	4.5	0.26	0.23	0.052	0.051
4	7.3	0.31	0.34	0.050	0.046
Area 13					
1	0.036	0.0052	0.0067	0.15	0.19
2	0.1	0.013	0.017	0.14	0.17
3	0.4	0.17	0.058	0.44	0.14
4	1.1	0.077	0.14	0.069	0.13
5	2.4	0.28	0.28	0.10	0.12
6	14.0	1.2	1.36	0.078	0.10

(a) From Romney *et al.* (1975).

(b) $y_v = 0.13\ C_s^{0.79}$ for Area GMX-5

$y_v = 0.07\ C_s^{0.89}$ for Area 13

Both equations are based on the strata means in Columns 2 and 3.

Assuming a steady-state between vegetation and soil, the vegetation/soil ratio can be expressed in the parameters of Equations (13), (14), and (15) as follows:

$$y_{ve}/C_s = V_d F_v L_a / \lambda_v \qquad (16)$$

where, $\lambda_v = \lambda_w + \lambda_g + \lambda_A$ is the effective decay rate coefficient for plutonium bearing soil particles externally deposited on vegetation.

As noted in the section on air, the deposition velocity is a function of particle size (Equation (4)). In the soils of Area 13 (Tamura, 1976), most of the plutonium is associated with coarse silt (20 to 53 μm) and the estimated deposition velocity (V_d) for particles of 50 μm diameter could be as high as 20 cm/sec or 1.73×10^6 cm/day.

The plant interception factor (F_v) determined by Miller and Lee (1966) for freshly deposited volcanic dust (50 to 100 μm) was 47.4 cm^2/g for dry exposure conditions.

For predictive purposes, Anspaugh (1974) has suggested that a mass loading factor (L_a) of 100 $\mu g/m^3$ (10^{-10} g/cm^3) be used. This is the amount of dust one would expect to find in the GMX area (Shinn and Anspaugh, 1975) when the wind velocity averages about 1.4 m/sec. (3 mph).

Substituting these values in Equation (16), assuming a vegetation/soil ratio of 0.1 and solving for λ_v indicates effective half-life of about 8.5 days. So,

$$\frac{y_{ve}}{C_s} = \frac{(1.73 \times 10^6\ \text{cm/day})\ (47.4\ \text{cm}^2/\text{g})\ (10^{-10}\ \text{g/cm}^3)}{\ln(2)/8.5}$$

$$= 0.10.$$

This exercise proves nothing. It merely demonstrates that Equation (16) might be adequate to explain the high vegetation/soil ratios observed at NTS. Shinn and Anspaugh (1975) have demonstrated that mass loading (L_a) increases with wind velocity. The effective half-life may decrease with wind velocity. Sehmel (1975) has shown that deposition velocity (V_d) decreases as particle size decreases for d_p >1μm. If small particles are more readily retained by vegetation than larger particles, the plant interception factor and the effective half-life may increase as particle size decreases. Variations of the factors of Equation (16) as functions of particle size and wind velocity may account for the range of vegetation/soil ratios implied by Equation (11), but there are still too many unknowns to develop a descriptive, dynamic model for the vegetation compartment.

For predictive purposes, we shall assume that the soil and vegetation compartments are in steady state and that the mean vegetation/soil ratio is 0.1. This ratio is somewhat conservative as it tends to overestimate the plutonium content of vegetation in areas where soil concentrations are > 100 pCi/g.

CATTLE

Transport Pathways

For present purposes, the only herbivore assumed to contribute to man's plutonium intake is the cow. Both dairy cattle and beef cattle are considered. The principal plutonium inputs to these herbivores (Figure 1) are by inhalation and by ingestion of contaminated soil and vegetation. Figure 2 illustrates the assumed pathways of plutonium transport to man via beef cattle and dairy cattle and provides estimates of some of the parameters required to estimate potential concentrations of plutonium in the muscle, liver, and milk of beef and dairy cattle maintained in a contaminated area at NTS.

Formulation of Cow Model

A general equation for the concentration of plutonium in the muscle, liver, or milk compartment of Figure 2 can be derived as follows:

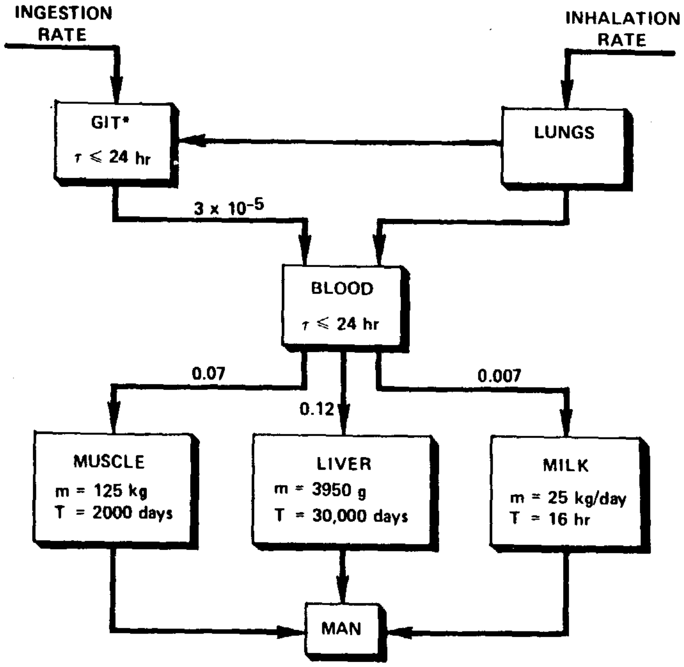
$$dy_i/dt = \frac{1}{m_i}(r_b f_{bi} - \lambda_i y_i) \tag{17}$$

$$y_i(t) = \frac{r_b f_{bi}}{m_i}\left(\frac{1 - e^{-\lambda_i t}}{\lambda_i}\right) \tag{18}$$

i = muscle, liver, milk.

where, y_i is the concentration of plutonium in compartment i at time t (pCi/g),
m_i is the weight of compartment i (g),
r_b is the rate at which plutonium enters blood (pCi/day),
f_{bi} is the fraction (dimension
to compartment i (Figure 2: 0.07, 0.12, and 0.007),
and λ_i is the effective elimination rate coefficient (day^{-1}),
for plutonium in compartment i (based on effective half lives,
T, in Figure 2),

Estimated values for some of the parameters of Equations (17) and (18) are given, as indicated above, in Figure 2. The transfer fractions from the gastrointestinal tract to blood and from blood to muscle, liver, and milk; the



*Gastrointestinal Tract.

FIGURE 2. PATHWAYS OF PLUTONIUM TRANSPORT TO MAN VIA BEEF CATTLE AND DAIRY CATTLE

weight of muscle and liver; and the effective half-life in milk are based on experimental results reported by Stanley *et al.* (1974). The other values were assumed (Martin and Bloom, 1976) for purposes of estimation. Equation (18) ignores the retention of plutonium in the lungs of cattle and assumes that all inhaled plutonium is transferred to the gastrointestinal tract or the blood within 24 hours.

If one assumes that the parameter values given in Figure 2 are reasonably close to the true values, the only parameters remaining to be determined are the plutonium ingestion and inhalation rates and the rate at which plutonium enters the blood (r_b).

Plutonium Ingestion Rates

Kleiber (1961) shows that the basal metabolic rate of mammals (heat production by a fasting animal) is proportional to the three-fourths power of body weight and that the feeding capacity (maximum energy intake) of domestic animals such as the cow is about 5 times the food intake required for basal metabolism. Data given in the Merk Veterinary Manual (Siegmund, 1967) for the digestible energy (DE) requirements for maintenance of mature cows are based on:

$$DE = 163.5\ W^{0.73} \qquad (19)$$

where, DE is digestible energy required for maintenance (kcal/day),
and W is the body weight (kg).

The additional DE requirement for milk production in the range from 20 to 35 kg (milk)/day at 5 percent butterfat is 1,850 kcal DE per kg milk. The additional DE requirement for growth ranges from about 8,600 to about 19,800 kcal/kg gained, depending also on body weight. According to McKell (1975), the average digestibility of desert vegetation is about 36 percent compared to 52 percent for good alfalfa hay and up to 80 percent for some concentrates (Siegmund, 1967). The average energy content of most plant materials is about 4.5 kcal/g dry weight (Golley, 1961), and the digestible energy content of desert vegetation is about 1.6 kcal/g dry weight. Based on these considerations, the vegetation ingestion rate for a cow grazing desert vegetation can be estimated as follows:

$$V_{ing} = \frac{163.5\ W^{0.73}\ \text{kcal/day}}{0.36 \times 4.5\ \text{kcal/g}} \qquad (20)$$

where, V_{ing} is the vegetation ingestion rate (g/day) for maintenance of a mature cow grazing desert vegetation. For cows which are gaining weight, producing milk, or pregnant, the energy requirement and thus the vegetation ingestion rate would be higher than estimated by Equation (20).

Grazing cattle also ingest soil. In earlier papers (Martin *et al.*, 1974; Martin and Bloom, 1976), we assumed that the soil ingestion rate might be as high as 2,000 g/day. Data recently reported by Smith (1977) indicate that this value is probably too high. The amounts of sediment (soil) recovered from the reticulum and rumen of three cows which had been grazing in Area 13 prior to sacrifice were 8.5, 57.3, and 278 g, respectively. As Smith points out: "These data suggest that the total amount of soil ingested is much less than 2 kg per day, and that a reasonable estimate would be between 0.25 and 0.5 kg."

Smith *et al.* (1976) estimate that a 409 kg cow which grazed for 177 days in the inner compound of Area 13 ingested a total of 100 µCi of ^{239,240}Pu or 0.565 µCi/day. This estimate was based on plutonium concentrations in the rumen contents of fistulated steers allowed to graze in the same compound. Gilbert *et al.* (1976) made an independent estimate of 0.620 µCi/day based plutonium concentrations in *Eurotia lanata* (34 percent of cow's diet) and *Atriplex canescens* (64 percent of cow's diet) as reported for Area 13 by Romney *et al.* (1975). The average wet weight of vegetation ingested by the fistulated steers was 30 kg/day and the average dry/wet ratio was about 0.2 (Gilbert, personal communication). In other words, it is estimated that the cow's vegetation ingestion rate was about 6 kg/day. Based on Equation (20), a 409 kg cow would have to ingest about 8 kg/day to meet its energy requirements for maintenance. Neither Smith *et al.* (1976) nor Gilbert *et al.* (1976) include soil ingestion in their estimates of the plutonium ingestion rate for the Area 13 cow.

Based on the methods outlined above, we would estimate this cow's plutonium ingestion rate as follows:

$$I_v = 8139\ \text{g(veg)/day} \times 0.1 \times 5.5 \times 10^{-4}\ \mu\text{Ci/g(soil)}$$

$$= 0.448\ \mu\text{Ci/day}$$

$$I_s = 250\ \text{g(soil)/day} \times 5.5 \times 10^{-4}\ \mu\text{Ci/g(soil)}$$

$$= 0.138\ \mu\text{Ci/day}$$

$$I_v + I_s = 0.585\ \mu\text{Ci/day.}$$

where I_v and I_s are the plutonium ingestion rates through vegetation and soil, respectively. In this calculation, 8,139 g/day is the vegetation ingestion rate, based on Equation (20) for a 409-kg cow; 0.1 is the assumed average vegetation/soil ratio; 5.5×10^{-4} µCi/g is the average soil concentration of the Area 13 inner compound (Gilbert and Eberhardt, 1976); and 250 g/day is the assumed soil ingestion rate. The resulting estimate of the cow's total plutonium ingestion rate, 0.585 µCi/day, is comparable to the independent, site-specific estimates of Smith *et al.*, 1976 (0.565 µCi/day), and of Gilbert *et al.*, 1976 (0.620 µCi/day). Considering the probable variability of the measurements and parameters involved, the agreement of results is remarkably good.

The principal difference between our method and the method of Gilbert *et al.* (1976) are: (a) our estimate of the vegetation ingestion rate based on energy requirements (8 kg/day) is higher than their estimate based on the rumen contents of a fistulated steer (6 kg/day), (b) our estimate of the average concentration of plutonium in vegetation is lower than theirs, 55 pCi/g versus 103 pCi/g, and (c) we included an input for soil ingestion.

If we assume that the digestibility of Area 13 vegetation is 49 percent instead of 36 percent, a ration of 6 kg/day would be adequate to meet the cow's maintenance energy requirements. The weighted mean vegetation/soil ratio for Area 13 is about 0.15 or 50 percent higher than average. If we use this ratio instead of 0.1 and 6 kg/day instead of 8 kg/day, $I_v = 0.495$ μCi/day and $I_s = 0.565 - 0.495 = 0.07$ μCi/day, or 127 g (soil)/day.

Inhalation Rate

Standard Man's respiration rate is $20 m^3$/day (ICRP, 1959) and his digestible energy requirement for maintenance metabolism (no weight gain or loss) is 2,600 kcal/day (NRC, 1968). We assume that for man and cattle, respiration rates are proportional to digestible energy maintenance requirements. The DE requirement (Equation (19)) for a 409-kg cow is 13,185 kcal/day. The cow's estimated respiration rate is therefore $13,185 \times 20/2,600 = 101\ m^3$/day. The cow's plutonium inhalation rate (I_a) can be estimated by

$$I_a = \frac{20\ DE}{2600} \times L_a \times C_s \tag{21}$$

where, I_a is the plutonium inhalation rate (pCi/day),
DE is the digestible energy required (Equation 19) for maintenance metabolism (kcal/day),
$20 m^3$/day divided by 2600 kcal/day = $7.69 \times 10^{-3}\ m^3$/kcal for Standard Man,
L_a is a mass loding factor (100 μg(soil)/m^3(air)) as recommended by Anspaugh (1974),
C_s is the average plutonium concentration (pCi/g) in the soil of the area grazed by the cow.

For the 409-kg cow of the Area 13 inner enclosure, where $C_s = 550$ pCi/g, $I_a = 5.56$ pCi/day.

Ingestion Versus Inhalation

It is obvious from the preceding discussion that $I_v >> I_a$, but the accumulation of plutonium in organs or tissues other than the gastrointestinal tract or lungs and the excretion of plutonium in milk depends (Figure 2 and Equation (17)) on the rate (r_b) at which plutonium reaches blood. The rate at which ingested plutonium reaches the blood is simply the plutonium ingestion rate (I_v) multiplied by the fraction transferred from the gastrointestinal tract to the blood. For the Area 13 cow discussed above,

$$r_{bv} = I_v f_{gb} = 565,000 \text{ pCi/day} \times (3 \times 10^{-5})$$
$$= 16.95 \text{ pCi/day}.$$

where r_{bv} is the rate at which ingested Pu reaches the blood (pCi/day)
and f_{gb} is the fraction transferred from the gastrointestinal tract to blood.

As this cow was left in the inner compound of Area 13 for 177 days before sacrifice (Smith, 1977), the total ^{239}Pu expected to have entered the blood via ingestion would be 3,000 pCi.

