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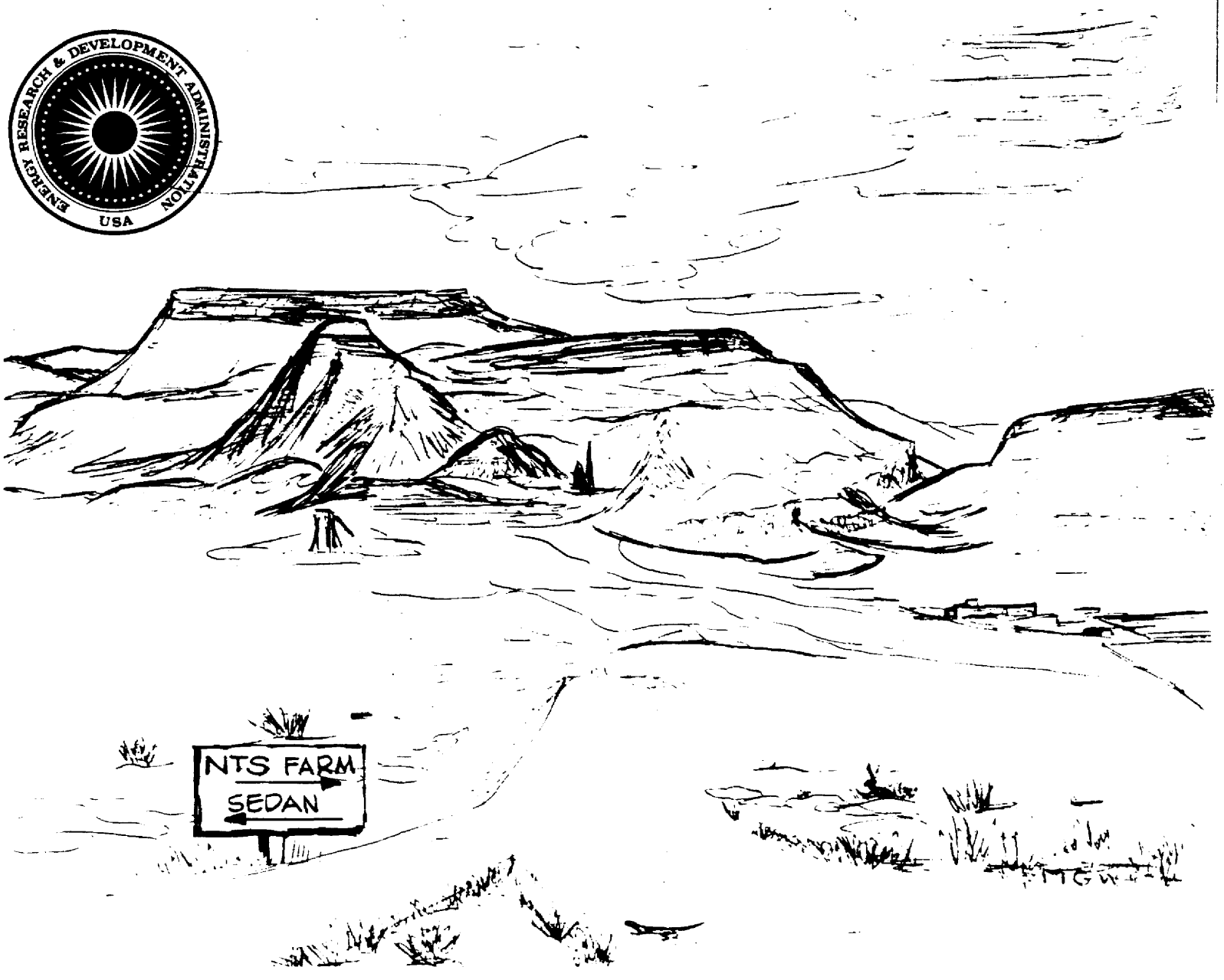
NEVADA APPLIED ECOLOGY GROUP PROGRESS REPORT (WORKSHOP SESSION - MAY, 1975)

MARCH 1976



UNITED STATES
ENERGY RESEARCH & DEVELOPMENT ADMINISTRATION
NEVADA OPERATIONS OFFICE
Las Vegas, Nevada

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AVAILABLE FROM:
NATIONAL TECHNICAL INFORMATION SERVICES, NTIS
U. S. DEPARTMENT OF COMMERCE
5285 PORT ROYAL ROAD
SPRINGFIELD, VA. 22161

PRICE: PRINTED COPY: $7.60
MICROFICHE: $2.25

NVO-159
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Edited by
M. G. WHITE and P. B. DUNAWAY

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Preface

PREFACE

This Nevada Applied Ecology Group publication is the result of a workshop held at the U.S. Federal Building, Las Vegas, Nevada, on May 6-7, 1975.

The document is comprised of summary progress reports of investigators and support activities of the Nevada Applied Ecology Group (NAEG), Nevada Operations Office, funded by the Division of Military Application, Headquarters, ERDA.

Essentially, the NAEG investigators have provided an update of various environmental studies currently in progress at the Nevada Test Site (NTS). The investigations are part of an integrated research program, designed: to provide information on the movement of plutonium and other radionuclides in the environment of NTS; to predict any possible associated hazard to man; and to recommend related cleanup and treatment, if required, for radioactively contaminated areas.

Previous publications of the Nevada Applied Ecology Group, ERDA, Nevada Operations Office, include Report NVO-142, *The Dynamics of Plutonium in Desert Environments*, July 1974; and Report NVO-153, *The Radioecology of Plutonium and Other Transuranics in Desert Environments*, June 1975. Numerous other reports by NAEG investigators have been provided in institutional publications, journals, symposia, etc.

The reader is also referred to presentations made by NAEG investigators at the IAEA/ERDA Symposium on Transuranium Nuclides in the Environment, November 1975, listed below by title and authors and to appear in the symposium proceedings:

1. Plutonium, Americium, and Uranium Concentrations in Nevada Test Site Soil Profiles. *E. H. Essington, L. L. Eberhardt, R. O. Gilbert, and E. B. Fowler.*

2. $^{239\text{-}240}Pu$ and ^{241}Am Contamination of Vegetation in Aged Fallout Areas. *E. M. Romney, R. O. Gilbert, and J. E. Kinnear.*

3. Plutonium Transport and Dose Estimation Model. *W. E. Martin and S. G. Bloom.*

4. Statistical Analysis and Design of Environmental Studies for Plutonium and Other Transuranics at NAEG "Safety Shot" Sites. *R. O. Gilbert, L. L. Eberhardt, E. B. Fowler, E. M. Romney, and E. H. Essington.*

5. Distribution of Ingested Transuranium Nuclides in Chickens and Subsequent Transport to Eggs. *Anita A. Mullen, Stephen R. Lloyd, and Robert E. Mosley.*

6. Grazing Studies on a Plutonium Contaminated Range of the Nevada Test Site. *Donald D. Smith, Julius Barth, and Robert G. Patzer.*

7. Plutonium Uptake by a Soil Fungus and Transport to its Spores. *W. F. Beckert and F. H. F. Au.*

8. Physical and Chemical Characteristics of Plutonium in Existing Contaminated Soils and Sediments. *Tsuneo Tamura.*

The cover of this progress report depicts the ERDA Experimental Farm at the Nevada Test Site. The farm, located at the foot of Flat Top Mountain, is operated for ERDA by the EPA Environmental Monitoring and Support Laboratory, Las Vegas. The NAEG large animal metabolism studies are performed at the farm.

We wish to acknowledge the continued support and encouragement of G. C. Facer, DMA, ERDA HQ; and R. L. Hitechew, R. Ray, C. E. Williams, and M. E. Gates (Manager), ERDA/NV. Special thanks are due to H. B. Gayle, P. G. Noblitt, and the Secretarial Center of Holmes & Narver, Inc., Las Vegas.

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

Soils

PLUTONIUM IN SURFACE SOILS OF AREA 13: A PROGRESS REPORT

Tsuneo Tamura

Environmental Sciences Division
Oak Ridge National Laboratory

ABSTRACT

Analyses of three additional soil samples taken along a north-south transect in Area 13 gave mean plutonium values of 324, 132, and 166 dpm/g for the samples taken at 1,910, 2,770, and 3,665 ft, respectively, from ground zero. In all three samples, more than 65% of the plutonium was in the 53-20 μm size fraction.

Comparison of particle size analysis of two soil samples (one under shrubbery and the other without vegetative cover), with and without ultrasonic dispersion, showed that in both soils, the silt and clay contents increased with the treatment. The major change in plutonium distribution following the treatment was an increase in contribution by the medium silt size fraction and a decrease in the fine sand contribution. Although the field survey instrument (FIDLER) recorded a higher reading for the sample under shrubbery as compared with nonvegetated soil, the analysis showed that the nonvegetated soil had a higher plutonium content in the 0-5 cm depth.

INTRODUCTION

This report covers research progress during the 6-month period of October 1974 through March 1975. The objectives and scope of this program have been presented in earlier reports (Tamura, 1974, 1975a).

During this period, research efforts were directed to two areas of study. First, analyses were completed of plutonium distribution among particle size fractions of three additional samples taken along the north-south transect. Second, particle size segregation and plutonium distribution in the different sizes of two closely situated samples were completed--one located in the bare area of the desert, the other beneath a saltbush 4 ft away from the first.

The additional three samples along the north-south transect were included because the distance between the previous consecutive sampling points 6 and 7 was 3,200 ft; and beyond point 7, the distance was 1,000 ft. For a better understanding of the plutonium concentrations in the surface soils as a function of distance from the point of detonation (GZ), more data points were desirable. The study of the two closely situated soils was initiated to determine differences, if any, in the plutonium distribution under these two different conditions.

EXPERIMENTAL METHODS

Quantitative analysis of plutonium was made using the hot HNO_3-HF technique, as described earlier (Tamura, 1975b). The technique used for particle size segregation of the NTS soils has also been described previously (Tamura, 1974). It should be mentioned that in the segregation treatment normally applied to these soils, no dispersant or vigorous mechanical treatment is used. However, in the two samples taken adjacent to each other, one aliquot received the usual treatment and a second aliquot received a 5-min ultrasonic treatment to induce dispersion. A Blackstone ultrasonic vibrator equipped with a 1/2 in. probe was used.

RESULTS AND DISCUSSION

North-South Transect Samples

In the earlier report (Tamura, 1975a), results of analyses of nine samples were given. In addition, the locations of three additional samples along the north-south transect in Area 13 were given. Analyses of 10-g subsamples of the three additional samples (labeled 6A, 6B, 6C) showed the plutonium concentrations to be 296, 136, and 145 disintegrations per minute per gram (dpm/g), respectively. The analyzed samples excluded gravel (greater than 2-mm particles), because the assumption was made that the plutonium contribution from gravel is negligible. Calculated concentrations of plutonium, however, did include the weight contributed by gravel in order to maintain consistency with other investigators on the project who normally grind the total soil sample prior to analysis.

In Fig. 1, the log of the plutonium activity is plotted against the log of the distance from GZ. The best-fit equation for the relationship was found to be the logarithmic relationship: $Y = 1.26 \cdot E + 9(X^{-2.07})$ where Y is the activity in dpm/g and X is the distance in feet; the term E + 9 is 10^9. In addition to the original nine samples along the transect and the three additional samples located from 1,900 to 3,600 ft from GZ, the mean concentration of plutonium in the samples taken near Site 2 for comparison of vegetated versus nonvegetated conditions is plotted. Note that Sample 6C contained 145 dpm/g, which is higher than anticipated; however, this type of anomaly is to be expected in large-scale field situations under variable atmospheric conditions.

Results of the particle size distribution of 6A, 6B, and 6C are given in Table 1. The particle size distribution of all three samples appears to be more closely related to that in the surface soil of Site 6 than of Site 7, which were previously reported (Tamura, 1975a). The plutonium distribution in size fractions less than 2 mm is given in Table 2. The activity (dpm/g) for each size fraction is presented in Row A. As in previously reported samples of the north-south transect, the 53-20 μm size contains the highest concentration of plutonium.

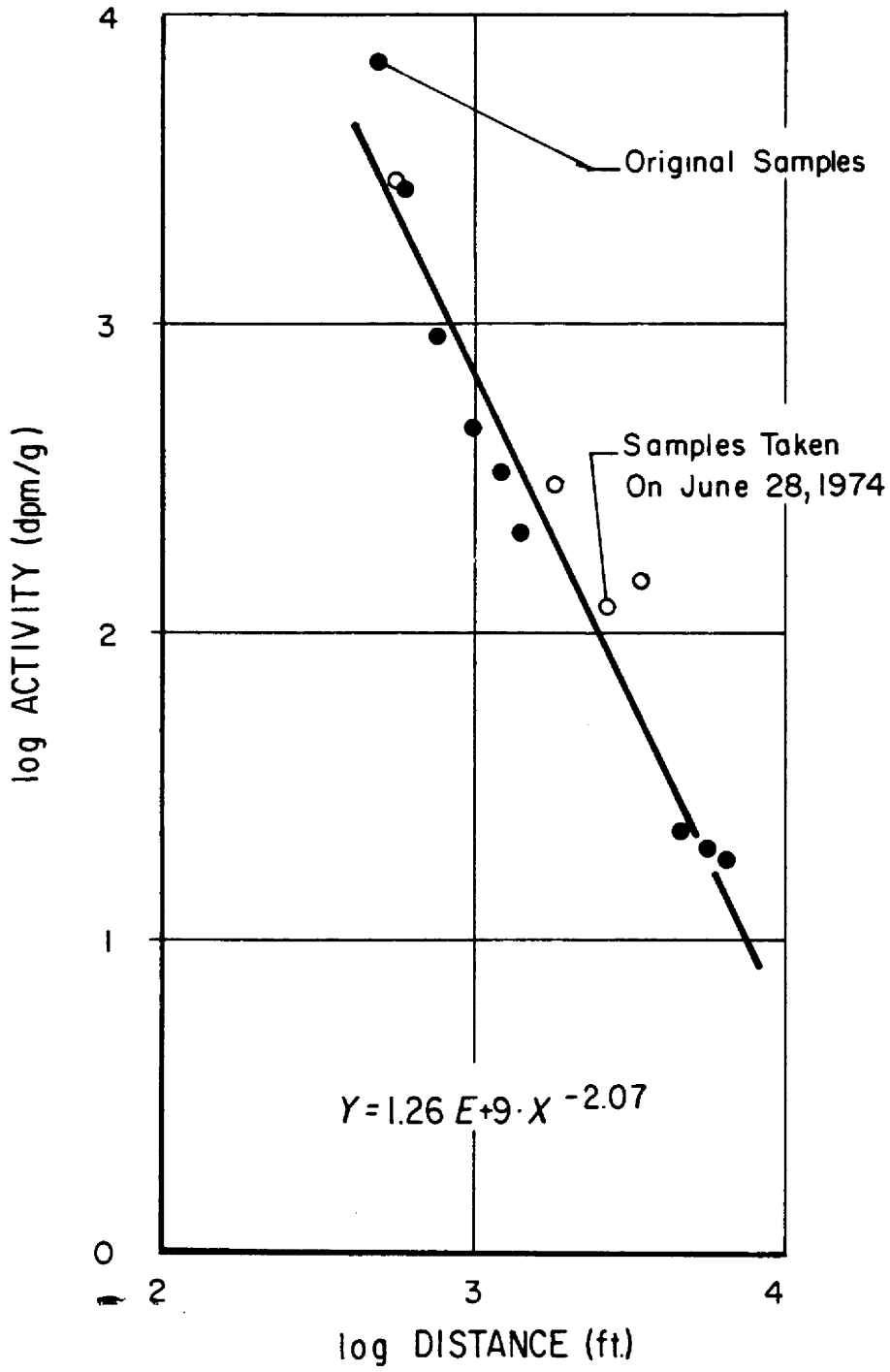

FIGURE 1. CONCENTRATION OF PLUTONIUM ALONG THE NORTH-SOUTH TRANSECT PLOTTED AS A FUNCTION OF DISTANCE FROM THE DETONATION POINT (GZ).

Table 1. Particle Size Distribution of Three Surface Soil Samples From Area 13 (Results Expressed in Percentage by Weight)

Size Range (μm)	Sample No. 6A	6B	6C
> 2000	10.3	10.1	12.6
2000-840	7.1	7.3	9.8
840-250	18.4	18.8	27.2
250-125	17.4	17.1	18.1
125-53	21.0	16.6	14.0
53-20	9.9	8.9	5.8
20-5	9.1	10.9	6.4
5-2	3.9	6.3	3.0
< 2	2.3	4.1	3.0
Total	99.4	100.1	99.9

Table 2. Distribution of Plutonium in Size Fractions and Contribution of Sizes to Total Soil Plutonium

Sample No.	Activity*	Size Range (μm)							
		2000-840	140-250	250-125	125-53	52-20	20-5	5-2	< 2
6A	A	6	6	5	208	2358	695	239	92
	B	0	1	1	44	233	63	9	2
	C	0.1	0.3	0.3	12.4	65.9	17.8	2.6	0.6
6B	A	10	13	6	7	1143	149	52	64
	B	1	2	1	1	102	16	3	3
	C	0.5	1.9	0.8	0.9	78.8	12.6	2.6	2.0
6C	A	4	6	4	155	2337	288	257	34
	B	0	2	1	22	136	19	8	1
	C	0.2	0.9	0.4	11.6	72.4	9.9	4.1	0.5

*Activity:

A = dpm/g in size fraction.

B = Contribution in dpm/g of soil, activity rounded off to nearest whole number.

C = Percentage contribution of activity in size fraction to soil.

In Row B, the activity for each size fraction is corrected for percent weight contributed by that fraction. The values have been rounded off to the nearest whole number. The sums of the activity of Samples 6A, 6B, and 6C are 353, 129, and 189 dpm/g of soil, respectively. These values may be compared with 296, 136, and 145 dpm/g, which are the concentrations based on analysis of the total soil. The mean values of the analyses are 324, 132, and 166 dpm/g, and the largest standard deviation is 18% for Sample 6C.

Row C shows the percentage contribution of each size fraction to the total soil activity. The highest contributor is the 53-20 μm size fraction.

Vegetated Versus Nonvegetated Areas

Formations of sandy mounds at the base of desert shrubbery are common at NTS. One characteristic of these mounds is that the portable field survey instrument (FIDLER) records a higher activity level over a mound than over the surrounding bare soil. The sample taken near Site 2 of the north-south transect (Fig. 1) was no different. Over the mound, the reading was 35,000 counts per minute (cpm); over the bare soil, it was 25,000 cpm. The samples taken represented the 0-5 cm depth. A noticeable feature was the uniform texture of the sandy mound in contrast to the more gravelly nature of the bare nonvegetated soil. Analysis of the gravel content showed that the bare soil contained 19.5% gravel by weight of total soil; the mound soil contained 2.7% gravel by weight of total soil.

The particle size distribution is given in Table 3. Note that the percentage distribution is based on the less than 2-mm particles, which excludes the gravel content. The total sand content of the two samples is almost equal (88%); the mound sample shows a high fine sand (53-250 μm) content as compared with the bare soil. The plutonium content, also shown in Table 3, is highest in the medium and in the coarse silt sizes (5-53 μm). Corrected for the relative abundance of these particle sizes, the medium and coarse silt sizes contribute 66% in the bare soil and 81% in the mound soil; in the latter case, 75% is in the coarse silt size. The activity is 3,800 and 2,573 dpm/g in the bare and mound soils, respectively. Analyses of 10-g soil samples

Table 3. Size Distribution and Plutonium Activity in Two Adjacent Soils in Area 13

Size Range (μm)	Abundance* (%)	Activity in Size Fraction (dpm/g)	Activity per Gram of Soil (dpm/g)	Contributed by Size Fraction (%)
		Nonvegetated Soil		
2000-840	9.0	195	18	0.5
840-250	30.2	190	57	1.5
250-125	26.2	206	54	1.4
125-53	21.9	4,645	1,016	26.7
53-20	6.4	26,556	1,686	44.4
20-5	3.0	28,350	836	22.0
5-2	1.8	5,540	97	2.6
< 2	1.6	2,250	36	1.0
Total	100.1		3,800	100.1
		Vegetated Soil		
2000-840	3.1	40	1	0.0
840-250	20.8	190	40	1.5
250-125	30.4	75	23	0.9
125-53	34.5	1,115	384	14.9
53-20	8.7	22,180	1,932	75.1
20-5	1.1	14,305	163	6.3
5-2	0.5	3,965	20	0.8
< 2	0.6	1,720	10	0.4
Total	99.7		2,573	99.9

*Abundance in percentage by weight.

showed 3,907 and 2,677 dpm/g, respectively. If the gravel contents are taken into account, the values are reduced by 2.7% for the mound samples and by 19.5% in the bare soil. The analyses show that the bare soil contains a slightly higher plutonium content even though the field readings showed the reverse situation.

These findings of plutonium association with particle size contrast with earlier studies of mound and bare soil samples from Area 5 (GMX) (Tamura, 1975a). There, the coarse silt in the mound samples contributed only about 11% of the activity as compared with 75% in the sample from Area 13. About 75% of the activity from the mound in GMX was in the coarser sand fractions. The likely explanation for the difference is the distance from GZ of the samples; at GMX, the distance was 250 ft; and at Area 13, 590 ft. At GMX, particles of coarse silt and larger sizes in the surrounding soil contributed 94% of the activity; at Area 13, the same particles contributed about 74%. Since closer distance to GZ would permit larger particles to be deposited, the difference in the bare soil and mound may be ascribed to this difference in source term.

To date, particle size segregations have been made without benefit of any dispersive treatment. In order to evaluate differences in particle size distribution and possible differences in plutonium distribution, the samples of bare soil and mound were given 5 min of ultrasonic treatment. The results of this treatment are given in Table 4. By comparing results in Tables 3 and 4, the notable feature of the size distribution is the reduction in sand content and the increase in silt and clay. Overall, the plutonium concentration in the respective size fractions was not greatly different in the bare soil. In the mound sample, the plutonium concentration in the size fractions appears to have decreased; this would suggest that the particles being contributed by the coarser fractions to the finer silt and clay were not highly contaminated particles and, thus, these particles proportionately changed the concentration in the different size fractions.

The effect of the ultrasonic treatment on the plutonium distribution in the soil is that the contribution from the sand fractions decreased and the silt and clay fractions increased. The largest increase in

Table 4. Size Distribution After Ultrasonic Treatment and Plutonium Activity in Two Adjacent Soils in Area 13

Size Range (μm)	Abundance* (%)	Activity in Size Fraction (dpm/g)	Activity per Gram of Soil (dpm/g)	Contribution by Size Fraction (%)
		Nonvegetated Soil		
2000-840	8.9	120	11	0.3
840-250	27.5	30	8	0.2
250-125	23.2	40	10	0.3
125-53	20.3	1,645	334	8.3
53-20	7.8	26,250	2,050	50.9
20-5	4.1	32,125	1,317	32.7
5-2	3.3	5,445	182	4.5
< 2	5.1	2,285	117	2.9
Total	100.2		4,029	100.1
		Vegetated Soil		
2000-840	3.0	80	2	0.1
840-250	20.1	20	4	0.1
250-125	25.4	20	6	0.2
125-53	28.9	565	163	5.9
53-20	10.4	19,495	2,031	73.2
20-5	3.3	12,700	413	14.9
5-2	3.1	3,120	116	4.2
< 2	5.6	735	41	1.5
Total	99.8		2,776	100.1

*Abundance in percentage by weight.

the contribution to the soil is in the medium silt fraction (20-5 μm); in the bare soil, it increased from a 22% contribution to almost 33%; and in the mound sample, it increased from 6 to almost 15%. The soil textural classification based on the dispersed sample place these soils at the lower limit of loamy sands, approaching sandy loam.

FUTURE PLANS

Plans are under way to sample soils from Area 11. Examination of the soils map and the FIDLER readings suggests that Site C in Area 11 might be interesting in terms of possible water transport during storms. Samples will be taken in Area 11 as functions of distance from GZ, runoff flow pattern, and depth of soil. Several samples will also be taken representing mounds and the adjacent bare soil.

Preliminary studies of NTS samples revealed that plutonium in the silt sizes was of high specific gravity (Tamura, 1974); it would be desirable to examine a few samples from the high specific gravity zone using density gradient segregation to detect the possible occurrence of plutonium oxide particles. Furthermore, preliminary studies of density gradient segregation of samples from Mound Laboratory, Miamisburg, Ohio, showed that organic material has adsorbed a significant fraction of the plutonium in the sediment. These results suggest that similar studies with samples taken beneath the shrubbery at NTS may reveal possible association of plutonium with vegetative material.

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NEVADA APPLIED ECOLOGY GROUP
SOILS ELEMENT ACTIVITIES FOR PERIOD
JULY 1, 1974, TO MAY 1, 1975

E. H. Essington and E. B. Fowler

Los Alamos Scientific Laboratory

ABSTRACT

During the period July 1, 1974, to May 1, 1975, LASL-H7 conducted assigned referee activities for the Nevada Applied Ecology Group (NAEG) Soils Element. Soil and vegetation samples were analyzed for various radionuclides, a tentative procedure for the analysis of uranium isotopes by alpha spectrometry was developed, and a review of the soil profile analyses for the intensive study sites was conducted. Recommendations regarding acquisition of additional data on profile sampling points are made.

SOILS ELEMENT ACTIVITIES

During this reporting period, LASL-H7 personnel have been working in four project areas:

1. Analysis of NAEG soil and vegetation samples.
2. Development of a procedure to determine U isotopes in soil and vegetation by alpha spectrometry.
3. Interlaboratory cross-calibration.
4. Preparation of papers for various meetings, and attendance at workshops.

Sample Analysis

During the reporting period, LASL-H7 has performed approximately 100 analyses for ^{238}U, ^{235}U, and ^{234}U; 150 analyses for ^{238}Pu and $^{239-240}Pu$; 200 analyses for ^{90}Sr; 100 analyses for ^{241}Am; and 100 gamma scans from which ^{60}Co and ^{137}Cs were determined.

Uranium Procedure Developemnt

During this same time period, a procedure to prepare alpha spectrometrically pure U plates from soil or large vegetation samples was developed. This effort is still in progress. However, a tentative procedure has been finalized and is being used to analyze NAEG soil and vegetation samples.

Most of the available procedures work quite well on solutions of low dissolved solids content, are long and involved, or are designed for fluorometric measurement of total U and produce an unsatisfactory plate for alpha spectrometry.

With the "tentative" method, small amounts of U can be isolated from a 10-g sample of soil or a 30-50 g sample of dried vegetation, in the presence of relatively large quantities of Pu and other interfering alpha emitters. The main interfering element is Fe, naturally present in the sample matrices to such an extent that serious alpha spectrum degradation occurs if Fe is not removed. Alpha spectrum interferences are avoided by this procedure since Pu, Am, Th, and most other alpha emitters are either removed from the sample or have alpha energies that do not interfere with the various natural or weapon U isotopes. As shown in the flow diagram (Fig. 1), the procedure includes (1) soil sample digestion by the LASL-HASL HF-HNO_3-HCl acid method, or vegetation digestion by the LFE method, (2) removal of much of the matrix mass by Fe scavenging, (3) ion exchange separation of U from most other elements which might interfere, (4) ether extraction to remove Fe, and (5) electrodeposition of U for alpha spectrometry using ^{232}U as a chemical yield tracer.

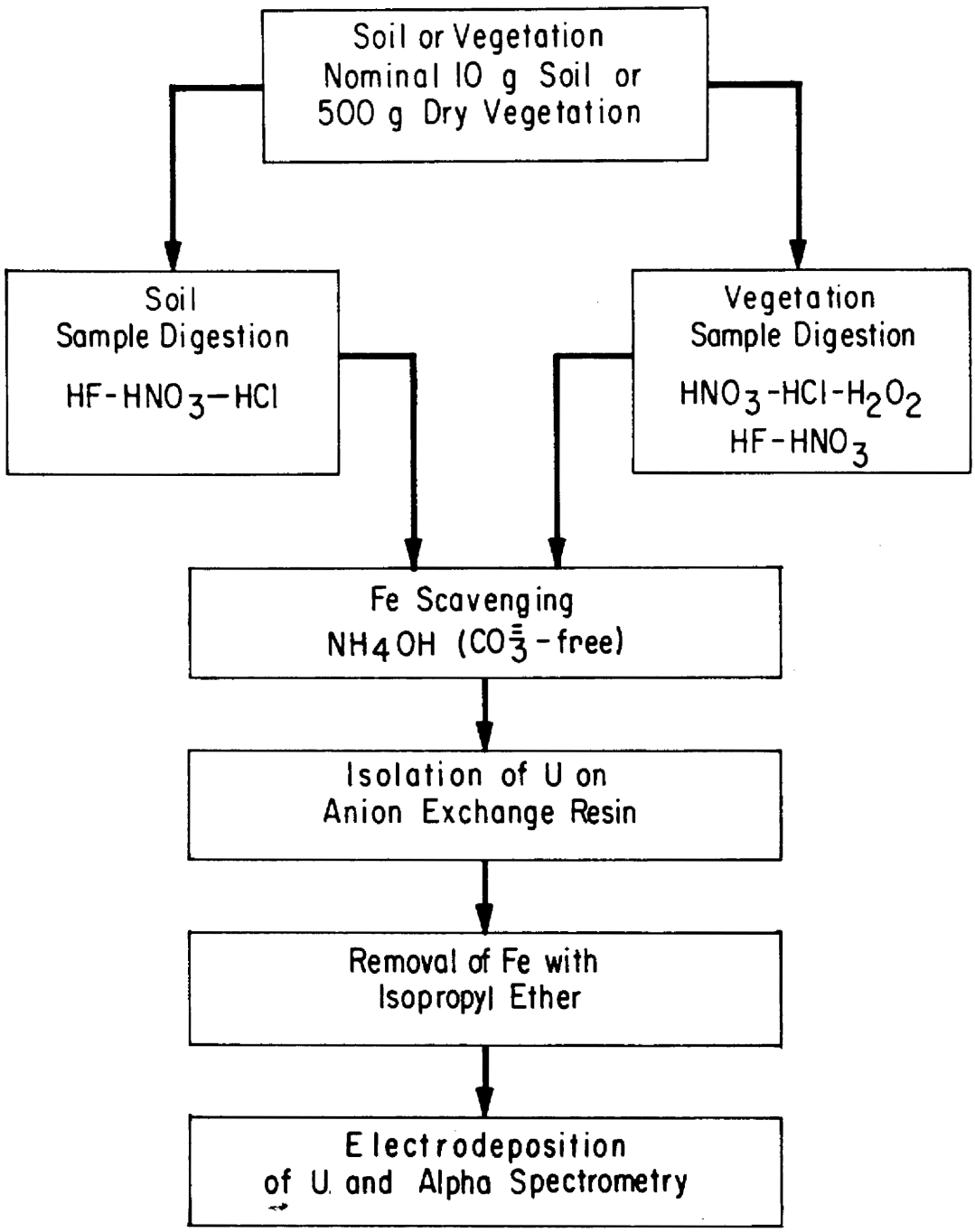

FIGURE 1. FLOW CHART OF TENTATIVE PROCEDURE TO ISOLATE ALPHA SPECTROMETRICALLY PURE U.

As an indication of the capability of this procedure to produce an alpha spectrometrically acceptable plate, the spectrum in Fig. 2 is presented as typical. This spectrum is one from an actual NAEG vegetation sample supplied to LASL-H7 by LFE. The figure shows the relative positions of ^{238}U, ^{235}U, ^{234}U, and the added ^{232}U. Typical U recoveries found to date on this type of vegetation sample range from 72-100%. The actual sample represented in the figure contained 300 d/m $^{239-240}Pu$; the amount of $^{239-240}Pu$ found in the U fraction was 0.16 d/m. This calculates to a .05% carry-over of $^{239-240}Pu$.

There are several problems yet to be worked out. For instance, this procedure with the current alpha spectrometer cannot quantitatively distinguish between ^{235}U and ^{236}U. If there is any ^{236}U to be found in NAEG samples, the ratio of ^{235}U to ^{236}U must first be determined by mass spectrometric techniques. The ratio can then be applied to the $^{235-236}U$ result from alpha spectrometric measurements to quantify the ^{235}U.

A second problem noted is that the plates are not as consistently clean as with the Pu procedure now in use. This is due to small amounts of Ni, Cu, Zn, and Fe present in the column eluate which seem quite difficult to remove prior to electrodeposition. However, of the nearly 100 plates thus far prepared only, 5 were unacceptable. It should be noted that the alpha spectrum presented in Fig. 2 represents only one resin column cleanup of the sample. It is believed that the problem of metal interferences can be adequately handled by a second column separation of U, and this feature is incorporated into the tentative procedure.

Laboratory Cross-Calibration

As part of the continuing effort to cross-calibrate the various NAEG-associated laboratories, LASL-H7 circulated a low-level Pu source for alpha spectrometry. Each laboratory was asked to report the amount of ^{238}Pu, $^{239-240}Pu$, and ^{242}Pu they found on the plate. To date, the source has been assayed by three LASL laboratories and three of the five NAEG-associated laboratories. The results (Table 1) show a good agreement for ^{242}Pu, but both $^{239-240}Pu$ and ^{238}Pu undergo substantial variation. For the purposes of NAEG soil analysis where there is an

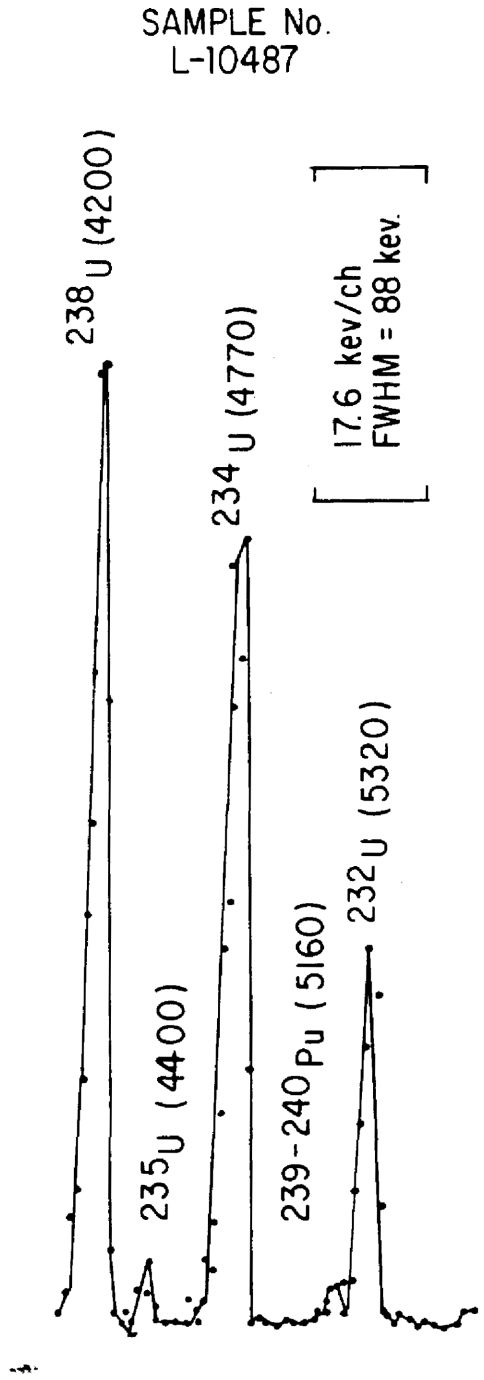

FIGURE 2. ALPHA ENERGY SPECTRUM OF ELECTRO-DEPOSITED URANIUM SEPARATED FROM VEGETATION.

Table 1. Results of Interlaboratory Calibration of an Electrodeposited Pu Source.

	^{242}Pu D/M	$^{239-240}Pu$ D/M	^{238}Pu D/M
	8.83	4.09	0.76
	8.80	4.57	0.78
	8.83	4.63	0.83
	8.42	4.57	0.72
	8.63	3.93	0.68
	8.24	5.00	0.77
Mean X =	8.62	4.46	0.76
One Standard Deviation =	0.247	0.390	0.052
Percent Deviation =	2.86	8.76	6.8

overriding influence of highly radioactive particles, these results are probably acceptable.

VEGETATION ANALYSIS PROBLEMS

LASL-H7 was asked to prepare a series of large vegetation samples for the purpose of checking the preparation procedure used by LFE and the analytical methods used by contributing laboratories for Pu. Am, and U analyses. In designing such a program, several problems have been encountered.

Details of the problems were presented to the NAEG in the hope that a method which best suits the needs of the group will be indicated.

PROFILE SAMPLING

Over the several years that the NAEG has been in existence, a large number of soil profile samples have been collected and analyzed. LASL-H7 has submitted a synopsis of a paper analyzing profile data for presentation at the IAEA Symposium on Transuranics in the Environment in November.

The paper will present a discussion of the profile data, attempting to explain noted irregularities and to discuss observations in relation to (1) rooting depth of local vegetation, (2) burrowing animal habitats, (3) surface redistribution by wind, water, and other mechanical activities, (4) leaching of radionuclides to various depths, and (5) decontamination and cleanup procedures.

