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THE DISTRIBUTION OF PLUTONIUM AND CESIUM IN ALLUVIAL SOILS OF THE LOS ALAMOS ENVIRONS

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Abstract. The alluvial soils of three liquid waste disposal areas at Los Alamos were sampled to determine plutonium and cesium distributional relationships and correlations with soil physical-chemical properties. Radionuclide concentrations were determined for soil samples as a function of soil depth and distance from the waste outfall. The cesium-plutonium data were correlated with levels of organic carbon, carbonates, exchangeable and water-soluble cations, pH, cation exchange capacity, bulk density, surface area and geometric particle size of these soils. The distribution patterns of soil plutonium and cesium were also compared to the waste use history of the three study areas.

Key words: ^{137}Cs; *intermittent streams;* ^{238}Pu; ^{239}Pu; *radioactive wastes; radionuclide distributions; sediments; volcanic soils.*

INTRODUCTION

In spite of the apparent environmental persistence of plutonium and the projected accumulations of over 1000 megacuries of plutonium by 2020 from the nuclear power industry (Jacobs et al. 1969), little is known about the distribution and behavior of plutonium in the environment.

Although cesium kinetics and reactions have been studied in the soils of lake and river ecosystems (Carrigan et al. 1967, Kornegay et al. 1963, Pickering 1969), the behavior of cesium and plutonium in the alluvium of intermittent streams has received little attention (Hakonson et al. 1973, 1974). The distribution of radionuclides in this ecosystem at Los Alamos is described by Hakonson et al. at this syposium and is influenced by environmental factors and by the activities of man, unlike many pristine ecological study sites.

The objective of the present study was to evaluate the horizontal and vertical distribution of plutonium and cesium in the soils of three canyons which have received radioactive liquid wastes from the Los Alamos Scientific Laboratory. The radionuclide distributons were also studied as influenced by soil physical and chemical properties and canyon use history.

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, near Los Alamos, New Mexico (Fig. 1). The alluvium of the intermittent streams in Mortandad, DP and Acid Canyons 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 have stream channel widths of less than 1 m and alluvium depths of less than 0.15 m, stream channels in the lower canyon areas are about 3 m wide and 0.15-30 m deep at the sampling locations. Although Griggs (1964) characterized the hydrology and geology of these areas, little is known about the properties of the soils of the entire plateau.

The canyons have been used as disposal areas for plutonium- and cesium-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 to 1963 (2.1 ± 1.8 (SD) mCi plutonium/year). Treated wastes were discarged into Mortandad Canyon since 1963 (4.2 ± 2.3 (SD) mCi plutionium/year) and into DP-Los Alamos Canyon since 1953 (1.6 ±1.2 (SD) mCi plutonium/year). No long-term data exists for the amounts of cesium added to these intermittent streams in the liquid wastes.

Sampling and Analytical Methods

Ten 30-cm soil cores were collected along the center of the stream channel at 30-cm intervals

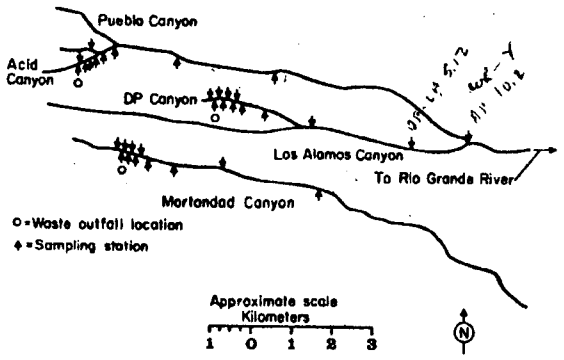

Fig. 1. Location of sampling stations in canyon study areas.

at each of 33 permanent sampling stations located in each canyon during May-July, 1973 (Fig. 1). Samples were taken 100 m above the waste outfall and at distances of 0, 20, 40, 80, 160, 320, 640, 1280, 2560, 5120, 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 device (2.4 cm i.d., schedule 89 pipe with a sharpened end), placed in a plastic bag, frozen, and cut into 0-2.5, 2.5-7.5, 7.5-12.5, and 12.5-30 cm sections. Five of the soil cores from each sampling location were composited by depth and assayed for their ^{137}Cs, ^{238}Pu and $^{239,240}Pu$ (the term Pu used herein designates total plutonium) content as previously described (Hakonson et al. 1973). A second composite was made of the remaining five cores which were analyzed for levels of exchangeable and water-soluble cations (sodium, potassium, magnesium, and calcium), calcium carbonate, and organic carbon, as well as cation exchange capacity, pH, bulk density and particle size distributon (U.S. Department of Agriculture 1972). The geometric mean particle size diameter and the surface area of the samples was determined from the particle size distribution data as described by Gardner (1956).