The transfer of inhaled plutonium to blood is considerably more complicated than indicated by Figure 2. Assuming resuspended particles to have an AMAD of 0.5 μm, and applying the ICRP-19 lung model (Figure 4 and Table 11) to the cow, we obtain the following expression for the rate at which inhaled plutonium would be expected to enter the blood:

$$r_{ba} = 0.0021\ I_a + 0.0833\ \lambda_1\ y_1 + \lambda_2 y_2 + 3 \times 10^{-5}\ y_3 \tag{22}$$

$$dy_1/dt = 0.18\ I_a - \lambda_1 y_1 \tag{23}$$

$$dy_2/dt = 0.225\ \lambda_1 y_1 - \lambda_2 y_2 \tag{24}$$

$$dy_3/dt = 0.75\ [I_a(0.2079 + 0.12/\lambda_o) + 0.667\ \lambda_1 y_1] \tag{25}$$

where, r_{ba} is the rate (pCi/day) at which inhaled plutonium enters the blood,
0.0021 is the fraction of inhaled plutonium transferred directly to blood from the upper respiratory tract,
0.0833 is the fraction of plutonium deposited in the lungs and then transferred to blood,
y_1 is the amount (pCi) present in the lung at time t,
λ_1 = ln(2)/500 days is the lung clearance rate,
y_2 is the amount (pCi) present in the lymph at time t,
λ_2 = ln(2)/1000 days is the lymph clearance rate,
y_3 is the amount (pCi) present in the gastrointestinal tract at time t,
0.18 is the fraction deposited in the lungs and cleared with a 500-day half-life,
0.225 is the fraction transferred from lung to lymph and then cleared to blood with a 1000-day half-life,
0.75 day is the average residence time of plutonium in the gastrointestinal tract,
0.2079 is the fraction transferred directly from the upper respiratory tract to the gastrointestinal tract,

0.12 is the fraction cleared from lungs to the gastrointestinal tract with a 1-day half-life,

$\lambda_o = \ln(2)/1$ day),

and 0.667 is the fraction cleared from lungs to the gastrointestinal tract with a 500-day half-life.

Note that Equation (22) consists of four terms. These terms represent transfers of inhaled plutonium to blood from the upper respiratory tract, the lungs, the pulmonary lymph, and the gastrointestinal tract, respectively. Equations (23), (24), and (25) are the differential equations for the long-term components of the lungs and lymph nodes and for the gastrointestinal tract, respectively. The solution to Equation (22) is:

$$\begin{aligned} r_{ba}(t) = I_a \Big\{ & 0.0021 + 0.015(1 - e^{-\lambda_1 t}) \\ & + 5.61 \times 10^{-5} (e^{-\lambda_2 t} - e^{-\lambda_1 t}) \\ & + 3 \times 10^{-5} [0.381 + 0.12(1 - e^{-\lambda_1 t})] \Big\} \end{aligned} \tag{26}$$

Integration of Equation (26) and evaluation of the integral from t = 0 to t = 177 days gives the amount reaching the blood in 177 days as 0.676 I_a. Applying this result to the Area 13 cow, the total ^{239}Pu expected to have entered the blood via inhalation would be 0.676 x 5.56 pCi/day = 3.76 pCi. Compared to 3,000 pCi for ingestion (see above), this is a negligible quantity.

Based on this comparison, we shall assume that plutonium concentrations in cow milk, muscle, and liver are due to ingestion only, or conversely, that the contribution from inhalation is negligible. Even after 10 years of exposure in Area 13, the contribution from inhalation would be no more than 0.56 percent of the total.

Comparison of Model Predictions and Field Data

Plutonium concentrations in lungs, liver, and muscle are given in Table 4 for four cows included in the Area 13 grazing study. Cow 2 (Smith *et al.*, 1976) was placed in the inner compound for 177 days. Cows 1, 4, and 6 grazed the outer compound for 433 days (Smith, 1977). Cow 1 weighed 252 kg.

Cow 4 weighed 300 kg, the weight of cow 6 is not known. We shall assume that the average weight of the outer compound cows was 275 kg. Tissue weights for these cattle were not reported; but we estimate, for a 275-kg cow, the average lung weight is about 2.1 kg and the average liver weight is 4.8 kg, based on a study made by Smith and Baldwin (1974). At 45 percent of body weight (Smith, 1977), the muscle weight for a 275-kg cow would be about 125 kg.

Table 4. Concentrations of ^{239,240}Pu (pCi/kg) in Tissues of Cattle Grazed in Area 13 (NTS)

Tissue	Outer Compound[(a)]				Inner Compound[(b)]	
	1	4	6	Avg.	2	M
Lungs	74.5	51.4	18.2	48.0	NR[(c)]	NR
Liver	14.5	15.8	10.9	13.7	38.9	6.13 kg
Muscle	0.05	0.195	lost	0.12	0.17	189 kg

Data extracted from:
(a) Smith *et al.*, 1976
(b) Smith, 1977
(c) Not reported

Lungs

Based on these and other considerations, the concentration of ^{239}Pu in the lungs of the cows from the outer compound of Area 13 can be estimated as follows:

$$C_{LUNG} = \frac{(76\ m^3/day)(10^{-4}\ g/m^3)(215\ pCi/g)(0.18)}{2.1\ kg}\left(\frac{1-e^{-433\lambda}}{\lambda}\right)$$

$$= 55\ pCi/kg\ (vs.\ 48\ pCi/kg,\ Table\ 4),$$

where, 76 m^3/day is the estimated respiration rate for a 275-kg cow (Equation 21),
10^{-4} g(soil)/m^3(air) is the estimated mass loading factor (Equation 21),
215 pCi/g is the average concentration of ^{239}Pu in the soil of Area 13*,
0.18 is the assumed fraction of inhaled plutonium deposited in the lungs and cleared with a 500-day half-life,
2.1 kg is the estimated weight of the lungs,
and λ = ln(2)/500 days.

Liver and Muscle

The plutonium ingestion rate for a 275-kg cow can be estimated as follows:

$$I_v = 6158g(veg)/day^{**} \times 0.1 \times 70\ pCi/g(soil)$$
$$+ 250g(soil)/day \times 70\ pCi/g(soil)$$
$$= 60{,}606\ p\ Ci/day.$$

The concentration in liver for cows in the inner compound can now be estimated as follows:

$$C_{LIV} = \frac{(60{,}606\ g/day)(3 \times 10^{-5})(0.12)}{4.8\ kg}\left(\frac{1-e^{-433\lambda}}{\lambda}\right)$$

$$= 19.6\ pCi/kg\ (vs.\ 13.7\ pCi/kg,\ Table\ 4),$$

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*The average soil concentration in the outer compound is 70 pCi/g, but some of the resuspended material in the air of the outer compound is assumed to come from the soil of the inner compound.

**Based on Equation (20) for a 275-kg cow.

where, 3×10^{-5} is the fraction transferred from the gastrointestinal tract to blood (Figure 2),
0.12 is the fraction transferred from the blood to the liver (Figure 2),
4.8 kg is the estimated weight of the liver,
and λ = ln(2)/30,000 days (Figure 2).

Similar calculations were made for the other cases given in Table 4. The observed values from Table 4 and the estimated values are compared below in pCi/kg wet weight.

	Outer Compound		Inner Compound	
	Observed	Estimated	Observed	Estimated
Lungs	48.0	55.0	NR	61.0
Liver	13.7	19.6	38.9	60.7
Muscle	0.12	0.4	0.17	1.12

These comparisons (above) suggest that the model for beef cattle may be somewhat conservative, but the order-of-magnitude agreement between observed and estimated values appears to be good, better than might be expected as a matter of fact. However, partial data for four cows, two areas, and two grazing times is hardly an adequate basis for model validation. The best we can conclude from these comparisons is that they provide no basis for rejecting the model. The discrepancy between the experimental values and values predicted by the model are less than an order of magnitude, and we have little reason to expect better than an order of magnitude accuracy.

The Milk Cow

The model milk cow is assumed to weigh 650 kg and to produce milk at a rate of 25 kg/day. Such an animal would require a digestible energy intake of 64,750 kcal/day, i.e., 18,500 kcal/day for maintenance plus 25 kg/day x 1,850 kcal/kg for milk production (Siegmund, 1967). To meet this high-energy requirement and, at the same time, provide a conservatively high estimate of plutonium transport to man via milk, we shall assume that the model milk cow consumes 10 kg/day of desert vegetation and 15 kg/day of alfalfa hay grown in the same contaminated area. The remainder of the diet consists of commercial concentrates containing no plutonium. For the model milk cow, the plutonium ingestion rate is estimated as follows:

$$I_v = C_s\ (250g(soil)/day + 0.1 \times 10{,}000g(veg)/day$$
$$+ 0.017 \times 15{,}000g(alfalfa)/day)$$
$$= 1505\ C_s\ (pCi/day),$$

where C_s is the soil concentration (pCi/g) and 0.017 is the alfalfa/soil ratio which is assumed to be one-sixth the desert vegetation/soil ratio due to plowing and mixing of the soil to a depth of 30 cm. The equation for estimating the concentration in milk is

$$C_{MILK} = \frac{(1505)(3 \times 10^{-5})(0.007)}{25\ \lambda_{MILK}}\ C_s \qquad (27)$$

$$= 1.37 \times 10^{-5}\ C_s\ \text{(pCi/kg)}.$$

where $\lambda_{MILK} = \ln(2)/0.75$ (Figure 2).

MAN

In the preceding discussion, we have considered the dynamics of the plutonium transport system (Figure 1) and attempted to establish mathematical relationships between compartments. Our present knowledge of the food chain kinetics of plutonium in contaminated areas at NTS is not adequate for modeling the dynamic aspects of all parts of the transport system. To simplify estimation of the plutonium inhalation and ingestion rates for herbivores (cattle), we assumed the system to be in steady-state and the intake rates to be constant. We now apply the same simplifying assumptions to estimate potential plutonium inhalation and ingestion rates for the hypothetical Standard Man.

Inhalation Rate

The Pu-inhalation rate (A_m) is defined as the product of the respiration rate (B_m) and the concentration of Pu in air. The concentration of Pu in air is, of course, quite variable; but, since it is due to resuspension of contaminated soil, it can be related to the average concentration in surface soil (C_s).

For predictive purposes, Anspaugh (1974) has suggested the use of a mean mass loading factor of 100 μg (soil)/m³ (air). We combine this factor with the further assumption that the specific activity of Pu in resuspended materials is the same as that in the associated soil, and estimate A_m as follows:

$$A_m = B_m L_a C_s = 0.002\ C_s \qquad (28)$$

where, A_m is the Pu-inhalation rate for man (pCi/day), B_m is the respiration rate (20 m³/day), L_a is the mass loading factor (100 μg/m³), and C_s is the average concentration of Pu (pCi/g) in the soil of the contaminated area.

The observed mass loading factor during cascade impactor runs at NTS was 70 μg/m³ (Anspaugh, 1974) and the specific activity of particles recovered from the impactors was about one-third as high as that of surface soil samples from the same locations (Phelps and Anspaugh, 1974). Compared with these observations, the estimate of A_m provided by Equation (28) may be conservatively high by a factor of about four, under average conditions. High winds or mechanical disturbances, such as vehicular traffic, plowing, etc., could cause the mass loading factor to increase temporarily to very high levels. However, a comparison of observed and predicted air concentrations based on L_a = 100 μg/m³ showed very good agreement (Anspaugh *et al.*, 1975).

Ingestion Rate

The Pu-ingestion rate is defined as the sum of products of the rates at which different kinds of contaminated materials are ingested and the concentration of Pu in each kind of material. The formula employed for estimating a probable ingestion rate for use in this study was

$$H_m = C_s \sum_{i=1}^{n=6} I_i D_i \qquad (29)$$

where H_m is the Pu-ingestion rate for man (pCi/day), I_i is the ingestion rate for substance i (g/day), and D_i is the discrimination ratio (dimensionless) for substance i.

The kinds of materials considered, their assumed ingestion rates (I_i), and associated discrimination factors (D_i) are listed, together with their products and sum, in Table 5. The methods, experimental data, and assumptions used to estimate the discrimination factors (D_i) are explained below.

Soil

The assumption that the Standard Man of the model accidentally ingests soil at an average rate of 0.01 g/day is purely speculative but not unreasonable considering the amount of dust that can be raised in desert environments by activities that disturb the soil surface.

Vegetation

To estimate the plutonium concentration in native vegetation, we assume an average vegetation/soil ratio of 0.1. As explained earlier, this ratio should tend to overestimate the concentration of plutonium growing in areas of relatively high soil concentration at NTS. To distinguish between native vegetation and cultivated plants (alfalfa hay), we assumed a sixfold dilution of soil concentration due to plowing to a depth of 30 cm, i.e., the plant/soil ratio for cultivated plants is 0.017 instead of 0.1. In preparing Table 1, we assumed that 90 percent of the external contamination of "leafy vegetables" and that 99 percent of the contamination associated with "other food plants" would be removed by washing, peeling, etc., during preparation for consumption. In spite of this assumed reduction, leafy vegetables and other food plants account for 90 percent of Standard Man's estimated plutonium ingestion rate (Table 5).

Table 5. Estimation of Standard Man's Plutonium Ingestion Rate

i	Substance Suggested	I_i (a)	D_i (a)	I_iD_i	Percent
1	Soil	0.01 (b)	1.0	1.0×10^{-2}	5.151
2	Leafy vegetables	81 (c)	1.7×10^{-3}	1.4×10^{-1}	70.935
3	Other food plants	222 (c)	1.7×10^{-4}	3.8×10^{-2}	19.441
4	Beef muscle	273 (c)	9.4×10^{-6}	2.6×10^{-3}	1.322
5	Beef liver	13 (d)	4.7×10^{-4}	6.1×10^{-3}	3.148
6	Cow milk	436 (c)	1.4×10^{-8}	6.1×10^{-6}	0.003
			$\Sigma\ I_iD_i = 1.9 \times 10^{-1}$		100.000

(a) See Equation (29) and explanation in text.
(b) Assumed accidental ingestion rate.
(c) U.S.D.A., Agricultural Statistics, 1973. U.S. GPO (1973).
(d) Food Consumption Statistics 1960-1968. Organization for Economic Cooperation and Development (1970).

Muscle, Liver, and Milk

The model beef cow weighs about 275 kg. Its plutonium ingestion rate (I_v), due to ingestion of 6.2 kg (native vegetation)/day and 0.25 kg (soil)/day, is about 870 C_s pCi/day. Given this ingestion rate and the parameters noted in Figure 2, the discrimination ratios for muscle and liver (Table 5) were estimated as follows:

$$\frac{C_{MUS}}{C_{SOIL}} = \frac{(870)(3 \times 10^{-5})(0.07)}{125{,}000} \left[\frac{1-\exp(-730(\ln(2)/2000))}{\ln(2)/2000}\right]$$

$$= 9.4 \times 10^{-6}$$

$$\frac{C_{LIV}}{C_{SOIL}} = \frac{(870)(3 \times 10^{-5})(0.12)}{4800} \left[\frac{1-\exp(-730(\ln(2)/30{,}000))}{\ln(2)/30{,}000}\right]$$

$$= 4.7 \times 10^{-4}.$$

In these examples, t was set equal to 730 days (2 years), and this is the assumed average age of beef cattle at the time of slaughter.

The method of estimating the discrimination factor for milk was described earlier in the description of the model milk cow (see Equation (27)).

Discussion

Models for calculating organ burdens and cumulative organ doses due to ingestion and inhalation of ^{239}Pu are discussed later in this report in considerable detail. Based on the ICRP-2 model for ingestion and the ICRP-19 model for inhalation, radiation doses to the respiratory system would be entirely due to inhalation and doses to the gastrointestinal tract would be primarily due to ingestion. Dose to organs receiving the radionuclide from blood (bone, liver, kidney, etc.) would be due to both ingestion and inhalation. The relative importance of inhalation versus ingestion can be compared by comparing the two components of organ burden after a period of chronic exposure. Such a comparison is provided in Table 6.

Based on our estimates for the hypothetical Standard Man at NTS (inhalation = 0.002 C_s pCi/day and ingestion = 0.2 C_s pCi/day), the ingestion/inhalation ratio would be 100 and ingestion would contribute about 5 percent of the 50-year bone burden. As indicated by Table 6, the relative importance of ingestion versus inhalation increases as the ingestion/inhalation ratio increases. Any factor tending to increase the transfer from the gastrointestinal tract to blood would have the same effect as an increase in the ingestion/inhalation ratio.

Table 6. Fractions of ^{239}Pu in Bone, Liver, or Kidney Due to Chronic Ingestion and Inhalation for a Period of 50 Years(a)

Ingestion/Inhalation	Fraction Due to Ingestion	Fraction Due to Inhalation
1	0.0005	0.9995
10	0.0053	0.9947
100	0.0506	0.9494
200	0.0964	0.9036
400	0.1758	0.8242
1,000	0.3478	0.6522

(a) Estimated burdens based on ICRP Publications 2 and 19.