The NAEG purpose of collecting profile samples from the intensive study sites was to determine the depth to which Pu had migrated. This information was used to establish a "surface sample" size (0-5 cm) and to determine whether significant downward movement of Pu had occurred. The information was used in inventory calculations for the

intensive study sites. To date, there are approximately 100 profiles from Area 13, Area 5-GMX, Area 11, and TTR (Fig. 3). More profile data will be available when analyses are completed.

In scanning through all the profiles, one is able to categorize each profile into one of three classes: (1) "high normal," where > 95% of the radionuclide content is localized in the top 5 cm; (2) "low normal," where there is insufficient radionuclide content to show profile development; and (3) "abnormal," where there occurs radionuclide profile development at some depth other than, or in addition to, the top 5 cm. Fig. 4 shows what is considered a "typical" or "high normal" profile. That profile was taken from Area 13 in the outermost stratum, furthest from GZ. As the graph shows, the majority of the Pu is in the top 5 cm. What appears to be profile development below 5 cm is open to question. The level of Pu found in each increment may be the result of Pu migration but is so low as to be questioned on at least three fronts: (1) cross-contamination during sampling, (2) cross-contamination during sample preparation, and (3) statistical variation in laboratory analyses. The procedures for sampling and sample preparation were designed to minimize cross-contamination, but the statistical variation in laboratory analysis may account for the observed results. It is believed that any Pu analysis resulting in radioactivities below 0.1 d/m/g from a 10-g sample should not be directly compared to a sample of similar radioactivity. Only after consideration of the variability generated in obtaining the analysis should comparison be made. Order-of-magnitude difference could occasionally be found, particularly if hot particles of Pu are present.

Closer observation of Fig. 4, however, does suggest that some Pu has moved to the region 5-20 cm by some mechanism such as: (1) ion exchange, (2) complex migration, or (3) particle migration.

The profiles, then, range from the "typical" (Fig. 4) to the "abnormal" (Fig. 5); a profile taken from Clean Slate 1, again in the stratum furthest from GZ. Here, in addition to a substantial portion of Pu at the surface, there appears to be two additional bands of Pu--one at 10 cm and one below 17.5 cm. There is evidence that much of the

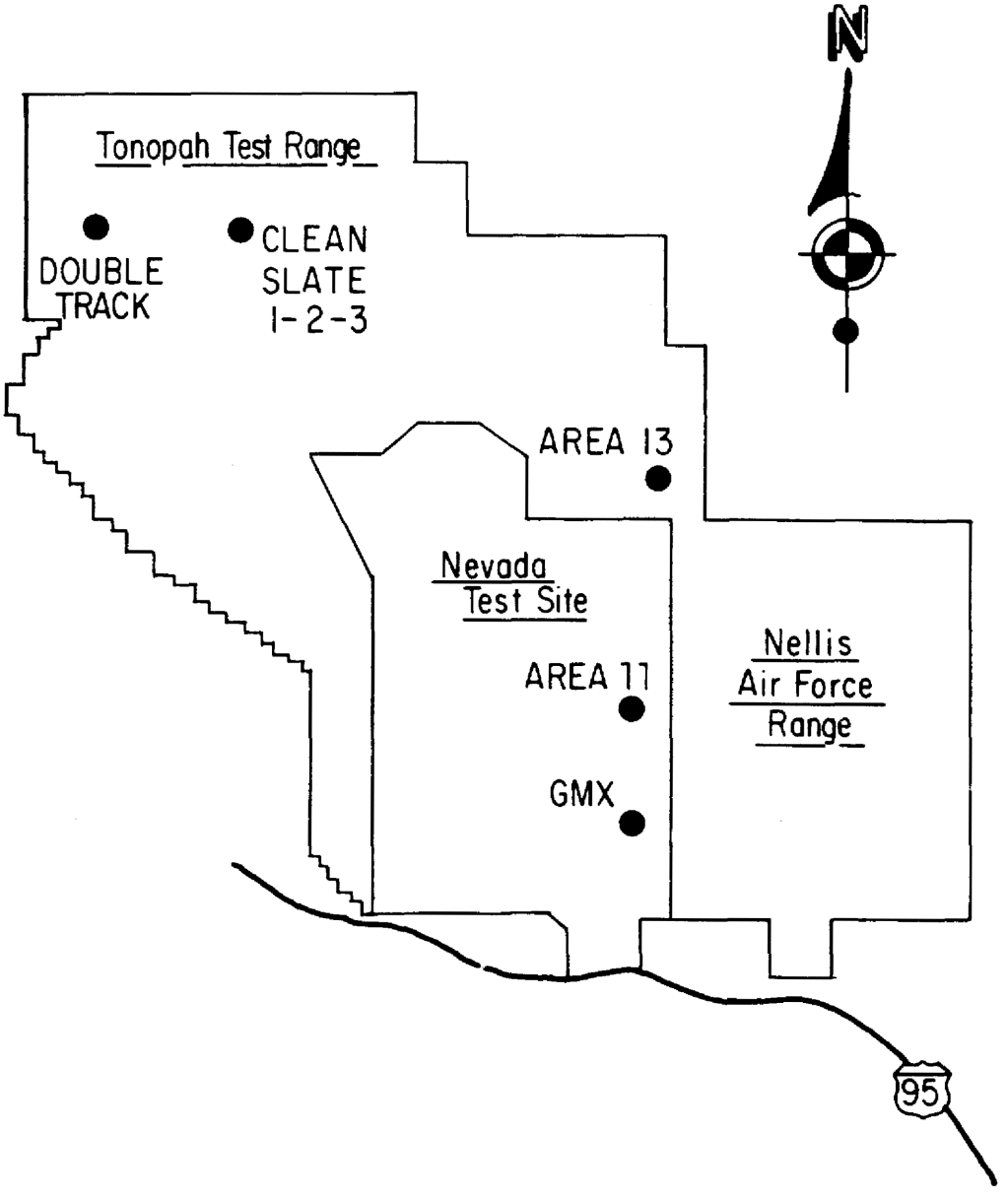

FIGURE 3. MAP OF NAEG INTENSIVE STUDY SITES ON THE NEVADA TEST SITE (NTS) AND THE TONOPAH TEST RANGE (TTR)

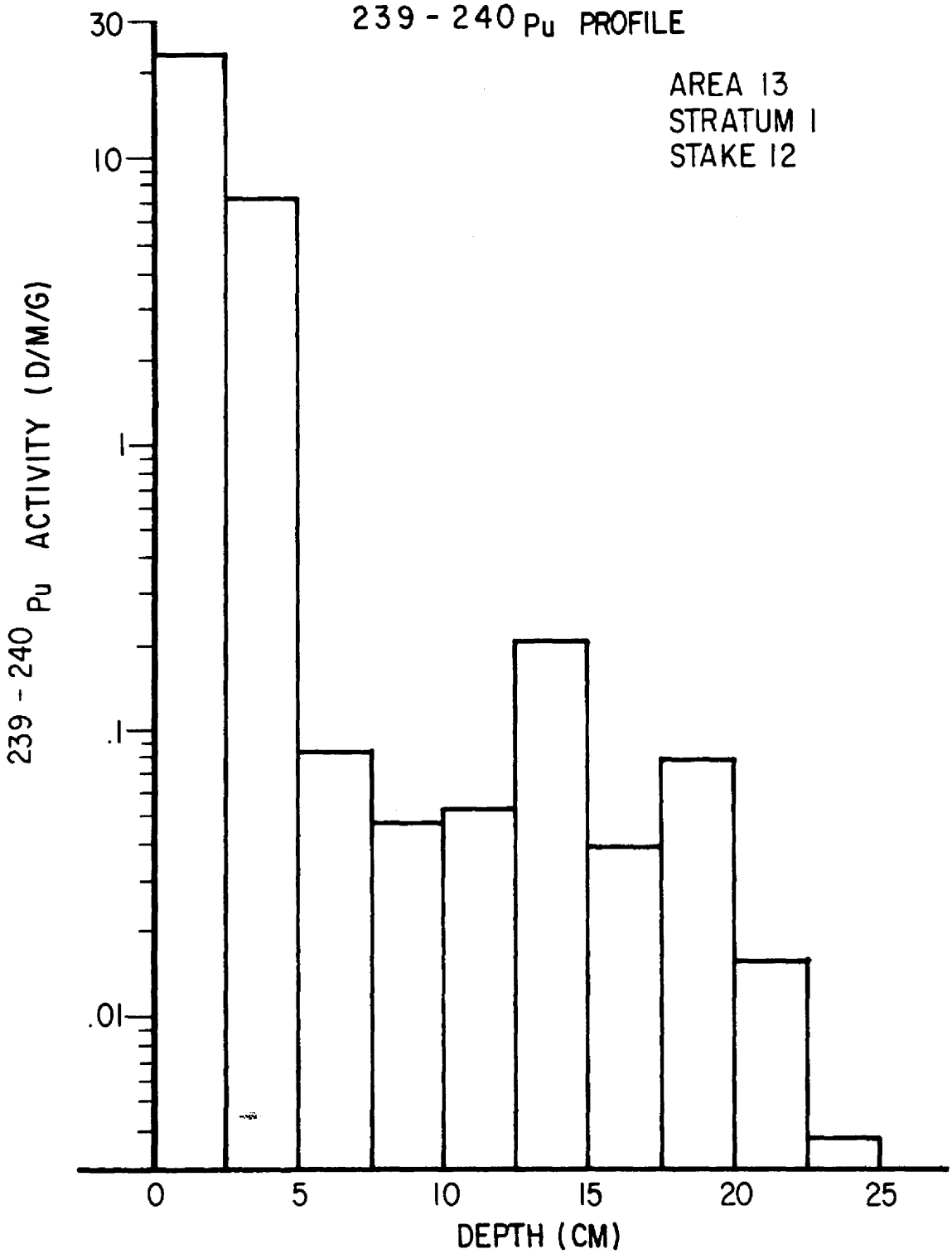

FIGURE 4. PLUTONIUM IN 25-cm SOIL PROFILE FROM AREA 13, NTS.

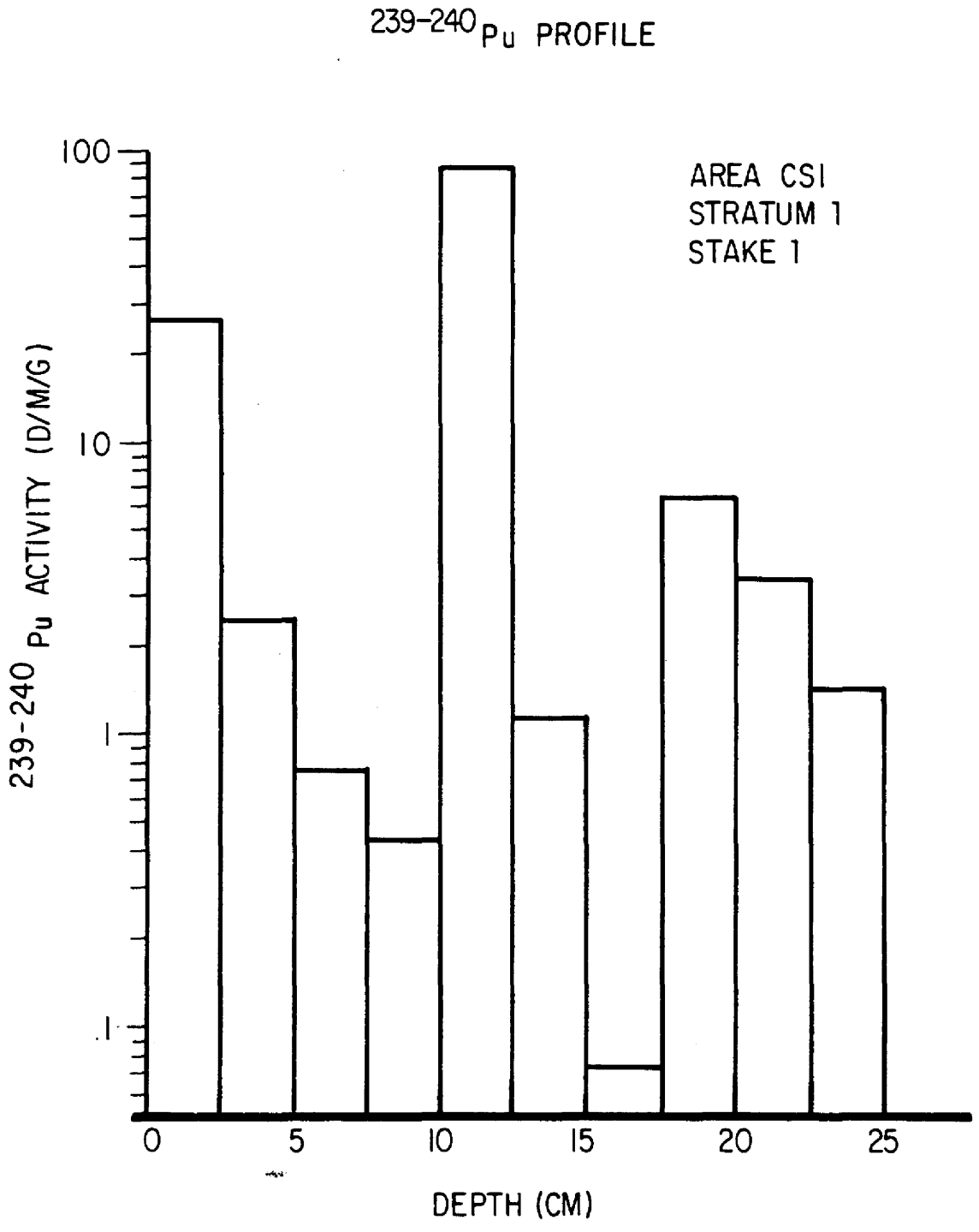

FIGURE 5. PLUTONIUM IN 25-cm SOIL PROFILE FROM AREA CLEAN SLATE 1, TONOPAH TEST RANGE

area around the Roller Coaster events had been mechanically disturbed for one reason or another shortly after the events. It appears that this profile reflects such mechanical disturbance. A number of the other TTR profiles show similar results. Until it can be resolved whether mechanical disturbance has occurred and, if whether unique soil properties are responsible for the observed development, this type of profile cannot properly be interpreted.

Selected soil profile sampling stations will be inspected later this spring to determine more precisely the soil characteristics with depth. For instance, gravel layers or clay layers within the sampling depth could significantly alter the interpretation of many of the profiles. Field notes are lacking in this respect.

Fig. 6 shows a profile of ^{235}U and ^{238}U from Area 11, Site A. The ^{238}U appears to be near natural background. The ^{235}U, presumably from the exploded devices, rapidly decreases with depth to more or less a continuum. Since natural ^{235}U levels are at least 10-20 times lower than those found in the 5-25 cm region, one must presume that device-^{235}U has migrated downward. A closer examination of the physical nature of the sampling site may allow confirmation of this presumption. This interpretation is based on the assumption that U contained in the device used at Area 11 Site A was predominately mass-235.

RECOMMENDATIONS

The following recommendations regarding profile sample collection and processing are made:

1. When taking profiles, make field notes on any kind of soil stratification observed. Also note any evidence of mechanical disturbance in the immediate location of the profile. Particular care should be taken to note microdrainage features.

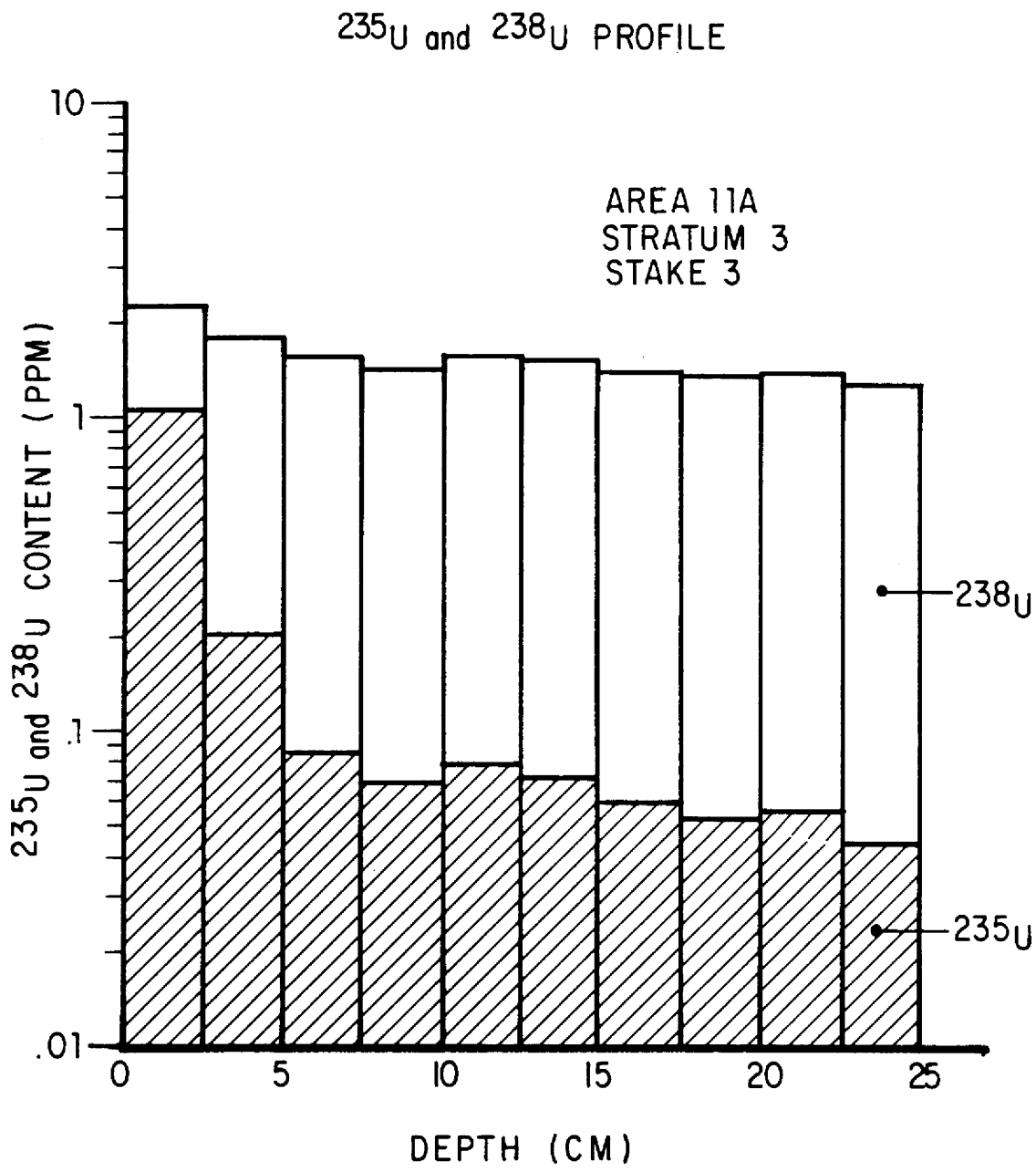

FIGURE 6. URANIUM IN 25-cm SOIL PROFILE FROM AREA 11A, NTS.

2. Return to several locations, to be later specified, and retake profiles to greater depths.

3. Review preparation laboratory procedures to be assured that a hot surface sample is not processed near other samples from the same profile.

Vegetation

NAEG PLUTONIUM PROGRAM VEGETATION STUDIES
STATUS REPORT, MAY 1975

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SCOPE OF PROGRAM

The significance of vegetation in any plutonium-contaminated area rests primarily upon its capacity to function as the carrier for plutonium and other transuranics in the food chains leading to grazing animals and man. Two different mechanisms of incorporation are involved in this transport process. First, the contaminant may become superficially entrapped upon vegetation through the processes of resuspension. This is expected to be the most important mechanism in the desert ecosystem, where environmental conditions are favorable to wind-driven forces. Second, the plutonium disseminated in soil may be taken up through plant roots and translocated to the above-ground vegetation. The Nevada Applied Ecology Group (NAEG) vegetation studies are expected to contribute information on how these two mechanisms of incorporation function in the vegetation-carrier transport of plutonium and other transuranics from soil to grazing animals and man.

DATA SUMMARY

Vegetation samples were collected in conjunction with soil samples from 10 study areas. Laboratory processing and radiochemical analysis of the vegetation samples essentially has been completed for Areas 13 and GMX-5 of the Nevada Test Site (NTS). About 60% of the samples

from the Area 11 sites at NTS and the Roller Coaster sites at the Tonopah Test Range (TTR) have undergone radiochemical analysis. Listed below are some pertinent findings:

1. The $^{239-240}Pu$ and ^{241}Am contamination levels varied from sample to sample collected within a given activity stratum defined by FIDLER survey instrument. These variations were of an order of magnitude attributable to differences in the amounts of resuspendable particulate material superficially entrapped upon plant foliage. The results also indicate that the activity levels found on vegetation may be attributed largely to superficial contamination in these aged fallout areas.

2. Reasonable agreement occurred between the mean activity levels of plutonium in vegetation and soil samples collected across different activity strata within each fallout area. The ratio of vegetation Pu to soil Pu tended to increase moving out from higher to lower activity strata.

3. Results indicate a reasonably constant Pu/Am ratio for vegetation samples collected from a given fallout area. This ratio, however, varied among the separate test events as the result of differences in the ingrowth of ^{241}Am within the aged fallout source materials.

4. The Pu/Am ratios were strikingly lower in vegetation samples than in soil samples from the Roller Coaster sites. A less pronounced trend was found in three of six activity strata sampled in Area 13 and at the Area 11 C and D sites. The reverse relationship (i.e., slightly higher ratios in vegetation than in soil samples) occurred at GMX-5 and Area 11 B. The real significance of these trends is not yet understood; however, the GMX-5 site had 22 test events, while the other sites each had a single event. One possible explanation for the Roller Coaster sites is that one of the more biologically available ingrowth products, ^{241}Am, has moved deeper into the root zone where greater root uptake has occurred relative to $^{239-240}Pu$. Results from soil profile samples should indicate how much movement has occurred.

Results from plant root uptake experiments, under glasshouse conditions, support this concept. Also, the annual precipitation is higher at the Roller Coaster sites than at other study sites.

5. The $^{239-240}Pu$ inventory estimates indicate that the standing vegetation contributes a rather insignificant portion of the total contaminant present in these aged fallout areas.

Four plant uptake experiments have been conducted under glasshouse conditions to determine the significance of root uptake of $^{239-240}Pu$ and ^{241}Am by alfalfa, barley, soybeans, and wheat. Radiochemical analysis has been completed for the barley samples (the other plant samples await analysis). Results showed discrimination factors ranging from 10^{-4} to 10^{-3} for $^{239-240}Pu$ uptake through roots. Pu/Am ratios for soil and plant samples also showed greater root uptake of the ingrowth product, ^{241}Am, relative to $^{239-240}Pu$ from the aged source materials.

EXISTING AND ANTICIPATED PROBLEMS

It has been desirous during the past three years of sample collection to obtain information on the annual plant species. During this period, however, the environmental conditions have been unfavorable for the production of annual plants in the study areas. Again this year, poor production of winter annuals is probable (those which germinate in late fall and winter and are so spectacular when flowering during April and May). Because of the late spring rains, there may be adequate summer annuals, grasses, and forbs to collect some samples for radiochemical analysis.

The supposition that the activity levels found on vegetation may be attributed largely to superficial contamination is based primarily upon two assumptions: (1) the residence particulate material is resuspendable and of small particle size (fallout particles entrapped upon plant foliage from aboveground nuclear testing generally ranged below 44-micron diameter), and (2) results from controlled plant

uptake experiments indicate much lower root uptake discrimination factors (Pu plant/Pu soil) than those Pu vegetation/soil ratios found in samples collected from the aged plutonium fallout areas. There is need to quantitatively verify the first of these assumptions through integration of findings from the resuspension and vegetation element studies. If it should result that the first assumption indicated above proves to be groundless, then the possiblility must be seriously considered that somehow, the perennial vegetation in these aged plutonium areas has been concentrating the transuranic elements in roots and stem tissues in forms that can be rapidly translocated to each new flush of foliage. Sampling to verify such a process will be difficult under field conditions due to cross contamination. Long-term root uptake studies now in progress with alfalfa and *Atriplex canescens* should contribute helpful information on this matter.

FUTURE ACTIVITY PLANS

Efforts shall be directed toward vegetation studies that will contribute information to help resolve those problems of concern mentioned above. Opportunities will be taken to report findings at scientific society meetings and symposia and to publish findings in the scientific literature.

Projects involving problems relative to revegetation and restoration of disturbed arid lands shall be conducted in conjunction with future cleanup activities in the aged plutonium fallout areas.

Large Vertebrates

SEASONAL EFFECTS ON THE SOLUBILITY IN *IN VITRO* BOVINE GASTROINTESTINAL FLUIDS OF PLUTONIUM INGESTED BY GRAZING CATTLE

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ABSTRACT

In order to study the alimentary solubility of plutonium, plutonium-contaminated rumen contents were collected from grazing cattle and were incubated in simulated bovine gastrointestinal fluids. During the spring and midsummer trials, a large increase in plutonium solubility and a marked reduction in the plutonium concentration of the rumen contents were observed concurrently with a reduction in intake of *Eurotia lanata* and an increase in the intake of *Oryzopsis hymenoides* or *Sitanion jubatum*. During the late summer, fall, and winter trials, comparatively high concentrations of plutonium, but low plutonium solubilities, were associated with high levels of *Eurotia lanata* in the rumen contents. Plutonium-238 was generally more soluble than plutonium-239 in these *in vitro* bovine gastrointestinal fluids.

INTRODUCTION

The primary purpose of this ongoing study is to determine the effects of grazing season, and the relative amounts of plant species ingested, on the solubility of plutonium-238 and plutonium-239 in an *in vitro* bovine digestive tract. Rumen contents, collected from grazing

*Now designated as the Environmental Monitoring and Support Laboratory-Las Vegas (EMSL-LV).

cattle allowed free choice of vegetation growing in a plutonium-contaminated area, provide an opportunity to study factors affecting the biological availability of the plutonium found in nuclear debris ingested at any particular time. Concurrently, biochemical factors affecting the availability of plutonium, such as abomasal and intestinal conditions including pH, bile, and enzymes, can be determined.

PROCEDURE

Rumen-fistulated cattle are allowed to graze periodically, and non-fistulated cattle permanently, in Area 13 of the Nevada Test Site. Samples of whole rumen contents are collected from the fistulated cattle following a 48-hr grazing period. Samples are collected from nonfistulated animals at time of slaughter.

Samples of whole rumen contents are added to digestion flasks with simulated abomasal juice consisting of HCl and pepsin, and the pH is adjusted to 3.0. Abomasal incubation in a water bath at 39.5^{o} C is allowed to proceed for three hrs. The contents of the flasks are then converted to simulate the duodenum by addition of NaOH to adjust the pH to 4.5 followed by incubation for about 10 min. The upper jejunum is simulated by the addition of bile, pancreatin, trypsin, and erypsin, and adjustment of the pH to 6.0, followed by a two-hr incubation period. The lower small intestine is simulated by adjustment of the pH to 7.5, followed by incubation for two more hrs.

Following each incubation period, the entire contents of one of the digestion flasks are separated into solid and liquid fractions by preliminary filtration through cheesecloth followed by centrifugation of the filtrate. The solid and liquid fractions are analyzed for plutonium-238 and plutonium-239.

FINDINGS TO DATE

The presently available data indicate that the qualitative response in *in vitro* bovine gastrointestinal tract of field-ingested plutonium, of uncertain chemical form, was in most cases similar to that of the plutonium-238 nitrate, plutonium-238 citrate complex, and plutonium-238 dioxide used in earlier *in vitro* studies. Increased plutonium solubility was generally noted when the pH was changed from 3.0 to 4.5. These respective pH values characterize a primary chemical difference between abomasal and duodenal contents. This was followed by a further increase after the addition of bile and enzymes and adjustment of the pH to 6.0 to simulate the upper jejunum. This increase is attributable to the action of bile.

In most cases, there was a considerable quantitative difference in the solubility of field-ingested plutonium and plutonium-238 dioxide used in earlier studies.

Field-ingested plutonium-238 was found to be generally more soluble than field-ingested plutonium-239 in these fluids. The differences were, however, quite variable.

During the late summer, fall, and winter trials, low plutonium solubilities were observed, accompanied by comparatively high concentrations of plutonium in the rumen contents. An analysis of the vegetal composition of the rumen contents indicated that this was associated with high levels of *Eurotia lanata* (winter fat). The minimum and maximum solubilities in the simulated duodenum, which is presently believed to be the most important stage, were 0.49 and 13.5% for plutonium-238. For plutonium-239, they were 0.14 and 0.76%.

During the spring and midsummer trials, a large increase in plutonium solubility was observed, which was accompanied by a marked reduction in total plutonium concentration in the rumen contents. Analyses of the vegetal composition of the rumen contents indicated a reduction in the proportion of *Eurotia lanata* and an increase in the proportion of *Oryzopsis hymenoides* (Indian ricegrass) or *Sitanion jubatum* (squirreltail grass). The minimum and maximum solubilities in the simulated

duodenum were 95.1 and 96.6% for plutonium-238; for plutonium-239, they were 44.5 and 90.0%.

This study is still in progress, and americium-241 analyses will be included in all future work. The major problem at present involves expediting the sample analyses for transuranium nuclides.

FUTURE ACTIVITY PLANS

1. A comparison of the solubilities of biologically incorporated and inorganic plutonium-238 in an artificial rumen and simulated gastrointestinal fluids.

   The results of the study previously described indicate the possibility that comparatively very small amounts of plutonium may be in an organic form, and that the biological availability may be much greater than that of inorganic forms. The digestion of plutonium-contaminated fungal tissue in an artificial rumen and *in vitro* bovine gastrointestinal fluids would provide a convenient means to compare the biological availability of biologically incorporated plutonium with inorganic plutonium.

   It is proposed that *Aspergillus niger* be cultured in a medium containing plutonium-238 nitrate. The fungal tissue will be collected and digested in an artificial rumen, followed by further digestion in simulated abomasal and intestinal fluids. Other digestion flasks will contain plutonium-238 nitrate for comparison.

   The objective of this study is to determine whether organic binding protects plutonium from being removed from solution by competing chemical reactions, such as adsorption, formation of insoluble salts, or polymerization.

2. The effect of soil microbiological activity on the solubility of plutonium and americium-241 derived from nuclear debris in an artificial rumen and simulated bovine gastrointestinal fluids.

It is proposed that contaminated soil be collected from Area 13 (NTS) and be divided into two portions. One portion will be autoclaved to destroy the soil microflora. A nutrient medium to support the growth of microorganisms will be added to both portions, and then will be incubated under optimal conditions. Periodically, plutonium and americium-241 solubility will be compared in an artificial rumen and simulated fluids of the abomasum and intestine.

STATUS REPORT ON GRAZING STUDIES ON A PLUTONIUM-CONTAMINATED RANGE OF THE NEVADA TEST SITE

D. D. Smith

National Environmental Research Center, Las Vegas*

INTRODUCTION

As has been reported previously, a grazing study on a plutonium-contaminated range in Area 13 of the Nevada Test Site (NTS) was initiated in May of 1973 and is continuing. Since that time, a herd of beef cattle (grazing ruminants) has been continuously maintained, and goats (browsing ruminants) have been periodically maintained within the study area. Their entire diet consists of native plants growing in the contaminated soil. The primary objective of this study is to determine the uptake and tissue distribution of plutonium by ruminants. Other objectives are: (1) to determine the dry matter and plutonium uptake of grazing animals at different seasons of the year; (2) to determine the relative contributions of plutonium incorporated within the plant versus that on the plant or ingested as foreign matter during the grazing processes; (3) to evaluate maternal-fetal transfer; (4) to determine uptake and distribution differences in young versus mature animals; (5) to compare levels of plutonium found in tissue from grazing animals with levels found in browsing animals; and (6) to compare these data with that collected from other contaminated areas.

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*See footnote, p. 35.

METHODS

Tissue samples were collected from three cows, two calves, and one fetus in October 1973; three cows and two fetuses in July 1974; and one cow and two yearling calves in January of 1975. Other animals sampled included three goats, three foxes, one jackrabbit, and one coyote. The wildlife species are considered to be transitory residents of the contaminated area.

All animals sampled were necropsied; selected tissue and organ samples were collected for actinide analysis and histopathological examination. Tissues sampled for actinide analyses included bone (femur and vertebra), lung, liver, muscle, tracheo-bronchial lymph nodes, gonads, blood, kidney, rumen ingesta, and reticulum sediment. Those collected for histopathological examination were adrenal, eye, cardiac and skeletal muscle, liver, lung, gonads, spleen, thyroid, kidney, and gross lesions. Botanical and radionuclide data obtained from the rumen ingesta will supplement that collected from the fistulated steer portion of the study.

In addition to those animals maintained in the contaminated area, fistulated steers are used as biological samplers to determine grazing habits as a function of plant availability and season, to estimate plutonium intake over specified periods of time, to determine the ratio of the actinide isotopes in the ingesta, and to estimate the percentage of digestable nutrients in the ingesta.

At monthly intervals, for the first 6 months of the study, and at quarterly intervals thereafter, four fistulated steers were placed in the fenced area of highest contamination for a 48-hr acclimation period. Following the acclimation period, their rumens were emptied of all ingesta and the steers were allowed to graze for a specific period of time. All ingesta from this controlled grazing period were removed for the various analyses required to meet the objectives of this portion of the study.

In July of 1974, a contractor (the University of Nevada, Reno) was selected to determine the dry matter and plutonium intake of grazing animals at various seasons of the year, to determine the digestibility of range plants grown on plutonium-contaminated soils, and to partition the quantity of ingested plutonium between that in the plant versus the plutonium on the exterior of the plant.

The 300 fecal, ingesta, and plant samples specified in the contract were collected during three 10-day sampling periods in July and October of 1974 and January of 1975. As of February 1, 1975, the University had completed 35% of the contract. This included 100% completion of sample collection, 30% completion of the sample preparation, and 10% completion of the sample analysis.

RESULTS AND DISCUSSION

With the single exception of the goat phases, the maintenance and sampling phases stated in the study protocol have been met. Because of coyote predation, we have been unable to maintain a viable goat herd.

As stated last year, the greatest problems with this study are concerned with actinide analysis.

Following the selection of an analytical laboratory, tissue and ingesta samples were shipped for analysis in September 1974 and February 1975. As funds are not available for further analysis until FY 1976, approximately 70 samples remain in cold storage. These include all the samples from the July 1974 sacrifice. Before the end of the fiscal year, approximately 135 samples from the University of Nevada, Reno, portion of the study will be submitted for actinide analysis. Hence, it is anticipated that 205 samples will be carried over and will be analyzed against FY 1976 funds.

The first raw data were received from the analytical laboratory in February and March of 1975. The key to the Nevada Applied Ecology Group computer code used to identify these data was received in

April. At present, the data have not been analyzed beyond a simple tabulation. Data will be statistically analyzed prior to preparing the paper for presentation at the International Symposium on Transuranium Nuclides in the Environment to be held in November at San Francisco.

In addition to the analyses for alpha-emitting actinides, selected tissue samples from these animals were analyzed for gamma-emitting radionuclides on the multiparameter system. It is interesting to note that detectable levels of americium-241 were found in rumen ingesta, 0.028 ± 0.003 pCi/gram, and in wildlife tissues; e.g., fox liver contained 0.36 ± 0.03 pCi/gram. At this time, comparison to other results is not possible, as the radiochemical data are not available.

Dietary analysis of rumen ingesta continues. As reported last year, the favored plant species were two grasses, *Hilaria jamesii* and *Orhyzopsis hymenoides*; two shrubs, *Eurotia lanata* and *Atriplex canescens*; and one forb, *Salsola paulsenii*. The graphical description of the diet of the fistulated steers is shown in Fig. 1 as relative percentages of the three types of plants.

Vegetation is sparse but apparently adequate to maintain a herd of six cattle remaining within the study area. One cow, one heifer, and two yearling bulls are in the outer compound, and one cow and one heifer are in the inner compound. All females are pregnant.

The study was originally designed as a five-year study, to be reevaluated at the end of two years. This reevaluation would determine whether to continue as designed, to modify, or to terminate the study due to lack of any measurable uptake of radionuclides. Unfortunately, the analytical data are insufficient at this time to make the evaluation and decide whether to continue as designed, or modify, which are the remaining two options. Hopefully, these data can be accumulated and interpreted during the next several months. After receiving this information, a decision can be made as to which animals to select and the number of animals to sample during the scheduled July 1975 sampling period.

