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CESIUM-137 AND PLUTONIUM IN LIQUID WASTE DISPOSAL AREAS AT LOS ALAMOS

T. E. Hakonson and K. V. Bostick

Los Alamos Scientific Laboratory, Los Alamos, New Mexico 87544

Abstract. The results of an ecological investigation of plutonium and ^{137}Cs are described for three canyons which have been used as liquid waste disposal areas at Los Alamos. Stream channel sediments were identified as the major reservoir of waste radioactivity based on the relatively high radionuclide concentrations measured in this component. Hydrological sediment transport processes in the respective canyons play a major role in the downstream movement of radioactivity. Statistically significant correlations between plutonium and ^{137}Cs in sediments downstream from the waste outfalls support a physical transport mechanism in the redistribution of sediment radioactivity. Substantially higher plant:sediment activity ratios were calculated for plutonium in the canyons than corresponding ratios derived by others through laboratory experimentation. Significant positive correlations between plutonium and ^{137}Cs concentrations in vegetation were observed in two canyons which currently receive wastes but not in the third canyon which has not received wastes for 10 years. Highest average concentrations of plutonium in small ground-dwelling rodents living adjacent to the contaminated streams were observed in lung and pelt tissue, indicating that resuspension is likely a major mechanism in the contamination of the rodents.

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Key words: Cesium-137; plutonium; radioecology; radioactive waste; plutonium uptake; cesium uptake; radionuclide transport.

INTRODUCTION

The Los Alamos Scientific Laboratory (LASL) has investigated various aspects of the nuclear energy field for over 30 years in a great number of fabrication and experimental facilities scattered over the Laboratory site. Liquid waste streams from some of these facilities contained low-level amounts of radionuclides such as plutonium and cesium, which after collection and treatment were released at about 0.5% of the maximum permissible concentration (ICRP 1962) to specific areas in the surrounding environment.

A discussion of environmental plutonium behavior, based on the few pertinent open literature publications, appears in two recent reviews (Hakonson 1974, Hanson 1974). It is evident that plutonium behavior in the environment has received little attention, especially in freshwater and terrestrial ecosystems. There are several aspects of plutonium movement through the environment which warrant investigation, especially the phenomena of adsorption, absorption, biological accumulation, trophic level increase or decrease, seasonal and long term variations, and how all of these factors vary with ecosystem type. Our approach at investigating some of these factors has been to compartmentalize the ecosystems under study. Such an approach required, as a first step, the identification of plutonium receptors in the ecosystems. This paper presents the results of a radioecological survey conducted in

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October and November 1972 in three canyons which received liquid wastes for various periods over the last 32 years. The primary objective of the study was to document the ^{238}Pu, ^{239}Pu, and ^{137}Cs levels in several canyon ecosystem components as a function of distance below waste discharge areas in order to identify some of the ecosystem components of importance in radionuclide redistribution. In the course of study, data were gathered on radionuclide concentration variability, plutonium and ^{137}Cs isotopic relationships, and the influence of canyon use history on radiocontaminant behavior. A small portion of the data appearing in this paper was presented elsewhere (Hakonson et al. 1974).

METHODS AND MATERIALS

Study Area

The Laboratory is located on the east flank of the Jemez Mountains, on a portion of the Pajarito Plateau, in north-central New Mexico. The plateau, which has pronounced elevational gradient in the east-west direction, was formed by successive ash flows from a volcanic area 32 km to the west. The Laboratory area has a semi-arid, continental mountain climate; precipitation averages about 46 cm at the higher elevations (~ 2200 m) on the west and about 18 cm at the lower elevations (~ 1650 m) on the east. Rainfall accounts for about 75% of the annual precipitation. Drainage from the 113 km^2 Laboratory site is via the many canyons which bisect the plateau.

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Nearly all of the liquid wastes generated by the Laboratory since its beginning in 1943, were collected by industrial waste lines, treated (since 1951) and released into one of three semi-drainage canyons (Fig. 1). Acid-Pueblo (AP) Canyon received untreated liquid waste from 1943 to 1964. The treatment facility was subsequently decommissioned and dismantled and AP Canyon has not received liquid wastes for about 10 years. DP-Los Alamos (DP) Canyon currently receives effluent from a treatment plant as it has since 1952. It is anticipated that this facility will be decommissioned within the next few years. Mortandad Canyon has received effluent from a waste treatment facility since 1963. Its use as a disposal area is expected to continue into the future.

In general, the physical features of the three canyons are similar. Near the waste outfalls at elevations of about 2200 m, the stream channels are narrow (< 1 m), rocky, and contain relatively thin layers of sediment (< 30 cm). The stream channels decrease about 200 m in elevation within 2000 m of the respective outfalls, where they widen to 2-3 m and the alluvium generally increases in depth (i.e. from 30 cm up to 35 m). In Mortandad Canyon alluvial deposits increase in depth abruptly, whereas in DP and AP Canyon, the increase is slight until after the two canyons join. The AP and DP Canyon systems join about 10000 m below the DP Canyon waste outfall and continue as one canyon for an additional 6000 m before entering the Rio Grande (Fig. 1). Mortandad Canyon, on the other hand, does not link with any major canyon on its path to the Rio Grande.

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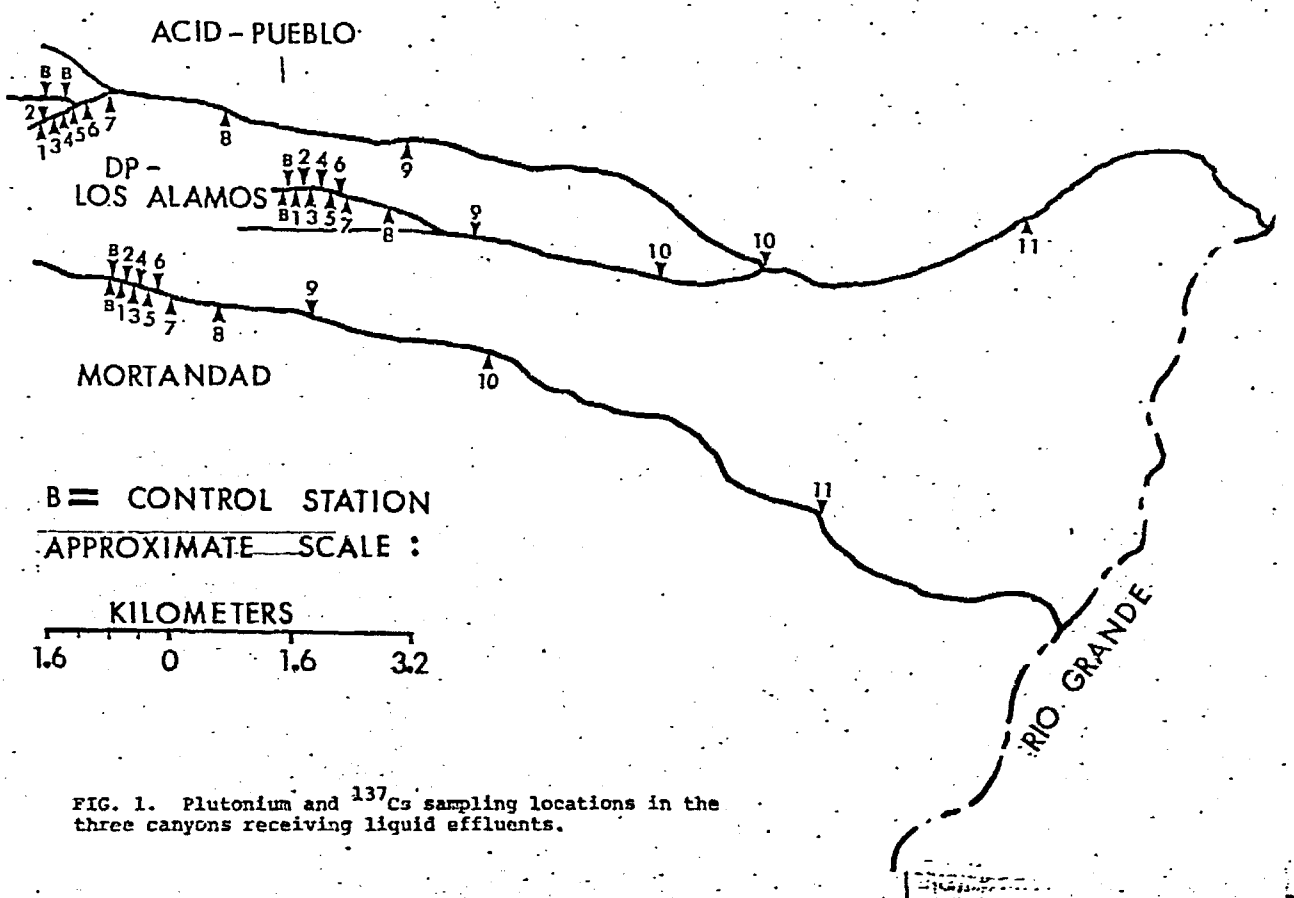