DOSE ESTIMATION MODELS

Plutonium reaches man by ingestion of contaminated food and water or inhalation of contaminated air. Part of this plutonium is distributed throughout the body where it may remain for some time. While it remains within the body, those organs that retain the plutonium will receive a radiation dose which depends on the weight of the organ, the amount of plutonium retained, and the time that the plutonium is retained. There are several models which we have used to estimate plutonium distribution in man and these are discussed in this section.

Dose and Dose Commitment

In all of the dose estimation models, the formula for estimating the radiation dose to a critical organ of man, i.e., one of those organs which tend to receive the highest radiation doses, is

$$dD/dt = E\ y/m \tag{30}$$

where

t is time, days
D is the dose to the reference organ (rems),
E = 51.2159 ε, is a dose rate factor (g rem/(μCi day)),
ε is the effective energy absorbed in the reference organ per disintegration of radionuclide (MeV/dis),
y is the plutonium burden in the organ (μCi),
m is either the mass of the organ if the organ is not part of the gastrointestinal tract or twice the mass of the contents if the organ is part of the gastrointestinal tract (g).

The values of the parameter in Equation (30) for ^{239}Pu and other transuranium elements are given in Table 7. Most of these values were reported by the ICRP (1959, 1964). The masses of deep lung and other portions of the respiratory tract are the values used by Snyder (1967) and Kotrappa (1968, 1969). The mass of thoracic lymph nodes was assumed to be the value (15g) reported by Pochin (1966). The mass of abdominal lymph nodes was assumed to be less than the thoracic lymph nodes and was arbitrarily set at 10g. The dose accumulated in the organ from the beginning of the exposure period (t=0) to some later time ($t=T_D$) is given by

$$D = (E/m) \int_0^{T_D} y\ dt. \tag{31}$$

If ingestion and inhalation of plutonium were halted at time T_D, and the individual were to live to some later time T_L, each organ would accumulate an

Table 7. Parameters for Calculating Radiation Doses From Transuranium Radionuclides

Organ(a) Mass, g		GIT 150(b)	Lung(c,d) 500	Bone 7000	Liver 1700	Kidney 300	TB 70000
Radionuclide	τ_A(e)	Effective Energy, MeV/dis (f)					
^{237}Np	8×10^{8}	0.62	49	250	49	49	49
^{239}Np	2.33	0.14	0.16	0.63	0.16	0.15	0.22
^{238}Pu	33000	0.55	57	284	57	57	57
^{239}Pu	8.9×10^{6}	0.52	53	266	53	53	53
^{240}Pu	2.4×10^{6}	0.52	53	266	53	53	53
^{241}Pu	4800	0.010	0.013	0.048	0.013	0.012	0.014
^{242}Pu	1.4×10^{8}	0.49	51	253	51	51	51
^{243}Pu	0.208	0.18	0.18	0.88	0.18	0.18	0.19
^{244}Pu	2.8×10^{10}	1.14	59	292	59	59	59
^{241}Am	1.7×10^{5}	0.56	57	283	57	57	57
^{242}Am	5.6×10^{4}	0.745	53.2	266	53.2	53.1	53.2
^{242}Am	0.667	0.734	53.1	266	53.1	53.1	53.1
^{243}Am	2.9×10^{6}	0.79	54.2	273	54.2	54.2	54.2
^{244}Am	0.0181	0.52	0.52	2.6	0.52	0.52	0.52
^{242}Cm	162.5	0.61	63	315	63	63	63
^{243}Cm	13000	0.61	60	299	60	60	60
^{244}Cm	6700	0.59	60	299	60	60	60
^{245}Cm	7.3×10^{6}	0.55	55	277	55	55	55
^{246}Cm	2.4×10^{6}	0.54	56	278	56	56	56
^{247}Cm	3.3×10^{10}	0.54	56	278	56	56	56
^{248}Cm	1.7×10^{8}	11.5	453	2244	453	453	453
^{249}Cm	0.044		0.31	27			5.2
^{249}Bk	290	0.026	0.026	0.13			0.026
^{250}Bk	0.134	0.41	0.52	1.5			0.83
^{249}Cf	1.7×10^{5}	0.63	60	301			60
^{250}Cf	3700	0.61	62	311			62
^{251}Cf	2.9×10^{5}	0.59	59	295			59
^{252}Cf	803	2.1	210	1100			210
^{253}Cf	18.0	0.78	68	343			68
^{254}Cf	56.0	120	3800	18900			3800

(a) GIT is gastrointestinal tract (principally, lower large intestine).
(b) Mass of contents of lower large intestine.
(c) ICRP-II used 1000g for mass of lungs. All other models use 500g.
(d) Effective energy to lymph nodes and all portions of the respiratory tract are assumed equal to lung. Masses for these organs are: nasopharyngeal region, 1.35g; tracheobronchial region, 400g; and abdominal lymph nodes, 10g.
(e) τ_A is radioactive half-life of radionuclide, days.
(f) Includes energy from daughter products with half-times less than 1 year.

additional dose from the plutonium already within the body at time T_D. The dose commitment is the sum of the dose accumulated to T_D plus the additional dose or

$$D_c = D + D_a \tag{32}$$

where

D_a is the additional dose (rems)
D_c is the dose commitment (rems).

ICRP Committee II Model

The report of the ICRP Committee II (1959) contained a model and data which was used to estimate maximum permissible concentrations (MPC) of radionuclides in air and water. The model, as applied to plutonium or other transuranics, is indicated in Figure 3. This model distinguishes between insoluble and readily soluble compounds that are inhaled. In the absence of other data, it is assumed that 25 percent of a soluble compound is exhaled, 50 percent is swallowed and reaches the gastrointestinal tract (GIT) almost immediately, and 25 percent is immediately transferred directly to the blood.

None of the soluble compound resides in the lung for any significant length of time and, therefore, does not contribute a radiation dose to the lung. For insoluble compounds, in the absence of other data, it is assumed that 25 percent is exhaled, 50 percent is swallowed and reaches the GIT immediately, and 25 percent is immediately transferred directly to tissues deep in the lung. Of the amount transferred to the deep lung, half (12.5 percent of that inhaled) is coughed up within 24 hours and is swallowed (reaches the GIT). The remainder (12.5 percent of that inhaled) resides in the deep lung with a half-time of 365 days and is then transferred to the blood. In order to avoid underestimating radiation doses, we use the insoluble parameters for calculating the transfer to the lung and GIT but use the soluble parameters for the transfer to all other organs.

On the basis of Figure 3 and the above discussion, the equations for the ICRP II model are

$$r_{GIT} = f_{URG} A_m + f_{URDLF} A_m + H_m \tag{33}$$

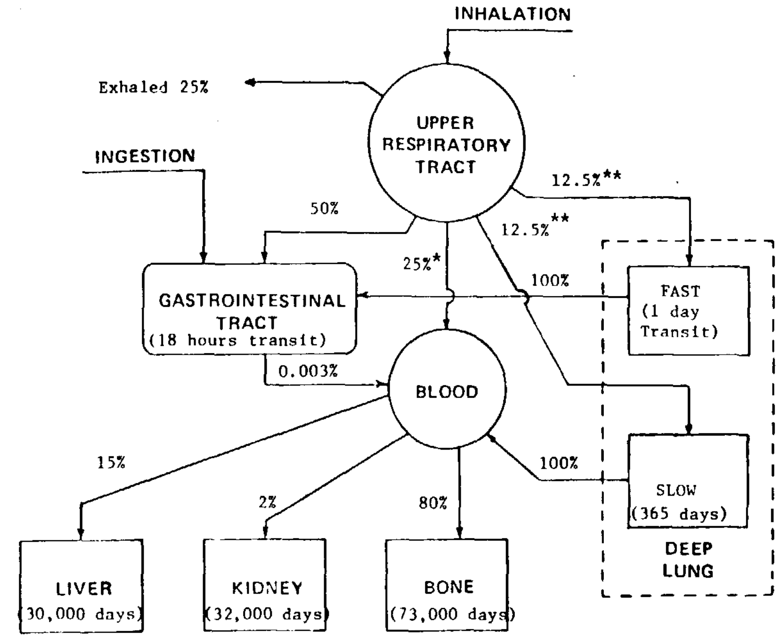
$$r_B = f_{URB} A_m + f_{GITB} r_{GIT} \tag{34}$$

$$y_{GIT} = r_{GIT} T_{GIT} \tag{35}$$

$$dy_{DLS}/dt = f_{URDLS} A_m - (\lambda_A + \lambda_{DLS}) y_{DLS} \tag{36}$$

$$y_{DL} = y_{DLS} + f_{URDLF} A_m T_{DLF} \tag{37}$$

$$dy_{LIVER}/dt = f_{BL} r_B - (\lambda_A + \lambda_L) y_{LIVER} \tag{38}$$



* Soluble Compounds
** Insoluble Compounds

Fraction From Bloom to Total Body is 100%
Biological Half-Time in Total Body is 65,000 days.

FIGURE 3. ICRP COMMITTEE II MODEL FOR PLUTONIUM

$$dy_K/dt = f_{BK}r_B - (\lambda_A + \lambda_K)y_K \tag{39}$$

$$dy_{BONE}/dt = f_{BBN}r_B - (\lambda_A + \lambda_B)y_{BONE} \tag{40}$$

$$dy_{TB}/dt = f_{BTB}r_B - (\lambda_A + \lambda_{TB})y_{TB} \tag{41}$$

where

r_B, r_{GIT} are the rates that plutonium reaches the blood and gastrointestinal tract (GIT), respectively (μCi/day),

A_m,H_m are the plutonium inhalation and ingestion rates, respectively (μCi/day),

f is the fraction of plutonium transferred from one location to another within the body with the subscript notation as follows: URG is upper respiratory tract (URT) to GIT; URB is URT to blood; GITB is GIT to blood; URDLF and URDLS are URT to the fast and slow portions, respectively, of the deep lung; BL, BK, BBN, and BTB are blood to liver, kidney, bone and total body, respectively. T_{DLF}, T_{GIT} are transit times for the fast portion of the deep lung and the GIT (lower large intestine), respectively.

λ is the biological elimination rate constant with the subscript notation as follows: DLS is deep lung (slow portion), TB is total body, L is liver, K is kidney, and B is bone (day^{-1}),

$\lambda = \ln(2)/\tau$ where τ is the biological half-time for the organ, days.

y is the plutonium burden in the organ with the subscript notation either self evident or identical to that for λ except DLF is the fast portion of the deep lung and DL is the total deep lung (μCi).

Values of the parameters in Equations (33) through (41) can be obtained from Figure 3 for plutonium. The values for other transuranic elements, if different from plutonium, are given in Table 8. Radioactive half-times for each transuranic nuclide are given in Table 7.

Task Group on Lung Dynamics Model

The ICRP Task Group on Lung Dynamics (Morrow *et al.*, 1966) provided a more detailed description of the inhalation pathway and arrived at the model indicated in Figure 4. A subsequent report (ICRP 19, 1972) provided specific data for applying this model to plutonium and other transuranics. The Task Group model treats the respiratory tract as a series of compartments in which the amount initially deposited depends on the particle size and the clearance rate (biological half-time) depends on the type of compound.

Table 8. Parameters Values Applicable to the ICRP II Model for Transuranics Other Than Plutonium[(a)]

Element	Neptunium	Americuim	Curium	Berkelium	Californium
		<u>Biological Half-Time, Days</u>			
Deep Lung (slow portion)	120	120	120	120	120
Liver	54000	3480	3000		
Kidney	64000	27000	24000		
Total Body	39000	20000	24000	65000	65000
		<u>Fraction From Blood to Organ</u>			
Liver	0.05	0.35	0.40		
Kidney	0.03	0.03	0.02		
Bone	0.45	0.25	0.30	0.80	0.80

(a) All other parameter values are identical to plutonium.

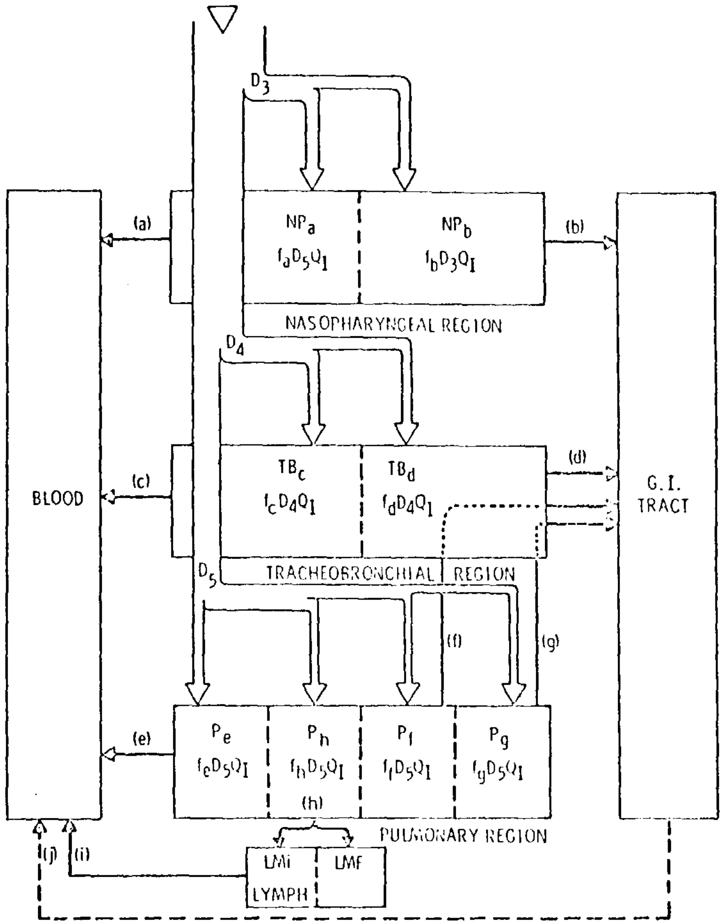


FIGURE 4. Schematic Diagram of the Task Group Lung Model From Houston *et al.*, 1975

On the basis of Figure 4, the equations for the compartments of the respiratory tract and lymph nodes plus the transfer to the gastrointestinal tract and blood are

$$r_{GIT} = \lambda_b y_{NPb} + \lambda_d y_{TBd} + \lambda_f y_{pf} + \lambda_g y_{Pg} + H_m \qquad (42)$$

$$r_B = \lambda_a y_{NPa} + \lambda_c y_{TBc} + \lambda_e y_{TBe} + \lambda_i y_{LMi} + f_j r_{GIT} \qquad (43)$$

$$dy_{NPa}/dt = f_a D_3 A_m - (\lambda_A + \lambda_a) y_{NPa} \qquad (44)$$

$$dy_{NPb}/dt = f_b D_3 A_m - (\lambda_A + \lambda_b) y_{NPb} \qquad (45)$$

$$y_{NP} = y_{NPa} + y_{NPb} \qquad (46)$$

$$dy_{TBc}/dt = f_c D_4 A_m - (\lambda_A + \lambda_c) y_{TBc} \qquad (47)$$

$$dy_{TBd}/dt = f_d D_4 A_m - (\lambda_A + \lambda_d) y_{TBd} \qquad (48)$$

$$y_{TBfg} = (\lambda_f y_{Pf} + \lambda_g y_{Pg}) T_{TBfg} \qquad (49)$$

$$y_{TB} = y_{TBc} + y_{TBd} + y_{TBfg} \qquad (50)$$

$$dy_{Pe}/dt = f_e D_5 A_m - (\lambda_A + \lambda_e) y_{Pe} \qquad (51)$$

$$dy_{Pf}/dt = f_f D_5 A_m - (\lambda_A + \lambda_f) y_{Pf} \qquad (52)$$

$$dy_{Pg}/dt = f_g D_5 A_m - (\lambda_A + \lambda_g) y_{Pg} \qquad (53)$$

$$dy_{Ph}/dt = f_h D_5 A_m - (\lambda_A + \lambda_h) y_{Ph} \qquad (54)$$

$$y_P = y_{Pe} + y_{Pf} + y_{Pg} + y_{Ph} \qquad (55)$$

$$dy_{LMi}/dt = f_i \lambda_h y_{Ph} - (\lambda_A + \lambda_i) y_{LMi} \qquad (56)$$

$$dv_{LMF}/dt = (1 - f_i) \lambda_h y_{Ph} - \lambda_A y_{LMF} \qquad (57)$$

$$y_{LM} = y_{LMi} + y_{LMF} \qquad (58)$$

where

r, λ, τ, y, H_m, f, and A_m	are defined previously in the ICRP II model and the subscripts, where different, refer to the compartments and the pathways listed in Figure 4 and Table 9.
T_{TBFg}	is the residence time of material following pathways f and g in the tracheobronchial region (days),
D_3, D_4, D_5,	are the fractions of inhaled material that are deposited in the nasopharyngeal, tracheobronichial, and pulmonary regions, respectively, of the respiratory tract (see Figure 4 and Table 9).

