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Distribution of Plutonium in Soil Particle Size Fractions of Liquid Effluent-Receiving Areas at Los Alamos[1]

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ABSTRACT

The alluvial soils of three liquid effluent-receiving areas at Los Alamos, New Mexico were sampled to determine the distribution of ^{238}Pu and $^{239,240}Pu$ in soil size fractions as a function of soil depth, distance from the waste outfall, and soil physical-chemical properties. Although the plutonium concentrations in the <53 μm soil size fraction were 10 times higher than plutonium levels in the 2-23 mm fraction, the largest portion of the plutonium inventory in the average soil sample was found in size fractions >105 μm. Plutonium concentrations decreased curvilinearly with distance from the waste outfall and with soil depth in the soils of discharge areas currently receiving treated liquid wastes. The <53 μm size fractions contained 7-fold larger surface areas and 3-fold larger cation exchange capacities than the 2-23 mm size fractions; plutonium concentrations in the size fractions were correlated with surface area in all three discharge areas.

Significant differences in the distribution of ^{238}Pu and $^{239,240}Pu$ were found in the soil size fractions from Mortandad canyon. Most soil separates from this canyon exhibited an approximate doubling of the $^{239,240}Pu/^{238}Pu$ ratio with increased soil depth and within 40 m from the waste outfall. The <53 μm soil size fraction had a 2-fold higher average $^{239,240}Pu/^{238}Pu$ ratio for each sampling station than the 2-23 mm size fraction.

The distribution of plutonium in the soil size fractions is discussed as a function of changing isotopic composition of the liquid wastes added to the canyons, movement of soil and plutonium in these intermittent streams, and potential isotopic differences in the behavior of ^{238}Pu and $^{239,240}Pu$.

Additional Index Words: aquatic ecosystems, intermittent streams, ^{238}Pu, ^{239}Pu, radioactive wastes, radionuclide distributions, sediments, volcanic soils.

The production rates of plutonium have been projected to increase to 20,000 kg per year by 1980 (International Atomic Energy Agency, 1965). The growing use of plutonium by the nuclear power industry may increase the probability of environmental contamination over that which has already occurred as a result of weapon tests, accidents, and the activities of fuel reprocessing, fabrication, and production facilities (Mays et al., 1970).

Treated low-level radioactive wastes generated by fuel reprocessing and fabrication facilities are commonly discharged to rivers or streams, where alluvial soils are considered as an integral part of the total waste treatment and handling system. In spite of the fact that many of the radionuclides in these liquid wastes become concentrated in specific particle size fractions of the alluvium, little is known about the distribution of plutonium in this type of an environment, most of the research having been performed in terrestrial ecosystems (Tamura, 1974). The distribution of plutonium in soil size fractions is an important consideration in setting meaningful radiation health standards for plutonium in soils since certain resuspended soil particles are respirable by man, and in waste management, because plutonium can be differentially transported downstream on soil particles of various sizes.

The objective of this research was to determine the distribution of ^{238}Pu and $^{239,240}Pu$ in size fractions of soil collected in three canyons used as liquid radioactive waste discharge areas for varying lengths of time. Plutonium concentrations in the soil separates were determined as functions of distance from the waste outfall, soil depth, and physical-chemical properties of the soil size fractions.

MATERIALS AND METHODS

Site Descriptions and Use Histories

The three study areas which have received treated radioactive liquid wastes are located in Mortandad, DP-Los Alamos, and Acid-Pueblo canyons in Los Alamos and Santa Fe counties, New Mexico (Fig. 1). The alluvium of the intermittent streams in Mortandad, DP and Acid canyons was formed from the rhyolitic volcanic rocks of the Bandelier Tuff. The alluvial soils in Pueblo and Los Alamos canyons were derived from the latite and quartz latite rocks of the Tschicoma Formation and from tuff particles from Acid and DP canyons. The lower reaches of Pueblo canyon also contain basaltic rocks in the stream channel matrix. Whereas the upper reaches of the canyons are characterized by stream channel widths of <1 m and alluvium depths of <0.15 m, the stream channels in the lower canyon areas are about 3 m wide and the alluvium is 0.15–30 m deep at the sampling locations. Although the hydrology and geology of these areas was characterized (Griggs, 1964), little is known about the properties of the soils of the area.

The canyons have been used as disposal areas for plutonium-containing liquid wastes for varying lengths of time. Acid-Pueblo canyon received untreated liquid wastes from 1944 to 1951, and treated waste products from 1951–1963 (2.1 ± 1.8 standard deviation [SD] millicuries [mCi] plutonium/year). Treated wastes have been discharged into Mortandad canyon since 1963 (4.2 ± 2.3 [SD] mCi plutonium/year) and into DP-Los Alamos canyon since 1953 (1.6 ± 1.2 [SD] mCi plutonium/year). The aging effects on the distribution of plutonium in the alluvium of these intermittent streams have not been characterized.

