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Characterization of Pu-Contaminated Soils From Nuclear Site 201 at the Nevada Test Site[1]

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Nevada Operation Office

U.S. Department of Energy

Las Vegas, Nevada

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ABSTRACT

Distribution and characteristics of Pu-bearing radioactive particles throughout five soil profiles from Nuclear Site (NS) 201 were investigated. Concentrations of $^{239,240}Pu$ and ^{241}Am decreased with depth and most of the contamination was contained in the top 5 cm except in profile 4 where it extended to 10 cm. The mean activity ratio of $^{239,240}Pu$ to ^{241}Am and its standard error were 5.8 ± 0.3 (N=42). Most of the total radioactivity of the soils was contributed by 0.25-2 mm sand size fraction which comprised 20-50% by weight of the soils. The radioactive particles in the 0.25-2 mm size fraction occurred as spherical glass particles or as glass coatings on sand particles. The glass coatings had gas voids in the matrix but were not as porous as the radioactive particles from NS 219. After impact grinding the >0.25-mm size fractions for one hour, 85% of the initial activity in a NS 201 sample remained with the particles on the 0.25 mm sieve, whereas in the NS 219 sample only 10% remained. The results show that the radioactive particles from NS 201 were much more stable against the impact grinding force than those from NS 219. Therefore, the NS 201 soils would be expected to have a lower probability of producing respirable-size radioactive particles by saltation during wind erosion.

INTRODUCTION

Since 1972, the Nevada Applied Ecology Group (NAEG) has been investigating various environmental and health aspects of the soils contaminated by nuclear tests at the Nevada Test Site (NTS). Significant progress through multidisciplinary group efforts has been made on the inventory, distribution, and various other aspects of the contamination at the safety-shot sites (Essington et al., 1976; Tamura, 1976; Gilbert and Essington, 1977) and nuclear sites (Church et al., 1974; Gilbert et al., 1977; Lee and Tamura, 1981; Lee et al., 1982).

This report covers research progress concerned with characterizing Pu distributions on NTS soils during the period from October, 1981 through December, 1982. During this period, research efforts were directed to the characterization of Nuclear Site (NS) 201 soils in area 18. Area 18 encompasses steep hills, significant drainage channels, rock outcrops, and alluvial soils (Gilbert et al., 1977). The nuclear test at site NS 201 was conducted on July 7, 1962 and the distribution of radionuclides in the site was surveyed for initial inventory estimation (Church et al., 1974; Brady and Church, 1975; Gilbert et al., 1977). Later, as a part of intensive soil sampling program, 22 soil profile samples were collected; five of which were shipped to Oak Ridge National Laboratory for extensive investigations of the contamination characteristics of the soils.

The objectives and scopes of NTS soil characterization study at ORNL have been (i) to define physicochemical parameters controlling the redistribution of Pu, (ii) to assess environmental and health hazards of Pu contaminated soils, and (iii) to develop techniques for clean-up or

treatment through characterization of Pu associations with host soils. This report presents the results of $^{239,240}Pu$ and ^{241}Am distributions and associations in NS 201 soils, and the physical characteristics of the radioactive particles.

SAMPLE DESCRIPTION AND EXPERIMENTAL TECHNIQUE

Locations of the soil profiles selected for this investigation are shown on a ^{241}Am concentration contour map of the NS 201 (Fig. 1) which is taken from Gilbert et al. (1977). Profiles 1 and 4 were located southeast, and profiles 5, 8 and 10 northwest of ground zero (GZ). The soil samples were collected from selected profiles in predetermined increments down to a maximum depth of 50 cm in accordance with procedure established by Essington (1978). Each sample was screened through a 2-mm sieve. Further size segregation was accomplished by suspending 100 g of the <2-mm fraction in water and passing through 0.25- and 0.106-mm sieves sequentially. The <0.106-mm fraction was separated into 0.106-0.005 and <0.005-mm fractions by gravity settling and centrifugation (Jackson, 1975).

