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USNRDL-TR-67-90 (EX)

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FALLOUT FROM NUCLEAR CRATERING SHOT DANNY BOY

I. Radiochemical Analysis and Some Physical Observations on Selected Samples

U. S. Naval Radiological Defense Laboratory
San Francisco, California

AD B951703

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REPORT DOCUMENTATION PAGE

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1. REPORT NUMBER: USNRDL-TR-67-90 (EX)
2. GOVT ACCESSION NO.: B951703
3. RECIPIENT'S CATALOG NUMBER:
4. TITLE (and Subtitle): Fallout From Nuclear Cratering Shot DANNY BOY. I. Radiochemical Analysis & Some Physical Observations on Selected Samples
5. TYPE OF REPORT & PERIOD COVERED:
6. PERFORMING ORG. REPORT NUMBER:
7. AUTHOR(s): Francis K. Kawahara, Edward C. Freiling, Leland R. Bunney, Glenn R. Crocker
8. CONTRACT OR GRANT NUMBER(s):
9. PERFORMING ORGANIZATION NAME AND ADDRESS: U.S. Naval Radiological Defense Laboratory, San Francisco, California
10. PROGRAM ELEMENT, PROJECT, TASK AREA & WORK UNIT NUMBERS:
11. CONTROLLING OFFICE NAME AND ADDRESS:
12. REPORT DATE: 2 October 1967
13. NUMBER OF PAGES: 36
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): Approved for public release; unlimited distribution.

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

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19. KEY WORDS (Continue on reverse side if necessary and identify by block number):
Danny Boy
Fallout
Radiochemistry
Debris
Particles

20. ABSTRACT (Continue on reverse side if necessary and identify by block number):
Radiochemical analyses were made on some samples of close-in fallout from the Danny Boy cratering event and some observations were made of physical properties of the particulate material. Contrary to observations on land-surface bursts, these samples were enriched in volatilely-behaving mass chains. Otherwise, the fractionation of the fission-product radionuclides was found to follow the same pattern which has been observed in land-surface bursts: Zr^{95}, Mo^{99} and Ce^{144} did not fractionate from each other; Sr^{89} and Cs^{137} fractionated strongly from Zr^{95}, while Sr^{90}, Te^{132}, Cs^{136} and Ba^{140} showed intermediate fractionation; particles smaller than 44 μ. The density of the radioactive particles ranged from 1.6 to 2.1 g/cm³. Most of the particles observed were irregularly shaped and

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most were light yellow in color with black inclusions on the surface. About a third of the radioactive particles studied were magnetic.

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per telecon w/Betty Fox (DNA Tech Libr, Chief), the classified references contained herein may remain.

R. LaChance (DDA-2
9-5-79

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

Radiochemical analyses were made on some samples of close-in fallout from the Danny Boy cratering event and some observations were made of physical properties of the particulate material. Contrary to observations on land-surface bursts, these samples were enriched in volatilely-behaving mass chains. Otherwise, the fractionation of the fission-product radionuclides was found to follow the same pattern which has been observed in land-surface bursts: Zr^{95}, Mo^{99} and Ce^{144} did not fractionate from each other; Sr^{89} and Cs^{137} fractionated strongly from Zr^{95}, while Sr^{90}, Te^{132}, Cs^{136} and Ba^{140} showed intermediate fractionation; particles smaller than 44 μ were more highly enriched in volatilely behaving mass chains than particles larger than 44 μ. The density of the radioactive particles ranged from 1.6 to 2.1 g/cm^3. Most of the particles observed were irregularly shaped and most were light yellow in color with black inclusions on the surface. About a third of the radioactive particles studied were magnetic.

SUMMARY

A limited number of close-in fallout samples from Danny Boy, a cratering event in basalt, were analyzed radiochemically for some important fission products. Observations of the chemical composition and of some of the physical properties of the fallout particles were also made. The samples showed strong fractionation of the fission products, being enriched in the volatile mass chains relative to the refractory mass chains. This condition is the reverse of that usually observed in close-in fallout from surface bursts, but the relative volatility of the volatile and intermediate mass chains remained the same.

INTRODUCTION

The Danny Boy event, conducted 5 March 1962 at the Nevada Test Site, was an underground nuclear explosion which produced a crater in basalt. The device, which was buried 33.5 m below the surface, had an explosive yield of 0.43 kt. The crater which resulted from the event was 18.9 m deep and 65.2 m in diameter. The fallout cloud which formed reached a height of 300 m and deposited fallout of high activity for several miles downwind. Studies of the crater and of the radiation survey made of the fallout field were reported by Nordyke and Wray.[1] Radiochemical analyses of samples of the cloud and of the deposited fallout were made by Bonner and Miskel,[2] in a study designed to estimate the partition of the fission products produced by the device. The U. S. Naval Radiological Defense Laboratory (NRDL) also received samples of fallout from Danny Boy which were used for radiochemical fractionation studies as well as for studies of the physical properties of the particles. This report contains the results of the NRDL studies, except for the gamma-ray spectra. The latter will be analyzed in the light of newly developed techniques and calibration data currently being accumulated. The spectra and the quantitative data extracted from them will form the subject of a second report.

DESCRIPTION OF SAMPLES

Five samples from the Danny Boy event were received by NRDL. Four of these were fallout samples collected by the University of California at Los Angeles (UCLA) School of Medicine, and the fifth was a soil sample collected by the Nuclear Defense Laboratory (NDL) in the area near the crater lip.

The UCLA collector and processing procedure have been described by Romney, et al.[3] The collector consisted of a 29-in. x 43-in. x 5/8-in. metal tray (Boyco Auto Drip Pan) which was divided by taping of a 1/2-in. x 1/2-in. x 28-in. wooden bar to the center of the tray. Each half of the tray was lined with 0.5-mil Mylar film and contained approximately

1.8 kg of polyethylene granules, approximately 1/8-in. to 3/16-in. in diameter, which formed a matrix for trapping the incident fallout debris.

Three of the samples collected by UCLA were received by NRDL in the processed condition. The fallout material had been separated from the polyethylene granules according to the following procedure: The Mylar film was gathered up and formed into a bag; isopropyl alcohol was added to the bag and the wet granules were transferred to a washer assembly, which subjected them to mechanical stirring and vibration for one minute. The isopropyl alcohol with the suspended fallout debris was then drained into a 6-liter enameled pot through a 3-in. diameter weighed sieve that retained all particles greater than 44 μ. This operation was repeated until no more than 10 % of the original radioactivity remained in the matrix. The material retained on the sieve was washed again with a pressure spray of isopropyl alcohol, dried under a heat lamp, weighed, and radioassayed for gamma activity. The material smaller than 44 μ remaining in the drain pot was then collected by passage of the liquid suspension through a weighed Millipore filter. This also was heat-dried, weighed, and radioassayed for gamma activity. Each processed granular-collector fallout sample received from UCLA thus consisted of two size fractions: > 44 μ and < 44 μ. The processed samples were designated Y-24, X-24 and T-18, according to the stations in the fallout field (see Fig. 1) at which they were collected.

One unprocessed, granular-collector fallout sample was also received from UCLA. This sample, designated W-10, consisted of the Mylar bag, the polyethylene granules and the trapped fallout material.

The sample received from NDL, designated DB-1, consisted of surface soil collected from an area 5-in. x 10-in. on frozen ground at the base of the crater lip to the northeast of ground zero. This sample was one of two collected for another investigation at this Laboratory[4] on particle-size distribution, leaching, desorption and readsorption. Sample DB-1 was given to this project for gamma-ray pulse-height distribution and radiochemical studies. When received, the sample was contained in 6 volumetric flasks and 10 test tubes. The material in the test tubes (designated TT) had been collected on various Tyler sieves by wet sieving. That in the flasks (designated VF) contained material of different particle-size fractions (less than 44 μ) obtained through a wet sedimentation technique (described in Ref. 4). The technique is such that each fraction contains particles of all sizes up to an indicated maximum.

Table 1 summarizes the sample fractions used in this project according to designation, micron size range, and the types of analysis to which each fraction was subjected.

