MARSHALL ISLANDS FILE TRACKING DOCUMENT

Record Number: 377

File Name (TITLE): Operation Nucleus Calculation Analysis of Atomic Debris

Document Number (ID): WT-932 (EX)

DATE: 7/1959

Previous Location (FROM): CIC

AUTHOR: C.K. Reed and W. Singleton

Addditional Information: _____

OrMIbox: 21

CyMIbox: _____

15202

WT-932 (EX)
EXTRACTED VERSION

OPERATION CASTLE

Project 7.4
Nuclear Calibration Analysis of Atomic Device Debris

March-May 1954

Headquarters Field Command
Defense Atomic Support Agency
Sandia Base, Albuquerque, New Mexico

July 9, 1959

NOTICE

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Extract version prepared for:

Director
DEFENSE NUCLEAR AGENCY
Washington, D.C. 20305

1 April 1981

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UNCLASSIFIED
SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered)

REPORT DOCUMENTATION PAGE		READ INSTRUCTIONS BEFORE COMPLETING FORM
1. REPORT NUMBER WT-932 (EX)	2. GOVT ACCESSION NO.	3. RECIPIENT'S CATALOG NUMBER
4. TITLE (and Subtitle) Operation CASTLE - Project 7.4 Nuclear Calibration Analysis of Atomic Device Debris		5. TYPE OF REPORT & PERIOD COVERED 6. PERFORMING ORG. REPORT NUMBER WT-932 (EX)
7. AUTHOR(s) C. K. Reed W. Singlevich		8. CONTRACT OR GRANT NUMBER(s)
9. PERFORMING ORGANIZATION NAME AND ADDRESS Headquarters Field Command Defense Atomic Support Agency Sandia Base, Albuquerque, New Mexico		10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS
11. CONTROLLING OFFICE NAME AND ADDRESS		12. REPORT DATE July 9, 1959 13. NUMBER OF PAGES 33
14. MONITORING AGENCY NAME & ADDRESS(if different from Controlling Office)		15. SECURITY CLASS. (of this report) Unclassified 15a. DECLASSIFICATION/DOWNGRADING SCHEDULE

16. DISTRIBUTION STATEMENT (of this Report)

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17. DISTRIBUTION STATEMENT (of the abstract entered in Block 20, if different from Report)

18. SUPPLEMENTARY NOTES

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number)

Operation CASTLE
Calibration Data
Nuclear Debris
Instrumentation

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

FOREWORD

This report has had classified material removed in order to make the information available on an unclassified, open publication basis, to any interested parties. This effort to declassify this report has been accomplished specifically to support the Department of Defense Nuclear Test Personnel Review (NTPR) Program. The objective is to facilitate studies of the low levels of radiation received by some individuals during the atmospheric nuclear test program by making as much information as possible available to all interested parties.

The material which has been deleted is all currently classified as Restricted Data or Formerly Restricted Data under the provision of the Atomic Energy Act of 1954, (as amended) or is National Security Information.

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It is the belief of the individuals who have participated in preparing this report by deleting the classified material and of the Defense Nuclear Agency that the report accurately portrays the contents of the original and that the deleted material is of little or no significance to studies into the amounts or types of radiation received by any individuals during the atmospheric nuclear test program.

ABSTRACT

The primary objective of Project 7.4 was to obtain calibration data on the nuclear and physical properties of solid, liquid, and gaseous matter associated with air-borne nuclear debris resulting from nuclear detonations. These data were obtained by the application of chemical, radiochemical, physical, and nuclear-physical analyses to the debris collected by specialized sampling devices. The calibration data were further extended by making similar measurements on nuclear debris collected at great distances from the site of detonation.

Nuclear-debris samples close-in to the detonation site were obtained utilizing sampling devices on F-84, WB-29 and B-36 aircraft. In addition, WB-29's similarly equipped operated out of Hawaii for the long-range calibration samples.

Sufficient fission product isotopes in particulate debris were determined from each detonation to establish fission-yield curves. Effects of the large fluxes of high-energy neutrons on the trough elements and right wing elements were observed; the significance of these effects are discussed.

Mass spectrometric analyses of plutonium and uranium isotopes showed evidence of thermonuclear reactions; plutonium isotopes up to Pu^{246} were easily measured in the debris.

Induced activities much higher than noted for fission devices were observed; notably higher than ever measured before were particularly in Shots 3, 4, and 5. Modal specific beta activity values for barge shots were much higher than for island shots.

Full-scale tests of gaseous debris samplers indicated that further engineering refinements were necessary although some useful samples were obtained. Measurements for C^{14}, A^{37}, Kr^{85}, H^{3}, and Xe^{133} did not show any consistent pattern as related to the devices tested. The variation of these data cannot be specifically attributed to sampling equipment, laboratory analysis, or natural fractionation of isotopes. It is qualitatively inferred that ratios were significantly higher for Castle shots than for Ivy-Mike. It is postulated that samples well above the troposphere are required for megaton shots to insure high-quality samples.

It is generally recommended that these calibration tests, both close-in and at long-range, be continued with emphasis on improving debris collection devices and refining analytical procedures used.

FOREWORD

This report is one of the reports presenting the results of the 34 projects participating in the Military Effects Tests Program of Operation Castle, which included six test detonations. For readers interested in other pertinent test information, reference is made to WT-934, "Summary Report of the Commander, Task Unit 13, Programs 1-9," Military Effects Program. This summary report includes the following information of possible general interest: (1) an overall description of each detonation, including yield, height of burst, ground zero location, time of detonation, ambient atmospheric conditions at detonation, etc., for the six shots; (2) discussion of all project results; (3) a summary of each project, including objectives and results; (4) a complete listing of all reports covering the Military Effects Tests Program.

PREFACE

This report is intended to present the high lights of factual information obtained from this project's participation in Operation Castle. Detailed evaluation of the data is minimized in this presentation; emphasis is placed on data presentation. Broad and intensive evaluation of the results of this report as related to the mission of Headquarters, United States Air Force, Washington 25, D. C. [ ] are included in other publications (Reference 1).

This report was prepared by the Office of the Technical Director, Headquarters, United States Air Force, Washington, D. C., under the overall command of Brigadier General Hooks and under the technical direction of D. L. Northrup.