Correlation coefficients (r) were designated as representing a "significant" correlation at the 5% level of significance.

RESULTS AND DISCUSSION

Horizontal and Vertical Distributons of Radionuclides

Plutonium and cesium-137 have accumulated with time in the alluvial soils of the three waste discharge areas to concentrations greater than worldwide fallout levels of up to 0.1 pCi Pu/g and 1 pCi ^{137}Cs/g found in New Mexico soils (Herceg 1972 and 1973, Kennedy et al. 1971, Schiager et al. 1974). In general, maximum concentrations of plutonium (Table 1) and cesium (Fig. 2) occurred in the upper canyon areas, usually within 2.56 km from the point of waste discharge. Fallout levels of cesium were found in Mortandad and Acid-Pueblo soils 5.12 and 10 km from the waste outfall, and at the pre-outfall sampling locations in DP-Los Alamos and Acid-Pueblo Canyons. Levels of cesium and/or

TABLE 1. Total plutonium concentrations of alluvial soils of the waste disposal areas

Distance from waste outfall	Total plutonium concentration (pCi/g)			
	0-2.5	2.5-7.5	7.5-12.5	Remainder[a]
	Mortandad Canyon			
-100 m[b]	2.69	0.771	0.117	
0	129	274	390	
20 m	158	189	70.9	
40 m	259	234	116	
80 m	61.6	33.3	16.9	
160 m	104.	84.6	12.5	
320 m	70.8	59.0	27.4	6.68
640 m	32.7	31.4	31.3	
1.28 km	13.8	18.2	13.0	9.64
2.56 km	--[c]	9.04	6.97	2.26
5.12 km	0.114	0.079	0.065	0.105
	DP-Los Alamos Canyon			
-100 m	0.036	0.036	0.044	0.051
0	957	1640		
20 m	24.8	16.4	2.63	
40 m	18.2	--	11.4	0.488
80 m	--	10.4	1.87	0.831
160 m	0.332	2.25	0.369	0.328
320 m	0.196	0.252	0.225	2.34
640 m	0.344	0.481	0.445	
1.28 km	0.864	0.878	0.644	1.78
2.56 km	0.183	0.0904	0.114	
5.12 km	0.599	0.186		
	Acid-Pueblo Canyon			
-100 m	0.122	0.137		
0	16.6	8.52		
20 m	16.8			
40 m	5.78	8.72	11.4	28.7
80 m	6.21	6.60	21.1	505
160 m	8.61	10.1	--	20.4
320 m	8.28	7.92	10.8	12.3
640 m	7.86	12.4	10.4	19.1
2.56 km	36.3	369	2250	
5.12 km	1.39	--	0.617	1.65
10.2 km	0.401	0.518	0.435	

[a] The depth of the remainder section varied from 12.5 to 30 cm maximum.

[b] Negative distances represent background locations upstream from the waste outfalls.

[c] Missing data.

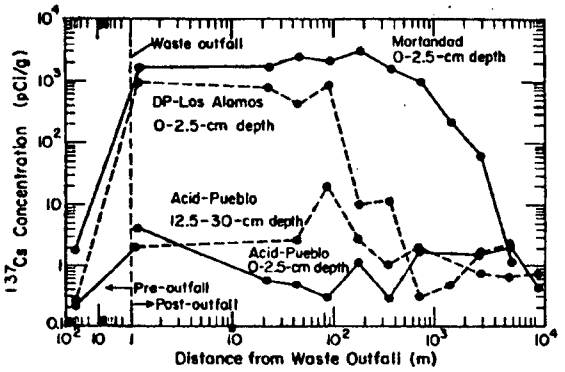

Fig. 2. Cesium-137 in alluvial soils of canyon study areas.

plutonium found in the soils 5.12 and 10 km from the DP-Los Alamos and Acid-Pueblo outfalls and at the pre-outfall station in Mortandad Canyon were greater than fallout levels of these radionuclides. Although the latter case represented a small pre-outfall source of contamination in Mortandad Canyon, the radionuclide levels observed in the other two canyons represented translocation of small amounts of laboratory wastes as far as 10 km away from their point of discharge.