A portion of each size fraction was packed into a circular plastic container (49 mm diameter x 9 mm height) for ^{241}Am photon analysis using a intrinsic germanium (IG) detector with a thin beryllium window (Larsen and Lee, 1983). After dry grinding to <0.1 mm with a blender mill, up to 2 g of each size fraction was assayed for $^{239,240}Pu$ using the HASL-LASL method described by Tamura (1975).

The ^{239,240}Pu-rich particles were sorted from the >0.25 mm fractions using an alpha survey meter for morphological characterization by scanning electron microscopy (SEM). To determine the relative physical strength of radioactive vs nonradioactive particles, 15 g of 2-0.25 mm fraction of the surface soil from profile 8 were transferred to a 30-ml tungsten carbide vial. The vial assembled without mill-balls was mounted on the Pica blender mill (Pitchford Scientific Instruments, Cannonsburg, PA) to generate a mild impact grinding condition (Lee and Tamura, 1981). After one hour of impact grinding, the sample was separated by dry sieving into >0.25, 0.25-0.125, and <0.125 mm fractions, which were analyzed for ^{241}Am (as a reference to ^{239,240}Pu concentration).

RESULTS

Soil Properties

Gravels comprise from 13 to 60% of the total weight of most of the soils at NS 201 (Table 1). The soils, formed on alluvium derived from rhyolite and tuff, ranged from light gray (10 YR 7/2) to reddish yellow (7.5 YR 8/6) in color. The soils were alkaline ranging in pH from 7 to 8 and did not effervescent visibly when treated with cold 0.1 N hydrochloric acid.

The textures of the <2 mm soil fractions were classified as sand (profile 1), loamy sand (profiles 4 and 5), and sandy loam (profiles 8 and 10). Particle size analyses revealed that very coarse to medium sand (2-0.25 mm) was a dominant fraction in the loamy sand soils, whereas very fine sand to medium silt (0.106-0.005 mm) was the dominant fraction in the sandy loam soils (Table 2). The fine silt to clay fraction (<0.005 mm)

was a minor component of the soils. The 0.106 mm particle size is near the upper size limit of resuspendable soil particles (Chepil, 1945) and the 0.005 mm is close to the upper limit for respirable size (Volchok et al., 1972).

The soil properties did not change systematically within the profiles except for profile 10. The surface (0-5 cm) of soil profile 10 was more acidic and brownish than the subsurface horizons and the 0.005 mm fraction of that soil increased with depth.

Radionuclide Association

The ^{241}Am concentration contour map of NS 201 (Fig. 1) showed that contamination (>24 dpm/g) extended to about 900 m north and 120 m south of ground zero (Gilbert et al., 1977). The width of contamination was about 100 m at the southern end and 400 m at the northern end. Soil profile 1 located just outside the southern boundary had radionuclide activities which were below the detection limits in this study (80 dpm/g for $^{239,240}Pu$ and 10 dpm/g for ^{241}Am). As indicated by Fig. 1 the surface sample (0-2.5 cm) of profile 8 had the highest radionuclide concentrations, profiles 10 and 4 were intermediate, and profile 5 had the lowest. The depth of the contamination in the soil profiles also varied; extending down to 10 cm in profile 4, 6 cm in profile 8, and less than 5 cm in profiles 5 and 10.

Distributions of radionuclides in the different size fractions showed that the 2-0.25 mm fraction of each soil (<2 mm) contributed the major portion of the total radioactivity, that is, most radioactivity was contributed by particles having this diameter (Table 2). A few

radioactive particles having diameters larger than 2 mm were found within the gravel fraction (>2 mm), but their contribution to the whole soil activity was minimal. At a given depth within the soil profiles, the activity distribution among the size fractions was fairly uniform except in soil profile 5. In profile 5, the 2-0.25 mm fraction contributed 93% of the total soil radioactivity in the surface soil (0-0.25 cm), but the same size fraction only contributed 62% of the total activity in the 2.5-5 cm depth increment. The pronounced decrease of total radioactivity in the soil at 2.5-5 cm depth suggests that the boundary of contamination should be near the top of the soil segment.