The conclusions as summarized in this report are based on the efforts of many individuals and organizations participating in this project. It is an impossible task to properly acknowledge each and every individual contribution to the efforts of this program; however, an attempt will be made to acknowledge some of the agencies and their key personnel who contributed to the overall success of the program.

Personnel of AFOAT-1 who participated in the planning, execution, and report preparation and review included: Dr. D. H. Rock, Dr. W. D. Urry, Lieutenant Colonel R. E. Heft, Captain D. N. Weiford, Captain O. J. Kvamme, J. W. Ponds, Major W. E. Scott, Major Robert S. Brundage, L. Sherrill, and Miss K. Harding. Captain F. F. Nicaise was officer-in-charge of gas sampling operations at Eniwetok. In addition, the program's success was greatly enhanced by the support given by many participating branches of the United States Air Force and the United States Atomic Energy Commission.

Dr. R. W. Spence and members of his staff of the Los Alamos Scientific Laboratory (LASL), Los Alamos, New Mexico, and Dr. K. Street and his staff members of the University of California Radiation Laboratory (UCRL), Livermore, California, contributed to this program by mutual exchange of samples, analytical data and ideas.

The assistance of Mrs. R. M. Ripley and Mrs. J. E. Kaul in the preparation of this report is gratefully acknowledged.

The following laboratories and their key personnel contributed to the Castle program:

Tracerlab, Inc., Boston, Massachusetts: Technical Director, Dr. W. C. Peacock; rare earth radiochemistry, Drs. R. Epple, J. W. Shearer, and H. Petrow; gas separation and counting, Drs. I. J. Berstein, R. Epple, and J. W. Shearer; physical studies, Dr. J. W. Shearer and C. H. Sherman.

Tracerlab, Inc., Berkeley, California: Technical Director, Dr. Lloyd R. Zumwalt; radiochemistry, Messrs. A. DeHaan, Jr., L. J. Beaufait, Jr., Leon Leventhal, and H. E. Menker.

Argonne National Laboratory, Chicago, Illinois: Technical Director, Dr. Winston Manning; radiochemistry, uranium and plutonium, Drs. Sherman T. Fried and Gray Pyle; gas purification, Dr. F. T. Hagemann.

Armour Research Foundation, Chicago, Illinois: petrographic analysis, Drs. W. McCrone and J. Krc.

U. S. Naval Radiological Defense Laboratory, San Francisco, California: radiochemistry, rare earths, uranium and plutonium, Drs. N. E. Ballou and L. R. Bunney.

The USAF McClellan Central Laboratory, McClellan Air Force Base, California: radiochemistry, fission products, rare earths, induced activities and uranium, Majors I. J. Russell, W. J. Worthington, Jr., G. M. Williams, H. O. Larson, J. Spencer, and Captains O. J. Kvamme and G. F. Jubber.

CONTENTS

Chapter I
INTRODUCTION

1.1 OBJECTIVES

The principal technical objectives of this project included the following:

1. To obtain the necessary data---utilizing chemical, radiochemical, physical, and nuclear-physical techniques on close-in nuclear debris---to establish reference or calibration points for analyses, using the same techniques, of debris from nuclear explosions of unknown origin, composition, and design.
2. To compare analyses on samples collected close-in to the detonation with those obtained at great distances, in order to study variability of debris composition with time and distance from detonation site.
3. To test the Squeegee gas-sampling device under full-scale operational conditions.

1.2 BACKGROUND

This experiment was an extension of a program established to monitor all United States nuclear explosions, in order to establish calibration or reference points based on analyses of air-borne nuclear debris collected under the best possible conditions. This program, under Headquarters, United States Air Force, Washington 25, D. C. (AFOAT-1), had actively participated in Operations Sandstone, Ranger, Greenhouse, Buster-Jangle, Tumbler-Snapper, Ivy, and Upshot-Knothole.

Data based on debris analysis from Trinity, with specific reference to capture-to-fission ratios and bomb efficiency, suggested the possibility that these types of analyses might be extended to give more diagnostic information about the source than had been thought possible. Therefore, serious efforts were expended in applying micro- and macro-radiochemical techniques, and other specialized analytical method to air-borne nuclear debris. These analyses yielded useful diagnostic information. It became possible to determine nuclear efficiency, [redacted] and other useful information required in a detection and analysis system.

A condensed review of results obtained using the techniques described here during Operations Sandstone, Ranger, Greenhouse, and---particularly---Buster-Jangle, Tumbler-Snapper, and Upshot-Knothole appear in [redacted] publications (References 2, 18, and 19).

Chapter 2
PROCEDURE

2.1 INSTRUMENTATION

A preliminary resume of the operational techniques, aircraft instrumentation and procedures used in the collection of gaseous debris from Castle have been briefly described in References 1, 2, 7, 8, 18, and 19. Close-in particulate and gaseous samples were obtained by F-84 and B-36 aircraft penetrating the cloud resulting from each detonation. The Air Weather Service WB-29 aircraft equipped with particulate and gaseous sampling devices collected samples at remote distances from the detonation site.

Five F-84G aircraft utilized the method of snap gas-sampling, which was the primary collection method for obtaining close-in samples during Operations Ivy and Upshot-Knothole (Reference 7). This consisted of an exterior stainless-steel probe in the nose of the aircraft which fed into a deflated polyethylene bag installed in the gun deck portion of the aircraft. Samples were taken by activating a valve and filling the polyethylene bag by ram pressure. On return of the aircraft to the ground, the sample was transferred from the bag by evacuation, using a diaphragm pump, and stored in a G-1 cylinder. The radioactive gases of interest were measured and the results compared with similar analyses of gases collected by the technique described in the following paragraph.

Ten F-84G's were equipped with a dual electrical compressor system feeding into two 500-in^3 compression cylinders (3,000 psi). All of the air sampled was bled from an intermediate stage of the axial compressor of the aircraft and fed into the dual compressors located in the gun-deck section. This method of collection---called the Squeegee method---had been tried experimentally during the Upshot-Knothole tests. Operation Castle provided the first full-scale operational test of this high-pressure system. In addition, several B-36's equipped with the Squeegee system were utilized. In these cases, intake air was bled from the upstream side of the large cabin pressurization filter to six compressors located in the bomb bay. Each compressor pumped into its individual 900-in^3 high-compression cylinder (3,000 psi).

Longer-range samples were obtained using WB-29 aircraft with associated C-1 foils for particulate samples, and a B-31 gas-sampling device for the gaseous debris (Reference 7).

