

THE ECODISTRIBUTION OF PLUTONIUM

IN LIQUID WASTE DISPOSAL AREAS AT LOS ALAMOS

T.E. Hakonson, L.J. Johnson and W.H. Purtymun  
Los Alamos Scientific Laboratory  
Los Alamos, New Mexico 87544

INTRODUCTION

Although the main function of the Los Alamos Scientific Laboratory has been in the field of weapons development, there are numerous other projects dealing with the use of nuclear energy. As may be expected, the nuclear programs have generated quantities of solid and liquid radioactive wastes. Although the liquid wastes are treated to remove impurities, a small but measurable fraction of the radioactive constituents remain in the effluent. Currently, these effluents are released into two canyons where they eventually disappear into the alluvium. In the past, an additional canyon was used as a liquid waste disposal area, but has not been used since 1964.

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A survey was initiated in each of these canyons to determine the concentrations of effluent-associated radionuclides in the alluvial sediments, water and some of the native biota. This paper will summarize preliminary findings on the ecodistribution of <sup>239</sup>Pu and <sup>240</sup>Pu in Mortandad Canyon, an area which has been used as a liquid effluent disposal area for 10 years.

**MASTER**

MATERIALS AND METHODS

The Los Alamos Scientific Laboratory occupies a 28,000

acre site on the Pajarito Plateau in northern New Mexico. The plateau, which is a shelf about 15 - 25 km wide and 70 km long on the eastern flank of the Jemez Mountains, has been deeply eroded by runoff with the result that the area consists of a series of mesas separated by canyons, many of which are several hundred feet deep.

The area has a semi-arid, continental mountain climate with an average annual precipitation of slightly more than 18 inches in the city of Los Alamos, of which 75 percent falls during the months of May through October. There are no permanent, natural streams flowing through the Laboratory area, although Frijoles Creek flows through Bandelier National Monument located on the southern border of the Laboratory site and the Rio Grande flows through White Rock Canyon on the eastern border. There are, however, intermittent streams flowing in the canyons during the rainy season, and surface water exists in certain of the canyons for a short distance below the discharge points for industrial or sanitary wastes.

Mortandad Canyon originates in the western portion of the Laboratory property at an elevation of about 2225 meters above sea level, and terminates about 15 km from its origin in the Rio Grande River on the eastern edge of the Laboratory property. The average slope over the entire length of the stream channel is 0.042.

Radionuclide bearing liquid effluents (containing  $^3\text{H}$ ,  $^{137}\text{CS}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{241}\text{Am}$ ) from a waste treatment plant

