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CHARACTERIZATION OF PLUTONIUM IN SURFACE SOILS FROM AREA 13 OF THE NEVADA TEST SITE*,**

Tsuneo Tamura

Oak Ridge National Laboratory
Environmental Sciences Division
Oak Ridge, Tennessee 37830

ABSTRACT

Total plutonium was determined in nine surface soil samples (0 - 5 cm) from area 13 in the Nevada Test Site (NTS). Particle size segregation was performed, and each particle size fraction of seven samples were analyzed for plutonium. The coarse silt fraction (53 - 20 μM) contained the highest percentage of plutonium in the soil (about 65%). Evidence of erosional translocation of plutonium was observed in one of the samples, and corroborative evidence was noted in describing the soil type.

Tests with 8 M nitric acid showed that about 13% of the plutonium was leached from the NTS sample as compared to about 70% in the leachate from sediments at Oak Ridge. In 0.1 M citric acid, about 1% of the plutonium was extracted from an NTS sample as compared to 25% from Oak Ridge samples. Implications of these results on the transport of plutonium are discussed.

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INTRODUCTION

Several areas in the Nevada Test Site (NTS) have been used for testing accidental detonation of subcritical atomic devices. These high explosive detonations without fission dispersed plutonium over the immediate landscape. One of these areas referred to as area 13 is being utilized as an ecological study area. An outer fence encloses approximately 1000 acres in area 13; and an inner fence, enclosing an area of about 250 acres, surrounds the most highly contaminated area.

The soil at the site has served as the primary repository of the dispersed plutonium. Since the initial deposition, the area has remained relatively undisturbed for about 17 years. The area is covered with desert shrubs at 5 - 10% plant density; the rainfall is approximately 4 - 8 inches per year. Experimental studies are being conducted in the area on the behavior of the plutonium, including resuspension by wind action, uptake by the desert vegetation, and concentration in mammals living and grazing in the area. To better understand plutonium translocation by these processes, information on the amount, distribution, and forms of the soil-deposited plutonium is necessary.

This progress report covers the results of studies of nine surface soil samples taken from a north-south transect in area 13. Results of similar studies of several sediment samples contaminated with plutonium in the waste disposal area of Oak Ridge National Laboratory (ORNL) are also presented for comparative purposes.

MATERIALS AND METHODS

The location where nine surface samples (0 - 5 cm) were taken initially in area 13 is shown in Fig. 1. The samples were taken by members of Reynolds Electrical and Engineering Company (REECO) as part of a coordinated sampling program of the Nevada Applied Ecology Group (NAEG). These samples differed from those supplied to other investigators in that they were not ball-mill ground. This allowed determination of particle size distribution. Later, three additional samples were taken at intermediate points between sampling point 6 and 7 (labeled 6A, 6B, and 6C). Analyses of these samples have not been completed at this time.

Ten-gram aliquots were used to determine the total plutonium in these samples. The extraction technique used is the HASL-LASL technique described elsewhere (Tamura, 1974). Approximately 75 grams of the sample were used to make size segregation of the soil. In order to minimize disturbance of the possible bonding association of plutonium with soil particles as it exists in the field, size segregation was made only with water and gentle trituration with a rubber policeman during screening. Sizes below 53 μM* were segregated by centrifugation. The fractions recovered were dried in a 40°C oven; the clay fraction, however, was obtained by freeze drying.

In addition to obtaining the total plutonium of unsegregated soils as described above, selected samples were extracted with 8 M HNO_3 for 1 hour at room temperature. Eight molar nitric acid was used since this