The collection of all close-in particulate samples was under the technical direction of the Los Alamos Scientific Laboratory (LASL), and the collection of gas samples was supervised by Headquarters, United States Air Force [redacted], The University of California Radiation Laboratory (UCRL) was responsible for gas separation and analyses of some samples at the test site.

The instrumentation, techniques, and procedures in the processing, separation, and assay of the nuclear particulate and gaseous debris---both

close-in and long-range---are of such magnitude and variation that it is not practical to itemize these in this report. Chemical procedures for separation and assay of the radioactive isotopes, specialized separation equipment, counting equipment, and other instruments are included in the detailed reports by agencies responsible for the separation and assay of these isotopes; the most pertinent are References 3, 4, 5, 13, 14, 15, 16, and 17.

2.2 OPERATIONAL PROCEDURES

2.2.1 Close-In Sampling. Close-in gas samples were collected during Castle at altitudes of 35,000 to 52,000 feet MSL. Sampling aircraft were directed into the nuclear cloud generally no sooner than two hours subsequent to the detonation and followed each cloud for approximately 5 to 7 hours, obtaining samples. To ensure no cross contamination of sampling equipment between shots, control samples were taken before and after a washdown of the sampling equipment. Gaseous-debris sample sizes collected varied from 10^{-15} to 10^{-17} bomb fraction. Duration of sample collection time varied from approximately 40 to 60 minutes in the case of the Squeegee method to less than 1 minute by the snap-sampling method. Squeegee gas samples in the high-pressure spheres were removed from the aircraft upon return to the ground and crated for shipment to the separation laboratories. Transfer of snap samples from the polyethylene bag to a G-1 cylinder was required prior to shipment.

Representative sections of each test could were sampled, but because of extreme cloud heights attained, sampling was conducted in only the lower portions of the cloud for the high-yield detonations.

2.2.2 Long-Range Samples. Long-range samples were collected by WB-29 aircraft staging out of Hickam Field, Guam, and McClellan Air Force Base (California). Samples were collected from approximately sea level to 20,000 feet altitude. Gas samples were obtained with B-31 collection equipment, which consisted of a Quincy compressor feeding into 5 J-1 gas-storage cylinders. The average sample size collected was approximately 500 ft^3. In addition, these aircraft were equipped with C-1 particulate samplers employing IPC paper as the filter medium.

2.3 ANALYTICAL PROCEDURES

2.3.1 Radiochemical; Particulate. Particulate nuclear debris collected by the filter-paper technique was radiochemically analyzed in order to provide the following information:

1. Sufficient fission-product data to establish a fission-yield curve with emphasis on studying the trough elements, peak elements, and those on the right wing of the fission-yield curve. About 30 fission products, from Zn^{72} through Tb^{161}, were chemically separated from the gross sample and assayed. These were then referred to Mo^{99} measured in the same sample.
2. Uranium and plutonium isotopic abundances were determined by

first chemically separating the plutonium and uranium from the gross sample and then submitting the separated fractions to mass spectrographic and pulse analyses. Extremely low levels of uranium and plutonium can be determined in this manner.

3. Certain induced activities such as iron, beryllium, nickel, cobalt, etc. were also chemically separated from the gross sample and individually assayed. These results are discussed in Chapter 3, Section 3.1.5. The detailed analytical and assay procedures for this complex array of data can be found in References 10, 11, 12, 13, 14, and 15.

2.3.2 Radiochemical; Gas. The principal gases of interest in Castle gas samples included C^{14} (measured as $C^{14}O_2$), A^{37}, Kr^{85}, Xe^{133}, and H^3 (measured as $H^3{}_2O$). Since the gas samples occurred in varying volumes, at least two separation systems capable of handling the varied volumes were required. A larger gas-separation system was utilized for the B-31 and the B-36 Squeegee samples, and a smaller separation train was utilized for the snap and F-84G Squeegee samples. Carrier for krypton and xenon was used in all samples separated; occasionally, samples were spiked with D_2O as a tracer for the tritium measurements. Separation, decontamination, and sample cross-contamination problems are discussed in detail in References 3, 4, and 5. Upon separation of the desired gas fractions, accurate assay or counting is required. Separations were accomplished at Tracerlab, Inc., and assay was principally done at the Argonne National Laboratory, Chicago, Illinois.

Current separation and assay procedures are sufficiently sensitive to measure background quantities (Reference 11). It was hoped that Castle tests would give gas fractions sufficiently high above background to explore and test the usefulness of these quantities in terms of interpreting phenomena associated with the nuclear explosions.

2.3.3 Physical and Petrographic. The primary prerequisite for physical and petrographic studies of particulate nuclear debris was the separation of the radioactive particles from the filter-paper medium and other inert particles. When individual particles were separated, they were observed under optical microscopes and their size determined. These individual particles were then examined for color, shape, and X-ray diffraction patterns, and also for specific beta and alpha activity. In some instances, the composition of the particles was measured when pertinent to the overall evaluation of these analyses.

Occasionally, individual particles were subjected to nuclear film studies to observe low-level alpha activity by studying the tracks produced by the radiations. This technique was sometimes useful for detecting the presence of polonium.

Chapter 3
RESULTS

3.1 RADIOCHEMICAL ANALYSES OF PARTICULATE DEBRIS

3.1.1 Fission Products. The fission-product results are reported in terms of R-values, where an R-value is defined by the relation:

$$R = \frac{(a_1/a_2)_S}{(a_1/a_2)_t} = \frac{Y_1 e_1 \lambda_1 / Y_2 e_2 \lambda_2}{(Y_1)_t e_1 \lambda_1 / (Y/2)_t e_2 \lambda_2} = \frac{(Y_1/Y_2)_S}{(Y_1/Y_2)_t} \qquad (3.1)$$

Where: $(a_1/a_2)_S$ = fission-product activity ratios of two isotopes measured in a debris sample. The activity is corrected for the decay between time of explosion and time of analysis

$(a_1/a_2)_t$ = fission-product activity ratios of the the same two isotopes from a sample of U^{235} irradiated using thermal neutrons; same procedures and equipment used as for determining $(a_1/a_2)_S$

e = counting efficiencies

Tables 3.1 and 3.2 list the fission-product data for the Castle tests. These tables present the Mo^{99} R-values obtained from samples collected close-in to the point of detonation and from samples collected at Hickam Field, Guam, and McClellan Air Force Base (California). In most instances, the values quoted are the weighted average of measurements made in three laboratories. Error limits shown are the standard deviations. Neither time nor facilities permitted extensive investigation of the characteristics of the debris as a function of distance from the detonation site. The long-range-debris (LRD) values quoted are based on a limited number of samples, and in some instances, there was a considerable spread in the values obtained for individual isotopes. No LRD values are given for Shot 3, since all LRD samples collected for this event were badly admixed with older debris.