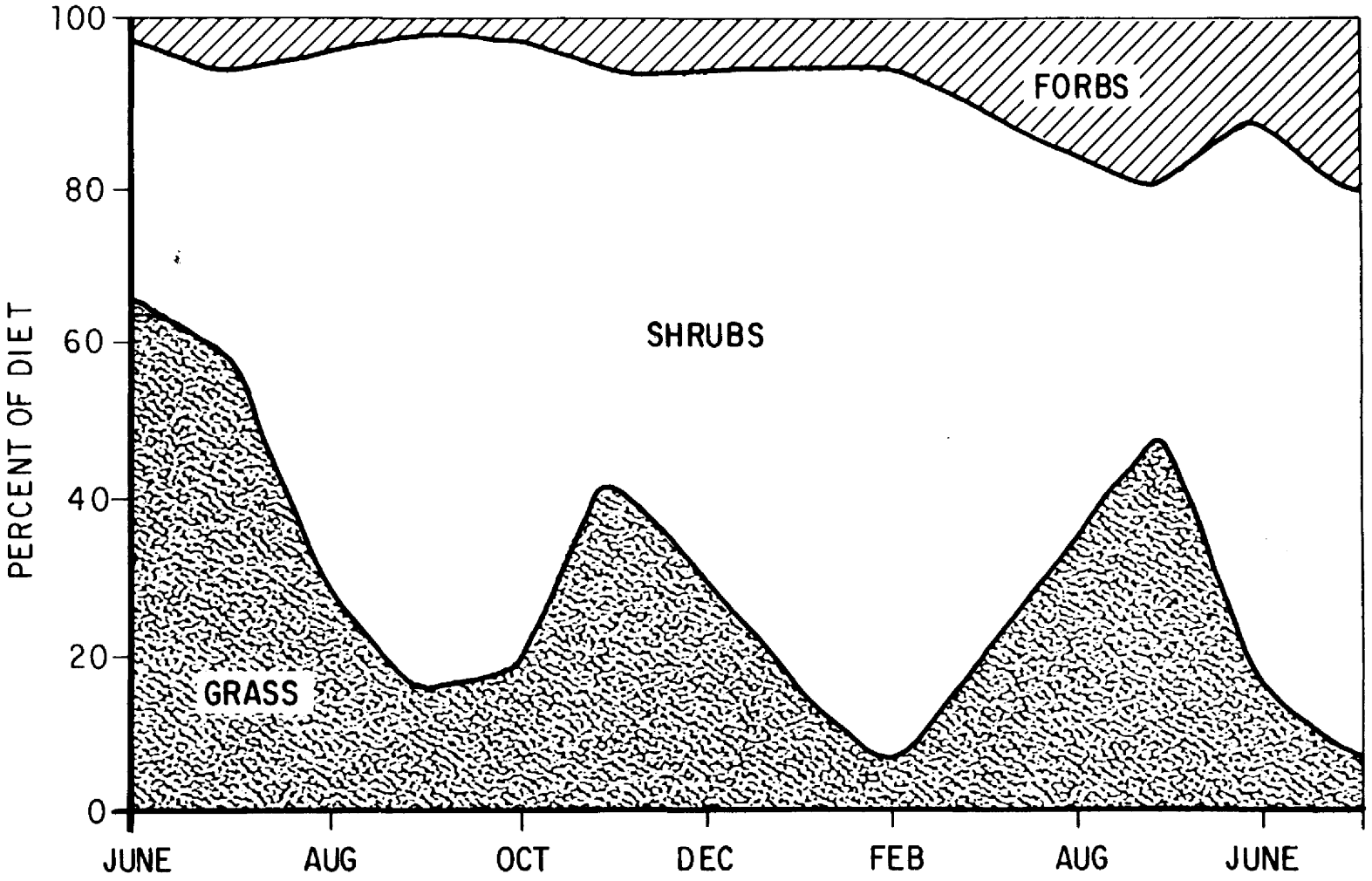

FIGURE 1. BOTANICAL COMPOSITION OF RUMEN CONTENT FROM FISTULATED STEERS GRAZING ON AREA 13, NEVADA TEST SITE.

PASSAGE OF SAND PARTICLES THROUGH THE GASTROINTESTINAL TRACT OF DAIRY COWS

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SUMMARY REPORT

Dairy cattle ingest moderate amounts of soil particles during normal foraging. These particles are ingested after becoming deposited on forage plants by air currents, or because they are associated with plant roots ingested by cattle. In addition, soil that becomes deposited on the snouts of cattle is licked off and ingested. With increasing frequency such soil contains toxic pollutants. The gastrointestinal absorption efficiency for a pollutant is related to the residence time of the material in the gut, and the residence time should vary for different sizes of particles which the animal may ingest.

Transuranic elements are usually in almost insoluble chemical forms when found in environmental media. It has been shown, however, that relatively insoluble forms of plutonium, for example, are solubilized to a considerable degree in the gastrointestinal environment of cattle. Information on the gastrointestinal passage rates for various soil particle sizes is, therefore, relevant to the potential absorption efficiency for substances entrained in debris ingested by animals grazing in contaminated areas.

This study was performed to elucidate the variability in bovine gastrointestinal passage rates following oral ingestion of sand particles of various sizes. Silica sand of four graded size ranges

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*See footnote, p. 35.

was obtained, and each size range was labeled with a gamma-emitting radioisotope. The sand sizes and isotopes used were:

Particle Size	Isotope Label
15-25 μm	^{141}Ce
74-88 μm	^{85}Sr
175-246 μm	^{54}Mn
417-495 μm	^{46}Sc

The particles were washed with dilute hydrochloric acid (pH 1), which removed less than 1% of the activity. Each size range was divided into four portions and placed in gelatin capsules. One capsule (20 g for the three smaller sizes, 40 g for the largest particles) of each size was orally administered to each of four lactating Holstein dairy cows maintained in metabolism stalls. The cows were fed their normal dairy ration of hay and pelleted food; water was provided *ad libitum*. Urine was collected by means of an indwelling catheter. Fecal material from each cow was collected, weighed, and mixed to obtain a 24-hr composite sample. Samples for each cow were analyzed by gamma-ray spectroscopy to determine the content of labeled particles. Samples of urine, blood, and milk were analyzed to make sure the tracer radionuclides did not leach from the particles *in vivo*.

Results of analyses on urine, blood, and milk showed that no significant *in vivo* leaching of the radionuclides from the particles occurred, as was intended in the study design.

The fecal composition was similar for the four cows, and, although the daily fecal outputs per cow varied by about 25%, the labeled sand excretion did not appear to be related to the amount of daily fecal output. Although variations were observed among the four cows in the retention times for the various sized particles, longer retention times were consistently associated with the larger particles.

The smallest particles (15-25 μm) were excreted rapidly in all cows. Half of these particles were excreted within 36 hrs and 90% within 91 hrs of dosing. These times corresponded to about 40 and 110 kg of feces elimination, respectively. The three larger size particle

groups were excreted more slowly and with large variations among cows. Up to 168 hrs were required to excrete 50% of the particles and up to 288 hrs to excrete 90%. These times correspond to fecal elimination of about 230 and 400 kg, respectively.

The data show that the passage time for soil particles through the gastrointestinal tract of dairy cows varies significantly with particle size. Some of the sand particles which cattle may ingest have gastrointestinal residence times of greater than a week. If the particles should contain relatively insoluble transuranic elements, this period would provide more time for reactions involved in gastrointestinal absorption, e.g., chelation and soluble compound formation, than would be derived from conventional studies carried out in ruminant digestion investigations.

BIOLOGICAL TRANSFER OF PLUTONIUM-238 VIA *IN VIVO* LABELED MILK

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Despite the fact that the biological transport of plutonium has been studied for many years, quantitative values for its transfer to milk, its distribution in tissues (both edible and nonedible), and its subsequent uptake by suckling calves or humans have not been established. The basis for this study, currently in progress at the NTS farm, is that people especially children consume large quantities of dairy products and beef (muscle and organs) so that any evaluation of radiological hazards associated with a plutonium-contaminated countryside must consider the biological availability of *in vivo* plutonium-labeled food (milk and meat).

During the range-finding portion of this study, a lactating dairy cow was given a single 83-mCi intravenous dose of plutonium-238 citrate. Milk, blood, urine, and feces were collected for analysis until the cow was sacrificed three days after dosing. It was determined that this amount of plutonium was too high for the objectives of this study.

Another lactating cow was then given an acute 10-mCi plutonium-238 citrate intravenous injection. As before, collections of milk, blood, urine, and feces were taken for analysis. In both cases, preliminary results indicate that slightly less than 1% of the original dose had been secreted in the milk 72 hrs after injection. The second animal has subsequently been given three additional intravenous doses (10 mCi per day for three consecutive days). Milk collections are being placed in suitable buckets and fed to two calves at a rate

*See footnote, p. 35.

of 10 pounds of milk per animal per day for five consecutive days. Prior to feeding, aliquots of milk are analyzed with the Phoswich detector. The calves will be sacrificed 96 hrs after the last treatment feeding, at which time tissues will be taken for analysis.

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Despite the fact that the biological transport of plutonium has been studied for many years, quantitative values for its transfer to milk, its distribution in tissues (both edible and nonedible), and its subsequent uptake by suckling calves or humans have not been established. The basis for this study, currently in progress at the NTS farm, is that people especially children consume large quantities of dairy products and beef (muscle and organs) so that any evaluation of radiological hazards associated with a plutonium-contaminated countryside must consider the biological availability of *in vivo* plutonium-labeled food (milk and meat).

During the range-finding portion of this study, a lactating dairy cow was given a single 83-mCi intravenous dose of plutonium-238 citrate. Milk, blood, urine, and feces were collected for analysis until the cow was sacrificed three days after dosing. It was determined that this amount of plutonium was too high for the objectives of this study.

Another lactating cow was then given an acute 10-mCi plutonium-238 citrate intravenous injection. As before, collections of milk, blood, urine, and feces were taken for analysis. In both cases, preliminary results indicate that slightly less than 1% of the original dose had been secreted in the milk 72 hrs after injection. The second animal has subsequently been given three additional intravenous doses (10 mCi per day for three consecutive days). Milk collections are being placed in suitable buckets and fed to two calves at a rate

*See footnote, p. 35.

of 10 pounds of milk per animal per day for five consecutive days. Prior to feeding, aliquots of milk are analyzed with the Phoswich detector. The calves will be sacrificed 96 hrs after the last treatment feeding, at which time tissues will be taken for analysis.

Small Vertebrates

ECOLOGICAL STUDIES OF SMALL VERTEBRATES IN PU-CONTAMINATED STUDY AREAS OF NEVADA TEST SITE AND TONOPAH TEST RANGE

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ABSTRACT

Various standard census methods were employed during the period March, 1972-May, 1975, to obtain a qualitative and quantitative inventory of the vertebrate biota in seven Nevada Applied Ecology Group (NAEG) intensive study areas of the Nevada Test Site (NTS) and Tonopah Test Range (TTR). In general, vertebrate populations in NAEG study areas correspond with earlier investigations of populations in uncontaminated study areas.

Animals known to be residents in the study areas for at least three months were sacrified and autopsied. Tissue samples for radioanalysis included pelt or skin, GI tract, and carcass. Over 300 tissue samples have been collected and shipped to laboratories for ^{239}Pu and ^{241}Am determinations. However, many of these data are forthcoming. Examination and discussion of many aspects of Pu and Am uptake await completion and receipt of laboratory analyses.

Data are presented on Pu and Am tissue burdens in *Dipodomys microps*, a granivorous rodent, from three study areas of NTS and TTR. Ratios of Pu/Am were significantly lower in the carcass than in the pelt or GI tract in this rodent, whereas ratios in tissue of lizards were uniform. A general trend of preferential Am uptake is suggested in certain rodents. It is suggested that the food habits and burrowing behavior of these rodents may be contributing factors to this apparent trend.

Some base line hematological data obtained from 50 rodents collected from uncontaminated control areas off-site are presented. These data will be used as a basis for comparison with blood values obtained from resident animals of NAEG intensive study areas.

Possible future plans are discussed, such as reducing the number of continuous study areas, and intensifying the investigation of certain areas to include preliminary observations on the availability of Pu and Am in rodent burrows.

INTRODUCTION

Ecological studies of vertebrates in the three Pu-contaminated areas of NTS began in spring, 1972, and were expanded to include four areas of TTR in fall, 1973. During the initial phases of this study, emphasis was on the development and improvement of standardized procedures for inventory, census, and collection of small vertebrates. This methodology is discussed in previous reports (Moor and Bradley, 1974; Bradley and Moor, 1975). This report includes analysis of data obtained during the contract period from the continuous study of seven areas of NTS and TTR. In addition, hematological studies of lizards and rodents began in fall, 1974. Emphasis during the report period has been on collecting vertebrates from areas off-site to obtain base line hematological data for comparisons with data which will be gathered from resident vertebrates of NAEG intensive study areas in the summer of this year.

MATERIALS AND METHODS

Census Methods

Various standard census methods were employed to obtain a qualitative and quantitative inventory of the vertebrate biota in each NAEG

intensive study area (Moor and Bradley, 1974). Permanent live-trapping grids were established in the study areas. Utilizing capture-recapture techniques and a system of toe-clipping to enable individual recognition of animals, data on population densities, biomass, and seasonal activities were gathered.

Radioanalysis

Individual animals known to be residents in the study areas for at least three months were captured, sacrificed, and taken to the CETO building in Mercury, where they were autopsied. Animals were also collected off-site for analysis following the same procedures used in NAEG study areas. Tissue samples for radioassay included pelt or skin, GI tract, and carcass. Laboratory procedures for preparation of tissue samples have been reported (Moor and Bradley, 1974) and included dipping animals in hot paraffin wax to minimize cross-contamination of respective subsamples. Levels of ^{241}Am and ^{239}Pu in the tissue samples were determined by LFE Environmental Analysis Laboratories, Richmond, California.

Hematological Studies

The following procedures were used to obtain base line hematological data from animals collected off-site and will be used to process resident animals collected in NAEG intensive study areas.

Animals were captured alive utilizing Sherman live traps or nooses and returned to the laboratory and examined within 24 hrs of capture. Blood was obtained from rodents by a cardiac puncture using ether as an anesthetic. Blood was obtained from lizards by decapitation. In all cases, blood was collected in heparanized (ammonium sulfate) syringes. All analyses were done in duplicate, and any test exceeding a 1% difference was repeated. All analyses were accomplished following standard methods (Helper, 1966).

RESULTS AND DISCUSSION

Vertebrate Census

Species composition, relative abundance, and seasonal status of vertebrates in NAEG study areas have been presented in earlier reports (Moor and Bradley, 1974; Bradley and Moor, 1975) and in general agree with other investigations at NTS (Hayward *et al.*, 1963; Tanner and Jorgensen, 1963; Jorgensen and Hayward, 1965).

Probably there have been major changes in the rodent fauna in two study areas of NTS since the last formal report. Table 1 presents population estimates of rodents in Area 5 and Area 11 in the springs of 1973, 1974, and 1975. The rodent fauna in Area 5 appears to have increased dramatically from 1973 (0.38 animals/hectare) to 1975 (3.39 animals/hectare). There is an apparent increase in two important granivores, *Dipodomys merriami* and *D. microps*, which now comprise over 90% of the rodent fauna in Area 5. This increase is attributed to the more abundant precipitation and production of green plants during and following 1973. The dependence of the desert fauna in southern Nevada upon green vegetation for successful reproduction has been reported previously (Beatley, 1971; Bradley and Mauer, 1971). Conversely, Area 13 had undergone an apparent drastic reduction in rodent species in richness and numbers. During the springs of 1973 and 1974, numbers of rodents were estimated at 3.5 animals/hectare. During spring of 1975, however, estimates were 0.52 animals/hectare. This reduction of rodents is probably due to the activities of large herbivores (cattle and goats) introduced as part of the grazing study in summer, 1973. Whereas more data need to be gathered before the extent of the impact of herbivores on the rodent fauna can be properly evaluated, definite destructive alteration of certain rodent habitats within the enclosure of Area 13 is evident.

Radioanalysis

One hundred resident vertebrates from NAEG intensive study areas and 20 control animals from off-site have been collected, autopsied, and

Table 1. Population Estimates of Rodents in Two Study Areas of NTS in Spring of 1973, 1974, and 1975. (Estimates are in number/hectare.)

	Area 5		Area 13		
Species	1973	1975	1973	1974	1975
Ammospermophilus leucurus	0.05	0.73	0.22	0.71	0.26
Dipodomys merriami	--	1.16	0.35	--	0.26
Dipodomys microps	0.09	1.95	1.70	0.69	--
Microdipodops megacephalus			0.15	--	--
Onychomys torridus	0.05	0.05	0.05	--	--
Perognathus longimembris	0.19	0.04	1.12	1.62	--
Peromyseus eremicus			0.09	--	--
Peromyscus maniculatus			--	0.37	--
Total Number of Species	4	5	7	4	2
Total Number of Animals/Hectare	0.38	3.39	3.68	3.39	0.52

shipped for radioanalysis. A summary of ^{239}Pu and ^{241}Am tissue burdens in vertebrates of NAEG study areas is contained in the last formal report (Bradley and Moor, in press). Few additional data were available for analyses to include in this report. Although many data await further analysis, certain trends appear evident and discussion appears justifiable.

Table 2 presents ^{239}Pu and ^{241}Am tissue burdens in *D. microps* from three study areas of NTS and TTR. An interesting trend is indicated here. Whereas Pu/Am ratios are variable in tissue samples, they are significantly lower in the carcass than in the pelt or GI tract ($P > .05$) of animals collected in the three study areas. For example, *D. microps* from Clean Slate 2 had significantly higher ratios ($P > .01$) in the pelt (27.6) and GI tract (18.2) than in the carcass (9.4). Preliminary results suggest preferential Am uptake is evident in rodents. Conversely, Pu/Am ratios in tissues of lizards were remarkably uniform. Nine lizards from Area 11 had Pu/Am ratios of 6.85 in the skin, 6.81 in the GI tract, and 6.85 in the carcass. There are several possible explanations for the differences in Pu/Am ratios in lizards and rodents. The rodents examined thus far have been granivores, the lizards insectivorous. Romney *et al.* (1974) indicate that preferential uptake and concentration of ^{241}Am through plant roots might have occurred in Area 13. Whereas the Pu/Am ratio is somewhate higher in vegetation than in animal tissue in Area 13, no data could be found on the distribution of Pu and Am within tissues and organs of individual plants. Additionally, no data on Pu and Am tissue burdens in insects could be found. These data should be available before further analyses of trophic transfer can be properly evaluated. Fowler and Essington (1974) indicated that Am is moving faster downward in the soil than Pu. Because rodents in general are more or less fossorial and lizards are not, perhaps Am is more available to rodents in the microhabitat of their burrows. The decrease in Pu/Am ratios may be explained on the basis of differential solubilities, and Am may be more available for absorption in the more advanced digestive tract of rodents.

Table 2. ^{239}Pu and ^{241}Am (NCi/g ash) in Tissue Samples of *Dipodomys microps* from Three Intensive Study Areas of NTS and TTR. (Range in parentheses.)

Area	Sample	^{239}Pu N	^{239}Pu $\bar{X}$ + SE	^{241}Am N	^{241}Am $\bar{X}$ ± SE	Pu/Am Ratio N	Pu/Am Ratio $\bar{X}$ ± SE
Area 11, NTS	Pelt	3	0.14 ± 0.10 (0.0027 - 0.338)	6	0.01 ± 0.004 (0.0018 - 0.047)	3	6.57 ± 0.91 (4.77 - 7.81)
	GI Tract	6	0.08 ± 0.06 (0.0045 - 0.068)	6	0.01 ± 0.008 (0.00086 - 0.052)	6	7.32 ± 1.46 (6.63 - 14.08)
	Carcass	6	0.0009 ± 0.0004 (0.00009 - 0.0035)	6	0.0002 ± 0.00004 (0.00007 - 0.0006)	6	3.29 ± 1.23 (0.78 - 8.13)
Area 13, NTS	Pelt	9	1.05 ± 0.26 (0.069 - 2.44)	10	0.10 ± 0.0283 (0.0036 - 0.287)	9	8.26 ± 0.16 (2.61 - 16.28)
	GI Tract	9	0.65 ± 0.48 (0.037 - 4.74)	10	0.084 ± 0.052 (0.0015 - 0.549)	9	5.56 ± 0.80 (3.85 - 8.63)
	Carcass	10	0.047 ± 0.002 (0.00018 - 0.0227)	1	0.0034	1	6.70
Clean Slate 2, TTR	Pelt	6	0.30 ± 0.06 (0.12 - 0.41)	10	0.009 ± 0.001 (0.0005 - 0.021	5	27.62 ± 2.45 (19.29 - 36.05)
	GI Tract	7	0.51 ± 0.38 (0.08 - 2.81)	7	0.15 ± 0.11 (0.001 - 0.826)	4	18.19 ± 1.50 (9.40 - 21.84)
	Carcass	6	0.007 ± 0.002 (0.0008 - 0.0204)	6	0.0007 ± 0.0004) (0.00004 - 0.0030)	3	9.36 ± 0.51 (8.46 - 10.24)

Hematological Studies

Base line hematological data obtained from 50 rodents (3 species) and 22 lizards (5 species) from off-site areas of southern Nevada have geen gathered. Blood cell counts and blood composition values for some rodents are presented in Tables 3 and 4. These and additional data from off-site areas will be used as a basis for evaluating blood values obtained from resident animals of NAEG intensive study areas. These animals will be sacrificed in the summer of this year, at which time they will have been residents in Pu-contaminated areas for at least three months.

FUTURE PLANS

After nearly three years of ecological study, the small vertebrate composition of seven NAEG intensive study areas is reasonably well known. Over 350 vertebrate tissue samples have been collected and shipped for radioanalysis. Based on preliminary data, certain problems and trends are apparent:

1. Access to certain NAEG study areas is limited and unpredictable due to other ERDA activities.

2. Climatic conditions strongly influence small vertebrate activities and population estimates.

3. Pu tissue burdens of animals collected from the same strata and sampling sites are not consistent and vary widely.

4. Pu/Am ratios vary between species, between individuals of the same species, and between tissues of individuals. Some data suggest that there is a preferential uptake of Am in at least certain species.

To facilitate a better understanding of population densities and trophic relationships as they relate to Pu and Am availability, uptake, and deposition, the following is proposed:

Table 3. Average Number and Range of Blood Cell Types for Some Vertebrates Found in Southern Nevada.

Species	N	Leukocytes $1000/cm^3$	Neutrophils (%)	Eosinophils (%)	Basophils (%)	Lymphocytes (%)	Monocytes (%)	Erythrocytes $1,000,000/cm^3$
Dipodomys merriami	8	12.5 (6.5-16.0)	22.0 (12.0-44.0)	2.4 (0-6.0)	0.5 (0-1.5)	73.0 (66.0-86.0)	3.0 (0-8.0)	9.1 (7.2-11.8)
Dipodomys microps	6	14.0 (8.0-21.0)	24.0 (10.0-35.0)	2.0 (0-5.0)	0.5 (0-1.0)	71.0 (52.0-83.0)	4.0 (0-15.0)	8.1 (7.5-9.6)

Table 4. Blood Composition Values for Some Vertebrates Found in Southern Nevada. (Mean, standard error, range.)

Rodents	N	Body Weight (g)	Packed Cell Volume (%)	Hemoglobin (g/100 ml)	Total Protein (g/100 ml)	Glucose (mg/100 ml)	Cholesterol (mg/100 ml)	Albumin (g/100 ml)
Dipodomys merriami	18	37.2 0.6 28.0-42.0	51.0 0.9 48.0-55.0	15.9 0.4 13.9-17.3	6.1 0.1 5.5-6.9	177.8 4.3 146.0-204.0	191.2 2.4 178.0-200.0	1.8 0.1 1.4-2.6
Dipodomys microps	14	56.0 1.5 47.0-63.0	46.8 1.3 40.0-55.0	15.7 1.1 14.0-17.0	6.2 0.7 4.5-7.4	181.0 3.0 160.0-193.0	185.1 2.8 165.0-200.0	
Peromysus maniculatus	18	19.8 0.7 16.4-25.0	40.5 0.1 32.0-48.0	12.4 1.8 9.0-13.8	7.0 0.9 6.0-8.0			

1. The establishment of permanent grids in one study area of NTS (Area 11) and two study areas of TTR (Double Track and Clean Slate 3). These areas are the most accessible and continuous sampling would be facilitated.

2. Each study area would have two separate permanent grids, one for establishing residency of animals which can then be sacrificed for radioanalysis, histopathology, or blood sampling and one for monitoring population densities including reproduction, recruitment, and mortality.

3. In study areas of low priority in which previous sampling has been done, the resident animals will be sacrificed to increase the number of samples for radioanalysis.

Data indicating preferential uptake of Am are interesting in light of data presented by Fowler and Essington (1974) on Am movement in the soil. An analysis of Pu and Am in rodent burrows would provide additional data on availability and uptake of Am in this microhabitat. Correction of sampling problems is now under consideration.

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Microorganisms

POSSIBLE INFLUENCE OF DESERT SOIL MICROBIAL CHANGES

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ABSTRACT

The soil microbial population was determined for soil depth segments from 0-3 cm, 3-6 cm, and 6-9 cm in a previously uncultivated high-desert area. Plots in this area were covered by miniature greenhouses and planted with vegetables. After harvesting the vegetable crops, another assessment was made of the microbial population.

The fungal population nearly doubled during the growing season, and the bacterial population increased by a factor of approximately 12. The fungal and bacterial live weights per hectare of the investigated desert soil were calculated and compared with those of agricultural soils. Possible consequences of the increase in microbial biomass are discussed for plutonium availability to plants, and for plutonium migration in soil.

INTRODUCTION

Soil fertility can be defined as the ability of the soil to supply nutrients to plants (Bollen, 1959). This ability depends largely on the activities of soil microorganisms. It is well known that soil microorganisms are responsible for the conversion of atmospheric and mineral constituents into forms which can be assimilated by plants.

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*See footnote, p. 35.

This explains why plants can be grown and harvested from the same plot year after year without the need for man to replace the nitrogen that is continuously removed by harvested crops (Stewart, 1966). In this case, most of the nitrogen used by plants enters the soil via fixation of atmospheric nitrogen by soil microorganisms. The agricultural soils of the Tigris and Euphrates valleys located in Iraq are examples of such agricultural practices. Even after centuries of tillage, these soils continue to yield crops, and their perpetual fertility can be largely attributed to nitrogen fixation and to the conversion of soil minerals into soluble forms by soil microorganisms (Chandra *et al.*, 1962).

Desert areas in temperate zones of the North American continent have been defined as regions "of hot daytime summer temperatures, higher than average evaporation rates, and . . . less than ten inches of precipitation annually" (Wheeler, 1971); consequently, they are envisioned by many people as barren wastelands. The newcomer to desert areas of the western United States may be surprised to see lush vegetation generated by seasonal precipitation in the higher elevations. Even in this arid climate, the existence of a viable plant community depends on microbial activities in soil to provide the plants with nitrogen from the atmosphere and with other inorganic plant nutrients. Thus, a "sterile desert," microbiologically speaking, is a misnomer because fungi and bacteria are present even in extremely arid soils of the Atacama desert of Chile, an area which is nearly devoid of vegetation (Cameron, 1969).

Nitrogen fixation has been reported for certain soil bacteria (Alexander, 1961) and for certain fungal strains of *Aspergillus*, *Mucors*, *Penicillium*, and *Botrytis* (Waksman, 1927; Griffin, 1972; Alexander, 1961). One of these nitrogen-fixers, *Aspergillus*, is the most prevalent fungus in some of the Iraqi soils mentioned earlier, as determined by microbial analyses (Chandra *et al.*, 1962); this fungus was also found in different soil types of the Nevada Test Site (NTS) (Durrell and Shields, 1960; Au, 1974).

It was reported earlier that plutonium when added as soluble and insoluble compounds to culture media was absorbed by *Aspergillus*

niger and transported to its spores (Au and Beckert, 1974; Au *et al.*, 1975). This suggests that soil fungi, and probably bacteria, could facilitate the transport of radionuclides in soil and enhance their transfer from soil to plants. Therefore, the laboratory became interested in fluctuations of soil microbial populations caused by agricultural use of a desert soil, and in their possible consequences for plutonium translocation in soil and plutonium uptake from soil by other organisms. Changes were assessed of the soil microbial biomass which resulted from controlled plant growth and irrigation under simulated agricultural conditions. These changes were studied as part of a field investigation designed to determine plutonium uptake by radishes and lettuce under controlled conditions from previously uncultivated soil in Area 13 of NTS.

METHODS AND MATERIALS

Small, transportable greenhouses were placed over two 1.2 m x 1.2 m plots in Area 13 of NTS. Trenches were dug for their emplacement so that the greenhouse structures extended approximately 30 cm below the soil surface. The access doors were sealed to prevent surface contamination by dust. The air in the greenhouse was recirculated and cooled or heated with portable temperature-modifying units. Each 1.2 m x 1.2 m plot was subdivided into four 0.6 m x 0.6 m subplots.

The soil surface of experimental areas was covered with plastic sheets with 2 cm holes arranged in an 8 cm grid pattern. These sheets eliminated surface contamination of the aerial plant parts from soil within the greenhouses. Radish and lettuce seeds were placed through the holes into undisturbed soil. Four different irrigants were used on the crops: distilled water, water containing the disodium salt of diethylenetriaminepentaacetic acid (DTPA), water containing inorganic fertilizer (ammonium nitrate plus phosphoric acid), and water containing both DTPA and the inorganic fertilizer. DTPA and inorganic fertilizer were applied at a rate corresponding to 2,750 kg per hectare for each of these components.

Prior to the placement of the greenhouses and planting of vegetable seeds, soil samples were taken from the 0-3, 3-6, and 6-9 cm segments of the vertical trench profile. Similarly, soil samples were collected during the harvest of mature plants from within the greenhouses at the same depth intervals. Forty-eight soil samples were collected. Previous experiments in NTS soils had shown that no major changes in the microbial population could be expected to occur during the time period of these experiments (Au, 1972). Accordingly, no further soil samples were collected from outside of the greenhouse structures. Soil moisture of the collected soil samples was determined in separate experiments according to standard procedures (Pramer and Schmidt, 1964).

Dilution plate counts were made of the collected soil samples to quantify and differentiate the fungal and bacterial populations. From each soil sample, an amount corresponding to 20 g of oven-dried soil was added to 100 ml of sterile distilled water and shaken for 10 min. Coarser particles were allowed to settle, and 1-ml samples of the soil suspension were used to prepare dilutions for plate counts according to standard procedures (Clark, 1965). The dilutions used were 5×10^{-2} and 5×10^{-3} for molds, and 5×10^{-4} and 5×10^{-5} for bacteria and *Streptomyces* counts. Five plates were poured of each dilution. Cooke's Rose Bengal agar with antibiotic added was used for fungal determinations, and sodium albuminate agar was used for bacteria and *Streptomyces* determinations. The plates were incubated at room temperatures; fungal and bacterial counts were made after 2 and 14 days, respectively. In addition to the microbial plate counts, the classification and the relative abundance (in percent of the total fungal and bacterial numbers) were determined for the fungi *Mucors*, *Aspergillus*, *Penicillium*, and *Dematiaceae* as well as for the bacterial genus *Streptomyces*.

RESULTS AND DISCUSSION

In this discussion, only the overall changes of microbial population caused by the agricultural use of the soil will be highlighted. More

Table 1. Differential Soil Microbial Populations for Three Soil Depth Segments Before Planting and at Harvest*

	Soil Depth Segment (cm)	FUNGI						BACTERIA		
		Total Numbers per Gram of Oven-Dry Soil (Thousands)	% *Mucors*	% *Aspergilli*	% *Penicillia*	% *Demataiceae*	% Others	Total Numbers per Gram of Oven-Dry Soil (Millions)	*Streptomyces* Numbers per Gram of Oven-Dry Soil (Millions)	*Streptomyces* %
Before Planting	0-3	4.2 ± 1.6	22 ± 6	7 ± 4	11 ± 4	22 ± 4	39 ± 7	3.9 ± 0.5	0.9 ± 0.02	24 ± 3
	3-6	9.6 ± 3.2	10 ± 2	25 ± 8	36 ± 10	2 ± 1	27 ± 5	4.7 ± 0.6	1.8 ± 0.02	39 ± 3
	6-9	12.4 ± 4.3	10 ± 4	27 ± 7	42 ± 12	1 ± 0.5	21 ± 5	4.5 ± 0.6	2.0 ± 0.02	44 ± 3
	0-9	8.7 ± 3.0	14 ± 4	20 ± 6	30 ± 9	8 ± 2	29 ± 6	4.4 ± 0.6	1.6 ± 0.02	36 ± 3
At Harvest	0-3	27.9 ± 8.1	48 ± 7	14 ± 4	22 ± 10	3 ± 1	17 ± 5	101.3 ± 30.2	9.1 ± 1.2	9 ± 4
	3-6	12.6 ± 3.1	28 ± 5	10 ± 4	7 ± 2	25 ± 9	29 ± 8	37.4 ± 13.4	4.5 ± 0.4	12 ± 3
	6-9	10.7 ± 3.7	26 ± 11	2 ± 1	31 ± 12	19 ± 7	23 ± 7	16.8 ± 5.1	3.0 ± 0.2	18 ± 3
	0-9	17.1 ± 5.0	34 ± 8	9 ± 3	20 ± 8	16 ± 6	23 ± 7	51.8 ± 16.2	6.7 ± 0.5	13 ± 3

*All values are expressed as the average ± 1 standard error.

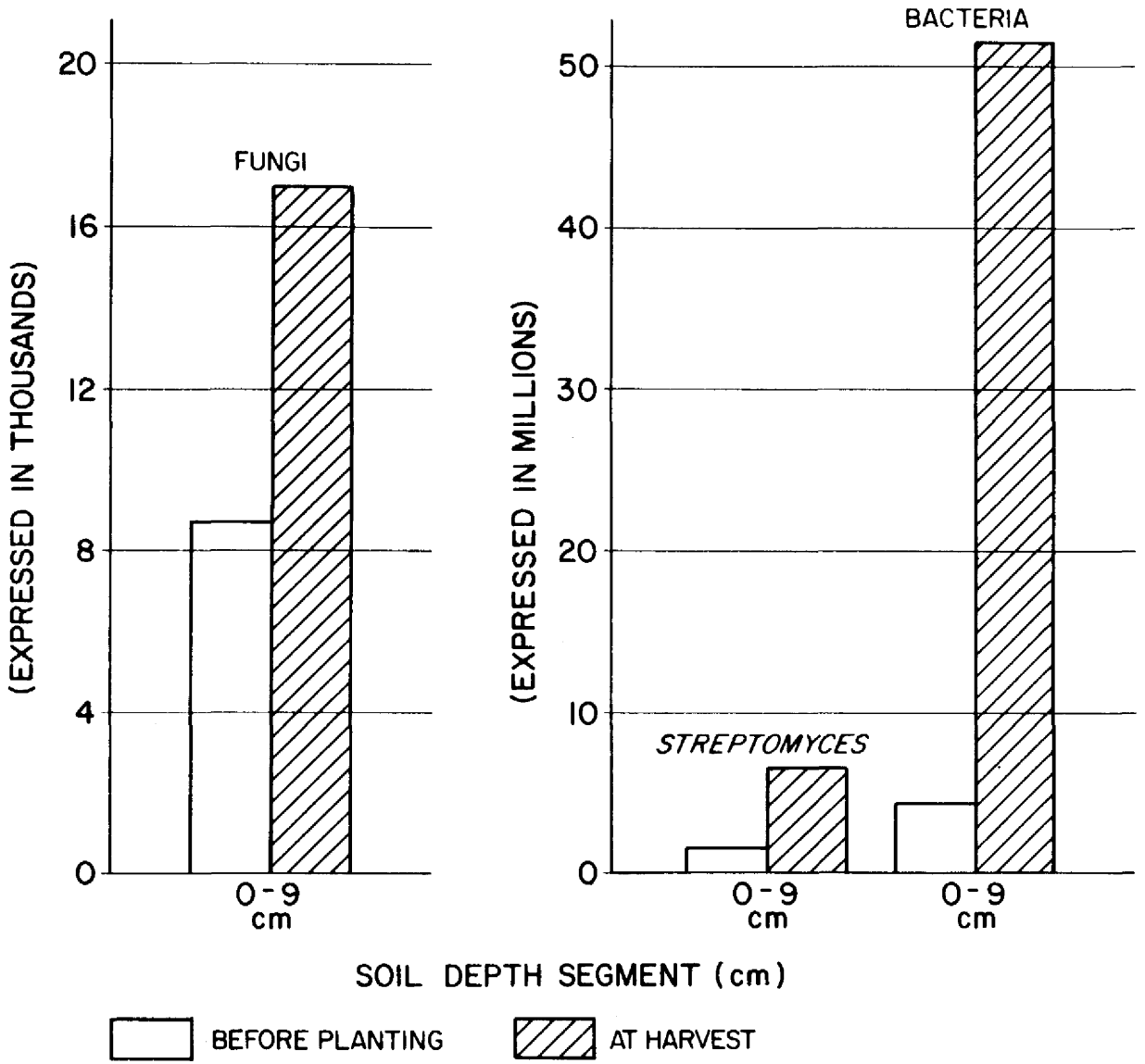

FIGURE I. TOTAL NUMBERS OF FUNGI, BACTERIA AND *STREPTOMYCES* PER GRAM OF OVEN DRY SOIL.