The concentrations of cesium and plutonium found in the soils were highly correlated with distance from the outfall in each of the canyon study areas (Table 2). Concentrations of plutonium and cesium decreased exponentially with distance from the waste outfall in Mortandad Canyon. Concentrations of plutonium and ^{137}Cs were significantly correlated with the inverse of distance from the outfall in DP-Los Alamos Canyon. Plutonium concentrations in Acid-Pueblo soils were also hyperbolically related to distance form the waste outfall. Except for a few residual pockets of cesium contamination in Acid-Pueblo Canyon soils, very little ^{137}Cs above fallout levels was detected in this canyon, resulting in a poor cesium-distance correlation.

The vertical distributions of plutonium and cesium were studied by evaluating their concentrations relative to the highest concentration observed at each sampling location, to minimize horizontal distributional effects (Table 1 and Fig. 2). Exponential decreases in plutonium and ^{137}Cs were found with increased soil depth in Mortandad canyon at pre-outfall station and 40-320 and 1280-2560 m from the outfall (Table 2). Plutonium and cesium-137 concentrations decreased exponentially with depth for soils located 20-80 m from the waste outfall in DP-Los Alamos Canyon (Table 2). No significant correlations of radionuclides with depth were found at the rest of the sampling locations in these two study areas because radionuclide concentrations were similar at all soil depths to 30 cm, i.e., vertical mixing-migration of radionuclides was optimized. No statistically significant radionuclide-depth correlations were found in Acid-Pueblo Canyon, because the plutonium (Table 1) and ^{137}Cs (Fig. 2) concentrations tended to increase abruptly with depth when a residual pocket of contamination occurred in the soil profiles, such as at the 80 m location.

The horizontal and vertical distributions of ^{238}Pu and ^{239,240}Pu were compared with one another for soil samples taken in each canyon to study the rates of mixing of the individual isotopes as a function of canyon use history. Since the first appreciable amounts of ^{238}Pu appeared in laboratory liquid wastes in 1968, the horizontal-vertical distribution of the ^{239,240}Pu/^{238}Pu ratio can be used to indicate mixing-migration rates of plutonium in soils (Table 3). Relatively complete mixing of ^{238}Pu into the soils in less that 5 years was indicated by a ^{239,240}Pu/^{238}Pu activity ratio of 0.26 ± 0.16 (SD) and 5.2 ± 4.5 (SD) for Mortandad (n = 27) and DP-Los Alamos (n = 29) samples collected as deep as 30 cm and as far as 2560 m from the outfalls. These ratios were not significantly different from the ratios in the treatment plant effluents in 1972 and 1973 (Schiager et al. 1974). It is noteworthy that the soils 40-80 m from the DP waste outfall have Pu ratios similar to the rest of the canyon, a distribution which took place in less than a year since the effluent discharge pathway was redirected to these relatively terrestrial locations in early 1972.

A limited amount of data indicate that about 60 ± 25% (SD) of the plutonium in the liquid wastes was in a "particulate" form (did not pass a 0.45 μm filter) when added to the canyons and that nearly all of the ^{137}Cs was soluble in the liquid wastes. Despite these differences in chemical form, a significant ^{137}Cs-Pu correlation was found for post-outfall soils in each study area. Whereas the log of the plutonium concentrations was directly correlated with plutonium levels in Mortandad Canyon (r = 0.94, n = 33), plutonium concentrations were exponentially related to soil cesium levels in DP-Los Alamos (r = 0.88, n = 31) and in Acid Canyon (r = 0.92, n = 22). Thus, in spite of any initial differences in "solubility" of radioisotopes in the waste solutions and any temporal effects on soil radionuclides, ^{137}Cs and plutonium concentrations seem to be correlated with one another and with distance from the outfall, a fact that might imply that similar mechanisms control the distribution of each isotope in the environ-

TABLE 2. Statistical correlations of soil radionuclide concentrations with spatial relationships and physical-chemical properties of the soils of the waste discharge areas