3.1.2 Rare Earths. The rare-earth data listed in Tables 3.3 and 3.4 are the most representative R-values available for the Castle shots. The accuracy of the data is such that no interpretive value should be

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placed on differences in comparative values of perhaps 25 percent or less.

3.1.3 Uranium Beta Emitters. Table 3.5 lists the heavy-element data for the six shots. This table presents the results obtained by radiochemical analysis of samples collected close-in to the point of detonation and of samples collected at Hickam Field, Guam, and McClellan (California) for LRD comparisons. The values given are the weighted average of measurements made in three laboratories and are expressed as atom ratios. The error limits shown are the standard deviations calculated from the average values of the individual determinations.

3.1.4 Plutonium and Uranium Alpha Emitters. Table 3.6 lists the pulse-analysis results obtained by Tracerlab, Inc. on close-in and LRD samples. Unless otherwise noted, fissions are based on Mo^{99}. Mass spectrometric measurements made at Argonne National Laboratory, Chicago, Illinois, on close-in and LRD samples are presented in Tables 3.7 and 3.8. Error limits shown are the calculated standard deviations from the average values of the individual determinations.

3.1.5 Induced Activities. Table 3.9 lists the induced-activity data obtained from close-in samples. The close-in values represent the measurements made in two laboratories. The error limits given are the calculated standard deviations from the average values of the individual determinations. The LRD samples for Shots 2, 3, and 6 were not analyzed for induced activities, and only very limited analyses were made in the LRD samples from the other shots. A comparison of these few LRD data with close-in data revealed a moderate spread in the values, but did not suggest any large degree of variability with distance from detonation site.

3.2 RADIOCHEMICAL ANALYSIS OF GAS SAMPLES

3.2.1 Background and Theoretical Data. The gas-sampling program was a continuation of a study to measure selected radioactive induced gases and fission-product gases associated with air-borne nuclear debris from Castle-type shots. Gases selected and measured during Upshot - Knothole indicated that the most promising gases---both from a diagnostic and detection point of view---included C^{14} as $C^{14}O_2$ (produced principally by n, p, on nitrogen), A^{37} as argon gas (produced by n, γ, on stable argon), fission-product gas Kr^{85} and Xe^{133}, and tritium present as H^3_2O formed during D + D and D + T reactions and/or formed by neutron capture by Li^6. Earlier experimental work during Ivy indicated that most or all tritium associated with nuclear debris was in the liquid physical state, i.e., water. Attempts were also made to measure the extent, if any, of absorbed and/or adsorbed gases in the particles of the debris.

Based on Upshot - Knothole tests, sampling was performed utilizing Squeegee equipment rather than the Ivy-type snap samplers. The xenon and krypton carrier was added to the high-compression sample cylinders prior to sample collection to aid in determining yields and recovery in the laboratory processing of the samples.

In general, to ensure no cross contamination within the sampling

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equipment itself, control gas samples were taken before and after decontamination of the sampling equipment. Equipment was decontaminated after each shot. Spot checks during Upshot - Knothole indicated decontamination factors exceeding 1,000 which were deemed satisfactory for close-in samples.

Gas samples were also collected at long-range in the vicinity of Hawaii using B-31 equipment. Variability, fractionation of gas isotopes with respect to each other and with respect to particulate debris, and rainout of tritium were to be studied.

Unfortunately---as experienced during Ivy and Upshot - Knothole---many samples, particularly the LRD samples, were compromised because of cross-contamination in the laboratory, particularly with respect to tritium. Due to the variation in size of the gas samples to be separated and assayed, two sets of separation equipment were used. Experiments conducted to determine the amount of holdover contamination in this equipment revealed that the large gas separation equipment used to assay the B-31 LRD gas samples and the B-36 Squeegees was not always effectively flushed after one separation. Redesign of traps and improved methods of steam flushing, followed by lengthy drying periods, removed the possibility of cross contamination of samples, but only after certain samples were lost or results were determined as invalid.

In the separation process it was also discovered by the University of California Radiation Laboratory (UCRL) personnel that tritium activity was being lost through an exchange of tritium in the sample with the plastic liner (heresite) of the Squeegee sample sphere. The tritium lost by this mechanism was recovered by treating the inside of the spheres with three separate rinses of hot, alkaline, potassium permanganate solution. The resultant mixture from each rinse was then processed and assayed. This result was added to the result obtained by assaying the water and water vapor in the sphere. Certain B-36 Squeegee samples in which assay of liner activities was not made are noted in Tables 3.10 through 3.15, and therefore do not represent the total tritium activity present in the sample.

Most of the values reported in the tables are believed to be reasonably valid. Those values wherein known cross contamination occurred have been deleted from the presented data.

Couriering of samples from the test site, separation, processing, and assay of all gas collected was handled by Headquarters, United States Air Force, Washington 25, D. C. [redacted] or by agencies responsible [redacted] under military contract. Procedures, instrumentation, and the processes of gas analysis are described in References 3, 4, and 5, and no attempt will be made to describe these methods here.

3.2.2 Definition of Units Expressing Results. In accordance with past procedures, and in order to standardize results, all activity results are expressed as atoms of a given isotope per unit volume of a given air sample at a specified temperature and pressure. The unit volume was defined in terms of moles: i.e., 1 equivalent mole air (EMA) is that volume occupied by 1 mole of air at 70°F and 760-mm pressure. The approximate volume of 1 mole equivalent air is 0.85 ft^3.