Sampling and Analytical Methods

Five 30-cm soil cores were collected along the center of the stream channel at 60-cm intervals at each of 33 permanent sampling stations located in each canyon during May-June 1973 (Fig. 1).

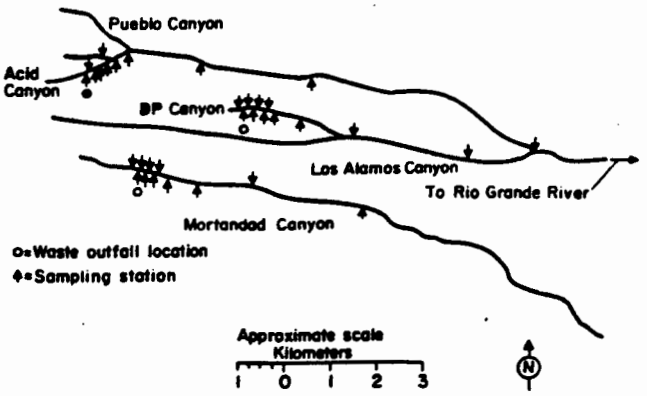

Fig. 1—Location of sampling stations in canyon study areas.

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Samples were taken 100 m above the waste outfall and at distances of 0, 20, 40, 80, 160, 320, 640, 1,280, 2,560, 5,120, and 10,240 m below the outfall along a sampling network described by Hakonson et al. (1973, 1974). Each soil core was taken with a polyvinylchloride coring tube (2.4 cm inside diameter [ID], schedule 89 pipe with a sharpened end), which was placed in a plastic bag, frozen, and cut into segments corresponding to 0–2.5, 2.5–7.5, 7.5–12.5, and 12.5–30 cm soil depths. When the alluvium was < 12.5 or 30 cm deep a partial core section was obtained and its length also measured. Individual soil samples were mechanically separated into six size fractions using a Model L3P sonic sifter (ATM Corp., Milwaukee, Wis.): < 53 μm, 53–105 μm, 105–500 μm, 500–1,000 μm, 1–2 mm, and 2–23 mm. Small soil particles were blown off of the larger soil separates with an air hose during the soil separations to minimize contamination of large soil particles with smaller particles. A < 2 μm particle size fraction of soil was obtained from 20 g aliquots of soil from each of three sampling locations (320 m and 2.56 km from the Mortandad waste outfall; 160 m from the DP outfall) for clay mineralogical analysis using x-ray diffraction techniques (U. S. Department of Agriculture, 1972). The cation exchange capacities of the particle size fractions obtained at these three locations were also determined (U. S. Department of Agriculture, 1972).

Soil from the 0–2.5 cm depths of the five cores collected at each sampling locatio:. was composited and particle size fractions from these composites were assayed for their ^{238}Pu and ^{239,240}Pu content (the term *total plutonium* used herein designates the sum of these three isotopes) as previously described (Hakonson et al., 1973). Soil collected from the other depths was composited, fractionated, and analyzed similarly. For plutonium analysis, 20-g soil samples were dissolved in HNO_3 and HF, and the plutonium was isolated on an anion exchange column, electrodeposited on a stainless steel disc and determined by alpha-particle spectrometry. Absolute amounts of ^{238}Pu and ^{239,240}Pu were obtained by correcting the counting data for chemical recovery, which was determined from the level of ^{242}Pu initially added to the sample.

RESULTS AND DISCUSSION

Plutonium in Soil Size Fractions

Maximum total plutonium concentrations were generally associated with small soil particles in Mortandad (Table 1), DP-Los Alamos (Table 2), and Acid-Pueblo (Table 3) canyon soils. The <53 μm soil size fractions had an average of 10-fold higher total plutonium concentrations than the 2–23 mm size fractions from all three discharge areas (0–2.56 km from the waste outfalls).

The distribution of plutonium concentrations within the soil size fractions varied significantly from discharge area to discharge area. The ratio (X) of the total plutonium concentrations in the <53 μm size fraction to those in the 2–23 mm fraction was significantly higher ($P < 0.01$) for soils collected 0–2.56 km from the Mortandad waste outfall ($X = 20$) than for similar samples from the other two canyons ($X = 6$–9). This distribution pattern suggests a correlation between the length of use of these discharge areas and the plutonium concentrations found in the soil size fractions, i.e., these alluvial soils contained increasingly lower plutonium concentrations in <53 μm fractions relative to plutonium levels in the 2–23 mm size fractions as time of usage increased. The gross alpha radiation measurements made on soil size fractions in Mortandad canyon in late 1964 by Purtymun (1966) also support this temporal relationship.