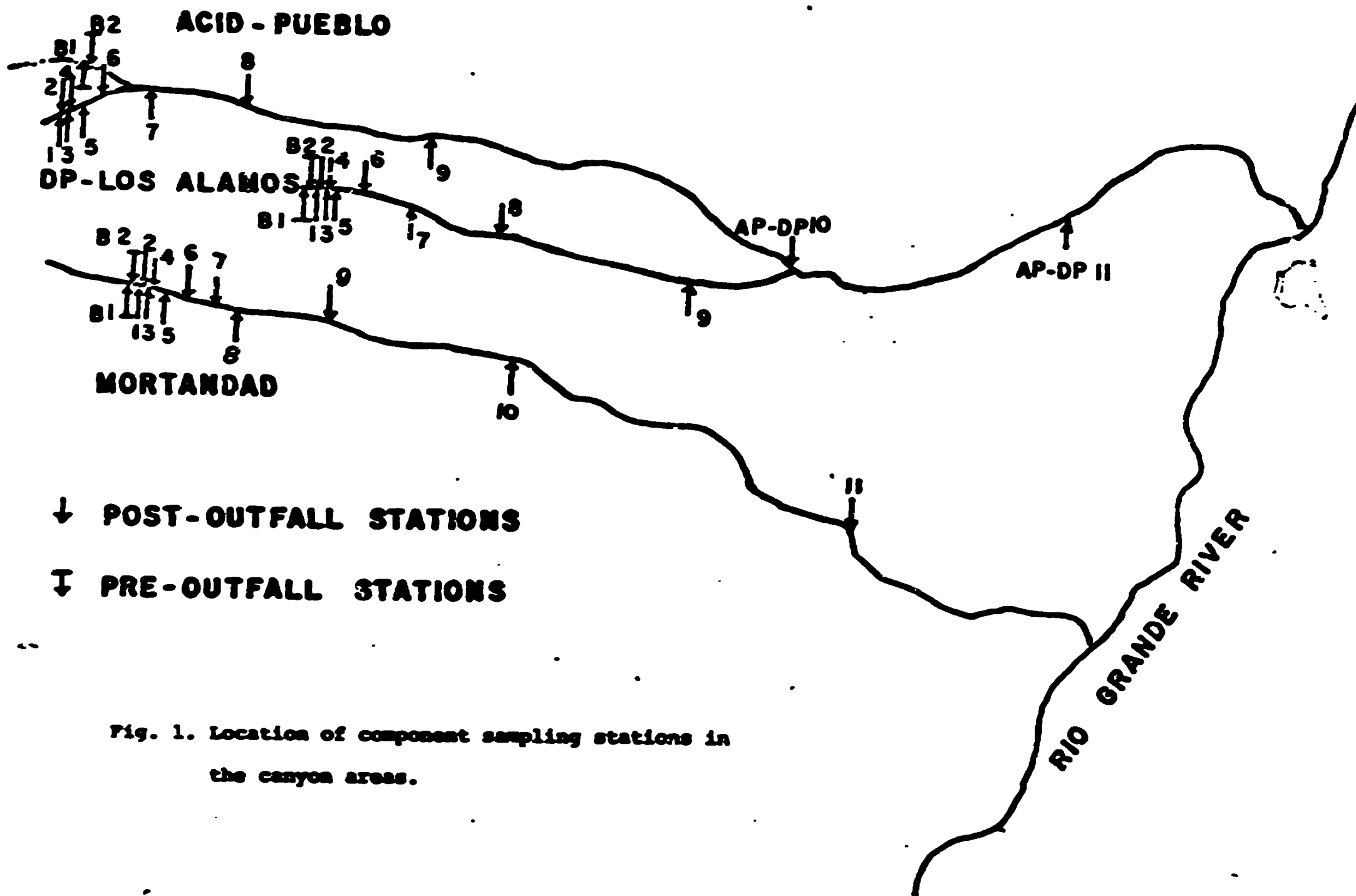


Fig. 1. Location of component sampling stations in the canyon areas.

located on a mesa adjacent to Mortandad Canyon enter the stream channel at an elevation of 2200 meters (i.e., near the origin of the canyon). The input of waste water over the last 10 years has been relatively constant at about 200 kiloliters per day. The effluent water along with a continuous supply of uncontaminated water ( $\approx 50$  kl/day) from a steam plant situated at the head of the canyon, moves as surface water over thin alluvial deposits ( $< 30$  cm deep) for a distance of from about 500 - 1300 meters below the effluent outfall. The effluent disappears into the alluvium and the remainder of the stream channel is dry at distances beyond 1300 meters where the canyon and stream channel widen with a corresponding increase in alluvium depth ( $> 30$  cm).

It was estimated that about 40 mCi of  $^{239-240}\text{Pu}$  was released into Mortandad Canyon from 1963 to 1973, and furthermore, that since 1970 at least 80 percent of the plutonium activity was  $^{239}\text{Pu}$  (i.e.,  $^{239}\text{Pu}/^{240}\text{Pu} \approx 4$ ).

A permanent sampling network was established in the canyon during the summer of 1972 (Fig. 1), at points 100 and 200 m above the waste discharge outfall (pre-outfall) to serve as a source of "background" samples and also at 0, 20, 40, 80, 160, 320, 640, 1280, 2460, 5120, and 10,240 m below the outfall (post-outfall). Considerably more sampling emphasis was placed on the areas immediately below the outfall since plutonium concentration gradients were expected to change rapidly in this region. The data presented in this paper were obtained from a sampling effort which was

made during a one week period in October 1972.