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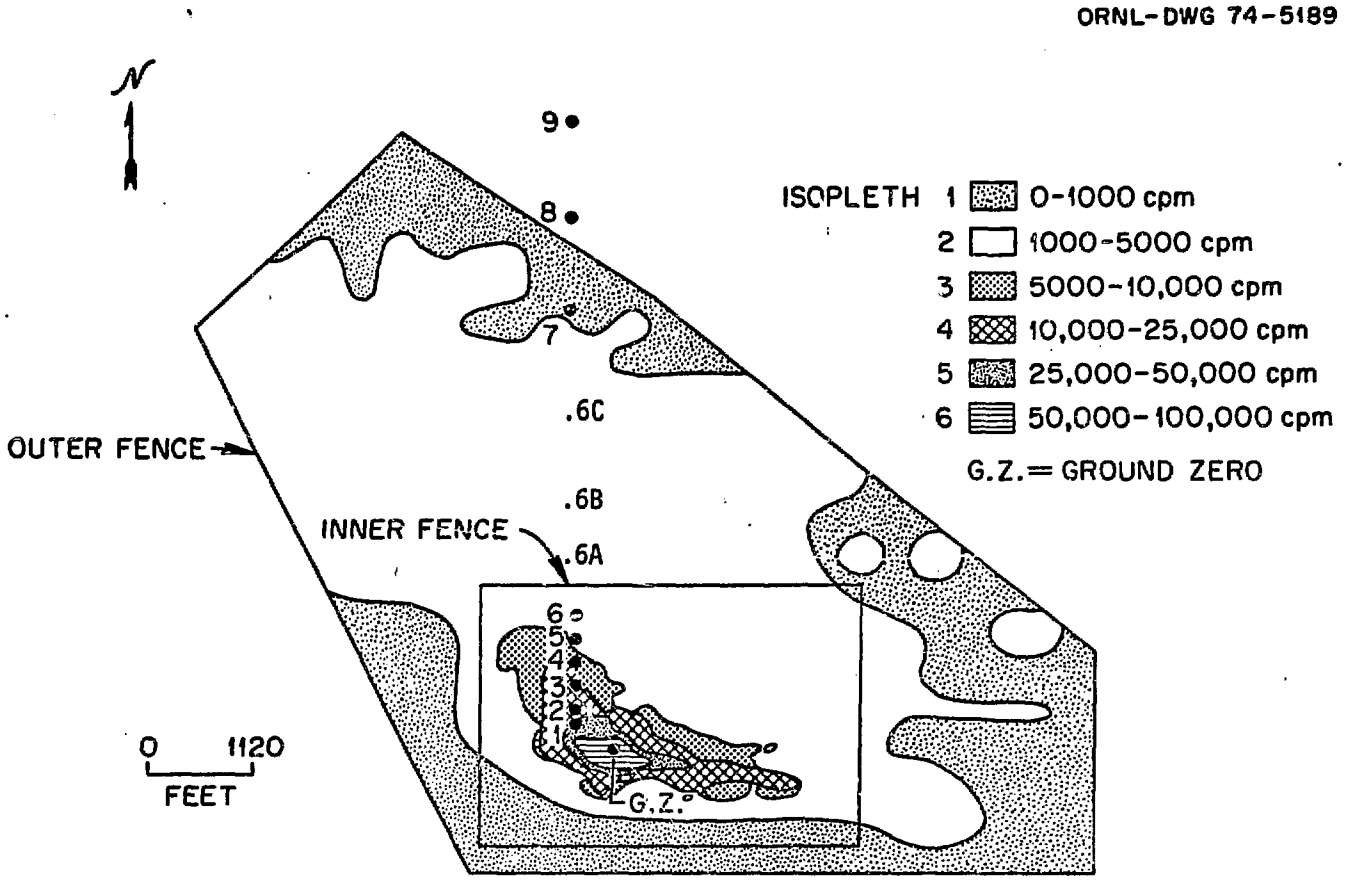
*For the sake of simplicity here, particle size in micrometer (μM) refers to mass median diameter of particles in a given soil fraction.

Fig. 1. Activity Isopleth and Sample Location in Area 13. Later samplings were made between sample points 6 and 7, marked 6A, 6B, and 6C. Isopleth map is taken from Gilbert and Eberhardt (1974).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N
9
8
7
.6C
.6B
.6A
OUTER FENCE
INNER FENCE
6
5
4
3
2
1
G.Z.
0 1120
FEET
ISOPLETH 1 0-1000 cpm
2 1000-5000 cpm
3 5000-10,000 cpm
4 10,000-25,000 cpm
5 25,000-50,000 cpm
6 50,000-100,000 cpm
G.Z. = GROUND ZERO

extraction is similar to that developed by REECO (1972), thus permitting the subsequent determination of plutonium without further analytical development. The technique differs only in that REECO recommends 4 hours of extraction at 90 - 95°C.