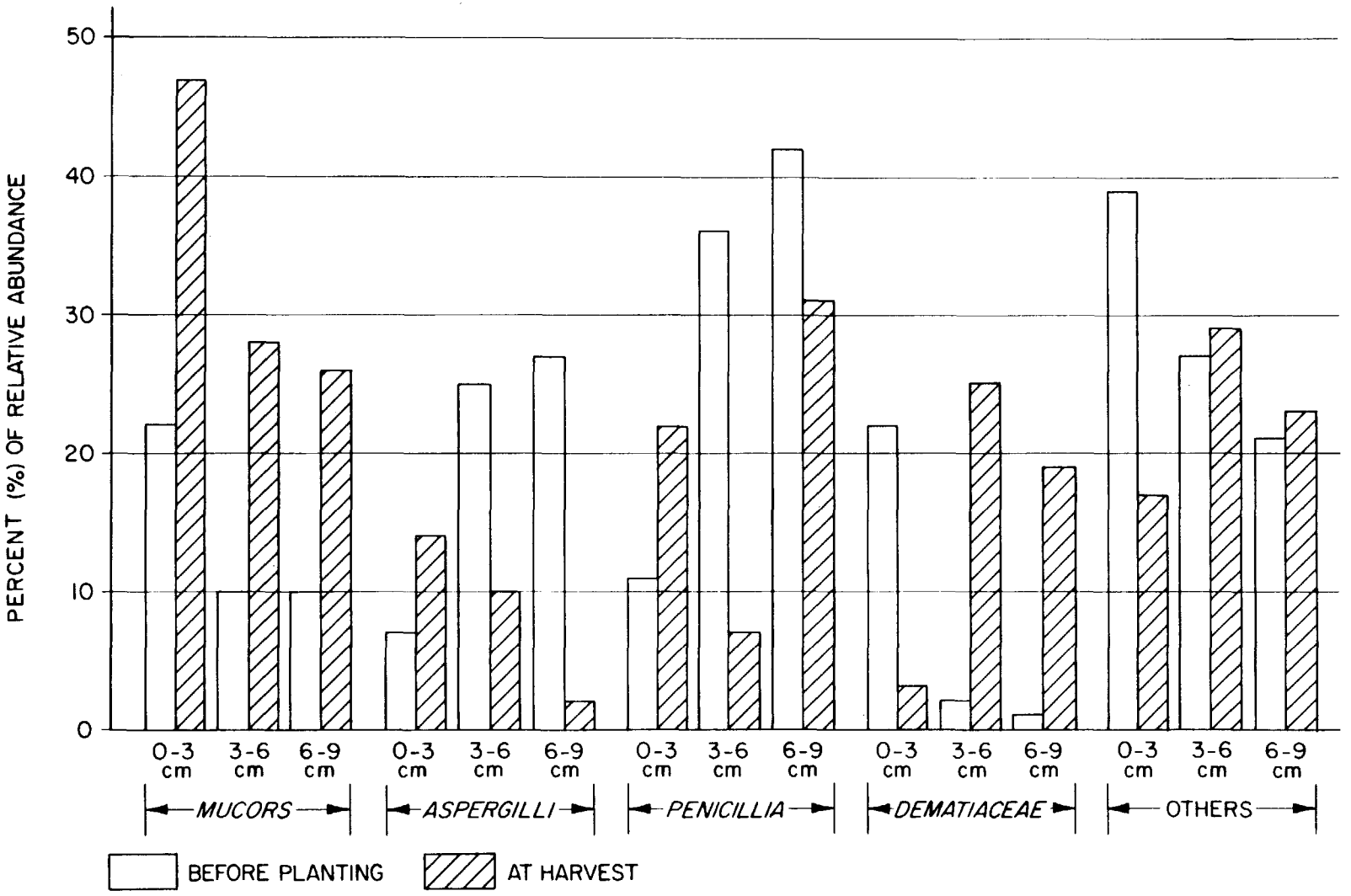

FIGURE 2. RELATIVE ABUNDANCE OF FUNGI AT 3 SOIL DEPTHS (0-3, 3-6, & 6-9 cm) BEFORE PLANTING & AT HARVEST.

subtle differences arising from the use of different irrigant treatments on the microbial population will be detailed elsewhere.

The average soil microbial populations of the eight subplots are presented in Table 1. A comparison of the total numbers of fungi and bacteria in the entire soil profile (0-9 cm) is plotted in Fig. 1. It shows that the fungal population nearly doubled and the bacterial population increased by a factor of approximately 12 during the growth period. The relative abundance of the various fungi identified for the three different soil depths is graphically shown in Fig. 2.

The relative abundance (Fig. 2) of *Mucors* before planting was highest in the 0-3 cm soil segment (22%) and about one-half of that value for the 3-6 cm and 6-9 cm depth segments (10%). At harvest, the relative abundance of *Mucors* had increased by a factor of two for all depth segments, while the distribution pattern as a function of depth was nearly the same as before planting.

In general, *Aspergillus* increased with soil depth from 7% at 0-3 cm to 27% at 6-9 cm before planting. At the time of harvest, *Aspergillus* had increased by a factor of 2 in the 0-3 cm depth segment (from 7 to 14%), but decreased to approximately 1/14 of its original value in the 6-9 cm depth segment (from 27 to 2%).

Before planting, the relative abundance of *Penicillium* increased with soil depth from 11% (0-3 cm) to 42% (6-9 cm). At harvest, it was twice as high (22%) in the 0-3 cm depth segment, but lower in the remaining segments.

The relative abundance of *Dematiaceae* before planting was highest at 22% in the 0-3 cm depth segment and lowest at less than 1% in the 6-9 cm depth segment. At harvest, the lowest count of *Dematiaceae* at 3% was in the 0-3 cm depth segment. It increased with depth to 25% in the 3-6 cm segment and to 19% in the 6-9 cm segment.

As for bacteria, the percentage of *Streptomyces* (Table 1) increased in general with soil depth before planting as well as at harvest.

It is quite revealing to compare the live weights of the desert microflora with those of more fertile soils. It has been estimated by Bollen (1959) that one hectare of fertile soil down to a depth of 17 cm (plough depth) can contain as much as 1,250 kg of bacteria and actinomycetes each and up to 2,500 kg of fungi, or a total of up to 5,000 kg of soil microorganisms. One can also estimate the live weight of desert microflora from data such as presented in Table 1. If one very conservatively assumes that for the 0-9 cm depth segment a moderately fertile soil has a live weight of only 288 kg of fungi per hectare, as cited by Alexander (1961), then one has about 35,000 fungi in every gram of this soil. Since experimentally 8,700 fungi per gram were found in NTS Area 13 soil, one can estimate a live weight of approximately 70 kg of fungi per hectare of the NTS soil before planting. Using the same basis of comparison for the 17,100 fungi per gram of NTS Area 13 soil found at harvest, the live weight would be approximately 140 kg per hectare, an increase of about 100%. Similarly, if one assumes a very conservative live weight of 175 kg of bacteria per hectare in the 0-9 cm depth segment of a moderately fertile soil at a population count of 115 million per gram of soil (Alexander, 1961; Clark, 1967), then the 4.4 million bacteria per gram determined for the NTS soil before planting would amount to a live weight of nearly 7 kg per hectare. Using the same method for the population of 52 million bacteria per gram at harvest, one may calculate a live weight of about 79 kg of bacteria per hectare, or a 12-fold increase in the live weight of bacteria in NTS soil.

The above results demonstrate that microbial activity in desert soil increases with increasing plant growth. This increase in activity is due at least in part to an increased supply of nutrients, especially root exudants. Since the common soil fungus, *Aspergillus niger*, can assimilate plutonium, it is reasonable to assume that other soil fungi, such as *Penicillium*, and probably soil bacteria, may possess similar abilities. Plutonium assimilation by soil organisms obviously requires the formation of complexes which can penetrate the microbial cell wall. Upon death of the microbial cells, the cellular contents

including any incorporated plutonium become available to other organisms. We do not know in what complexed form or forms plutonium is stored in the microbial cells, but whatever form(s) these may be, it is very probable that this complexed plutonium when released from the microbial cells is more soluble than the relatively insoluble plutonium compounds thus far found deposited in soil; consequently, it will be more biologically available. This suggests that an increase of microbial population during the growing season could result in a sudden increase in the amount of plutonium available to other trophic levels.

Besides making plutonium more available to other organisms, soil microorganisms can also play a role in plutonium transport in soil. Krasilnikov, who found that naturally occurring actinides are absorbed by soil microorganisms (1958), claimed that the movement of soil microbial cells containing natural radioactive elements determined the migration of these substances (1967). Thus, the soil fungus *Aspergillus niger*, which absorbs and translocates plutonium, and other plutonium-absorbing soil microorganisms, could be moved to other locations within the soil system. Upon death of the microbial cells, any cellular plutonium not assimilated by another organism would become available for translocation by water, possibly in a chemical form which would facilitate this translocation.

The activities of mobile predators such as nematodes and protozoa further complicate the picture. These predators and other animals feed on soil microorganisms and thus spread any plutonium that is incorporated in their food. Obviously, the number of these predators will be approximately proportional to the number of microorganisms present in the soil.

It was pointed out earlier that the typical desert of western United States is not a barren wasteland, but supports some vegetation. The above results and discussion show that increased plant activity results in an increase in the soil microbial population. This in turn, in plutonium-contaminated soils, could increase the amount of biologically available plutonium during the growing season as well as cause a seasonal increase in plutonium translocation in the soil.

Very similar mechanisms for plutonium transformation and translocation are probably equally valid for soils of other areas, a factor to be considered in selections of nuclear sites for intensive study.

ACKNOWLEDGMENT

The technical assistance of John G. Homewood is gratefully acknowledged.

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Distribution and Inventory

PLUTONIUM DISTRIBUTION IN THE ENVIRONS OF THE NEVADA TEST SITE--STATUS REPORT, APRIL, 1975

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ABSTRACT

The U.S. Environmental Protection Agency's National Environmental Research Center at Las Vegas (NERC-LV) has been investigating plutonium in the environs of the Nevada Test Site (NTS) since 1970. Sampling and analysis of soil, air, water, and vegetable material continued through Fiscal Year 1975. This report summarizes the results of these efforts through April of Fiscal Year 1975.

Plutonium concentrations in soil and air correspond to expected levels, i.e., similar to those reported at the 1974 Plutonium Information Meeting--Nevada Applied Ecology Group. No plutonium was detected in fruits and vegetables grown near the NTS; however, positive values possibly as great as 24 pCi/kg wet weight were detected in samples of vegetables prepared for human consumption and purchased from a local commercial distributor.

INTRODUCTION

The NERC-LV has been investigating plutonium in the environs of NTS since 1970. Sampling and analysis of soil and air, as well as vegetable material from related surveillance studies noted at the 1974

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*See footnote, p. 35.

Plutonium Information Meeting continued through Fiscal Year 1975. The additional efforts and results through April of Fiscal Year 1975 are summarized in this report.

PLUTONIUM IN SOIL

Analyses were completed on 230 samples during the period July 1974-April 1975. Preliminary analysis of the data shows no unusual results. Results from samples analyzed prior to Fiscal Year 1975 ranged from background to 105 nCi/m^2, whereas results from samples analyzed in Fiscal Year 1975 ranged from background to 41 nCi/m^2. The distribution of plutonium in soil is shown in Fig. 1, 2, and 3. Sampling designed to close existing gaps in the offsite sample array is continuing.

PLUTONIUM IN AIR

Analysis of air filters from eight locations in the western United States is continuing. Composites of filters collected monthly at Barstow and quarterly at the other locations from September 1973, to November 1974, and representing approximately 2,500 m^3 of air, are presently undergoing analysis. The results will be reported when available. To determine ambient levels of plutonium in air around the NTS with emphasis on selected areas containing fallout from nuclear tests, plutonium analyses will be routinely performed on composites of filters, representing approximately 10,000 m^3 of air, collected from Diablo, Lathrop Wells, and Las Vegas, Nevada, beginning with filters collected in January, 1975. This study is proposed to begin by the end of the present calendar year.

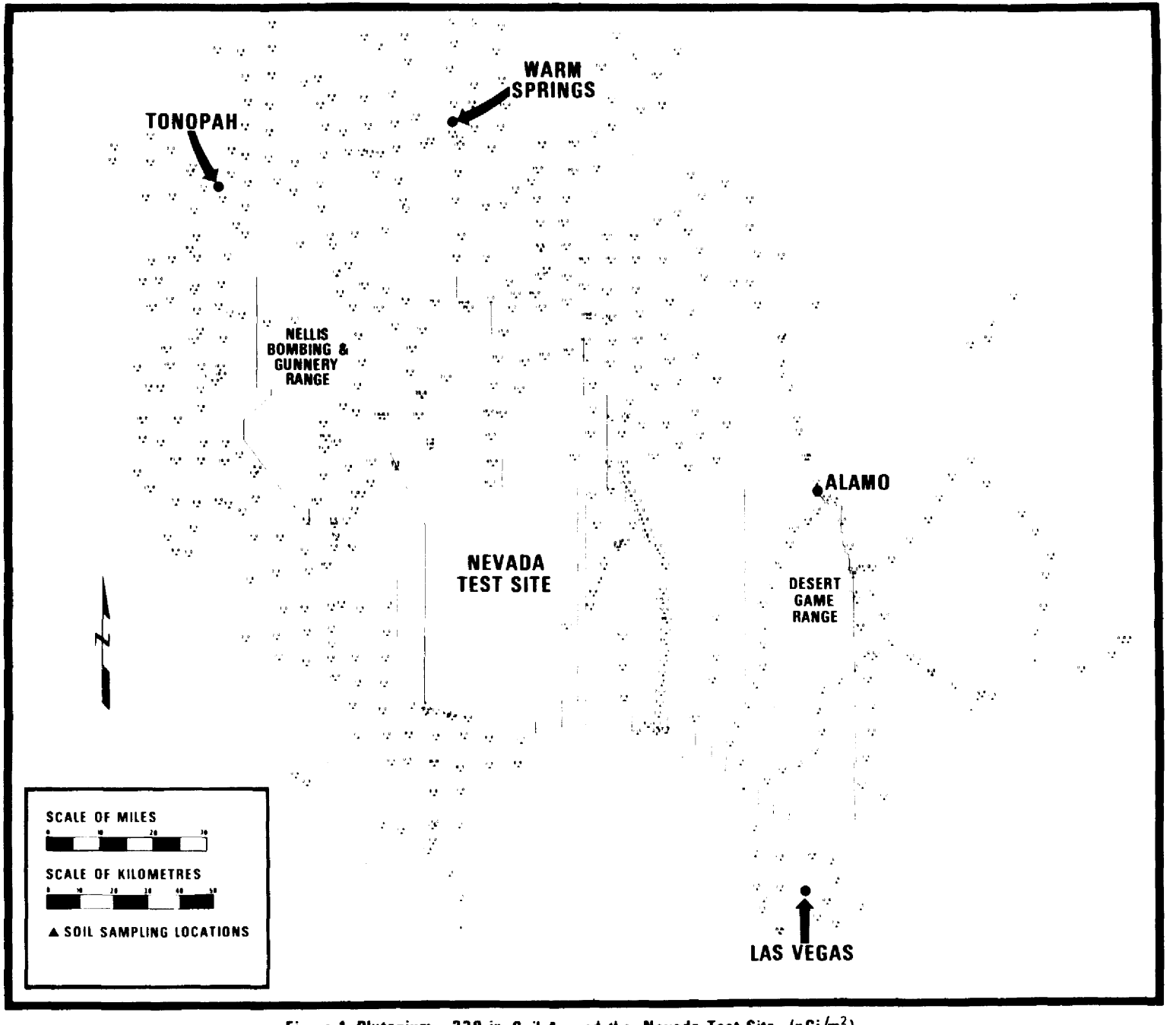

Figure 1. Plutonium - 239 in Soil Around the Nevada Test Site (nCi/m^2)

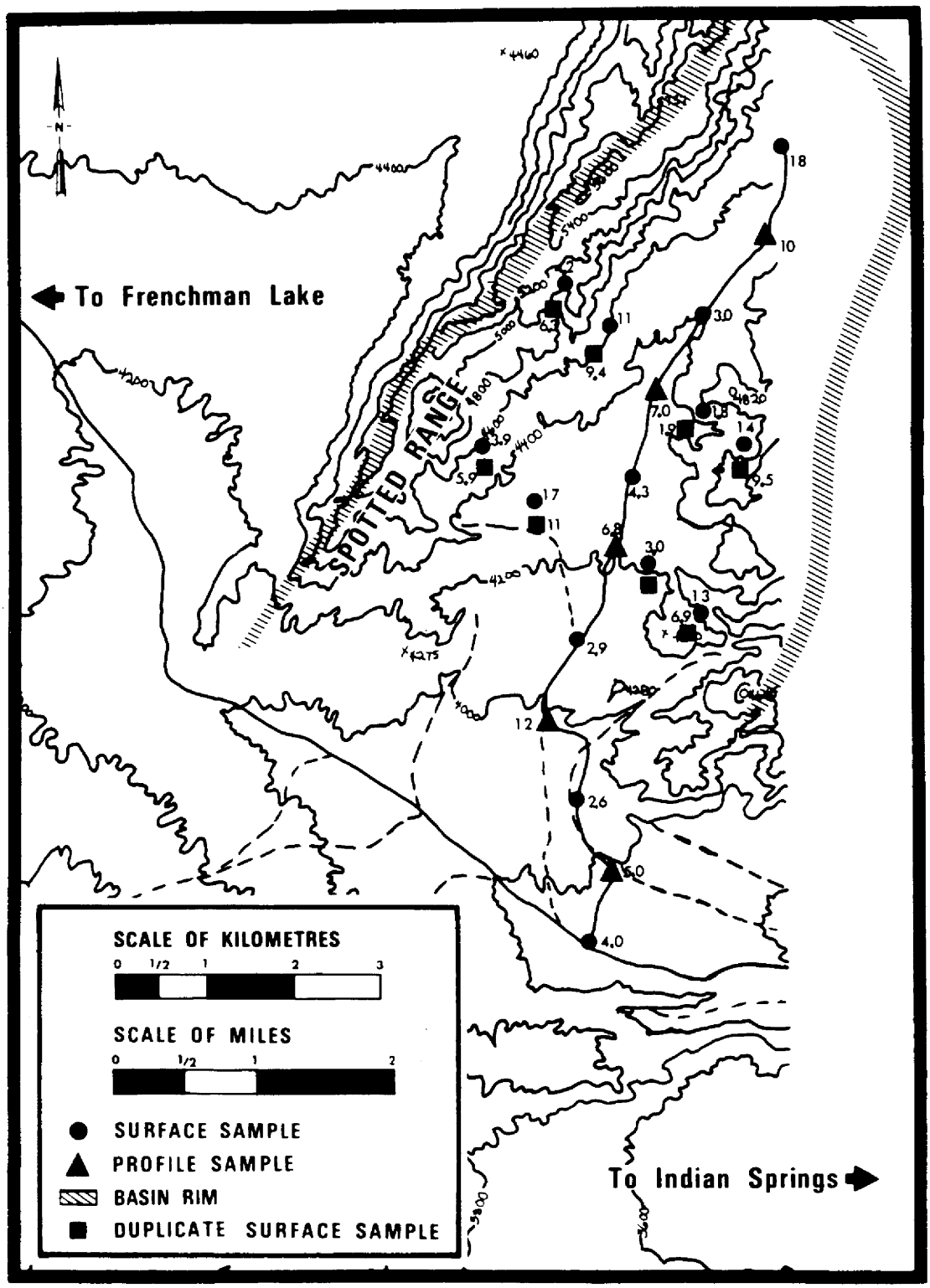

Figure 2. Plutonium–239 in Soil (nCi/m^2) – Basin Study Sampling

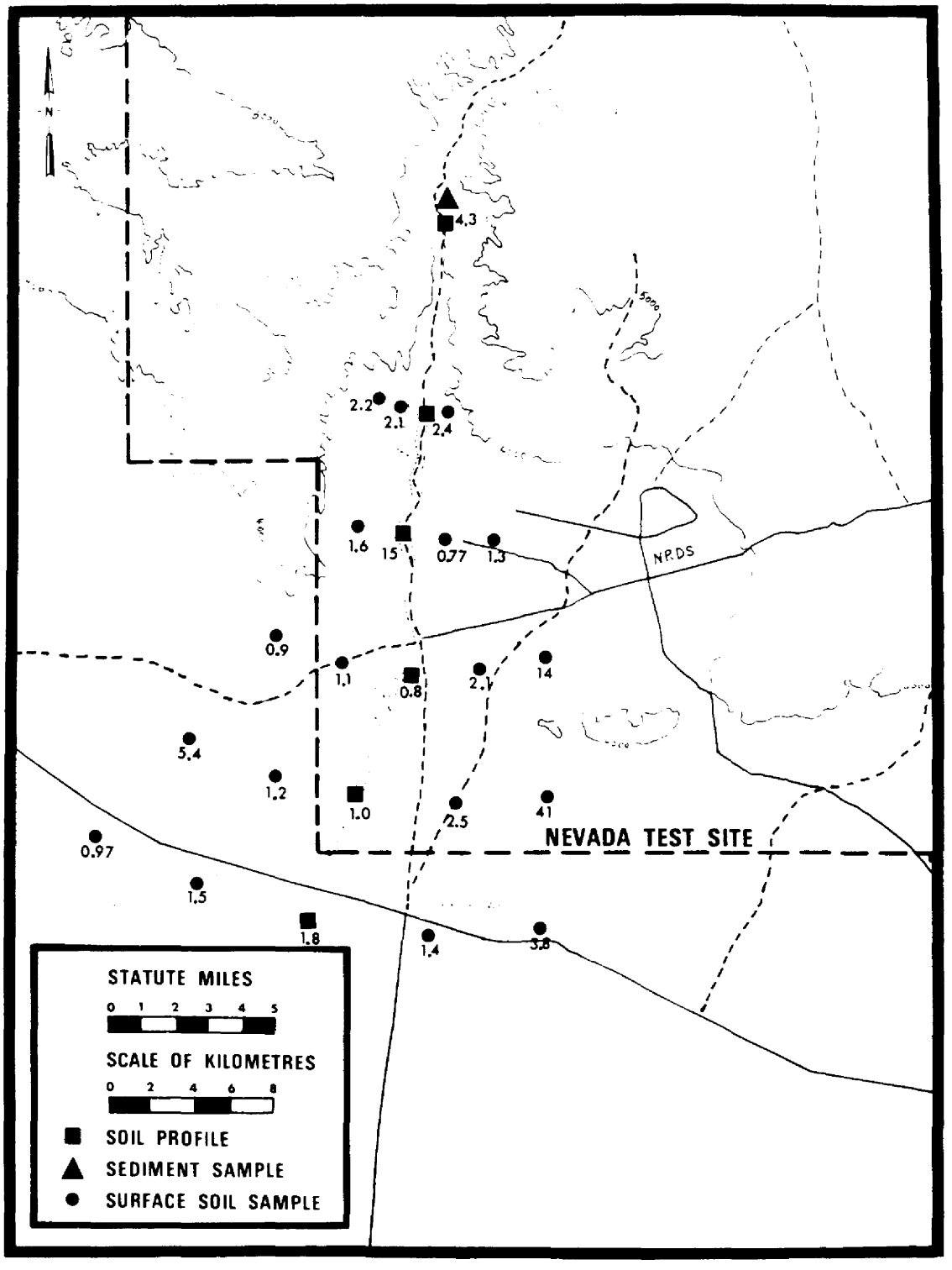

Figure 3. Plutonium - 239 in Soil Around the Nevada Test Site (nCi/m^2) Fortymile Canyon

RELATED PROJECTS

Basin Studies

Soil from a drainage basin located approximately 11 km southeast of Frenchman Flat, NTS, an area of known plutonium contamination, was sampled as a preliminary phase of a project to investigate movement of plutonium with surface water drainage. The sampling pattern and results are shown in Fig. 2. These results do not show movement or concentration of plutonium within the basin.

Fortymile Canyon Study

The Fortymile Canyon Study was initiated to investigate the possibility that radioactivity deposited on the NTS might be carried to the off-site area by surface runoff. Two water and sediment collectors have been emplaced in the wash of Fortymile Canyon, which drains a large portion of the western NTS. Due to the lack of sufficient rainfall, no water or sediment samples have been collected. Soil sampling was completed in October, 1974, and sample analyses were completed in March, 1975. Results from these samples show no definitive patterns of radionuclide movement or concentration as a result of hydraulic transport. All samples were analyzed by NaI(Tl) gamma counting and selected samples were analyzed by Ge(Li) counting. Neither method detected gamma emitters other than background levels of ^{137}Cs and ^{40}K. The sampling array and plutonium results are shown in Fig. 3. The sediment collectors will remain in place until samples can be collected and analyzed.

Vegetable and Fruit Sampling Study

Concurrently with the above studies, the radionuclide content of fruits and vegetables grown in home orchards and gardens near the NTS was investigated. No detectable quantities of plutonium were found in vegetable material collected near the NTS; however, ^{239}Pu concentrations possibly as great as 24 pCi/kg wet weight were detected in some samples

prepared for human consumption and purchased locally from a commercial distributor. These results are being investigated further. Table 1 shows the results of those samples purchased commercially. Details of this survey will be covered in a separate report.

FUTURE ACTIVITY PLANS

Two high-volume air sampling systems have been obtained and will be placed in the field northeast of the NTS by the end of the present calendar year. Wind speed and direction will be recorded using National Oceanic and Atmospheric Administration--Air Resources Laboratory instrumentation.

Several drainage basins approximately 160 km northeast of the NTS are presently being considered as a second sampling area to investigate concentrating effects of radionuclides in soil. A basin with high sheer walls which act as impingement collectors is being sought.

Soil sampling and analysis will continue in order to close existing gaps in the data array. Collecting these samples will require more time than previous samplings due to the remoteness of the sampling area and the difficulties caused by terrain.

During the summer of 1975, a study will be executed in the off-NTS area to evaluate the hypothesis that radionuclides accumulate near vegetation where erosion products are deposited.

Table 1. Plutonium-239 in Vegetable and Fruit Samples

Sample Type	Pu-239 (pCi/kg fresh wt.)*	Remarks
Peaches	2.6 ± 0.33**	
	2.5 ± 0.2	Original plating reanalyzed.
Apricots	<0.049	
	<0.10	Second aliquot of ash.
Plums	0.021 ± 0.020	
	<0.073	Original plating reanalyzed.
Carrots	24 ± 2.2	
	5.9 ± 5.2	Second aliquot of ash.
Lettuce	0.071 ± 0.027	
	<0.10	Original plating reanalyzed.
Cabbage	0.026 ± 0.023	
	<0.19	Second aliquot of ash.
Corn	<0.016	
	<0.079	Second aliquot of ash.
Turnips	<0.02	
	<0.049	Second aliquot of ash.
Turnips	8.3 ± 0.65	
	7.6 ± 0.86	Second count of above.
	8.5 ± 0.76	Second aliquot of ash.
Turnip Greens	0.14 ± 0.071	

*Preliminary results under review.

**2-sigma counting error.

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Resuspension

RESUSPENSION ELEMENT STATUS REPORT, MAY, 1975*
NEVADA APPLIED ECOLOGY GROUP

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ABSTRACT

The resuspension-inhalation pathway is generally regarded as the critical pathway for a few radionuclides of very low biological availability, such as plutonium. The primary goal of the resuspension element program is to develop a general model for the prediction of the average concentration of resuspended contaminants. This report briefly summarizes past accomplishments, current studies, and future plans of the resuspension project being conducted at the Nevada Test Site. Past accomplishments have been: measurement of the concentration of resuspended ^{239}Pu over short time periods of consistent meteorology with concomitant detailed micrometeorological measurements; development of two interim predictive models and derivation of protective guidelines for soil contamination with ^{239}Pu; derivation of parameterizations of resuspended soil concentrations and fluxes as functions of friction velocity and soil erodibility; test of applicability of these parameterizations to the resuspension of ^{239}Pu; and development of an integrated approach to a general resuspension model. Current studies include: test of the resuspended soil concentration and flux parameterizations at a variety of Test Site locations; development of an airborne platform for the study of mass fluxes produced by episodic events such as dust devils; development of a computer-controlled sampling system for testing model validity; addditional data analysis of collected data; and development of

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*A preprint of this report appeared as UCRL-76823.

improved detectors and methods for *in situ* quantitation of ^{239}Pu and ^{241}Am in soils. Future plans are: a major experimental program in FY 1976, at the GMX location in Area 5 under a wider variety of meteorological conditions; participation in proposed trials of clean-up procedures; and further efforts to develop a general resuspension model.

INTRODUCTION

Long-term health hazards associated with the release and terrestrial deposition of pollutant material of low food-chain transfer are partially attributable to the respiration of particles resuspended in air due to atmospheric forces, or by disturbances resulting from human activity. This pathway is critically important in the evaluation of health hazards associated with processing of uranium ore, liquid-metal fast breeder reactor technology, accidents involving nuclear weapons, reoccupation of contaminated land areas such as Enewetak and Bikini Atolls, and the environmental impact of current and future uses of the U.S. Energy Research and Development Administration's (ERDA) Nevada Test Site (NTS).

The primary goal of the resuspension studies conducted in collaboration with the Nevada Applied Ecology Group (NAEG) is to develop a general model for prediction of average concentration of pollutant particles resuspended as a function of surface characteristics and meteorological parameters following their initial ground deposition. It is anticipated that independent variables in the model necessarily will include the areal size and magnitude of deposition, particle-size distribution of the contaminant, particle-size distribution of the host surface, soil-surface characteristics and vegetation cover, time since deposition, appropriate climatological descriptions of the area, and meteorological parameters that affect dispersion and redeposition.

A specific, secondary goal of the project is to provide input to the NAEG, Nevada Operations Office, regarding hazard assessments of

radionuclides within the NTS, and to assist in providing guidance for the possible cleanup of plutonium-contaminated areas.

Progress in this project has resulted primarily from experiments carried out at the intensive study location at the GMX site in Area 5. In addition, less elaborate measurements have been conducted at several additional areas of plutonium contamination. Off-site experiments concerning parameterization of dust fluxes and mass loadings have been conducted near Plains, Texas. Interim models developed from these studies have been utilized in radiological assessment of the reoccupation of Enewetak Atoll and in the environmental assessment of liquid-metal fast breeder reactor technology.

PAST ACCOMPLISHMENTS

Nine reports (see References 1 through 9, this report) have been published or submitted for publication since July 1, 1974. These publications cover in detail the experimental measurement programs, results of measurements, and the data analysis including model development which has resulted. Additional data have also been collected which are not covered by these publications and include measurements of the concentration of the plutonium in air at other NTS locations in addition to the GMX.

Some highlights of these past accomplishements are listed below:

1. Concentration of resuspended plutonium in air has been measured over short time periods of consistent meteorology with the use of ultrahigh-volume air samplers (1,500 m^3/hr). Simultaneous measurements of wind speed and temperature profiles, in addition to other meteorological variables, have provided the first well-documented set of resuspension measurements made over short time periods. Important meteorological parameters, friction velocity (u*), surface roughness length (z_o), and Richardson number (Ri)

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*The resuspension factor is the concentration in air divided by the surface deposition and has units of m^{-1}.

are derived from the data. These measurements are the cornerstone of the resuspension program. Measurements have also been made of particle-size distributions of airborne mass and ^{239}Pu.

2. A time-dependent resuspension factor* model was developed as an interim, conservative model for prediction of the resuspended-plutonium concentrations. The only independent parameter in this model is time, so it was considered appropriate that the model be consistent with higher reported measurements of the resuspension factor. This model differs from other such models in that time-dependency is more complex and consistent with actual measurements over aged plutonium sources. This model was used to develop a protective guideline for soil contamination immediately after deposition; the result is 1 $\mu Ci/m^2$ for a 50-yr dose commitment of 75 rem.

3. A mass-loading resuspension model was developed which is appropriate for reoccupation of contaminated land several years after deposition, and at which time the contaminant may be assumed to be intimately associated with the host soil. Using the model, predicted results were found to be in good agreement with experimental data. A protective guideline was also developed for this situation; the result is 7 nCi/g in the top 1 cm of soil for a 50-yr dose commitment of 75 rem.

4. The eddy correlation method was used for the first time to derive a quantitation of dust flux. This study also resulted in a more generally useful method of predicting resuspended soil concentrations and fluxes when it was ascertained that both are expressible as a power function of friction velocity. The power, however, varies from location to location and was tentatively parameterized as a function of the Chepil Soil Erodibility Index (Chepil and Woodruff, 1959).

5. The above results were tested for applicability to ^{239}Pu resuspension at the GMX location. Generally good agreement was found between predicted and measured concentrations at sampling locations close to the area of maximum ground deposition. At farther locations, the complicating effect of a nonhomogeneous source

made comparison more ambiguous and demonstrated the need for a source-specific model of atmospheric transport and diffusion. Such a model had been derived for the GMX location by J. Healy (1974) of LASL, and through its use, it was possible to derive resuspension rates (the fraction of the areal source that is resuspended per unit time) for the GMX source. These results were also consistent with those predicted by the mass flux model.

6. The mass flux model and Healy's resuspension rate model were combined to derive an integrated approach to development of a general predictive model of resuspension. An interesting result is the derivation of an expression which relates resuspension rate to the older, frequently used parameter resuspension factor. This relationship is general for locations close to the maximum ground deposition and involves three micrometeorological parameters.

STUDIES IN PROGRESS

1. A mobile field laboratory has been established for measurement of suspended dust concentrations and micrometeorological parameters at various locations at the NTS. This equipment is being used to test previously derived parameterizations which relate concentration of suspended dust to friction velocity and soil erodibility. These results, if verified, will also have general applicability to prediction of the resuspension of aged radionuclide deposits. Experiments with this system have been conducted at three sites.

2. An isokinetic air sampler and appropriate meteorological sensors are being installed on a small aircraft under the direction of P. Sinclair of Colorado State University. This system will be used to measure mass loadings in dust devils at the NTS. This data, in combination with data on the occurrence frequency of dust devils, will be used to assess the significance of such episodic events upon overall resuspension process.

3. A computer-controlled sampling system is undergoing final development at Livermore. This system will monitor and record a variety of meteorological parameters and can turn samplers on and off according to preset constraints in terms of meteorological conditions. This system will be used for long-term studies designed to verify predictive models. Field operation of this system should begin early in FY 1976.

4. Additional data analysis is being performed on measurements made at the GMX site. This includes our own measurements for which radionuclide analyses only recently became available and data collected by Reynolds Electrical & Engineering Company, Inc. (REECo), at this site before our own measurement program started.

5. Analysis of data collected at other NTS locations is also under way. Data will be reported for the concentration of resuspended mass and ^{239}Pu, and for their distributions with particle size.

6. The resuspension element has been actively supporting development of a more sensitive *in situ* detector for ^{239}Pu and/or ^{241}Am. In the past, we have reported a general calibration method (Anspaugh *et al.*, 1972) for the FIDLER and have also reported on the use of a large Ge(Li) *in situ* detector which is an order of magnitude more sensitive than the FIDLER. A prototype of a more desirable Ge(Li) detector configuration has been fabricated and is undergoing laboratory testing. If these are successful, field trials in the GMX location will be conducted in early FY 1976.

FUTURE STUDIES

A major experimental program at the GMX site is scheduled to begin in July, 1975. The airborne sampling system will be used to measure mass concentrations in dust devils in order to estimate the mass flux per dust devil. The frequency of occurrence of dust devils will also be recorded by ground observers. At the same time, surface air sampling for ^{239}Pu determination will be conducted with emphasis on studying resuspension during a range of meteorological conditions

that have not yet been experienced during previous measurement periods. Simultaneous measurements of mass loading will be made with the use of fast-response instrumentation.

It is also planned to participate in proposed trials of clean-up procedures for contaminated soils. Such studies will provide a valuable opportunity to document the influence of artificial disturbances on the resuspension of plutonium. Such data are needed for input to cost-benefit analyses of plutonium cleanup, particularly for aged sources.

Additional experimental work will also continue in order to further define and verify the tentative (but critically important) parameterization of the mass flux of suspended soil as a power function of friction velocity with power depending upon the Soil Erodibility Index.

More effort will be devoted to developing and publishing a useful, general model of the resuspension process. Part of this effort will be to adapt a model of atmospheric transport and diffusion so that it will be particularly useful in predicting the concentration of resuspended contaminants downwind from a localized source of any geometrical configuration. A critical lack in the development of a general model is an adequate description of the weathering process which involves the attachment of deposited contaminant to host soil and its downward migration through the soil. Future efforts will be devoted to modeling this process from a fundamental basis, and additional measurement programs at appropriate sites will also be designed to examine this process.