Although maximum total plutonium concentrations were generally found in size fractions < 105 μm, the largest percentage of the plutonium in the whole soil samples was usually found in coarser size fractions due to larger amounts of material in size fractions with diameters > 105 μm (Table 4). Almost 80% of the plutonium in samples from Mortandad canyon was associated with soil particles > 105 μm, which composed 96% of the Mortandad alluvium by weight. The soils of Acid-Pueblo canyon and most of DP-Los Alamos canyon followed the same pattern as the post-outfall soils from Mortandad canyon. The distribution of total plutonium in the size fractions of samples collected 40 and 80 m from the DP-Los Alamos outfall did not fit this pattern, i.e., 45% of the alluvium consisted of particles with diameters < 105 μm and accounted for 50% of the plutonium in these soils (Table 4).

The dry sieving technique used to produce the size fractions for plutonium analyses may have resulted in contamination of large soil particles with small particles, accounting for a portion of the plutonium inventory associated with large particles. This problem was minimized for most of the samples because (i) there were small amounts of fine particles in most of the soil samples (Table 4), and (ii) small soil particles were blown off of the larger soil size fractions with the air hose during the soil separations. In spite of these precautions, the large size fractions of finer-textured soils (such as the soils collected 40 and 80 m from the DP-Los Alamos outfall) were probably contaminated with small soil particles resulting in an overestimation of the plutonium inventory in the large soil size fractions.

The plutonium concentrations of the soil separates were compared with the surface area of the size fractions collected in each waste disposal area. The surface area of each particle size fraction was calculated from the fraction's mean effective particle diameter, a procedure shown to underestimate the surface area of particles < 0.2 μm (Kohnke, 1968). The surface area of the size fractions ranged from 1.7 to 12.5 cm^2/g as the mean effective particle diameter varied from 12.5 mm (2–23 mm size fraction) to 26.5 μm (<53 μm size fraction). Plutonium levels were expressed as a percentage of plutonium concentrations in the 2–23 mm size fraction in these comparisons to minimize the effect of the horizontal and vertical plutonium concentration gradients. Thus, the log of the relative ^{238}Pu (Mortandad canyon) or ^{239,240}Pu (DP-Los Alamos and Acid-Pueblo canyons) concentrations was significantly correlated ($P < 0.05$) with the log of the surface area of soil size fractions from Mortandad (r [correlation coefficient] = 0.82, n = 198), DP-Los Alamos (r = 0.46, n = 186), and portions of Acid-Pueblo canyon (80–2,560 m from the outfall; r = 0.57, n = 132). No significant plutonium-surface area correlations were found for soils in portions of Acid-Pueblo canyon, probably due to dilution of radioactive sediments with noncontaminated soil and translocation of plutonium-contaminated soils since the cessation of waste additions to this canyon in 1963.

Mineralogical and chemical reactivity differences in the soil size fractions were also considered as influencing the distribution of plutonium in the soil separates. Whereas the 2–23 mm particles in soils formed in Bandelier Tuff (Mortandad, DP and Acid canyons) were composed of relatively unweathered tuff fragments, this size fraction contained mainly latites and quartz latites in Los Alamos and Pueblo canyon (Fig. 1). Quartz and sanidine crystals

Table 1—Total plutonium concentrations in soil separates from Mortandad canyon

Soil depth	Total plutonium concentration in size fraction (pCi/g)*					
cm	<53 μm	53–105 μm	105–500 μm	500–1,000 μm	1–2 mm	2–23 mm
			100 m preoutfall			
0 - 2.5	1.5	1.9	5.5	0.66	2.3	2.6
2.5- 7.5	1.1	1.2	1.0	0.57	0.15	0.29
7.5-12.5	0.32	0.26	0.15	0.69	0.046	0.047
			0–40 m from outfall			
0 - 2.5	1,500 (250)†	1,300 (360)	610 (140)	310 (150)	87 (57)	69 (39)
2.5- 7.5	1,400 (610)	1,200 (350)	540 (130)	230 (80)	230 (68)	91 (39)
7.5-12.5	1,100 (720)	710 (430)	420 (360)	320 (360)	90 (87)	68 (65)
			80–1,280 m from outfall			
0 - 2.5	380 (320)	390 (190)	140 (110)	62 (40)	45 (29)	27 (13)
2.5- 7.5	240 (140)	140 (75)	76 (47)	43 (25)	27 (25)	29 (18)
7.5-12.5	120 (46)	94 (46)	39 (17)	20 (13)	12 (5.5)	11 (6.1)
12.5-22.1	39 (17)	31 (11)	16 (2.1)	7.4 (.85)	4.5 (.71)	3.6 (1.8)
			2.56 km from outfall			
0 - 2.5	24	16	7.6	2.4	1.5	– ‡
2.5- 7.5	49	32	18	6.4	3.9	6.2
75.-12.5	37	20	8.3	7.1	2.9	2.9
12.5-24	13	7.1	2.5	1.5	1.4	1.0
			5.12 km from outfall			
0 2.5	1.1	1.0	0.18	0.080	0.045	0.015
2.5- 7.5	0.25	0.63	0.088	0.079	0.028	0.0063
7.5-12.5	0.30	0.38	0.11	0.062	0.034	0.0063
12.5-22.4	0.41	0.39	0.17	0.056	0.047	0.017

* Each value represents the total plutonium concentration in a composite of five soil core segments.
† Numbers in parentheses represent the standard deviation of the mean total plutonium concentrations.
‡ Missing data.