Interest in ^{241}Am distribution in the soils is two-fold: (1) the possible radiological health hazard of the nuclide (Fowler and Essington, 1974) and (2) the possible application of the ^{241}Am distribution as a reference for ^{239,240}Pu distribution provided the ^{239,240}Pu to ^{241}Am activity ratio for selected samples in a given contaminated area is relatively constant within acceptable error. The ^{241}Am analysis by gamma spectrometry is simpler to perform and is cost effective compared to ^{239,240}Pu analysis by radiochemical separation and alpha spectrometry (Larsen and Lee 1983). In the NS 201 soil profile, the linear correlation coefficient (r^2) between ^{239,240}Pu and ^{241}Am activity was 0.93 (N=42) among the samples having ^{241}Am activity greater than 10 dpm/g (Fig. 2). The mean activity ratio of ^{239,240}Pu to ^{241}Am and its standard error (one sigma) of the samples were 5.8 ± 0.3 (N=42). The mean activity ratio is smaller than a previously reported value of 8.1 ± 0.7 (N=23) by Gilbert et al (1977), which may be attributed to ingrowth of ^{241}Am activity from decay of ^{241}Pu during the period from

1977 to 1982. Because only four contaminated profiles were studied and all were located near ground zero, more samples from different areas should be analyzed to test the validity and applicability of the activity ratio.

Physicochemical Properties of Radioactive Particles

A number of radioactive particles were separated from the larger than 2 mm fraction of the soils in order to conduct morphological and structural investigations. The particles were spherical or irregular in shape and translucent to opaque in appearance. Most of the particles were partially or completely covered with a glassy, fused material.

The scanning electron micrographs (Fig. 3) shows: (A) varying sizes of spherical particles formed on a fine-grain-mineral aggregate, (B) a marble-like large glass particle, (C) smoothness of the surface of glass coatings and particles, and (D) a cross-section surface of glass coating on a coarse fragment. The spherical glass particles were usually solid but the glass coatings frequently contained gas voids. The size of gas voids (or pores) is highly variable, but were often smaller near the glass-mineral interphase and larger near the surface of the coating. The physical continuity of the interphase and similarity of bulk chemical composition of the phases indicated that the glass phase was derived by melting the crystalline mineral surface.

The impact grinding and sieving separation showed that 85% of the ^{241}Am activity was unaffected by the grinding process and remained with the initial 2-0.25 mm size fraction which comprised 90% of the total weight (Table 3). Only 4% of the total ^{241}Am activity was associated with the

<0.125 mm fraction (3% by weight) generated by the grinding process. The differences between the percent weight and activity for each size fraction was small and not sufficient to recognize a preferential grinding process between radioactive glass and nonradioactive mineral particles. These results are in contrast to those of a previous study of NS 219 soils (Lee and Tamura, 1981), which showed a large difference between the percent weight and activity (Table 3). After impact grinding of 3.36-4.75 mm fraction, the <0.25 mm fraction which comprised only 18% of the total weight had 90% of the total activity. If the radioactive glass had less physical strength to withstand impact grinding than the non-radioactive particles, a larger activity contribution relative to the weight contribution from a finer size fraction and a smaller activity contribution from the coarser size fraction would be expected.

DISCUSSION

An initial radionuclide distribution resulted from the nature of the nuclear tests and environmental conditions could be changed through erosion and weathering processes. The significance of the erosional process to the redistribution of radionuclides has been well established through characterization of desert pavement and mound soils in area 11 (Tamura, 1977; Essington et al., 1977). Soil properties such as pH, color, and texture were determined to correlate with the radionuclide distribution among surface soils and within each soil profile. The texture of soils varied from sand to sandy loam but it was relatively uniform within each profile (Table 1). The uniformity of the soil texture, particularly between contaminated and noncontaminated zone

within a soil profile, indicated that the texture of surface soils was not affected by the fall-out materials caused by detonation. However, as was suggested by the varying contamination depths in the profiles (Fig. 1), the contaminated surface soils were locally redistributed after the deposition of contaminated particles. An extensive mixing of surface soils, probably by wind, was apparent from the absence of abrupt changes of soil properties and size distributions of radioactive particles in the contaminated zone. The variation of contamination depth further complicates the estimation of radionuclide inventory of the site. For a better estimation, the average depth of contamination should be determined from an expanded profile sampling scheme based on the topographical and morphological features of the area.