Values of most of the parameters in the above equations are given in Table 9 (USNRC, 1976). T_{TBfg} is assumed to one hour or 1/24-day (Snyder, 1967; Kotrappa, 1968, 1969), while f_j is identical to f_{GITB} (0.003 percent) in the ICRP II model. The equations for GIT, liver, kidney, bone, and total body are identical to the ICRP II model with identical parameter values except for the following values which apply to all transuranic elements: $f_{BL} = 0.45$, $f_{BBN} = 0.45$, $\tau_L = 40$ years, and $\tau_B = 100$ years.

Stuart, Dionne, and Bair (SDB) Model

Stuart *et al.* (1968) developed models to describe the distribution and retention of plutonium in the body following a single inhalation. These models were based on the results of several studies with dogs and these results were extrapolated to where they might apply to man. The short-term model is shown in Figure 5, while the long-term form is shown in Figure 6. Stuart *et al.* (1971) revised the long-term model and these revisions are incorporated into Figure 6. Stuart *et al.* (1971) combine the nasopharyngeal and tracheobronchial regions of the Task Group model into one compartment but expand the pulmonary region into two compartments: one with a constant biological half-time of 3 years and another with a variable half-time.

They also add compartments for abdominal lymph nodes and treat the transfers from the pulmonary region to lymph nodes in a slightly different manner than the Task Group model. In addition, they include feedback pathways for liver to the gastrointestinal tract (GIT) and for liver, kidneys, and skeleton to blood. Since they only applied this model to inhalation, they did not consider the transfer from GIT to blood since the fraction transferred is so small (less than 0.01 percent).

On the basis of Figures 5 and 6, the equations for the compartments of the respiratory tract and lymph nodes plus the transfer to GIT and blood are

Table 9. Task Group Lung Model Parameter Values

FRACTION OF INHALED PARTICLES DEPOSITED IN THE RESPIRATORY SYSTEM VERSUS PARTICLE DIAMETER(a)

Particle Size AMAD Micrometers	Fraction of Inhaled Quantity Retained		
	Nasopharyngeal Region, D_3	Tracheobronchial Region, D_4	Pulmonary Region, D_5
0.05	0.001	0.08	0.59
0.1	0.008	0.08	0.50
0.3	0.063	0.08	0.36
0.5	0.13	0.08	0.31
1.0	0.29	0.08	0.23
2.0	0.50	0.08	0.17
5.0	0.77	0.08	0.11

CLEARANCE PARAMETER VALUES(b)

Compartment	Translocation Class						
		D		W		Y	
	k	T_k^b	f_k	T_k^b	f_k	T_k^b	f_k
NP	a	0.01	0.50	0.01	0.10	0.01	0.01
	b	0.01	0.50	0.40	0.90	0.40	0.99
TB	c	0.01	0.95	0.01	0.50	0.01	0.01
	d	0.20	0.05	0.20	0.50	0.20	0.99
P	e	0.50	0.80	50	0.15	500	0.05
	f	n.a.	n.a.	1	0.40	1	0.40
	g	n.a.	n.a.	50	0.40	500	0.40
	h	0.50	0.20	50	0.05	500	0.15
L	i	0.50	1.00	50	1.00	1000	0.90

(a) Estimated from data of Task Group on Lung Dynamics (Morrow 1966).

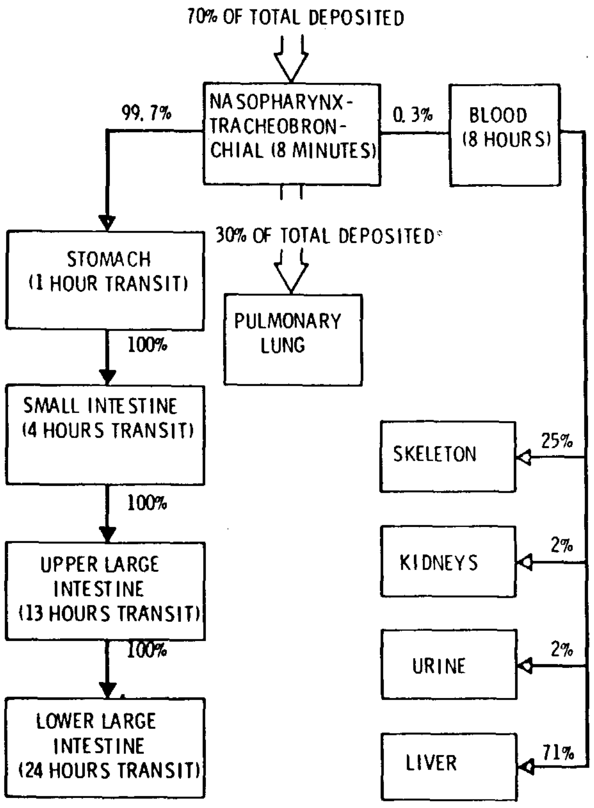
(b) As amended by ICRP Publication 19 (1972)

D.W.Y. = Days, Weeks, Years

l.c. = Clearance pathway (See Figure 4)

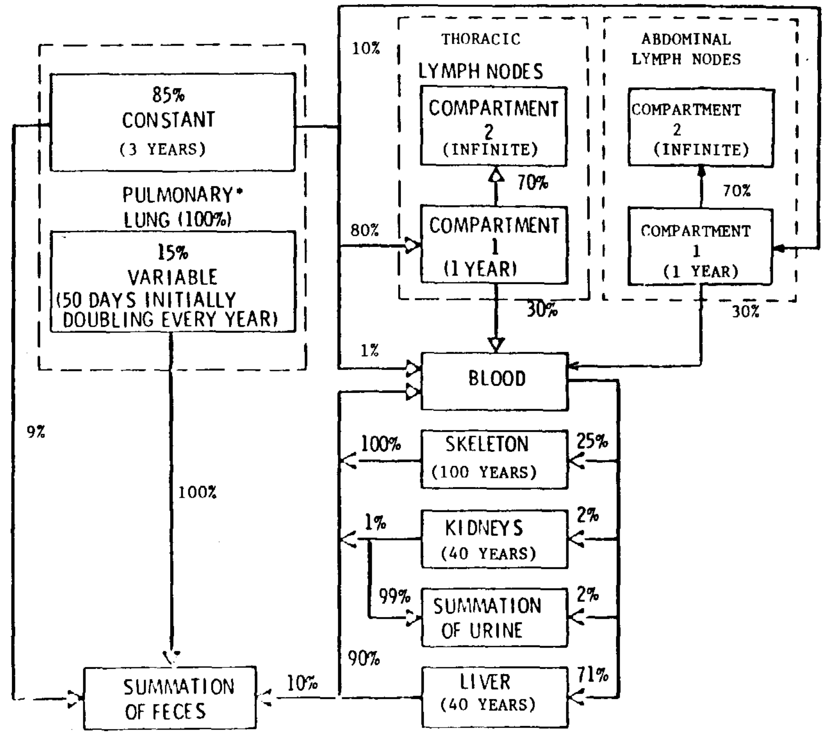
T_k^b = Biological half-time (days) for pathway k

f_k = Fraction cleared by pathway k



* INITIAL ALVEOLAR DEPOSITION

FIGURE 5. BLOCK DIAGRAM OF THE SHORT-TERM FORM OF THE SDB MODEL



* BASED ON 100% OF THE INITIAL ALVEOLAR DEPOSITION.

FIGURE 6. BLOCK DIAGRAM OF THE LONG-TERM FORM OF THE SDB MODEL

$$r_{GIT} = f_{URG}\lambda_{NPTB}y_{NPTB} + f_{LCG}\lambda_{LC}y_{LC} + \lambda_{LV}y_{LV} + f_{LG}\lambda_{L}y_{LIVER} \tag{59}$$

$$\begin{aligned} r_B = {} & f_{URB}\lambda_{NPTB}y_{NPTB} + f_{LCB}\lambda_{LC}y_{LC} \\ & + f_{TLB}\lambda_{TL}y_{TLNM} + f_{ALB}\lambda_{AL}y_{ALNM} + f_{LB}\lambda_{L}y_{LIVER} \\ & + f_{KB}\lambda_{K}y_{K} + \lambda_{B}y_{BONE} \end{aligned} \tag{60}$$

$$dy_{NPTB}/dt = (0.7)(D_3 + D_4 + D_5)A_m - (\lambda_A + \lambda_{NPTB})y_{NPTB} \tag{61}$$

$$dy_{LC}/dt = (0.3)(0.85)(D_3 + D_4 + D_5)A_m - (\lambda_A + \lambda_{DC})y_{LC} \tag{62}$$

$$dy_{LV}/dt = (0.3)(0.15)(D_3 + D_4 + D_5)A_m - (\lambda_A + \lambda_V)y_{LV} \tag{63}$$

$$\lambda_{LV} = [\ln(2)/50]\exp(\ln(2)t/365) \tag{64}$$

$$y_P = y_{LC} + y_{LV} \tag{65}$$

$$dy_{TLNM}/dt = f_{LCTL}\lambda_{LC}y_{LC} - (\lambda_A + \lambda_{TL})y_{TLNM} \tag{66}$$

$$dy_{TLNR}/dt = f_{TLTL}\lambda_{TL}y_{TLNM} - \lambda_A y_{TLNR} \tag{67}$$

$$dy_{ALNM}/dt = f_{LCAL}\lambda_{LC}y_{LC} - (\lambda_A + \lambda_{AL})y_{ALNM} \tag{68}$$

$$dy_{ALNR}/dt = f_{ALAL}\lambda_{AL}y_{ALNM} - \lambda_A y_{ALNR} \tag{69}$$

where

r, λ, y, f, D and A_m are as defined previously in the ICRP II and Task Group models. The subscripts, where different, are as follows: NPTB refers to the combined nasopharynx-tracheobronchial region; LC refers to that portion of the pulmonary lung with a constant biological half-time, LV refers to the variable half-time portion, and P refers to the combined portions; TLNM and ALNM refer to those portions of the thoracic and abdominal lymph nodes, respectively, in which plutonium is mobile while TLNR and ALNR refer to the corresponding portions where plutonium is retained indefinitely; LCG refers to transfer from LC to GIT, LG is from liver to GIT, LCB is from LC to blood, TLB is from TLNM to blood, ALB is from ALNM to blood, LB is liver to blood, KB is kidney to blood, LCTL LC to TLNM, and LCAL is LC to ALNM.

Values of the parameters in the above equations can be taken from Figures 5 and 6. The equations for GIT, liver, kidney, and bone are identical to the ICRP II and Task Group models, but the paramerer values are those indicated in Figure 6. Stuart *et al.* (1971) did not give estimates for total body.

Modifications to the SDB Model

We (Bloom and Martin, 1976) modified the SDB, long-term model to include ingestion and the possibility of transfer from the blood to lungs and to lymph nodes. We also incorporated many of the long-term transfers of the Task Group model. However, we also simplified the model somewhat by removing the variable half-life lung compartment from the SDB model. The resulting model is shown in Figure 7. In this figure, the Upper Respiratory Tract (URT) refers to the nasopharyngeal and tracheobroncheal regions, while Deep Lung (DL) refers to the long-term component of the pulmonary compartment in the Task Group model.

The equations for our modifications to the SDB model are

$$r_{GIT} = (D_3 + D_4 + D_5)f_{URG}A_m + f_{DLG}\lambda_{DL}y_{DL} + f_{LG}\lambda_L y_{LIVER} + H_m \qquad (70)$$

$$\begin{aligned} r_B = (D_3 + D_4 + D_5)f_{URB}A_m &+ f_{GITB}r_{GIT} + f_{DLB}\lambda_{DL}y_{DL} \\ &+ f_{TLB}\lambda_{TL}y_{TLNM} + f_{ALB}\lambda_{AL}y_{ALNM} + f_{LB}\lambda_L y_{LIVER} \\ &+ f_{KB}\lambda_K y_K + \lambda_B y_{BONE} \end{aligned} \qquad (71)$$

$$dy_{DL}/dt = (D_3 + D_4 + D_5)f_{URDL}A_m + f_{BDL}r_B - (\lambda_A + \lambda_{DL})y_{DL} \qquad (72)$$

$$dy_{TLNM}/dt = f_{DLTL}\lambda_{DL}y_{DL} + f_{BTL}r_B - (\lambda_A + \lambda_{TL})y_{TLNM} \qquad (73)$$

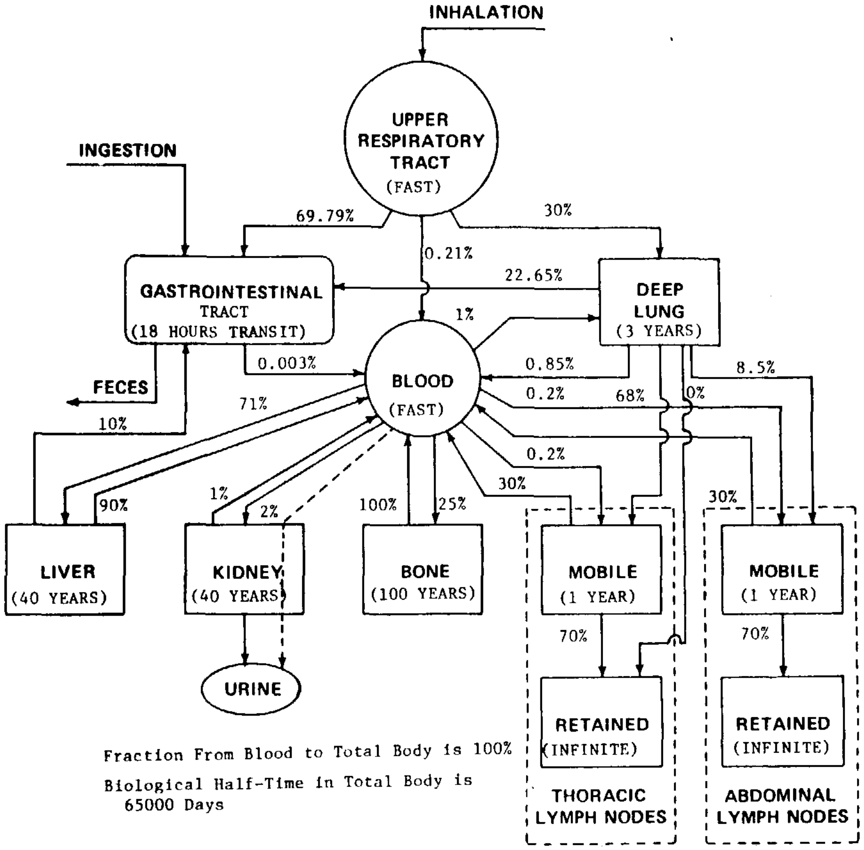
$$dy_{TLNR}/dt = f_{TLTL}\lambda_{TL}y_{TLNM} + f_{DLTR}\lambda_{DL}y_{DL} - \lambda_A y_{TLNR} \qquad (74)$$

$$dy_{ALNM}/dt = f_{DIAL}\lambda_{DL}y_{DL} + f_{BAL}r_B - (\lambda_A + \lambda_{AL})y_{ALNM} \qquad (75)$$

$$dy_{ALNR}/dt = f_{ALAL}\lambda_{AL}y_{ALNM} - \lambda_A y_{ALNR} \qquad (76)$$

$$dy_{LIVER}/dt = f_{BL}r_B - (\lambda_A + \lambda_L)y_{LIVER} \qquad (77)$$

$$dy/dt = f_{BK}r_B - (\lambda_A + \lambda_K)y_K \qquad (78)$$



Fraction From Blood to Total Body is 100%

Biological Half-Time in Total Body is 65000 Days

FIGURE 7. BLOCK DIAGRAM FOR MODIFICATIONS TO THE SDB MODEL

$$dy_{BONE}/dt = f_{BBN}r_B - (\lambda_A + \lambda_B)y_{BONE} \quad (79)$$

$$dy_{TB}/dt = f_{BTB}r_B - (\lambda_A + \lambda_{TB})y_{TB} \quad (80)$$

where

r, λ, y, f, D, A_m, and H_m are as defined previously in the ICRP II, Task Group, and SDB models. The subscript URDL refers to transfer from URT to DL, BDL is from blood to DL, DLTL is from DL to TLNM, BTL is from blood to TLNM, DLTR is from DL to TLNR, DLAL is from DL to ALNM, and BAL is from blood to ALNM.