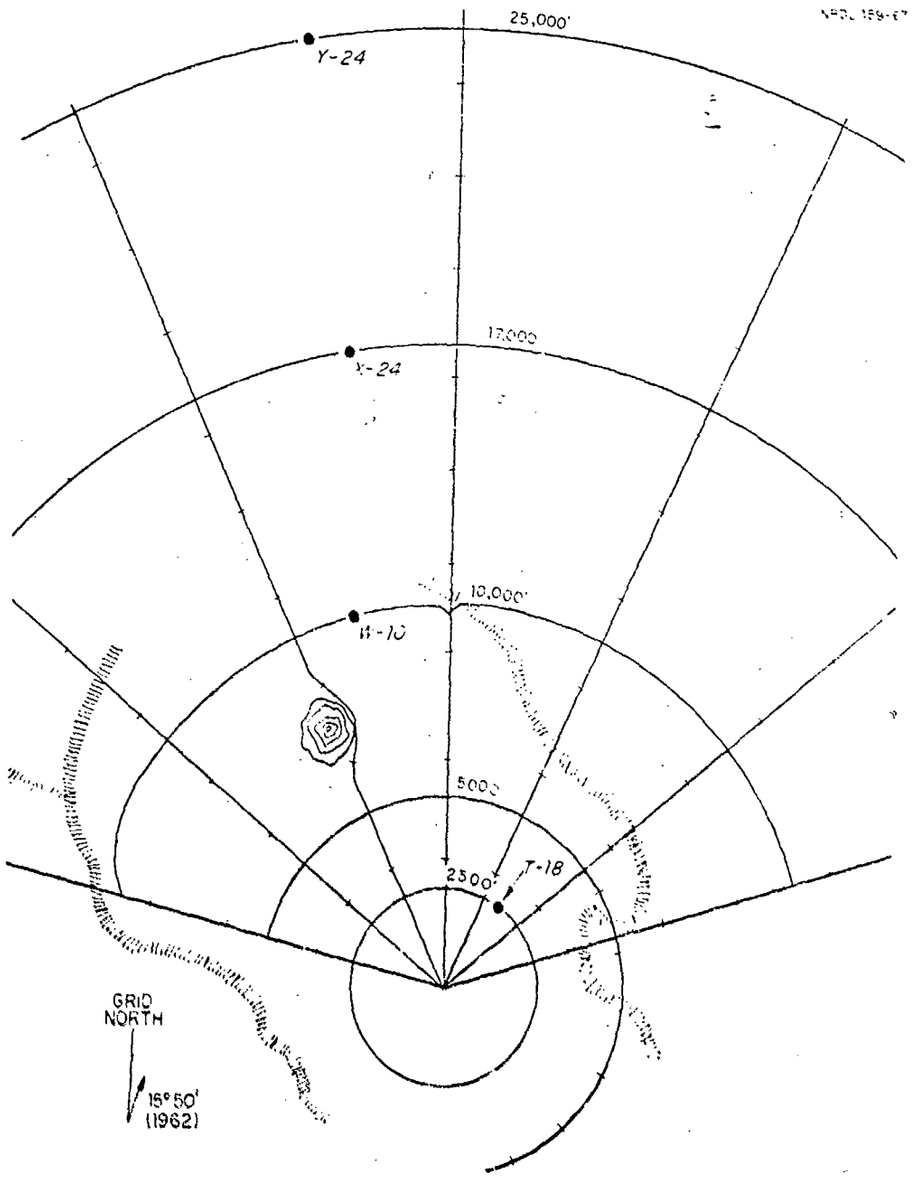

Fig. 1 Schematic Map of the Danny Boy Collecting Stations.

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TABLE 1

Samples Received From Event Danny Boy

Sample Designation	Size Range (μ)	Type of Analysis[a]
	UCLA Granular Collector Samples	
Y-24a	> 44	Radiochemical
Y-24a (1-4)[c]	< 44	Radiochemical
X-24a	> 44	Radiochemical
X-24a (1-4)	< 44	Size distribution, and radiochemical
T-18a	> 44	Radiochemical
T-18a (1-4)	< 44	Radiochemical
W-10	Gross	Size distribution, density, electron microbeam probe analysis, ratio of active to inactive particles.
	NDL Soil Sample DB-1	
TT-1	colloid	Decay[b]
TT-2	< 44	Decay[b]
TT-3	44-62	Radiochemical
TT-4	44-62	Decay[b]
TT-5	62-74	Decay[b]
TT-6	74-149	Decay[b]
TT-7	1190-2380	Decay[b]
TT-8	2380-4760	Decay[b]
TT-9	> 4760	Radiochemical
TT-10	149-1190	Decay[b]
VF-1	0-30	Radiochemical
VF-2	0-20	Decay[b]
VF-3	0-10	Decay[b]
VF-4	0-5	Decay[b]
VF-5	0-3	Decay[b]
VF-6	0-1	Radiochemical

a. In addition to the analyses listed, gamma-ray pulse-height distributions were determined on all samples.

b. Gamma-ray decay measured on a 4-pi ionization chamber. In the light of radiochemical results these samples were subsequently judged to be perturbed, and the data are therefore not reported.

c. (1-4) indicates the number of isopropyl alcohol washes that were combined to make the sample.

EXPERIMENTAL PROCEDURES

Radiochemical Analyses

All radiochemical analyses were performed by Tracerlab/West (TLW) according to standard radiochemical procedures described in Appendix A. All results were reported in terms of the number of U^{235} thermal-neutron fissions which would have produced the activity found in the total sample analyzed. For the purposes of this report, these values were converted into equivalent fissions of the device per sample in the manner described in Appendix B. The equivalent device-fissions for any mass chain in a given sample is the number of atoms in the device which underwent fission in order to produce the quantity of the mass chain observed in the sample.

Sample Treatment Prior to Physical and Chemical Property Observations

The X-24a(1-4) sample fraction, the < 44 μ fraction of one of the processed samples from UCLA, was used by TLW to determine whether the successive isopropyl-alcohol washings in the processing technique caused a significant change in the particle-size distribution. The sample fraction had been washed and filtered four times by UCLA. The particle-size distributions were obtained by smearing of some particles from each of the four filter papers on to microscope slides. The slides were examined under a 430X microscope and the diameter of each particle detected was measured.

Sample W-10, the unprocessed sample from UCLA, was used for a number of physical and chemical property determinations. The sample consisted of fallout material along with 1.8 kg of polyethylene granules. The fallout was separated from the granules by dry sieving in a Tyler sieving unit. The sieving unit had a 20-mesh screen (833-μ opening) on top to retain the granules but not the particles. Three other screens, with 147-μ, 104-μ, and 44-μ openings, were used. Particles retained on each sieve were brushed onto aluminum foil, and samples for further investigations were selected from these separations. The material was used by TLW for determination of the ratio of the number of magnetic to non-magnetic particles, the ratio of the number of radioactive to non-radioactive particles, the densities of selected particles, and the chemical composition of so of the particles as determined by electron-microbeam probe analysis.

Physical and Chemical Property Observations at TLW

For separation of magnetic from non-magnetic particles, the particles were arranged in a 1-in. ring on a Petri dish. A magnetic stirrer under the Petri dish agitated the particles and attracted the magnetic ones to the center. The magnetic particles were removed from the center with a stainless steel rod to which they were attracted. A brush was used for removal of the particles from the rod. The separated magnetic and non-magnetic particles were then counted.

The radioactive particles were located by radioautography with Kodak Blue Brand X-ray film. The number of inactive particles in an area approximately 900 microns square around the radioactive particles was then counted under 100X magnification. While under the microscope, the radioactive particles were also classified by color, shape, and size.

The density of a few selected particles was measured by a sedimentation method based on Stokes' law. Since streamline flow could hardly occur with irregularly-shaped particles, Stokes' law does not strictly apply, but it does present a rough estimate of the true value. Particles were cleaned in acetone and dropped through a 12-in. pyrex tube filled with trichloroethylene. A white light shining down from the top illuminated the particle (Rayleigh scattering) so that its terminal velocity could be measured visually without a microscope.

Electron-microbeam probe analysis was used for determination of the chemical composition of six selected particles, one magnetic and five non-magnetic. The American Metals Research Corporation instrument was used. The selected particles were embedded in plastic and hand-polished. After removal from the plastic by application of heat and with the aid of a pick, they were placed on a beryllium disc and coated with a light layer of carbon by vacuum deposition. Because of particle porosity and rounded surfaces (the latter created by the hand polishing), the operators of the electron microbeam probe analyzer were forced to scan the particles for usable points.