In the soils investigated, the 2-0.25 mm sizes were dominant and the size distribution of radioactive particles was fairly constant. A smaller size of radioactive particles was expected to dominate in the soils located farther away from ground zero as noted from another nuclear site (NS 219) (Lee and Tamura, 1981). However, at the location of profile 10, which was farthest from ground zero among the profiles selected on the NS 201 transect, was not sufficiently distant to reveal any reduction in particle size.

Although the size of radioactive particles from NS 201 was relatively smaller than of those from NS 219 (Table 3), NS 201 particles were less porous than the NS 219 particles. Physical strength of the radioactive particles is closely related to their porosity. Experimentally, the relative strength of the particles was measured by impact grinding (Lee

and Tamura, 1981). If radioactive particles had the same physical strength as the nonradioactive particles, their size reduction should be in the same proportion after impact grinding. If such particles were different in physical strength, the finer fraction should be dominated by physically weaker particles after grinding. As suspected from morphological studies of the radioactive particles, there was no significant preferential size reduction between radioactive and nonradioactive particles in NS 201 soil. Conversely, there was a significant preferential size reduction of radioactive particles compared to nonradioactive particles in NS 219. Since the results of the impact grinding experiments suggested that the radioactive particles from NS 201 are much more resistant to impact grinding than those from NS 219 (Table 3), the NS 201 soils are expected to have a lower probability of producing respirable-size radioactive particles by saltation during wind erosion.

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LIST OF TABLES AND FIGURES

Table 1. Soil Properties of Profile Samples Selected from NS 201

Profile No.	Library No.	Depth cm	>2 mm size %	Texture[a]	Color air dry	pH 1:1 ratio	Am-241[b] dpm/g
1	17269	0-5	38	s	10YR7/2	8.1	N.D.[c]
	17270	5-10	55	s	10YR7/2	7.8	N.D.
	17271	10-15	56	s	10YR7/2	7.8	N.D.
	17272	15-20	60	s	10YR7/2	7.2	N.D.
	17273	20-25	35	s	10YR7/2	7.3	N.D.
	17274	25-30	47	s	10YR7/2	7.5	N.D.
4	17275	0-2.5	21	ls	10YR7/3	7.3	1296
	17276	2.5-5	20	ls	10YR7/3	7.5	1296
	17277	5-10	20	ls	10YR7/3	7.6	810
	17278	10-15	40	ls	10YR7/3	7.6	720
	17279	15-20	28	ls	10YR7/3	7.5	72
	17280	20-25	22	ls	10YR7/3	7.2	6
	17281	25-30	24	ls	10YR7/4	7.1	6
	17282	30-50	-	Rock	-	-	N.D.
5	17283	0-2.5	26	ls	10YR7/3	7.3	300
	17284	2.5-5	26	ls	10YR7/3	7.3	300
	17285	5-10	34	ls	10YR7/3	7.7	36
	17286	10-22.5	-	Rock	-	-	6
	17287	22.5-27.5	21	ls	10YR7/4	7.2	N.D.
	17288	27.5-32.5	29	ls	10YR7/4	7.2	N.D.
	17289	32.5-37.5	30	ls	10YR7/4	7.0	N.D.
	17290	37.5-42.5	31	ls	10YR7/4	7.3	N.D.
8	17291	0-2.5	25	sl	10YR7/4	7.5	6726
	17292	2.5-5	13	sl	10YR7/4	7.9	6726
	17293	5-6	13	sl	10YR7/4	7.8	198
	17294	6-13	-	Rock	-	-	N.D.
	17295	13-18	27	sl	10YR7/4	7.4	N.D.
	17296	18-25	-	Rock	-	-	N.D.
10	17297	0-2.5	51	sl	10YR7/4	5.6	4350
	17298	2.5-5	26	sl	10YR7/4	6.4	4350
	17299	5-10	28	sl	7.5YR8/4	7.2	144
	17300	10-15	40	sl	7.5YR8/4	7.1	N.D.
	17301	15-20	32	sl	7.5YR8/6	7.2	N.D.
	17302	20-22.5	55	sl	7.5YR7.6	6.9	N.D.