Surface and ground water samples were obtained at several locations down the length of the canyon and a 500 ml aliquot of each sample was filtered through Whatman 40 filter paper prior to analysis. Surface samples were collected at 0, 160, and 320 m post-outfall and subsurface samples were collected at 640 (4), 1280 (11), 2560 (12), and 5120 (21) m post-outfall. The parenthetical values indicate the depth from which subsurface water was collected.

Since the alluvial sediments in the canyon are generally very sandy (and not rocky), it was decided that a core sampling device, which was designed to collect a maximum of about 300 grams of sediment, would best suit the objectives of the study. A disposable section of 2.4 cm diameter plastic pipe, which was chosen as the coring device, was sharpened on one end and was gently driven into the sediment to a maximum depth of about 30 cm with a hammer. The coring device, which was rotated as it was being driven into the ground, compacted the contained core by about 10 percent or less. The sampling depth of each core, in many cases, was less than 30 cm especially in the upper portions of the canyons where the sediments were shallow.

Each core sample was frozen and was sectioned into a 0 - 2.5 cm layer, a 2.5 - 7.5 cm layer, a 7.5 - 12.5 cm layer and the remainder (i.e. below 12.5 cm) to provide data on the vertical distribution of plutonium.

A sample of the most abundant grass, shrub, and tree

species was collected at each sampling station. Only those species which were directly rooted in the stream channel or that were likely to be inundated during high runoff periods were sampled. The samples consisted of the complete above ground portions of the grasses and the terminal leaves and stems of the shrub and tree species. Dust residues were not removed from the exterior of the plant surfaces prior to assay for plutonium.

Since it was not practical from a time and analytical standpoint to sample small mammals at each station, collections were made only in the two areas above the effluent outfall (i.e., 100 and 200 meters pre-outfall) and at distances of 0, 2560, and 10,240 meters post-outfall. 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. Peanut butter was used as bait and each station was trapped for three nights (i.e., 288 trap-nights) without pre-baiting treatment. Rodent samples were dissected into four portions including the lung, liver, hide, and the eviscerated and skinned carcass. Species caught included Peromyscus maniculatus, P. truei and Reithrodontomys megalotis.

Tritium and  $^{137}\text{Cs}$  concentrations were determined in all sample materials utilizing liquid scintillation and gamma spectroscopic techniques. All sample materials to which tracer quantities of  $^{242}\text{Pu}$  and  $^{243}\text{Am}$  were added, were subjected to a hydrofluoric-nitric acid leach, an ion exchange

separation, electrodeposition, and alpha-ray spectroscopy to quantify the plutonium content. The total sample was carried through the chemical procedures to eliminate any errors associated with aliquoting a complex matrix such as sediment.

The plutonium content of all sample materials except rodents was sufficient to reduce the relative standard deviations of the determination to less than 30 percent (i.e., based on counting statistics). However, the generally low levels of plutonium in rodent tissues in combination with the small sample masses resulted in relative standard deviations usually in excess of 30 percent. The minimum detectable amount of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  based on a 23 hour count was 0.03 pCi/sample (at 95 percent confidence).

## RESULTS AND DISCUSSION

The plutonium content of water, vegetation and the 0 - 2.5 cm layer of the alluvial sediments as a function of distance from the effluent outfall in Mortandad Canyon is presented in Figures 2 and 3. Since the composition of the vegetation changed with distance down the canyon, it was not possible to observe plutonium concentration gradients in any one species. Consequently, the data for plant samples were grouped according to the growth form of the species (i.e., grasses, shrubs and trees) and the grouped data were plotted as a function of distance post-outfall. Obviously, grouping in this manner disregards individual species