RESULTS

Radiochemical Data

Table 2 presents the results of radiochemical analyses of three of the granular collector samples, Y-24, X-24 and T-18. The $< 44\ \mu$ fraction and the $> 44\ \mu$ fraction of each of these samples were analyzed

separately. The data are presented in terms of equivalent fissions of the device per sample (see Appendix B), and are averages of duplicate determinations unless otherwise noted. The percent standard deviation is shown in the table only when calculated to be greater than $\pm$ 2 %. In those cases where only a single value is reported (footnotes b, d, and h), the standard counting error is reported if greater than $\pm$ 2 %.

The radiochemical procedures utilized and the nuclear constants used in reducing the data are presented in Appendix A. Comments by the contractor (TLW) on the radiochemical analyses are included.

Table 3 presents the radiochemical results in terms of ratios of the type $r_{i,95}$, described in Appendix B.

Particle-Size Distributions After Successive Washings

Figure 2 shows the particle-size distributions of the < 44 μ sample fraction designated X-24a(1-4), obtained after each of four successive isopropyl-alcohol washings. The distributions were obtained by microscopic examination of smears from the filter papers. The largest dimension of the particles was estimated to the nearest micron, by use of a reticle with 5 μ rulings. The estimated error was of the order of a micron. The distributions may be considered representative but not very accurate, since the number of particles measured for each sample was rather small.

Individual Particle Studies

Table 4 shows the size distribution of the magnetic and non-magnetic radioactive particles from sample W-10, the unprocessed granular collector sample. Fifty-four radioactive particles were found in this sample.

In white transmitted light most of the non-magnetic radioactive particles were light yellow, with small black spots included or fused to the surface. Of all the radioactive particles found, only one had a rather regular shape. It was medium-sized, light brown, and oval. Six of the 54 particles had a decidedly sponge-like appearance. Large internal bubbles and hemispheric surface depressions were characteristic of this group. One particle had long needle-like bubbles inside it.

The magnetic particles also fell into two groups. The first, the large magnetic particles, had inclusions similar to those in the first group of non-magnetic particles, but the black inclusions were significantly larger. The second group of magnetic particles, all less than 40 μ, were black and had very little radioactivity.

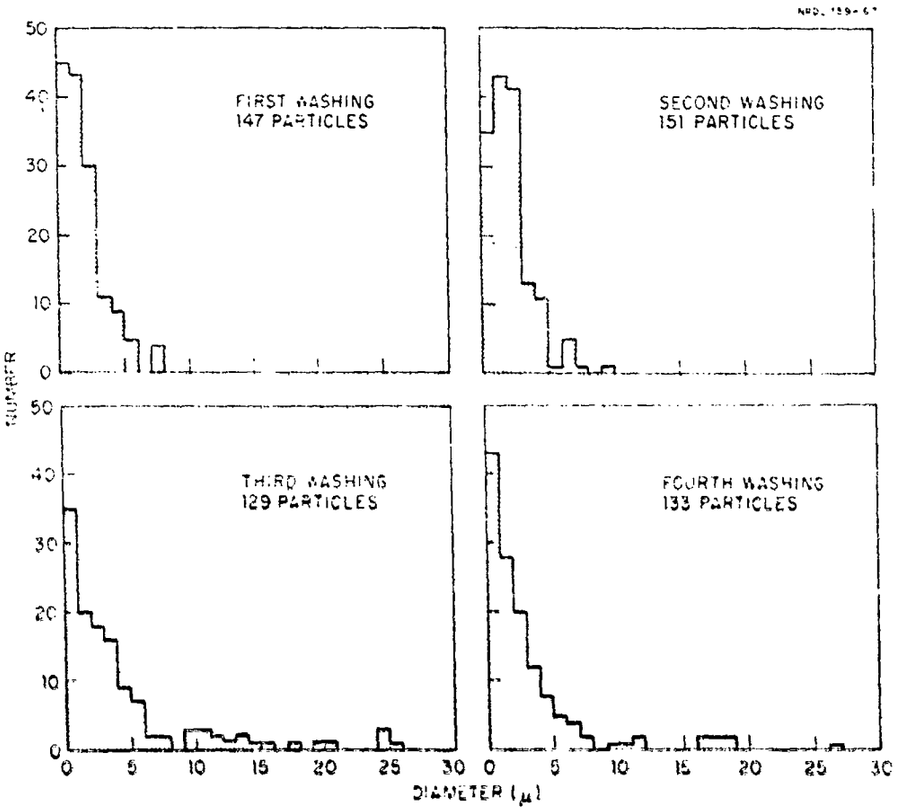

Fig. 2 Particle Size Distributions for Sample Fraction X-24a(1-4) After Each of Four Successive Washings with Isopropyl Alcohol.

TABLE 4

Size Distribution of Radioactive Particles from Sample W-10

Size Range (μ)	Number of Non-magnetic Particles	Number of Magnetic Particles
> 250	7	2
150-250	9	0
100-150	10	3
50-100	12	1
< 50	0	10
Totals	38	16

Density Measurements

The particle densities were all relatively low, even those for particles having large black inclusions. No particle density was greater than 2.1 and most were about 1.8. The density of the one oval shaped magnetic particle was 1.77. Translucent silica has a density of 2.07, but the particles bore scant resemblance to marine sand. However, the density of basalt is between 2.5 and 3.1, so the particles are probably closer to silica. It is possible that the presence of very small interior bubbles could have made the density measurements misleading.

Chemical Composition by Electron Microbeam Probe Analysis

The chemical composition results are shown in Table 5. The table shows the percent composition for Ca, Mg, Al, Si, and Fe, expressed both as elements and as oxides. The last column shows the sum of the oxides. The deviation of these values from 100 % indicates the unsuitability of the particles, because of their porosity and the rounded edges caused by hand polishing, to this kind of analysis. Particles A, B, C, D, and E had the distinctive sponge-like appearance, while Particle F contained many colored spots in a clear matrix. Within the limitations of their reliability, the data indicate the particles have compositions resembling basalts.[5]

TABLE 5

Chemical Composition by Electron Microbeam Probe Analysis

	Percent Composition										Sum of Oxides (%)
	Ca	CaO	Mg	MgO	Al	Al_2O_3	Si	SiO_2	Fe	Fe_2O_3	
Particle A: non-magnetic, light yellow, 151μ-166μ.											
Peak 1	3.3	4.6	2.2	3.6	13.5	25.6	28.5	61.0	5.7	8.1	102.9
Peak 2	3.3	4.6	5.5	9.1	14.5	27.5	31.0	66.4	6.5	9.3	117.9
Peak 3	1.5	2.1	2.0	3.3	5.5	10.4	18.5	39.6	5.5	7.9	63.3
Particle B: non-magnetic, light yellow with various colored spots, 182μ-185μ.											
	1.5	2.1	-	-	15.7	29.8	37.3	79.6	4.3	6.1	117.6
Particle C: magnetic, light yellow with large black spots, 264μ-288μ.											
Peak 1	7.0	9.8	2.0	3.3	4.5	8.5	35.0	74.8	7.4	10.6	107.0
Peak 2	9.2	12.9	2.5	4.1	9.5	18.0	13.6	29.1	8.6	12.3	76.7
Peak 3	9.0	12.6	2.5	4.1	4.5	8.6	14.6	31.0	7.9	11.3	67.6
Particle D: non-magnetic, light yellow, 174μ-189μ.											
Peak 1	7.0	9.8	4.0	6.6	15.0	28.5	34.5	73.7	3.6	5.1	123.7
Peak 2	3.6	5.0	2.0	3.3	17.0	32.2	33.8	72.3	5.7	8.1	120.9
Peak 3	4.0	5.6	1.5	2.5	17.3	32.8	29.0	62.0	2.6	3.7	106.6
Particle E: non-magnetic, light yellow with a black spot, 177μ-315μ.											
Peak 1	6.0	8.4	5.1	8.4	9.0	17.1	35.0	74.8	10.8	15.4	114.1
Peak 2	3.0	4.2	2.0	3.3	9.2	17.5	14.0	30.0	15.2	21.8	76.8
Particle F: non-magnetic, light yellow with many black spots, 166μ-359μ.											
Peak 1	4.5	6.3	17.0	28.1	17.8	33.6	35.0	74.9	18.9	27.1	170.0
Peak 2	10.4	14.5	10.2	16.9	3.0	5.7	18.5	39.6	15.9	22.8	99.5

DISCUSSION

Correlation of Radiochemical Results

The kind of radionuclide fractionation observed in the local fallout from the Danny Boy event was different from that shown by the data on local fallout from near-surface bursts correlated by Freiling[6] and by Crocker.[7,8] The samples from the near-surface bursts were depleted in volatile mass chains, such as mass 89 and mass 137, relative to mass 95. In the Danny Boy samples, on the other hand, enrichment in the volatile mass chains relative to the mass-95 chain was usually observed. However, in both types of event the smaller particles were found to be the more highly enriched in the volatilely behaving nuclides. The probable explanation is that the refractory mass-95 chain (along with the other refractory chains) condenses so quickly that very little of it escapes from the immediate area of the crater. Bonner and Miskel estimated, from the analysis of cloud samples and close-in fallout samples from Danny Boy, that only a few tenths of 1 % of the refractory mass chains escaped into local and worldwide fallout, but that 10 to 20 % of the volatile mass chains escaped.