Table 2—Total plutonium concentrations in soil separates from DP-Los Alamos canyon

Soil depth	Total plutonium concentration in size fraction (pCi/g)*					
cm	<53 μm	53–105 μm	105–500 μm	500–1,000 μm	1–2 mm	2–23 mm
			100 m preoutfall			
0 - 2.5	9.1	0.68	0.050	0.021	0.022	0.015
2.5- 7.5	1.7	0.91	0.041	0.028	0.022	0.0091
7.5-12.5	0.23	0.16	0.055	0.028	0.038	0.0094
12.5-22.4	0.12	0.18	0.047	0.059	0.032	0.0068
			0 m from outfall			
0 - 2.5	650	720	1,000	1,000	1,000	930
2.5- 7.5	2,000	4,300	2,600	80	2,000	1,500
7.5-12.8	730	420	– †	1,100	220	53
			20–80 m from outfall			
0 - 2.5	38 (21)‡	33 (15)	50 (6.4)	26 (13)	9.4 (4.7)	19 (6.8)
2.5- 7.5	22 (11)	24 (10)	39 (23)	19 (19)	8.5 (5.7)	4.9 (6.5)
7.5-12.5	5.0 (5.2)	13 (13)	7.3 (9.3)	2.8 (3.3)	2.4 (2.8)	4.3 (6.7)
12.5-22.4	0.90 (0.86)	0.83 (0.24)	0.69 (0.20)	0.27 (0.07)	0.21 (0.13)	0.32 (0.26)
			160–1,280 m from outfall			
0 -2.5	1.1 (0.45)	1.4 (0.77)	0.90 (0.43)	0.47 (0.34)	0.24 (0.20)	0.41 (0.40)
2.5- 7.5	2.0 (0.80)	1.7 (0.30)	1.3 (0.59)	0.47 (0.25)	0.31 (0.16)	2.6 (4.0)
7.5-12.5	1.9 (0.29)	1.8 (0.62)	1.1 (0.45)	0.50 (0.13)	0.24 (0.08)	0.36 (0.17)
12.5-22.6	3.6 (2.7)	3.4 (3.0)	2.1 (1.7)	2.2 (3.0)	0.67 (0.40)	0.43 (0.27)
			2.56 km from outfall			
0 - 2.5	0.61	1.1	0.23	0.23	0.28	0.060
2.5- 7.5	0.46	0.49	0.28	0.075	0.035	0.041
7.5-11.6	0.77	0.36	0.28	0.099	0.069	0.049
			5.12 km from outfall			
0 - 2.5	0.05	0.45	0.76	0.058	0.052	0.83
2.5- 7.5	0.94	0.71	0.46	0.14	0.13	0.084

* Each value represents the total plutonium concentration in a composite of five soil core segments.
† Missing data.
‡ Numbers in parentheses represent the standard deviation of the mean total plutonium concentrations.

were the major components of the 500–2,000 μm size fractions, whereas the smaller fractions contained the weathered volcanic ash component of the tuff. The x-ray diffraction analysis of the soils from Mortandad (320 m and 2.56 km from the waste outfall) and DP (160 m from waste outfall) canyons demonstrated that amorphous clays (allophane) made up most of the < 2 μm portion of the < 53 μm soil size fraction, with trace amounts of clay mica and smectite also present. The average cation exchange capacities of the 1–2 mm and < 53 μm size fractions collected at these three locations were 0.060 and 0.21 eq/kg, respectively. Thus, the smaller soil size fractions not only contained 7 times more surface area than the largest soil particles, but also about 3 times greater cation exchange capacities. Although the surface area and cation exchange capacity of a particle size fraction are important in determining how plutonium reacts with a soil separate, the ratio of plutonium concentrations in small vs. large size fractions was usually larger than the ratio of cation exchange capacities in small vs. large soil separates. This may imply that surface area is more important than cation exchange capacity of these soils in influencing the distribution of plutonium in various soil size fractions.