TABLE 3.10 SQUEEGEE AND SNAP GAS SAMPLES, SHOT 1

Sample No.	Date and Altitude (ft.msl.)	Collection Time After Shot (Hrs & Min)	GROSS ATOMS PER EMA†				
			$C^{14}O_2 \times 10^8$	$A^{37} \times 10^3$	$Kr^{85} \times 10^8$	$H^3 \times 10^8$	$Xe^{133} \times 10^8$
CB-1-SS-2	2-28-54 40,000	2 + 49	-5.2 ± 8.3	---	12.90 ± 0.40	19,800 ± 200	---
CB-1-FQ-2	2-28-54 40,000	2 + 51	11.9 ± 0.9	4.7 ± 12.0	0.47 ± 0.02	1,150 ± 30	---
CB-1-BQ-2	2-28-54 50,000	3 + 49	2.1 ± 0.1 2.7 ± 0.1	0.5 ± 0.3	0.84 ± 0.08	313 ± 23 * 817 ± 2*	3.0 ± 0.3
CB-1-FQ-1	2-28-54 35,000	4 + 24	2.7 ± 1.3	11.0 ± 6.3	0.34 ± 0.02	53 ± 2	0.25 ± 0.03
CB-1-FQ-5	2-28-54 39,000	4 + 27	5.0 ± 1.5	486.0 ± 155.0	0.11 ± 0.01	6.6 ± 2.0	---
CB-1-FQ-3	2-28-54 42,000	4 + 29	3.4 ± 2.1	---	0.04 ± 0.01	1.7 ± 0.2	---
CB-1-BQ-1	2-28-54 51,000	5 + 04	2.4 ± 0.1 2.7 ± 0.1	1.7 ± 0.6	0.98 ± 0.10	558 ± 14	---

* H^3 activity does not include activity contained in Squeegee liner and is probably low.
† EMA is equivalent mole air or 22.4 liters STP air.

TABLE 3.11 SQUEEGEE AND SNAP GAS SAMPLES, SHOT 2

Sample No.	Date and Altitude (ft.msl.)	Collection Time After Shot (Hrs & Min)	GROSS ATOMS PER EMA†				
			$C^{14}O$, x 10^8	A^{37} x 10^3	Kr^{85} x 10^8	H^3 x 10^8	Xe^{133} x 10^8
CR-2-FQ-11	3-26-54 39,000	2 + 04	169.0 ± 3.0	3,470 ± 20	0.66 ± 0.03	1,000 ± 19	3.0 ± .03
CR-2-FQ-12	3-26-54 40,000	3 + 03	4.2 ± 1.3	235 ± 27	0.02 ± 0.02	14 ± 6	---
CR-2-FQ-18	3-26-54 42,500	3 + 44	256.0 ± 3.0	3,980 ± 60 4,360 ± 20	0.44 ± 0.02	332 ± 7	---
CR-2-BQ-4	3-26-54 51,000	3 + 47	2.3 ± 0.2 3.5 ± 0.1	1.8 ± 1.2	0.02 ± 0.00	454 ± 34*	2.1 ± 0.2
CR-2-SS-5	3-26-54 39,000	3 + 51	-11.9 ± 13.8	2,980 ± 770	---	2,290 ± 90	---
CR-2-FQ-7	3-26-54 39,500	3 + 53	6.6 ± 1.1	193 ± 119	---	666 ± 20	---
CR-2-BQ-3	3-26-5 51,000	7 + 15	8.0 ± 0.2 8.4 ± 0.1	71 ± 1 91 ± 1	0.01 ± 0.00	114 ± 5	1.2 ± 0.1

* H^3 activity does not include activity contained in Squeegee liner and is probably low.
† EMA is equivalent mole air or 22.4 liters STP air.

TABLE 3.12 SQUEEGEE AND SNAP GAS SAMPLES, SHOT 3

Sample No.	Date and Altitude (ft.msl.)	Collection Time After Shot (Hrs & Min)	GROSS ATOMS PER EMA†				
			$C^{14}O_2 \times 10^8$	$A^{37} \times 10^3$	$Kr^{85} \times 10^8$	$H^3 \times 10^8$	$Xe^{133} \times 10^8$
CK-3-FQ-16	4-6-54 40,000	2 + 15	62.4 ± 1.3	2,670 ± 30 3,680 ± 60	0.03 ± 0.01	225 ± 4	---
CK-3-SS-6&7	4-6-54 38,500	3 + 15	17.2 ± 15.1	449 ± 375	3.51 ± 0.60	4,950 ± 420	---
CK-3-BQ-6	4-6-54 52,500	3 + 59	174.0 ± 1.0 180.0 ± 0.5	7,990 ± 90 10,100 ± 20	1.45 ± 0.07	110 ± 5*	10.1 ± 1.0
CK-3-BQ-5	4-6-54 45,000	5 + 05	11.9 ± 0.3 12.3 ± 0.2	296 ± 3 495 ± 2	0.17 ± 0.01	1,620 ± 30	0.23 ± 0.01

* H^3 activity does not include activity contained in Squeegee liner and is probably low.
† EMA is equivalent mole air or 22.4 liters STP air.

TABLE 3.13 SQUEEGEE AND SNAP GAS SAMPLES, SHOT 4

Sample No.	Date and Altitude (ft.msl.)	Collection Time After Shot (Hrs & Min)	GROSS ATOMS PER EMA *				
			$C^{14}O_2 \times 10^8$	$A^{37} \times 10^3$	$Kr^{85} \times 10^8$	$H^3 \times 10^8$	$Xe^{133} \times 10^8$
CU-4-FQ-29	4-25-54 41,500	2 + 47	211.0 ± 8.0	20,000 ± 200 23,900 ± 700	---	927 ± 11	5.1 ± 0.5
CU-4-SS-11	4-25-54 40,000	3 + 20	47.3 ± 12.6	---	4.01 ± 0.12	51,500 ± 400	---
CU-4-FQ-36	4-25-54 37,500	3 + 48	98.4 ± 1.7	548 ± 40	0.47 ± 0.01	4,360 ± 140	---
CU-4-FQ-32	4-25-54 37,500	3 + 48	55.2 ± 1.3	5,350 ± 63	0.43 ± 0.01	3,660 ± 100	---
CU-4-FQ-31	4-25-54 37,500	3 + 48	---	5,920 ± 30 5,280 ± 50	---	4,150 ± 60	---
CU-4-BQ-8	4-25-54 51,000	4 + 03	2.5 ± 0.1 2.3 ± 0.1	-2 ± 4	0.01 ± 0.00	349 ± 7	2.8 ± 0.2

* EMA is equivalent mole air or 22.4 liters STP air.