FIG. 1. Plutonium and ^{137}Cs sampling locations in the three canyons receiving liquid effluents.

Surface water exists in portions of all three canyons as a result of Laboratory effluents and/or domestic sewage, but these flows occur only in small segments of the canyons near the outfall areas. Relatively large flows occur in all three canyons during storm runoff events. Storm runoff reaches the Rio Grande in the AP-DP Canyon system whereas the runoff water rapidly soaks into the thick alluvial deposits in Mortandad Canyon and seldom reach post-outfall distances exceeding 3 km.

A detailed chemical characterization of the liquid wastes currently released to DP and Mortandad Canyons has not been attempted. We know that the wastes are alkaline (pH averages about 11) as a consequence of the treatment process and that large, but unquantified amounts of chelating agents such as EDTA are present in the effluent. Volumes of effluent entering the canyons average about 2×10^5 l/d into Mortandad Canyon and 2×10^4 l/d into DP Canyon. Releases into Mortandad Canyon occur on a daily basis, whereas released into DP Canyon are made about once a week. The effluents rapidly soak into the alluvium below the waste outfalls and disappear underground about 800 m post-outfall in Mortandad Canyon and about 80 m post-outfall in DP Canyon.

Available treatment plant records were used in estimating the amounts of plutonium released to the three canyons (Hakonson et al. 1973). Plutonium additions to AP Canyon were estimated as 170 mCi from 1943-1964. About 80% of this quantity was released prior to 1950 and virtually all of the plutonium was $^{239-240}Pu$ (hereafter referred to as ^{239}Pu). An unknown amount of radioactive soils/sediments were removed from AP Canyon in the immediate area of the

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outfall during decontamination and cleanup of the treatment plant in 1964. DP Canyon received an estimated 32 mCi through 1972 and, at the present time, the $^{239}Pu/^{238}Pu$ activity ratio in the effluent measures about 1, although most of the plutonium released to this canyon was ^{239}Pu. The release into Mortandad Canyon was about 42 mCi through 1972, and since 1968 most of the Pu on an activity basis was ^{238}Pu. Prior to 1968, nearly all of the plutonium released to DP and Mortandad Canyon was ^{239}Pu since ^{238}Pu was not handled in quantity at the Laboratory until 1968.

Total ^{137}Cs additions to the canyons could not be estimated with available data. During 1973, about 300 mCi were released to Mortandad Canyon and 1 mCi to DP Canyon. Of the three canyons, it is thought that Mortandad Canyon received the largest total amounts of this nuclide.

Sampling

A sampling network was established in the canyons during the summer of 1972 (Fig. 1) at two locations above the waste discharge outfalls to serve as control areas and also at 0, 20, 40, 80, 160, 320, 640, 1280, 2560, 5120, and 10240 m below the respective waste outfalls. Considerably more sampling emphasis was placed on the area immediately below the outfalls because radionuclide concentrations were expected to change rapidly in this region.

A core sampling technique was used in the collection of sediment samples. A section of 2.4 cm diameter plastic pipe was sharpened on one end and was gently driven with a hammer into the sediment to a maximum depth of 30 cm. Each station was sampled at the center and two lateral positions in the stream channel for a total

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of three cores per station. The coring device resulted in about a 10% compaction of the contained core. In many cases, sampling depths were less than 30 cm, especially in the upper portions of the canyons where alluvial deposits were shallow. Sampling depths also varied from core to core at each station although in most cases were about equivalent ~~20 cm~~. Individual sediment samples were sealed in a plastic bag to prevent cross-contamination and they were frozen until the radionuclide analyses could be initiated. The frozen core sample from the center of the stream channel was sectioned into a 0-2.5 cm, 2.5-7.5 cm, 7.5-12.5 cm and a remainder (below 12.5 cm) section to provide data on the vertical distribution of ^{137}Cs and plutonium.

A sample of the most abundant grass, shrub, and tree species was collected at each sampling station to determine the key plant receptors of plutonium and ^{137}Cs. In addition, mosses and crustose lichens were sampled when available. Only those plants which were directly rooted in the stream channel, or that were likely to be inundated during storm runoff events, were collected. The samples consisted of the complete above-ground portions of the grasses, mosses, and lichens and the terminal leaves and stems of the shrub and tree species. Particulates were not removed from the exterior of the plant surfaces prior to radiochemical analysis. A considerable amount of soil was associated with moss and lichen samples. Each sample was bagged and sealed immediately upon collection to prevent sample cross-contamination. Most species were identified to genus prior to analyses.

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Small mammal collections were made in the control area above the respective outfall locations, and at the 0, 2560, and 10240 m post-outfall sampling stations. Snap traps were positioned on a 6 x 8 grid network at each station with two traps at each grid point for a total of 96 traps per station. Eight grid points were established at 5 m intervals parallel to the stream channel, and six were established perpendicular to the stream channel at 10 m intervals. Peanut butter was used as bait and each station was trapped for three nights (288 trap-nights) without pre-baiting treatment. Each specimen was weighed, measured, and examined according to procedures outlined by Packard (1971) to provide data on various physical characteristics of the species. The pelt was first removed from the animal to prevent contamination of the remainder of the carcass with pelt debris. The skinned carcass was cleaned with a damp paper towel to further reduce the probability of cross-contamination. The lung, liver, and evicerated carcass were saved, along with the pelt, for individual radionuclide analysis.

Radionuclide analysis

Vegetation and rodent carcasses were oven-dried at 100°C for 24 h and then were muffled at 450°C until a white ash was obtained. Ashed biotic samples were dissolved in 50 ml of 7.2 N HNO_3 in a 600-ml Pyrex beaker and were analyzed for ^{137}Cs on a 7.6 x 7.6 cm NaI (Tl) scintillation detector. Oven-dried sediment samples were counted in a 600-ml Pyrex beaker and the height of the contained volume was recorded to the nearest millimeter. Standards containing known quantities of ^{137}Cs were prepared in the proper geometry

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to compare with sample materials. Some samples contained measurable ^{134}Cs and ^{40}K and the channels ratio method was used to correct the ^{137}Cs photopeak area for ^{134}Cs and ^{40}K contributions.

The minimum ^{137}Cs sensitivity ($P < 0.05$) of our gamma detection system was:

Sediment (50-gram sample)	-	0.09 pCi/g
Vegetation (100-gram sample)	-	0.40 pCi/g
Mouse Carcass (15-gram sample)	-	0.30 pCi/g

All sample materials to which tracer quantities of ^{242}Pu were added, were subjected to hydrofluoric-nitric acid digestion, ion exchange separation, electrodeposition, and alpha-ray spectroscopy to quantify the plutonium content. The total sample was carried through the plutonium chemistry to eliminate any errors associated with aliquoting a complex matrix, such as the sediments. The minimum detectable amount of ^{238}Pu and ^{239}Pu based on the variation in counts due to background was 0.05 pCi/sample assuming a 50% recovery of the ^{242}Pu tracer. Tracer recoveries for sample data reported in this paper varied from 30-100% and averaged about 45% since the procedures used were not optimized for biological samples. As procedure improvements were developed, low activity samples with recoveries below 30% were either reanalyzed or excluded from the following data analysis.