Concentrations of total plutonium in the six size fractions were also studied as a function of distance from the waste outfall and soil depth. Levels of plutonium found in the soils 5.12 and 10 km from the DP-Los Alamos and Acid-Pueblo outfalls and at the preoutfall station in Mortandad canyon were greater than worldwide fallout levels of up to 0.1 picocuries [pCi] Pu/g found in New Mexico soils (Herceg, 1972 and 1973; Kennedy and Purtymun, 1971; Schiager and Apt, 1974). Although the latter case represented a small source of contamination above the Mortandad waste outfall, the radionuclide levels observed in the other two canyons represented transport of small amounts of laboratory wastes as far as 10 km away from their point of discharge. Plutonium concentrations in the soil samples collected in Mortandad canyon decreased exponentially with distance from the waste outfall whereas levels of plutonium were significantly correlated with the inverse of distance from the outfall in DP-Los Alamos and Acid-Pueblo canyon soils, reflecting a migration of plutonium into Los Alamos and Pueblo canyons[3]. Plutonium concentrations decreased exponentially with soil depth in Mortandad canyon soils, whereas the plutonium concentrations in Acid-Pueblo

[3] J. W. Nyhan, F. R. Miera, Jr., and R. J. Peters. 1976. The distribution of plutonium and cesium in alluvial soils of the Los Alamos environs. Submitted to Proc. of the Fourth Nat. Symp. on Radioecology.

Table 3—Total plutonium concentrations in soil separates from Acid-Pueblo canyon

Soil depth	Total plutonium concentrations in size fraction (pCi/g)*					
cm	<53 μm	53–105 μm	105–500 μm	500–1,000 μm	1–2 mm	2–23 mm
			100 m preoutfall			
0 - 2.5	2.7	2.2	0.50	0.18	0.099	0.035
2.5- 7.5	1.7	2.1	0.58	0.23	0.16	0.024
			0–40 m from outfall			
0 - 2.5	85 (74)†	60 (25)	25 (9.1)	8.8 (3.7)	7.9 (4.2)	25 (31)
2.5- 7.5	90 (78)	45 (26)	41 (48)	13 (11)	14 (11)	19 (26)
7.5–12.5	33	39	21	8.7	6.1	5.6
12.5–21	110	70	39	21	16	5.8
			80 m from outfall			
0 - 2.5	42	56	17	4.3	4.9	3.5
2.5- 7.5	26	25	15	5.4	3.3	3.1
7.5–12.5	20	60	39	16	11	24
12.5–22	1,700	1,500	900	340	190	110
			160–640 m from outfall			
0 - 2.5	50 (38)	53 (28)	10 (9.6)	8.2 (1.6)	6.7 (2.5)	5.5 (2.3)
2.5- 7.5	46 (12)	46 (6.0)	20 (1.5)	9.5 (2.7)	6.1 (0.95)	8.9 (6.2)
7.5–12.5	42 (4.2)	67 (16)	25 (3.5)	7.9 (0.57)	6.6 (2.3)	12 (4.6)
12.5–21	57 (12)	58 (9.6)	35 (6.8)	13 (3.7)	10 (0.58)	12 (6.3)
			2.56 km from outfall			
0 - 2.5	97	110	110	44	21	6.7
2.5- 7.5	140	110	1,000	160	350	195
7.5–12.5	2,300	2,000	2,000	3,100	3,200	1,600
			5.12 km from outfall			
0 - 2.5	77	38	3.1	0.84	0.46	0.54
2.5- 7.5	14	4.5	2.1	0.34	0.028	–‡
7.5–12.5	4.8	3.6	1.3	0.14	0.49	0.50
12.5–22	3.5	3.5	1.8	0.48	0.35	3.7
			10.2 km from outfall			
0 - 2.5	2.7	1.6	1.1	<0.0025	0.30	0.44
2.5- 7.5	3.8	2.9	1.1	0.65	0.31	<0.0025
7.5–12.5	3.2	2.4	0.54	0.24	0.42	0.47
12.5–22	4.1	6.3	–	1.1	0.39	0.43

* Each value represents the total plutonium concentration in a composite of five soil core segments.
† Numbers in parentheses represent the standard deviation of the mean total plutonium concentrations.
‡ Missing data.

soils occurred as residual pockets of contamination deep in the soil profile[3].

Differential Distribution of Plutonium Isotopes in Soils

The horizontal and vertical distributions of ^{238}Pu and $^{239,240}Pu$ were compared with one another for the soil size fractions from each canyon. Differences in the $^{239,240}Pu$-^{238}Pu activity ratios in these soil samples could result due to: (i) a change in the isotopic ratios or chemical forms of plutonium in the liquid wastes which were added to the canyons, (ii) relocation of plutonium-containing soil particles with soil movement in the stream channel, and (iii) differential solubility, adsorption, and physical mixing of plutonium isotopes in the soils of these intermittent streams. Thus, there are physical properties of ^{238}Pu that might account for its enhanced solubility and mobility in soils compared to $^{239,240}Pu$ (Hanson, 1975).

