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A SURVEY OF PLUTONIUM CONCENTRATION VARIABILITY IN SOIL SAMPLES FROM VARIOUS LOCATIONS By T.E. HARONSON

omedical and Environmental Research Program

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Sector	Area (na)	17/2							/
Arctic Gas	6	12	10	10	7	1	1	2 /	5
North	9	1	, 8	5	5	• 0	2	2/	1
West	20	4	/ 5	5	4	5	4	Æ	2
Southwest	25	3 /	5	1 \	3	2	3	2	1
Southeast	15	3/	5	2	2	5	5	2	2
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TABLE I

NESTING BIRDS IN VARIOUS SECTORS OF THE PRUDHOE BAY STUDY AREA

comparison with current research results; this will not only extend the data base but will also facilitate predictions about long-term effects and recovery of tundra ecosystem components from petroleum resource developments.

The 1976 nesting season idr tundra birds was considerably less successful than during 1975 because of inclement weather that maintained a lingering snow cover and large expanses of standing water in low-center polygon and Carex meadow habitats and a high degree of predation. Numbers of nests of semipalmated sandpipers and Lapland longspurs found on the 82-ha study frea at Prudhoe Bay are shown in Table I to illustrate the shifting of these species in occupation of available nesting habitat and increased nesting during the 1973 optimum conditions. A total of 92 nesting birds of 4 species were banded during the 1976 season, and 12 birds banded in previous years were recaptured, providing new data on nest site and pairing fidelity.

Small-mammal population studies were again conducted by live-trapping within 17 grids of 0.116 ha each within the Alaskan arctic gas study area and its periphery. A population density of 7.1 collared (or varying) lemmings per ha was estimated from 8200 trap-nights of effort during July and August; this was a decrease from the 19.2 lemmings per ha estimated last year. Greater mobility of the lemmings, particularly adult males, was noted from observations of tagged individuals that repeatedly crossed pipeline berms and made journeys of 300 to 500 m straight-line between points of capture.

Study areas were established during early May near Franklin Bluffs, 50 km south of Prudhoe Bay, and near Happy Valley, 100 km south of Prudhoe Bay, for purposes of determining the impact of the installation and operation of/the trans-Alaska pipeline and haul road upon smalf mammals and tundranesting birds. The Happy Valley site was abandoned on May 29 after repeated damage to the personnel tent by a grizzly bear that had been pauperized by pipeline construction activities. The Franklin Bluffs study area consisted of a 400-x 2500-m gridded bird study plot and 32 small-mammal trapping grids of 0.146 ha each, situated in such a manner to measure changes in/animal populations that would be affected by pipeline and road activities, if such occur. The inclement weather conditions that prevailed over most of the North Slope were apparently responsible for the very low nesting densities of the bird species (13 nests/100 ha) compared to those at Prudhoe Bay (60 nests/100 ha), although the Franklin Bluffs site contained a greater diversity of species. Forty-three birds representing 6 species were banded and coldr-marked for continuing studies of population dynamics of those species.

Live-trapping revealed that the collared (or varying) lemming, <u>Dicrostonyk groenlandicus</u>, was the most abundant small mammal in the Franklin Bluffs habitats, and a population density of about 4.3 animals/ha was estimated from tagging and recapturing the animals. Next in importance was the tundra vole, <u>Microtus oeconomus</u>, followed by the singing vole, <u>Microtus miurus</u>, and the brown lemming, Lemmus trimucronatus.

A Survey of Plutonium Concentration Variability in Soil Samples from Various Locations

[T. E. Hakonson]

The large variability associated with environmental plutonium data is currently recognized as a major problem in designing field studies of this element. The large sample sizes required for acceptable statistical control of an experiment place severe restrictions on the kinds of research questions that can be addressed within the limits of time and money.

The purpose of this review is to summarize some of the available data on plutonium concentration variability in terrestrial soil components and to identify potential sources of this variability, to serve as guidance in designing studies that are both efficient and effective in achieving desired goals. Plutonium data from 7 geographical regions representing 15 terrestrial study sites were selected for review based on the availability of data, source of study area plutonium, sampling methodology, and regional climate. The intent in selecting specific study areas was to present plutonium concentration variability estimates from a diverse array of study-related factors. Description of Some Environmental Plutonium Study Areas.--The 7 sites chosen to represent the varied conditions under which plutonium can exist in the environment are listed in Table I. Sources of plutonium in these areas include industrial liquid effluents (Los Alamos), accidental releases from industrial and military sources (Rocky Flats; Thule, Greenland), fallout from both single and multiple weapons tests (Trinity Site; Glenn and Janet Islands, Eniwetok Atoll), and nonfission explosive tests with plutonium devices (Nevada Test Site).