RESULTS AND DISCUSSION

Radionuclides in sediments

The mean ^{238}Pu, ^{239}Pu and ^{137}Cs concentrations in sediments along with variability estimates as a function of distance from the

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liquid waste outfalls are presented in Table 1. Concentration means for the control area were based on a sample size of six, whereas post-outfall concentration averages were based on a sample size of three.

Maximum concentrations of 160 pCi $^{238}Pu/g$, 70 pCi $^{239}Pu/g$ and 2200 pCi $^{137}Cs/g$ were measured in sediments from Mortandad Canyon, the area that has been used for liquid waste disposal for the least amount of time (10 years). Concentrations in Mortandad Canyon sediments typically averaged 5-10 times higher than those in DP and AP Canyons at comparable post-outfall distances. This observation is noteworthy in that both DP and AP Canyons received about as much or more plutonium than Mortandad Canyon.

Concentration patterns with distance post-outfall were similar between canyons in that highest values were measured near the waste outfalls with lower concentrations occurring further downstream. The highest concentrations of plutonium and ^{137}Cs in AP Canyon sediments were measured at the 80 m post-outfall location in an area of sediment accumulation.

Concentrations of plutonium and cesium in most post-outfall sediment samples reflect the addition of liquid wastes to considerable distances downstream even though surface water does not exist in the lower reaches of the canyons through most of the year. Plutonium and cesium concentrations about 10 times control values were measured at the 2560 m sampling station in Mortandad Canyon which was about 1500 m downstream from the last permanent surface water in the canyon. The downstream edge of the contamination was located about 3500 m post-outfall with the aid of gamma radiation survey

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TABLE 1. Average cesium-137 and plutonium concentrations in sediments from control and liquid waste contaminated areas during 1972.

Canyon-Sampling Location	^{238}Pu (pCi/g dry)	^{239}Pu (pCi/g dry)	^{137}Cs (pCi/g dry)
Mortandad			
Control	0.17*(99)**	0.47(52)	2.0 (85)
0***	160 (40)	59 (22)	2200 (43)
20	110 (94)	70 (86)	510 (100)
40	65 (91)	26 (120)	530 (94)
80	39 (62)	9.6 (55)	630 (14)
160	86 (87)	38 (110)	1200 (30)
320	20 (93)	3.3 (81)	290 (74)
640	17 (6)	4.0 (28)	320 (19)
1280	6.8 (42)	2.4 (38)	90 (30)
2560	8.6 (110)	2.5 (110)	91 (71)
5120	0.05 (140)	0.06(73)	0.57(26)
10280	0.01 (120)	0.02(130)	0.31(11)
DP-Los Alamos			
Control	0.02(150)	0.03(58)	0.31(100)
0	2.1 (66)	28 (68)	1700 (74)
20	1.5 (37)	7.9 (89)	190 (52)
40	0.06(76)	0.58(120)	5.9 (80)
80	0.20(84)	2.5 (120)	49 (140)
160	0.15(46)	0.61(43)	24 (22)
320	0.17(54)	0.5 (75)	15 (33)
640	0.23(77)	0.70(46)	51 (22)

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TABLE I. (Continued)

Canyon-Sampling Location	^{239}Pu (pCi/g dry)	^{238}Pu (pCi/g dry)	^{137}Cs (pCi/g dry)
DP-Los Alamos			
2560	0.03(66)	0.17(69)	13 (12)
5120	0.03(130)	0.36(82)	3.6 (42)
10280	0.05(64)	0.13(35)	1.6 (28)
Acid-Pueblo			
Control	0.009(100)	0.20 (100)	0.36(78)
0	0.02 (50)	2.3 (120)	0.53(15)
20	0.09 (150)	2.2 (38)	0.02(140)
40	0.03 (100)	6.9 (99)	1.5 (140)
80	1.0 (76)	54 (64)	0.74(100)
160	0.08 (35)	8.5 (87)	14 (42)
320	0.08 (7.5)	12 (23)	1.1 (6.4)
640	0.08 (25)	11 (29)	1.5 (21)
2560	0.02 (100)	1.5 (19)	1.1 (30)
5120	0.01 (100)	0.35 (80)	0.31(2.6)
10280	0.02 (50)	0.74 (66)	0.20(30)

* Control means based on six samples, post-outfall means based based on three samples

** Coefficient of variation ($\frac{\text{standard deviation}}{\text{mean}} \times 100$)

*** Meters below waste effluent outfall.

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instruments. Elevated ^{238}Pu levels measured at the 2560 m location, demonstrate that the downstream transport of this isotope occurred within a four-year period, since significant ^{238}Pu was not released to Mortandad Canyon until 1968.

Plutonium-239 and ^{137}Cs concentrations significantly ($P < 0.05$) above control station values, were measured at the 10230 m sampling location in DP Canyon, some 3000 m below the confluence of ~~AP~~ DP and AP Canyon and about 4000 m downstream from any permanent surface water. Relatively low sediment concentrations of ^{238}Pu were measured in the DP Canyon study area indicating that either low quantities of this isotope were released to the canyon and/or that substantial redistribution had occurred.

Storm runoff, especially rain, has been implicated as an important vector in the transport of radionuclide contaminated sediments down the canyons (Kingsley 1947, Purtymun 1974). Surface flows following a rainfall event on the upper DP-AP Canyon watershed often reach the Rio Grande. Data presented elsewhere (Purtymun et al. 1966) indicated that sediment concentrations of plutonium in the upper reaches of DP Canyon increased through the fall-winter months as wastes were released to the canyon and decreased through the summer months as storm (rain) runoff events moved the contaminated sediments downstream. Hydrological data gathered over the last 16 years show that an average of four such events reach the Rio Grande River each year. On the other hand, storm runoff in the Mortandad Canyon watershed has never reached post-outfall distances of over 3500 m in the 16 years of observation due to unsaturated alluvial deposits up to 35 m in depth beginning about 2000 m

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post-outfall. Thus, hydrological and accompanying sediment transport characteristics of these semi-drainage canyons appear to be an important factor in the downstream movement of liquid effluent radiocontaminants.

If a physical transport process (i.e. sediment transport) is operative in the downstream movement of radioactivity, then one could speculate that the relative distribution of plutonium and cesium with distance might be similar, since differences in chemical behavior would be at least partially negated. The correlation between individual sediment concentrations of the respective isotopes of plutonium and ^{137}Cs, varying over 2 to 3 orders of magnitude, were significant ($P < 0.05$) for most comparisons in Mortandad, DP, and AP Canyons, as determined by linear least squares regression.