Dependent variable: radionuclide concentration (pCi/g soil)	Independent variable	Correlation coefficient	Locations of samples used in comparison
	Radionuclide-spatial correlations		
Mortandad Canyon			
log ^{137}Cs	distance from outfall	-0.94	0-5.12 km[a], n = 33
log Pu	distance from outfall	-0.93	0-5.12 km[a], n = 33
log ^{137}Cs	soil depth	-0.79	-100 m, 40-320 m, 1.28-2.56 km, n = 20
log Pu	soil depth	-0.76	-100 m, 40-320 m, 1.28-2.56 km, n = 20
DP-Los Alamos Canyon			
^{137}Cs	inverse of distance from outfall	0.66	0-5.12 km, n = 31
Pu	inverse of distance from outfall	0.96	0-5.12 km, n = 31
log ^{137}Cs	soil depth	-0.87	20-80 m, n = 9
log Pu	soil depth	-0.91	20-80 m, n = 9
Acid-Pueblo Canyon			
Pu	inverse of distance from outfall	0.94	0-10.2 km, n = 31
	Radionuclide-soil property correlations		
Mortandad Canyon			
log ^{137}Cs	geometric mean particle diameter	0.71	0-5.12 km, n = 33
log Pu	geometric mean particle diameter	0.70	0-5.12 km, n = 33
log ^{137}Cs	log cation exchange capacity	0.50	0-2.56 km, n = 29
log ^{137}Cs	log % $CaCO_3$	0.80	0-5.12 km, n = 33
log ^{137}Cs	Exchangeable Ca percentage	0.82	0-5.12 km, n = 33
DP-Los Alamos Canyon			
log ^{137}Cs	log cation exchange capacity	0.67	0-5.12 km, n = 31
Pu	inverse of % organic carbon	0.75	0-5.12 km, n = 31
log ^{137}Cs	log % $CaCO_3$	0.75	0-5.12 km, n = 31
log ^{137}Cs	exchangeable Ca percentage	0.74	0-5.12 km, n = 31
Acid Pueblo Canyon			
log Pu	cation exchange capacity	0.82	0-10.2 km, n = 31
log ^{137}Cs	cation exchange capacity	0.70	0-10.2 km, n = 31
log Pu	% organic carbon	0.57	0-10.2 km, n = 31

[a]Represents distance from waste outfall; negative distances represent background locations upstream from waste outfall.

ment. However, complications arise with this interpretation if the annual release rate of ^{137}Cs has varied greatly in the liquid wastes, a variable which cannot be evaluated due to the absence of ^{137}Cs determinations on the waste solutions dumped during the histories of the canyons. If the assumptions are made that (1) the levels of ^{137}Cs in Acid-Pueblo Canyon soils were comparable to the present cesium levels in the other two canyon systems when the use of Acid Canyon ceased 12 years ago (as evidenced by the level of 22 pCi ^{137}Cs/g found in the 80 m soil in Fig. 2) and (2) annual release rates of cesium were constant (as supported by data in Schiager et al. 1974), then the distributive mechanisms for cesium and plutonium could be different, i.e. - most of the cesium has disappeared and relatively more plutonium has persisted in Acid-Pueblo Canyon soils (Table 1, Fig. 2).

TABLE 3. Ratios of $^{239,240}Pu/^{238}Pu$ in alluvial soils of waste disposal areas

Distance from waste outfall	Soil depth (cm) 0 - 2.5	2.5 - 7.5	7.5 - 12.5	Remainder[a]
Mortandad Canyon				
-100 m[b]	20	13	3.5	
0	0.16	0.16	0.44	
20 m	0.25	0.36	0.30	
40 m	0.33	0.84	0.66	
80 m	0.12	0.13	0.13	
160 m	0.15	0.17	0.25	
320 m	0.16	0.20	0.31	0.25
640 m	0.12	0.16	0.24	
1.28 km	0.17	0.18	0.19	0.33
2.56 km	--	0.24	0.28	0.50
5.12 km	1.1	4.2	1.7	1.9
DP-Los Alamos Canyon				
-100 m	1.4	0.93	0.55	0.49
0	15	14		
20 m	4.9	4.3	0.41	
40 m	4.1	--	3.0	2.0
80 m	--	2.9	2.0	1.9
160 m	3.7	11	1.6	2.0
320 m	2.0	3.1	1.9	11
640 m	2.7	2.9	3.1	
1.28 km	3.4	3.2	3.0	8.5
2.56 km	17	11	4.5	
5.12 km	0.39	11		
Acid-Pueblo Canyon				
-100 m	1.2	0.56		
0	390	200		
20 m	83			
40 m	210	200	190	95
80 m	140	130	76	49
160 m	130	170	--	190
320 m	190	180	190	170
640 m	130	140	140	126
2.56 km	230	163	82	
5.12 km	120	--	18	43
10.2 km	81	100	62	

[a] The depth of the remainder section varied from 12.5 to 30 cm maximum.