<u>Plutonium Concentration Variability in Study</u> <u>Area Soils</u>.--The variability in soil plutonium concentrations, expressed as the coefficient of variation (CV, standard deviation/mean), ranged from 0.21 to 3.2, while plutonium concentrations varied from 0.15 to 460 000 pCi/g (Table II). The pattern was similar in whole soil samples to various depths in

TABLE I

SOME CHARACTERISTICS OF ENVIRONMENTAL PLUTONIUM STUDY AREAS

Location	Source of Plutonium	Mean Annual Precipitation (cm)	Reference
	•		
LOS ATAMOS			
Mortandad Canyon	Industrial liquid effluent	46	1-3
DP-Los Alamos Canyon		46	1-3
Acid-Pueblo Canyon		46	1-3
Trinity Site	Single weapons test		
Ground Zero	1 km from Ground Zero	15	4,5
Area 21	44 km from Ground Zero	20	4.5
Eniwetok Atoll	Weapons test		
Janet	Multiple ground zeros	145	6
Glenn	Multiple fallout	145	6
Rocky Flats	Unintentional		
Macroplot 1	Release from leaking drums	40	7,3
Macroplot 2		40	7,8
<u>Nevada Test Site</u>			
Area 13	Safety test shots		
Strata 1		8	9,10
Strata 6		8	9,10
Area 5 (CMX)			
Strata 1		8	9,10
Strata 4		8	9,10
Thule, Greenland	Aircraft accident	13	11

		0 to 2.5 cm			To Depth	
		mean		n	mean	
L <u>ocation</u>	n	(pCi/g)	CV	(depth, cm)) (pCi/g)	CV
Los Alamos						
Mortandad Canyon	5	140	0.52	15 (30)	90	0.79
DP-Los Alamos Canyon	6	0.42	0.63	21 (30)	0.73	1.6
Acid-Pueblo Canyon	7	10	0.48	21 (30)	21	1.7
Trinity Site						
Ground Zero	8	0.44	0.82	8 (25)	0.07	0.68
Area 21	8	2	0.48	8 (33)	0.14	0.68
Eniwetok Atoll						
Janet	12	18	1.9	138 (15)	16	1.3
Glenn	3	0.15	0.21	29 (15)	0.11	0.62
<u>Nevada Test Site</u>						
Area 13						
Strata 1	4	880	1.2	39 (5)	36	1.4
Strata 6	3	460 000	0.67	47 (5)	14 000	3.1
Area 5						
Strata l	5	2 200	1.5	41 (5)	59	1.4
Strata 4	2	150 000	0.8	23 (5)	7 300	1.1
Thule, Greenland				6 (5)	0.16	1.5
<u>Rocky Flats</u>						
Macroplot 2	12	116 ^a	3.2			

TABLE II

PLUTONIUM CONCENTRATION AND VARIABILITY ESTIMATES IN SOME STUDY AREA SOILS

^aConcentration in \leq 45-µm size fraction to depth of 3 cm.

that the range in CVs (0.62 to 3.1) was relatively small compared to the range in plutonium concentrations (0.11 to 14 000 pCi/g).

There were no significant linear relationships between the magnitude of the CV and the corresponding plutonium concentration within the ranges of the data. Generally lower variability was associated with fallout (Trinity Site) and liquid effluent (Los Alamos) sources of plutonium (Table III), while higher CVs were associated with safety shot (NTS) and Ground Zero areas (Trinity, Janet). This pattern seems reasonable based on suspected physical forms of the plutonium in the respective study areas. However, other site-related factors such as study area climate (and its effects on plutonium weathering), topography, and sampling methodology contribute to the total variability and confound any attempts to TABLE III

PLUTONIUM CONCENTRATION COEFFICIENT OF VARIATION IN SURFACE SOILS

Location	Plutonium Source	<u>cv</u>
Glenn	Fallout	0.21
Trinity, Aeea 21	Fallout	0.48
Mortandad	Liquid effluent	0.52
DP-Los Alamos	Liquid effluent	0 .63
NTS-A-13, Strata 6	Safety shot	0 .67
NTS-A-5, Strata 4	Safety shot	0.80
Trinity, GZ	Ground zero	0.82
Acid-Pueblo	Liquid effluent	0.87
NTS-A-13, Strata 1	Safety shot	1.2
NTS-A-5, Strata 1	Safety shot	1.5
Thule, Greenland	Aircraft accident	1.5
Janet	Multiple ground zeros	1.9
Rocky Flats		
Macroplot 2	Industrial accident	3.2