The least squares regression line for ^{238}Pu vs ^{137}Cs in 27 samples from Mortandad Canyon (Fig. 2) was $Y = 7.7 \pm 11$ (SE) + 0.08 ± 0.01 X ($r = 0.63$). The regression intercept (7.7) was not significantly different ($P < 0.05$) from zero. Even though the data were relatively well-distributed along the regression line, there was an obvious clumping of lower concentrations near the origin. This tendency was even more pronounced for the DP and AP Canyon data comparisons, indicating that samples from areas of intermediate concentrations were needed to better define the plutonium-cesium relationship. Correlations were improved by log transformation of the concentration data as shown by the ^{238}Pu vs ^{137}Cs comparison for Mortandad Canyon in Fig. 3. Although the interpretation of the latter comparison was not immediately obvious in terms of

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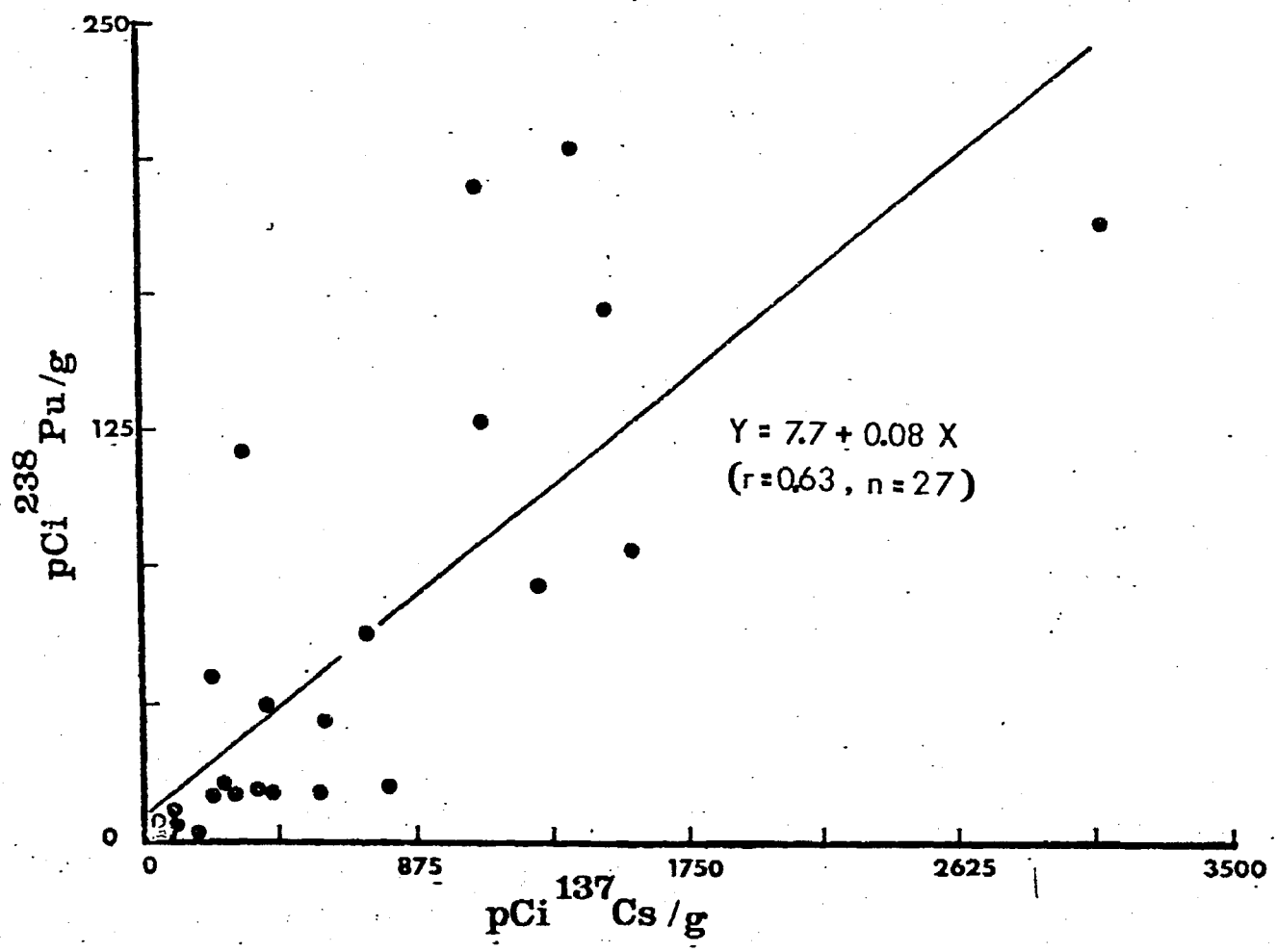

FIG. 2. The relationship between ^{137}Cs and ^{238}Pu concentrations in Mortandad Canyon sediments from the 0-2560 m segment below the liquid waste outfall.

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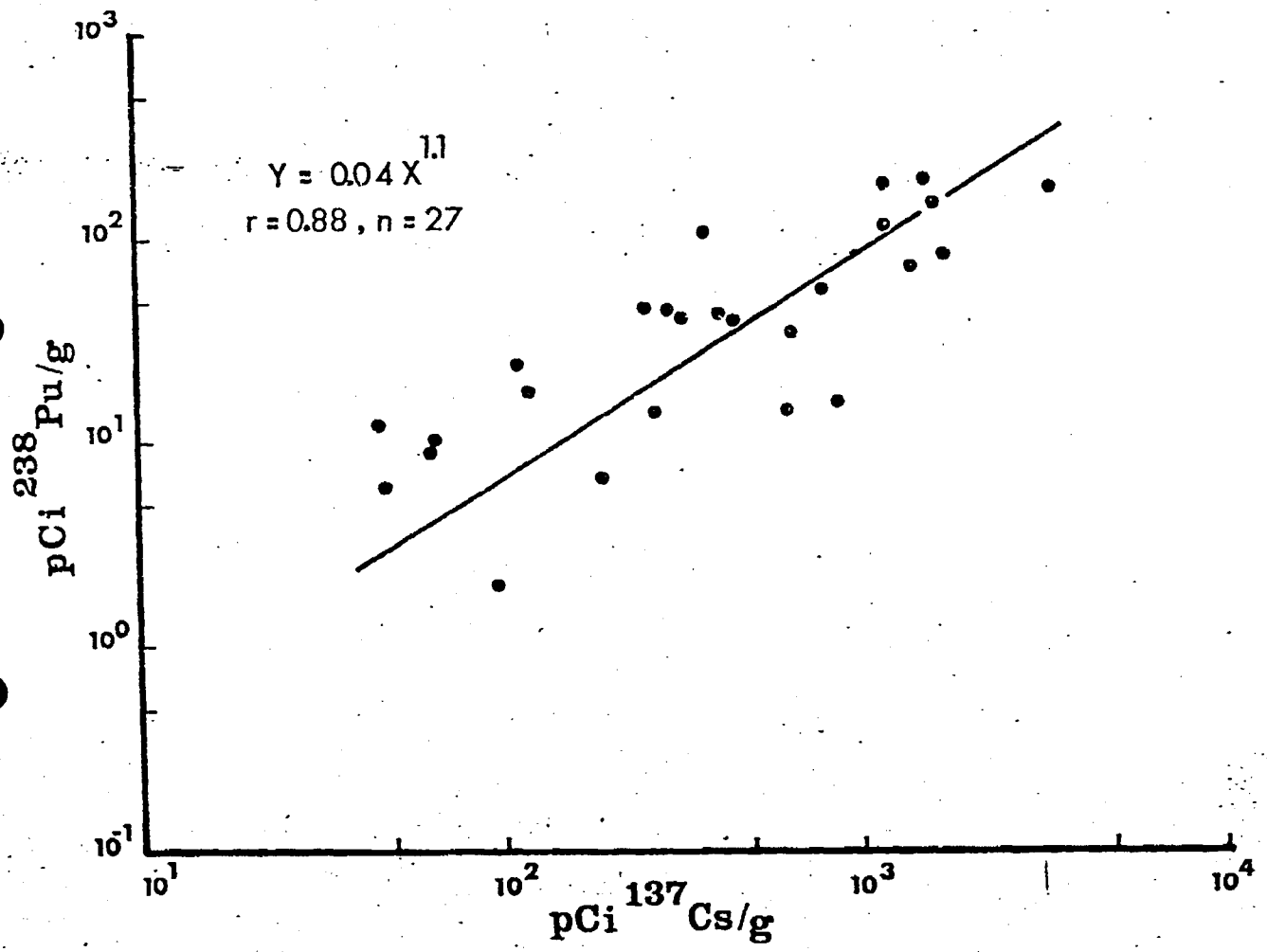

FIG. 3. The relationship between log transformed ^{137}Cs and ^{238}Pu concentrations in Mortandad Canyon sediments from the 0-2560 m segment below the liquid waste outfall.

distributional relationships, the regression may be useful for predictive purposes.