[a] s = sand, ls = loamy sand, sl = sandy loam.

[b] Provided by Reynolds Electrical and Engineering Company, Inc.

[c] Not detectable.

Table 2. Particle Size and Radionuclide Distribution in the Selected NS 201 Soil Profiles

Profile No.	Depth cm	Size range mm	Size distribution weight %	$^{239,240}Pu$ activity dpm/g	$^{239,240}Pu$ contribution[a] %	Am^{241} activity dpm/g
1	0-5.0	2-0.25	72	b	c	d
		0.25-0.16	11	b	c	d
		0.106-0.005	11	b	c	d
		<0.005	6	b	c	d
	5-10	2-0.25	77	b	c	d
		0.25-0.106	8	b	c	d
		0.106-0.005	10	b	c	d
		<0.005	5	b	c	d
4	0-2.5	2-0.25	45	12241	84	1929
		0.25-0.106	32	2247	11	797
		0.106-0.005	21	1484	4	301
		<0.005	2	1058	1	747
	2.2-5	2-0.25	50	11309	80	2744
		0.25-0.106	30	3465	15	579
		0.106-0.005	19	1828	4	380
		0.005	1	3005	1	889
	5-10	2-0.25	41	13217	79	1899
		0.25-0.106	33	3402	16	567
		0.106-0.005	24	1230	4	248
		<0.005	2	2400	1	440
	10-15	2-0.25	44	450	c	110
		0.25-0.106	30	b	c	23
		0.106-0.005	24	b	c	d
		<0.005	2	b	c	d
5	0-2.5	2-0.25	52	24620	93	2324
		0.25-0.106	23	2488	4	426
		0.106-0.005	23	944	2	159
		<0.005	2	1914	1	294
	2.5-5	2-0.25	42	620	62	90
		0.25-0.106	28	310	20	38
		0.106-0.005	26	180	10	35
		<0.005	4	910	8	113
	5-10	2-0.25	51	b	c	d
		0.24-0.106	25	b	c	d
		0.106	22	b	c	d
		<0.005	2	240	c	36

Table 2 (continued)

Profile No.	Depth cm	Size range mm	Size distribution %	$^{239,240}Pu$ activity dpm/g	$^{239,240}Pu$ contribution[a] weight %	Am^{241} activity dpm/g
8	0-2.5	2-0.25	24	74530	96	11557
		0.25-0.106	21	578	1	194
		0.106-0.005	46	893	2	176
		<0.005	9	1690	1	259
	2.5-5	2-0.25	20	29000	91	4900
		0.25-0.106	21	1800	6	280
		0.106-0.005	48	290	2	55
		<0.005	11	630	1	80
	5-6	2-0.25	22	6100	94	1200
		0.25-0.106	20	110	2	15
		0.106-0.005	47	89	3	11
		<0.005	11	170	1	18
10	0-2.5	2-0.25	31	21724	92	4777
		0.25-0.106	21	1076	3	191
		0.106-0.005	45	588	4	86
		<0.005	3	1782	1	293
	2.5-5	2-0.25	26	18429	97	4039
		0.25-0.106	22	237	1	51
		0.106-0.005	45	91	1	14
		<0.005	7	639	1	124
	5-10	2-0.25	21	b	c	d
		0.25-0.106	19	b	c	d
		0.106-0.005	48	b	c	d
		<0.005	12	b	c	d
	10-15	2-0.25	21	b	c	d
		0.25-0.106	19	b	c	d
		0.106-0.005	44	b	c	d
		<0.005	16	b	c	d

[a]Percent activity contribution of each size range to the <2-mm fraction (radionuclide activity in a fraction x fractional abundance of size range/total activity of 2-mm fraction).

[b]$^{239,240}Pu$ activity was <80 dpm/g (counting error was 7% at 89 dpm/g).

[c]Not calculatable.

[d]^{241}Am activity was <10 dpm/g (counting error was 13% at 11 dpm/g).