The radiochemical data on the Danny Boy samples were correlated according to the log-log plotting method of Freiling,[6] which is described in Appendix C. The correlation offered an opportunity of comparing the NRDL radiochemical data with the results reported by Bonner and Miskel for ground-collected close-in samples. The latter appear to be companion samples to the NRDL granular-collector samples, and were probably obtained from the same source. The two sets of radiochemical results are compatible, since they follow the same correlation lines, as shown in Figs. 3 to 8.

It seems clear from the figures that the radiochemical data on the fractions of the soil sample, DB-1, cannot be correlated with the data on the granular-collector samples and the samples of Bonner and Miskel. It is believed that the wet sieving process, to which sample DB-1 was subjected, resulted in leaching appreciable amounts of some activities from the debris. Since the DB-1 data points for Sr^{90}, Ce^{141}, Ba^{140}, and Cs^{136} lie above the correlation lines, it is necessary to postulate preferential loss of Sr^{89} in the leaching. In the case of Cs^{137}, where the DB-1 points correlate with those for the other samples, it is possible that a compensating loss of Cs^{137} occurred. Note that the foregoing explanation of the DB-1 data points requires the assumption that Sr^{90} differs from Sr^{89} and that Cs^{136} differs from Cs^{137} in leachability. Such an assumption is not tenable on chemical grounds, but would be justified if the processes by which the radionuclides were incorporated

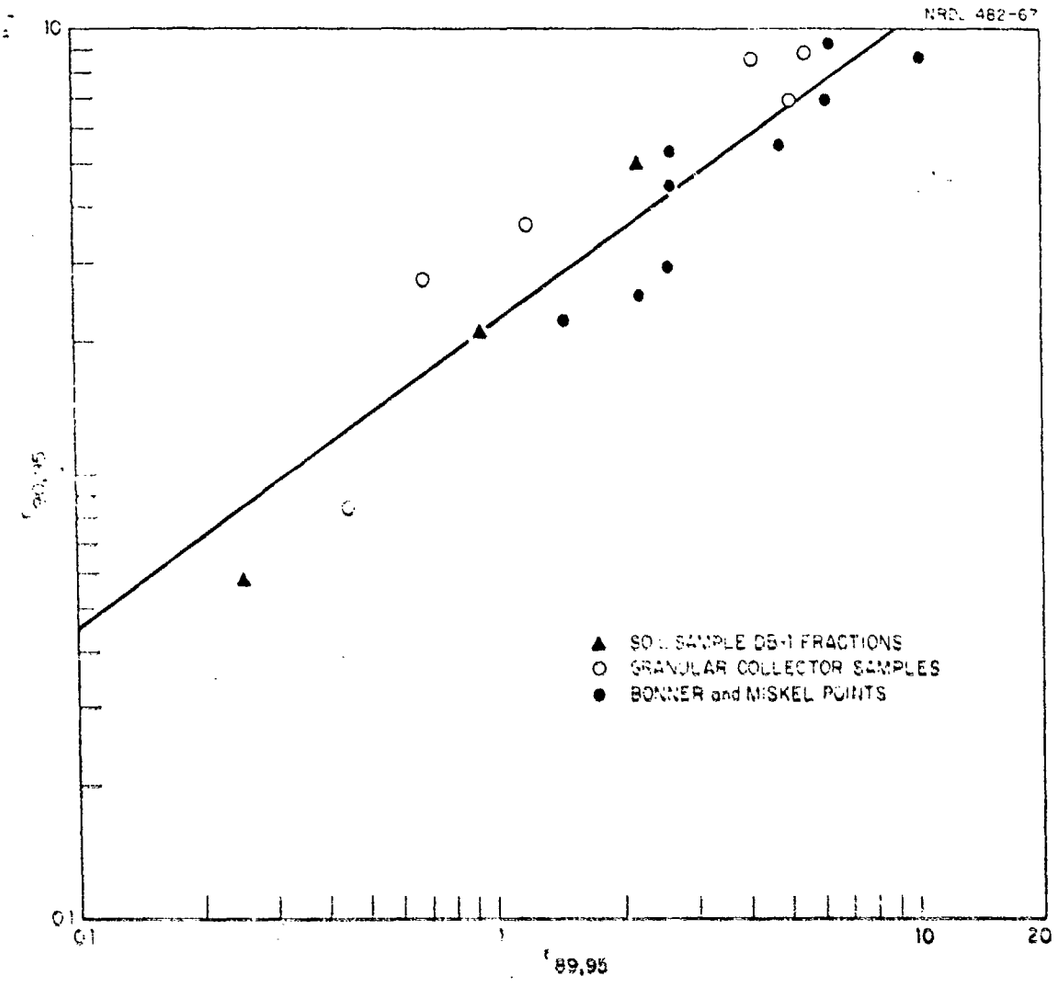

Fig. 3 Fractionation Correlation Plot for Sr^{90} Data from Danny Boy Samples.

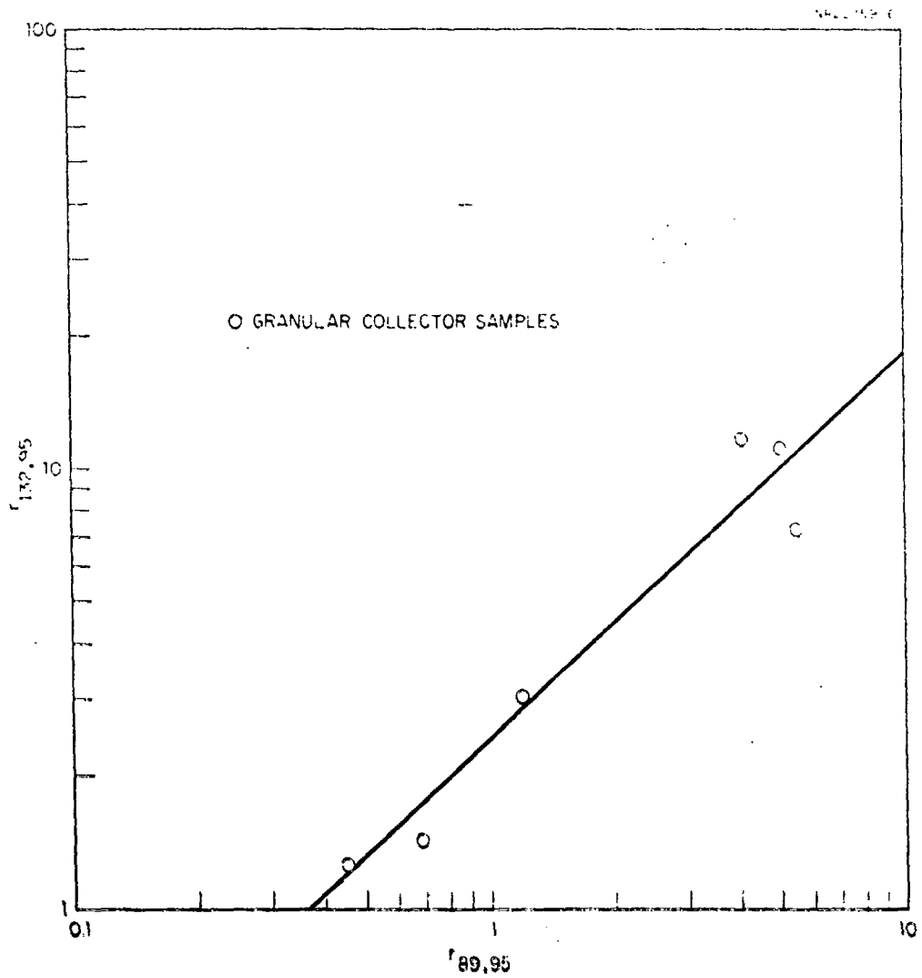

Fig. 4 Fractionation Correlation Plot for Te^{132} Data From Danny Boy Samples.