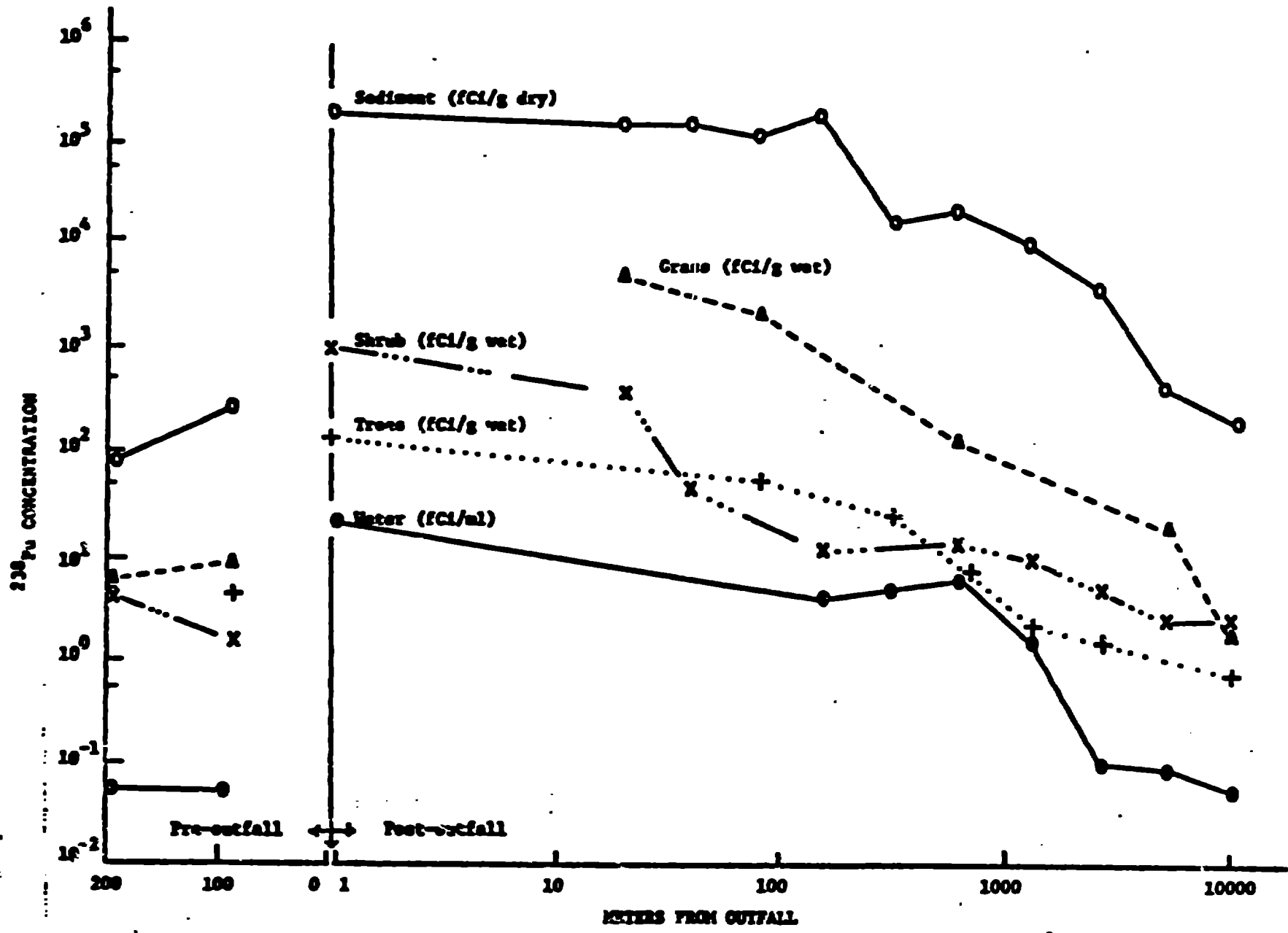


Figure 2. The  $^{238}\text{Pu}$  concentrations in sediment (0-2.5 cm layer), vegetation and water from Hordand Canyon in October 1972.