Several samples were also extracted with 0.1 M citric acid for 30 minutes at room temperature. This reagent was selected since the more soluble forms of plutonium are extractable in citric acid. Subsequent to extraction in citric acid, the solution was acidified to 1 to 4 M with nitric acid. The plutonium was extracted in tertiary amine nitrate, stripped in perchloric acid, and reextracted and counted in a scintillator solution (PBBO-naphthalene-HDEHP in toluene) using a high resolution liquid scintillation detector. The extraction steps following acidification to 1 to 4 M HNO_3 were developed by McDowell et al. (1974). Spiking tests with known plutonium standards verified that acidification of the citric acid with nitric acid enabled the tertiary amine nitrate to extract soluble plutonium from the citric acid media.

In addition to the NTS samples, several sediment samples were obtained from a former impoundage on White Oak Creek at Oak Ridge National Laboratory (ORNL). The results reported herein are from sediment samples taken at 0- to 12.4-cm depth.

RESULTS AND DISCUSSION

Total Plutonium and Plutonium According to Particle Size Distribution

Prior to segregating the soils, ten grams of each sample were analyzed for total plutonium. After segregating the soil sizes in seven

of the nine samples, each fraction was analyzed for total plutonium. Particles greater than 2000 μM were not analyzed and in calculating the plutonium concentration in soil, this fraction was assumed to contain no plutonium. The total plutonium in the unsegregated and segregated soil was averaged. The mean value for each sample is shown in Table 1. Since aliquots of the same original sample were analyzed by Los Alamos Scientific Laboratory and the LFE corporation, their results are also included in Table 1*.

Plutonium levels are seen to decrease as the distance from G.Z. increases (Table 1). Furthermore, the plutonium levels are consistent with the Fidler** readings taken in the field (see Fig. 1). However, variability in analytical results from the different laboratories is large; the variability is ascribed to the particle size of plutonium in samples. This variability as related to the particle size is discussed more fully later.

The particle size analyses of the nine samples are given in Table 2. Normally, particle size analysis in soils is reported for particles less than 2000 μM; however, since plutonium analyses by other participants in the program is reported for soil, particle size percentages are reported on the entire sample for consistency of reporting. The size analyses show that the soils are high in sands (2000 - 53 μM) and low in clay (< 2 μM). The highest content of sands, i.e., approximately 87%, is

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*Data taken from letter dated November 13, 1972, from E. B. Fowler, LASL, to R. O. Gilbert, Battelle--Pacific Northwest Laboratory.

**The Fidler is a portable field instrument which measures the ^{241}Am gamma radiation which is correlated with plutonium content.

Table 1. Plutonium concentration in nine surface soil samples from north-south transect in area 13. Activity of soil and standard deviation of the mean.

Sample No.	Grid Coordinate*	Distance from Ground Zero	Plutonium Activity (dpm/g)		
			ORNL	LASL	LFE
1	936400 N	500	7267 ± 4.6%	6416 ± 7.5%	6720
2	936550 N	602	2666 ± 12.0%	1067 ± 15%	2050
3	936800 N	806	915 ± 14.8%	-	787
4	937050 N	1031	480 ± 23.1%	213 ± 39%	866
5	937300 N	1265	334 ± 5.7%	396 ± 15%	705
6	937550 N	1504	210 ± 29.0%	163 ± 31%	254
7	940800 N	4718	22 ± 4.5%	41 ± 24%	25
8	941800 N	5715	20	11 ± 21%	30
9	942800 N	6713	18	13 ± 19%	16

*Listed coordinate is north-south; east-west coordinate is 721000 E. Coordinates for Ground Zero (G.Z.) are 936098.8 N and 721402.9 E.

**ORNL refers to analysis by Oak Ridge National Laboratory (author); LASL refers to Los Alamos Scientific Laboratory; and LFE refers to LFE Corporation in Richmond, CA.

Table 2. Particle size distribution of nine surface soil samples from area 13. Results expressed in percentage by weight.