[b] Negative distances represent background locations upstream from the waste outfalls.

Correlations of Soil Properties with Radionuclide Distributions

The relationship between soil plutonium and cesium concentrations and selected soil physical properties was studied for each canyon. Bulk density was compared with radionuclide distributions because this soil physical property reflects differences in particle densities, particle size distributions and soil compaction, soil characteristics which influenced the transport of isotopes in aquatic ecosystems (Kornegay et al. 1963). Whereas the tuff-derived soils of Mortandad, DP and Acid Canyon soils exhibited bulk densities of 1.3-1.5 g/cm^3, the soils of lower Pueblo and Los Alamos Canyons, demonstrated bulk densities greater than 2 g/cm^3, due to their composition of relatively dense latites and granitic materials. No statistical correlations were detected between bulk density and radionuclide levels because a large proportion of the samples exhibited bulk densities of 1.3-1.5 g/cm^3 and high radionuclide concentrations. Although bulk density was inversely related to soil concentrations of ^{137}Cs and plutonium, this trend was not influential in the distribution of radionuclides within soils formed from similar parent materials.

An attempt was made to relate geometric mean particle size and surface area to radionuclide levels in the alluvium (Table 4). The geometric mean particle size of an alluvial soil is related to watershed size, nature of the suspended materials transported in runoff events and degree of weathering of the soil materials, which are important variables in understanding radionuclide transport in aquatic ecosystems (Kornegay et al. 1963). The log of the concentration of either Pu or ^{137}Cs was positively correlated with geometric mean particle size diameter in Mortandad canyon soils (Table 2). This relationship is a result of the relatively coarse nature of the tuff particles in the upper reaches of Mortandad Canyon (Table 4) as well as a relationship of Pu-^{137}Cs to distance from the outfall. Although no statistically significant radionuclide-particle or radionuclide-surface area correlations were found when the soils data from the other two canyons was analyzed for all sampling locations, particle sizes were inversely related to vertical distributions of plutonium concentrations in Acid-Pueblo (2.56 km) and DP-Los Alamos (80 m) Canyons. Surface area of the soil samples was directly related to plutonium levels at these same locations (Table 4).

Absorption and exchange reactions of radionuclides with soil colloidal complex influenced the natural distribution and behavior of cesium and other isotopes (Carrigan et al. 1967, Kornegay et al. 1963, Pickering 1969). Since the soil colloidal complex consists of inorganic and organic constituents, the soil cation exchange capacity (CEC) and organic carbon content was compared with the ^{137}Cs and plutonium content of the study area soils. Increases in the log of CEC were significantly correlated with elevated levels of the log of plutonium (Fig. 3) and

TABLE 4. Selected physical properties of soils of the waste disposal areas

Distance from waste outfall	Geometric mean particle diameter (mm)	Percent soil (by weight) in <53 μm fraction	Surface area (cm²/g)
	Mortandad Canyon		
0	3.1 (1.1)[a]	2.2 (1.1)	49 (19)
20 m	1.8 (0.35)	1.9 (0.90)	51 (12)
40 m	1.4 (0.15)	2.6 (0.71)	60 (7.6)
80 m	1.7 (0.51)	3.4 (2.2)	69 (31)
160 m	1.5 (0.44)	2.6 (1.4)	65 (25)
320 m	1.5 (0.05)	2.3 (1.7)	59 (21)
640 m	1.4 (0.38)	0.75 (0.42)	42 (10)
1.28 km	0.97 (0.11)	1.3 (1.1)	56 (12)
2.56 km	0.89 (0.18)	2.8 (1.2)	78 (21)
5.12 km	0.79 (0.11)	2.9 (1.7)	82 (21)
	DP-Los Alamos Canyon		
0	2.0 (0.70)	8.6 (6.0)	120 (72)
20 m	0.91 (0.27)	6.6 (2.8)	120 (44)
40 m	0.43 (0.42)	24 (17)	350 (210)
80 m	0.10 (0.05)	38 (12)	530 (140)
160 m	1.1 (0.09)	1.1 (0.36)	49 (6.7)
320 m	1.1 (0.13)	0.58 (0.37)	41 (7.8)
640 m	1.1 (0.10)	0.48 (0.50)	43 (5.4)
1.28 km	1.1 (0.10)	1.0 (0.77)	46 (9.6)
2.56 km	1.6 (0.21)	0.83 (0.57)	42 (9.9)
5.12 km	2.4 (0.92)	0.80 (0.86)	36 (20)
	Acid-Pueblo Canyon		
0	1.0 (0.10)	1.5 (1.1)	60 (17)
20 m	1.2 (0.21)	1.8 (1.3)	60 (22)
40 m	0.95 (0.22)	2.4 (2.7)	75 (38)
80 m	1.1 (0.19)	2.3 (1.8)	69 (32)
160 m	1.1 (0.13)	2.1 (2.0)	64 (23)
320 m	1.2 (0.10)	0.95 (0.68)	44 (7.1)
640 m	1.2 (0.20)	1.5 (0.82)	53 (13)
2.56 km	0.83 (67)	14 (12)	230 (170)
5.12 km	1.1 (0.21)	0.95 (0.71)	53 (9.5)
10.2 km	1.0 (0.15)	0.75 (0.40)	48 (5.4)

