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Radiochemical, Chemical, and Physical Analysis
of Atomic Bomb Debris

Project 7.3 (Buster), Project 7.1 (Jangle)

Armed Forces Special Weapons Project
Washington, D.C.

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October—November 1951

NOTICE

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"Radiochemical, Chemical and Physical Analysis of Atomic
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Director

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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.

This report has been reproduced directly from available copies of the original material. The locations from which material has been deleted is generally obvious by the spacings and "holes" in the text. Thus the context of the material deleted is identified to assist the reader in the determination of whether the deleted information is germane to his study.

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.

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ACKNOWLEDGMENTS

The National Institute of Health has made a radiochemical study of soil samples obtained in and near the bomb craters of the JANGLE shots. The results, submitted by Dr. Maxwell of National Institute of Health, were of considerable value in supporting some of the tentative conclusions based on radiochemical analyses. It is understood that the National Institute of Health is planning to submit a detailed report of their analytical work.

The fall-out samples mentioned in Table 5 of this report were made available by the National Institute of Health.

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ABSTRACT

Radiochemical analyses of atomic bomb debris were made on samples collected from the atomic bomb explosions of Operations BUSTER and JANGLE (October and November, 1951). Samples were collected close-in to detonation point and about 1200 miles away for the entire series; in addition, samples were also taken at several altitudes for the JANGLE shots. Fission product activity ratios were obtained for all BUSTER shots. The BUSTER Baker shot offered the first opportunity to obtain important calibration ratios

Throughout the BUSTER series, apparent significant differences were noted between individual analyses of the close-in samples and constitute indications of some fractionation. The average of these individual analyses, however, indicated fission product ratios which were in good agreement with the expected values. More pronounced differences were also noted between individual analyses of long-range samples and constituted further indication of fractionation. Unfortunately, the number of long-range samples analyzed was too few to support a real comparison with the close-in samples. Extreme variation in fission product ratios were noted during the JANGLE series; in general, it appeared that fractionation (non-homogeneity of sample) increased with increasing altitude and distance from the point of detonation. The variability in the measured ratios for the underground shot was more pronounced than the surface shot. A study of all the fission product ratios of the JANGLE measurements indicates that the Mo^{99} may be one of the isotopes most susceptible to the fractionation. The wide variation in fission product activity ratios is indicative of a surface or underground atomic detonation.

Data are included in this report reviewing all the fission product calibration data available to date. Key fission product activity ratios of bomb debris from all United States atomic bomb explosions to date are summarized. It is recommended that greater emphasis be placed upon long-range sampling in future atomic tests.

1.0 OBJECTIVE

The objectives of this project included the accumulation of the maximum quantity, within practical accomplishments, of nuclear and other pertinent data related to debris collected from the different types of explosions in the BUSTER and JANGLE tests. These data are then used as calibration points or as reference points in evaluating collected debris resulting from a foreign atomic explosion with respect to the composition and type of atomic bomb exploded and in general afford a technical means of interpreting and assessing foreign developments in the nuclear field of science and their military applications.

This project of the BUSTER-JANGLE tests indirectly contributed to the knowledge of the military effects of these particular explosions by indicating the relative specific activity of atomic explosion debris resulting from air bursts and underground bursts.

2.0 HISTORICAL BACKGROUND

Theoretically, by thorough and complete radiochemical, chemical and physical analyses of atomic bomb debris, it is possible by astute interpretation of the data obtained to determine the nature of the fissionable material used in the bomb, the efficiency and utilization of the fissionable material, the date of the explosion, the presence of uranium as the tamper material and the conditions of the test such as an air burst, surface or underground shot.

Radiochemical analyses were first employed as tools in determining characteristic properties of atomic explosions when the efficiency of the utilization of the fissionable material and the capture-to-fission ratios were measured during the TRINITY operations, the first A-bomb explosion. Similar analyses have been made on all subsequent atomic explosions in the United States. During Operation SANDSTONE, a large-scale program was initiated in employing radiochemical analyses of atomic bomb debris to determine the practical aspects of deducing as much information as possible about a particular atomic explosion by this technique. The results obtained during the SANDSTONE tests were of a qualitative nature at that time but were sufficiently positive to warrant thorough investigations into the specialized techniques and calibrations needed to obtain sound and reliable quantitative radiochemical assays of the extremely minute amounts of atomic bomb debris collected by the various sampling techniques available.