Size Range (μM)	Sample No.								
	1	2	3	4	5	6	7	8	9
>2000	1.60	6.80	10.35	7.04	10.08	8.50	3.51	3.55	3.47
2000-840	3.69	7.64	8.17	5.81	6.99	7.41	4.66	6.14	4.25
840-250	26.41	26.86	26.65	22.51	23.12	15.48	32.34	33.31	24.04
250-125	27.37	22.95	20.01	22.05	15.78	17.01	29.92	29.04	31.42
125- 53	22.99	17.95	19.62	24.23	16.69	20.67	20.74	18.44	18.82
53- 20	7.67	7.02	6.07	8.67	8.73	10.29	4.05	5.89	4.39
20- 5	3.92	4.14	3.51	3.71	9.52	10.39	1.37	1.65	5.70
5- 2	2.17	2.44	2.14	2.64	5.05	6.26	0.65	0.82	4.02
<2	3.47	3.10	1.89	3.28	3.34	4.53	2.72	1.30	3.90
Total	99.29	98.90	98.41	99.94	99.30	100.54	99.96	100.14	100.01

in samples 7 and 8; the highest clay content (4.5%) is in sample 6. Sample 6 also contains the lowest sand content (53%).

The concentrations of plutonium in each of the particle size ranges less than 2000 μM are presented in Table 3. The alpha activity expressed as dpm/g in each size fraction is in row A. Because of the low concentration of plutonium in samples 8 and 9 and the similarity in plutonium distribution, these size fractions were not analyzed. The fraction with the highest level of plutonium is in the 20 - 5 μM size of sample 1; however the highest contributor (rows B and C) to total soil plutonium is the 53 - 20 μM size (coarse silt). The contribution to total soil plutonium takes into account the weight distribution of each fraction given in Table 2. Although plutonium concentration in the medium silt fractions (20 - 5 μM) of samples 1 and 7 are higher than in the coarse silt, the lower weight of these fractions decreases their contribution to total soil plutonium.

An interesting distribution of plutonium is seen in sample 6. In all other samples the clay fraction contributed less than 2% to total soil plutonium; in sample 6, the clay fraction contributed 9%. Sample 6 was also the highest in clay content (Table 2). An inspection of the soil survey map of area 13 (Leavitt, 1974) reveals the possible cause of this plutonium distribution (Fig. 2). Note that sample 6 is taken in an area where the soil type is labeled 405. According to the soil description, soil type 405 is similar to 401 except for a heavier texture and evidence of wind and water erosion. If erosion had removed the coarser materials and thereby enriched the clay content in the sample, the plutonium in the clay fraction would be expected to be between 48

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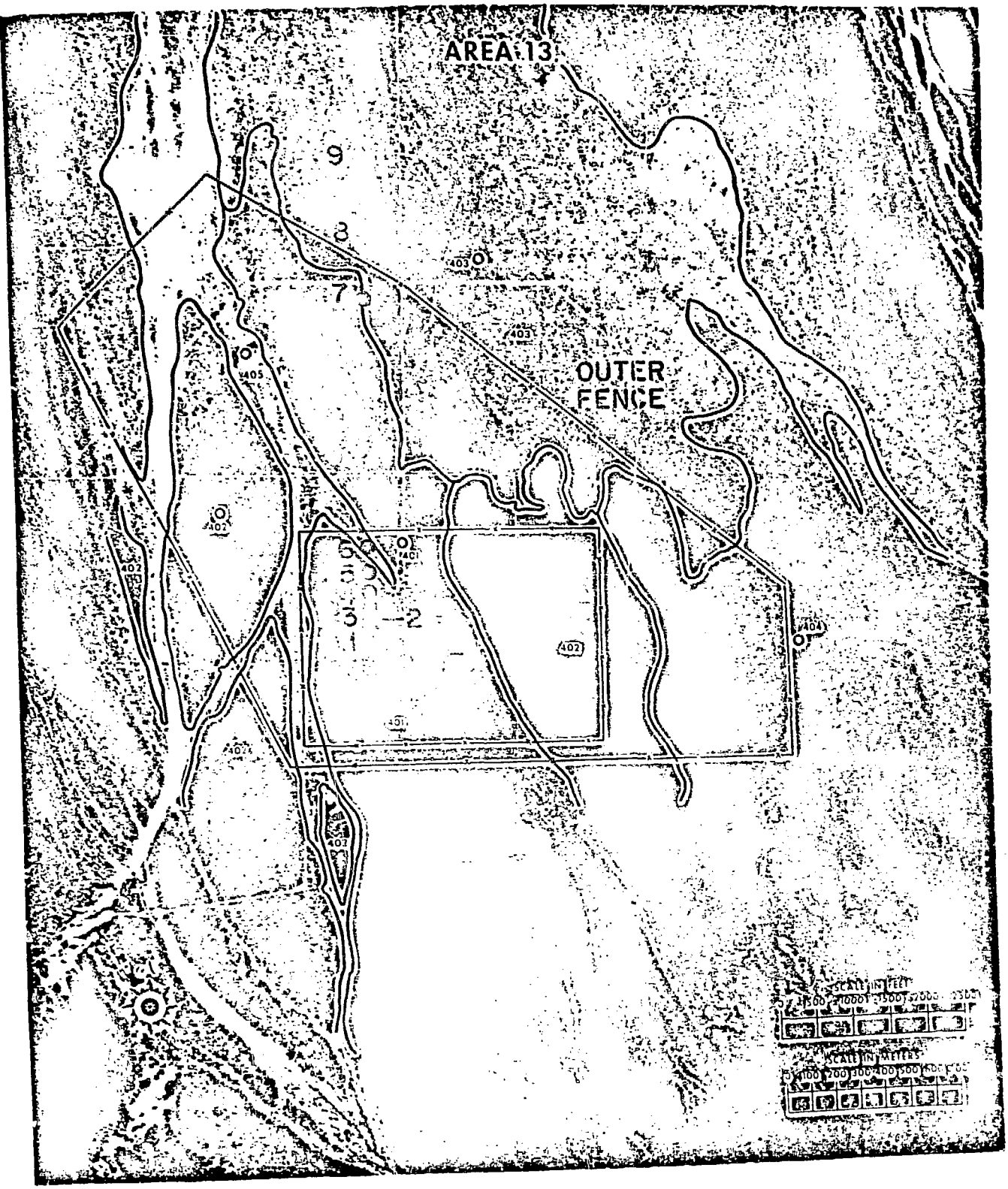