Similar relationships between plutonium and ^{137}Cs concentrations are under study in the terrestrial ecosystem at Trinity Site (Hakonson and Johnson 1974) where the world's first nuclear weapon was detonated in 1945. If a physical process (i.e. resuspension) is operative in the redistribution of weapons-deposited radioactivity in the arid Trinity Site environs, then a relationship may exist between plutonium and ^{137}Cs concentrations in various ecosystem components.

Plutonium and ^{137}Cs concentrations as a function of sediment depth for samples collected in 1972 will not be presented at this time since another paper at this symposium treates the subject in detail (Nyhan et al, this symposium). In general, plutonium and ^{137}Cs were relatively well-mixed with depth at most sampling locations. The fact that ^{238}Pu had penetrated to the lower sampling depths (> 12.5 cm) in Mortandad Canyon indicated that the mixing process was rapid compared to vertical penetration rates for plutonium in terrestrial ecosystems (Francis 1973). In Mortandad Canyon, ^{238}Pu penetration to the 12.5-30 cm depth profile had occurred within a four-year period.

The variability in plutonium and cesium concentrations in replicate sediment samples was generally quite large. The coefficient of variation (CV) for plutonium measurements averaged about 80% in a range of 6-150% for all three canyons. The CV for sediment ^{137}Cs measurements averaged about 50% in a range of 3-140% for all canyons indicating that the ^{137}Cs may be more homogeneously distributed throughout the stream channels than plutonium. Differences

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TABLE 2. Mean Concentrations of plutonium and cesium in sediments and vegetation from a control and two contaminated segments of Mortandad Canyon during 1972.

Sample Type*	No. Samples	Control (pCi/g dry) ^{238}Pu	^{239}Pu	^{137}Cs	No. Samples	0-160 m Post-outfall (pCi/g dry) ^{238}Pu	^{239}Pu	^{137}Cs	Samples	160-2560 m Post-outfall (pCi/g dry) ^{238}Pu	^{239}Pu	^{137}Cs
Sediment	6	0.17 (99)	0.47 (52)	2.0 (85)	15	90 (79)	40 (95)	980 (78)	12	12 (92)	2.6 (69)	200 (76)
Grass	4	0.02 (68)	0.003(58)	1.1 (110)	5	17 (64)	1.7 (82)	76 (81)	2	0.22	0.07	74 (110)
Shrub	6	0.004(79)	0.003(42)	0.22(32)	13	0.71 (170)	0.14 (150)	9.5 (190)	7	0.02 (84)	0.008(44)	5.4 (190)
Tree	3	0.004(100)	0.005(44)	0.09(79)	6	0.18(82)	0.06 (150)	0.94(100)	8	0.009(140)	0.008(120)	0.37(70)
Moss	2	0.08 (66)	0.08 (88)	0.62(130)	4	3.4 (150)	1.5 (120)	36 (140)	3	4.3 (120)	2.6 (100)	200 (64)
Lichen	2	0.1	0.6	8.0	3	18 (39)	6.6 (29)	110 (68)	1	1.4	0.67	23

* Genera comprising the vegetation types were: Grass-Poa, Bouteloua; Shrub-Quercus, Artemisia, Berberis, Chrysothamnus, Prunus, Ribes, Physocarpus, Rhus, Salix; Tree-Acer, Juniperus, Pinus, Pseudotsuga; moss and crustose lichens-not identified.

in the chemical behavior of the two elements would be expected because ^{137}Cs is likely more soluble than plutonium in post-outfall sediments.

The $^{239}Pu/^{238}Pu$ activity ratios in sediment averaged 0.31 ± 0.12 (SD), 4.6 ± 2.4, and 50 ± 25 for Mortandad (from 0-2560 m post-outfall) DP and AP Canyons, reflecting past relative additions of the two radionuclides.

Radionuclides in vegetation

The plutonium and ^{137}Cs concentration data for vegetation were grouped into rather general categories due to the lack of similarity in plant species composition down the canyons and to the small number of samples collected from individual locations (Tables 2-4). The contaminated portion of each canyon was divided into two segments; one near the waste outfall, where sediment radionuclide concentrations were relatively high, and one further downstream where sediment radionuclide concentrations averaged about a factor of 10 lower. Average sediment concentrations for the various stream channel segments also appear in the Tables.

The variability in plutonium and ^{137}Cs concentrations within vegetation types was large. The coefficient of variation range about 12% to 270%. At least some of this extreme variability was the result of grouping the data by vegetation types and by areas of relative sediment radionuclide concentrations. The data were not sufficient to compare concentrations by species and by individual sampling locations.

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TABLE 3. Mean concentrations of plutonium and cesium in sediments and vegetation from control and two contaminated segments of DP-Los Alamos Canyon during 1972.

Sample	No.	Control (pCi/g dry)			No.	0-20 m Post-outfall (pCi/g dry)			No.	20-10280 m Post-outfall (pCi/g dry)		
Type*	Samples	^{238}Pu	^{239}Pu	^{137}Cs	Samples	^{238}Pu	^{239}Pu	^{137}Cs	Samples	^{238}Pu	^{239}Pu	^{137}Cs
Sediment	6	0.02 (150)	0.03 (58)	0.31(100)	6	1.8 (58)	18 (94)	920 (120)	21	0.12 (100)	0.73 (162)	19 (124)
Grass	2	0.04 (110)	0.07 (81)	0.05(140)	4	0.76 (120)	1.8 (160)	194 (43)	9	0.03 (71)	0.09 (110)	2.4 (110)
Shrub	2	0.11 (130)	0.07 (81)	0.28(40)	--	--	--	--	8	0.02 (160)	0.02 (82)	0.74(99)
Tree	3	0.01 (7.4)	0.03 (28)	0.18(12)	--	--	--	--	13	0.007(110)	0.05 (170)	0.41(93)
Moss	2	0.06	0.30	1.0	--	--	--	--	4	0.23 (73)	1.5 (41)	28 (57)
Lichen	--	--	--	--	--	--	--	--	--	--	--	--

* Genera of vegetation types were: Grass-Poa, Muhlenbergia; Shrub-Berberis, Quercus, Fallugia, Salix, Rhus; Tree-Pinus, Populus, Juniperus; Moss and crustose lichens-not identified.

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TABLE 4. Mean concentrations of plutonium and cesium in sediments and vegetation from control and two contaminated segments of Acid-Pueblo Canyon during 1972.

Sample Type*	No. Samples	Control (pCi/g dry) ^{238}Pu	^{239}Pu	^{137}Cs	No. Samples	0-640 m Post-outfall (pCi/g dry) ^{238}Pu	^{239}Pu	^{137}Cs	No. Samples	640-10280 m Post-outfall (pCi/g dry) ^{238}Pu	^{239}Pu	^{137}Cs
Sediments	6	0.009(100)	0.20 (100)	0.36(78)	21	0.28 (190)	21 (170)	3.3 (170)	9	0.02 (60)	0.84 (74)	0.22(67)
Grass	2	0.05 (140)	0.03	0.33	7	0.03 (97)	4.2 (120)	0.68(120)	3	0.02	0.33 (71)	0.60(39)
Shrub	4	0.01 (120)	0.03 (170)	0.22(100)	9	0.05 (160)	1.6 (270)	0.66(33)	10	0.009(150)	0.009(70)	0.27(47)
Tree	--	--	--	--	--	--	--	--	8	0.003(57)	0.006(71)	0.26(67)
Moss	--	--	--	--	4	1.8 (86)	65 (92)	34 (150)	--	--	--	--
Lichen	--	--	--	--	2	0.82 (140)	2.1 (84)	8.3	--	--	--	--

* Genera of vegetation types were: Grass-Poa; Shrub-Artemisia, Berberis, Prunus, Chrysothamnus, Rosa, Quercus, Fallugia, Ribes, Salix, Physocarpus, Rhus; Tree-Juniperus, Pinus, Psuedotsuga; Moss and crustose lichen-not identified.