TABLE 3.14 SQUEEGEE AND SNAP GAS SAMPLES, SHOT 5

Sample No.	Date and Altitude (ft.msl.)	Collection Time After Shot (Hrs & Min)	GROSS ATOMS PER EMA *				
			$C^{14}O_2 \times 10^8$	$A^{37} \times 10^3$	$Kr^{85} \times 10^8$	$H^3 \times 10^8$	$Xe^{133} \times 10^8$
CY-5-FQ-23	5-4-54 40,500	2 + 05	3.0 ± 0.4	41.0 ± 9.0 0.9 ± 9.2	---	1,670 ± 30	---
CY-5-FQ-43	5-4-54 40,000	2 + 27	6.9 ± 0.4	10.0 ± 5.0 11.0 ± 7.0	---	206 ± 3	---
CY-5-SS-13	5-4-54 38,000	3 + 00	28.2 ± 8.3	---	6.59 ± 0.20	60,000 ± 5500	---
CY-5-BQ-9	5-4-54 52,600	3 + 50	332.0 ± 3.0 356.0 ± 0.1	2,390 ± 30 2,810 ± 9.0	0.19 ± 0.01	253 ± 7	6.53 ± 0.65
CY-5-FQ-5	5-4-54 39,000	4 + 00	7.4 ± 1.0	-0.1 ± 0.2	0.22 ± 0.33	750 ± 9	---
CY-5-FQ-40	5-4-54 38,000	4 + 13	31.3 ± 1.1	---	---	842 ± 15	---

*EMA is equivalent mole air or 22.4 liters STP air.

TABLE 3.15 SQUEEGEE AND SNAP GAS SAMPLES, SHOT 6

Sample No.	Date and Altitude (ft.msl.)	Collection Time After Shot (Hrs & Min)	GROSS ATOMS PER EMA†				
			$C^{14}O_2 \times 10^8$	$A^{37} \times 10^3$	$Kr^{85} \times 10^8$	$H^3 \times 10^8$	$Xe^{133} \times 10^8$
CN-6-BQ-7	5-13-54 51,000	3 + 40	45.2 ± 4.5	13,600 ± 40 13,700 ± 200	1.75 ± 0.05	441 ± 14	4.85 ± 0.48
CN-6-SS-14	5-13-54 40,000	3 + 55	11.9 ± 26.3	---	0.14 ± 0.01	1,890 ± 20	---
CN-6-BQ-10	5-13-54 48,500	4 + 53	25.6 ± 0.2 22.8 ± 0.2	587 ± 4 1,020 ± 20	0.07 ± 0.01	73 ± 1*	1.03 ± 0.05
CN-6-FQ-37	5-13-54 36,000	6 + 25	5.6 ± 0.5	52.0 ± 7.3 21.9 ± 5.6	---	650 ± 18	---
CN-6-FQ-41	5-13-54 36,000	6 + 25	36.2 ± 1.7	463 ± 21	---	276 ± 3	---

*H^3 activity does not include activity contained in Squeegee liner and is probably low.
†EMA is equivalent mole air or 22.4 liters STP air.

To calculate the results as shown in Section 3.2.3, the following constants were accepted as standard (Reference 6).

Isotope	Natural Abundance	Half-life	λ, Min^{-1}
$C^{14}O_2$	3.3×10^{-2}	5,720.0 years	2.30×10^{-10}
A^{37}	9.3×10^{-1}	34.1 days	1.41×10^{-5}
H^3	---	12.4 years	1.06×10^{-7}
K_r^{85}	1.12×10^{-4}	10.7 years	1.23×10^{-7}
X_e^{133}	8.7×10^{-6}	5.27 days	0.91×10^{-4}

The following backgrounds were chosen as being representative of the general test area:

Isotope	Background Atoms per Equivalent Mole Air
$C^{14}O_2$*	2.2×10^{8}†
K_r^{85}	2.0×10^{5}†
A^{37}*	Essentially Zero
H^3	Between 10^5 and 10^6 at 20,000 feet
X_e^{133}	Zero

*These specific activities reported in Los Alamos Scientific Laboratory Report, LA-1102, as 180 and 0.03 dis/min.

†Experimentally determined values based on unpublished data of Headquarters, United States Air Force [redacted] For LRD B-31 samples taken at 20 degrees to 30 degrees N latitude, values for K_r^{85} background are taken as 2.4×10^5 atoms per equivalent mole air.

Methods used to compute atoms per equivalent mole air for a specific gas of a sample in accordance with separation and counting techniques are described in Reference 5. All isotopic quantities reported have been corrected for decay which occurred during the interval prior to assay.

3.2.3 Individual Results. Tables 3.10 through 3.23 summarize results for all six shots. Close-in sampling results are presented in Tables 3.10 through 3.21, and LRD sampling results in Tables 3.22 and 3.23. All individual isotope concentrations are expressed as atoms per equivalent mole air. For the close-in sampling, quantities of any specific gas show concentrations which are normally well above background. The LRD sample results are not sufficiently higher than background in most cases to justify the validity of the computed ratios or any conclusions which are reached therefrom. A great deal of the LRD sample data has been omitted wherein isotopic concentrations were at background level. As presented in the tables, sample numbers indicate the type and collection method of the particular sample. The sample Code FQ refers to Squeegee samples collected by the F-84G, BQ the Squeegee samples collected from the B-36, and SS the F-84G snap sample collections.

Calculated fissions in any one sample are based on the yield of the Kr^{85} fission product. The Castle series is estimated to have yielded 0.22 percent of this gaseous isotope per fission.

3.2.4 Atom and Other Ratios. In order to correlate quantities of a particular isotope present in a sample, atom ratios have been taken and are shown in Tables 3.16 through 3.21, and 3.23. Ratios were also taken between the induced activities A^{37} and C^{14}. This ratio has been calculated to be approximately 1.4×10^4 (Reference 9). Ratios relating the activities of H^3 and C^{14} with respect to fissions have also been taken with view towards a correlation with the excess neutrons released by the Castle type of nuclear explosions.

A calculation of the tritium residue of each shot has been made wherever possible as determined by the H^3/f ratio within each particular sample. These calculations are based on total fission estimates by the Los Alamos Scientific Laboratory (LASL).

In all cases, the individual results represent gross-activity measurements; the backgrounds reported in Section 3.2.2 were subtracted, where significant, when computing these ratios.