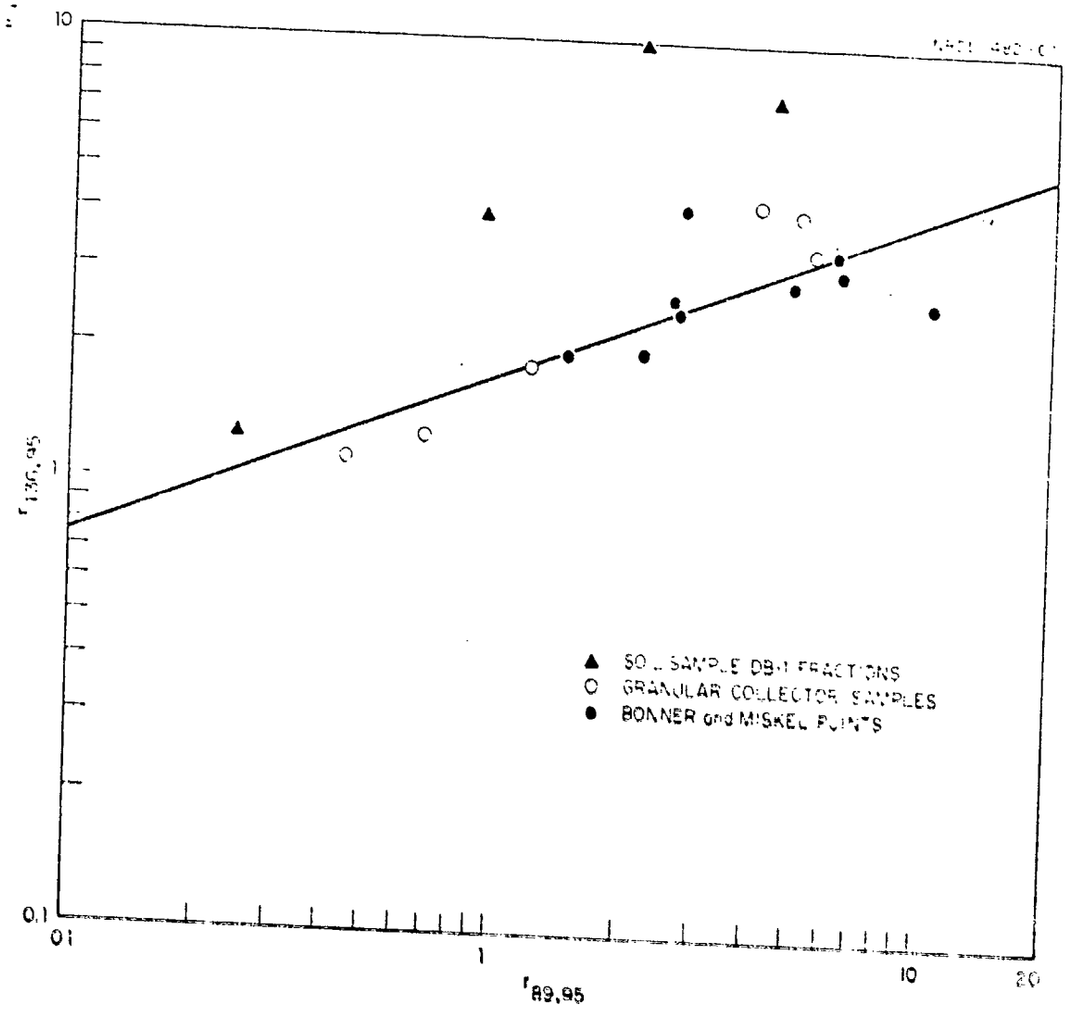

Fig. 5 Fractionation Correlation Plot for Cs^{136} Data From Danny Boy Samples.

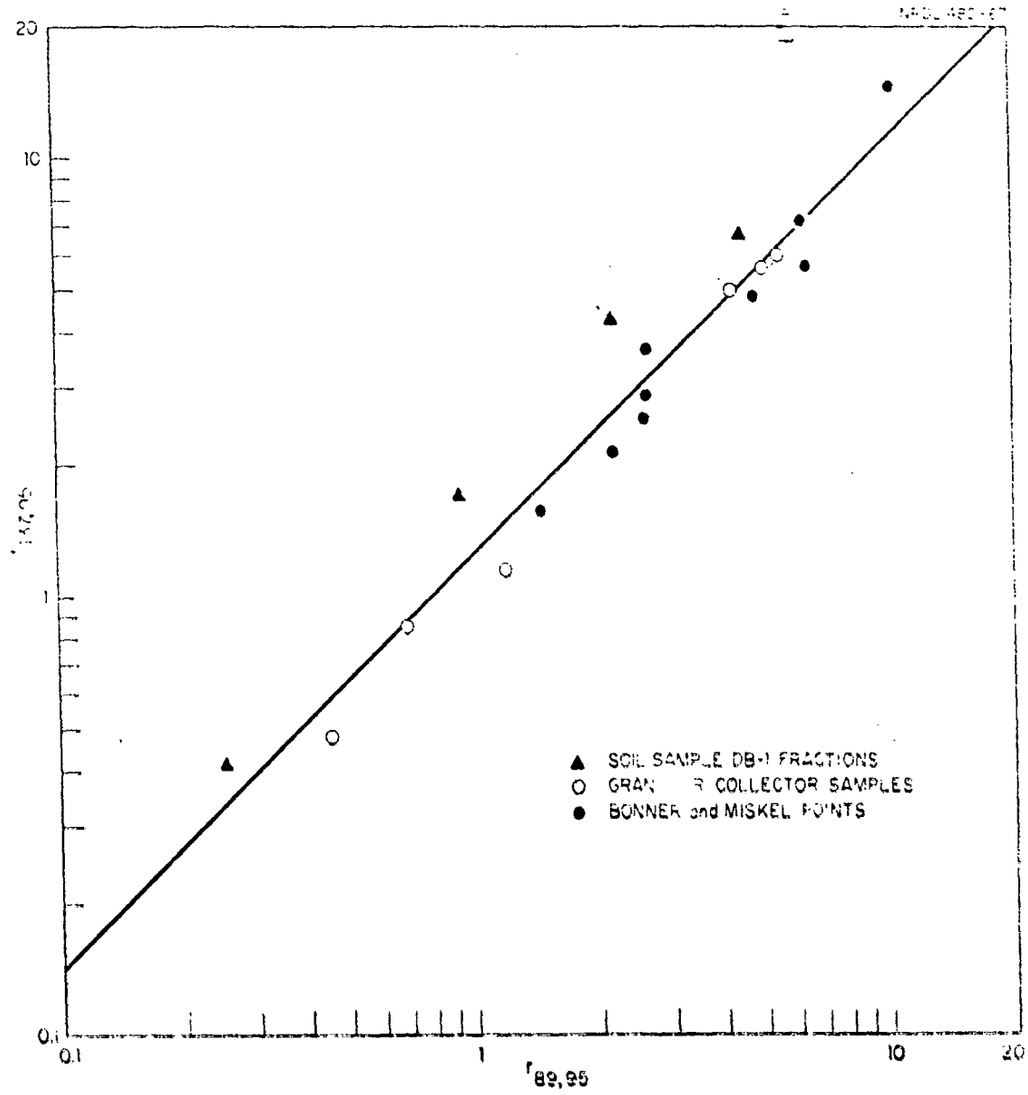

Fig. 6 Fractionation Correlation Plot for Cs^{137} Data From Danny Boy Samples.