In connection with Operations RANGER and GREENHOUSE, the radiochemical analyses and joint Los Alamos Scientific Laboratory - AFOAT research had been developed to the point where emphasis could be placed on the observation of departures from previously measured quantities as

an indication of a special feature of the explosion

Some work on particle studies were sufficiently far along to suggest data analyses looking for a relation between the particle size distribution and radiochemical composition and the properties of a particular type of atomic detonation.

The present project was designed to continue observations on the effect of changing one or more characteristics of an A-bomb on the nuclear properties of the debris and to study further the effect of the condition of the explosion; i.e., underground, tower, or air burst.

3.0 INSTRUMENTATION

No special instrumentation other than the regular airborne filter samplers was employed in the field. Laboratory equipment was for the most part standard radiochemical laboratory equipment including standard equipment for microscopy, petrography, and microchemistry. Some special counting equipment for extremely weak samples was developed. Any special instrumentation developed for this project will be described in detail in the contractors' annual report on this subject to be issued in June 1952.

4.0 OPERATIONS

4.1 General

The detailed aspects of the overall operations involved in the collection of the necessary samples for radiochemical analyses and related studies are reviewed in separate summary¹ and will not be discussed in this section. Relative sampling locations such as high and low altitude, close-in to bomb detonation point, etc. are included with the results reviewed in the attached tables.

In general, close-in samples were taken with B-29 aircraft equipped with type C-1 and A-1 filtering devices; some were taken by T-33 planes equipped with air-filtering devices (wing tank samplers); long-range samples (about 1200 miles) were taken with B-29 planes equipped with the type C-1 filtering devices.

¹"Sampling Tactics, Operation BUSTER", by H. F. Plank
Group V-11, Los Alamos Scientific Laboratory, 12 October 1951.

4.2 Participating Agencies

The principal contributor to the data summarized in this report was the Western Division of Tracerlab, Inc. Some particle work was done by the Boston office of Tracerlab.

A portion of the data of this report, namely the data in Table 6, was submitted by the U. S. Naval Radiation Defense Laboratory, San Francisco, California. The sampling program was conducted by the Los Alamos Scientific Laboratory - Atomic Energy Commission by agreement with AFOAT

No formal completed and final reports specifically devoted to the BUSTER-JANGLE tests have been submitted by the contractor to this office but will be included when such reports are submitted. The summaries submitted are compiled from the contractors' original data.

5.0 RESULTS

5.1 General; R Value Defined

In order that intercomparison could be made between the results obtained by the various laboratories participating, an agreement was reached to express the measured radiochemical ratio as an R value which is defined by the relation:

$$R_{12} = \frac{A_1/A_2}{(A_1/A_2)_c} = \frac{Y_1 e_1 \lambda_1 / Y_2 e_2 \lambda_2}{Y_{1c} e_1 \lambda_1 / Y_{2c} e_2 \lambda_2} = \frac{Y_1/Y_2}{(Y_1/Y_2)_c}$$

A_1/A_2 is the measured activity ratio for two fission products, the activities having been corrected for decay between the time of explosion and time of measurement. $(A_1/A_2)_c$ is the activity ratio of the same two fission products measured under the same laboratory conditions, but produced by the slow-neutron-irradiation of U^{235} . The combination of counting efficiencies and counting geometries (e) may vary widely between laboratories, but when the results are reported as defined above such effects cancel out. This requires, of course, that each laboratory determine $(A_1/A_2)_c$ by a detailed analysis of a slow-neutron-irradiated sample of U^{235} , but it eliminates the uncertainties of absolute beta-ray counting. A summary of the results obtained from the radiochemical analysis of the debris from the BUSTER-JANGLE series of tests is included in Tables 1 through 6, along with results obtained from other U. S. atomic bomb tests. These results are expressed as R values wherever possible. Ratios including elements other than fission products, specifically: U^{237} and Np^{239} , are expressed as activity ratios corrected

to zero time. The capture-to-fission ratios are expressed on an atom basis.