Since the first appreciable amounts of ^{238}Pu appeared in laboratory liquid wastes in 1968, the $^{239,240}Pu/^{238}Pu$ ratios can be used to indicate horizontal-vertical mixing-migration rates of plutonium in these soil separates (Fig. 2, 3, and 4) as a function of canyon use history. Partial mixing of ^{238}Pu into the alluvium had occurred in < 5 years (1968–1973), since no significant differences ($P < 0.05$) were found between the $^{239,240}Pu/^{238}Pu$ ratios of soil samples collected as far as 2.56 km from the waste outfalls and the ratios currently found in the waste treatment plant effluents (Schiager and Apt, 1974). Since the DP-Los Alamos effluent discharge pathway was redirected in 1972 to the 40- and 80-m sampling stations, the migration of ^{238}Pu into these soils occurred in < 1 year. These observations probably reflect a fast reaction between plutonium in the liquid wastes and the alluvial soils, as well as downstream movement of plutonium and soil during snowmelt and summer runoff.

The horizontal and vertical variations in the $^{239,240}Pu/^{238}Pu$ ratios of each particle size fraction were studied in each waste discharge area. Two-way analysis of variance of the plutonium soil size fraction data from each discharge area was used to detect differences between the horizontal and vertical distributions of $^{239,240}Pu$ and ^{238}Pu in these alluvial soils.

Statistically significant differences ($P < 0.01$) in the distribution of $^{239,240}Pu$ and ^{238}Pu were found in Mortandad and DP-Los Alamos canyon soils as a function of distance from the waste outfall. The $^{239,240}Pu/^{238}Pu$ ratios of the < 53 μm, 53–105 μm, 105–500 μm, and 2–23 mm size fractions in Mortandad canyon were generally higher in the separates collected 40 m from the waste outfall than elsewhere in the upper 2.56 km of this canyon (Fig. 2). Similarly, the plutonium ratios in DP-Los Alamos canyon size fractions were higher (Fig. 3) for samples collected at the waste outfall and at 2.56 and 5.12 km from the waste outfall than in the remainder of this canyon. Since the soils of Mortandad and DP-Los Alamos canyons received most of their ^{238}Pu after 1968,

Table 4—Particle size distribution data and total plutonium inventories of soil fractions (average of values from each soil depth per sampling location)

Distance from waste outfall and parameter studied	Size fraction					
	<53 μm	53–105 μm	105–500 μm	500–1,000 μm	1–2 mm	2–23 mm
Mortandad canyon						
100 m preoutfall						
Weight % soil	23	9.3	24	17	17	9.7
Percent plutonium in fraction	28	12	39	8.8	7.4	4.8
0–2.56 km						
Weight % soil	2.2	1.8	14	21	26	35
Percent plutonium in fraction	14	6.0	27	21	16	16
5.12 km						
Weight % soil	2.9	1.0	23	35	33	5.1
Percent plutonium in fraction	16	7.4	34	28	14	0.61
DP-Los Alamos canyon						
100 m preoutfall						
Weight % soil	2.0	2.0	24	34	27	11
Percent plutonium in fraction	60	9.5	11	11	7.3	1.1
0–20 m and 160–5,120 m						
Weight % soil	1.9	1.1	13	26	32	26
Percent plutonium in fraction	5.7	7.3	24	22	20	21
40–80 m						
Weight % soil	30	15	26	12	12	5.0
Percent plutonium in fraction	30	20	36	7.0	5.0	2.0
Acid-Pueblo canyon						
100 m preoutfall						
Weight % soil	0.72	0.85	8.4	18	26	46
Percent plutonium in fraction	7.0	6.0	26	22	27	12
0–5.12 km						
Weight % soil	3.0	3.0	16	26	28	24
Percent plutonium in fraction	7.0	7.0	31	19	17	19
10.2 km						
Weight % soil	0.60	1.4	16	31	26	25
Percent plutonium in fraction	4.4	5.6	35	19	22	14

soil samples from these canyons with relatively high ratios may represent: (i) the remains of soil present in the canyons prior to 1968, (ii) soils which have adsorbed only small amounts of the ^{238}Pu-rich wastes added to the canyons after 1968, or (iii) soils which have received relatively more radioactive wastes before 1968 than after this time due to relocation of the waste discharge pathway. The soil samples collected at the Mortandad 40-m locations may represent remnants of severely eroded portions of the channel alluvium. The soils located at the DP outfall also contained higher plutonium ratios than the rest of the post-outfall soils, probably due to a relocation of the waste discharge pathway at this location after 1968. The relatively high plutonium ratios found 2.56 and 5.12 km from the waste outfall in DP canyon probably reflect migration of plutonium into these locations from upper Los Alamos canyon, an area known to contain alluvium with $^{239,240}Pu/^{238}Pu$ ratios as high as 18 (Purtymun, 1971).