Parameters for the above equations can be obtained from Figure 7. Most of these values are identical to the SDB model (Stuart *et al.*, 1971). Our modifications are designed to make the model more applicable to chronic inhalation and to include ingestion, transfer of plutonium from blood to DL, and from blood to the lymph nodes. The value for the transfer from GIT to blood is identical to the ICRP II value. The values for the other parameters are based on data reported by Ballou *et al.* (1972) for the translocation of a soluble form of plutonium (plutonium citrate) in dogs. Stuart *et al.* (1971) recommended a variable half-time for part of the transfer rate from the lungs to blood but it was felt that a constant, 3-year half-time for the entire lung was an adequate representation of their dog data. It was also much easier to use the constant half-time for the mathematical description of chronic inhalation.

Like the SDB model, we assumed that 30 percent of the material initially deposited in URT is rapidly transferred to DL. Since the short-term form of the SDB model transfers 99.7 percent of the material in URT to GIT in a short time (8 minutes), the effective transfer from URT to GIT is 69.79 percent (0.997 x 0.70). The corresponding transfer from URT to blood is 0.21 percent (0.003 x 0.70).

MODEL COMPARISONS

The preceding models were compared by computing organ burdens and radiation doses resulting from unit intakes by ingestion or inhalation. The units were either 1 μCi as a single intake or 1 μCi/day continuous intake and the calculations were carried to 50 years. The resulting organ burdens and doses after 50 years are given in Tables 10 and 11.

Table 10. Comparison of Calculations for 1 μCi Single Intake Using Various Models

Model[b]	Path	URT[c]	GI Tract	Lung	Lymph Nodes Thoracic	Lymph Nodes Abdom.	Liver	Kidney	Bone	Total Body
50-Year Organ Burdens, μCi										
ICRP II	Ingestion		0.00	0.00			2.95E-6[d]	4.03E-7	2.01E-5	2.47E-5
Task Group	Ingestion	0.00	0.00	0.00	0.00		5.67E-6	4.03E-7	9.53E-6	2.47E-5
SDB	Ingestion	0.00	6.37E-11	0.00	0.00	0.00	1.74E-5	4.91E-7	9.10E-6	2.70E-5
Modified	Ingestion		6.37E-11	1.47E-8	2.90E-7	9.54E-8	1.74E-5	4.91E-7	9.10E-6	4.11E-5
ICRP II	Inhalation[e]		0.00	1.11E-16			2.49E-2	3.37E-3	1.68E-1	2.05E-1
Task Group	Inhalation	7.26E-17	1.31E-15	1.89E-12	4.64E-3		1.22E-2	8.30E-4	1.95E-2	4.98E-2
SDB	Inhalation	0.00	8.30E-8	1.70E-5	7.45E-2	9.38E-3	2.27E-2	6.39E-4	1.14E-2	1.19E-1
Modified	Inhalation		8.30E-8	2.06E-5	7.45E-2	9.38E-3	2.27E-2	6.39E-4	1.14E-2	5.10E-2
50-Year Cumulative Dose to Organs, Rem										
ICRP II	Ingestion		6.66E-2	0.00			1.07E-1	8.18E-2	7.94E-1	1.93E-2
Task Group	Ingestion	0.00	2.50E-2	0.00	0.00		2.63E-1	8.18E-2	4.05E-1	1.93E-2
SDB	Ingestion	0.00	6.66E-2	0.00	0.00	0.00	5.62E-1	8.96E-2	2.99E-1	2.02E-2
Modified	Ingestion		6.66E-2	4.00E-3	7.48E-1	3.82E-1	5.62E-1	8.96E-2	2.99E-1	2.55E-2
ICRP II	Inhalation[e]		4.16E-2	1.79E+2			9.04E+2	6.82E+2	6.61E+3	1.61E+2
Task Group	Inhalation	7.27E-1	3.04E-2	7.29E+2	2.59E+4		5.03E+2	1.54E+2	7.59E+2	3.50E+1
SDB	Inhalation	1.97E-2	2.67E-2	1.13E+3	2.28E+5	4.30E+4	6.42E+2	1.03E+2	3.36E+2	8.73E+1
Modified	Inhalation		2.67E-2	1.34E+3	2.28E+5	4.30E+4	6.42E+2	1.03E+2	3.36E+2	2.84E+1

(a) Particle size assumed to be 0.5 μm activity mean aerodynamic diameter.

(b) ICRP is International Commission on Radiological Protection, Report of Committee II (1959); Task Group is Task Group on Lung Dynamics (Morrow, 1966) as modified in ICRP-19 (1972); SDB is Stuart, Dionne, and Bair (1968, 1971); Modified is our modifications to the SDB model (Bloom and Martin, 1976).

(c) URT is upper respiratory tract which includes the nasopharyngeal and tracheobronchial regions.

(d) Numbers following E are exponents of 10.

(e) Particles assumed insoluble for lung and GI tract, soluble for other organs.

Table 11. Comparison of Calculations for 1μ Ci/Day Intake(a) Using Various Models

Model(b)	Path	URT(c)	GI Tract	Lung	Lymph Nodes Thoracic	Lymph Nodes Abdom.	Liver	Kidney	Bone	Total Body
50-Year Organ Burdens, μCi										
ICRP II	Ingestion		7.50E-1(d)		0.00		6.70E-2	9.04E-3	4.02E-3	4.97E-1
Task Group	Ingestion	0.00	7.50E-1	0.00	0.00		1.65E-1	9.04E-3	2.08E-1	4.97E-1
SDB	Ingestion	0.00	7.50E-1	0.00	0.00	0.00	3.50E-1	9.90E-3	1.52E-1	5.12E-1
Modified	Ingestion		7.50E-1	7.37E-4	3.41E-3	1.41E-3	3.52E-1	9.91E-3	1.54E-1	6.57E-1
ICRP II	Inhalation(e)		4.69E-1	6.58E+1			5.66E+2	7.54E+1	3.55E+3	4.14E+3
Task Group	Inhalation	1.07E-1	3.42E-1	1.35E+2	1.42E+2		3.15E+2	1.70E+1	3.90E+2	9.26E+2
SDB	Inhalation	2.92E-3	3.00E-1	2.48E+2	1.26E+3	1.58E+2	4.02E+2	1.13E+1	1.72E+2	2.25E+3
Modified	Inhalation		3.00E-1	2.48E+2	1.26E+3	1.58E+2	4.02E+2	1.13E+1	1.72E+2	7.38E+2
50-Year Cumulative Dose to Organs, Rem										
ICRP II	Ingestion		1.22E+3	0.00			1.04E+3	7.96E+2	7.45E+3	1.82E+2
Task Group	Ingestion	0.00	1.22E+3	0.00	0.00		2.74E+3	7.96E+2	3.97E+3	1.82E+2
SDB	Ingestion	0.00	1.22E+3	0.00	0.00	0.00	5.28E+3	8.48E+2	2.62E+3	1.87E+2
Modified	Ingestion		1.22E+3	5.52E+1	6.03E+3	3.19E+3	5.30E+3	8.46E+2	2.64E+3	2.21E+2
ICRP II	Inhalation(e)		7.59E+2	3.17E+6			8.82E+6	6.63E+6	6.21E+7	1.51E+6
Task Group	Inhalation	1.33E+4	5.48E+2	1.28E+7	3.05E+8		4.74E+6	1.36E+6	6.79E+6	3.09E+5
SDB	Inhalation	3.60E+2	4.82E+2	2.24E+7	1.94E+9	3.66E+8	5.43E+6	8.66E+5	2.67E+6	8.17E+5
Modified	Inhalation		4.82E+2	2.24E+7	1.94E+0	3.66E+8	5.43E+6	8.66E+5	2.67E+6	2.22E+5

(a) Particle size assumed to be 0.5 μm activity mean aerodynamic diameter.

(b) ICRP is International Commission on Radiological Protection, Report of Committee II (1959); Task Group is Task Group on Lung Dynamics (Morrow, 1966) as modified in ICRP-19 (1972); SDB is Stuart, Dionne, and Bair (1968, 1971); Modified is our modifications to the SDB model (Bloom and Martin, 1976).

(c) URT is upper respiratory tract which includes the nasopharyngeal and tracheobronchial regions.

(d) Numbers following E are exponents of 10.

(e) Particles assumed insoluble for lung and GI tract, soluble for other organs.

Stuart *et al.*, 1971, applied their model to single inhalations and the variable half-time in the pulmonary lung did not present any difficulty. However, there is some doubt as to the interpretation of the variable half-time for the chronic case. A strict interpretation would imply that a significant fraction (15 percent) of material continuously deposited in the pulmonary lung is eliminated with a half-time which exceeds 140 years after 10 years of chronic inhalation. The lung burden could thus reach very high levels. There may indeed be a fraction which is retained with a long half-time, but we find it difficult to believe this fraction could be so high (15 percent). We therefore examined the long-term behavior of the burden in this portion of the lung resulting from a single inhalation. Equations (63) and (64) can be solved to yield

$$y_{LV} = 0.15y_P(0)\exp(-\lambda_A t)\exp(-7.3[1-\exp(-0.001899t)]) \quad (81)$$

where

$y_P(0)$ is the initial amount deposited in the pulmonary lung.

Neglecting radioactive decay (λ_A=0), the initial fraction (15 percent) is reduced to 0.4 percent after one year, 0.013 percent after five years, and assymptotically approaches a constant value of about 0.01 percent. On the basis of these results, we assumed this fraction (0.01 percent) is retained indefinitely, while the remaining 14.99 percent is removed with a three year biological half-time.

Upon examining the calculated results in Tables 10 and 11, it is apparent that inhalation is the critical pathway for plutonium to all organs except GIT. The organ burdens and radiation doses from inhalation are generally 1,000 to 10,000 times greater than the corresponding burdens and doses from ingesting the same amount of plutonium. This is due to the relatively large fraction (0.2 to 25 percent) which reaches the blood directly from inhalation versus the relatively small fraction (0.003 percent) from ingestion. For ingestion, the bone is the critical organ for the ICRP-II and Task Group models, while liver is critical for the SDB and Modified models. This difference is explained by the fraction transferred from blood to the organ which is 71 percent to the liver and 25 percent to bone for the SDB and Modified models, while the corresponding values for the ICRP-II model are 15 and 80 percent and those for the Task Group model are 45 and 45 percent. Where the fractions are equal, the bone has the larger burden and dose because it has the larger biological half-time.

For inhalation, the lung is the critical organ for all models except ICRP-II. The dose to lymph nodes is actually higher, but ICRP (1959) does not recognize lymph nodes as critical organs. For the ICRP-II model, the bone is the critical organ because this model has the highest fraction of inhaled material which reaches the blood immediately (25 percent) and the shortest biological half-time in lung (365 days).

In spite of differences in translocation pathways and biological half-times, the radiation doses to critical organs are surprisingly similar for a given intake situation. This leads us to use the Task Group model because it is recognized by ICRP (ICRP-19, 1972) and the results using this model are not too different from the more elaborate SDB and Modified models. Although still the official model of ICRP, the ICRP-II model is generally considered to be outdated. The Task Group model was used to calculate the accumulated doses and dose commitments (to 70 years) due to constant intake rates [$A_m = 0.002\ C_s$ (pCi/day) and $H_m = 0.19\ C_s$ (pCi/day)], and the results are shown in Figures 8 and 9. Detailed tables of calculations based on 1 pCi/day ingestion or inhalation are given in Appendix A.

PRACTICAL APPLICATIONS

Our purpose in this discussion is to show how the results of a transport and dose estimation model may be applied to the practical problem of deciding whether and to what extent environmental decontamination might be required to limit or reduce potential health hazards. The procedure suggested for this purpose and outlined below is analogous to the procedure followed by ICRP in calculating maximum permissible concentrations of radionuclides in air and water. The principal steps involved are: (1) identification of the critical exposure pathway, (2) identification of the critical organ or organs, (3) selection of maximum permissible dose criteria, (4) calculation of the corresponding maximum permissible concentration of Pu in soil $(MPC)_s$, and (5) comparison of the $(MPC)_s$ with estimated inventories of Pu in the surface soils of contaminated areas at NTS.

CRITICAL PATHWAY

The estimated plutonium ingestion rate for a hypothetical Standard Man living in a contaminated area at NTS is about 100 times the estimated inhalation rate; but, owing to the very small fraction of plutonium transferred from the gastrointestinal tract to blood (3×10^{-5}), the gastrointestinal tract is the only organ that receives a significant dose from ingested plutonium. The preferred dose estimation model based on ICRP recommendations (the Task Group model, Figure 4) shows that inhalation accounts for 100 percent of the plutonium deposited in the lungs and thoracic lymph nodes; and, for an ingestion/inhalation ratio of 100, inhalation accounts for about 95 percent of the plutonium in bone, liver, and kidney after 50 years of chronic exposure (Table 6). Clearly, inhalation is the critical pathway.