ACKNOWLEDGMENTS

The resuspension work at the Nevada Test Site is a joint effort with Lawrence Livermore Laboratory funding provided by the Division of Biomedical and Environmental Research, U.S. Energy Research and Development Administration. The Nevada Operations Office provides

additional funding for the participation of the Air Resources Laboratory-Las Vegas, National Oceanic and Atmospheric Administration; the Nevada Applied Ecology Group, Nevada Operations Office, provides funding for support by Reynolds Electrical and Engineering Co., Inc., and for radiochemical analyses. All meteorological measurements are the responsibility of Mr. N. C. Kennedy of the Air Resources Laboratory.

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Statistics

SOME STATISTICAL DESIGN AND ANALYSIS ASPECTS FOR NEVADA APPLIED ECOLOGY GROUP STUDIES

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ABSTRACT

Some of the design and analysis aspects of the Nevada Applied Ecology Group (NAEG) studies at safety-shot sites are reviewed in conjunction with discussions of possible new approaches. The use of double sampling to estimate inventories is suggested as a means of obtaining data for estimating the geographical distribution of plutonium using computer contouring programs. The lack of estimates of error for plutonium contours is noted and a regression approach discussed for obtaining such estimates. The kinds of new data that are now available for analysis from A site of Area 11 and the four Tonopah Test Range (TTR) sites are outlined, and the need for a closer look at methods for analyzing ratio-type data is pointed out. The necessity for thorough planning of environmental sampling programs is emphasized in order to obtain the maximum amount of information for fixed cost. Some general planning aspects of new studies at nuclear sites and experimental clean-up plots are discussed, as is the planning of interlaboratory comparisons.

INTRODUCTION

The NAEG has been active during the past few years with the collection and analysis of more than 2,000 soil, vegetation, small mammal, cattle, and air samples at 10 safety-shot sites on the Nevada Test Site (NTS) and the Tonopah Test Range (TTR). The authors have been

actively involved in the statistical design and analysis aspects of these studies and takes the opportunity here to review some of these methods and to suggest some new approaches that might be useful in future work. This review may be particularly helpful at this time since new studies at nuclear test sites at NTS are expected to begin soon, as are experimental plots to evaluate the feasibility of several alternative clean-up treatments in desert environments. Some of the general planning aspects of these new studies are also discussed.

SAMPLING FOR INVENTORY AND GEOGRAPHICAL DISTRIBUTION OF Pu

Review and Possible New Approaches

The FIDLER (Field Instrument for the Determination of Low-Energy Radiation) has been successfully used as a survey instrument at most safety-shot sites to delineate the approximate distribution of ^{241}Am in surface soil. The only exception is A site in Area 11, where the FIDLER could not be used since the principal contaminant is ^{235}U. (At this site, soil samples were collected on a grid system and analyzed for ^{235}U on the Ge(Li) system.) FIDLER survey readings give a general notion of the geographical distribution of ^{241}Am contamination in surface soil and are useful for defining strata for estimating plutonium inventory in soil. Strata maps drawn from these FIDLER survey data are given in Figs. 4 through 14 in Gilbert *et al.* (1975). While a major advantage of the FIDLER instrument is the relative ease and speed at which a study site can be surveyed, it has been our experience that two surveys of an area are usually required to obtain the necessary detail in distribution desired for establishing strata: an initial survey to learn the general pattern of dispersal, followed by a second survey on a finer grid to obtain greater detail. The only comment here is that greater use of any prior survey information that may exist on the dispersal of contaminants could reduce the time necessary to obtain the more detailed survey information needed for current studies.

A difficulty with the strata maps obtained using the FIDLER is that the isopleth lines defining the strata are in units of counts per minute (1,000 cpm, 5,000 cpm, etc.) which cannot be related directly to nCi/gm concentrations of $^{239-240}Pu$ or ^{241}Am in surface soil. One approach to this problem would be to choose a subset of the survey-sample grid points covering a wide range of expected concentrations, and for these locations, to collect a soil sample immediately after the FIDLER survey reading was taken. These samples would then be counted for ^{241}Am on the Ge(Li) system, and, hopefully, a sufficiently accurate regression relationship between the FIDLER readings and the Ge(Li) scans would result to enable one to associate concentrations of ^{241}Am with the FIDLER isopleth lines. This is still one step from the goal of relating FIDLER isopleth lines to $^{239-240}Pu$ concentrations in soil, but Figs. B1 through B20 in Gilbert *et al.* (1975), indicate a good linear relationship exists between ^{241}Am and $^{239-240}Pu$ in surface soil at the safety-shot sites, especially for higher concentration levels. Hence, it may be possible to obtain at least a rough idea as to plutonium soil concentrations along FIDLER isopleth lines. However, there is considerable doubt at this time that this approach would result in <u>accurate</u> estimates, since data collected to date at random locations indicate the correlation between FIDLER readings and $^{239-240}Pu$ concentrations are often low, especially for low concentration areas (see Tables 18, 19, and 20, and Figs. 33-36 in Gilbert *et al.*, 1975). These data need to be examined more closely to determine the feasibility of the approach outlined above. An advantage of this regression approach is that it would allow for placing limits of error on ^{241}Am concentrations associated with FIDLER contour lines.

The method used thus far to estimate concentration contours of plutonium in soil is to use the $^{239-240}Pu$ concentration data for soil samples collected at random locations within strata (the samples collected to estimate Pu inventory in surface soil) in a computer contouring program called SURFACE II Graphic Systems (Sampson, 1973). This yields computer-drawn contour maps and 3-dimensional representations of the present geographical distribution of plutonium (Figs. 16-27, Gilbert *et al.*, 1975) using a nearest-neighbor estimation scheme. Unfortunately, the program does not allow for estimating the precision of its estimated contour lines. Whether or not this is possible using the nearest-neighbor approach remains to be determined.

It was pointed out in Gilbert *et al.* (1975, Fig. 28) that the contour lines obtained to date for Area 13 and GMX are biased, due in part, it is believed, to the nearest-neighbor estimatiom method and to the use of soil samples collected at random locations within strata (stratified random sampling) resulting in inadequate coverage of portions of the field. One approach that may reduce the bias in these contours is to fit contours to the log-transformed data. This might be effective since the large concentration peaks would be scaled down, it is presumed, for making more accurate fits to the less variable data. This bias might also be reduced by the use of more strata or the division of strata into substrata within which samples would still be taken at random. This approach would tend to reduce the size of areas where no samples were collected, and hence reduce the bias due to such gaps in information.

Another approach would be to increase the number of locations at which samples are taken. This could be done as part of a "double sampling" scheme for estimating plutonium inventory in soil so that costs would not be expected to be greater than for presently used sampling designs. The general idea, as it might be applied to a safety-shot site, would be to define strata using the FIDLER and to collect a relatively large number of soil samples within strata. All samples would be analyzed for ^{241}Am using the Ge(Li) system and a small proportion would also be analyzed for $^{239-240}Pu$ using wet chemistry techniques (both Pu and Am analyses being done on the same aliquot from the soil sample). This kind of sampling procedure is known as double sampling. Gilbert and Eberhardt (1975) evaluated its effectiveness for Area 13 and GMX and concluded that except for the two lowest plutonium concentration strata, the technique can yield estimates of plutonium inventory with greater precision than possible with plutonium analyses alone for a fixed cost. This result has implications relative to the estimation of unbiased concentration contours since Gilbert and Eberhardt (1975) estimated that about twice as many soil samples would need to be collected using double sampling than were used to estimate Pu inventory using stratified random sampling. All of these soil samples would be Ge(Li) scanned for ^{241}Am, but less than 20% would also need to be analyzed for $^{239-240}Pu$ (keeping total cost equal to that in estimating inventory using

stratified random sampling). The important point here is that if inventory were estimated using this kind of double sampling scheme, a rather large number of soil samples analyzed for ^{241}Am (about 340 in Area 13 and 220 in GMX) would be available for estimating ^{241}Am contours using the computer contouring program. Since there is a strong linear relationship between ^{241}Am and $^{239-240}Pu$ at the safety-shot sites, this relationship could be used to transform the ^{241}Am contours into $^{239-240}Pu$ contours within some limit of error (which could be estimated). This approach in combination with the above-mentioned techniques (using log-transformed data and more strata) has been discussed here as they might be applied to the safety-shot sites. Their applicability to nuclear sites needs to be carefully considered before sampling plans are finalized for these areas.

Doubling sampling using Ge(Li) scans for ^{241}Am has been suggested above as a method whereby more precise estimates of inventory may be obtained at safety-shot sites with no increase in costs over presently used methods. Gilbert and Eberhardt (1975) also found for Area 13 and GMX that costs could be reduced on the order of 20 to 30% using double sampling, if the precision of present estimates of inventory (Tables 4, 5, and 6 in Gilbert *et al.*, 1975) are adequate. While Gilbert and Eberhardt did not look in detail at the remaining safety-shot sites (Area 11 and the four TTR sites), it appears that double sampling using Ge(Li) scans could be applied there also. The same general conclusions were found by Gilbert and Eberhardt to apply to a double sampling scheme wherein field FIDLER cpm readings take the place of Ge(Li) scans for ^{241}Am in the above description of the method. Again, the method is not feasible for low-concentration strata (Stratum 1 and 2 in Area 13 and GMX). For these low-level areas, it appears there is no substitute for wet chemistry Pu analyses.

An analysis approach not tried as yet by the authors is to examine the regression relationship between distance from ground zero (GZ) and $^{239-240}Pu$ concentrations in soil and vegetation. Since concentrations decrease quite rapidly with distance from GZ, this suggests one might look at the feasibility of estimating $^{239-240}Pu$ concentrations in soil and vegetation using this variable in conjunction with Ge(Li) scans for ^{241}Am. The rate at which plutonium concentrations drop off also depends on compass direction, suggesting this

variable might also be included. If these distances and direction variables improved significantly the ability to predict plutonium concentrations in soil, then these variables might be incorporated into a double sampling plan to obtain yet more accurate and precise estimates of plutonium inventory. As indicated above, this has yet to be examined for its applicability.

Gilbert *et al.* (1975) discuss the information available to date on plutonium amounts below the surface 5 cm of soil and within blow-sand mounds on the safety-shot sites. A closer look at the profile samples is needed and a comprehensive blow-sand mound sampling program is being planned. Considerable thought by several NAEG scientists has been given to how one might sample to estimate both the distribution and total amount of plutonium within mounds. Recommendations have been made but the problem is complex, and progress may be largely a trial and error approach.

Concerning profile samples, an examination of present data strongly suggests that more detailed field notes should be taken at the time of sampling regarding the nature of the soil at the sample location (evidence of physical disturbance, erosion by wind or water, animal signs and burrows, etc.), both on the surface and at each depth in the profile. Information on the soil type (according, perhaps, to the classifications given by Leavitt (1974)) at each depth should also be recorded since soil type is a variable that may have an effect on the relative amounts of Pu in surface soil. This information should be recorded at each soil sample location.

Consideration might also be given toward devising a field experiment to estimate the amount of cross contamination between different levels of the profile due to the sampling method itself. It is necessary to know how much of the plutonium being seen at depths is due to environmental weathering forces and how much to the sampling method. Such an experiment may be difficult to devise, but could be useful in the interpretation of these profile data.

Anticipated Analyses of New Data

Since January 1, 1975, additional data from the safety-shot sites have been reported by the analytical laboratories. This includes analyses for $^{239\text{-}240}Pu$, ^{241}Am, ^{234}U, ^{235}U, ^{236}U, and ^{238}U for soil and/or vegetation samples taken from A site of Area 11. While incomplete, these data will be used to obtain preliminary estimates of inventory for $^{239\text{-}240}Pu$, ^{235}U, and ^{238}U in the surface soil of A site. Results for four profile samples have also been reported for this site. Additional soil and vegetation analyses for ^{238}Pu, $^{239\text{-}240}Pu$, and ^{241}Am have also been reported for the TTR sites. Whenever possible, these will be used to update the estimates of inventory reported for these sites in Gilbert *et al.* (1975). Attempts to estimate $^{239\text{-}240}Pu$ concentration contours using the SURFACE II computer program will also be made using the new TTR data in combination with that previously available. Hopefully, less biased estimates can be obtained than reported by Gilbert *et al.* (1975) for Area 13 and GMX. Also, the data from these latter two sites will be reexamined with the same goal in mind.

ESTIMATION OF RATIOS

The methods used up to the present time to estimate (1) $^{239\text{-}240}Pu$ to ^{241}Am ratios in soil and vegetation and (2) vegetation to soil ratios of $^{239\text{-}240}Pu$ concentrations for paired vegetation-soil samples are given in Gilbert *et al.* (1975) and Romney *et al.* (1975), along with the estimated ratios themselves. Gilbert *et al.* concluded that the estimated ratio

$$\hat{\beta}_1 = \sum_{i=1}^{n} Y_i \Big/ \sum_{i=1}^{n} X_i$$

was preferable to the average of the ratios

$$\hat{\beta}_2 = \frac{1}{n} \sum_{i=1}^{n} Y_i/X_i$$

on the grounds of a better linear regression fit through the origin for the latter. It is felt, however, that the general topic of ratio estimation for plutonium and other transuranics needs further consideration, particularly in view of the skewed nature of these environmental data. Some thought needs to be directed toward optimum methods of estimating ratios when both numerator and denominator are, say, lognormally distributed. For example, if both are lognormal, then the individual ratios Y_i/X_i are also lognormally distributed, which suggests a method efficient for estimating lognormal means might be considered (Aitchison and Brown, 1969, p. 44). A related problem is the bias that is known to be present to some extent in $\hat{\beta}_1$ and $\hat{\beta}_2$ when the ^{241}Am data are subject to error (Snedecor and Cochran, 1967, p. 164).

The new data from A site, Area 11, will permit the estimation of ^{235}U to ^{238}U, and $^{239\text{-}240}$Pu to ^{241}Am ratios in soil and vegetation. The ^{235}U to ^{238}U ratio will decrease with increasing distance from GZ since the device was made up predominately of ^{235}U. The ^{238}Pu to $^{239\text{-}240}$Pu ratios can also be computed for the soil samples analyzed by Los Alamos Scientific Laboratory (LASL). Some additional plutonium and americium data from TTR and Area 11 sites will also permit the update of estimated ratios given in Gilbert, *et al.* (1975).

Some new analyses of the ratio data may also be useful. For example, there has been no attempt to explain the variability observed in vegetation to soil plutonium ratios. It would prove informative to investigate whether high Pu concentrations in vegetation tend to be associated with low Pu concentrations in soil. This is suggested by the Stratum 3 Double Track vegetation and soil data (Gilbert *et al.*, 1975, Fig. 32). If so, this could be a reflection of the particulate nature of Pu in the soil samples versus the less variable (more homogenous) vegetation solutions from which aliquots for analysis are drawn.

SAMPLING AT NUCLEAR TEST SITES

The initiation of environmental sampling porgrams at nuclear event sites on the NTS must begin, as always, with precisely written statements of the objectives for sampling. It might be helpful to have written copies of these objectives sent to all participating scientists so that decisions made during the course of these studies will not be in conflict with established objectives. Some guidance relative to the level of precision desired in estimated quantities should be given to permit the selection of adequate numbers of samples and appropriate survey and laboratory measurement techniques.

Stratified random sampling is expected to be useful at the nuclear sites in estimating total amounts of contaminates present, although a field survey instrument other than the FIDLER will, it is presumed, be required to define strata. The usefulness of (1) double sampling to estimate total amounts at these sites and (2) whether any of the modifications in sampling plans suggested above for the safety-shot sites might be useful in these new areas, needs to be evaluated. A thorough study of maps, surveys, and other data available on these nuclear sites well in advance of actual field sampling should result in better sampling plans in terms of obtaining more information for a fixed cost. Consultations between NAEG personnel and those familiar with field activities at the test sites since the time of detonation is strongly encouraged. It is also necessary to become familiar with the kinds of survey and laboratory analytical equipment appropriate for these sites.

Consideration should be given to the adequacy of the present NAEG data bank system relative to the needs associated with sampling at nuclear sites. Since modifications may be required, someone familiar with the system should be involved in planning sessions where sampling designs are discussed. Consideration should be given toward modifications to the data bank that will permit greater interaction between the scientist and the data.

The design of small mammal studies should receive careful considerations at the nuclear sites. The designs presently in use at the safety-shot areas yield data that are difficult to relate to soil and vegetation concentrations. These difficulties arise primarily from the mobile nature of the animals and the lack of detailed knowledge relative to the levels of contamination to which individuals are exposed. Explaining the variability observed among animals captured at the same trap location is, therefore, a difficult task.

The procedures used to prepare soil before being sent out to the laboratories for analysis need careful review before sampling begins at the nuclear sites. The practice of sending ball-milled samples to some laboratories and sieved (after ball-milling) samples to other laboratories (the case for TTR and Area 11 soil samples) has created problems of the comparability of soil results between laboratories. Hence, this practice should not be continued for the nuclear sites. All aliquots of soil leaving the soils preparatory lab should be prepared in the same way, regardless of which analytical laboratory is to receive the aliquot for analysis. Furthermore, the recommended procedure is to ball-mill but not to sieve the soil samples. This recommendation is based on data from the safety-shot sites that suggest concentrations in sieved soil aliquots are greater than those in unsieved (but still ball-milled) aliquots. This effect would presumably also be present for soils at nuclear sites.

EXPERIMENTAL CLEAN-UP PLOTS

Plans are being made to establish small experimental clean-up plots on nuclear and/or safety-shot sites for purposes of choosing among several clean-up procedures on the basis of cost, effectiveness, and posttreatment stabilization of the areas. The design and layout of these plots will require particular attention to the specification of detailed objectives, variables to be measured, kinds of field and laboratory instrumentation to be used, control (if possible) of cross contamination of the treatment plot from the contaminated sites, and the selection of the sampling and experimental design. The placement

of plots relative to one another should take into account any environmental factors such as prevailing wind directions and field drainage patterns that would tend to confound (mix up) the clean-up treatment effects. The use of two or more control plots in appropriate locations is advisable. Also, an outline of the pertinent summary tables and probable results expected from the study would be useful at the planning stage, as would an outline of the statistical analyses to be performed. Careful attention to detail at the planning stage should yield benefits over the long run.

One feature of the study will be, apparently, to measure the degree of revegetation that occurs following the various clean-up treatments. The particular variables used to measure this effect must be selected with care to ensure that measurements obtained over time are truly comparable. Also, the variability in revegetation behavior from plot to plot that will be induced by the probable inability to apply a clean-up treatment with equal force to different plots suggests the need to have replicate plots for each treatment. Specifications of the optimum number of replicate plots per treatment will be difficult to ascertain due to lack of prior information. Slight differences in revegetation due to treatment effects are likely to go undetected.

INTERLABORATORY COMPARISONS

There is a need to continue the comparison of laboratories regarding their analytical results for plutonium or other radioisotopes. But there also needs to be a greater awareness of the limitations of such studies, relative to being able to detect differences between laboratories, when environmental samples are used.

There are basically two types of samples that can be used for interlaboratory comparisons: "cross-check" and "standard" samples. Cross-check samples are environmental samples (soil, vegetation, etc.) collected either during the course of an ongoing field study, such as at an NAEG study site, or as special field samples collected with the express purpose of comparing laboratories' analyses. Standard

samples are specially prepared in the laboratory so that all aliquots from a given sample are known to have the same radioisotope concentration.

The purpose of both kinds of interlaboratory comparison samples is basically the same; namely, to see if all participating laboratories obtain analytical results that are "alike" to some acceptable degree for a given sample. If there are large, consistent differences (biases) between laboratories, then the use of "standard" samples is, in general, the better of the two methods to detect and estimate this bias. This is primarily due to the homogeneity of the "standard" soil or vegetation relative to the greater variability that can occur between aliquots of the same "cross-check" sample due, perhaps, to the presence of hot plutonium particles or some other unknown environmental factor(s).

The great variability between aliquots of the same environmental sample tends to obscure differences between labs. Hence, when designing an interlaboratory comparison study, two important aspects are (1) the number of samples (n) that need to be aliquoted and sent out to the laboratories and (2) the number of replicate determinations (m) that should be performed at each laboratory on each of the n aliquots. As concerns cross-check (environmental) samples, it appears reasonable to use enough samples (n) in order that a broad range of concentrations and field conditions (e.g., distance from ground zero, desert pavement as well as blow-sand mound samples, etc.) can be represented. This can be achieved by selecting cross-check samples according to some randomization scheme from the total number of field samples collected. The percentage of field samples chosen to be cross-check samples will depend on cost factors and the total number of field samples collected. Concerning "standard" samples, n should be large enough that a broad range of concentrations could be represented in order to detect biases that may be dependent on concentration level (e.g., some labs could be biased on samples with large plutonium concentrations, but not for lower concentration samples).

Gilbert *et al.* (1975) and Eberhardt and Gilbert (1972) discuss the selection of the number m of replicate analyses on cross-check samples that should be used to detect differences between labs. The optimum

value for m, among other things, depends on the within-laboratory variability (variability between replicate analyses) and the size of the actual difference between labs. Gilbert *et al.* (1975) conclude that for soil samples from the safety-shot sites, the use of only three replicate analyses per sample by each laboratory is not likely to detect even large differences between labs. They suggest that the use of less than 10 replicates might be largely a waste of time and money if one wants a fairly good chance of detecting even a ratio of 2 between the high and low lab. Since environmental vegetation samples are presumed to be less affected by the hot particle problem (the vegetation sample is dissolved, Major *et al.*, 1974), the within-lab variability should be less than for soil samples, allowing the use of fewer replicate analyses per sample. Some data is now becoming available for estimating this within-lab variability for vegetation. Hopefully, the hot particle problem in soil will not be as great at the nuclear study sites, allowing for a smaller number m of replicate analyses per lab. Since "standard" samples are prepared so that all aliquots should have the same plutonium concentration, an m of 3 per laboratory for these samples may not be unreasonable, although we have yet to examine the question with actual data.

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STATISTICAL ANALYSIS OF "A SITE" DATA AND INTERLABORATORY COMPARISONS FOR THE NEVADA APPLIED ECOLOGY GROUP

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ABSTRACT

Preliminary estimates of the inventory of ^{235}U and ^{238}U in surface (0-5 cm) soil of A Site in Area 11 of the Nevada Test Site (NTS) are 1,045 $\pm$ 274 ($\pm$ S.E.) and 10,500 $\pm$ 1,150 grams, respectively. The inventory of ^{235}U due to the device alone, i.e., after subtracting off the estimated inventory of natural ^{235}U, is estimated to be 983 $\pm$ 274 grams. The like estimate for ^{238}U is 1,920 $\pm$ 1,150 grams. The estimated inventory of $^{239-240}Pu$ in surface soil for A Site is 0.0347 $\pm$ 0.012 curies. Average concentrations of ^{235}U, ^{238}U, and $^{239-240}Pu$ are estimated for both soil and vegetation. The ratio of these means (vegetation/soil) for ^{235}U decrease from 0.16 to 0.0039 with decreasing distance from ground zero. The decrease is from 0.028 to 0.0066 for ^{238}U. The concentrations of ^{234}U and ^{236}U for vegetation are about 100 times less than for ^{235}U or ^{238}U for vegetation. The data from four profile samples are plotted and show the rapid decline in concentrations of ^{235}U with depth. This does not occur with ^{238}U, however, presumably because most ^{238}U present is natural uranium.

Data are presented to compare $^{239-240}Pu$ concentrations in "ball-mill" versus "sieved" (<100 mesh) soil fractions from soil samples analyzed by two laboratories. Most data examined here via simple regression techniques suggest that for soil samples collected in relatively-high-

activity strata, the $^{239-240}Pu$ concentrations in sieved soil aliquots can be considerably higher than obtained for ball-mill (BM) aliquots. Ratios of sieved to BM Pu activities as high as 25 are reported. For very low-level samples, however, the concentrations in BM aliquots are often higher than those in sieved aliquots. That is, the ratio of BM to sieved Pu concentrations does not appear in most cases to be constant over all activity levels. These data suggest that estimates of inventory in high-activity strata that are based only on sieved aliquots could be elevated 15 to 20 times above estimates obtained using BM fraction aliquots. Elevations on the order of 2.5 times appear possible for lower-activity strata. Pu concentrations in <100 mesh and >100 mesh soil fractions from the same soil sample are also compared. Eighteen of 21 ratios (<100 mesh/>100 mesh) were greater than 1, the highest ratio observed being 119. Regression analyses indicate the <100 mesh concentrations are significantly elevated over the >100 mesh concentrations and that the ratio is not constant over the range of activities encountered.

An attempt is made to "correct" the <100 and >100 mesh Pu concentrations back to field conditions by taking into account the proportion by weight of these soil fractions in the total soil sample collected and then computing a weighted mean Pu concentration. These corrected concentrations are more nearly equal to corresponding BM concentrations from the same soil sample, but the lack of a complete correction suggests that differences between laboratories as well as differences between the actual amounts of plutonium in soil fractions may in part account for differences between BM and sieved soil results. Some possible options for future action in obtaining final estimates of $^{239-240}Pu$ inventory in the Tonopah Test Range (TTR) and Area 11 are discussed relative to these results.

INTRODUCTION

In Gilbert *et al.* (1975), the available $^{239-240}Pu$ and ^{241}Am concentrations in soil samples from 9 of the 10 safety-shot study sites were analyzed relative to estimating the inventory and geographical distribution of plutonium in surface soil. Data from the remaining study

site, A Site in Area 11 (Plutonium Valley), have since become available and are analyzed in the present report. These data include $^{239\text{-}240}Pu$, ^{241}Am, ^{234}U, ^{235}U, ^{236}U, and ^{238}U for soil and/or vegetation. Approximately 50% of the data have still to be reported, but the available results (analyzed by LFE Laboratories) are used here to estimate: (1) average concentrations of $^{239\text{-}240}Pu$, ^{235}U, and ^{238}U in random soil and associated (adjacent) vegetation samples; (2) average concentrations of ^{234}U, ^{236}U, and total U ($^{234}U + ^{235}U + ^{236}U + ^{238}U$) in vegetation samples; (3) the inventory of ^{235}U, ^{238}U, $^{235}U + ^{238}U$, and $^{239\text{-}240}Pu$ in surface (0-5 cm depth) soil; (4) the relationship between soil and associated vegetation samples for ^{235}U and ^{238}U; and (5) the change in concentrations of ^{235}U, ^{238}U, and $^{239\text{-}240}Pu$ with depth of soil for four profile samples.

The remainder of this report concerns a comparison of plutonium and americium concentrations in different "fractions" of the soil matrix in samples collected from Area 13 and the Tonopah Test Range as part of the inventory sampling program. This information is particularly relevant at the present time since the three analytical laboratories currently performing radiochemical analyses for the NAEG have been sent either ball-mill (BM) or sieved soil fractions for analysis. Hence, the question arises whether the reported results from the three laboratories are really comparable, i.e., do the different fractions contain (on the average) different levels of plutonium concentration? That this might be the case is suggested by Tamura's (1974) work on the distribution of plutonium in different soil fractions. If there are differences in plutonium concentrations in different soil fractions, then changes may be required in soil preparation procedures so that all laboratories receive the same soil fraction for analysis.

Information is given here on plutonium and americium concentrations in the following soil fractions: "ball-mill," "<100 mesh," and ">100 mesh." A BM aliquot is one taken from the sample after the sample has been oven dried at 65° C. for 12 hours (allowing the sample to reach standard dry weight) and then ball-milled for 5 hours in a gallon can with ten 1-inch manganese forged steel balls per sample. A "<100 mesh" ("sieved") aliquot is one taken from BM soil that has passed through a 100 mesh sieve (2 or 8 inch). Finally, a ">100

mesh" aliquot is one taken from the ball-milled soil that will not pass through the 100 mesh sieve. LFE Laboratories, which processes about 60% of the total NAEG soil samples, receives 10 gram BM fraction aliquots for analysis. Los Alamos Scientific Laboratories (LASL) received BM fraction aliquots from samples collected in Areas 13 and 5 (GMX) and sieved aliquots from the four sites on TTR (Double Track, Clean Slates 1, 2, and 3) and from A, B, C, and D Sites in Area 11. Lab X has received only sieved samples.

For TTR and Area 11, a number (about 7% of the total samples) of "cross-check" soil aliquots were sent out for analysis, wherein more than one laboratory received an aliquot from the same sample collected in the field. Aliquots from these samples that were sent to LFE were from the BM fraction, while those going to LASL and Lab X were aliquots from the sieved fraction. On a number (about 3%) of soil samples from TTR and Area 11, as much of the total BM soil sample as possible was sieved so that the entire sample as it came from the ball mill was divided into two fractions, <100 mesh and >100 mesh. The weights of these two fractions were recorded and aliquots from both fractions sent to LASL for analysis. (As a general rule, only enough sample is passed through the sieve so that a 50 gram aliquot of the "fines" of <100 mesh soil is obtained for shipment to LASL or Lab X.) In addition, inter-intralaboratory samples (about 2% of the total from TTR and Area 11) were sent to all three laboratories to look for differences between laboratories and to estimate the within-lab variability (between aliquots from the same sample). These provide us with additional information on differences between $^{239\text{-}240}Pu$ concentrations in BM and sieved aliquots from the same soil sample. This paper analyzes the data from Area 13 and TTR that have been reported to date by LFE and LASL. Results are not yet available from Lab X for TTR.

The conclusions reached from the analyses of these BM, <100 mesh, and >100 mesh data are relevant to the question of how to proceed in obtaining final estimates of plutonium inventory and distribution in surface soils on the TTR and in Area 11. As noted above, Gilbert *et al.* (1975) have reported preliminary estimates of $^{239\text{-}240}Pu$ inventory for the TTR and Area 11 safety-shot sites. These are based on results

from about 60% of the collected soil samples from these areas that were analyzed for $^{239-240}Pu$ on BM aliquots by LFE laboratories. For the remaining 40% of the samples, aliquots from the sieved soil were sent to LASL and LFE for analyses, some of which have not yet been analyzed. If there are significant differences in plutonium concentrations between BM and sieved aliquots, then at least three options are available for future action: (1) have LASL and Lab X analyze the sieved aliquots already shipped to them, adjusting, if possible, their plutonium concentrations to what would be expected if they had analyzed BM fraction aliquots; (2) recall the sieved aliquots from LASL and Lab X, replacing them with new BM fraction aliquots from stored library samples; or (3) accept as final the preliminary estimates of plutonium inventory for TTR and B, C, and D Sites in Area 11 given in Gilbert *et al.* (1975) and for A Site of Area 11 given in this report. These alternatives will be discussed following our presentation of the data.

Estimates of $^{239-240}Pu$ inventory reported in Gilbert *et al.* (1975) for Area 13 (Project 57) on the NTS are based entirely on BM aliquots and hence are not in need of correction. This is also true for the estimated inventory reported for Area 5 except for 18 samples in stratum 2 analyzed by Lab X on sieved aliquots. Hence, the discussion in this paper is also relevant to these 18 samples.

ANALYSIS OF DATA FROM "A SITE," AREA 11

The land area defined here as A Site is shown in Fig. 1 and the three strata drawn on the basis of Ge(Li) scans for ^{235}U on soil collected on a grid system are given in Fig. 2. Estimates of average concentrations of $^{239-240}Pu$, ^{235}U, and ^{238}U in random soil and associated vegetation are given in Table 1. Table 2 gives average concentrations in vegetation for ^{234}U, ^{235}U, and total U (^{234}U + ^{235}U + ^{236}U + ^{238}U). Table 3 gives the estimated inventories (total amounts) of ^{235}U, ^{238}U, ^{235}U + ^{238}U, and $^{239-240}Pu$ in surface soil (0-5 cm) at A Site. The uranium inventories are estimated separately for natural uranium and for uranium from the safety-shot device.

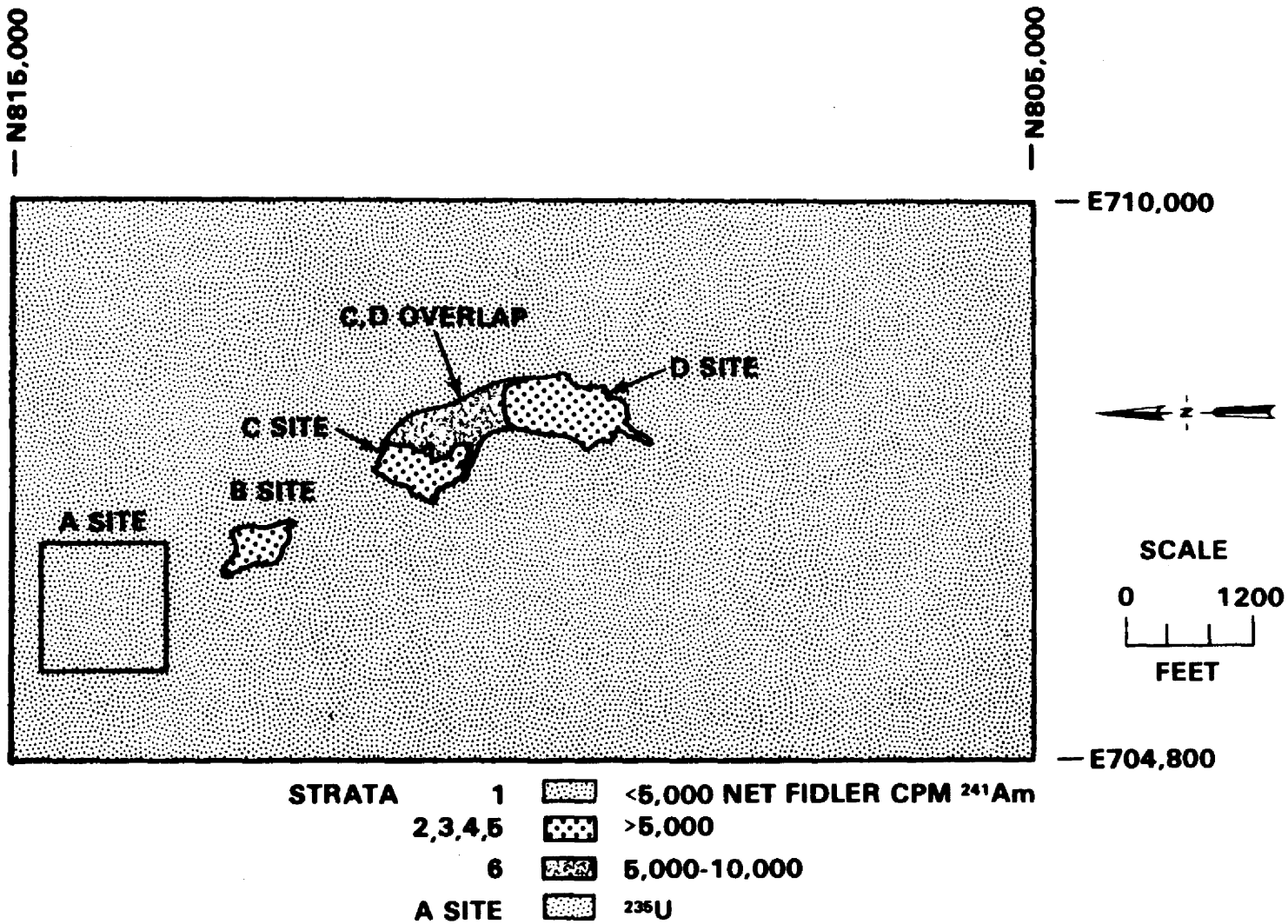

FIGURE 1. AREA 11 (PLUTONIUM VALLEY)

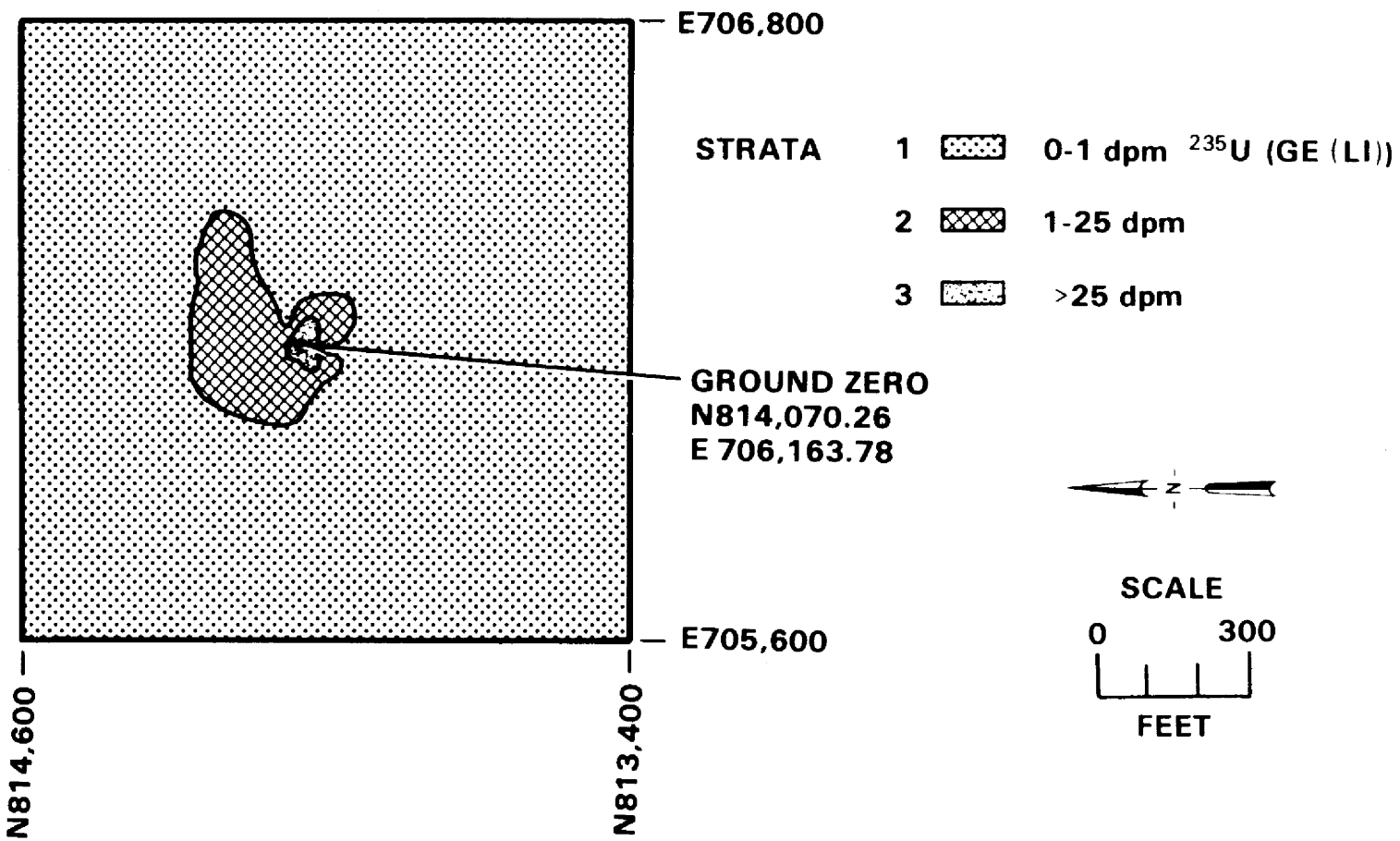

FIGURE 2 AREA 11 — A SITE
STRATA USED IN SAMPLING FOR INVENTORY

Table 1. Estimated[††] Concentrations of $^{239-240}Pu$, ^{235}U, and ^{238}U in Random Soil and Associated Vegetation Samples From A Site of Area 11, NTS

Isotope	Strata	Soil n	Soil Mean ± S.E.	Soil c[†]	Soil Min.	Soil Max.	Vegetation n	Vegetation Mean ± S.E.	Vegetation c[†]	Vegetation Min.	Vegetation Max.	Veg. Mean/ Soil Mean
$^{239-240}Pu$ (pCi/g dry wt.)	1	12	5.92 ± 2.16	1.3	0.955	25.8	12	1.47 ± 0.569	1.3	0.0562	7.32	0.25
	2	14	1.46 ± 0.335	0.8	0.124	4.08	18	0.640 ± 0.0989	0.7	0.234	1.87	0.44
	3	9	32.4 ± 31.4	2.9	0.101	284	6	1.04 ± 0.377	0.89	0.126	2.83	0.032
	Total	35	11.0 ± 8.07	4.4	0.101	284	36	0.982 ± 0.209	1.3	0.126	7.32	0.089
^{235}U (ng ^{235}U/g dry wt.)	1	14	30.0 ± 13.9	1.7	2.41	210	12	4.07 ± 1.31	1.1	0.795	13.1	0.14
	2	14	1,090 ± 454	1.6	52.3	5,500	20	49.1 ± 12.2	1.1	2.45	178	0.045
	3	10	17,200 ± 7,090	1.3	270	67,000	6	74.2 ± 15.7	0.5	37.8	135	0.0043
^{238}U (ng ^{238}U/g dry wt.)	1	14	1,670 ± 198	0.44	155	2,630	12	41.4 ± 5.69	0.48	8.57	71.3	0.025
	2	14	2,120 ± 206	0.36	1,410	4,160	20	71.8 ± 8.76	0.55	18.2	158	0.034
	3	10	11,700 ± 3,610	0.98	1,820	36,300	6	81.3 ± 11.4	0.34	35.7	114	0.0069

[†] c = coefficient of variation = $s/\bar{x}$

[††] Preliminary estimates. About 50% of results have been reported.