5.2 BUSTER Series

The average R values obtained from the analyses of the close-in samples are shown in Table 2 along with the same data obtained from other atomic bomb tests, and are indexed in accordance with the composition of the fissionable material used. The averages obtained for BUSTER Charlie, Dog and Easy agree reasonably well with those obtained from other composite bomb tests.

Table 3 repeats the average values for the fission product ratios obtained from the close-in samples, and in addition indicates the standard deviation observed over the samples analyzed. Table 4 is a summary comparison of the fission product activity ratios of samples collected close-in to the bomb detonation point and at distances approximately 1200 miles from the Nevada Test Site.

5.3 JANGLE Series

The wide variation observed in the measured fission product ratios for both the JANGLE shots makes the use of average values meaningless and therefore Table 2 shows the maximum range of values. It was possible, however, to find a trend in the data with respect to the location at which the samples were collected. Table 5 is a summary of the JANGLE R values for the key fission products measured in samples taken at the ground, close-in at low and high altitudes, and at long-range (about 1200 miles from the source).

The extreme variation found for the JANGLE samples was confirmed by analyses of bomb debris by personnel of the U. S. Naval Radiological Defense Laboratory; these data are reviewed in Table 6 and in general follow the same trend as indicated in Table 5.

5.4 Fission Product Activity Ratios, R Values, Calibration Data

Table 1 lists the measured significant fission product activity ratios expressed as R values for various reactions on U^{233} , U^{235} , U^{238} , and Pu^{239} .

Additional work is underway to confirm some values indicated in Table 1, to complete Table 1, and to extend the calibration data to other isotopes of particular interest in connection with thermonuclear reactions.

6.0 DISCUSSION

6.1 BUSTER Baker;

IR. W. Spence, Los Alamos Scientific Laboratory reported preliminary R values for

6.2 BUSTER Shots; Indication of Fractionation

The standard deviations listed in Table 2 are, in general, considerably larger than are inherent in the methods of analysis, and are interpreted to indicate that the atomic cloud was not a homogeneous mixture. The average number of independent determinations in each case was about six, so that too much emphasis cannot be placed upon the magnitude of the individual standard deviations. The consistency of the high standard deviations observed for the $\text{Sr}^{89}/\text{Mo}^{99}$ ratio, however, constitutes a strong indication of fractionation with respect to this particular ratio. Analyses of debris from the BUSTER Charlie and the BUSTER Easy shots generally indicate that more fractionation occurred with these shots than in the case of BUSTER Baker and BUSTER Dog.

Accepting the analyses for the BUSTER Charlie and BUSTER Easy, some interesting comparisons can be made.

Table 4 presents the analyses of close-in and distant samples to indicate the extent of possible fractionation effects with distance from the source. The number of independent determinations of distant samples, however, was seldom more than two; therefore, although differences do appear between close-in and long-range samples, they are not necessarily significant. It should be mentioned that there was an appreciable number of cases where two independent long-range samples

gave results that differed by more than a factor of two. Thus the number of samples studied is too few to permit any meaningful comparison between close-in and long-range samples. However, the averages of close-in and long-range samples do not exhibit differences of such magnitude as to give rise to completely false evaluations of the nuclear ratios of the weapons being tested.

6.3 The JANGLE Shots; Severe Fractionation Effects

From the data of Table 5 it is apparent that fractionation increased with increasing height of sampling and increasing distance from the point of explosion. The extent of fractionation was considerably more pronounced for the JANGLE Underground shot.

This observation is borne out by a review of the preliminary results of analyses of soil and other local samples by the National Institute of Health under Program 2, Operation JANGLE.

Conversely, there is a reasonable probability that Sr^{89} and Ba^{140} exist, at least partially, as Kr^{89} and Xe^{140} at the time of condensation and, since there is no tendency for these rare gases to condense with the other debris, it might be expected that Ba^{140} and Sr^{89} , under certain conditions of condensation, would be associated with the fine particles of the debris. Further, the half-life of Kr^{89} (2.6 minutes) is longer than that of Xe^{140} (16 seconds) so that there is a greater probability of Sr^{89} existing as its precursor, a rare gas, over a longer period of condensation. Thus one can reason that the Sr^{89} isotope would be associated with particles still finer than those associated with Ba^{140} .