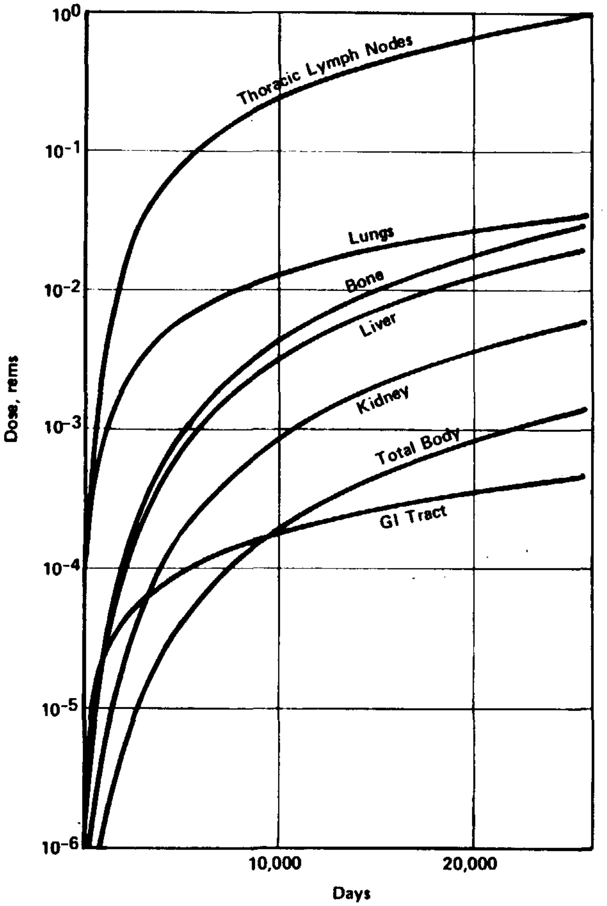


FIGURE 8. PREDICTED CUMULATIVE DOSES DUE TO ^{239}Pu IN DIFFERENT ORGANS OF STANDARD MAN

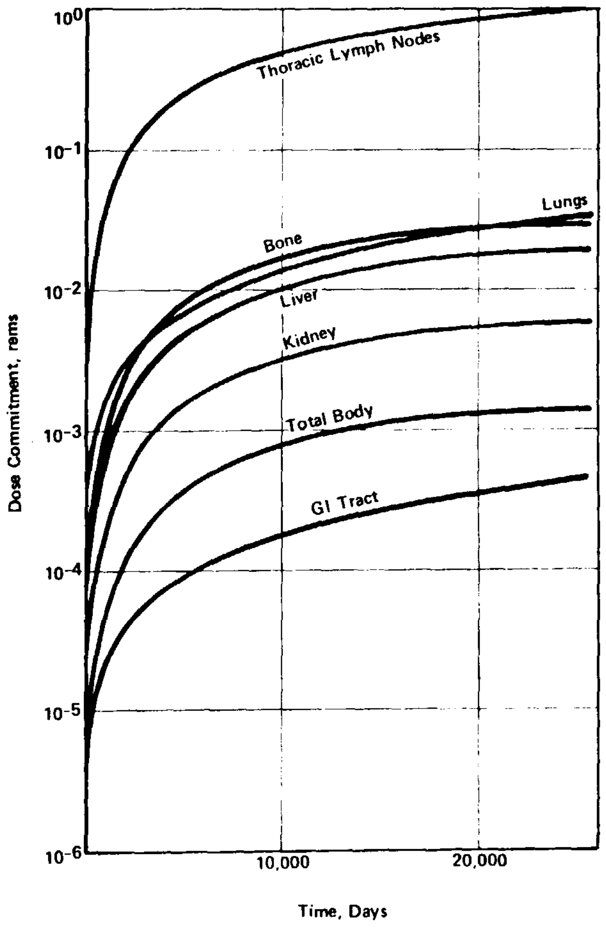


FIGURE 9. PREDICTED DOSE COMMITMENTS DUE TO ^{239}Pu IN DIFFERENT ORGANS OF STANDARD MAN

CRITICAL ORGAN

According to the Task Group model (Figure 4 and Appendix A), thoracic lymph nodes receive the highest dose (Figures 8 and 9) but the critical organs recognized by ICRP (1959) are bone, if the Pu is "soluble," and lung if the Pu is "insoluble." For relatively short exposure times, the model, which makes no distinction between "soluble" and "insoluble," predicts that the cumulative dose to lungs would be considerably higher than the cumulative dose to bone (Figure 8), but the cumulative doses to 70 years are about the same. For exposure times longer than 70 years, the dose to bone would be higher than the dose to lungs. This is due to the relatively short biological half-life of plutonium in lungs (500 days) compared to that of bone (36,500 days). As the estimated dose to lungs is higher than the estimated dose to bone, the exposure periods < 70 years, and the permissible dose to lungs is lower (see below), the lung (i.e., the pulmonary region of the respiratory tract) is the critical organ.

PERMISSIBLE DOSE CRITERIA

Current ICRP (1966) recommendations concerning "dose limits for individual members of the public" indicate that the dose to lungs should not exceed "1.5 rems in a year." Annual dose rates to a given organ can be estimated on the basis of predicted organ burdens as a function of exposure time. For present purposes, we shall consider only the equilibrium lung burden which, for practical purposes, is constant for chronic exposure times in excess of about 10 years.

ESTIMATION OF ASC

The acceptable soil concentration (ASC) is defined as the average concentration of ^{239}Pu in the soil of an area occupied by the hypothetical Standard Man which would result in a dose to the lungs equal to or less than the permissible dose. This value is estimated as follows:

$$\mathrm{ASC} = (1.5/365)/(E\ Y_{eq}/m) = 2817\ \mathrm{pCi}(^{239}\mathrm{Pu})/\mathrm{g(soil)}$$

where

(1.5/365) is the "permissible dose rate" in rem/day,

E for ^{239}Pu is 51.2159 x 53 = 27144 (rem/day)/μCi or 0.0027144 (rem/day)/pCi, m is 500 g for lung,

$Y_{eq} = 0.002\ C_s y$,

y is the lung burden after 50 years = 134.34 μCi per μCi/day inhaled (see Tables 11 and A-1).

and

$0.002\ C_s$ is the plutonium inhalation rate (pCi/day), i.e., 20 m^3(air)/day x 10^{-4} g(soil)/m^3(air), and C_s is the average soil concentration (pCi/g).

COMPARISON OF ASC AND SOIL INVENTORY DATA

Gilbert *et al.* (1975, p. 379) have summarized the estimated inventories of $^{239,240}Pu$ in the surface soils (0-5 cm depth) at NTS. In each contaminated study area, soil sampling was stratified according to contour intervals (strata) previously established by field instrument (FIDLER) surveys. The pertinent results for Area 13 are given in Table 12.

Table 12. Estimated Inventory of $^{239,240}Pu$ in Surface Soil (0-5 cm Depth) in Area 13

Strata(a)	Area (m^2)(a)	Mean Pu (μCi/m^2)(a)	Soil Concentration (pCi/g)(b)
1	874,500	1.9	34
2	2,552,400	5.8	103
3	81,100	23.0	411
4	73,700	54.0	952
5	20,900	110.0	1,847
6	23,400	820.0	14,245
Total	3,626,000	Mean	213

(a) From Gilbert *et al.* (1975, p. 379).
(b) From Table 1, this report.

Only stratum 6 exceeds ASC = 2,800 pCi/g. Complete decontamination of stratum 6 (0.65 percent of Area 13) would remove about 43 percent of the total plutonium in Area 13 and reduce the average soil concentration from 213 to 121 pCi/g. If it were decided to decontaminate all areas at NTS in which the average soil concentration exceeds 2 nCi/g, decontamination would be required for about 0.2 km^2 (about 50 acres) of the 11.4 km^2 (about 2,812 acres) included in the soil inventory study (Table 1). If the decontamination criteria were further reduced to 1 nCi/g, the area requiring decontamination would be less than 0.4 km^2 (100 acres). In other words, the plutonium contamination at NTS is so concentrated in areas near ground zero sites that decontamination of from 2 to 4 percent of the total soil inventory area would reduce average soil concentrations by 40 to 50 percent. However, it should be noted that effective decontamination of these areas may be extremely difficult (Wallace and Romney, 1975; Rhoads, 1976).

DISCUSSION

Based on the results derived above (ASC = 2,800 pCi/g) and the mass loading factor of 100 μg(soil)/m^3(air), the expected air concentration would be 2.8 x 10^{-13} μCi/cm^3. The maximum permissible concentration in air (MPC_a) indicated by ICRP Publication 2 (10^{-12} μCi/cm^3) is higher than this by a factor of 3.6. Based on $MPC_a = 10^{-12}$ μCi/cm^3 and a mass loading factor of 100 μg/m^3, the acceptable soil concentration would be 10 nCi/g instead of 2.8 nCi/g which would be equivalent to assuming a mass loading factor of 355 instead of 100 μg (soil)/m^3(air).

Another conservative factor in our estimate of ASC is that the lung deposition factor ($D_s = 0.31$) is based on the assumption that the mean size of resuspended soil particles of 0.5 μm (AMAD). The value obtained from cascade impactor studies in the GMX area was 3 μm (AMAD) which would indicate $D_s \le 0.2$. Changing only this parameter would increase the estimate of ASC, by a factor of 1.55, to 4,266 pCi/g.

The least conservative factor involved in arriving at ASC = 2.8 nCi/g is the assumed mass loading factor of 100 μg(soil)/m^3(air). As demonstrated by Shinn and Anspaugh (1975) and Anspaugh *et al.* (1975), this estimate appears to be adequate for undisturbed areas and normal winds, but high winds or mechanical disturbances such as vehicular traffic, plowing, excavation, etc., might increase the mass loading factor to several mg/m^3. If we assume, for example, that the hypothetical Standard Man at NTS were exposed, for one reason or another, to mass loading factors of 5,000 μg/m^3 during 30 days each year, the average mass loading factor would increase to about 500 μg/m^3, and our estimate of ASC would decrease to about 560 pCi/g.

The point of this discussion is that the notion of an "acceptable soil concentration" is not fixed, but very much dependent on how man plans to utilize a contaminated area. Under present conditions, the ASC for contaminated areas at NTS is 2.8 nCi/g. If these same areas were to be used for agricultural purposes or for any other purpose which would tend to increase the average mass loading factor, a lower ASC would be indicated.

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APPENDIX A

EQUATIONS FOR ESTIMATING ORGAN BURDEN, RADIATION DOSE, AND DOSE COMMITMENT DUE TO INHALATION AND INGESTION OF TRANSURANICS AT CONSTANT RATES

Definitions

- Figure 1 is a schematic diagram of the deposition and clearance model proposed by the Task Group on Lung Dynamics (TGLD) of the International Commission on Radiological Protection (ICRP)[1].
- The respiratory tract is divided into three regions: the nasoypharyngeal (NP), the tracheobronchial (TB), and the pulmonary (P).
- Radioactive dust deposited in the different regions of the respiratory tract is cleared by various mechanisms and pathways to the gastrointestinal tract (GIT), to the thoracic lymph nodes (LM), or to the blood (B). Radioactive materials entering the GIT and LM may be transferred to B and from B to other organs of the body.
- Q_I is the radionuclide inhalation rate (pCi/day).
- Q_F is the radionuclide ingestion rate (pCi/day).
- D_3, D_4, and D_5 are the fractions of radioactive dust deposited initially in the NP, TB, and P regions respectively.
- f_k is the fraction cleared by pathway k
- k = a, b, ...h
- f_1 is the fraction of the radionuclide entering the GIT which is cleared to B

- f'_{2n} is the fraction of the radionuclide entering B which is transferred to organ n
- n = bone, liver, kidney, etc.
- T_r is the half-life of the radionuclide (day).
- T_k is the biological half-life of radioactive dust deposited in subcompartment k of the respiratory tract (day)
- T_n is the biological half life of the radionculide deposited in organ n (day).
- $\ln(2)/T_r = \lambda_r(\mathrm{day}^{-1})$
- $\ln(2)/T_k = \lambda_k^b(\mathrm{day}^{-1})$
- $\ln(2)/T_n = \lambda_n^b(\mathrm{day}^{-1})$
- $\lambda_k = \lambda_k^b + \lambda_r$
- $\lambda_n = \lambda_n^b + \lambda_r$
- $R_n = 5.1216 \times 10^{-5}\ E_n/M_n$ is the dose conversion factor

  (rem/pCi·day) for region or organ n.
- E_n is the energy deposited by radionuclide in region or organ n (MeV/dis). Same as ΣEF(RBE)n in Table 5 of Refs.[4,5].
- M_n is the mass of region or organ n (g)
- M_{NP} = 1.35 g, M_{TB} = 400g, M_P = 500g [6,7]. For other organs see Refs. [3,4,5].

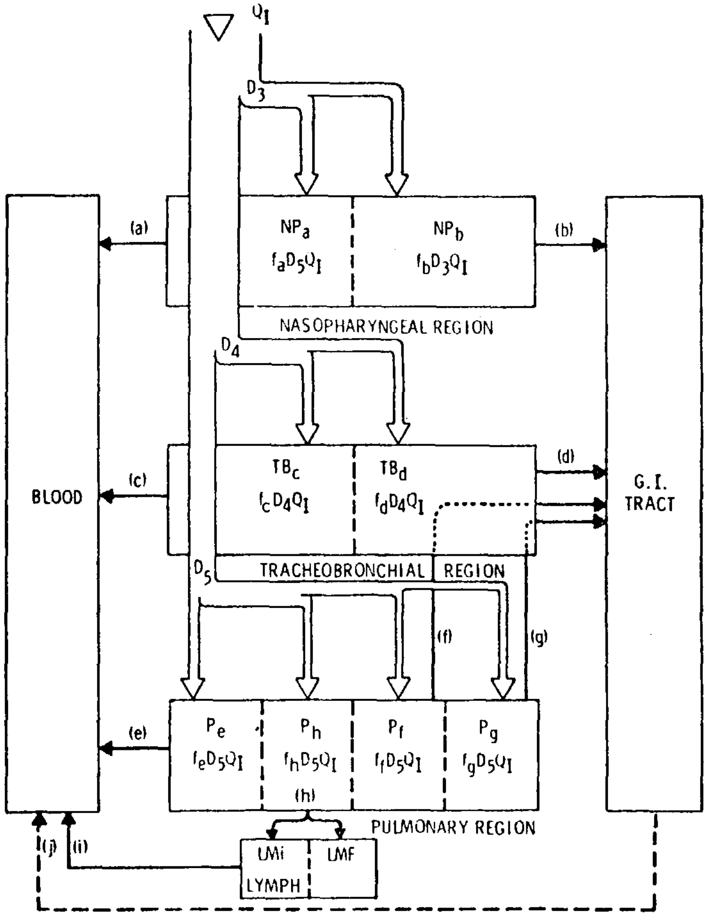

FIGURE 1. Schematic Diagram of the Task Group Lung Model (8)

Table I. Task Group Lung Model Parameter Values

FRACTION OF INHALED PARTICLES DEPOSITED IN THE RESPIRATORY SYSTEM VERSUS PARTICLE DIAMETER$^{(a)}$

Particle Size AMAD Micrometers	Fraction of Inhaled Quantity Retained		
	Nasopharyngeal Region, D_3	Tracheobronchial Region, D_4	Pulmonary Region, D_5
0.05	0.001	0.08	0.59
0.1	0.008	0.08	0.50
0.3	0.063	0.08	0.36
0.5	0.13	0.08	0.31
1.0	0.29	0.08	0.23
2.0	0.50	0.08	0.17
5.0	0.77	0.08	0.11

CLEARANCE PARAMETER VALUES$^{(b)}$

Compartment		Translocation Class					
		D		W		Y	
	k	T_k^b	f_k	T_k^b	f_k	T_k^b	f_k
NP	a	0.01	0.50	0.01	0.10	0.01	0.01
	b	0.01	0.50	0.40	0.90	0.40	0.99
TB	c	0.01	0.95	0.01	0.50	0.01	0.01
	d	0.20	0.05	0.20	0.50	0.20	0.99
P	e	0.50	0.80	50	0.15	500	0.05
	f	n.a.	n.a.	1	0.40	1	0.40
	g	n.a.	n.a.	50	0.40	500	0.40
	h	0.50	0.20	50	0.05	500	0.15
L	i	0.50	1.00	50	1.00	1000	0.90

(a) Estimated from data of Task Group on Lung Dynamics.(9)

(b) As amended by ICRP Publication 19 (2).

D.W.Y. = Days, Weeks, Years

1.c. = Clearance pathway (See Figure 1)

T_k^b = Biological half-time (days) for pathway k

f_k = Fraction cleared by pathway k

PARAMETER VALUES

Typical values of D_3, D_4, and D_5 and recommended values of f_k and T_k[3] are given in Table 1. Table 3 of Ref. [1] gives the pulmonary clearance classification (D,W,Y) of inorganic compounds. For suitable values of f_1, f_2', T_r, T_n, M_n, and E_n(= ΣEF(RBE)n), see Refs. [3,4,5].