Statistically significant ($P < 0.05$) differences in the $^{239,240}Pu/^{238}Pu$ ratios of soil separates collected at various depths were found only in Mortandad canyon, the youngest of the three waste disposal areas. Higher $^{239,240}Pu/^{238}Pu$ ratios were found in the < 53 μm, 53–105 μm, 105–500 μm, and 2–23 mm particle size fractions with increasing depth (Fig. 5). Although only part of the plutonium ratio data for this canyon are presented in Fig. 5, most of the soil separates exhibited an approximate doubling of the $^{239,240}Pu/^{238}Pu$ ratio proceeding from the top to the bottom of the soil profile, due to larger concentrations of ^{238}Pu than $^{239,240}Pu$ in the surface layers of the soil. Thus, the large amounts of ^{238}Pu added to this canyon from 1968 to 1973 have not completely mixed with the alluvial soil separates deep in the soil profile, as they have in the the soils of the other waste disposal areas. This could be due to: (i) the fact that Mortandad canyon is a smaller watershed than the DP-Los Alamos or Acid-Pueblo drainage areas, resulting in less mixing and migration of soil and recently added plutonium during runoff events in Mortandad canyon than in the other study areas, and (ii) potential isotopic differences in behavior of $^{239,240}Pu$ and ^{238}Pu discussed previously.

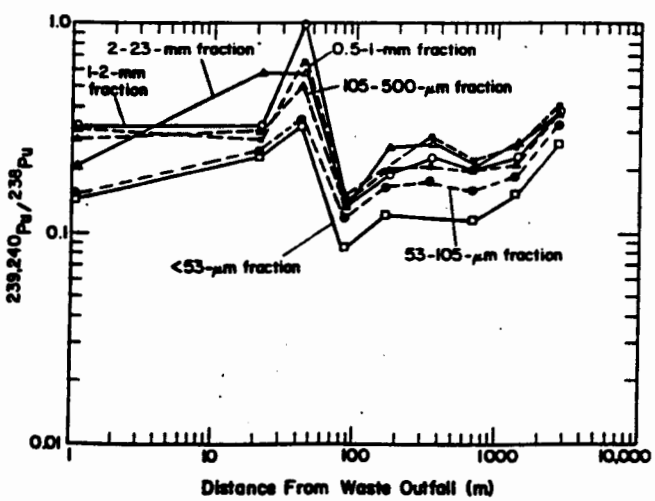

Fig. 2—Relationship of $^{239,240}Pu/^{238}Pu$ activity ratios of soil size fractions to distance from the waste outfall in Mortandad canyon.

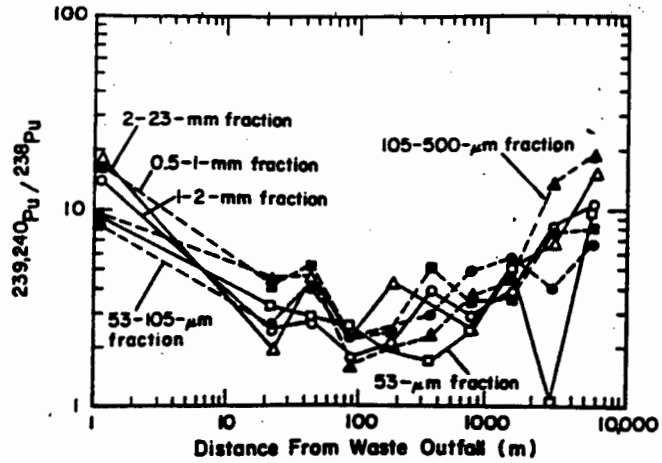

Fig. 3—Relationship of $^{239,240}Pu/^{238}Pu$ activity ratios of soil size fractions to distance from the waste outfall in DP-Los Alamos canyon.

The $^{239,240}Pu/^{238}Pu$ ratios of each particle size fraction from each canyon were compared to determine possible fractionation of plutonium isotopes on specific soil separates. Statistically significant differences in plutonium ratios between size fractions were detected only in Mortandad canyon soils ($P < 0.10$); relatively high and statistically similar $^{239,240}Pu/^{238}Pu$ ratios were found in the coarser size fractions (0.5–1.0 mm, 1–2 mm, and 2–23 mm) of Mortandad canyon soils, with progressively lower, statistically different ($P < 0.05$) plutonium ratios found in progressively smaller soil separates (Fig. 2). Thus, a larger portion of the more recently added ^{238}Pu was associated with smaller soil particles than larger particles in Mortandad canyon soils during the spring months. Although isotopic differences between ^{238}Pu and $^{239,240}Pu$ could result in a distribution pattern such as found in Mortandad canyon size fractions, a more comprehensive explanation would take into account the mean residence times of soil particles within each of the discharge areas. The distribution of plutonium isotopes observed in Mortandad canyon soils could also have occurred if large tuff fragments moved through this discharge area more