[a] Number in parentheses represents one standard deviation of the mean value observed at all soil depths per location.

^{137}Cs (Table 2) in Mortandad and DP-Los Alamos Canyons, whereas the plutonium and cesium levels of Acid-Pueblo soils were exponentially related to CEC (Table 2). Since cation exchange capacity was directly related to percentages of fine particles (less than 53-μm diameters) in these soils, these CEC-radionuclide relationships are related to the ability of the soil inorganic phases to absorb radionuclides. The levels of soil ^{137}Cs were insensitive to fluctuations in organic carbon content, but plutonium concentrations were exponentially related to levels of soil organic carbon in Mortandad (Fig. 4) and Acid-Pueblo (Table 2) Canyons and hyperbolically related to organic carbon in DP-Los Alamos Canyon (Table 2). Thus, specific reactions may have occurred between plutonium and the organic constituents of the alluvial soils independent of CEC reactions. This observation could be partially explained by the fact that algae, a constituent of the soil organic carbon complex, are known to concentrate Pu in these ecosystems (Hakonson et al. 1974).

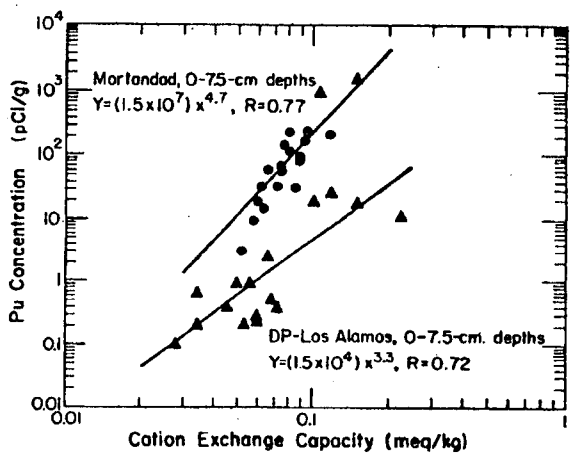

Fig. 3. Relationship of soil cation exchange capacity to levels of plutonium in alluvial soils of Mortandad (n = 33) and DP-Los Alamos (n = 31) Canyons.

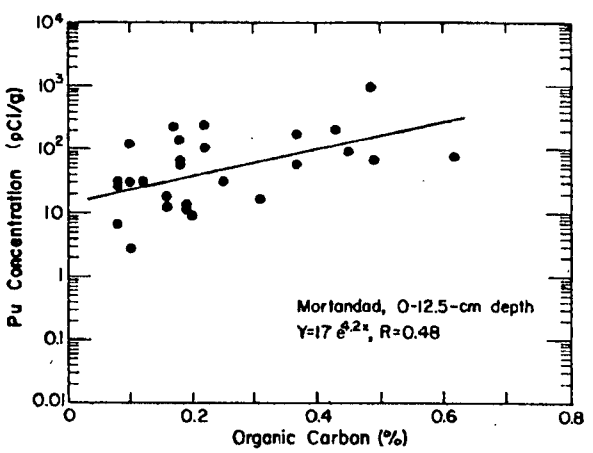

Fig. 4. Relationship of organic carbon content to levels of plutonium found in alluvial soils of Mortandad Canyon (0-2.56 km from waste outfall, n = 29).