GENERAL TREATMENT

The subcompartments of the respiratory tract are identified by clearance pathways (K = a,b,...h). Equations for calculating burden, dose, and dose commitment for a given subcompartment are as follows:

Burden = Q_k

$$\frac{dQ_k}{dt} = f_k D_n Q_I - \lambda_k Q_k$$

$$S\bar{Q}_k + \lambda_k \bar{Q}_k = f_k D_n Q_I / S, \text{ if } Q_I \text{ is constant}$$

$$\bar{Q}_k = f_k D_n Q_I / S(S + \lambda_k)$$

$$Q_k = (f_k D_n Q_I / \lambda_k)(1 - \exp(-\lambda_k t)) \tag{1}$$

Dose = D_k

$$D_k = R_k f_k D_n Q_I / \lambda_k \int_o^t (1 - \exp(-\lambda_k t))dt$$

$$D_k = (R_k f_k D_n Q_I / \lambda_k)\left(t - \frac{1 - \exp(-\lambda_k t)}{\lambda_k}\right) \tag{2}$$

Dose Commitment = C_k

If exposure (inhalation) should stop at time t, the additional dose (D_{Ak}) delivered to subcompartment k by the radionuclide present at time t would be

$$D_{Ak} = (R_k f_k D_n Q_I/\lambda_k)(1-\exp(-\lambda_k t))\int_o^{T-t} \exp(-\lambda_k(T-t))dt$$

$$D_{Ak} = (R_k f_k D_n Q_I/\lambda_k)(1-\exp(-\lambda_k t))[1-\exp(-\lambda_k(T-t))/\lambda_k[$$

where T is any time subsequent to t, i.e., T > t

$$C_k = D_k + D_{Ak}$$

$$C_k = (R_k f_k D_n Q_I/\lambda_k)[t- \frac{1-\exp(-\lambda_k t)}{\lambda_k} \exp(-\lambda(T-t))] \qquad (3)$$

EQUATIONS USED FOR NTS PLUTONIUM MODEL

Pulmonary Region = Lungs

$$Q_P = D_5 Q_I[(f_e/\lambda_e)(1-\exp(-\lambda_e t)) + (f_f/\lambda_f)(1-\exp(-\lambda_f t))$$
$$+ (f_g/\lambda_g)(1-\exp(-\lambda_g t)) + (f_h/\lambda_h)(1-\exp(-\lambda_h t))] \qquad (4)$$

As $\lambda_e = \lambda_g = \lambda_h$, $f_e + f_g + f_h = 0.6$, and $f_f = 0.4$, Eq. (4) reduces to

$$Q_P = D_5 Q_I \left[0.6 \frac{1-\exp(-\lambda_h t)}{\lambda_h} + 0.4 \frac{1-\exp(-\lambda_f t)}{\lambda_f}\right]$$

For $t \geq 10$, $1-\exp(-\lambda_f t) \rightarrow 1$, and

$$Q_P = D_5 Q_I \ [0.6(1-\exp(-\lambda_h t))/\lambda_h + 0.4/\lambda_f] \qquad (5)$$

is an accurate approximation.

$$D_P = R_P D_5 Q_I \left[\frac{0.6}{\lambda_h}\left(t - \frac{1-\exp(-\lambda_h t)}{\lambda_h}\right) + \frac{0.4t}{\lambda_f}\right] \qquad (6)$$

$$C_P = R_P D_5 Q_I \left[\frac{0.6}{\lambda_h}\left(t - \frac{1-\exp(-\lambda_h t)}{\lambda_h} \exp(-\lambda_h(T-t)\right)\right.$$
$$\left. + \frac{0.4t}{\lambda_f}\right] \qquad (7)$$

THORACIC LYMPH NODES

The thoracic lymph nodes (L) receive radionuclide from P_h at the biological elimination rate, λ_h^b.

For class Y compounds, there are two subcompartments, Li and Lr. Li receives 90 percent of the radionuclide in P_h and transfers it to B at the biological elimination rate, λ_i^b. The remaining 10 percent is retained in Lr from which it is lost by radioactive decay, λ_r, only.

$$Q_L = Q_{Li} + Q_{Lr}$$

$$\frac{dQ_{Li}}{dt} + \lambda_i Q_{Li} = f_i \lambda_h^b Q_h; \ \frac{dQ_{Lr}}{dt} + \lambda_r Q_{Lr} = (1-f_i)\lambda_h^b Qh$$

$$\bar{Q}_{Li} = f_i \lambda_h^b \bar{Q}_h/(S+\lambda_i); \ \bar{Q}_{Lr} = (1-f_i)\lambda_h^b \bar{Q}_h/(S+\lambda_r)$$

$$\bar{Q}_h = f_h D_5 Q_I/S(S+\lambda_h)$$

$$\bar{Q}_L = \lambda_h^b f_h D_5 Q_I \left[\frac{f_i}{S(S+\lambda_h)(S+\lambda_i)} + \frac{1-f_i}{S(S+\lambda_h)(S+\lambda_r)}\right] \qquad (8)$$

Let $K_L = \lambda_h^b f_h D_5 Q_I$

Expansion of Eq. (8), letting $S = 0, -\lambda_h, -\lambda_i$, and $-\lambda_r$, gives

$$Q_L = K_L f_i \left(\frac{1}{\lambda_h \lambda_i} - \frac{\exp(-\lambda_h t)}{\lambda_h(\lambda_i - \lambda_h)} - \frac{\exp(-\lambda_i t)}{\lambda_i(\lambda_h - \lambda_i)} \right)$$

$$+ K_L(1-f_i) \left(\frac{1}{\lambda_h \lambda_r} - \frac{\exp(-\lambda_h t)}{\lambda_h(\lambda_r - \lambda_h)} - \frac{\exp(-\lambda_r t)}{\lambda_r(\lambda_h - \lambda_r)} \right) \qquad (9)$$

Rearranging Eq. (9) gives

$$Q_L = \frac{K_L f_i}{\lambda_h - \lambda_i} \left[\frac{1-\exp(-\lambda_i t)}{\lambda_i} - \frac{1-\exp(-\lambda_h t)}{\lambda_h} \right]$$

$$+ \frac{K_L(1-f_i)}{\lambda_h - \lambda_r} \left[\frac{1-\exp(-\lambda_r t)}{\lambda_r} - \frac{1-\exp(-\lambda_h t)}{\lambda_h} \right] \qquad (10)$$

$$D_L = R_L \left\{ \frac{K_L f_i}{\lambda_h - \lambda_i} \left[\frac{1}{\lambda_i} \left(t - \frac{1-\exp(-\lambda_i t)}{\lambda_i} \right) - \frac{1}{\lambda_h} \left(t - \frac{1-\exp(-\lambda_h t)}{\lambda_h} \right) \right] \right.$$

$$\left. + \frac{K_L(1-f_i)}{\lambda_h - \lambda_r} \left[\frac{1}{\lambda_r} \left(t - \frac{1-\exp(-\lambda_r t)}{\lambda_r} \right) - \frac{1}{\lambda_h} \left(t - \frac{1-\exp(-\lambda_h t)}{\lambda_h} \right) \right] \right\} \qquad (11)$$

$$C_L = R_L \left\{ \frac{K_L f_i}{\lambda_h - \lambda_i} \left[\frac{1}{\lambda_i} \left(t - \frac{1-e^{-\lambda_i t}}{\lambda_i} e^{-\lambda_i (T-t)} \right) \right. \right.$$

$$\left. - \frac{1}{\lambda_h} \left(t - \frac{1-e^{-\lambda_h t}}{\lambda_h} e^{-\lambda_h (T-t)} \right) \right] + \frac{K_L(1-f_i)}{\lambda_h - \lambda_r}$$

$$\left. \left[\frac{1}{\lambda_r} \left(t - \frac{1-e^{-\lambda_r t}}{\lambda_r} e^{-\lambda_r (T-t)} \right) - \frac{1}{\lambda_h} \left(t - \frac{1-e^{-\lambda_h t}}{\lambda_h} e^{-\lambda_h (T-t)} \right) \right] \right\} \qquad (12)$$

T = 70 years = 25,550 days.

The GIT is treated as an open tube, and the lower large intestine (LLI) is the critical organ.

The contents of the LLI (M_G) are assumed to weigh 150 g and to remain in the LLI an average of 18 hours or 0.75 day. The dose to LLI is one half the dose to its contents.

The radionuclide enters the GIT (Fig. 1) by ingestion (Q_F) and by clearance from subcompartments of the respiratory tract. As the biological half-times for pathways b, d, and f are $\leq$ 1 day, the amounts of radionuclide in subcompartments b, d, and f reach equilibrium in $\leq$ 10 days. Thereafter, the rate of transfer from these subcompartments to GIT is constant. Transfer from subcompartment g (T_g = 500 days) is significantly delayed.

$$\frac{dQ_G}{dt} = 0.75\ [Q_F + Q_I(f_b D_3 + f_d D_4 + f_f D_5)]$$

$$+ 0.75\ [\lambda_g^b f_g D_5 Q_I - \lambda_g Q_g]$$

Let $G_1 = Q_F + Q_I(f_b D_3 + f_d D_4 + f_f D_5)$, constant input

$G_2 = \lambda_g^b f_g D_5 Q_I$, variable input

$$\bar{Q}_G = 0.75 \left(\frac{G_1}{S} + \frac{G_2}{S(S+\lambda_g)} \right)$$

$$Q_G = 0.75 \left[G_1 + G_2 \left(\frac{1-\exp(-\lambda_g t)}{\lambda_g} \right) \right] \qquad (13)$$

$$D_G = \frac{0.75\ R_G}{2}\left[G_1 t + \frac{G_2}{\lambda_g}\left(t - \frac{1-\exp(-\lambda_g t)}{\lambda_g}\right)\right] \qquad (14)$$

$$C_G = \frac{0.75\ R_G}{2}\left[G_1 t + \frac{G_2}{\lambda_g}\left(t - \frac{1-\exp(-\lambda_g t)}{\lambda_g}\exp(-\lambda_g(T-t))\right)\right] \qquad (15)$$

ORGANS RECEIVING RADIONUCLIDE FROM BLOOD

$$dQ_n/dt = f'_{2n} r_B - \lambda_n Q_n \qquad (16)$$

where r_B is the rate at which the radionuclide enters blood.

$$r_B = \underline{\lambda_a^b Q_a} + \underline{\lambda_c^b Q_c} + \lambda_e^b Q_e + f_1(\underline{\lambda_b^b Q_b} + \underline{\lambda_d^b Q_d} + \underline{\lambda_f^b Q_f} + \lambda_g^b Q_g)$$

$$+ \lambda_i^b Q_{Li}$$

The underlined components ($T_k > 1$ day) are assumed to be in equilibrium and therefore constant.

$$S\bar{r}_B = K + \lambda_e^b \bar{Q}_e + f_1 \lambda_g^b \bar{Q}_g + \lambda_i^b \bar{Q}_{Li}$$

where $K = f_a D_3 + f_c D_4 + f_1(f_b D_3 + f_d D_4 + f_f D_5)$

Substituting $\lambda_h^b = \lambda_e^b = \lambda_g^b$ and combining terms,

$$\bar{r}_B = \lambda_h^b D_5 Q_I \left(\frac{f_e + f_1 f_g}{S(S+\lambda_h)} + \frac{f_h f_i \lambda_i^b}{S(S+\lambda_h)(S+\lambda_i)}\right) + \frac{K}{S} \qquad (17)$$

From Eq. (16)

$$S\bar{Q}_n + \lambda_n \bar{Q}_n = f'_{2n}\ \bar{r}_B;\ \bar{Q}_n = f'_{2n}\ \bar{r}_B/(S+\lambda_n) \qquad (18)$$

Substituting Eq. (17) in Eq. (18) and rearranging terms,

$$\bar{Q}_n = f'_{2n} Q_I \left[\frac{K}{S(S+\lambda_n)} + \frac{L}{S(S+\lambda_h)(S+\lambda_n)} + \frac{M}{S(S+\lambda_h)(S+\lambda_i)(S+\lambda_n)}\right] \qquad (19)$$

where,

$$K = f_a D_3 + f_c D_4 + f_1(f_b D_3 + f_d D_4 + f_f D_5)$$

$$L = \lambda_h^b D_5 (f_e + f_1 f_g)$$

$$M = \lambda_h^b \lambda_i^b f_h f_i D_5$$

Expansion of Eq. (19) letting $S = 0, -\lambda_h, -\lambda_i, -\lambda_n$, gives

$$\begin{aligned} Q_n = f'_{2n} Q_I \Bigg[& \left\{\frac{K}{\lambda_n} + \frac{L}{\lambda_h \lambda_n} + \frac{M}{\lambda_h \lambda_i \lambda_n}\right\} \\ & + \left\{-\frac{L}{\lambda_h(\lambda_n-\lambda_h)} - \frac{M}{\lambda_h(\lambda_i-\lambda_h)(\lambda_n-\lambda_h)}\right\}\exp(-\lambda_h t) \\ & + \left\{-\frac{M}{\lambda_i(\lambda_h-\lambda_i)(\lambda_n-\lambda_i)}\right\}\exp(-\lambda_i t) \\ & + \left\{-\frac{K}{\lambda_n} - \frac{L}{\lambda_n(\lambda_h-\lambda_n)} - \frac{M}{\lambda_n(\lambda_h-\lambda_n)(\lambda_i-\lambda_n)}\right\}\exp(-\lambda_n t)\Bigg] \end{aligned} \qquad (20)$$

The concise forms for the burden, dose, and dose commitment equations are:

$$\begin{aligned} Q_n = {} & f'_{2n} Q_I [A_o + A_1 \exp(-\lambda_h t) + A_2 \exp(-\lambda_i t) \\ & + A_3 \exp(-\lambda_n t)] \end{aligned} \qquad (21)$$

where, A_0, A_1, A_2, A_3 are the bracketed expressions in Equation (20).

$$D_n = R_n f'_{2n} Q_I \left[A_o t + A_1 \frac{1-\exp(-\lambda_h t)}{\lambda_h} + A_2 \frac{1-\exp(-\lambda_i t)}{\lambda_i} + A_3 \frac{1-\exp(-\lambda_n t)}{\lambda_n} \right] \tag{22}$$

$$C_n = R_n f'_{2n} Q_I \left[A_o t + A_1 \left(\frac{1-\exp(-\lambda_h t)}{\lambda_h} \exp(-\lambda_h (T-t)) \right) + A_2 \left(\frac{1-\exp(-\lambda_i t)}{\lambda_i} \exp(\lambda_i (T-t)) \right) + A_3 \left(\frac{1-\exp(-\lambda_n t)}{\lambda_n} \exp(\lambda_n (T-t)) \right) \right] \tag{23}$$

where A_o, A_1, A_2, and A_3 are the constants calculated as shown in Eq. (20).

Equations (21), (22), and (23) give the burden, dose, and dose commitment to organ n due to inhalation of a Class Y compound such as $^{239}PuO_2$. Equations (24), (25), and (26) give the corresponding values due to ingestion of the radionuclide.

$$Q_{nF} = (f_1 f'_{2n} Q_F / \lambda_n)(1-\exp(-\lambda_n t)) \tag{24}$$

$$D_{nF} = R_n \frac{f_1 f'_{2n} Q_F}{\lambda_n} \left(t - \frac{1-\exp(-\lambda_n t)}{\lambda_n} \right) \tag{25}$$

$$C_n F = R_n \frac{f_1 f'_{2n} Q_F}{\lambda_n} \left(t - \frac{1-\exp(-\lambda_n t)}{\lambda_n} \exp(-\lambda (T-t)) \right) \tag{26}$$

RESULTS

Sample results based on these equations are given in Tables A.1 to A.3.