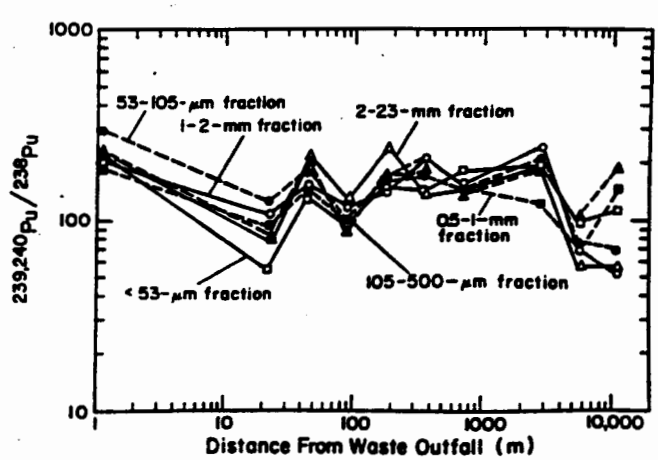

Fig. 4—Relationship of $^{239,240}Pu/^{238}Pu$ activity ratios of soil size fractions to distance from the waste outfall in Acid-Pueblo canyon.

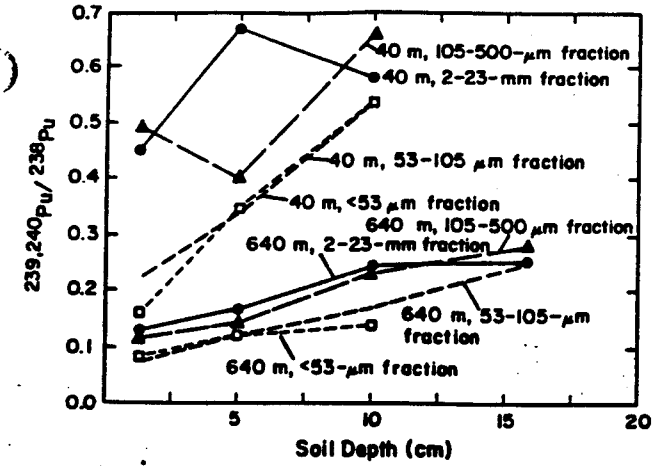

Fig. 5—Relationship of $^{239,240}Pu/^{238}Pu$ activity ratio of particle size fractions to soil depth for samples collected 40 and 640 m from the waste outfall in Mortandad canyon.

slowly than small soil particles, which would then more nearly reflect the $^{239,240}Pu/^{238}Pu$ ratios in the current effluents. In contrast, all the soil particles in the other discharge areas may have had similar residence times. This pattern may vary seasonally as the plutonium adsorbed to large and small soil particles is redistributed in turbulent late summer runoff events in these intermittent streams (Purtymun, 1974).

SUMMARY AND IMPLICATIONS

The ^{238}Pu and $^{239,240}Pu$ content of soil size fractions from three liquid effluent-receiving areas at Los Alamos was determined as a function of soil depth, distance from the waste outfall, and soil physical and chemical properties. Although the largest plutonium concentrations were found in size fractions < 105 μm, larger amounts of plutonium in whole soil samples were found in larger size fractions due to greater amounts of material in > 105 μm size fractions. Soil plutonium concentrations in the size fractions were correlated with surface area of soil particles and decreased with distance from the waste outfall and with soil depth in canyons currently receiving treated liquid wastes.

Soil size fractions were examined for differences in $^{239,240}Pu/^{238}Pu$ activity ratios as a function of distance from the waste outfall and soil depth. The differential distribution of plutonium isotopes was considered to be the result of the changing isotopic composition of the liquid wastes added to the canyons, subsequent movement of soil and plutonium within these intermittent streams, and potential isotopic and chemical differences in the behavior of ^{238}Pu and $^{239,240}Pu$.

Although some physical-chemical properties of the soil separates have been correlated with plutonium distributions, much additional research is needed in many ecosystems before the distribution of plutonium in soils is thoroughly understood. Although plutonium concentrations in the size fractions demonstrated specific horizontal and vertical distributional relationships, additional work is needed to determine seasonal changes in these patterns and in the movement of plutonium-containing soil particles. Changes in plutonium inventories at a specific location may occur due to the late summer runoff events, changes in environmental conditions and nature of the radioactive liquid wastes, biological activity in the sediments, and man's activities. Comparisons of the distribution of plutonium will have to be made across different ecosystems and chemical species present in the environment pathways to man can be knowledgeably interpreted and predicted.

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