Table 2. Estimated[†††] Concentrations of ^{234}U, ^{236}U, and Total[†] Uranium for Vegetation at A Site of Area 11, NTS

Isotope	Strata	n	Mean ± S.E.	c[††]	Min.	Max.
^{234}U (ng ^{234}U/g dry wt.)	1	7	0.0929 ± 0.0274	0.78	0.0137	0.198
	2	20	0.548 ± 0.136	1.1	0.0279	1.99
	3	6	0.822 ± 0.168	0.50	0.419	1.43
^{236}U (ng ^{236}U/g dry wt.)	1	7	0.0246 ± 0.00817	0.88	0.00417	0.0527
	2	20	0.203 ± 0.0502	1.1	0.0103	0.731
	3	6	0.312 ± 0.0650	0.51	0.156	0.550
Total U[†] (ng/g dry wt.)	1	12	45.6 ± 6.56	0.50	15.2	84.3
	2	20	121 ± 20.4	0.75	20.7	321
	3	6	141 ± 26.9	0.47	74.0	251

†^{234}U + ^{235}U + ^{236}U + ^{238}U = Total U (see Table 1 for ^{235}U and ^{238}U vegetation results)

††$c = s/\bar{x}$

†††Preliminary estimates. About 50% of results have been reported.

Table 3. Estimated[†††] Inventory of ^{235}U, ^{238}U, $^{235}U + ^{238}U$, and $^{239-240}Pu$ in Surface Soil (0-5 cm Depth) at A Site in Area 11

Isotope	Strata	Size of Area m^2	Size of Area Percent	n	Natural Uranium + Safety-Shot Uranium: Mean ± S.E. (g/m^2)	Estimated Inventory ± S.E. (grams)	Percent Inventory in Strata	Estimated 95% C.L. on Inventory (grams) Lower	Estimated 95% C.L. on Inventory (grams) Upper	c† of Inventory Estimate
^{235}U	1	125,592	93.9	14	0.00142 ± 0.000698	178 ± 88	17	-12	368	0.49
	2	7,714	5.8	14	0.0434 ± 0.0168	335 ± 130	32	54	616	0.39
	3	475	0.4	10	1.12 ± 0.473	532 ± 225	51	24	1,040	0.42
	Total	133,781	100.1	38		1,045 ± 274	100	457	1,630	0.26
^{238}U	1	125,592	93.9	14	0.0754 ± 0.00910	9,470 ± 1,140	90	7,000	11,900	0.12
	2	7,714	5.8	14	0.0952 ± 0.00845	734 ± 65	7	594	874	0.09
	3	475	0.4	10	0.725 ± 0.249	344 ± 118	3	77	611	0.34
	Total	133,781	100.1	38		10,500 ± 1,150	100	8,020	13,000	0.11
$^{235}U + ^{238}U$	1	125,592	93.9	14	0.0768 ± 0.00924	9,650 ± 1,160	83	7,140	12,200	0.12
	2	7,714	5.8	14	0.139 ± 0.0239	1,070 ± 184	9	673	1,470	0.17
	3	475	0.4	10	1.84 ± 0.721	876 ± 342	8	102	1,650	0.39
	Total	133,781	100.1	38		11,600 ± 1,220	100	8,960	14,200	0.11
					($\mu Ci/m^2$)	(Curies)		(Curies)		
$^{239-240}Pu$	1	125,592	93.9	12	0.265 ± 0.0955	0.0333 ± 0.0120	96	0.00688	0.0597	0.36
	2	7,714	5.8	14	0.0674 ± 0.0153	0.00052 ± 0.00012	1.5	0.00026	0.00078	0.33
	3	475	0.4	8	1.77 ± 1.73	0.000841 ± 0.000822	2.4	-0.00110	0.00279	0.98
	Total	133,781	100.1	34		0.0347 ± 0.012	99.9	0.0083	0.061	0.35

Table 3. (Continued)

Isotope	Strata	Natural Uranium: Estimated[††] Mean ± S.E. (grams/m^2)	Natural Uranium: Estimated Inventory ± S.E. (grams)	Natural Uranium: Estimated 95% C.L. on Inventory (grams) Lower	Natural Uranium: Estimated 95% C.L. on Inventory (grams) Upper	Uranium From Safety-Shot Device: Estimated Inventory ± S.E. (grams)	Uranium From Safety-Shot Device: Percent Inventory in Strata	Uranium From Safety-Shot Device: Estimated 95% C.L. on Inventory (grams) Lower	Uranium From Safety-Shot Device: Estimated 95% C.L. on Inventory (grams) Upper
^{235}U	1	0.000462 ± 0.000028	58.1 ± 3.5	50.5	65.7	120 ± 88	12.2	-70	310
	2	0.000462 ± 0.000028	3.57 ± 0.22	3.09	4.05	331 ± 130	33.7	50	612
	3	0.000462 ± 0.000028	0.22 ± 0.013	0.19	0.25	532 ± 225	54.1	24	1,040
	Total	0.000462 ± 0.000028	61.9 ± 3.5	54.3	69.5	983 ± 274	100.0	395	1,570
^{238}U	1	0.0645 ± 0.000377	8,100 ± 47.3	8,000	8,200	1,370 ± 1,140	71.4	-1,090	3,830
	2	0.0645 ± 0.000377	498 ± 2.9	492	504	236 ± 65	12.3	96	376
	3	0.0645 ± 0.000377	30.6 ± 0.18	30	31	313 ± 118	16.3	46	580
	Total	0.0645 ± 0.000377	8,630 ± 47.4	8,530	8,730	1,920 ± 1,150	100.0	-564	4,400
$^{235}U + ^{238}U$	1	0.0650 ± 0.000378	8,160 ± 47.5	8,060	8,260	1,490 ± 1,160	51.4	-1,040	4,020
	2	0.0650 ± 0.000378	501 ± 2.9	495	507	569 ± 184	19.6	168	970
	3	0.0650 ± 0.000378	30.9 ± 0.18	30	31	845 ± 342	29.1	56	1,630
	Total	0.0650 ± 0.000378	8,690 ± 47.6	8,590	8,790	2,900 ± 1,220	100.1	240	5,600

[†]c = coefficient of variation

[††]Estimated using data from the 10 samples (2.5-cm increments) of profile 3, stratum 1, A site. The $\bar{x}$ ± S.E. atom percent ^{235}U for these samples was 0.720 ± 0.0064. Atom percent of natural ^{235}U is 0.720. More data is needed to confirm these estimates of natural ^{235}U.

[†††]Preliminary estimates. About 50% of results are reported.

The average concentration of $^{239-240}Pu$ in A Site over all three strata is 11.0 $\pm$ 8.07 pCi/g ($\bar{X} \pm$ S.E.) which is close to the 21 $\pm$ 6.6 pCi/g reported in Gilbert *et al.* (1975, Table 16), for the <5,000 cpm region in Area 11, of which A Site is a part (Fig. 1). The plutonium levels in A Site are low compared with those at B, C, and D Sites in Area 11 (Table 16, Gilbert *et al.*, 1975). The ratio of vegetation mean to soil mean for $^{239-240}Pu$ in A Site is 0.095 (Table 1). If one considers only the 14 paired soil, vegetation Pu concentrations, the ratio of vegetation mean to soil mean is 0.064 $\pm$ 0.096, which is similar to that reported in Gilbert *et al.* (1975, Table 17) for Area 11. The paired vegetation-soil data for ^{235}U and ^{238}U for all strata combined are plotted in Figs. 3 and 4. The estimated linear correlation between soil and vegetation is 0.93 for ^{235}U, but only 0.35 for ^{238}U. The higher correlation for ^{235}U could be related to the preponderance of this isotope in the device, whereas the ^{238}U present is primarily natural uranium.

Three models have been fit to these data in Figs. 3 and 4.

1. $y_i = \beta_1 x_i + e_i$, where $\hat{\beta}_1 = \sum_{i=1}^{n} y_i / \sum_{i=1}^{n} x_i$

2. $y_i = \beta_2 x_i + e_i$, where $\hat{\beta}_2 = \sum_{i=1}^{n} (y_i/x_i)/n$

and

3. $y_i = \beta_3 + \beta_4 x_i + e_i$,

the usual linear regression model (see Snedecor and Cochran, 1967, p. 167, for discussions on Models 1 and 2). It is clear that Model 2 is inappropriate for both ^{235}U and ^{238}U, that Model 1 is considerably better, but that Model 3 is the best of the three for ^{235}U, it too being inadequate for ^{238}U. The poor fits using the commonly used ratio estimates $\hat{\beta}_1$ and $\hat{\beta}_2$ suggest that calculation of such ratios without first plotting the data and checking for adequate fits can give misleading results. Gilbert *et al.* (1975) report similar poor fits using $\hat{\beta}_1$ and $\hat{\beta}_2$ for $^{239-240}Pu$ concentrations in adjacent soil and vegetation samples at the other safety-shot sites. Model 3 was also fit to the logs of the soil and vegetation pairs, but the

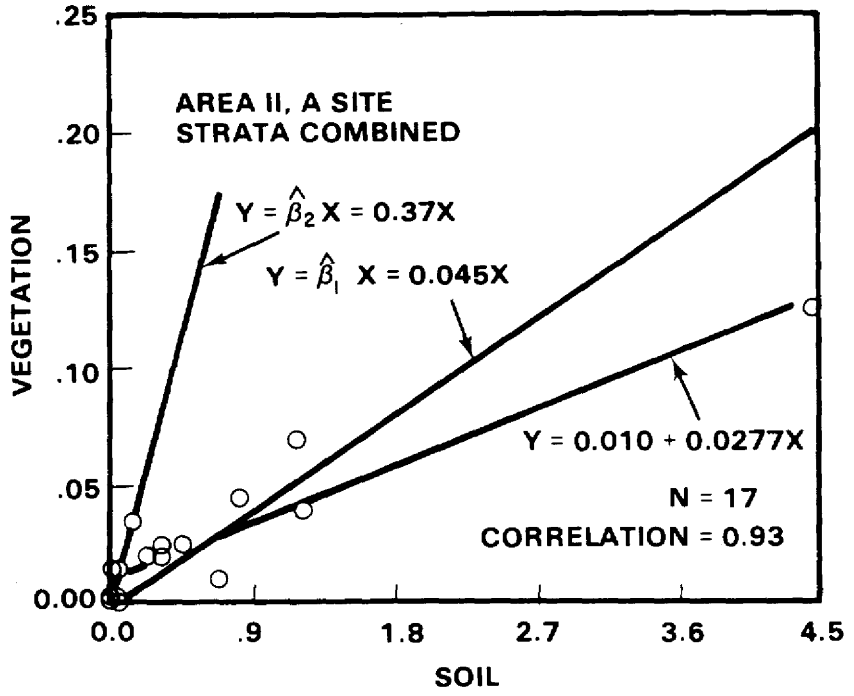

FIGURE 3 VEGETATION VS SOIL
^{235}U (μg/g DRY)

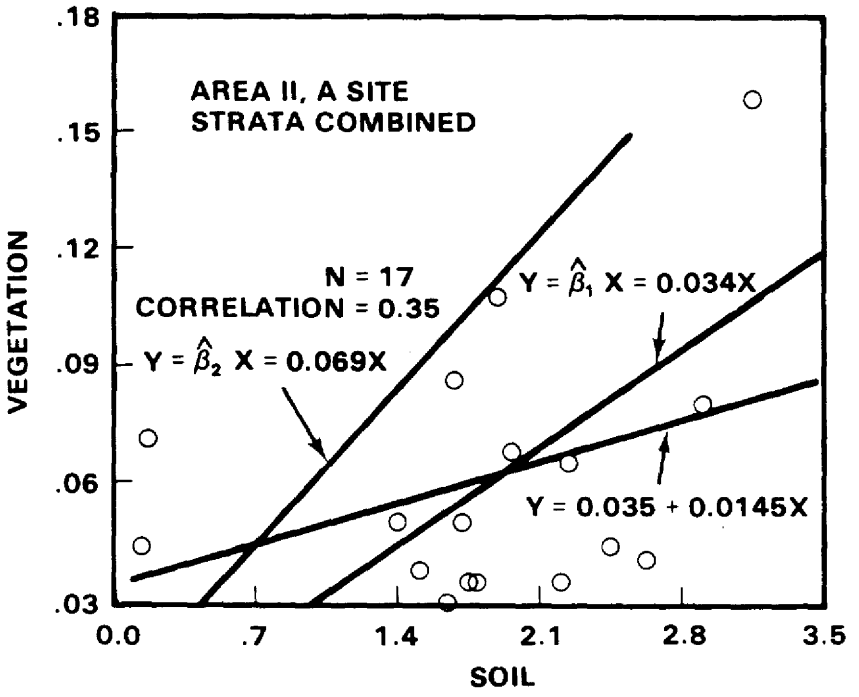

FIGURE 4 VEGETATION VS SOIL
^{238}U (μg/g DRY)

resulting fits were worse than when the original data were used (correlation of logs was 0.77 and 0.11 for ^{235}U and ^{238}U, respectively).

Both ^{235}U and ^{238}U concentrations for soil and vegetation increase as one approaches ground zero. The increase in vegetation concentration proceeds at a lower rate, however, than that for soil. This is reflected in the decreasing ratio of vegetation mean to soil mean given in the last column of Table 1 for ^{235}U and ^{238}U. This same phenomenon was reported for $^{239-240}Pu$ in vegetation and soil by Romney *et al.* (1975, Table 2) for the other safety-shot sites. Romney attributes this decreasing ratio in higher activity strata to differential particle size distribution that occurred within the fallout patterns of these events. The ratios themselves are of the same order of magnitude as observed for $^{239-240}Pu$ at the other sites.

The average concentrations of ^{234}U and ^{236}U (Table 2) for vegetation are approximately 100 times less than the vegetation concentrations for ^{235}U and ^{238}U (Table 1). There is a modest increase in these concentrations for strata near ground zero. The average vegetation concentrations for total U is about the same for strata 2 and 3, which are about twice the level of stratum 1 (Table 2).

The estimated inventories of ^{235}U and ^{238}U in the surface 5 cm of soil for A Site are given in Table 3 as 1,045 $\pm$ 274 and 10,500 $\pm$ 1,150 grams, respectively. Using the 10 samples (2.5-cm increments) from a single profile sample in stratum 1 of A Site, which we believe to contain only natural uranium (see the footnote to Table 3 and the discussion in the next paragraph), the total amount of natural ^{235}U and ^{238}U present in the top 5 cm of A Site soil was estimated to be 61.9 $\pm$ 3.5 and 8,630 $\pm$ 47.4 grams, respectively. By subtraction of the strata inventories, the inventory resulting from only the safety shot was found to be 983 $\pm$ 274 and 1,920 $\pm$ 1,150 grams for ^{235}U and ^{238}U, respectively. Note, however, that the standard errors of these latter estimates are very large, resulting in approximate 95% confidence limits of from 395 to 1,570 grams for ^{235}U, and from -564 (or zero) to 4,400 grams for ^{238}U. More information at other locations in A Site is needed to verify our estimates of mean concentrations

for natural uranium before these estimates of device-related uranium can be accepted as accurate.

Four profile samples from A Site are currently available for study. Figs. 5, 6, 7, and 8 are plots of the ^{235}U, ^{238}U, and $^{239-240}Pu$ results from the 10 levels (2.5-cm increments) from each profile (the data from two other profiles of A Site are not yet available). Figs. 5 and 6 show the decrease with depth in atom percent ^{235}U and concentration of ^{235}U (μg/g dry soil), respectively. The device used in the safety shot at A Site was largely ^{235}U, and the decrease in concentration with increasing distance from GZ is reflected in Figs. 5 and 6. Concerning stake 3 in stratum 1 (210 feet from GZ), the atom percent averages 0.720 over the 10 depths and plots as a horizontal line in Fig. 5. As noted above, since the atom percent of natural ^{235}U is 0.72, the ^{235}U concentrations for this profile (plotted in Fig. 6) have been used to approximate the inventory of natural ^{235}U (Table 3). The concentrations for ^{238}U for this profile were used in a similar manner. Fig. 7 is a plot of ^{238}U concentrations in these same profile samples. The slightly elevated concentrations in the first 2.5 cm of soil for those three profiles nearest ground zero suggest a relatively small percentage increase of ^{238}U to the surface soil from the device. This contrasts with the rather large percentage increase in ^{235}U concentrations in surface soil for profiles near ground zero (Fig. 6). Fig. 8 gives the available profile data for $^{239-240}Pu$. Most of the plutonium appears to be in the top 2.5 cm, with perhaps a gradual decline with depth below 5 cm. The plutonium present here could have come in part from the safety-shot device at B Site which is some 1,600 feet or so away. The lines in Figs. 5 and 8 were fit by eye. In Fig. 6, the data for two profiles were fit by the sum of two exponential terms (parameter estimates statistically significant at $\partial = 0.05$ or less).

CONCENTRATIONS IN DIFFERENT SOIL FRACTIONS

Analysis of plutonium and americium concentrations in ball-mill, <100 mesh, and >100 mesh soil fractions was approached by asking, and attempting to answer, the following three questions:

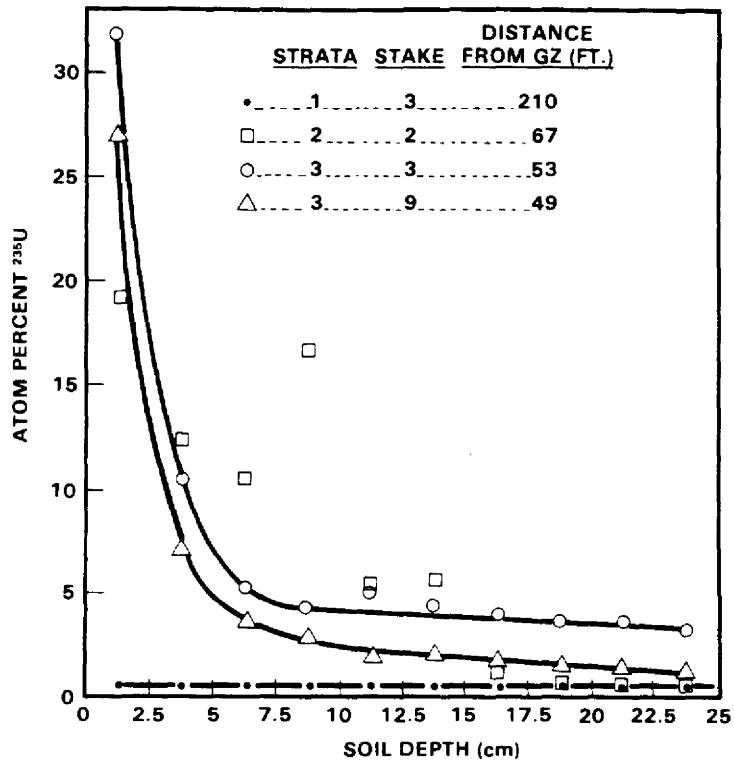

FIGURE 5 ATOM PERCENT ^{235}U IN SOIL PROFILES A SITE, AREA II

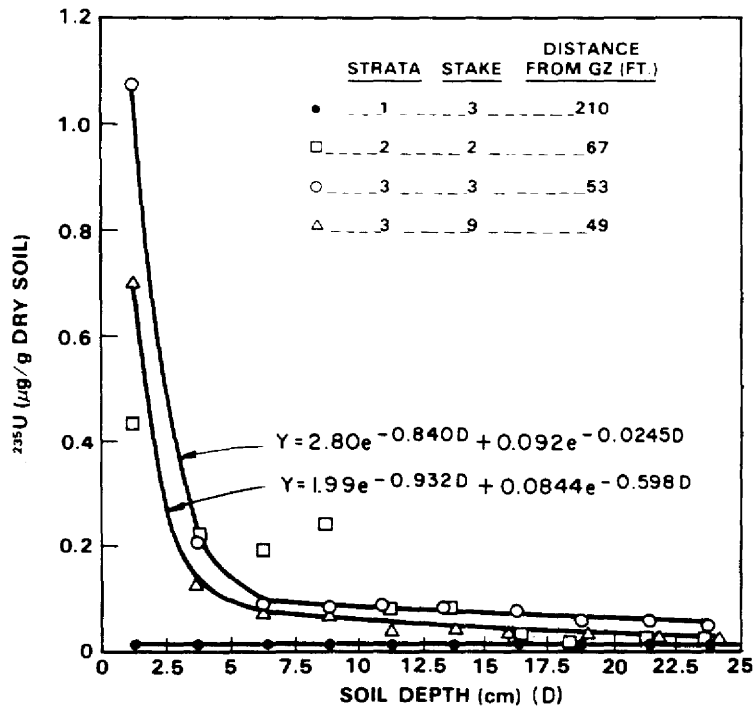

FIGURE 6 ^{235}U CONCENTRATIONS IN SOIL PROFILES A SITE, AREA 11

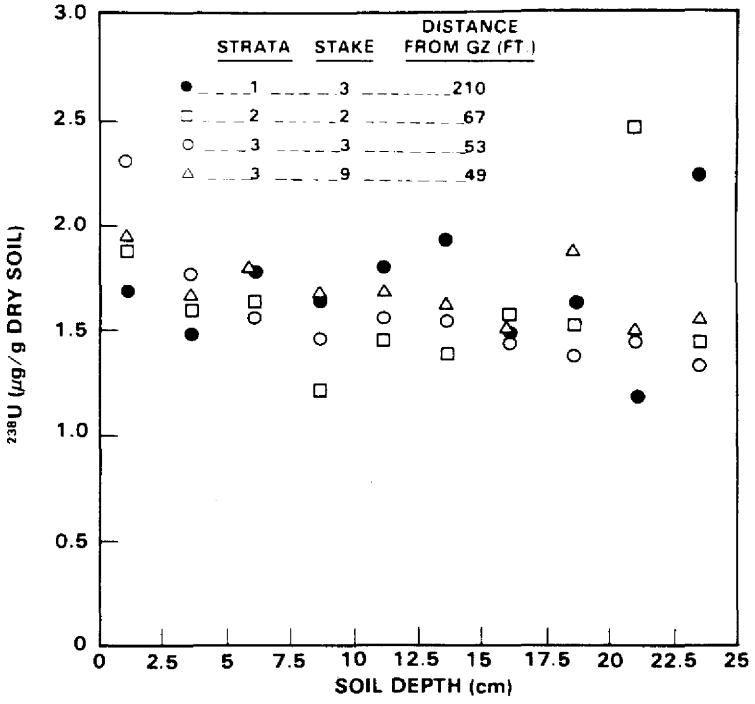

FIGURE 7 ^{238}U CONCENTRATIONS IN SOIL PROFILES A SITE, AREA 11

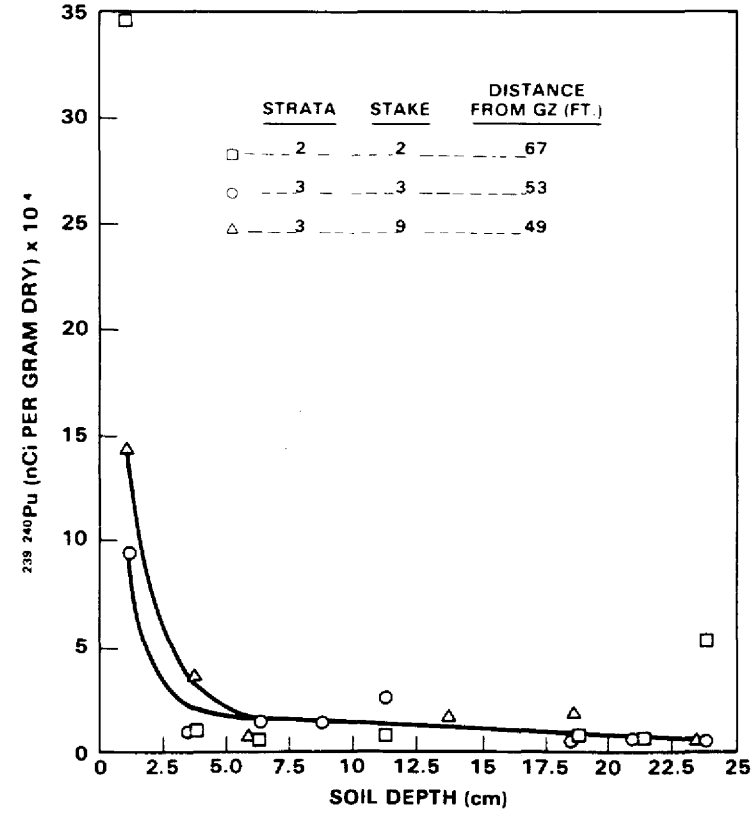

FIGURE 8 $^{239\text{-}240}Pu$ CONCENTRATIONS IN SOIL PROFILES A SITE, AREA 11

1. Do ball-mill (BM) and <100 mesh (sieved) soil fractions contain equal plutonium concentrations?

2. If the answer to Question 1 is no, then what is the error in estimating inventory by using concentrations from sieved aliquots in place of those from BM fractions?

3. If additional information on the weights and concentrations of the <100 mesh and >100 mesh fractions is available, can they be used to "correct" the <100 mesh and >100 mesh concentrations to what would be expected if BM fractions had been analyzed?

Throughout this discussion, it is assumed that BM fractions are the preferred fraction to be analyzed for plutonium and if differences in plutonium concentrations between BM and sieved aliquots are found, then the proper approach is to try and adjust the results on sieved aliquots to those obtained on BM fraction aliquots.

The largest single body of data available to investigate Question 1 is given in its entirety in Table 4. These are $^{239-240}Pu$ concentrations (nCi/g dry) in BM and sieved soil fractions from profile samples collected on the TTR. The library number in Column 1 defines a soil sample from which LFE was sent a 10-gram aliquot from the BM fraction, and LASL a 50-gram or larger aliquot from the sieved fraction. The data are tabulated by depth of soil sample and ordered from largest to smallest LASL plutonium concentrations within each depth. The most striking feature of these data are given by the ratio of LASL to LFE mean concentrations in the last column. For each soil depth, this ratio is seen to decrease as the LASL average concentration decreases. That is, the relationship between BM and sieved Pu concentrations is not a constant over all levels of contamination. LASL results are up to 20 or 25 times greater than reported by LFE for the highest sieved concentrations. Note, however, that the ratio is less than 1 for the lower LASL (sieved) concentrations, so that for these low-level samples, LFE is reporting higher concentrations than LASL. The logs of the mean data (Columns A and B in Table 4) are plotted in Fig. 9 and a linear regression equation is fitted. It is clear that the fitted line is not coincident with the line that should

Table 4. $^{239-240}Pu$ Concentrations (nCi/g) in "Ball-Mill" and "Less Than 100-Mesh" Soil Fractions for Soil Profile Samples From the Tonopah Test Range

Library No.	Site	Stake No.	Stratum	Depth (cm)	Less Than 100-Mesh†				Ball-Mill Fraction††		
					$^{239-240}Pu$	Percent Counting Error††††	Mean (A)	c†††	$^{239-240}Pu$ (B)	Percent Counting Error††††	A/B
L11553	DT	14	4	0- 2.5	6.08	1.7	6.08		0.442	2	13.8
L11590	CS2	24	4	2.5- 5.0	16.1	2.1	21.7	0.36	1.14	2	19.0
					27.3	1.6					
L11630	CS2	1	4	2.5- 5.0	2.18	1.6	2.08	0.068	0.477	4	14.6
					1.98	1.7					
L11604	CS3	3	3	2.5- 5.0	1.88	1.6	1.88		0.620	2	3.0
L11582	CS2	5	3	2.5- 5.0	1.79	1.8	1.79		0.392	3	4.6
L11623	CS1	3	4	2.5- 5.0	0.622	1.9	0.622		0.370	2	1.7
L11613	CS3	2	4	2.5- 5.0	0.0307	4.1	0.0438	0.42	0.0189	3	2.3
					0.0568	4.2					
L11543	DT	2	3	2.5- 5.0	0.0148	4.4	0.0148		0.00496	4	3.0
L11614	CS3	16	4	5.0- 7.5	6.04	1.8	6.04		0.238	2	25.4
L11619	DT	3	4	5.0- 7.5	0.432	2.0	0.432		0.418	3	1.0
L11572	CS1	11	4	5.0- 7.5	0.000509	4.2	0.000509		0.00101	8	0.50
L11561	CS1	11	3	5.0- 7.5	0.0000680	8.3	0.0000680		0.000374	20	0.18
L11617	DT	14	4	7.5-10.0	0.626	2.1	0.626		0.173	3	3.62
L11589	CS2	24	4	7.5-10.0	0.100	5.8	0.182	0.75	0.0234	3	7.78
					0.274	4.2					
					0.318	5.1					
					0.0349	4.9					
L11635	CS3	2	4	7.5-10.0	0.125	2.2	0.125		0.158	3	0.79
L11624	CS1	3	4	7.5-10.0	0.0404	8.1	0.0404		0.114	2	0.35
L11534	DT	12	2	7.5-10.0	0.000443	8.4	0.000346	0.39	0.000413	12	0.84
					0.000250	5.7					
L11596	CS3	15	2	7.5-10.0	0.000184	6.5	0.000184		0.00330	4	0.06
L11631	CS2	1	4	10.0-12.5	0.631	1.7	0.631		0.166	2	3.80
L11603	CS3	3	3	10.0-12.5	0.00550	4.4	0.00550		0.00367	5	1.50

Table 4. (Continued)

Library No.	Site	Stake No.	Stratum	Depth (cm)	Less Than 100-Mesh†				Ball-Mill Fraction††		A/B
					$^{239-240}$Pu	Percent Counting Error††††	Mean (A)	c†††	$^{239-240}$Pu (B)	Percent Counting Error††††	
L11620	DT	3	4	12.5-15.0	4.23	2.0	2.52	0.96	0.923	3	2.73
					0.802	2.0					
L11591	CS2	24	4	12.5-15.0	0.662		0.662		0.141	3	4.70
L11583	CS2	5	3	12.5-15.0	0.0372	3.9	0.0380	0.52	0.0267	3	1.42
					0.0229	3.7					
					0.0662	3.9					
					0.0254	4.6					
L11562	CS1	11	3	12.5-15.0	0.0205	4.0	0.0205		0.0273	2	0.75
L11636	CS3	2	4	12.5-15.0	0.00505	7.5	0.00505		0.0131	2	0.38
L11544	DT	2	3	12.5-15.0	0.000332	5.2	0.000486	0.45	0.000260	18	1.87
					0.000640	4.1					
L11573	CS1	11	4	12.5-15.0	0.0000168	22.6	0.0000254	0.48	0.000155	23	0.17
					0.0000339	13.2					
L11554	DT	14	4	15.0-17.5	0.141	2.4	0.141	2.4	0.0561	2	2.51
L11592	CS2	24	4	15.0-17.5	0.0514	5.5	0.138	1.2	0.0326	2	4.23
					0.323	4.3					
					0.0400	15.6					
L11535	DT	12	2	15.0-17.5	0.104	4.1	0.0762	0.55	0.0257	3	2.96
					0.0273	3.6					
					0.0937	4.0					
L11615	CS3	16	4	15.0-17.5	0.00527	3.6	0.00527		0.00111	8	4.75
L11625	CS1	3	4	15.0-17.5	0.000622	4.7	0.000477	0.43	0.000800	12	0.60
					0.000332	4.4					
L11605	CS3	3	3	15.0-17.5	0.0000536	11.3	0.0000536		0.00269	3	0.02
L11563	CS1	11	3	15.0-17.5	0.0000374	10.7	0.0000374		0.00360	6	0.01
L11584	CS2	5	3	17.5-20.0	0.00806	3.6	0.00903	0.15	0.000522	11	17.3
					0.0100	4.5					
L11545	DT	2	3	17.5-20.0	0.000172	6.6	0.000172		0.000935	9	0.18
L11621	DT	3	4	20.0-22.5	1.07	2.2	1.07		0.475	2	2.25
L11594	CS2	24	4	20.0-22.5	0.124	2.3	0.124		0.113	2	1.10
L11536	DT	12	2	20.0-22.5	0.000928	4.7	0.000928		0.00818	3	0.11

Table 4. (Continued)