TABLE IV

SOIL SAMPLING AND ANALYTICAL METHODOLOGY AT VARIOUS STUDY SITES

Location	Sampling Technique	Sampling Area (cm ²)	Sample Pretreatment	Amount Analyzed (g)
Los Alamos	Core	4.5	None	Whole sample ~ 25
Trinity	Core	4.5	None	Whole sample ~ 25
Eniwetok	Core	30	Ball-milled	10-50
Nevada Test Site	Core-excavation	100-127	Ball-milled	10
Thule, Greenland	Excavation	100	Remove > 0.6 cm 5 samples composited	~ 100
Rocky Flats	Excavation	25	Remove > 0.5 cm	5-10

TABLE V

SPATIAL RELATIONSHIPS OF 238 Pu CONCENTRATION VARIABILITY IN MORTANDAD CANYON SOILS

		Within St	rata	Within Plot		
	<u>Overall</u>	0 to 160	160 to 2560	<u>0 to 160</u>	160 to 2560	
Coefficient of variation	1.2	0.79	0.94	0.75 (0.31)	0.85 (0.60)	
n	27	15	12	5	4	

relate variability to any specific factor. The difficulties in comparing soil plutonium data between sites can be appreciated by examining some of the potential components of variability at the various sites.

Soil sampling and analytical methodology employed at several sites are summarized in Table IV to emphasize the potential for these factors as components of the total variability. Although chemical procedures for plutonium analysis between sites are similar in that they involve HNO₃-HF digestion, followed by column separation and alpha spectrometry, considerable differences exist in sampling techniques, sample preparation, and in the size of soil aliquot analyzed for plutonium.

One of the sources of field sampling variability for soil plutonium is the heterogeneous distribution of the element within the study areas. An example of the effect of spatial heterogeneity on variability is given in Table V for Mortandad Canyon at Los Alamos where a plutonium concentration gradient exists with distance down the canyon. The CVs in Table V were calculated for all samples (n = 27) collected within a 2560-m segment of stream channel, for two strata (0 to 160 and 160 to 2560 m) within that segment, and for triplicate samples taken at each of 9 plots within that segment. The variability was considerably reduced by examining the data from smaller spatial units as achieved through stratification. The concentration CV was calculated as 1.2 when all the data from the contaminated portion of the canyon were used, whereas the data from two segments of stream channel reduced the CV to 0.79 and 0.94 for the respective strata. Within-plot variability based on triplicate samples from each sampling location within the 2560m segment averaged 0.75 and 0.85 for the two strata, indicating that closely spaced replicate samples did not greatly reduce the variability as measured over much larger areas as represented by the two strata.

The relationships between soil plutonium concentration variability, soil particle size fraction, and soil depth were investigated at those sites where data were available (i.e., Los Alamos and Rocky Flats) to examine further the components of plutonium concentration variability. The data in Table VI illustrate the relationship of the CV with soil particle size fraction and soil depth for soils from Mortandad Canyon at Los Alamos. There were no significant relationships between CV and soil particle size fractions in either study area as inferred by analysis of variance. However, in all cases, there was a significant change ($\rho \leq 0.05$) in CV with soil depth.

TABLE VI

RELATIONSHIP OF PLUTONIUM CONCENTRATION VARIABILITY (COEFFICIENT OF VARIATION) WITH SOIL PARTICLE SIZE FRACTION AND SOIL DEPTH IN MORTANDAD CANYON

	Soil Particle Size Fraction									
Depth Profile (cm)	(< 53 µm)	(53 to 105 µm)	(105 to 500 µm)	(500 to 1000 µm)	(1 to 2 mm)	(2 to 23 mm)				
0-2.5	0.84	0.49	0.79	0.65	0.64	0.48				
2.5-7.5	0.58	0.54	0.62	0.58	0.93	0.62				
7.5-12.5	0.38	0.49	0.44	0.65	0.46	0.55				
12.5-22	0.44	0.35	0.13	0.11	0.16	0.50				

All available data were examined using linear least-squares techniques to determine the significance of plutonium concentration vs depth relationships (Table VII). Only four comparisons were significant ($\rho \leq 0.05$): those from Glenn, Trinity Site (Ground Zero and Area 21), and macroplot 1 at Rocky Flats. Regression slopes for the first three areas were positive, whereas the slope was negative for the Rocky Flats area. It would appear that a clear definition of such relationships would be instrumental in efficient study design and would provide insight as to the mechanisms of soil plutonium mobility.