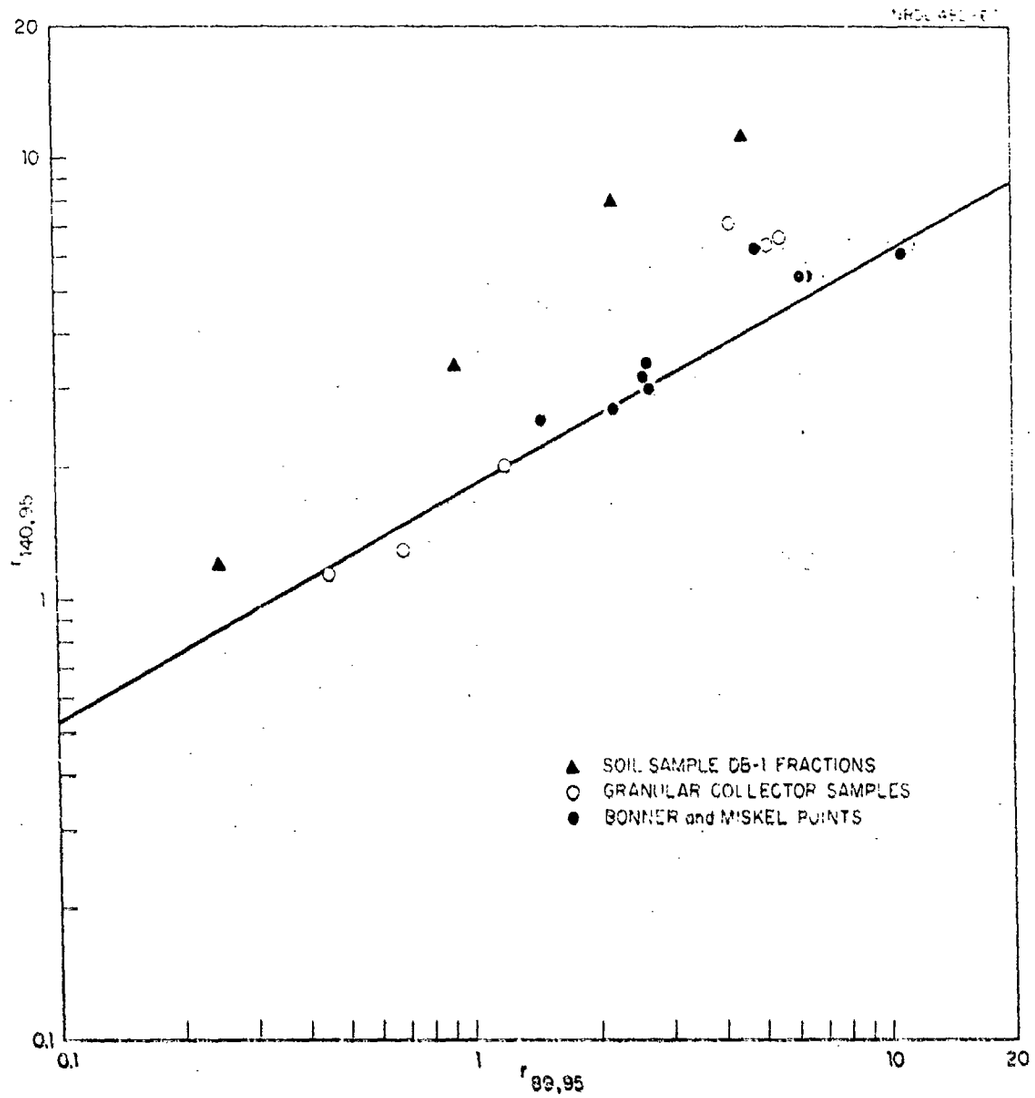

Fig. 7 Fractionation Correlation Plot for Ba^{140} Data From Danny Boy Samples.

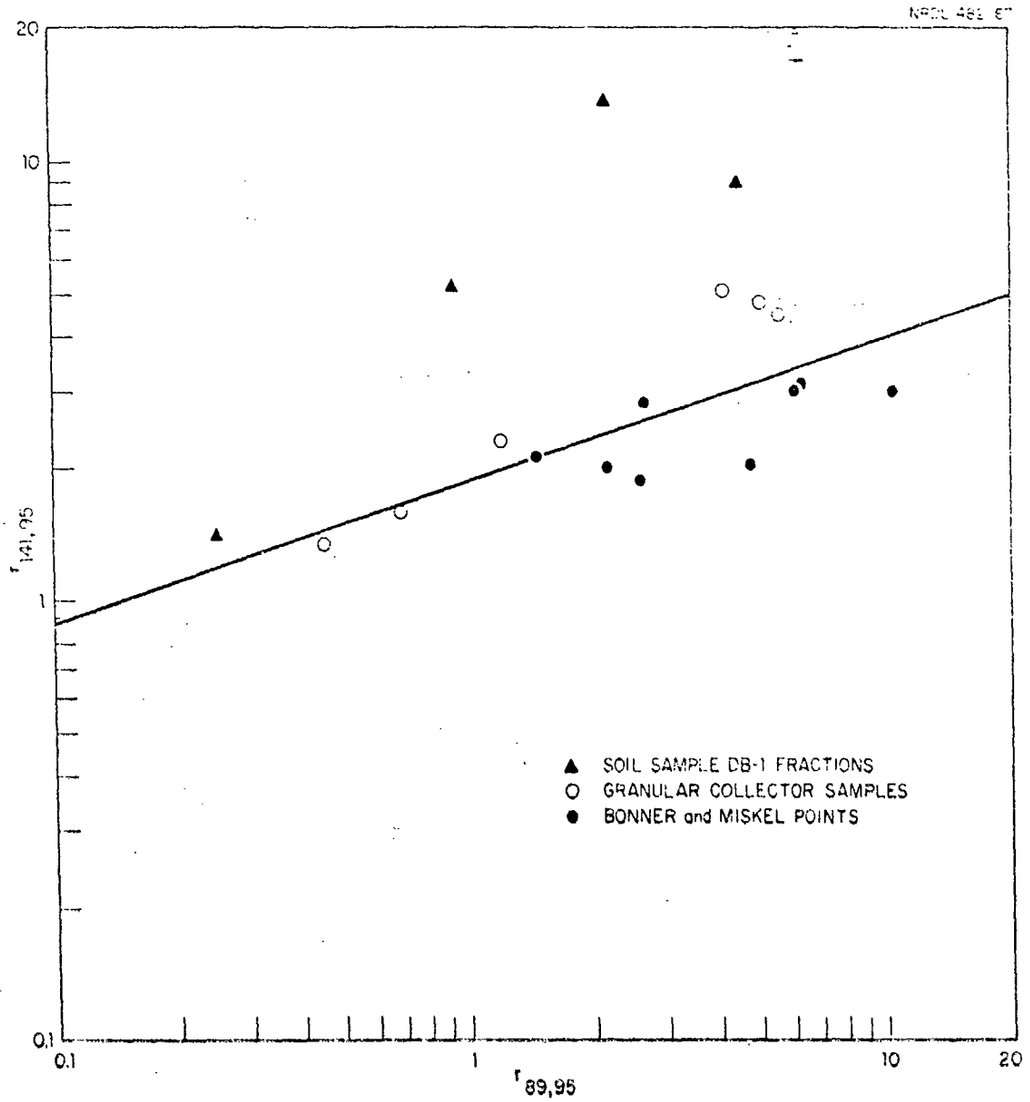

Fig. 8 Fractionation Correlation Plot for Ce^{141} Data From Danny Boy Samples.

into the fallout differed; i.e., perhaps Sr^{89} and Cs^{137} are mainly present on the surfaces of the particles while Sr^{90} and Cs^{136} have penetrated further into the interior.

The slopes of the correlation plots indicate the extent of fractionation from Zr^{95} of the nuclides to which they refer, relative to the fractionation of Sr^{89} to Zr^{95}. The slope for a particular nuclide is thus a measure of the relative volatility of its behavior. Table 6 gives the slope, the 95 % confidence limits, and the coefficient of correlation (COC) for the nuclides determined on the Danny Boy samples, along with comparable parameters from previous correlations of data on other events. The data points for the soil sample, DB-1, were not used in calculating the parameters, but the data points of Bonner and Miskel were included.