Library No.	Site	Stake No.	Stratum	Depth (cm)	Less Than 100-Mesh†				Ball-Mill Fraction††		A/B
					$^{239-240}$Pu	Percent Counting Error††††	Mean (A)	c†††	$^{239-240}$Pu (B)	Percent Counting Error††††	
L11564	CS1	11	3	20.0-22.5	0.000210	5.7	0.000210		0.00125	8	0.17
L11574	CS1	11	4	20.0-22.5	0.0000313	13.3	0.0000332	0.079	0.00640	4	0.005
					0.0000350	12.1					
L11618	DT	14	4	22.5-25.0	0.0300	4.2	0.0275	0.13	0.0208	3	1.32
					0.0250	4.5					
L11616	CS3	16	4	22.5-25.0	0.0232	1.1	0.0232		0.0342	1	0.68
L11637	CS3	2	4	22.5-25.0	0.00734	3.7	0.00734		0.00533	2	1.38
L11546	DT	2	3	22.5-25.0	0.00150	4.1	0.00150		0.000873	13	1.72
L11581	CS2	5	3	22.5-25.0	0.00155	4.1	0.00140	0.15	0.00122	9	1.15
					0.00126	4.2					
L11626	CS1	3	4	22.5-25.0	0.000127	7.8	0.000127		0.000374	17	0.34
L11606	CS3	3	3	22.5-25.0	0.0000358	13.5	0.0000287	0.35	0.00139	7	0.02
					0.0000216	19.1					

†Analyzed by LASL

††Analyzed by LFE

†††Coefficient of variation = s/mean, where s = standard deviation

††††Percent counting error = 100σ/x, where σ = counting error

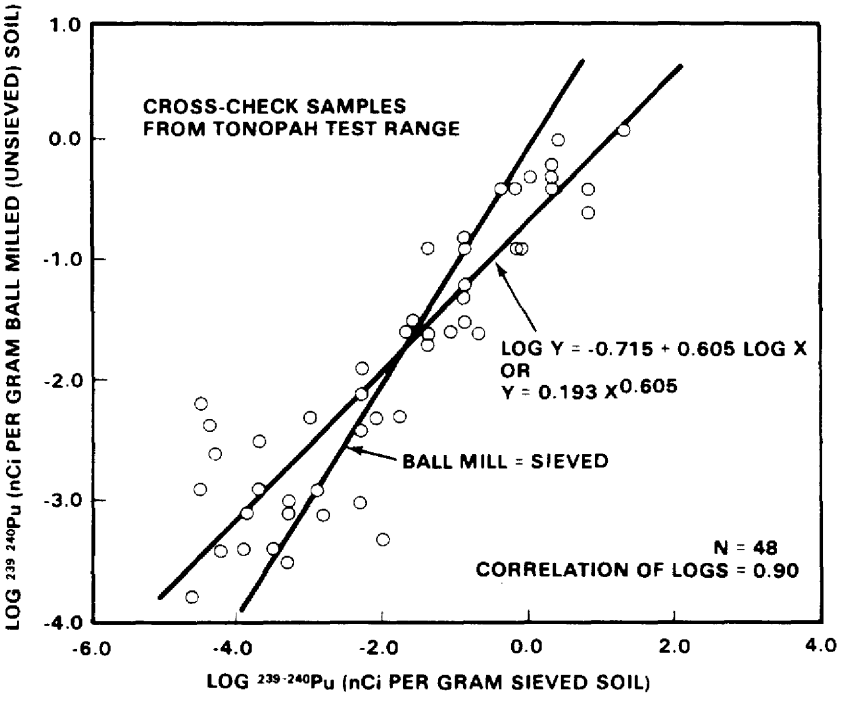

FIGURE 9 BALL MILL VERSUS SIEVED (<100 MESH) $^{239-240}Pu$ CONCENTRATIONS IN SOIL

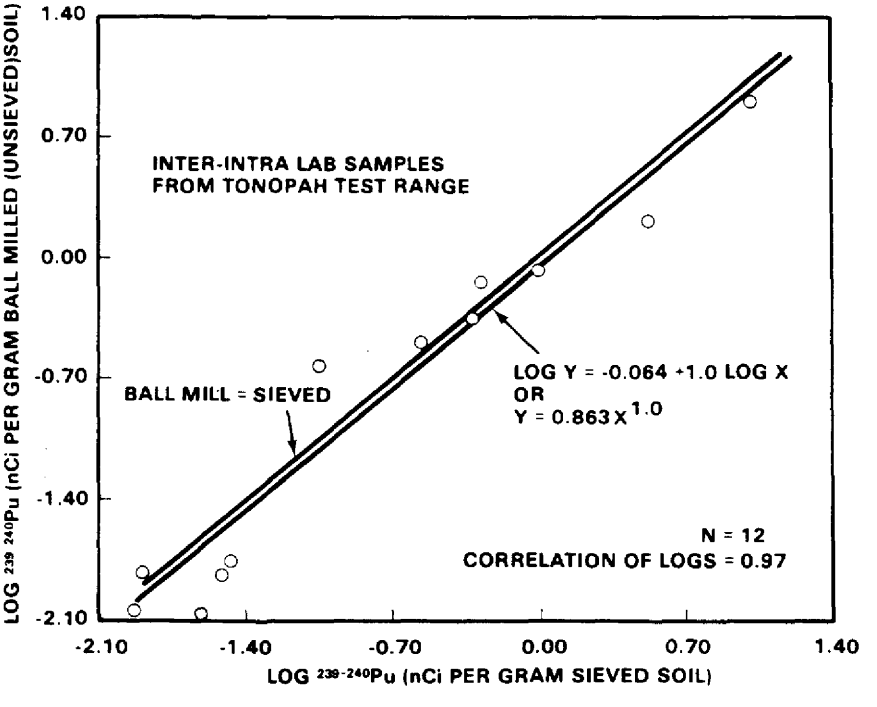

FIGURE 10 BALL MILL VERSUS SIEVED (<100 MESH) $^{239-240}Pu$ CONCENTRATIONS IN SOIL

result if Pu concentrations in BM and sieved aliquots from the same soil sample were equal over the wide range of concentrations encountered. If the Pu in BM and sieved fractions were equal, then in the linear model

$$\log y = \beta_0 + \beta_1 \log x ,$$

where y and x are the BM and sieved Pu concentrations, respectively, $\beta_0 = 0$ and $\beta_1 = 1$ so that log y = log x, or equivalently, y = x. However, $\beta_0 = 0$, $\beta_1 = 1$ are not within the computed 95% confidence region for β_0, β_1 for the mean data (labeled A and B) in Table 4 (see Draper and Smith, 1967, page 64, for computational details). Hence, the null hypothesis that $\beta_0 = 0$ and $\beta_1 = 1$ for these data was rejected.

Additional information relative to Question 1 is given by the data in Table 5. These are $^{239\text{-}240}$Pu concentrations (nCi/g dry) in BM and sieved soil fractions of surface (0-5 cm) samples analyzed by LFE and LASL. As was the case in Table 4, the mean LFE and LASL results are paired since for a given stake number the various aliquots (from BM or sieved fractions) are from the same soil sample. The logs of the mean data (Columns A and B) are plotted in Fig. 10. The fitted line is parallel to the line expected when BM = sieved, i.e., when $\beta_1 = 1$, but the sieved concentrations tend to be higher on the average than do the BM concentrations. Contrary to the case in Fig. 9, however, the points $\beta_0 = 0$, $\beta_1 = 1$ <u>are</u> within the joint 95% confidence region for β_0, β_1. Hence, for these data, the differences in elevation and slope between the two lines in Figure 10 are not statistically significant.

Looking at Figs. 9 and 10, one might ask why there is a parallel situation between the fitted and BM = sieved lines in Fig. 10 but not in Fig. 9. One difference between these two data sets is that the Pu concentrations in Fig. 9 are on profile samples, while those in Fig. 10 are restricted to surface (0-5 cm) samples. Hence, the data in Fig. 9 include much lower concentration levels than occur in Fig. 10. Could it be that a parallel situation exists only for higher-level concentrations? To investigate this possibility, only those data pairs from Table 4 for which the LASL Pu concentrations were approximately in the range of the LASL data used in Fig. 10

Table 5. $^{239-240}Pu$ Concentrations (nCi/g) in "Ball-Mill" and "Less Than 100-Mesh" Soil Fractions for Surface Soil Samples From the Tonopah Test Range; Inter-Intralaboratory Samples

Site	Stake No.	Stratum	Less Than 100-Mesh†					Ball-Mill Fraction††					A/B
			Library No.	$^{239-240}Pu$	Percent Counting Error	Mean (A)	c†††	Library No.	$^{239-240}Pu$	Percent Counting Error	Mean (B)	c†††	
DT	1	1	L11727	0.482	2.1	0.482		L11655	0.337	2	0.471	0.43	1.0
								56	0.702	2			
								57	0.373	2			
DT	1	2	L11728	0.136	5.9	0.0914	0.69	L11658	0.322	9	0.219	0.50	0.42
				0.0468	3.8			59	0.232	3			
								60	0.103	3			
DT	14	3	L11729	0.0264	4.3	0.0226	0.23	L11661	0.00812	5	0.00843	0.35	2.7
				0.0189	3.9			62	0.00567	4			
								63	0.0115	3			
CS1	2	1	L11730	0.0117	4.4	0.0117		L11664	0.00334	11	0.00983	0.97	1.2
								65	0.00535	5			
								66	0.0208	4			
CS1	1	2	L11731	1.05	1.8	0.991	0.084	L11667	1.08	2	0.900	0.18	0.92
				0.932	1.7			68	0.754	2			
								69	0.867	2			
CS1	1	3	L11732	4.55	7.1	3.38	0.49	L11670	1.74	2	1.73	0.041	2.0
				2.20	3.4			71	1.65	2			
								72	1.79	3			
CS2	2	1	L11733	0.0129	5.5	0.0129		L11673	0.0106	8	0.0162	0.52	0.80
								74	0.0121	6			
								75	0.0259	5			
CS2	2	2	L11734	0.536	2.4	0.536		L11676	0.800	3	0.718	0.10	0.75
								77	0.656	2			
								78	0.698	2			
CS3	24	1	L11735	0.282	2.1	0.282		L11679	0.315	3	0.331	0.12	0.85
								80	0.304	2			
								81	0.375	2			
CS3	35	1	L11736	0.0323	10.8	0.0323		L11682	0.0216	3	0.0149	0.39	2.2
								83	0.0111	2			
								84	0.0120	3			

Table 5. (Continued)

Site	Stake No.	Stratum	Less Than 100-Mesh†					Ball-Mill Fraction††					A/B
			Library No.	$^{239-240}Pu$	Percent Counting Error	Mean (A)	c†††	Library No.	$^{239-240}Pu$	Percent Counting Error	Mean (B)	c†††	
CS3	1	3	L11737	11.1 9.64	2.6 2.0	10.4	0.099	L11685 86 87	7.38 8.14 7.97	2 3 3	7.83	0.051	1.3
CS3	3*	3	L11738	0.0327	4.2	0.0327		L11688 89 90	0.0209 0.0135 0.0207	2 3 3	0.0184	0.23	1.8

*Profile sample; depth of 5.0-7.5 cm

†Analyzed by LASL

††Analyzed by LFE

†††Coefficient of variation = c = s/mean

were considered. The fit to these 31 data pairs is shown in Fig. 11. As in Fig. 9, the two lines are not parallel nor coincident, although β_0 is slightly nearer 0 and β_1 slightly nearer 1. Statistically speaking, however, the null hypothesis that $\beta_0 = 0$ and $\beta_1 = 1$ was still rejected. Hence, the inclusion of profile samples with low-level Pu concentrations does not appear to materially affect the regression relationship between the logs of Pu concentrations in BM and sieved soil fractions.

The comparison between BM and sieved results in Figs. 9 and 10 is complicated by the fact that one laboratory analyzed the BM aliquots and another the sieved aliquots. That is, part of the differences between BM and sieved results may be due to differences between laboratories. Table 6 gives $^{239-240}$Pu concentrations on BM and sieved aliquots of surface soil samples from stratum 2 of Area 13, all of which were analyzed by LASL. In Table 7, plutonium concentrations are given for BM and sieved aliquots from the 10 levels of a single profile sample in stratum 3 of Area 13, all of which were analyzed by LFE. The logs of these data are plotted in Figs. 12 and 13, and linear fits made to the data. Concerning the LASL data on surface samples in Fig. 12, the Pu concentrations in sieved samples are consistently higher than in BM fraction aliquots, and β_0, β_1 are significantly different from zero and 1, respectively, so that the two lines in Fig. 12 are neither parallel nor coincident. The range of concentrations in Fig. 12 is about the same as that in Fig. 11 where this same conclusion was reached. LASL also reported ^{238}Pu for these aliquots. These data are not given here, but a plot of the data and the fitted line look almost identical to that for $^{239-240}$Pu in Fig. 12 with identical conclusions. The LFE data from Table 7 (plotted in Fig. 13) exhibit considerable scatter since 8 of the 10 samples are at depths greater than 5 cm with resulting low Pu concentrations in both BM and sieved soil fractions. Note that the scatter below log concentrations of -2.0 for both BM and sieved aliquots is about the same in Fig. 13 as it is in Fig. 9. The fitted line in Fig. 13 suggests that sieved aliquots tend to yield Pu concentrations greater than obtained on BM aliquots. However, there are no statistically significant differences between the fitted

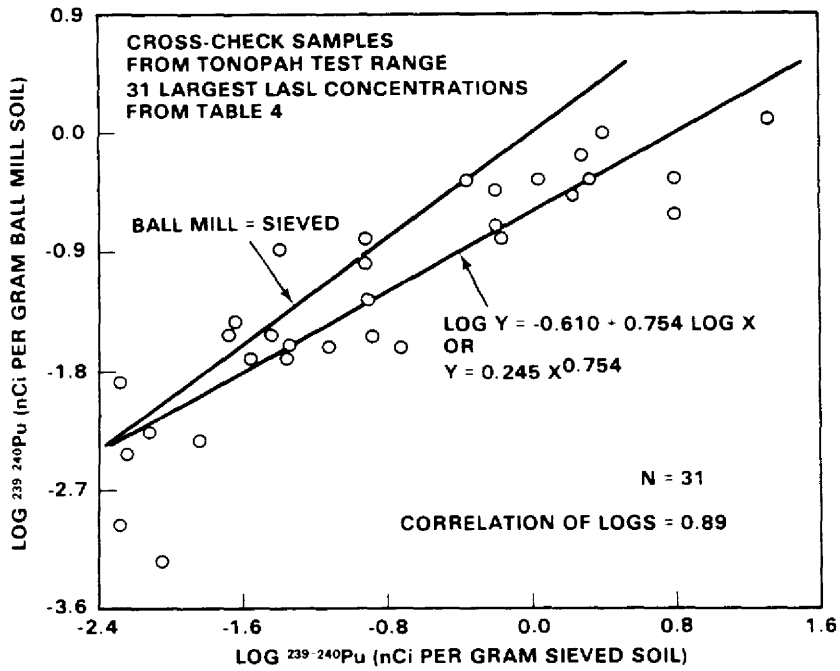

FIGURE 11 BALL MILL VERSUS SIEVED (<100 MESH) $^{239-240}Pu$ CONCENTRATIONS IN SOIL

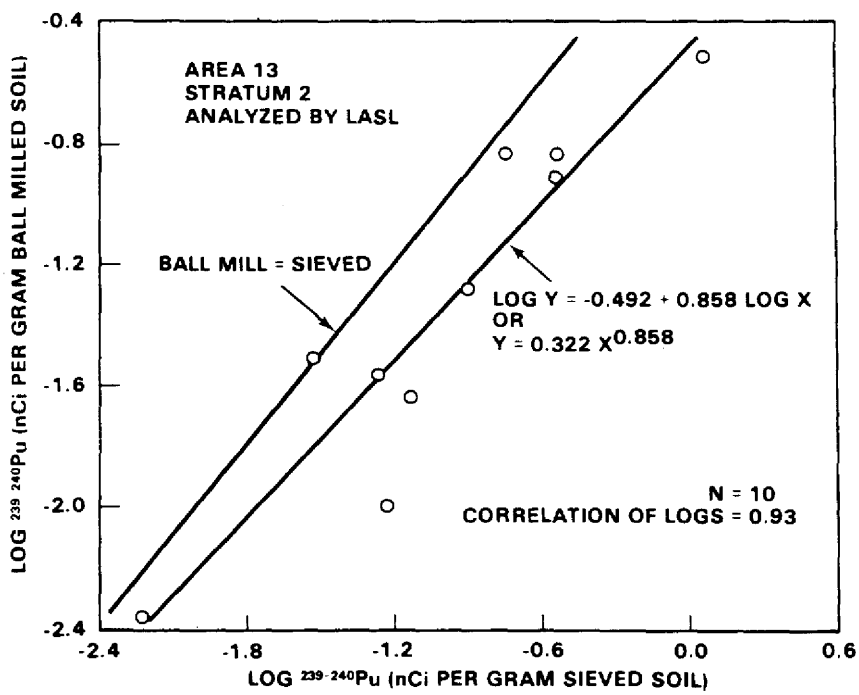

FIGURE 12 BALL MILL VERSUS SIEVED (<100 MESH) $^{239-240}Pu$ CONCENTRATIONS IN SOIL

Table 6. $^{239-240}Pu$ Concentrations (nCi/g) in "Ball-Mill" and "Less Than 100-Mesh" Soil Fractions for Surface Soil Samples From Stratum 2 of Area 13; Analyzed by LASL

Library No.	Stake No.	Less Than 100-Mesh				Ball-Mill Fraction				A/B
		$^{239-240}Pu$	Percent Counting Error	Mean (A)	c†	$^{239-240}Pu$	Percent Counting Error	Mean (B)	c†	
L00986	10	0.000932	4.4	0.00616	0.84	0.00304	3.7	0.00423	0.69	1.5
		0.00626	4.0			0.00757	3.4			
		0.0113	4.0			0.00208	3.9			
L00987	15	0.0377	4.6	0.0305	0.47	0.0334	3.8	0.0316	0.083	0.96
		0.0141	4.0			0.0297	3.9			
		0.0397	0.6							
L00988	16	2.05	1.6	1.12	0.59	0.202	1.6	0.299	0.37	3.8
		1.10	1.3			0.274	1.5			
		0.581	2.8			0.421	2.6			
		0.739	1.9							
L00989	18	0.144	1.7	0.298	0.93	0.140	1.7	0.139	0.47	2.1
		0.617	1.7			0.0730	1.6			
		0.134	2.0			0.205	1.8			
L00990	21	0.174	1.7	0.184	0.31	0.141	1.4	0.149	0.38	1.2
		0.132	1.8			0.189	1.6			
		0.246	2.3			0.193	1.7			
						0.0730	3.0			
L00991	23	0.048	3.9	0.0607	0.80	0.00865	3.2	0.0101	0.13	6.0
		0.0253	4.4			0.0105	3.8			
		0.116	1.9			0.0112	1.2			
L00992	25	0.286	1.7	0.290	0.02	0.113	1.6	0.115	0.47	2.5
		0.295	1.7			0.0613	1.7			
						0.170	1.9			
L00993	27	0.0316	2.2	0.124	0.69	0.0437	1.7	0.0535	0.45	2.3
		0.141	1.8			0.0811	1.8			
		0.199	2.4			0.0358	2.2			
L00994	33	0.0283	2.2	0.0552	0.60	0.0208	2.3	0.0271	0.81	2.0
		0.0455	2.1			0.00896	3.1			
		0.0919	2.0			0.0514	1.8			
L00995	35	0.0635	1.8	0.0734	0.92	0.0394	2.1	0.0227	0.64	3.2
		0.0116	3.0			0.0137	3.1			
		0.145	2.0			0.0151	2.9			

†c = coefficient of variation = s/mean, where s = standard derivation of n observations

Table 7. $^{239-240}Pu$ Concentrations (nCi/g) in "Ball-Mill" and "Less Than 100 Mesh" Soil Fractions for a Single Profile Sample From Stratum 3 of Area 13; Analyzed by LFE Laboratories

Library No.	Depth in Soil (cm)	Less Than 100 Mesh		Ball-Mill Fraction		A/B
		$^{239-240}Pu$ (A)	Percent Counting Error†	$^{239-240}Pu$ (B)	Percent Counting Error†	
L01048	0- 2.5	0.226	2	0.288	2	0.78
L01049	2.5- 5.0	0.0111	3	0.0115	28	0.97
L01050	5.0- 7.5	0.000711	3	0.00206	18	0.35
L01051	7.5-10.0	0.000817	3	0.000190	11	4.30
L01052	10.0-12.5	0.00197	19	0.0000685	12	28.8
L01053	12.5-15.0	0.000713	8	0.0000327	13	21.8
L01054	15.0-17.5	0.000125	9	0.0000288	16	4.34
L01055	17.5-20.0	0.000124	9	0.00440	6	0.028
L01056	20.0-22.5	0.00110	3	0.0000497	12	22.1
L01057	22.5-25.0	0.000163	6	0.00200	3	0.082

†Percent counting error = 100 σ/x, where σ = counting error

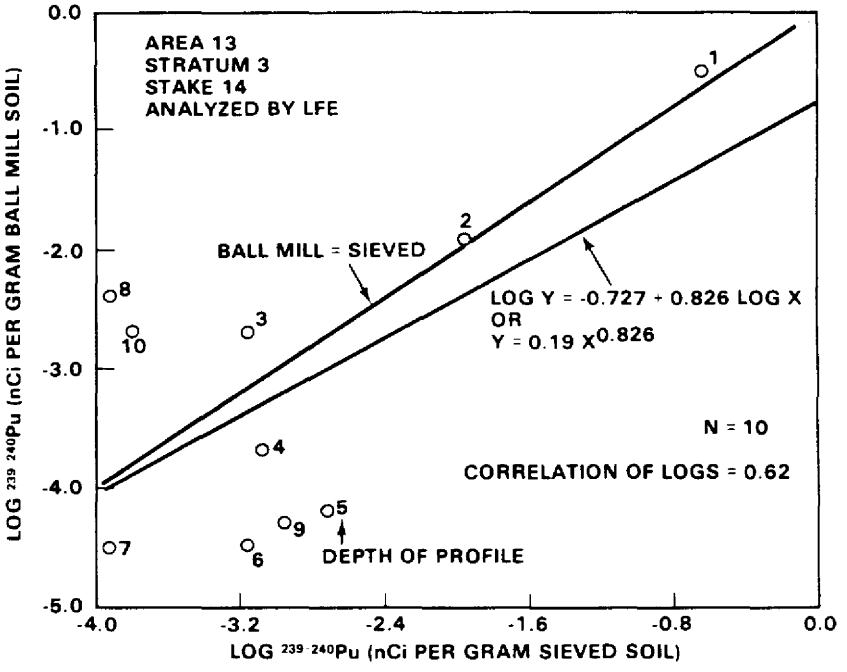

FIGURE 13 BALL MILL VERSUS SIEVED (<100 MESH) $^{239-240}Pu$ CONCENTRATIONS IN A SOIL PROFILE

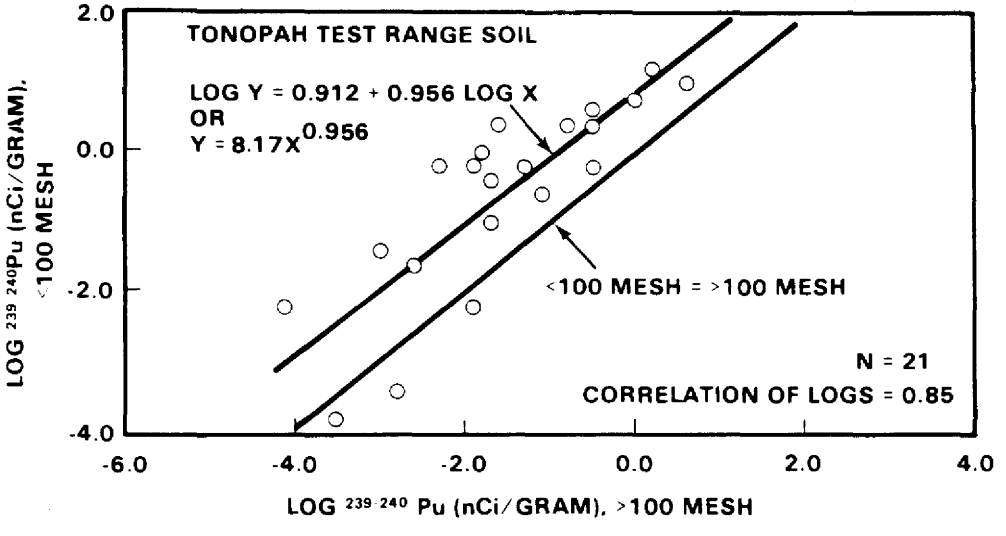

FIGURE 14 <100 MESH VERSUS >100 MESH $^{239-240}Pu$ IN SOIL

and BM = sieved line. This conclusion is not surprising in view of the large scatter due to low Pu concentrations. At these low levels, considerably more data would be required to detect differences between BM and sieved results.

Some information is also available on plutonium concentrations in <100 mesh (sieved) and >100 mesh fractions of the same soil sample. The BM fraction can be separated into these two fractions, and if it were shown that their Pu concentrations were not different, this would cast doubt on the results reported above that sieved and BM fractions do yield different Pu concentrations. In Fig. 14, the logs are plotted for 21 pairs of mean $^{239\text{-}240}$Pu concentrations obtained on <100 and >100 mesh soil fractions of profile samples. The <100 mesh data are given in Table 4, Library numbers L11617 through L11637, inclusive. The mean >100 mesh concentrations for 14 of the 21 samples are given in Column e of Table 8. The remaining mean $^{239\text{-}240}$Pu results for >100 mesh are given in the footnote on this page.[†] In all but three samples (L11625, L11626, and L11637 in Table 8, Columns b and e), the <100 Pu concentration is greater than the corresponding >100 Pu result for the same sample; i.e., the ratio A/B (Column k, Table 8) is greater than 1. Some of these ratios are very large, considerably larger in most cases than the ratio of <100 mesh to BM fraction (Column 1, Table 8). This is expected since the BM fraction contains both the <100 and >100 mesh fractions. In Figs. 15 and 16 are plotted the available ^{238}Pu and ^{241}Am results for these same soil samples. The general conclusions are the same as for the $^{239\text{-}240}$Pu data. In all three figures, the estimated 95% confidence region for β_0 and β_1 does not include $\beta_0 = 0$, $\beta_1 = 1$. In other words, the <100 mesh concentrations are significantly elevated over the >100 mesh concentrations, and the ratio is not constant over the range of concentrations considered.

What can one tentatively conclude relative to Question 1? First, for most data sets investigated, the Pu concentrations in BM fraction aliquots are not equal to those in <100 mesh aliquots, at least for

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[†]L11622, 0.0784; L11627, 3.80; L11628, 0.290; L11629, 2.49; L11632, 1.45; L11633, 0.325; L11634, 0.955. Units are nCi/g dry.

Table 8. $^{239-240}Pu$ Concentrations (nCi/g) in Different Profile Soil Fractions Before and After "Correction" to Field Conditions

Library No.	Site	Stake No.	Depth (cm)	Total Dry Wt. (g)	a Wt. (g)	b Less Than 100 Mesh $^{239-240}Pu$ Uncorrected† (A)	c Less Than 100 Mesh $^{239-240}Pu$ Corrected†††
L11617	DT	14	7.5-10.0	279	215	0.626	0.520
L11618	DT	14	22.5-25.0	309	230	0.0275	0.0221
L11619	DT	3	5.0- 7.5	400	212	0.432	0.238
L11620	DT	3	12.5-15.0	356	233	2.52	1.70
L11621	DT	3	20.0-22.5	379	230	1.07	0.688
L11623	CS1	3	2.5- 5.0	340	295	0.622	0.570
L11624	CS1	3	7.5-10.0	339	255	0.0404	0.0344
L11625	CS1	3	15.0-17.5	333	230	0.000477	0.000374
L11626	CS1	11	20.5-22.5	298	220	0.000127	0.0000941
L11630	CS2	1	2.5- 5.0	361	281	2.08	1.69
L11631	CS2	1	10.0-12.5	399	314	0.631	0.511
L11635	CS3	2	7.5-10.0	427	315	0.125	0.0944
L11636	CS3	2	12.5-15.0	517	354	0.00505	0.00358
L11637	CS3	2	22.5-25.0	466	339	0.00734	0.00546

Table 8. (Continued)

Library No.	d Wt. (g)	e Greater Than 100 Mesh $^{239-240}Pu$ Uncorrected† (C)	f Greater Than 100 Mesh $^{239-240}Pu$ Corrected††††	g $^{239-240}Pu$ Total Corrected*	h $^{239-240}Pu$ Ball-Mill†† (B)	i % <100 Mesh By Wt.**	j % $^{239-240}Pu$ in <100 Mesh****	k A/C	l A/B
L11617	44	0.0133	0.00226	0.522	0.173	83	99.6	47	3.6
L11618	56	0.00245	0.000480	0.0226	0.0208	80	97.9	11	1.3
L11619	172	0.0178	0.00797	0.246	0.418	55	96.8	24	1.0
L11620	113	0.0247	0.00808	1.70	0.923	67	99.5	102	2.7
L11621	128	0.0163	0.00583	0.693	0.475	64	99.2	66	2.3
L11623	27	0.00523	0.000439	0.570	0.370	92	99.9	119	1.7
L11624	44	0.000950	0.000140	0.0346	0.114	85	99.6	43	0.35
L11625	63	0.00155	0.000333	0.000708	0.000800	78	52.9	0.31	0.60
L11626	77	0.000286	0.0000742	0.000168	0.000374	74	55.9	0.44	0.34
L11630	65	0.142	0.0267	1.72	0.477	81	98.4	15	4.4
L11631	74	0.0509	0.00971	0.520	0.166	81	98.1	12	3.8
L11635	102	0.0192	0.00470	0.0991	0.158	76	95.3	6.5	0.79
L11636	145	0.0000865	0.0000251	0.00361	0.0131	71	99.3	58	0.39
L11637	117	0.0120	0.00308	0.00854	0.00533	74	63.9	0.61	1.4

†As reported by LASL

††As reported by LFE

†††Computed as (ba)/(a+d) = c

††††Computed as (ed)/(a+d) = f

*Computed as c + f

**Computed as 100a/(a+d)

***Computed as 100c/g

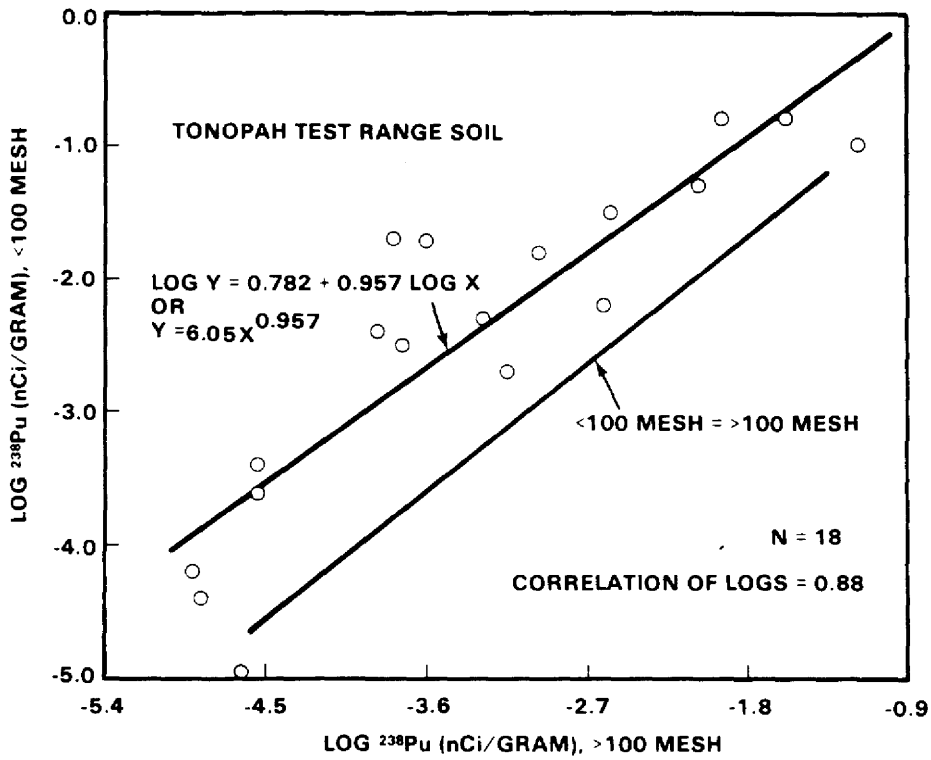

FIGURE 15 <100 MESH VERSUS >100 MESH ^{238}Pu IN SOIL

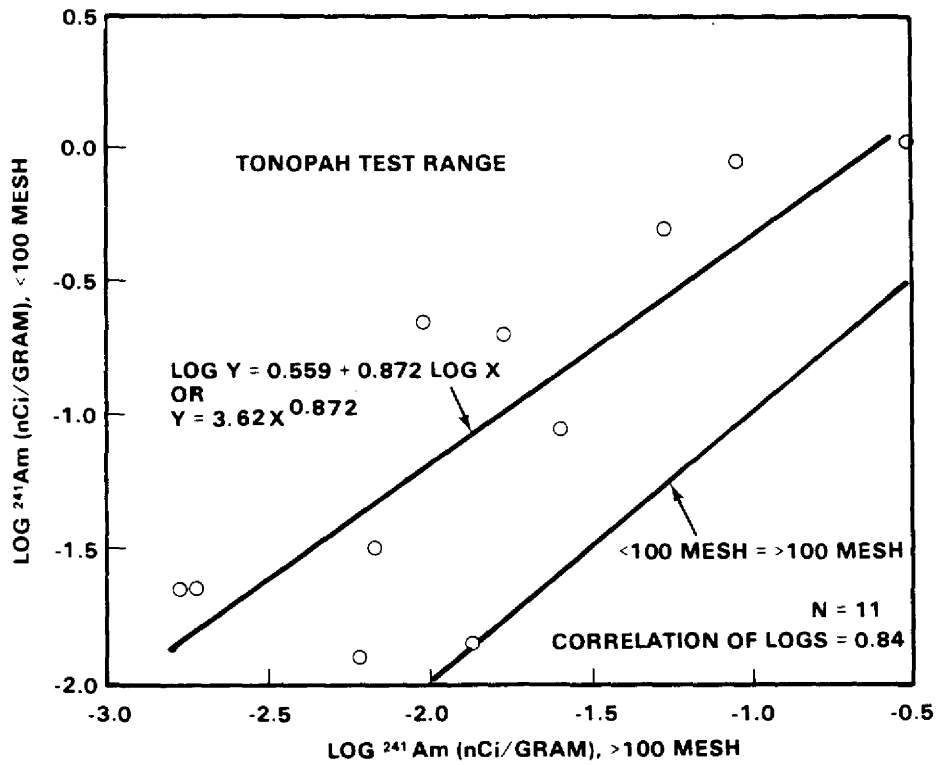

FIGURE 16 <100 MESH VERSUS >100 MESH ^{241}Am IN SOIL

relatively high contamination levels found in surface soil, where <100 mesh results tend to be considerably higher than those for BM fractions. Second, the ratio of BM to <100 mesh is not, in most cases, constant over all levels of contamination. Concerning Question 2, the error in estimating inventory using Pu concentrations from <100 mesh rather than BM aliquots depends on the ratio of concentrations in these two soil fractions which (as noted above) is not constant over all concentration levels. Probably a conservative estimate of the average effect can be obtained from the Pu data in Table 6. These are surface samples from the second stratum of Area 13, a low-level stratum as compared to strata closer to GZ at that site. For these data, the average ratio (sieved/BM) is 2.6 ± 0.5 ($\bar{X} \pm$ S.E.) so that the estimated inventory would be elevated by a factor of 2.6. For strata nearer ground zero, one sees from Table 4 for soil samples in the top 0-2.5 or 2.5-5.0 cm in stratum 4 (the highest activity stratum for the TTR sites) that the ratio can be expected to be closer to 15 or 20. Hence, the estimated inventories for these high-activity strata would be similarly elevated.

Turning to Question 3 as to whether Pu concentrations on sieved samples can somehow be corrected back to BM fraction or "field" conditions, the data in Table 8 is presented for consideration. These are profile samples from the TTR (a subset of those in Table 4) for which $^{239-240}$Pu concentrations were obtained on BM, <100 mesh and >100 mesh aliquots. After the 10-gram BM fraction was removed from the total BM sample, as much as possible of the remaining ball-milled soil was passed through a 100 mesh screen, and the weights of the total <100 and >100 mesh fractions recorded. A $^{239-240}$Pu concentration "corrected" to field conditions can be obtained by computing

$$g = b\left(\frac{a}{a+d}\right) + e\left(\frac{d}{a+d}\right) = c + f \quad , \qquad (1)$$

where the letters correspond to the labeled columns in Table 8; i.e., a = weight of <100 fraction, d = weight of >100 fraction, b = uncorrected Pu concentration in <100 fraction, e = uncorrected Pu concentration in the >100 fraction, c = Pu concentration of <100 mesh corrected to field conditions, and f = Pu concentration of >100 mesh corrected to field conditions. Hence, g is a "weighted" average, where a/(a+d) = proportion by weight of <100 mesh soil, and d/(a+d) = proportion

by weight of >100 mesh soil. One may think of ba/(a+d) and ed/(a+d) as the <100 and >100 mesh fraction Pu concentrations, respectively, that have been corrected back to field conditions by taking into account the proportion of <100 and >100 mesh soil in the total soil sample. [Note that a+d as reported to the authors are slightly less than the total BM fraction weight (98.0 $\pm$ 0.62 percent). The assumption is made here that no bias is introduced into the corrections by this fact. The total dry weight (before ball-milling) is given in Table 8].