In the process of treating the low level radioactive wastes, sodium carbonate, calcium hydroxide, and ferric sulfate are added to the liquid wastes to concentrate plutonium activity in a ferric hydroxide floc at a relatively high pH. The resulting supernatant, containing low levels of Pu and ^{137}Cs, is then filtered and discharged to the canyon. Thus, we investigated the influence of the nonradioactive chemicals in the wastes on soil radiouclide distributions for the soils of the waste discharge areas.

An attempt was made to relate plutonium and cesium concentrations to the pH and levels of carbonates in the canyon soils. Whereas the average soil pH for the soils of the upper 1.28 km of Acid-Pueblo Canyon was 7.5 (range of 6.4 to 8.2), the average pH of soils within this distance of the Mortandad and DP-Los Alamos outfalls was 8.9 (range of 8.0 to 9.7, reflecting substantial additions of sodium and carbonates to the canyons in the liquid wastes (Schiager et al. 1974). Although no significant pH-radionuclide correlations were detected in any canyon, the log of either the plutonium (Fig 5) or ^{137}Cs concentrations (Table 2) were significantly correlated with log of calcium carbonate levels in Mortandad and DP-Los Alamos Canyons. Radionuclide concentrations were not significantly correlated with carbonate levels in Acid-Pueblo Canyon because most of the soil samples contained background levels of carbonates and ^{137}Cs, making a statistically-significant relationship nondetectable. However, the samples collected deep in the soil profiles in Acid Canyon did demonstrate high levels of carbonates where residual pockets of radionuclides were found.

The carbonate-radionuclide correlation found in these alluvial soils has significant implications in the fields of environmental monitoring and radioecology. This correlation has application to environmental monitoring since the extent of potential radionuclide migration down the canyon can be determined in any location by testing for soil carbonates. This is also important in the radioecology of this ecosystem, since plutonium carbonate complexes are more "soluble" (for potential plant uptake) or mobile (in potential soil migration) than plutonium oxide-hydroxide compounds (Andelman et al. 1970, Wick 1967) potentially present in other ecosystems. This relationship also suggests a radionuclide distribution or retention mechanism involving both ion exchange processes described above and carbonates.

The percent of the exchange complex saturated with Ca, Mg, K and Na was calculated, since the amounts and types of cations on the soil cation exchange complex and in soluble form affect the behavior and distribution of radioactive cations in soils (Wahlberg et al. 1962). Cesium (Table 2) and plutonium (Fig. 6) concentrations in the alluvium were exponentially correlated with exchangeable calcium percentages in Mortandad and DP-Los Alamos Canyon soils. This significant correlation was to be expected due to the contributions of calcium from calcium carbonate present in the soils. Plutonium and cesium levels in the alluvium were insensitive to fluctuations in exchangeable or water-soluble magnesium and potassium.

Levels of exchangeable and soluble sodium were correlated with the distribution of ^{137}Cs and Pu in Mortandad Canyon soils. This relationship was not

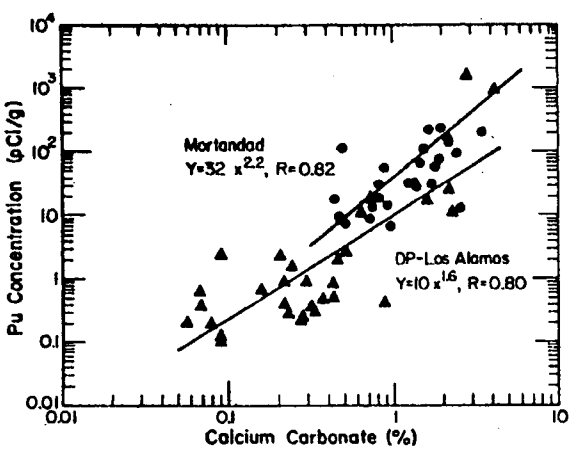

Fig. 5. Relationship of calcium carbonate content to levels of plutonium in Mortandad (n = 33) and DP-Los Alamos (n = 31) Canyons.