The parameters for Danny Boy indicate that Cs^{137} fractionated from Zr^{95} to about the same extent as did Sr^{89}; the fractionation of Sr^{90} and Te^{132} was slightly less; and the fractionation of Cs^{136}, Ce^{141} and Ba^{140} was markedly less but still appreciable. There was no fractionation of Mo^{99} and Ce^{144} from Zr^{95}. The Danny Boy parameters agree within the 95 % confidence limits with the parameters for the other three silicate-soil events listed in the table--Sedan, Johnie Boy, and Smallboy. Sedan was a relatively large (about 100 kt) cratering shot in alluvium, scaled to the same depth as Danny Boy. Johnie Boy and Smallboy were low-yield near-surface shots; the former was detonated 23 inches below the surface in basalt and the latter 10 feet above the surface of alluvium. The parameters for Johnie Boy and Smallboy appear to be slightly higher, in general, than those for Danny Boy and Sedan. Nonetheless, the parameters for all four events agree within the confidence limits except for the high value of the Cs^{136} slope in Johnie Boy and the low values for the Ba^{140} and Te^{132} slopes in the Sedan event. The first two of these (Cs^{136} and Ba^{140}) are only slightly out of range and the last is probably an artifact of the linear regression method of fitting (see Appendix C). A "visual" fit of the Te^{132} data from Sedan would prefer a slope in the range of 0.7 to 0.8. The parameters for the coral-surface events also agree within the confidence limits with those for the silicate-surface events except in the case of Cs^{136}. This nuclide showed moderate fractionation in the silicate events, but no fractionation in the coral-surface event.

TABLE 6

Correlation Parameters for Danny Boy and Some Other Events

Nuclide	Event				
	Danny Boy	Sedan[c]	Johnie Boy[c]	Smallboy[c]	Coral Surface
Sr^{90}	0.69 ± 0.21 COC = 0.89	0.54 ± 0.18 COC = 0.68	0.73 ± 0.10 COC = 0.91	0.73 ± 0.09 COC = 0.66	0.59 ± 0.08[a] COC = 0.95
Mo^{99}	0.01 ± 0.02 COC = 0.40	0.04 ± 0.09 COC = 0.29	-0.02 ± 0.05 COC = -0.22	0.04 ± 0.11 COC = 0.10	-0.08 ± 0.03[a] COC = -0.68
Te^{132}	0.89 ± 0.33 COC = 0.97	0.43 ± 0.21 COC = 0.73	1.08 ± 0.38 COC = 0.89	0.90 ± 0.26 COC = 0.86	0.60 ± 0.21[b] COC = 0.96
Cs^{136}	0.37 ± 0.17 COC = 0.80	0.31 ± 0.29 COC = 0.51	0.83 ± 0.22 COC = 0.93	0.65 ± 0.44 COC = 0.60	0.05 ± 0.08[a] COC = 0.25
Cs^{137}	0.94 ± 0.11 COC = 0.98	0.99 ± 0.06 COC = 0.99	1.11 ± 0.33 COC = 0.91	1.19 ± 0.10 COC = 0.98	1.00 ± 0.05[a] COC = 0.99
Ba^{140}	0.53 ± 0.38 COC = 0.64	0.43 ± 0.07 COC = 0.96	0.61 ± 0.08 COC = 0.98	0.51 ± 0.08 COC = 0.95	0.64 ± 0.79[b,d] COC = 0.99
Ce^{141}	0.32 ± 0.19 COC = 0.73	0.41 ± 0.44 COC = 0.46	0.39 ± 0.38 COC = 0.56	0.43 ± 0.13 COC = 0.83	-
Ce^{144}	0.03 ± 0.06 COC = 0.43	0.02 ± 0.08 COC = 0.70	0.00 ± 0.04 COC = -0.06	0.03 ± 0.11 COC = 0.07	0.08 ± 0.12[b] COC = 0.62

a. Based on data from an event of Operation Hardtack, Reference 7.
b. Based on data from an event of Operation Redwing, Reference 6.
c. Based on data reported in Reference 8. The parameter for Ce^{144} in Sedan given in Reference 8 is in error and has been corrected in this table.
d. These values are based on only 3 data points.

CONCLUSIONS

The number of samples received by NRDL from the Danny Boy event was limited and there was initially some doubt as to the validity of the radiochemical results, since all of the samples had been subjected to solvent action at some point in the processing. In the case of the granular-collector samples the solvent was isopropyl alcohol. The soil sample, DB-1, on the other hand, was subject to possible leaching action by water in the wet-sieving process; in addition, it was reported to have been exposed to an indeterminate amount of wetting by snow prior to collection. Unfortunately no samples of the isopropyl alcohol or of the water from the wet-sieving were available for analysis. However, the results of the correlations of the radiochemical data provide some basis for judgment. It appears that the isopropyl alcohol had no appreciable leaching effect on the fission-product radionuclides, since the data from the granular-collector samples correlate reasonably well, the slopes of the correlation lines agree with those obtained for other silicate-surface events, and the agreement with the results of Bonner and Miskel is satisfactory. On the other hand, the wet-sieving process (or possibly the wetting by snow) of sample DB-1 appears to have altered some of the fission-product concentrations, as discussed earlier.

Except for the presence of larger particles in later washes, the particle-size distribution studies of fractions from successive isopropyl-alcohol washings of the $< 44\ \mu$ fraction of sample X-24 do not indicate any significant differences.

The limited observations of the physical properties of the Danny Boy debris particles suggest that they were similar to particles from surface bursts. The proportion of radioactive particles of regular (spherical or spheroidal) shape was comparatively low, and the ratio of magnetic to non-magnetic particles was somewhat high. Spherical or spheroidal particles result from complete melting of the material in the fireball. Since the fallout from cratering events contains a much larger proportion of inactive soil than does that from surface bursts, the scarcity of regular particles is not surprising. It is more difficult to account for the proportion of magnetic particles in the Danny Boy debris. In surface bursts, magnetic particles are usually associated with large amounts of steel, such as steel towers, on the site of the detonation. Since the magnetic particle data on Danny Boy are fragmentary (only 54 radioactive particles were examined) it is probably unsafe to draw conclusions.

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APPENDIX A

RADIOCHEMICAL ANALYSIS PROCEDURE USED BY TRACERLAB/WEST

Chemistry

The granular collector samples were received as solutions in 100-ml volumetric flasks. However, the solution levels were below the mark at the time of receipt. The following amounts of water were added to the volumetric flasks in order to bring them up to the mark (at 20°C) and to permit the samples to be divided equally:

Y-24a, > 44 μ	no water added
Y-24a, < 44 μ	0.6 ml
X-24a, > 44 μ	0.25 ml
X-24a, < 44 μ	no water added
T-18a, > 44 μ	0.45 ml
T-18a, < 44 μ	0.25 ml

Two 50-ml aliquots were taken from these solutions, and sequential analyses for the following nuclides were performed: $Sr^{89,90}$, Zr^{95}, Mo^{99}, Te^{132}, $Cs^{136,137}$, Ba^{140}, and $Ce^{141,144}$.

The carriers were equilibrated with activity with HNO_3 and then 5 ml of H_2SO_4 were added. Barium and strontium were removed as sulfates. Tellurium metal was precipitated from a 2N HCl (nitrate-free) solution by bubbling SO_2 gas through it. After boiling and treatment with Br_2 water, the hydroxides of Zr and Ce were precipitated with NaOH. The supernate as acidified with HCl and put through a Dowex 1-X10 column for separation of Cs from Mo. Zirconium was separated from Ce by precipitation of $ZrPO_4$. All fractions were put through standard decontamination procedures.

Samples VF-1, VF-6, TT-3 and TT-6 were dissolved in 100-ml teflon beakers by means of the NRDL "Rock Dissolution Procedure" (see Appendix D). These samples were analyzed in duplicate for the following nuclides: $Sr^{89,90}$, Ba^{140}, $Ce^{141,144}$, Zr^{95}, $Cs^{136,137}$, and Te^{132}. The chemical separation was identical to that used on the granular collector samples except for the separation of Mo and Cs with the Dowex 1-X10 column, since Mo was not analyzed in these samples.

Counting

All purified samples were beta-counted. Most samples were counted on methane-end-window (MEW) proportional flow detectors. These detectors use 99 % C.P. methane gas and a window of 1.75 mg/cm^2 aluminum. Samples of low activity levels were counted on Tracerlab CE-14 low background end-window flow counters which use 99 % helium, 1 % isobutane gas, and a 0.9 mg/cm^2 aluminized mylar window.

Standard counting methods were used for assay of the various nuclides as appropriate: i.e., direct counting (at least three consecutive counting measurements), two-component decay resolution, and aluminum beta-absorption curves. The proper corrections were then made for sample self-absorption, chemical yield, decay, growth, and counter efficiency. Briefly, the techniques for the various isotopes were:

Sr^{89}, Sr^{90}: Direct-count Sr^{89} and Sr^{90} immediately after Y^{90} has grown in, milk off Y^{90} and count. Make appropriate subtraction to determine Sr^{89}.

Zr^{95}, Ba^{140}: Direct-count immediately and make correction for daughter growth.