activity.), the activity of a pure 8 μM spherical $^{239}PuO_2$ particle would approximate 360 dpm. The value of 328 dpm/g (Table 3) was based on a 2.0-gram sample; hence the measured activity was 656 dpm/2 grams. This activity level would suggest that 1.8 particles were present. If, in taking a 2-gram sample the aliquot contained three $^{239}PuO_2$ particles, the activity contributed by the fraction would be 24 dpm instead of 13 dpm, and the total soil plutonium would be 34 dpm/g. On the other hand, if the sample had one particle, the activity contributed by the fraction would be 8 dpm and the total soil plutonium would be 18 dpm/g. In taking a 10-gram sample of unsegregated soil for analysis, the 53- to 20-μM fraction would make up 0.4 gram. In only 0.4 gram there is a likelihood that no particles of plutonium would be present; in this case, assuming other size fraction contributions remain constant, the total soil plutonium could be as low as 10 dpm/g. At soil plutonium levels of this magnitude and with only a few particles to work with, there is a need for analyzing larger samples. For example, in analyzing 2 grams of the 53- to 20-μM fraction, this was equivalent to analyzing 50 grams of unsegregated soil. Furthermore, this helps to explain the wide range of values reported by the three laboratories.

Selected Leaching Tests of Plutonium

Earlier work with NTS soil samples suggested that most of the plutonium extracted is plutonium oxide of high specific gravity (Tamura, 1974). During exploratory field work at ORNL plutonium was extracted from sediment samples taken in an old former lake bed. In contrast to the plutonium in NTS soils, which is associated primarily with silt size particles, the ORNL samples showed that a substantial fraction of the

total sediment plutonium was associated with the clay size (40%). In order to compare differences in the behavior of plutonium in these two types of environmental samples, selected samples of each were leached with 8 M nitric acid for 1 hour at room temperature (28°C). The fractions leached in the acid are shown in Table 4: approximately 10 - 15% of the plutonium from the NTS samples and approximately 60 - 75% from the ORNL samples. This shows that the plutonium in ORNL sediment is more soluble than that in NTS soils.

To further characterize the behavior of plutonium in the NTS and ORNL samples, citric acid was used to extract the plutonium. The extraction data in Table 5 reinforce the results of the nitric acid leach; viz., that "NTS soil plutonium" is less soluble than "ORNL sediment plutonium." Note also that the plutonium in the ORNL sample is about 20 times more soluble than the NTS sample in citric acid. Furthermore, in comparing the citric and nitric acid leaches, the plutonium in NTS samples is about 10 times more soluble in 8 M nitric acid than in 0.1 M citric acid; but, in the ORNL samples, the plutonium is only about three times more soluble in 8 M nitric than in 0.1 M citric acid. These differences suggest that the chemical form of the plutonium is different in the two types of samples. If size distribution of the plutonium were the only difference, the citric acid solubilization in the clay size fractions (< 2 μM) of the two samples would have shown similar extractable percentages (not 3.9 and 15.6% as seen in Table 5). Further characterization of these samples and studies will be needed to define the chemical and physical forms of the plutonium in these environments.