<u>Conclusions</u>.--Plutonium concentration variability in soils from 7 geographical regions ranged from 0.48 to 3.2. The CV was relatively consistent, considering that plutonium concentrations varied through 6 orders of magnitude and appeared to be

TABLE VII

LINEAR REGRESSION OF SOIL COEFFICIENT OF VARIATION WITH DEPTH

Location	a 	a ₁	<u>r</u> ²
Glenn	0.38	0.02	0.59 ^ª
Trinity Ground Zero	0.93	0.05	0.83 ^a
Trinity Area 21	0.75	0.04	0.82 ^ª
NTS-A-13, Strata 1	1.8	-0.02	0.17
NTS-A-13, Strata 6	0.63	0.02	0.16
NTS-A-5, Strata 1	0.98	0.01	0.06
NTS-A-5, Strata 4	1.1	-0.02	0.28
Janet	1.7	0.02	0.27
Rocky Flats			
Macroplot 1	1.6	-0.06	0.53 ^a
Macroplot 2	2.2	-0.05	0.22

Equation of the form $y = a + a_1 x$, where y = CVnd x = soil depth (cm). independent of the magnitude of plutonium concentration within the ranges of observed data. Vertical and horizontal inhomogeneity in plutonium contamination within study areas comprises a part of the total observed variability, while soil particle size fractions do not appear to contribute significantly to the overall variability at those sites examined.

The relationship between UV and soil depth was statistically significant in the study areas at Trinity and Glenn and at macroplot 1 at Rocky Flats. The CV increased with depth at the Trinity Site and Glenn study areas and decreased with depth in macroplot 1 at Rocky Flats.

Lower CV values were generally associated with fallout and effluent sources of plutonium. However, the diversity of environments and "age" of the plutonium at the various study sites likely contribute to overall variability and certainly complicate interpretation of data between sites.

Highest CVs were associated with accidental releases of plutonium. This type of release would be the most likely source of plutonium to presently uncontaminated ecosystems.

Differences in methodologies at the various study sites make comparison of the data difficult and emphasize the need for coordination of effort between sites to improve the utility and comparability of the data.

Studies should be designed to look specifically at the components of overall variability. Data from Rocky Flats and Los Alamos indicate that the analytical component of overall variability contributes less than 35% to the total. Other factors which might be considered include variability in mass of soil particle size fractions within study plots and the relationship of plutonium CV to the variability in other soil physical-chemical properties.

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Rainout Collateral Damage Study

[S. Barr and J. D. Klett]

<u>Precipitation Morphology</u>.--Studies of precipitation structure in the atmosphere have been initiated in connection with an assessment of collateral damage due to precipitation scavenging of nuclear weapon debtis. The insights gained from these studies and the computational model developed as the vehicle for expressing these insights promise to be valuable additions to the assessment technology in a wide variety of practical problems involving precipitation effects.

The fundamental premise is that precipitation exhibits a high degree of variability in time and space from less than 60 sec and 100 m out to the scales of large-scale synoptic weather systems. In applications which depend on the interaction of several physical processes, each of which have small-scale variations, the preliminary smoothing of one process prior to estimating its covariant interactions with the other processes can lead to errors \in the result. For example, the scavenging effects on pollutant clouds which have dimensions of less than a few kilometers depend on the interaction of \that pollutant with a precipitation cell. If the precipitation field also has variability on the same scale, it is incorrect to assume that the field is uniform. The data bases typically available for assessment projects have a spacing between observation points of 10 to 100 km. In problems that are driven by higher resolution effects (e.g., some scavenging problems, drainage basin runoff, air-frame damage, electromagnetic energy transmission), a subgrid scale simulation is necessary.

The simulation of fine-scale precipitation variability has been addressed in a computer code (TEMPEST) which uses Monte Carlo techniques to estimate the fundamentally stochastic aspects of rainfall morphology. The basic unit adopted in the model is a cell, and the current representation of the major elements of cell morphology are given in Table I.

TEMPEST incorporates a sophisticated geometry package, MCN,² which allows a very flexible specification of number and shapes of precipitating zones. The input parameters for rainfall character are zonedependent so that it is a straightforward matter to construct a synoptic storm with its regions of stratiform precipitation, drizzle frontal showers, and post-frontal showers.

In preliminary tests of the code, we have simulated the time history of rainfall collection at a stationary rain gauge with a resolution of 0.1 h. These were compared with high resolution experiments^{3,4} to select the first round of input parameters. The ability of the model to produce integral statistics on 1-, 3-, and 6-h accumulations was then tested against traditional climatological data to identify the spectrum of rain-area-fractions and intensity distributions for a selected geographic site.