Mo^{99}: Direct-count equilibrium mixture of Mo^{99}-Tc^{99m}. Follow beta-decay for purity check.

Te^{132}: Follow decay until long-lived Te component is left. Least-squares calculation on IBM 7090 computer to resolve Te^{132}.

Cs^{136}, Cs^{137}: Follow decay for sufficient time to permit least-squares calculation on IBM 7090 computer to resolve each component.

Ce^{141}, Ce^{144}: Obtain aluminum absorption curve to resolve Pr^{144} daughter of Ce^{144}. Subtract equivalent count rate of Ce^{144} and Pr^{144} from count at zero absorber to determine Ce^{141}.

The nuclear constants used in reduction of data are listed in Table A.1.

The beta-emission rates encountered in the samples were usually adequate for reasonable counting precision. The lowest count rates were found in the samples T-18a (> 44 μ), and TT-9 (a fraction of the DB-1 soil sample), but even these varied by several orders of magnitude for different isotopes.

TABLE A.1

Nuclear Constants Used in Reduction of Data

Nuclide	Half-Life (days)	Decay Constant (day^{-1})	Fission Yield[a] (percent)
Sr^{89}	52.9	0.0131	4.8
Sr^{90}	1.01×10^{4}	6.86×10^{-5}	5.9
Zr^{95}	64.8	0.0107	6.4
Mo^{99}	2.75	0.2518	6.1
Te^{132}	3.24	0.214	4.4
Cs^{136}	12.9	0.0537	0.006
Cs^{137}	9.71×10^{3}	7.14×10^{-5}	5.9
Ba^{140}	12.79	0.0542	6.3
Ce^{141}	32.5	0.0213	6.0
Ce^{144}	282	0.00246	6.1

a. For thermal-neutron fission of U^{235}.

The count-rate levels of the milked Y^{90} samples were all below 100 cpm and required low-background beta counting. The count-rate levels of Sr^{89} were adequate.

The Mo^{99} samples were measured at count-rate levels varying from 5 cpm to 1000 cpm for different samples at 22 to 24 days after the Danny Boy event.

The Te^{132} count rates for the granular collector samples were all greater than 300 cpm, usually several thousand cpm. In the case of the soil samples, the Te^{132} could not be resolved from the relatively high levels of long-lived Te because these samples were received and processed about 38 days after the Danny Boy event. For this reason, results for the latter samples are reported as limits.

The counting rates of the Cs^{136} and Cs^{137} samples were generally less than 100 cpm but ranged from 4 to 300 cpm. The combined counting rate for the two isotopes was not less than 12 cpm. The beta-decay data for these samples was taken on the MEW counter, because of an overload

of very-low-counting samples on the low background counters at the time. For this reason, the errors on some of the Cs^{136} values are rather high. The lowest-counting Cs^{137} samples were later recounted on low background counters for better statistics.

It is possible that the results for the long-lived isotopes Sr^{90}, Cs^{137}, and Ce^{144} were perturbed by unknown amounts of pre-shot background in the soil at the test site.

It is believed that one aliquot of the TT-9 fraction of the DB-1 soil sample received cross-contamination from another sample during the separation of the Cs-Zr-Ce fraction. For this reason only the results from the second aliquot of TT-9 were reported for these nuclides.

APPENDIX B

BACKGROUND INFORMATION ON METHOD FOR EXPRESSING RADIOCHEMICAL RESULTS IN TERMS OF EQUIVALENT FISSIONS

Consider a fission event which consists of F simultaneous fissions and which produces A_i atoms of mass i. The total chain-yield of the mass-i chain is defined as

$$Y_i = A_i/F$$

If the total number of atoms A_i were determined for this event, and if Y_i were known from previous experience with similar events, F could be calculated.

Frequently, as in the case of nuclear bursts, only a sample of the products is available for analysis. If α_i represents the number of mass-i atoms in this sample, the number of fissions represented by these atoms is

$$f_i = \alpha_i/Y_i$$

Here, f_i is the number of equivalent fissions for the mass-i chain. If the sample were representative, this value could be calculated for each fission product mass chain, and the values so calculated would all be equal. Because samples of nuclear bomb debris are not completely representative but have been enriched in some fission products or depleted in others by so-called fractionation processes, values of f_i calculated from the determinations of different mass chains will not all be equal, though some may be. As a consequence, the equivalence of these values for different mass chains determined in the same sample indicates that the mass chains to which they refer did not fractionate from each other in the debris formation process. Conversely, the lack of equivalence of these values indicates fractionation. Since the mass-95 and mass-89 chains differ widely in behavior, the ratio of their equivalent-fission values

$$r_{89,95} = f_{89}/f_{95}$$

is a useful measure of how badly a given sample is fractionated. It is called the fractionation ratio.

APPENDIX C

BACKGROUND INFORMATION ON RADIOCHEMICAL DATA CORRELATIONS

The ratio $r_{89,95}$, described in Appendix B, indicates the extent to which Sr^{89} has fractionated from Zr^{95}. The fractionation of any other nuclide i from Zr^{95} can then be related to $r_{89,95}$ in a simple way by log-log plotting. Values of $r_{89,95}$ for a set of samples are used as abscissas and corresponding values of $r_{i,95}$ are used as ordinates. The points can usually be fitted with a fair degree of confidence to the line:

$$\log r_{i,95} = a_i + b_i \log r_{89,95}$$

Values of a_i and b_i can be determined by linear regression, along with coefficients of correlation and limits of confidence. The slope, b_i, indicates the degree to which the nuclide i fractionates from Zr^{95} relative to the fractionation of Sr^{89} from Zr^{95}. If i does not fractionate at all, the slope will be near zero; if i fractionates like Sr^{89}, the slope will be near one; intermediate fractionation yields slopes between zero and one, while degrees of fractionation greater than that of Sr^{89} will give slopes greater than one. Negative values of the slope are sometimes encountered. The implication of a negative slope for nuclide i is that Zr^{95} fractionated from i in the same direction that Sr^{89} fractionated from Zr^{95}.

The significance of the intercepts, a_i, is less clear-cut. If all the correlation lines pass through the point ($r_{89,95} = 1$, $r_{i,95} = 1$), then some part of the debris may exist which has the same relative proportions of radionuclides as the unfractionated fission-product mixture. Interpretation of the significance of the intercept values is complicated by the fact that they may be strongly affected by calibration errors in the determinations as well as by errors in the nuclide yield estimates for the device.

APPENDIX D

ROCK DISSOLVING PROCEDURE

10 g finely ground rock in 250 ml Pt crucible.

1. Add 30 ml of conc. HF. After initial exothermic reaction has ceased, boil to dryness (or until spattering starts).

2. Repeat HF treatment.

3. Add 30 ml conc. $HClO_4$, 10 ml conc. HNO_3 and 10 ml conc. HF. Boil to strong fumes of $HClO_4$. Cool. Wash fume hood or change to new one.

4. Rinse down the sides of the crucible with 6 N $HClO_4$ and add 10 to 20 ml of conc. $HClO_4$. Fume to dryness, or until spattering starts.

5. Add 50 ml of 6 N HCl. Boil gently for four or five minutes. Cool.

6. Centrifuge ~ 5 minutes and pour supernate through No. 40 filter into pot.

7. Add 20 ml 6N HCl to ppt. and warm. (Centrifuge cone containing HCl may be warmed by turning on evaporators and holding the cone in the evaporator well for one or two minutes. Watch for bumping!) If all ppt. goes into solution, cool and if salt reprecipitates, repeat washing ppt. with 20 ml portions of 6 N HCl until all ppt. is dissolved. (Also try warm H_2O).

8. If residue remains, transfer from centrifuge cones to platinum crucible with water, add 10-20 ml conc. HF, and boil to dryness. Repeat HF treatment. Wash down walls of crucible with ~ 20 ml 6 N $HClO_4$ and fume down to dryness or until spattering starts. Repeat $HClO_4$ fuming. Wash fume hood.

9. Repeat step 8 until the residue has all dissolved. Then repeat steps 5-7.

10. Transfer filter paper to small Vycor beaker, add 20 ml conc. HNO_3 and 20 ml conc. $HClO_4$. Wet ash filter. Fume to dryness or until spattering starts, add 20 ml 6 N HCl and boil gently two or three minutes, cool and centrifuge. Transfer supernate to pot. Treat any residue by steps 8 and 9; however, no filtration should be required at this stage.

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