Table 3. Radioactive Particle Separation by Grinding and Sieving Method

Particle size mm	weight distribution %	Activity distribution dpm/g	%
	Nuclear site 201[a]		
2-0.250	90	2354	85
0.250-0.125	7	4117	11
<0.125	3	3276	4
	Nuclear site 219[b]		
<2	74	22	1
2-0.250	8	2612	9
0.250-0.125	9	10560	45
<0.125	9	11712	45

[a]Start with 15 g of 2-0.25-mm size fraction.

[b]Start with 10 g of 3.36-4.75-mm size fraction.

ORNL–DWG 83–1947

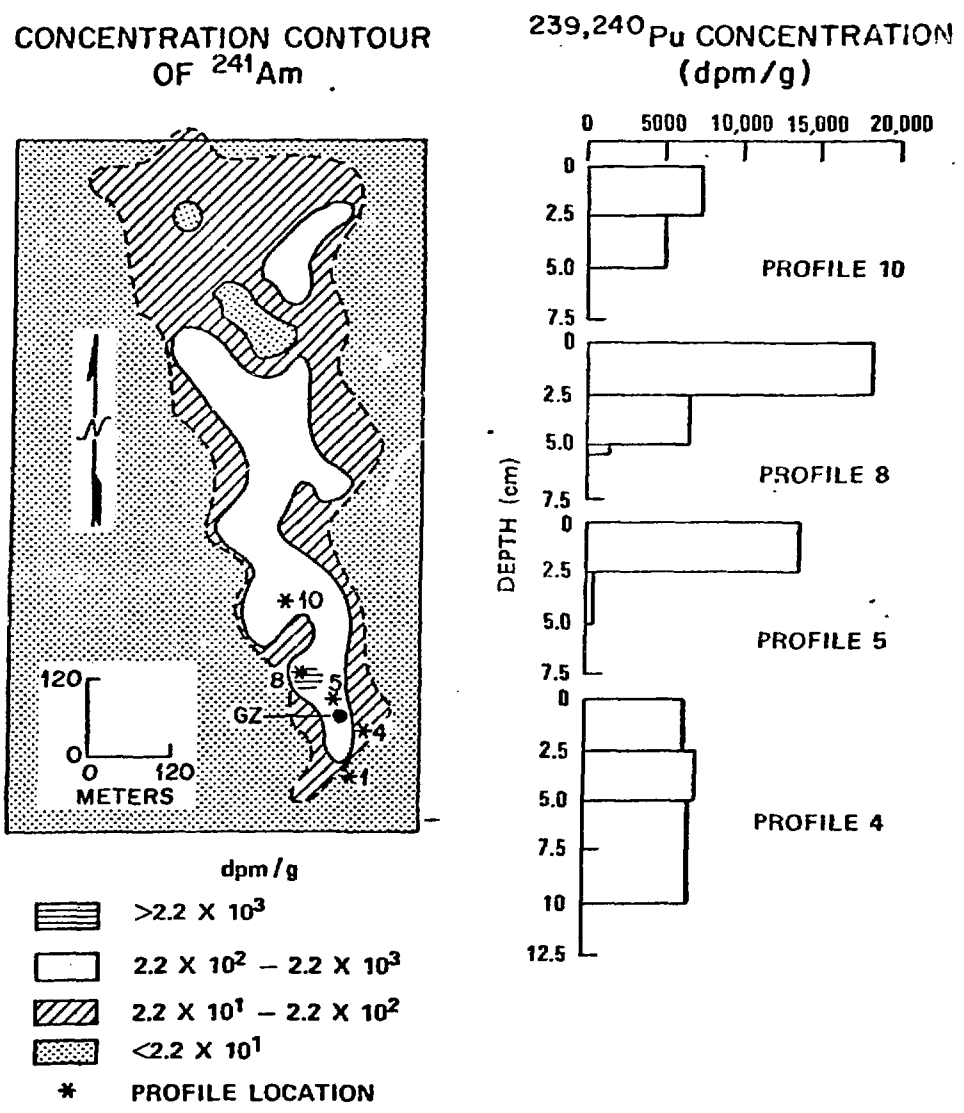

Fig. 1

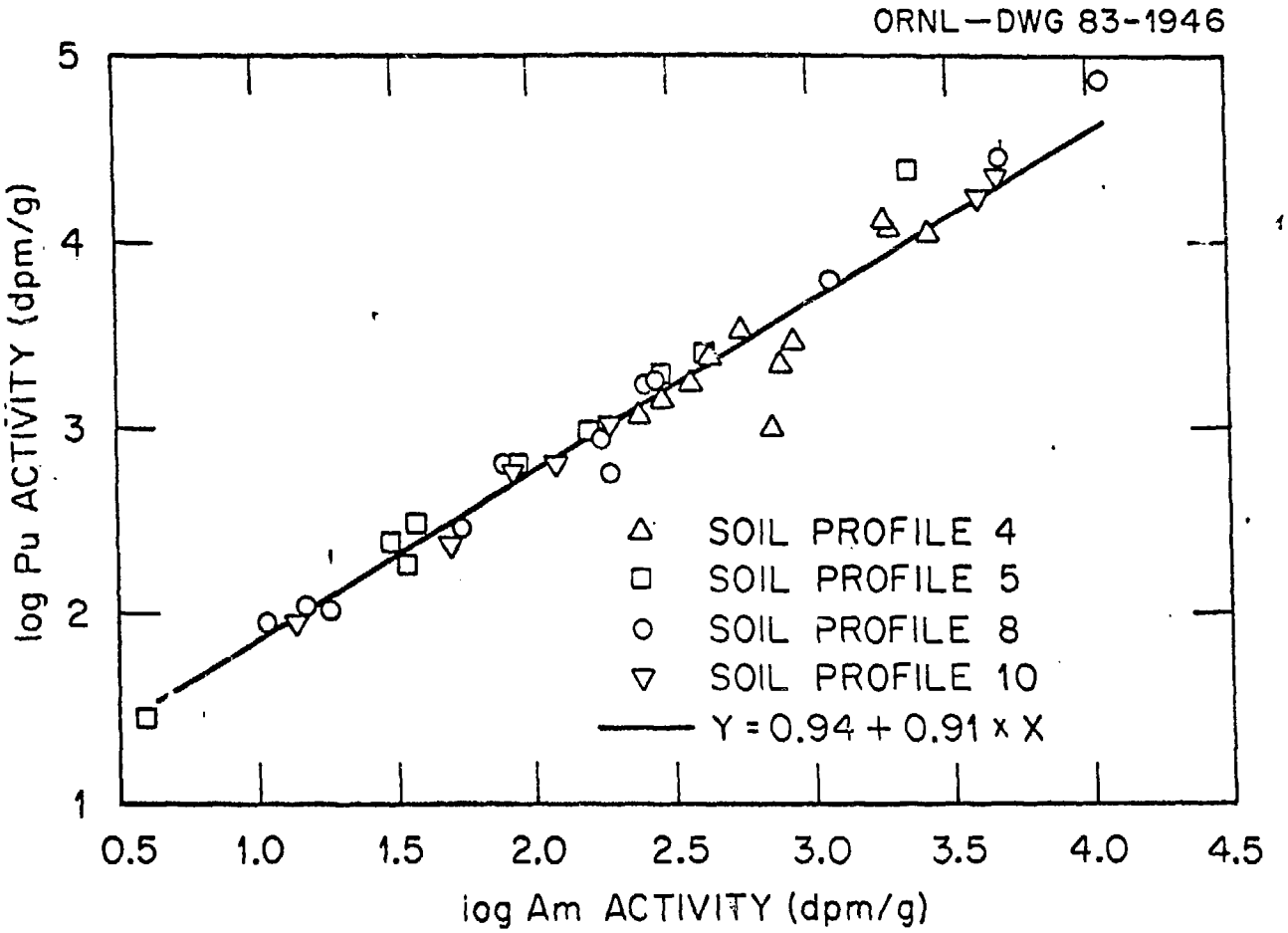
ORNL—DWG 83-1946
log Pu ACTIVITY (dpm/g)
log Am ACTIVITY (dpm/g)
5
4
3
2
1
0.5
1.0
1.5
2.0
2.5
3.0
3.5
4.0
4.5
SOIL PROFILE 4
SOIL PROFILE 5
SOIL PROFILE 8
SOIL PROFILE 10
Y = 0.94 + 0.91 x X