Table A.1. INHALATION: 1.0 pCi/day ^{239}Pu, AMAD = 0.5 μm
(Burden = pCi; Dose & Commitment = rem)

Time, years	LUNGS Burden	LUNGS Dose	LUNGS Commitment	LYMPH Burden	LYMPH Dose	LYMPH Commitment
1	5.3457 E+1	5.7574 E-2	2.6620 E-1	3.3820 E+0	7.4591 E-2	1.1545 E+1
2	8.5577 E+1	1.9800 E-1	5.3241 E-1	1.0780 E+1	5.3759 E-1	2.2991 E+1
3	1.0494 E+2	3.8837 E-1	7.9861 E-1	1.9551 E+1	1.5300 E+0	3.4310 E+1
4	1.1662 E+2	6.0886 E-1	1.0648 E+0	2.8337 E+1	3.1203 E+0	4.5531 E+1
5	1.2366 E+2	8.4750 E-1	1.3310 E+0	3.6501 E+1	5.2577 E+0	5.6624 E+1
10	1.3349 E+2	2.1400 E+0	2.6620 E+0	6.4771 E+1	2.2515 E+1	1.1045 E+2
15	1.3427 E+2	3.4680 E+0	3.9931 E+0	7.9785 E+1	4.6600 E+1	1.6149 E+2
20	1.3434 E+2	4.7988 E+0	5.3241 E+0	9.0185 E+1	7.4726 E+1	2.0972 E+2
25	1.3434 E+2	6.1298 E+0	6.6551 E+0	9.9213 E+1	1.0602 E+2	2.5516 E+2
30	1.3434 E+2	7.4608 E+0	7.9861 E+0	1.0785 E+2	1.4022 E+2	2.9780 E+2
35	1.3434 E+2	8.7918 E+0	9.3172 E+0	1.1637 E+2	1.7724 E+2	3.3763 E+2
40	1.3434 E+2	1.0123 E+1	1.0648 E+1	1.2486 E+2	2.1708 E+2	3.7466 E+2
45	1.3434 E+2	1.1454 E+1	1.1979 E+1	1.3334 E+2	2.5972 E+2	4.0885 E+2
50	1.3434 E+2	1.2785 E+1	1.3310 E+1	1.4181 E+2	3.0515 E+2	4.4015 E+2
55	1.3434 E+2	1.4116 E+1	1.4641 E+1	1.5029 E+2	3.5339 E+2	4.6828 E+2
60	1.3434 E+2	1.5447 E+1	1.5969 E+1	1.5876 E+2	4.0442 E+2	4.9236 E+2
65	1.3434 E+2	1.6778 E+1	1.7261 E+1	1.6723 E+2	4.5825 E+2	5.0962 E+2
70	1.3434 E+2	1.8109 E+1	1.8109 E+1	1.7570 E+2	5.1488 E+2	5.1488 E+2

Time, years	GASTROINTESTINAL TRACT* Burden	GASTROINTESTINAL TRACT* Dose	GASTROINTESTINAL TRACT* Commitment	GASTROINTESTINAL TRACT** Burden	GASTROINTESTINAL TRACT** Dose	GASTROINTESTINAL TRACT** Commitment
1	1.0359 E+0	3.3016 E-5	3.5381 E-5	2.8585 E-1	8.7144 E-6	1.1079 E-5
2	1.0581 E+0	6.6972 E-5	7.0762 E-5	3.0812 E-1	1.8368 E-5	2.2158 E-5
3	1.0715 E+0	1.0149 E-4	1.0614 E-4	3.2154 E-1	2.8587 E-5	3.3237 E-5
4	1.0796 E+0	1.3636 E-4	1.4152 E-4	3.2963 E-1	3.9148 E-5	4.4316 E-5
5	1.0845 E+0	1.7143 E-4	1.7691 E-4	3.3451 E-1	4.9915 E-5	5.5396 E-5
10	1.0913 E+0	3.4789 E-4	3.5381 E-4	3.4133 E-1	1.0487 E-4	1.1079 E-4
15	1.0919 E+0	5.2476 E-4	5.3072 E-4	3.4187 E-1	1.6023 E-4	1.6619 E-4
20	1.0919 E+0	7.0167 E-4	7.0762 E-4	3.4192 E-1	2.1563 E-4	2.2158 E-4
25	1.0919 E+0	8.7857 E-4	8.8453 E-4	3.4192 E-1	2.7102 E-4	2.7698 E-4
30	1.0919 E+0	1.0555 E-3	1.0614 E-3	3.4192 E-1	3.2642 E-4	3.3237 E-4
35	1.0919 E+0	1.2324 E-3	1.2383 E-3	3.4192 E-1	3.8181 E-4	3.8777 E-4
40	1.0919 E+0	1.4093 E-3	1.4152 E-3	3.4192 E-1	4.3721 E-4	4.4316 E-4
45	1.0919 E+0	1.5862 E-3	1.5921 E-3	3.4192 E-1	4.9261 E-4	4.9856 E-4
50	1.0919 E+0	1.7631 E-3	1.7691 E-3	3.4192 E-1	5.4800 E-4	5.5396 E-4
55	1.0919 E+0	1.9400 E-3	1.9460 E-3	3.4192 E-1	6.0340 E-4	6.0935 E-4
60	1.0919 E+0	2.1169 E-3	2.1228 E-3	3.4192 E-1	6.5879 E-4	6.6471 E-4
65	1.0919 E+0	2.2938 E-3	2.2993 E-3	3.4192 E-1	7.1419 E-4	7.1967 E-4
70	1.0919 E+0	2.4707 E-3	2.4707 E-3	3.4192 E-1	7.6958 E-4	7.6958 E-4

*Includes ingestion of 1.0 pCi/day.

**Inhalation only.

Table A.2. INHALATION: 1.0 pCi/day of ^{239}Pu; AMAD = 0.5 μm
(Burden in pCi; Dose & Commitment in rem)

Time,	BONE			LIVER		
years	Burden	Dose	Commitment	Burden	Dose	Commitment
1	1.2764 E+0	3.7907 E-4	3.7782 E-1	1.2710 E+0	3.0544 E-4	2.2751 E-1
2	3.9058 E+0	2.1672 E-3	7.5101 E-1	3.8748 E+0	1.7415 E-3	4.5309 E-1
3	7.8075 E+0	6.3179 E-3	1.1195 E+0	7.7177 E+0	5.0631 E-3	6.7673 E-1
4	1.2825 E+1	1.3695 E-2	1.4834 E+0	1.2631 E+1	1.0945 E-2	8.9837 E-1
5	1.8780 E+1	2.5038 E-2	1.8425 E+0	1.8427 E+1	1.9958 E-2	1.1180 E+0
10	5.7261 E+1	1.5887 E-1	3.5660 E+0	5.5075 E+1	1.2488 E-1	2.1845 E+0
15	1.0160 E+2	4.4459 E-1	5.1663 E+0	9.5638 E+1	3.4443 E-1	3.1950 E+0
20	1.4658 E+2	8.9212 E-1	6.6392 E+0	1.3491 E+2	6.8090 E-1	4.1442 E+0
25	1.9063 E+2	1.5003 E+0	7.9800 E+0	1.7151 E+2	1.1281 E+0	5.0267 E+0
30	2.3335 E+2	2.2650 E+0	9.1841 E+0	2.0524 E+2	1.6777 E+0	5.8364 E+0
35	2.7467 E+2	3.1812 E+0	1.0247 E+1	2.3623 E+2	2.3216 E+0	6.5668 E+0
40	3.1460 E+2	4.2439 E+0	1.1163 E+1	2.6465 E+2	3.0519 E+0	7.2107 E+0
45	3.5317 E+2	5.4480 E+0	1.1928 E+1	2.9072 E+2	3.8617 E+0	7.7603 E+0
50	3.9042 E+2	6.7888 E+0	1.2536 E+1	3.1462 E+2	4.7442 E+0	8.2075 E+0
55	4.2641 E+2	8.2616 E+0	1.2983 E+1	3.3655 E+2	5.6934 E+0	8.5439 E+0
60	4.6117 E+2	9.8620 E+0	1.3269 E+1	3.5665 E+2	6.7038 E+0	8.7635 E+0
65	4.9475 E+2	1.1585 E+1	1.3403 E+1	3.7508 E+2	7.7704 E+0	8.8684 E+0
70	5.2718 E+2	1.3428 E+1	1.3428 E+1	3.9199 E+2	8.8884 E+0	8.8884 E+0

Time,	KIDNEY			TOTAL BODY		
years	Burden	Dose	Commitment	Burden	Dose	Commitment
1	5.6706 E-2	7.7141 E-5	7.4698 E-2	2.8399 E+0	1.6549 E-5	1.8057 E-2
2	1.7346 E-1	4.4093 E-4	1.4851 E-1	8.6993 E+0	9.4695 E-5	3.5869 E-2
3	3.4662 E-1	1.2851 E-3	2.2143 E-1	1.7408 E+1	2.7627 E-4	5.3436 E-2
4	5.6918 E-1	2.7848 E-3	2.9346 E-1	2.8625 E+1	5.9932 E-4	7.0757 E-2
5	8.3316 E-1	5.0902 E-3	3.6457 E-1	4.1961 E+1	1.0966 E-3	8.7830 E-2
10	2.5356 E+0	3.2256 E-2	7.0630 E-1	1.2869 E+2	6.9863 E-3	1.6945 E-1
15	4.4899 E+0	9.0141 E-2	1.0243 E+0	2.2980 E+2	1.9635 E-2	2.4473 E-1
20	6.4633 E+0	1.8062 E-1	1.3175 E+0	3.3373 E+2	3.9573 E-2	3.1355 E-1
25	8.3873 E+0	3.0332 E-1	1.5851 E+0	4.3700 E+2	6.6853 E-2	3.7578 E-1
30	1.0244 E+1	4.5724 E-1	1.8259 E+0	5.3863 E+2	1.0139 E-1	4.3129 E-1
35	1.2032 E+1	6.4126 E-1	2.0388 E+0	6.3839 E+2	1.4305 E-1	4.7994 E-1
40	1.3750 E+1	8.5422 E-1	2.2228 E+0	7.3622 E+2	1.9170 E-1	5.2160 E-1
45	1.5403 E+1	1.0950 E+0	2.3768 E+0	8.3215 E+2	2.4721 E-1	5.5613 E-1
50	1.6991 E+1	1.3625 E+0	2.4995 E+0	9.2621 E+2	3.0944 E-1	5.8341 E-1
55	1.8517 E+1	1.6558 E+0	2.5899 E+0	1.0184 E+3	3.7826 E-1	6.0335 E-1
60	1.9985 E+1	1.9738 E+0	2.6478 E+0	1.1088 E+3	4.5354 E-1	6.1600 E-1
65	2.1396 E+1	2.3155 E+0	2.6750 E+0	1.1975 E+3	5.3516 E-1	6.2189 E-1
70	2.2751 E+1	2.6801 E+0	2.6801 E+0	1.2844 E+3	6.2299 E-1	6.2299 E-1

Table A.3. INGESTION: 1 pCi/day of ^{239}Pu
(Burden in pCi; Dose & Commitment in rem)

Time,	BONE			LIVER		
years	Burden	Dose	Commitment	Burden	Dose	Commitment
1	4.9105 E-3	1.7724 E-6	1.9596 E-4	4.8851 E-3	1.4276 E-6	1.1602 E-4
2	9.7870 E-3	7.0732 E-6	3.8971 E-4	9.6862 E-3	5.6778 E-6	2.3118 E-4
3	1.4630 E-2	1.5878 E-5	5.8124 E-4	1.4405 E-2	1.2702 E-5	3.4545 E-4
4	1.9439 E-2	2.8163 E-5	7.7055 E-4	1.9042 E-2	2.2453 E-5	4.5883 E-4
5	2.4215 E-2	4.3903 E-5	9.5760 E-4	2.3600 E-2	3.4883 E-5	5.7128 E-4
10	4.7606 E-2	1.7361 E-4	1.8586 E-3	4.5242 E-2	1.3564 E-4	1.1193 E-3
15	7.0200 E-2	3.8621 E-4	2.7010 E-3	6.5087 E-2	2.9681 E-4	1.6419 E-3
20	9.2024 E-2	6.7886 E-4	3.4827 E-3	8.3286 E-2	5.1338 E-4	2.1368 E-3
25	1.1310 E-1	1.0489 E-3	4.2016 E-3	9.9974 E-2	7.8074 E-4	2.6015 E-3
30	1.3347 E-1	1.4935 E-3	4.8554 E-3	1.1528 E-1	1.0947 E-3	3.0332 E-3
35	1.5314 E-1	2.0104 E-3	5.4420 E-3	1.2931 E-1	1.4513 E-3	3.4290 E-3
40	1.7214 E-1	2.5969 E-3	5.9588 E-3	1.4218 E-1	1.8472 E-3	3.7857 E-3
45	1.9049 E-1	3.2508 E-3	6.4035 E-3	1.5398 E-1	2.2789 E-3	4.0997 E-3
50	2.0821 E-1	3.9697 E-3	6.7735 E-3	1.6480 E-1	2.7436 E-3	4.3670 E-3
55	2.2534 E-1	4.7514 E-3	7.0662 E-3	1.7472 E-1	3.2385 E-3	4.5836 E-3
60	2.4188 E-1	5.5938 E-3	7.2787 E-3	1.8382 E-1	3.7611 E-3	4.7447 E-3
65	2.5785 E-1	6.4948 E-3	7.4085 E-3	1.9216 E-1	4.3091 E-3	4.8455 E-3
70	2.7329 E-1	7.4524 E-3	7.4524 E-3	1.9982 E-1	4.8804 E-3	4.8804 E-3

Time,	KIDNEY			TOTAL BODY		
years	Burden	Dose	Commitment	Burden	Dose	Commitment
1	2.1814 E-4	3.6068 E-7	3.8673 E-5	1.0928 E-2	7.7390 E-8	9.4185 E-6
2	4.3456 E-4	1.4385 E-6	7.6928 E-5	2.1814 E-2	3.0915 E-7	1.8719 E-5
3	6.4927 E-4	3.2291 E-6	1.1476 E-4	3.2656 E-2	6.9468 E-7	2.7901 E-5
4	8.6229 E-4	5.7256 E-6	1.5217 E-4	4.3456 E-2	1.2334 E-6	3.6964 E-5
5	1.0736 E-3	8.9228 E-6	1.8914 E-4	5.4212 E-2	1.9246 E-6	4.5908 E-5
10	2.1057 E-3	3.5229 E-5	3.6747 E-4	1.0736 E-1	7.6482 E-6	8.8819 E-5
15	3.0977 E-3	7.8244 E-5	5.3454 E-4	1.5948 E-1	1.7096 E-5	1.2867 E-4
20	4.0513 E-3	1.3732 E-4	6.8990 E-4	2.1057 E-1	3.0196 E-5	1.6541 E-4
25	4.9679 E-3	2.1184 E-4	8.3309 E-4	2.6066 E-1	4.6876 E-5	1.9897 E-4
30	5.8490 E-3	3.0119 E-4	9.6360 E-4	3.0977 E-1	6.7066 E-5	2.2928 E-4
35	6.6959 E-3	4.0482 E-4	1.0809 E-3	3.5792 E-1	9.0698 E-5	2.5629 E-4
40	7.5100 E-3	5.2215 E-4	1.1846 E-3	4.0513 E-1	1.1770 E-4	2.7992 E-4
45	8.2926 E-3	6.5266 E-4	1.2739 E-3	4.5141 E-1	1.4802 E-4	3.0011 E-4
50	9.0448 E-3	7.9585 E-4	1.3484 E-3	4.9679 E-1	1.8157 E-4	3.1679 E-4
55	9.7678 E-3	9.5122 E-4	1.4075 E-3	5.4128 E-1	2.1831 E-4	3.2989 E-4
60	1.0463 E-2	1.1183 E-3	1.4505 E-3	5.8490 E-1	2.5817 E-4	3.3934 E-4
65	1.1131 E-2	1.2966 E-3	1.4768 E-3	6.2766 E-1	3.0108 E-4	3.4506 E-4
70	1.1773 E-2	1.4858 E-3	1.4858 E-3	6.6959 E-1	3.4698 E-4	3.4698 E-4

APPENDIX A

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