Table 4. Plutonium leachability of selected soils and sediments in 8 M nitric acid. One-hour contact at room temperature (26°C).

Site Location	Soil Designation	Activity Leached (dpm/g)	Total Activity in Sample (dpm/g)	Fraction Leached (%)
Nevada Test Site	No. 2	306	2666	11.5
	No. 6	31	210	14.7
Oak Ridge National Laboratory	IRB - 1	347	452	76.8
	EG - S180 - 1	122	196	62.2
	W45 - S302 - 1	143	201	71.1

Table 5. Extraction of plutonium from selected soils and sediments in 0.1 M citric acid. *Thirty minutes' extraction at room temperature (28°C).*

Site Location	Soil Designation	Amount Extracted (dpm/g)	Total Activity in Sample (dpm/g)	Fraction Extracted (%)
Nevada Test Site	No. 2 (unsegregated)	12	915	1.3
	No. 3 (53 - 20 μM)	124	11084	1.1
	No. 3 (20 - 5 μM)	37	3968	0.9
	No. 3 (<2 μM)	35	896	3.9
Oak Ridge National Laboratory	EO - S180 - 1	32	135	23.7
	IRB - 1 (53 - 20 μM)	78	313	24.9
	IRB - 1 (20 - 5 μM)	75	301	24.9
	IRB - 1 (5 - 2 μM)	70	329	21.3
	IRB - 1 (<2 μM)	79	506	15.6

Implications of Findings in Area 13

The relatively uniform distribution of plutonium in the coarse silt fraction of all seven samples and the relatively constant fraction leached by 8 M nitric acid suggest that the uptake factor* for the same plant species in the area should be relatively constant. An increased uptake may be expected in plants growing in areas where soil is similar to sample 6 because of the comparatively high concentration of plutonium in the clay size fraction. The heavier textured soil of sample 6 contained 9% of the plutonium activity in the clay fraction as compared to less than 1.8% in the other samples.

The distribution of plutonium in different particle sizes should be useful in interpreting resuspension phenomena. Plutonium concentrations in the larger particles, such as the coarse silt and sands, would be useful in understanding plutonium accumulation under bushes by saltation and creep phenomena. Plutonium concentrations in the finer sizes, which could become airborne, would be valuable in interpreting the potential hazard of plutonium which might be moving off site.

The leaching data obtained with mineral and organic acids both on the NTS and ORNL samples should be useful in evaluating the potential hazard of the plutonium as it exists in these environments. The lower leachability in nitric acid and lower solubility in citric acid of the NTS samples, as compared with the ORNL samples, would suggest a lower uptake factor in plants grown in NTS soils.

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*Uptake factor is defined as the ratio of activity per gram of plant to that per gram of soil.

FUTURE PLANS

Plans for future studies include the following:

1. Complete segregation and analysis of three surface samples (6A, 6B, and 6C) taken between samples 6 and 7 of the north-south transect and which lie between the inner and outer fence in area 13. These samples will provide needed data to define mathematically the relationship of activity with distance from G.Z.

2. Continue chemical characterization studies of soil plutonium using NTS soils as well as other environmental samples representing different sources of plutonium. These studies will enable better prediction of the long-term behavior of plutonium in the environment and more reliable assessment of the potential hazard.

3. Continue physical characterization of soil plutonium in area 13, especially with regard to nearness to the site of detonation. Preliminary results suggest that the plutonium on particles near ground zero (600 feet or closer) are more tightly bonded to the soil particles than plutonium in soils farther away.

4. Study the distribution of plutonium in soil as a function of depth. Since surface plutonium occurs largely in the coarse silt sizes, the size distribution of plutonium at greater depths would provide information on the mode of downward transport. Leach tests of the plutonium as a function of depth and particle size would also aid in understanding the mechanism of downward movement.

5. Sample and analyze plutonium in soil type 405 from area 13 to establish and ascertain the extent of movement of plutonium by erosion.

ACKNOWLEDGMENTS

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