A positive relationship was observed between sediment and vegetation radionuclide concentrations, with maximum plutonium and ^{137}Cs concentrations in vegetation corresponding to areas of highest mean sediment radionuclide concentrations. Concentrations of the three radionuclides were generally higher in Mortandad Canyon vegetation than in vegetation from the other two canyons which corresponds to the concentration pattern observed in sediments. The values in Mortandad Canyon vegetation ranged to maximums of 18 pCi $^{238}Pu/g$, 6.6 pCi $^{239}Pu/g$ and 200 pCi $^{137}Cs/g$ dry weight. Corresponding maximums for DP Canyon were 0.76 pCi $^{238}Pu/g$, 1.8 pCi $^{239}Pu/g$ and 190 pCi $^{137}Cs/g$ whereas the values for AP Canyon were 1.8 pCi $^{238}Pu/g$, 65 pCi $^{239}Pu/g$ and 34 pCi $^{137}Cs/g$.

Average plutonium concentrations in vegetation from the low concentration post-outfall segment in each canyon were generally not significantly different than control station values, with the exception of moss and lichen samples where concentrations remained relatively high. On the other hand, the ^{137}Cs concentrations in all post-outfall vegetation samples were significantly higher than control station values with the exception of the 640-10280 m post-outfall segment of AP Canyon.

Grass species, mosses, and lichens, usually averaged higher plutonium and ^{137}Cs concentrations than shrubs. Concentrations in tree species averaged lowest, although the differences between vegetative types in some cases were not significant ($P < 0.05$). A similar pattern was noted at Trinity Site where grass species contained higher ^{239}Pu concentrations than other vegetative types

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(Hakonson and Johnson 1974). Although additional data are needed to substantiate these observations, one could speculate that higher concentrations in the low growth forms (i.e., grasses, mosses, lichens) may be due to surface area relationships if the contamination is on the plant surfaces or that rooting characteristics and/or physiological differences between the species account for the differences in plutonium and ^{137}Cs concentration between species. Seasonal variations in plutonium and ^{137}Cs concentrations in plant species should be studied in conjunction with the food habits of herbivores, to identify periods of greatest potential transfer of the radionuclides to animals living in the canyons.

The relationship between the plutonium and ^{137}Cs concentrations in all vegetation samples were compared by canyon using linear least squares regression techniques. Significant relationships ($P < 0.05$) between the respective plutonium isotopes and ^{137}Cs were observed in the Mortandad and DP Canyon study areas but not in AP Canyon where ^{137}Cs and ^{238}Pu concentrations in sediments and vegetation were generally near pre-outfall (control) levels. Although the correlation between ^{238}Pu and ^{137}Cs in Mortandad Canyon vegetation (Fig. 4) was significant ($r = 0.79$, $n = 39$), the clumping of data near the lowest concentrations, as observed for plutonium-^{137}Cs comparison in sediments, biased the regression and indicated the need for data in the higher concentration ranges. Both physical and biological processes may be operative in the movement of the plutonium and ^{137}Cs into the vegetation (Hakonson 1975), although a physical process (i.e., external contamination of plants with resuspended material) would seem more likely.

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The slope of the regression line in Fig. 4 represents the average $^{238}Pu/^{137}Cs$ activity ratio in vegetation. The value of 0.1 compares with an average $^{238}Pu/^{137}Cs$ activity ratio of 0.08 in Mortandad Canyon sediments (slope of regression line in Fig. 2). The similar activity ratios in sediments and vegetation suggest that a physical transport mechanism, such as resuspension of radioactivity, could be operative in plutonium and ^{137}Cs movement to the vegetation component. If a biological mechanism (root uptake) were involved then the $^{238}Pu/^{137}Cs$ ratio in plants would likely be smaller than in sediments since ^{137}Cs is the more soluble of the two nuclides and therefore, would be incorporated into plant tissues more readily than plutonium. This interpretation is obviously speculative, however, the exercise illustrates the potential use of isotopic ratios in studying the mechanisms of radionuclide redistribution in the environment.

Log transformation of the vegetation concentration data improved the correlation between plutonium and ^{137}Cs only slightly for the Mortandad Canyon comparison (Fig. 5), but resulted in considerable improvement for the DP and AP Canyon comparisons. The log relationships were significant ($P < 0.05$) for all canyons.

Estimates of plant-sediment activity ratios (i.e., $\frac{\text{pCi/g dry plant}}{\text{pCi/g dry sediment}}$) were made for each isotope to determine the relative movement of plutonium and ^{137}Cs from sediment to streambank vegetation. The ratios were significantly higher ($P < 0.05$) in post-outfall moss and lichen samples than in the remaining vegetation types in the respective canyons. The ratio varied from about 0.01 to 10 in mosses and lichens and from 0.0001 to 1 for the

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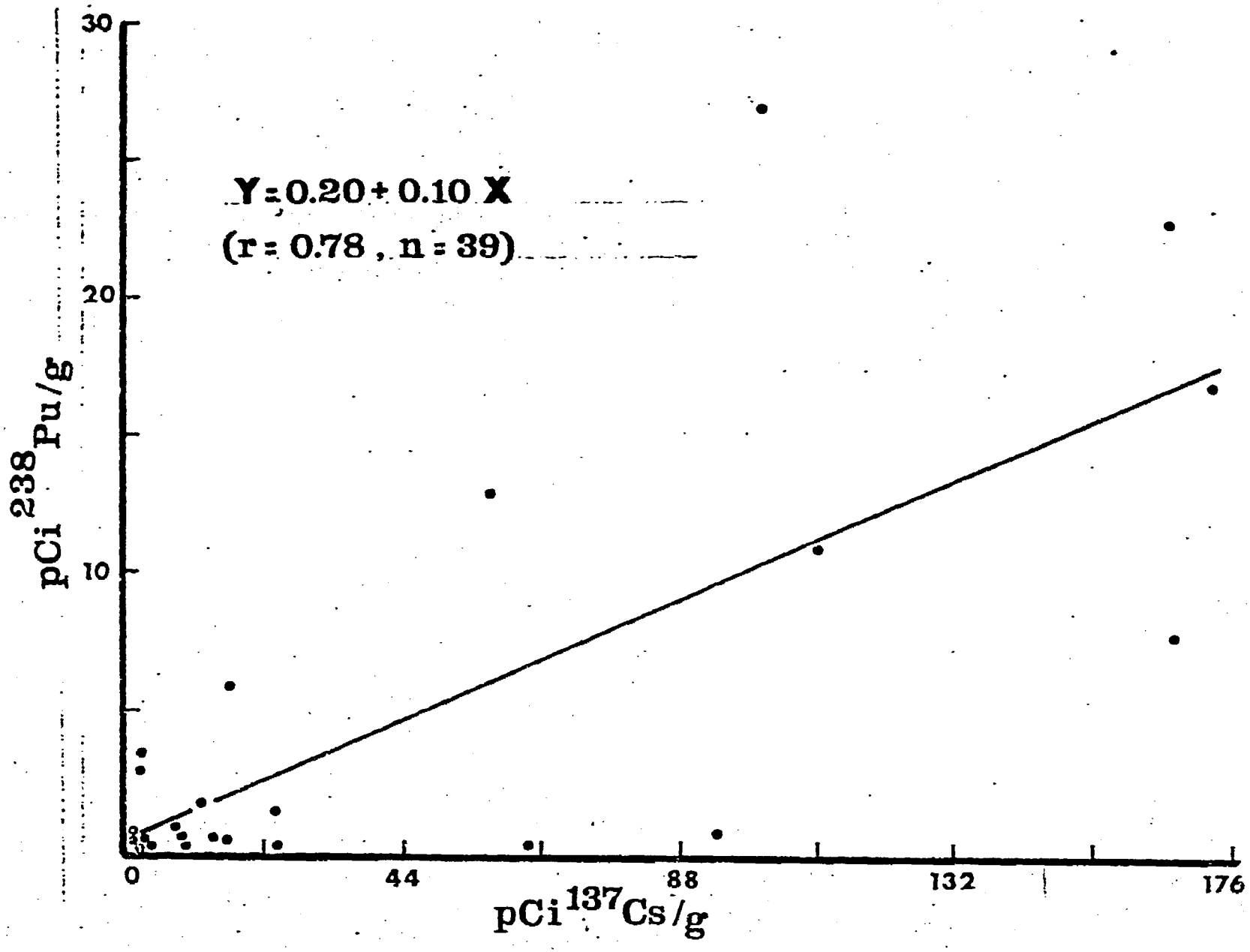

FIG. 4. The relationship between ^{137}Cs and ^{238}Pu concentrations in Mortandad Canyon vegetation samples.