3.2.5 Operation of the Squeegee Sampler. Castle was the first full-scale operational testing of the small-size, high-pressure Squeegee, although sufficient experimentation had been accomplished during Upshot - Knothole to indicate that this method was successful. This method proved ideal for ease of removal of sample from contaminated aircraft and handling enroute to processing laboratory. During the Castle tests, the main malfunctions of the system consisted of: (1) high-pressure leaks from fittings and connections resulting in the loss of certain samples, (2) compressor difficulties, and (3) faulty check-valve operation due to freeze up at high altitudes, resulting in either loss of sample or no collection being made. These defects were corrected, as Castle tests progressed, with improved operational procedures and maintenance. Of all Squeegee flights during Castle, 68 percent resulted in successful missions and 18 percent were only partially successful in sample collection; 14 percent of the missions failed. The size of most good

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samples collected was adequate for assay and separation, showing much improvement in this respect over the snap-sample volume.

3.3 PHYSICAL ANALYSIS OF PARTICULATE DEBRIS

3.3.1 Petrographic Analyses. Petrographic studies were made of individual radioactive debris particles collected from each of the six shots of the Castle series. For these tests the major constituents of the carrier material fall roughly into three groups as shown in Table 3.24. Further details as to refractive index studies and observations detailing the size, color and shapes of the individual particles observed can be found in Reference 12.

3.3.2 Specific Beta-Activity Measurements. Table 3.25 lists the modal specific beta activity determined for each of the Castle detonations. The modal values are only very roughly known, since the observed frequency distributions covered a broad spectrum of specific activities with no pronounced peaks (for further details, see Reference 13).

3.3.3 Gross Activity Measurements. Beta and alpha measurements were made by Tracerlab, Inc. on gross samples from each of the Castle detonations. These measurements together with estimates of the Pu/Qf ratio are presented in Table 3.26.

Chapter 4
DISCUSSION

4.1 FISSION PRODUCT ANALYSES

Fission-product data for the events indicate that the close-in debris was not seriously fractionated. In some cases, the long-range results differed considerably from the close-in results; however, no clear-cut pattern of variability of isotopic ratios with distance from origin site is displayed by the data.

4.2 RARE EARTH ANALYSES

With the exception of Shot 3, which cannot be considered a representative thermonuclear event, the rare-earth ratios were relatively constant, even though the yield of the events ranged from 2 to 14 megatons. It appears, then, that the rare-earth ratios can be used only in a qualitative manner to indicate a thermonuclear event. For example, if the heavy-element data shows that plutonium fissioning was not a significant consideration,

However, the LRD Eu^{156}/Sm^{153}, Tb^{116}/Sm^{153}, and Tb^{161}/Eu^{156} ratios for Shot 1 are consistent with the close-in results within the limits of reliability of the data.

It was not possible to determine Gd^{159} on LRD samples because of its low fission yield and short half-life.

4.3 ISOTOPIC ANALYSES, HEAVY ELEMENTS

A comparison of close-in and LRD uranium data shows that with the exception of Shot 4 Guam LRD values, uranium-to-fission ratios agree as well as the internal uranium ratios. The low atom-to-fission ratios for Shot 4 Guam LRD values can perhaps be explained: Since collections were made at approximately 2,000 feet, the debris collected in this case may have been from the stem portion of the cloud. It is possible that the close proximity of the water resulted in more-rapid cooling of this portion of the cloud. Since molybdenum is among the first elements to condense, and total fissions are calculated from this isotope, low atom-to-fission ratios would be expected.

Fractionation of the internal uranium isotopic ratios would not be ordinarily expected, as the decay of these isotopes is not significant during the time required for cooling of the fireball. However, the LRD and close-in values for these ratios differ by as much as 15 percent in several instances. Since this difference could not be accounted for in terms of analytical error, it is possible that this apparent fractionation occurs during a carrier-free dissolution of close-in samples, at which time the U^{239} is determined by analyzing for its decay product, Np^{239}.

These unusually high values are indicative of the large fluxes of high-energy neutrons generated in the explosions. Additional evidence of the thermonuclear nature of these tests was the presence in the debris of such multiple neutron capture products; only thermonuclear events can supply the large neutron flux necessary for multiple n, γ reactions of this magnitude.

Large amounts of depleted uranium were placed in close proximity to the Castle devices, with the consequence that the uranium mass-spectrometric results cannot be interpreted in terms of bomb-reaction products. In fact, the U^{235} abundance in the mass-spectrometer samples is less than the normal abundance in natural uranium. In comparing mass-spectrometric close-in and LRD data, the U^{235} to U^{238} LRD ratios closely approach the natural abundance of these isotopes---thus demonstrating the contribution of atmospheric uranium background. For the events for which mass-spectrometric data is available, it is possible to account for all of the observed Pu^{239} as the decay product of U^{239}.

4.4 INDUCED ACTIVITIES

The isotope Mn^{54}, produced primarily by the (n,p) reaction on Fe^{54}, may be indicative of the amount of iron present in the device.

4.5 GAS ANALYSES

Although detectable quantities of gaseous radioactive isotopes of interest were measured in approximately all close-in gas samples, the results were disappointing, since there was extreme variation and inconsistencies in the proportionate concentrations of these isotopes throughout the samples obtained. Analysis of long-range gas samples were equally disappointing. In addition to the inconsistencies observed close-in, long-range samples were further complicated by lower concentrations of debris radioactive gases in the presence of significant background levels, particularly with respect to Kr^{85}, which was to be used as a fission reference.

The variability of radioactive gas atom ratios for the close-in samples is probably due to unrepresentative samples of the cloud, as all samples were taken at altitudes well below the altitude attained by the main cloud. There is no guarantee that this variability would be eliminated by sampling at 75,000 to 80,000 feet for the megaton shots, however, as there are still insufficient data with respect to fractionation of the debris gases either with respect to each other or with respect to the particulate portion of the cloud. The environment of the explosion---water or barge shots at Eniwetok and Bikini---carry such large quantities of water into the atmosphere that serious effects in attempting to get representative and quantitative tritium measurements under these conditions would be expected, particularly during the first twelve hours after detonation when rainout and/or fallout is very prevalent. This factor appears significant, as the extreme variations in H^3/Kr^{85} ratios are normally not observed in sampling shots at the Nevada Test Site---e.g., as observed during Upshot-Knothole. This comparison is not absolute, since megaton shots have never been fired at Nevada. However, during Operation Teapot, analysis of about ten shots gave gas atom ratios that were quite reasonable within theoretical expectations. From this latter fact, it was concluded that the sampling equipment and laboratory analysis for the close-in samples were not the principal sources of the unreasonable variations observed in the gas data. The long-range samples may be compromised within the sampling equipment itself, since it is known that recovery of tritium from the sample containers, quantitatively, is open to serious question. Hence, the overall comparison of close-in analyses with long-range analyses is not considered completely valid, because of the differences in the sampling equipment used. It is anticipated that for Operation Redwing, sampling equipment will be completely converted to Squeegee type---both close-in and at long-range.