A particularly important requirement exists for some fission product that does not fractionate with respect to neptunium. This would make possible consistent and accurate determinations of the capture-to-fission ratio. From the data in Table 5 it is obvious that Mo^{99} is not suitable in the case of surface and underground explosions since the capture-to-fission ratios calculated on the basis of Mo^{99} varied widely, and exceeded theoretical limits. Unfortunately, none of the isotopes studied was satisfactory in this respect.

The JANGLE tests offered an excellent opportunity to observe that the $\text{U}^{237}/\text{Np}^{239}$ ratio remains unaffected even in cases involving severe fractionation. The measurement of this activity ratio was consistent throughout with the normal value, as to be expected, since the precursor of neptunium is undoubtedly uranium at the time of condensation.

7.0 CONCLUSIONS

Variations observed between individual determinations of the R values indicated that fractionation accompanied the BUSTER shots to some extent and this is particularly noticeable for the BUSTER Charlie and BUSTER Easy shots. However, in the case of air bursts, the variation of the fission product ratios from expected values and between samples is not of such magnitude as to invalidate completely the evaluation of the nuclear nature of the weapon being tested.

Extreme variation occurs in the fission product activity ratios for the two JANGLE shots. This variation is attributed to the location of the shot; one on the surface of the ground and the other underground.

In general, the fractionation in the JANGLE filter samples appeared to increase with increasing altitude and increasing distance from the point of detonation.

The fractionation observed was more pronounced for the underground than for the surface shot.

The JANGLE shots showed that the capture-to-fission ratio, when based on Mo^{99} , varied widely and did not agree with the expected value. None of the fission products studied gave the expected capture-to-fission ratio.

Extreme fractionation of the fission products is indicative of a surface or underground explosion.

8.0 RECOMMENDATIONS

It is recommended that all future atomic bomb tests continue to be monitored by the current known techniques. Additional calibration data for a pure plutonium weapon with no uranium tamper is desirable.

The ever-present possibility of fractionation dictates the need for more extensive sampling, particularly at long-range.

It is apparent that more emphasis must be placed in the future on the analysis of samples for the residual fissionable materials and the products of various reactions such as n, γ and $n, 2n$ reactions

Such analyses have been strictly limited in the past because of the small size of samples obtainable at distance and the limitations of analytical techniques. Recently, it has been possible to obtain larger samples at long-range and to extend the observational limits of the analytical procedures so that the capability to study the residual fissionable materials and reaction products should exist in the near future.

TABLE 1

Fission Product Activity Ratios - Calibration Data

R Values

Reaction	$\frac{95}{Zr} \frac{99}{Mo}$	$\frac{103}{Ru} \frac{99}{Mo}$	$\frac{106}{Ru} \frac{103}{Ru}$	$\frac{111}{Ag} \frac{99}{Mo}$	$\frac{115}{Cd} \frac{99}{Mo}$	$\frac{140}{Ba} \frac{99}{Mo}$	$\frac{144}{Ce} \frac{99}{Mo}$	$\frac{111}{Ag} \frac{115}{Cd}$
U^{235} , fast - n	1.1	1.0	1.1	1.6	1.5(2)	1.0	0.9	1.06
U^{235} , capsule - n	-	-	-	3.8(1)	4.1(2)	-	-	0.92
U, normal, fast - n	-	-	2.5	2.6	-	-	-	-
U, normal, capsule - n	-	-	-	4.2(2)	-	-	-	-
U^{238} , fast n	1.0	1.3	4.5	4.6	-	1.1	1.0	-
U^{238} , capsule n	-	-	-	4.2(2)	3.2(2)	-	-	-
Pu^{239} , thermal n	0.85	1.45	5.2	15.8	3.2(2)	0.9	0.7	4.9
Pu^{239} , fast n	0.85	1.7	5.5	19.2	4.3(2)	0.9	0.7	4.4
Pu^{239} , capsule n	0.80	1.6	6.8	25.0	7.1(2)	1.1	-	3.5
U^{233} , thermal n(1)	1.25	0.3	2.0	1.6	1.9	1.3	0.85	0.84
U^{233} , fast n	1.3	0.5	1.8	3.3	-	-	1.0	-

(1) Values based on published yield data taken from NNES, Coryell and Sugarman, McGraw-Hill, 1951.

(2) Values given by R. W. Spence, Los Alamos based on counting rate ratios.