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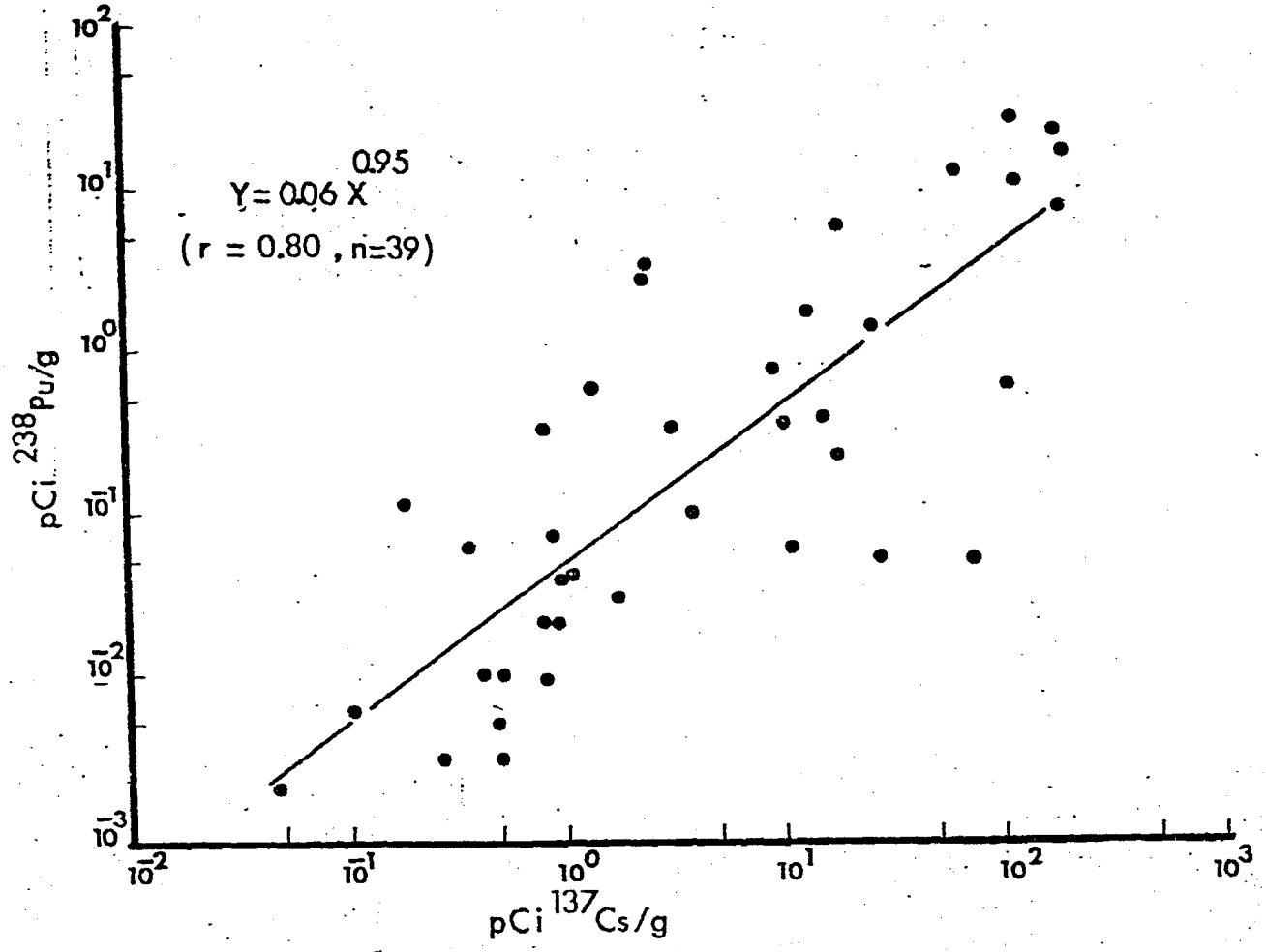

FIG. 5. Relationship between log transformed concentrations of ^{137}Cs and ^{238}Pu in Mortandad Canyon vegetation samples.

remaining vegetation types. Relatively large amounts of soil particulates were associated with the moss and lichen samples compared to the remaining vegetation samples ~~and~~ may ~~particularly~~ account for the higher activity ratios in the mosses and lichens. There were no significant differences ($P < 0.05$) ~~in the~~ between ^{137}Cs and plutonium plant-sediment activity ratios within a particular canyon indicating that the relative movement of the respective isotopes from sediments to vegetation were similar. However, the ^{239}Pu (and ^{238}Pu) plant-sediment satios in DP and AP Canyons were significantly higher ($P < 0.05$) than corresponding ratios in Mortandad Canyon indicating that a relatively greater transfer of plutonium to vegetation had occurred in the two canyons that have received wastes over the longest times. The differences in ^{137}Cs plant-sediment activity ratios between canyons were not significant.

Although the data comparing isotope ratios are certainly not conclusive, such ratios may be useful in advancing our knowledge on environmental plutonium behavior.

Radionuclides in rodents

Average concentrations of plutonium were highest in the lung and/or pelt tissues of rodents from the three canyons (Table 5). Concentrations of plutonium in liver and carcass tissue were generally at the analytical detection limits. Elevated ^{137}Cs concentrations were measurable in carcass tissues, with maximums of 103 ~~116~~ pCi/g dry ~~wet~~ occurring in DP outfall samples. Concentration

TABLE 5. Mean concentrations of plutonium and cesium in rodents from the liquid disposal areas during 1972.

pCi/g dry weight

Location-Sample Type*	No. Samples	^{238}Pu		^{239}Pu		^{137}Cs	
		CONTROL					
DP-Los Alamos							
Lung	6	0.04	(130)	0.04	(190)	---	
Pelt	6	0.06	(50)	1.3	(70)		
Liver	6	0.004	(160)	0.02	(110)		
Carcass	6	0.002	(110)	0.02	(100)	3.2	(140)
Mortandad							
Lung	2	0.02	(140)	0.02	(60)		
Pelt	2	0	(0)	0.03	(80)		
Liver	2	0.004	(140)	0.002	(140)		
Carcass	2	0.001	(140)	0	(0)	1.2	(12)
Acid-Pueblo							
Lung	4	0.04	(170)	0	(0)		
Pelt	4	0.003	(110)	0.01	(70)		
Liver	4	0.008	(90)	0.02	(80)		
Carcass	4	0.0	(200)	0.007	(150)	0.20	(210)

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TABLE 5., Continued

Location-Sample Type*	No. Sample	pci/g dry weight ^{238}Pu		^{239}Pu		^{137}Cs	
		OUTFALL					
DP-Los Alamos							
Lung	19	0.08	(130)	0.04	(130)	---	
Pelt	19	0.11	(110)	0.74	(60)		
Liver	19	0.04	(160)	0.08	(120)		
Carcass	19	0.01	(140)	0.07	(200)	103	(130)
Mortandad							
Lung	5	1.3	(160)	0.57	(130)		
Pelt	5	1.9	(70)	0.28	(60)		
Liver	5	0.12	(65)	0.03	(60)		
Carcass	5	0.07	(100)	0.01	(110)	1.6	(130)
Acid-Pueblo							
Lung	5	0.08	(90)	0.02	(150)		
Pelt	5	0.03	(110)	0.36	(70)		
Liver	5	0.08	(170)	0.03	(130)		
Carcass	5	0.004	(70)	0.04	(140)	0.35	(80)