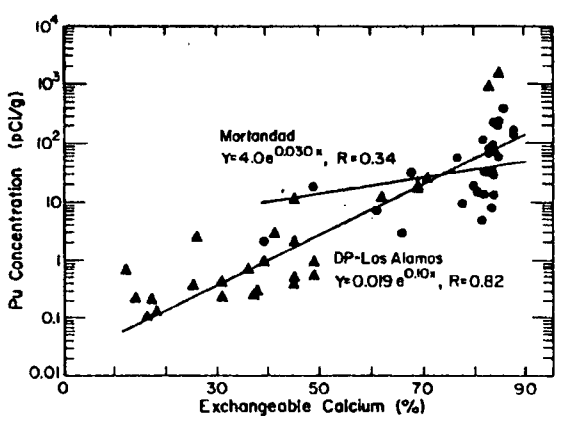

Fig. 6. Relationship of exchangeable calcium percentage to levels of plutonium found in alluvial soils of Mortandad (n = 20) and DP-Los Alamos (n = 26) Canyons.

detected in the soils of the other study areas, because of the smaller amounts of sodium added to these canyons and their larger drainage areas relative to Mortandad Canyon. Using cesium for the example, the mass-action equation for this reaction is:

$$[CsX] + [Na^+] \rightleftharpoons [NaX] + [Cs^+]$$

where:

[CsX] = concentration of the cesium adsorbed on the soil
[NaX] = concentration of the sodium adsorbed on the soil
$[Cs^+]$ = concentration of cesium in solution
$[Na^+]$ = concentration of sodium in solution

Thus, during a discharge of liquid wastes, ^{137}Cs adsorbed on the canyon soils may be displaced by soluble sodium in the waste solution and levels of exchangeable sodium gradually increase. When the soluble sodium were normalized to levels of exchangeable sodium, using the ratio of $[NaX][Na^+]$, plutonium and cesium (Fig. 7) concentrations in Mortandad samples were found to be exponentially correlated to this sodium ratio (r = 0.85 and 0.84 for Pu and ^{137}Cs, n = 33). This correlation resulted because of the high soluble sodium concentrations found close to the waste outfall and the infrequent migration of wastes past the 1.28 km sampling station.

Although several physical-chemical properties of the soils have been correlated with distribution patterns of plutonium and cesium, much additional research is needed before the distribution of these radioisotopes in soils is thoroughly understood. Much of the variation in plutonium and cesium concentrations was not explained by variations in bulk density, geometric mean particle size and surface area in these alluvial soils. Whereas soil chemical properties such as organic carbon content were correlated only with levels of plutonium, cation exchange capacity, carbonate concentrations and levels of exchangeable calcium were correlated with plutonium as well as ^{137}Cs concentrations. Turbulent mixing and transport of alluvium and radionuclides during late summer runoff events also influence radionuclide distributions, as observed by Purtymun (1974) and by the plutonium ratios presented in Table 3. The biological properties of the alluvium should also be considered in future research as well as the distribution of these radionuclides and americium in the stream bank soils. The influence of man's activities on the migration of plutonium and cesium

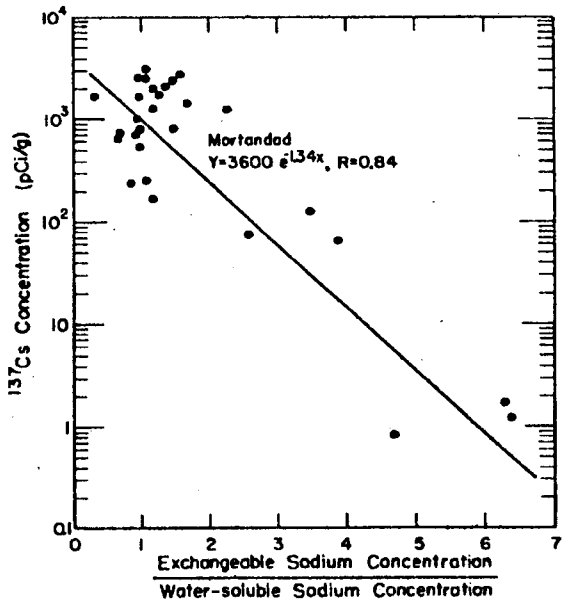

Fig. 7. Influence of exchangeable/water-soluble sodium concentrations on cesium levels in alluvial soils of Mortandad Canyon (n = 22).

in the outfall soils should also be examined, i.e. - chelating agents have been discharged into the canyons during cooling-tower cleaning operations, which could make plutonium more mobile in the alluvium. Comparisons of the distribution and behavior of plutonium and cesium will have to be made across different ecosystems and soil types before environmental pathways to man can be knowledgeably interpreted and predicted.

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