Fig. 2

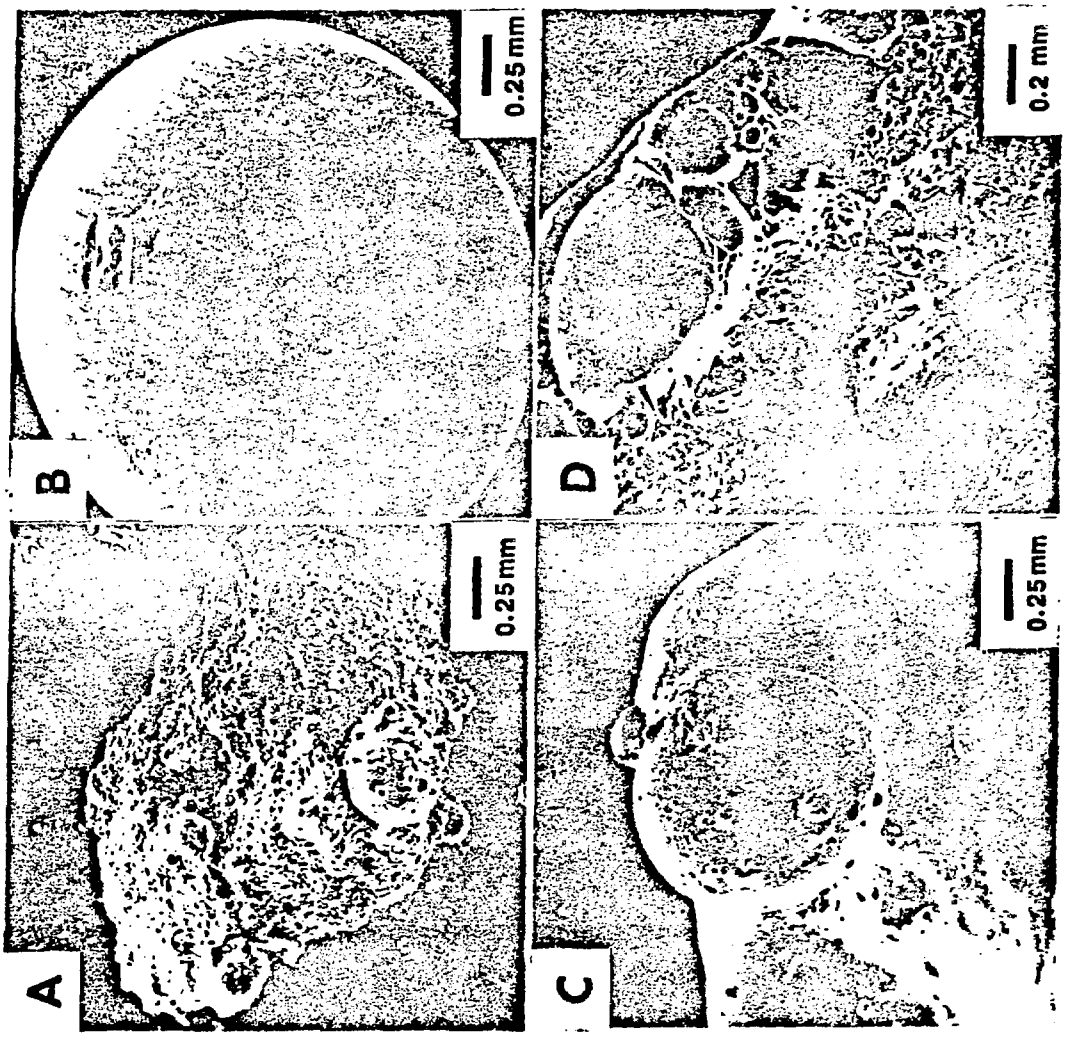

Fig. 3