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TABLE 5., Continued

Location-Sample Type*	No. Sample	^{238}Pu		^{239}Pu		^{137}Cs	
		pCi/g dry weight					
2560 m POST-OUTFALL							
DP-Los Alamos							
Lung	5	0.06	(90)	0.06	(190)	—	
Pelt	5	0.01	(74)	0.04	(100)		
Liver	5	0.02	(160)	0.02	(180)		
Carcass	5	0.001	(190)	0.01	(130)	0.30	(230)
Mortandad							
Lung	8	3.1	(250)	0.32	(220)		
Pelt	8	0.22	(230)	0.08	(240)		
Liver	8	0.02	(110)	0.02	(120)		
Carcass	8	0.006	(160)	0.002	(120)	1.0	(70)
Acid-Pueblo							
Lung	16	0.28	(220)	15	(410)		
Pelt	16	0.01	(130)	0.08	(190)		
Liver	16	0.08	(170)	0.03	(110)		
Carcass	16	0.002	(120)	0.006	(120)	14	(150)

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TABLE 5., Continued

Location-Sample Type*	No. Sample	^{238}Pu		^{239}Pu		^{137}Cs	
		pci/g dry weight					
10 280 m POST-OUTFALL							
DP-Los Alamos							
Lung	10	0.04	(150)	0.06	(100)	—	
Pelt	10	0.06	(120)	0.03	(90)		
Liver	10	0.01	(210)	0.01	(67)		
Carcass	10	0.001	(90)	0.001	(90)	3.9	(77)
Mortandad							
Lung	13	2.9	(300)	1.4	(340)		
Pelt	13	0.50	(460)	1.3	(360)		
Liver	13	0.04	(250)	0.08	(220)		
Carcass	13	0.01	(270)	0.004	(210)	1.2	(290)
Acid-Pueblo							
Lung	4	0.02	(120)	0.02	(110)		
Pelt	4	0.01	(130)	0.08	(70)		
Liver	4	0.04	(120)	0.01	(70)		
Carcass	4	0.002	(80)	0.004	(80)	0.69	(154)

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patterns with distance post-outfall were not consistent with the patterns observed for vegetation and sediments. Maximum concentrations of all three radionuclides in rodent tissues in some cases occurred in samples from the downstream sampling locations.

A relatively high mean concentration of ^{137}Cs was measured in rodents from the ~~0-20 m post~~ outfall ~~segment~~ location ~~of~~ in DP Canyon where sediment concentrations averaged 920 pCi/g. The stream channel in this portion of DP Canyon is relatively undefined, resulting in a large lateral contamination of sediments with radioactivity. The rodents living in this region ~~have~~ had a more intimate contact with the radioactivity than ~~do~~ did the rodents in the 0-160 m segment of Mortandad which has a narrow, well-defined stream channel with sediment ^{137}Cs concentrations comparable to the 0-20 m post-outfall segment of DP Canyon. Mortandad Canyon rodents from the 0-160 m segment averaged only 0.84 pCi ^{137}Cs/g carcass, which indicates the importance of determining the ecological relationships in contaminated areas. The plutonium concentrations in most rodent tissues were not significantly different between canyons. The coefficient of variation for mean concentrations of all three radionuclides generally ranged from 100% to as high as 410%. Factors such as rodent species differences, food preferences and movement patterns in the contaminated areas are likely important components of this extreme variability.

CONCLUSIONS

Based on radionuclide concentrations, it appears that sediments are the major reservoir of the plutonium and ^{137}Cs released to the three canyon study areas. The downstream distribution of these radionuclides is influenced by hydrological characteristics

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of the canyons ~~and that~~ The transport of sediments during storm runoff events ~~is~~ appears to be a major vector in the downstream movement of the radiocontaminants. Statistically significant ($P < 0.05$) correlations between sediment plutonium and ^{137}Cs provide supporting evidence for a physical transport process. Data indicate that transport distances are considerable in the DP-AP Canyon system but are relatively minor in Mortandad Canyon due to hydrological differences between canyons.

Movement of plutonium and ^{137}Cs into the vegetation component of the canyon ecosystems was detectable. Plant-sediment activity ratios of 10^{-4} - 10 were observed, indicating that relative contamination of plants under natural conditions is greater than would be predicted by greenhouse studies. Data indicate that plant-sediment plutonium activity ratios are higher in the areas which have been used for waste disposal over the longest time.

Highest mean concentrations of plutonium in the lung and pelt of rodents from the canyons suggest that a physical mechanism is operative in the movement of this element to small ground-dwelling mammals.

The variability in plutonium and ^{137}Cs concentrations ~~of~~ in sample groups was large. The coefficient of variation was smallest for sediments and largest for vegetation and rodent tissues. The extreme variability indicates the need for large sample sizes to demonstrate differences between study treatments.

A major impact of time on the distribution of waste contaminants has been in the downstream transport of waste contaminants.

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The estimates of residual plutonium in the AP and DP Canyon systems, based on the sediment radionuclide concentration data for 1972, indicate that more than 90% of the estimated input has flushed out of the canyons. In Mortandad Canyon, the waste plutonium is confined to the 3500 m segment immediately below the waste outfall. Some of the data indicates increased plant uptake of plutonium in the oldest waste disposal areas. Such differences cannot be demonstrated for rodents with data presently available.

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LITERATURE CITED

Francis, C. W. 1973. Plutonium mobility in soils and uptake in plants - A review. J. Environ. Quality 2(1):67-60.

Hakonson, T. E., J. W. Nyhan, L. J. Johnson, and K. V. Bostick. 1973. Ecological investigation of radioactive materials in waste discharge areas at Los Alamos. Los Alamos Scientific Laboratory report, LA-5282-MS, Los Alamos, New Mexico. 47 p.

Hakonson, T. E. and L. J. Johnson. 1974. Distribution of environmental plutonium in the Trinity Site environs after 27 years, p. 242-247. In W. S. Snyder (ed) Proc. of the Third Int. Congress of the Int. Radiation. NTIS report CONF-730907-P1.

Hakonson, T. E., L. J. Johnson, and W. D. Purtymun. 1974. The distribution of plutonium in liquid waste disposal areas at Los Alamos, p. 248-253. In W. S. Snyder (ed) Proc. of the Third Int. Congress of the Int. Radiation Protection Association. NTIS report CONF-730907-P1.

Hakonson, T. E. 1975. Environmental pathways of plutonium into terrestrial plants and animals. Health Phys. (In Press)

Hanson, W. C. 1975. Ecological considerations of the behavior of plutonium in the environment. Health Phys. (In Press)

International Commission on Radiological Protection (ICRP). 1962. Recommendations of the International Commission on Radiological Protection, ICRP Publ. 6. Pergamon Press, New York.

Kingsley, W. H. 1947. Survey of Los Alamos and Pueblo Canyons for radioactive contamination and radioassay tests run on sewer-water samples and water and soil samples taken from Los Alamos and Pueblo Canyons. Los Alamos Scientific report LA-516.

LASL-AEC OFFICIAL

Langham, W. H. 1972. Biological implications for the transuranium elements for man. Health Phys. 22:943-952.

Packard, R. L. 1971. Small mammal survey on the Jornada and Pantex sites. Int. Biol. Program Grassland Biome, Tech. report 114.

Purtymun, W. D., G. L. Johnson, and E. C. John. 1966. Distribution of radioactivity in the alluvium of a disposal area at Los Alamos, New Mexico. U. S. Geol. Survey Prof. Paper 550-D.

Purtymun, W. D. 1974. Storm runoff and transport of radionuclides in DP Canyon, Los Alamos County, New Mexico. Los Alamos Scientific Laboratory report, LA-5744.

LASL-AEC-OFFICIAL