In reviewing the data on an overall and qualitative basis, it appears that ratios for the Castle shots are significantly higher than those observed for Ivy-Mike. The range of values for Ivy-Mike is ratio all Castle shots indicate ratios greater in the majority of the samples analyzed. Theoretically, based on reactions in-

volved in both the Mike-type device [redacted] and Castle-type devices [redacted] larger quantity of H^3 left over or unburned in the Castle-type devices should be expected. This difference appears to be suggested by the data in a broad, qualitative sense.

A study of the H^3/Kr^{85} atom ratios determined experimentally shows an intolerable variation, with most values being unreasonably higher than theoretical expectations. Some results also appear to be too low. Many explanations can be offered for these variations, although none is completely satisfactory. The high values can be caused by tritium rain-out at time of sampling, while the low values could represent sampling immediately after rainout where the atmosphere may be momentarily scrubbed of the tritium. An attempt was made to correlate the H^3/Kr^{85} ratios with respect to time of sampling. A plot of this correlation is included for what it is worth in Figure 4.1. No specific conclusions can be drawn based on the data available.

The C^{14}/Kr^{85} ratio exhibits the same variation within samples collected from the same shot and throughout the entire test series. No consistent variations with altitude or sampling time are observed.

For each sample in which a successful separation and assay of a detectable amount of Kr^{85}, C^{14}, and H^3 were found, the C^{14} and H^3 formed per fission have been calculated. Only a small number of these computed ratios appear compatible with expected theoretical ratios computed for nuclear reactions of this type. No general observations resulted from an analysis of these ratios.

Average values of the C^{14}/A^{37} ratio calculated for each shot of the series are:

[redacted]

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indicates that the water or barge shots have ratio values which increase with yield. The very-low values of this ratio for the two coral island shots might be significant. If the reaction ($CA^{40}(n,d)A^{37}$) contributed any quantity of A^{37} to the nuclear cloud, such an effect would tend to lower the C^{14}/A^{37} ratio.

In Table 3.22, only those long-range data are shown in which isotopic concentrations are sufficiently high above background to warrant inclusion. No observations or correlations with close-in data are made.

4.6 PETROGRAPHIC ANALYSES

All shots resulted in the formation of microspheres; these particles represent the non-crystalline constituents and presumably include compounds from the bomb, fission products, bomb casing, and bomb support. All shots except Shot 6 resulted in collection of one or more of the following crystalline compounds: (oxide, hydroxide, and carbonate) of calcium, magnesium oxide, and sodium chloride. Shots 1 and 3 show only calcium compounds, indicating that little, if any, sea water was vaporized. Shots 2 and 4 show principally sodium chloride and magnesium oxide from sea water, although Shot 4 shows some calcium compounds, indicating that a small percentage of island material was vaporized in this shot.

Sodium and calcium compounds were absent as major constituents of the debris for Shots 5 and 6. It is significant, perhaps, that rain was reported subsequent to both tests, which may have resulted in the leaching of these compounds from these two events.

4.7 SPECIFIC BETA ACTIVITY

From a plot of the number of particles per unit logarithmic interval of disintegrations per minute divided by the cube of the particle diameter in microns, a modal value for specific beta activity can be obtained from the apparent normal distribution curve. The modal values for the Castle shots are only rough estimates since the observed frequency distributions covered a broad spectrum of specific activities with no pronounced peaks. Modal values for the barge shots were much greater than those from island shots.

Chapter 5
CONCLUSIONS and RECOMMENDATIONS

5.1 CONCLUSIONS

The most striking difference between the fission-product [redacted] obtained for the Castle tests occurs in the region of the trough of the fission yield curve. [redacted]

With the exception of Shot 3, [redacted] it can be shown from the heavy-element data that plutonium fissions were a negligible fraction of the total fissions in the Castle detonations.

The U^{237}/f ratios for the series clearly indicate that the events were of a thermonuclear nature. [redacted]

The presence of measurable amounts of the heavier plutonium isotopes such as Pu^{246} in the debris is evidence of the thermonuclear nature of the Castle events.

The isotope Pb^{203} was reported for Shots 3, 4, and 5, [redacted]

Only rough estimates could be made of the modal specific beta activity values, since the observed frequency distributions covered a broad spectrum of specific activities with no pronounced peaks. Modal values for the barge shots were much greater than those from island shots.

The [redacted] gas sampling system proved to be a satisfactory collection system, provided certain operational and maintenance techniques were employed in its use.

Radioactive gases of interest resulting from the explosions were detected close-in to the site of detonation. [redacted]

No correlations could be made between radioactive gas concentrations and the characteristics of the particular device under test due to the extreme variations of these quantities. The causes of these variations are not readily apparent, but may be due to non-representativeness of samples, fractionation of gaseous debris, cross-contamination of an inadvertent nature in the sampling equipment or in laboratory analysis.

5.2 RECOMMENDATIONS

All future U. S. nuclear tests should be monitored employing present

detection and collection techniques, and expanding the techniques where improvements can be obtained.

Whenever possible, all close-in calibration data should be correlated with identical measurements of samples collected at locations remote from the test site, in order to simulate long-range sample conditions that would be expected from debris collections of a foreign nuclear explosion.

Sampling for particulate and gaseous debris in tests of thermonuclear magnitude should be conducted in the stratosphere, in order to obtain representative samples.

Laboratory and processing techniques should be improved and developed to the point where cross contamination between gaseous debris samples is negligible.

Certain of the physical studies, i.e., petrographic studies, X-ray diffraction, etc., should be continued to explore possible effects useful for diagnostic studies.

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