The BM fraction $^{239-240}$Pu concentrations (Column h) are plotted against "total corrected" concentrations (Column g) in Fig. 18. The resulting estimated linear regression line may be compared with that in Fig. 17 where these same BM fraction data are plotted versus the uncorrected sieved Pu concentrations (Column b). That is, by correcting the <100 and >100 mesh Pu concentrations to field conditions, does one obtain concentrations equivalent or closer to those obtained on the BM fraction? If the BM and "total corrected" concentrations are indeed "equal," then the two nonparallel lines in Fig. 17 should become parallel and coincident in Fig. 18; i.e., one should find $\hat{\beta}_0 \simeq 0$ and $\beta_1 \simeq 1$ in Fig. 18. One sees that $\hat{\beta}_0$ does get closer to zero (-0.355 changes to -0.252) and $\hat{\beta}_1$ is closer to 1 (0.744 changes to 0.817), but $\hat{\beta}_0 = -0.252$ and $\hat{\beta}_1 = 0.817$ are still significantly different from 0 and 1, respectively. Hence, the "corrected" concentrations obtained using Equation (1) above did not, at least in this instance, fully correct to BM fraction results, although some improvement was noted. This raises the question as to whether differences between laboratories might be partly responsible for the lack of agreement between BM and sieved results in Fig. 9. The "corrections" applied above would not remove any such laboratory bias.

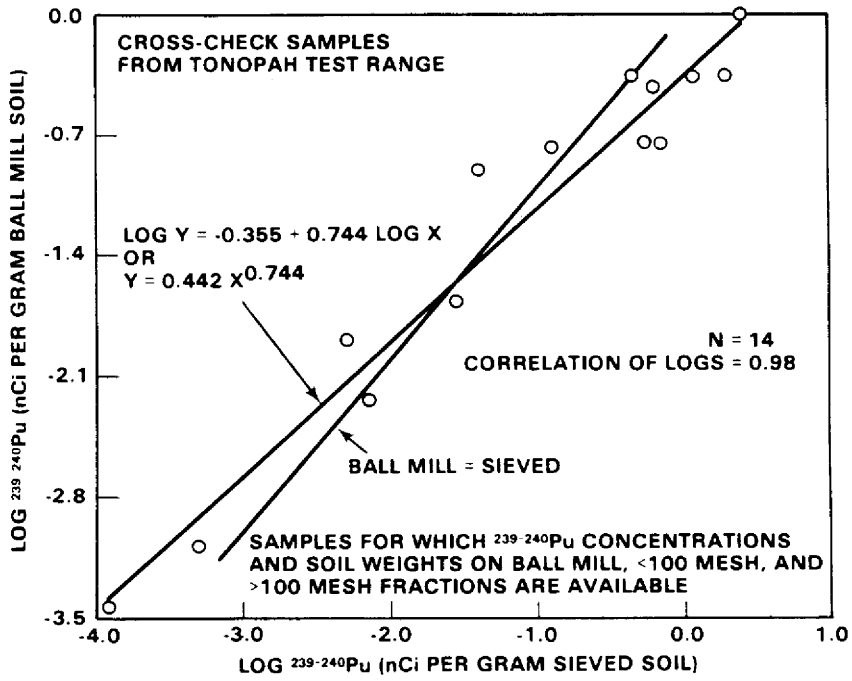

FIGURE 17 BALL MILL VERSUS SIEVED (<100 MESH) $^{239-240}Pu$ CONCENTRATIONS IN SOIL

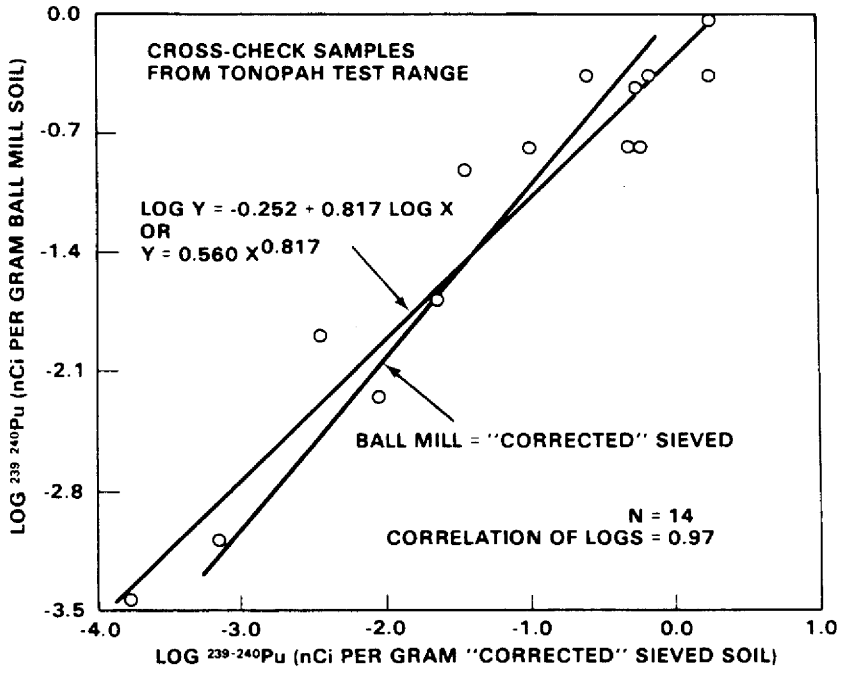

FIGURE 18 BALL MILL VERSUS "CORRECTED" SIEVED $^{239-240}Pu$ CONCENTRTIONS IN SOIL

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Support Activities

INFORMATION SUPPORT TO THE NEVADA APPLIED ECOLOGY GROUP

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ABSTRACT

The Nevada Applied Ecology Information Center (NAEIC) has initiated additional approaches to selecting and disseminating information to the Nevada Applied Ecology Group (NAEG). Attempts to keep the NAEG and its contractors informed on developments reported in literature include: (1) sending a monthly reference listing of new documents, (2) alerting researchers to incoming documents that match needs, and (3) conducting bimonthly or quarterly individualized updated searches from the computerized information file. Information from classified documents is being selected, cleared, and entered into the computerized file. The NAEIC is collaborating with the scientific staff of the Comparative Animal Research Laboratory, ORNL, on a computerized file of tabular data on plutonium in mammals.

DISCUSSION

The NAEIC has initiated additional approaches for selecting and disseminating information to the NAEG. The primary objective of the information center is to keep the NAEG and its contractors informed about literature in their fields. The Information Center now attempts to supplement this objective by sending a monthly reference listing of new documents (over 100/month) to NAEG contractors who have requested it. From the reference listing, contractors can request copies of documents of interest and the Information Center will

supply those documents not available to the researchers. This service will be limited to the NAEG and its contractors. When NAEG researchers notify the Information Center of their interests and research projects, they are automatically alerted to incoming documents that match their needs. Another new approach in use is to send NAEG researchers bimonthly or quarterly individualized updated searches from the computerized information file. For these procedures to be effective, feedback on their usefulness is necessary in each case in order to select subsequent specific applicable information.

Selecting and computerizing information within the scope of the NAEG project from the classified literature is a second aspect of the project. This classified literature reposes in the vault of the Technical Information Center in Oak Ridge. From the classified literature, bibliographic and descriptive information has been selected, cleared, and entered into the computerized file. This activity is a logical extension of a continuing project to locate and extract pertinent information from early literature. The information extracted from the classified documents is, however, necessarily scant. Some of the data contained within a document can be described by subject categories and key words, which have been cleared by a security officer, i.e., the radionuclide, the type of study, the methods, and the nonsensitive results will be noted.

The collaboration of the NAEG Information Center with the Comparative Animal Research Laboratory (CARL) in Oak Ridge is a recent project. A handbook of tabular data extracted by the CARL scientific staff on pluntonium in mammals will be published soon by CARL. It will be sent to interested NAEG contractors. The CARL data were designed to merge with the NAEG Information Center computerized information file and will be available for specific searches fitting NAEG needs.

Since the NAEIC began in January of 1972, the scope of the project has grown as needs and interests of the NAEG have expanded. The initial interest and continuing primary concern are plutonium in the environment, particularly that of the Nevada Test Site. Table 1 relates some of the scope changes to the publication dates of the bibliographies. Ecology of the Nevada Test Site was added before the first publication in September, 1972. Uranium was added in time to

*NUMBER OF REFERENCES BY SUBJECTS IN THE NAEG INFORMATION CENTER BIBLIOGRAPHIES

PUBLICATIONS	TOTAL REFERENCES	TOTAL SUBJECTS	PRE-1962 TOTAL SUBJECT/REFERENCES %	PLUTONIUM TOTAL SUBJECT/REFERENCES %	URANIUM TOTAL SUBJECT/REFERENCES %	OTHER TRANSURANICS TOTAL SUBJECT/REFERENCES %	NEVADA TEST SITE TOTAL SUBJECT/REFERENCES %
72-21 (1) SEPTEMBER, 1972	785	786	84/5%	621/79%	64/9%	46/6%	55/7%
73-21 (2) AUGUST, 1973	880	1091		758/86%	132/15%	60/7%	141/16%
74-21 (3) FEBRUARY, 1974	435	461	306/70%	242/56%	114/26%	6/1%	99/23%
74-21A (4) DECEMBER, 1974	528	705	157/10%	516/98%	84/16%	55/10%	50/9%
75-1 (5) JUNE, 1975	594	748		505/85%	101/17%	108/18%	34/6%
75-2 (6) JULY, 1975	505	755		500/99%	116/23	132/26%	7/1%

*EACH REFERENCE MAY BE INDEXED ON SEVERAL SUBJECTS

TABLE - 1

be included in the second publication in August, 1973. The third bibliography added much of the unclassified and declassified pre-1962 literature and other transuranic elements. Environmental aspects of technetium were added for the fourth bibliography. The fifth bibliography of about 500 references is in press; the sixth is in preparation.

Since the October NAEG progress report, 500 references have been added to the computerized information file to bring the total to about 4,000 searchable references. The level of funding, however, has restricted the extraction of information from documents in preparation for input, so that there is a constant backlog of approximately 500 documents. Priorities are set within this backlog in order to ensure that NAEG needs are met. Only those documents of immediate interest to NAEG and its contractors are chosen for entering into the computerized information file. Documents with low priority are on subjects such as waste management, routine monitoring, and environmental aspects of the nuclear fuel cycle (transportation and storage). When support levels permit, these documents will be included.

PLANNED ACTIVITIES

Several symposia on the actinide elements are scheduled this year and individual participants in these meetings will be contacted with a request to provide abstracts of their papers for dissemination.

Classified documents will continue to be searched for environmental information on uranium and the transuranics as a second priority. Pre-1974 literature on neptunium and the transplutonic elements will also be addded as permitted by obligations of staying current and user requests. New procedures to maximize the value of the NAEIC will be sought and evaluated.

ANALYTICAL DATA FOR NEVADA APPLIED ECOLOGY GROUP SAMPLES (OCTOBER, 1974-MAY, 1975)

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SUMMARY REPORT

Since the October, 1974, meeting of the Nevada Applied Ecology Group (NAEG), a number of projects have been implemented and completed by LFE Environmental. The principal function of the laboratory during the reporting period was to provide NAEG contractors with analytical data for samples collected in the field or prepared in the laboratory.

Approximately 2,300 data results for soil, vegetation, and mammals were sent to NAEG-NV. Approximately 500 pieces of data for soft tissue, bone, and rumen contents in large grazing mammals were also transmitted to NAEG-NV. Results from 1,000 data for filters, saltations, soils, and vegetation were also transmitted to NAEG-LLL. Computer programs were devised for reporting these data to each group.

All the above projects required analysis for a variety of nuclides, such as $Pu^{238,239}$, Am^{241}, Pb^{210}, Sr^{90}, Fe^{55}, total U, Is U, Ra^{226}, Th^{230}, Ce^{144}, and Cs^{137}. Analysis included radiochemical, instrumental, and mass spectrometer methods. Procedures for ashing, dissolution, and analysis of nuclides in filters, soils, vegetation, saltations, and small mammals were greatly improved through many hours of research and development work. Americium-243 tracer recovery, for example, was increased from an average 30 to 60%, in addition to a considerable improvement in the quality of the electroplate.

Two new NAEG projects were started and completed during the period. The first was the Large Animal Grazing Study which required the analyses of large cattle tissue and bone samples for Pu^{239}, Am^{241}, Sr^{90}, total U, and is U. Special procedures were developed for the ashing, dissolution, and analyses of these samples.

The second project involved the simulated radiochemical analysis of large cattle tissue and bone samples in the NAEG hot sample facility at NRDS, Jackass Flats, Nevada, adjacent to Nevada Test Site facilities. The purpose of building this facility was to have available a remote area where large mammal tissue and bone samples, containing high-level radioactivity transuranics, could be prepared for radiochemistry or instrument analysis. In the simulated analysis, tissue and bone samples were spiked with Co^{60} and ashed and/or dissolved with special procedures. Work areas were monitored after each step of the procedure to check for spread of contamination. Results showed no such occurrence. A report of the simulated test is being prepared.

A few minor projects such as preparation of vegetation solutions for shipment to other laboratories, analyses of calibration samples, preparation of dry and ash weight tables of vegetation and small mammals, consulting on the NAEG hot sample facility's construction and outfitting, a computer program for data output of the Grazing Mammal project, and installation of two new hoods and four new alpha spectrometer systems at LFE were implemented and in most cases completed during the period.

A paper describing the details of the new and improved ashing, dissolution, and analysis procedures is in preparation. Highlights of the paper include a new method of dissolving large tissue samples which bypasses the dry ashing step, a method for separating Am-Cm in any matrix using HDEHP in toluene and DDCP in 12$\underline{N}$ HNO_3, and a Dowex-50 cation column using EtOH-HCl elution.

REECO ACTIVITIES AND SAMPLE LOGISTICS IN SUPPORT OF THE NEVADA APPLIED ECOLOGY GROUP

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ABSTRACT

Logistical support activities by Reynolds Electrical & Engineering Co., Inc. (REECo), are discussed for the collection, preparation, and shipment of the Nevada Applied Ecology Group's (NAEG) soil, vegetation, small animal, and large animal samples. The samples were taken from the NAEG plutonium program intensive-study areas at the Nevada Test Site (NTS) and the Tonopah Test Range (TTR) during the period of September 1974 to March 1, 1975.

REECo activities in support of the NAEG high-level radioactivity sample facility at NRDS, NTS, are also discussed.

DATA SUMMARY

During the period September 1974 to March 1, 1975, the following operations were completed:

Three hundred twenty (320) soil samples were collected, 484 aliquots were prepared, and 463 aliquots were shipped to analysis laboratories.

From previous collections of 38 animals, 114 small animal tissue samples were prepared and shipped for analysis. One hundred forty-one large animal samples were shipped to Eberline Instrument Corporation for analysis and 196 to LFE Environmental Analysis Laboratories.

The total number of NAEG intensive-study area aliquots which have been prepared for radiological analysis as of March 1975 is soil aliquots, 2,662; vegetation aliquots, 1,214; small animal tissues, 327; and large animal samples, 337.

During March, REECo personnel supported a dry run for large animal sample preparation at the NAEG hot sample (high-level radioactivity) facility at NRDS, Jackass Flats. The dry run, performed under an NAEG contract to LFE, consisted of performing sample preparation on large animal tissue and bone samples. Cobalt-60 tracer was used in the dissolution and ashing process to check radioactivity handling and processing procedures. The two major objectives were to determine what preparation problems might develop under different laboratory and equipment conditions, and to determine health physics measures necessary to prevent radioactive contamination of personnel and equipment. As a result of the dry run, protocols are being developed for the preparation at the hot sample facility of large animal samples containing transuranics.

Considerable REECo effort has been expended on the NAEG data bank. The culmination of some of this effort is indicated by the NAEG intensive-study area data printout, distributed to NAEG element managers and investigators during this period.

FUTURE PLANS

Plans for REECo support activities for Fiscal Year 1976 include the following:

1. At least 12 trips to the TTR are anticipated for collection of small animals, soil, vegetation, and for resuspension studies.

2. On-NTS activity is expected to be of a larger magnitude than in the past. Primary support activity will be the sample preparation and/or shipment of about 2,000 large animal samples.

3. Support requirements for the collection, preparation, and shipment of approximately 1,800 soil, vegetation, and small animal samples are also anticipated.

4. REECo handling of NAEG data is currently limited to soil, vegetation, and small animal data. For the past several months, REECo has been in the process of recruiting an Environmental Analyst to input and proof NAEG Data Bank data. REECo handling of large animal sample data is currently pending approval of NV management.

5. Considerable REECo engineering field support is expected to be required to support proposed decontamination and treatment studies, and surveying and mapping for intensive study of nuclear sites, scheduled to begin during the last half of Fiscal Year 1976.

Miscellaneous

A WISE RESOLUTION OF THE PU-CONTAMINATED LANDS PROBLEM IN CERTAIN PARTS OF NTS MAY BE TO ISOLATE AND MAINTAIN THEM WITHOUT FURTHER DISTURBANCES: A POSITION PAPER

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Editor's note:

Although this paper was not part of the NAEG workshop discussions, the thoughts expressed by Dr. Rhoads result from his participation in NAEG activities and are included as his position statement.

Readers are also referred to the views expressed by Wallace and Romney, "Feasibility and Alternate Procedures for Decontamination and Post-Treatment Management of Pu-Contaminated Areas in Nevada," *in* Report NVO-153, 1975, *The Radioecology of Plutonium and Other Transuranics in Desert Environments* (White and Dunaway, Eds.).

INTRODUCTION

There are a number of areas of limited extent on, or adjacent to, the Nevada Test Site (NTS) which have been contaminated with the radioactive element plutonium. In most cases, these areas have resulted from tests of Pu-containing nuclear devices for safe storage and handling as an essential part of the national defense. In most cases, also, scattering of Pu was caused by chemical high

explosives, and are not due to fission reactions. Because of the toxicity of the scattered material, however, both from the standpoint of its chemical and radioactive properties, and because its long half-lives (^{238}Pu, 87.4 yrs; ^{239}Pu, 24,390 yrs; ^{240}Pu, 6,600 yrs), it is essential to prevent Pu from reaching man on both short- and long-term bases.

"Long-term" infers that methods essential to prevent Pu from reaching man should be effective from 10's of years to time periods bordering the geological time scale. As to which preventative methods are chosen to achieve this end, the final decision must be predicated on both efficiency and cost, as well as such environmental parameters as disruption or destruction of the biota in affected areas and the potential recovery of the areas.

Plutonium's characteristics make it particularly important to exclude the element from man's food chain, or drinking water, but most importantly to prevent inhalation of plutonium particulates by man.

From time to time, a number of theoretical methods have been proposed for preventing plutonium in contaminated NTS areas from reaching man or his domestic animals, or animals which contribute directly or indirectly to his food supply. Among these proposed methods are the following:

1. Scraping and removal of the soil surface from contaminated areas. This could be accomplished at relatively large expense, although the methods would likely not be efficient in areas which have very shallow soils with intermittent rock outcrops. Removed materials would be stored, presumably, in locations not subject to wind, water, or human or animal exposures. The stored areas themselves would, of necessity, become exclusion areas with problems paralleling those of the present with exception as to localization.

2. A method has been proposed of producing, in situ, by thermal and mechanical means, relatively insoluble iron-rich silica compounds like those which appear as natural end products of soil and rock weathering in certain parts of the world. This would require the Pu-contaminated surface layers to be picked up, fired to temperatures sufficient to fuse iron-silica mixes which would carry the Pu into inert, chemically inactive compounds, thereby effectively removing Pu from wind, weathering, and biotic processes. This would likely be expensive, disruption of the environment would be complete, and recovery would be uncertain.

3. Leaving the Pu on the soil and rendering the soil surface immobile by surface agglomeration and fixation or by covering with a macadam or concrete surface, also intended to remove Pu from biotic processes, has been proposed.

All these proposals have certain characteristics in common. Their objectives are to remove Pu from contact with biotic cycles, and from the further widespread distribution by wind and water. Most of the proposed methods would require extensive disturbance of the areas. Some would also present the hazard of contributing to further distribution of the element, even if on a short term, and some would leave earth surfaces denuded of native vegetation and subject to severe erosional processes on whatever Pu may have been left subsequent to the operation. All proposals are based on the hypothesis that Pu in the field in its present state under present conditions is a hazard which should be eliminated or at least properly mitigated.

There is a concomitant with this position paper, a solicitation of proposals for pilot projects to undertake cleanup of Pu-contaminated areas and other areas contaminated with other nuclear debris. No doubt there will be other methods proposed or combinations of the above-suggested methods. This paper is a response to that solicitation for proposals, but supports, instead, the hypotheses that

isolation, nonmanipulation, and protective nondisturbance may be an adequate treatment of Pu-contaminated areas. This hypothetical treatment will, however, be restricted to those contaminated areas of NTS which are within closed drainage basins as, for example, both Yucca and Frenchman basins. Some arguments may be applicable, however, to other nearby areas with similar adaphic and environmental conditions.

The position of this paper is that the present hazards of Pu in the NTS environment are minor and are diminishing, that protecting these areas from further disturbance will accelerate the rate of diminishing hazards. That is, the bioenvironmental processes will, in the long term, contribute to the continued decrease in the availability of Pu to the biotic cycles. In the very long term, a combination of environmental, geochemical, and geological processes will reduce the hazard in insignificance.

Wallace and Romney (1975) have reviewed the literature and decontamination procedures as a basis for a report on the feasibility of decontamination of Pu-contaminated areas in Nevada. They have also suggested the alternative that the areas not be further disturbed, and have provided support for that alternative. In order to avoid duplication of their material, this paper attempts to provide further support not given by Wallace and Romney and to provide further discussion of some points not detailed in their report. The Wallace and Romney report should therefore be used as a supplement to information provided herein.

BIOENVIRONMENTAL PARAMETERS AFFECTING PU AVAILABILITY TO FOOD CHAINS

Movement of Pu within ecosystems is determined by many factors. Because of the distribution of Pu on or near the soil surfaces at NTS, and because it is thought to be in the metallic or oxide state, its movement is determined by wind and weather within the

conditions and boundaries determined by the topography and geology of the area. Plant uptake as a basic for transfer to food chains (to be discussed later) is no longer considered significant. For these reasons, it is useful to examine not only certain aspects of the geology of the area but also the topography, hydrology, and soils of the area.

Topography, Hydrology, and Soils

Among those areas of interest at NTS, because of Pu contamination, are Yucca, Frenchman, and Groom valleys, which are all closed basins as far as surface drainage is concerned. That is, rainfall into any of these basins and its erosive products, insofar as water per se is concerned, remain there and do not flow away from the area into streams which eventually reach the Pacific as generally occurs outside such basins. In addition, because of the low rainfall in the region, soil surfaces outside the actual drainage patterns do not receive sufficient water to reach the deep subsurface drainage patterns. Moreover, movement in subsurface aquifers is very slow, 7 to 730 ft per yr (Anonymous, 1973), beneath Yucca Flat. Although water moves faster in Frenchman's aquifers to reach downward to the Paleozoic aquifer, it apparently must pass through Tertiary tuffs where the rate of movement is 0.2 ft per yr or less. Paleozoic aquifer depths vary from a few hundred feet to a few thousand feet, depending on the depth of the alluvial materials which cover the basins' floors; i.e., both these valleys, and presumably Groom also, are filled from a few hundred feet to a few thousand feet with alluvial material eroded from the surround ing mountains, it is thought, during earlier pluvial periods correlated with times of extensive worldwide glaciers (Fernald *et al.*, 1968). Near the mountains, these pluvial fans are dissected by more recent washes.

Soils in these areas are often ill-defined as to horizons and frequently underlain by caliche hardpans. Those soils derived from limestone outcrops on the surrounding mountains are often strongly calcareous as compared to those derived from volcanic materials.

As we will attempt to demonstrate, many of these factors are likely to be important in restricting the distribution of Pu and slowing its movement into food chains.

PRESENT CONDITIONS IN PU-CONTAMINATED AREAS

There is at present a large amount of information describing the extent of contaminated areas, concentrations of Pu present, its distribution within the soil profile, and the nature of the Pu particles themselves. There have been extensive studies concerning the movement of Pu by wind (Phelps and Anspaugh, 1975; Shinn and Anspaugh, 1975), and although there have been no formal studies on the subject, there is some indication of movement of Pu by surface water. For example, radiological surveys of Area 11 have shown a finger of Pu activity toward the south, beginning to follow a drainage channel from the Pu-valley site.

In general, however, these studies have shown that Pu is not evenly distributed in and on the soils but is likely to be concentrated about the bases of clumps of shrubs, a distribution attributed to movement by wind and entrapment where there is a reduced wind velocity under and around shrubs. It should be pointed out that these conditions have developed over a period of 10 to 20 yrs, depending on the time of initial Pu distribution; these conditions are therefore indicative of what is likely to continue to occur in the future.

Tamura (1975) has noted that Pu in the NTS area is probably in the oxide form, that it is associated with the silt fraction of the soils, and that its solubility was very low, less than 0.5%. Its occurrence in the top five cm was shown by Essington *et al.* (1975). Tamura also noted that the fraction of Pu moved by wind might be tightly bonded with silicate particles rather than as separate Pu oxide particles.

In studies on windborne material, Shinn and Anspaugh (1975) have shown that dust flux is sensitive to wind speed, likely due to the avalanching effect from saltation. Saltation is a process whereby large particles moving at the ground surface sandblast the surface, thereby ejecting smaller particles which may be suspended in the air, even though the saltating particles rise no more than one meter above the ground. They compared a highly erodible agricultural soil surface in Plains, Texas, to the low saltation, less erodible surface at Frenchman Flat and found that the airborne dust concentration was proportional to an exponential power of wind speed greater than six. In Nevada conditions, it was closer to a power of two. This suggests that with the same wind speed in both disturbed and relatively undisturbed areas, the airborne material should be decreased by a factor of about 10^3.

In addition, Phelps and Anspaugh (1975) have developed a time-dependent resuspension factor to describe long-term contaminated areas. They showed that the resuspension rate after 20 yrs was much lower than initially. In a practical sense, this infers that the worst of resuspension by wind at all the Pu-comtaminated areas of NTS are long past. They cite values for initial resuspension factors at 10^{-4} m^{-1}; after 17 yrs, the resuspension factor should approximate 10^{-9} m^{-1}, a decrease of 10^{-5}.

Romney *et al.* (1975) have investigated the surface contamination of vegetation in Pu-contaminated areas of interest to this study. The ratios of the amounts of Pu on vegetation that is primarily on the surface of vegetation, compared to that with soil beneath, tended to be greater on the peripheries of the contaminated areas where the particles of Pu initially deposited by the tests were smaller sizes. Inventory estimates indicated that standing vegetation provides a rather insignificant portion of the total contaminant remaining in the fallout areas.

It should be noted in passing that surficial contamination of the shrub species could occur from having soil carried to leaves and stems by wind or some other physical process.

To date, there does not appear to be any data which clearly define the amount of Pu taken up by plants by their roots, largely because of the difficulties in delineating between that Pu on the surface and that within the leaves and stems. Some laboratory work by Romney *et al.* not yet published (1975) indicated factors of 10^{-3} to 10^{-4} between concentraions of Pu in soils compared to that within plants grown on soils taken from NTS to the laboratory where surface contamiation could be prevented with confidence. To what extent these factors represent the field cannot yet be evaluated, although general experience with such experiments indicates laboratory values are generally higher than field values.

That leaf surface materials deposited from the environment may be absorbed within plant tissue has been recognized for some time. Cataldo *et al.* (1975) have provided some information on Pu translocation to other tissue subsequent to leaf contamination. In the absence of a solution vector, Pu was relatively immobile with respect to translocation to root and seed tissues. Subsequent to a simulated rainfall of 0.4 cm in 7 mins, they found translocation dependent on period of the life cycle in beans for aged Pu oxide. The values ranged from less than 8.6×10^{-6} to 1.8×10^{-4}. There does not appear to be any way to evaluate these numbers in terms of the more heavily cutinized tissues of NTS, but it might be supposed that for desert vegetation, the values might be even lower. Even if these values are accepted and extrapolated to the precipitation values of 10 cm for NTS, and the ratios of soil Pu to plant Pu are considered, the final values would probably not be detectable. In any event, for NTS vegetation values presented by Romney *et al.* (1975) would include Pu within plants as well as on the surface of plants and these values ranged from a high of 1.2 nCi/g dry tissue down to 0.012 nCi/g dry tissue.

These values thus represent the ranges in amounts that grazing animals would have entering their digestive systems from shrubs to which must be added amounts from direct ingestion of contaminated soils and from grasses also in the area. Smith *et al.*

(1975) have studied the uptake and tissue distribution of Pu by cattle. On one cow which was confined solely to the contaminated area for 177 days, the total intake of Pu was about 100 U Ci/day. They found no lesions not encountered in cattle on similar noncontaminated ranges. The skeletal tissue of the 177-day cow contained 24 times the background amounts in cattle elsewhere, 9.5 times the amounts in muscle tissues of cattle elsewhere, and in the liver, 260 times the background amounts. The total content was 238 pCi in a liver from this animal weighing a little more than 6 kg.

In brief, conditions around NTS can be summed up as follows:

In several areas within NTS, there are relatively large amounts of Pu contaminating the soils. This condition has existed for up to 20 yrs, during which time the Pu, thought to be primarily in the oxide form, has migrated to a few centimeters depth in the soils on NTS proper and to greater depths in other test areas. Within soils Pu appears to be associated with the silt fraction, with a low solubility. It has been moved by wind and weather in a redistribution pattern which now shows most of the Pu associated with the soils adjacent to, and under, shrubs and shrub clumps. The amounts of Pu moved by winds have been reduced sharply 38during the time since the tests. Wind movement and redistribution appear to result from saltation of larger soils particles which may not be carried to an altitude more than a meter above the surface, and which, when they strike the surface, dislodge smaller Pu particulates by an avalanching effect; and these smaller Pu particles may be more readily airborne. With distance, however, Pu-air concentrations decrease very rapidly. Plants growing in the Pu-contaminated areas have Pu contaminating their surfaces and likely have small amounts within their tissues from uptake from their roots and possibly from surface absorption through aboveground tissue. Cattle, restricted in their food to grazing these areas, have Pu in their tissues within a few 10's of times background values except in liver, where the concentration reached 260 times background.

POSSIBLE DOSE TO MAN FROM SUCH AREAS

Martin and Bloom (1975) have utilized this kind of information to produce a theoretical model to predict how much Pu might possibly reach man from such an area. Their model was based on several assumptions which, although useful for such projects, have otherwise little in common with reality. For example, their hypothetical subject for dose calculation purposes was a man who lived for 50 yrs within a major contaminated area of NTS where he received most of his sustenance in the form of meat and vegetables grown within the same contaminated area. Even with these unrealistic assumptions, a relatively samll part of the NTS area which was the subject of the model contained sufficient Pu to provide a dose to critical human tissue above the maximum permissible dose for the public at large, based on ICRP (1959) standards.

CONCLUSIONS AND RECOMMENDATIONS

The Martin Bloom model is to date the only overall evaluation of possible hazards from the Pu-contaminated areas of NTS. Its utilization of unrealistic parameters is recognized by the builders, and these parameters were used as a worst conceivable case. Even if one assumes these worst possible cases as being a serious hazard, it would appear to be useful, before a cleanup of these areas occurs, for the parameters to be examined more closely to find whether there may be ways of modification short of removal or immobilization of soil surfaces. The alternative to active cleanup or soil immobilization is to do nothing. What follows then is a defense of the proposal to do nothing other than exclude as much physical activity in Pu-contaminated areas as is possible.

Let us then examine that action and the advantages and disadvantages of such action.

The first and most obvious action appears to be to exclude men from the contaminated areas and second, to exclude animals which contribute to his food supply directly. Since the area is not in cultivation, a recommendation for some positive action is not needed, and a recommendation to maintain it in its present uncultivated state is the equivalent of no action. Exclusion of men and grazing animals will provide for not only reducing the exposure of men and prevent a Pu contribution to his food supply, but will undoubtedly also decrease the amount of Pu carried with surface materials into the atmosphere. This will occur because there will be a reduction in the amounts of smaller Pu-bearing particles carried into the atmosphere from this cause. The small particle disturbance contribution to atmospheric movement would undoubtedly also be reduced.

It has to be recognized that some provisions must be made to carry out this exclusion for a very long time if the half-life of Pu isotopes are taken into consideration. It is difficult to conceive that this is a serious problem in light of present conditions. NTS is an arid region with little potential for food production above the minimal need for a few families, and the possibility of population pressures from our culture seem remote as long as large-scale irrigation is not feasible. Moreover, we appear to be at a point of decreasing population pressures due to a decreasing birthrate. Even with population pressures, the areas required would scarcely be significant from overall productivity. If the area continues into the future to be considered as a grazing area, which seems its greatest potential at present, there may be some positive benefits to exclude plots from grazing whether or not they are contaminated because these areas may serve as seed production and distribution centers. Overgrazing is at present a common problem in much of the Western United States, particularly in the more arid regions. Exclusion areas might in fact serve also as sites for maintaining native species of animals as well, principally, small mammals and birds. Wild horses and burros should be excluded because of the physical disturbance caused by these animals. Antelope and deer might be allowed access if the surrounding area is not overgrazed and the

deer and antelope are protected against hunting. These are attractive possibilities under present drives for preservation of native species.

Surface waters in the area of interest to this proposal all empty into closed basins as has been pointed out previously, and these receiving points for runoff generally are close; that is, within a few to tens of kms. In addition, the rates of movement of soil particles and Pu particles, possibly because of the greater density of the latter, will undoubtedly be different. It seems reasonable that Pu will likely move much slower than clay fractions, and possibly less rapidly than the silt fractions. Movements into the dry lakes (the playas), if this occurs, leave some unaswered questions, but the movement will undoubtedly be very slow and dilutions of the Pu contaminant will likely bring it nearer to worldwide fallout concentrations. However, the characterstics of rainfall in this region of NTS, which often occurs as intense, local, and short-lived storms, provide some further uncertainties.

There appear to be natural forces at work which will further reduce Pu movement. Pu, on the soil surfaces, will undoubtedly continue to reach greater depths in the soils with weathering, primarily wetting and freeze-thawing. As pointed out by Essington *et al.* (1975), this will continue to reduce the quantities of material available for wind distribution and, in fact, may be the primary factor in the reduction of airborne material noted by Phelps and Anspaugh (1975), previously mentioned.

Within the soils, if Pu is in the oxide or hydroxide, as suggested by Tamura (1975), then there is a possibility of chemical inactivation over intermediate time periods as suggested by Wallace and Romney (1975), a process which may further reduce the amounts of Pu for wind dispersion and plant and animal uptake.

Much of the NTS area is underlain by caliche hardpans, which appear at the depth to which precipitation may reach, the result of carrying small amounts of calcium salts in solution downward, salt precipitation by water evaporation into the soil atmosphere, and then loss to the open atmosphere above. Wallace and Romney have pointed out the possibility that Pu may also be taken into this cycle, which is, of course, an efficient route for removing an element from the mineral cycles of vegetation occurring there.

The possibility that Pu may reach the subsurface aquifers on a time scale short of geological time appears very remote, or if it did, that that water would be moved to a locality where drinking or irrigation occurs is also remote.

In the very long terms that approach short geological time periods, there appear to be other forces at work which may reduce Pu availability to bioenvironmental cycles. This is the general geological process known as orogenic isostacy. By this process, mountains rise, are eroded into the valleys, filling the valleys at approximately the same rates as the mountains rise. As a result, present valley surfaces will eventually be covered by weathered materials from the surrounding mountains and, with the covering, the Pu-contaminated surface will also be covered. The possibility is only of interest because of the very long half-life of Pu, which is itself approaching geological time.

Finally, there is the consideration of cost. Fencing, which may or may not be needed, is certainly a very small expenditure relative to most of the methods considered at present. In addition, large amounts of soils will need to be moved and places will have to be found to deposit it at further expense. There will be losses in transport, equipment will be contaminated, and there will still be a potential problem in preventing redistribution even after the soils are at a new location. In addition to the fiscal costs, there may be other costs. Wallace and Romney (1975) have pointed out some of the possibilities of long-term damage to ecosystems and those arguments will not be repeated here.

In conclusion, we maintain there is sufficient information to warrant the decision to leave the Pu-contaminated areas discussed in this paper as exclusion areas. There are a few uncertainties in maintaining exclusion areas among which appears to be the possibility of movement of Pu into the playas, even though the dilution factors may negate the hazard in this area. There may be some positive advantages to these exclusion areas, and the possibility of decreasing Pu availability to food chains or to other human exposures with time making this alternative well worth considering.

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