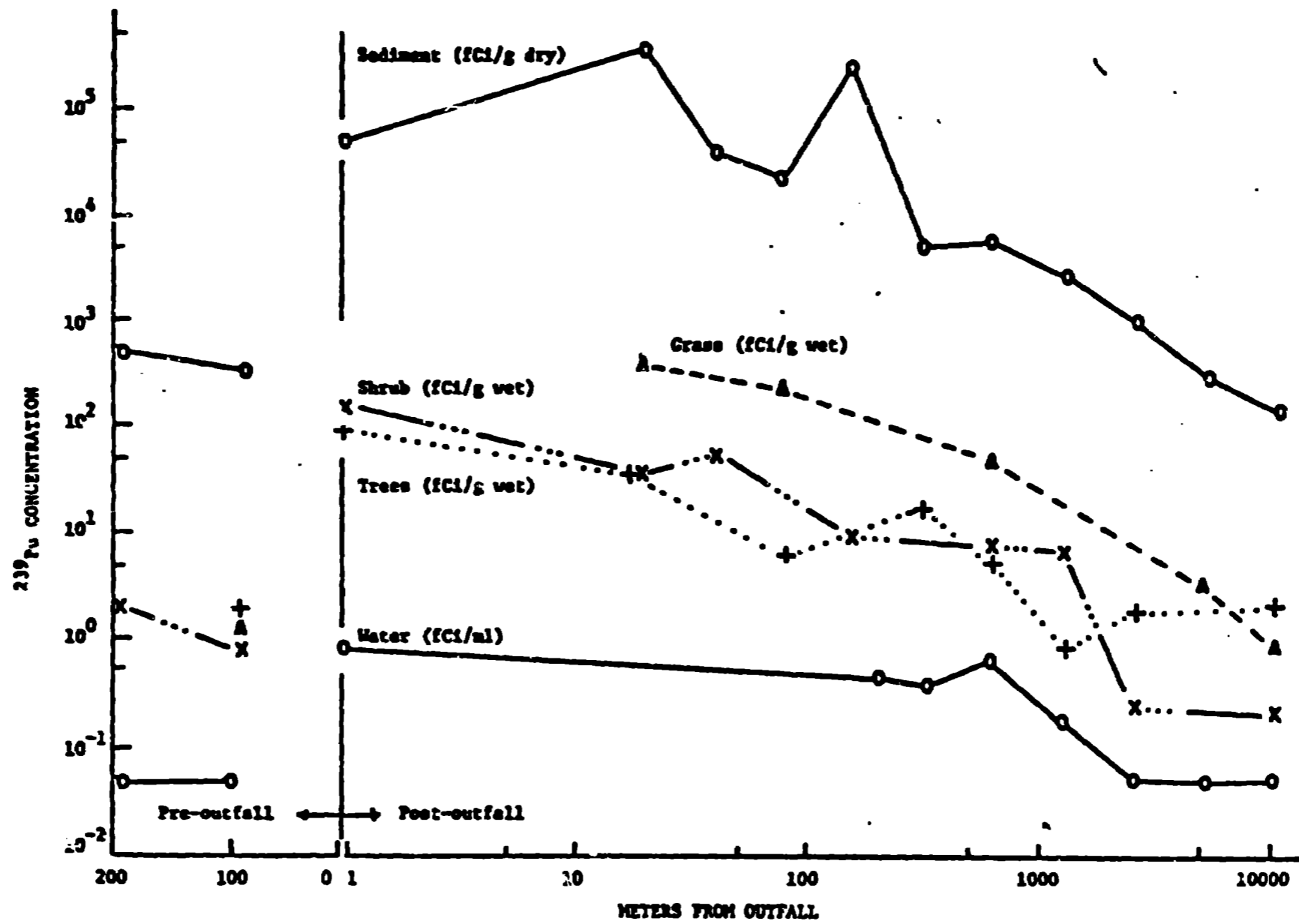


Figure 3. The  $^{239}\text{Pu}$  concentrations in sediment (0-2.5 cm layer), vegetation and water from Mortandad Canyon in October 1972.

variation. All the grass samples analyzed were from the genus Poa with the exception of the 5120 meter post-outfall sample which was Bouteloua gracilis. The shrub category consisted of Artemesia tridentata, Berberis fendleri, Chrysothamnus parryihowardi, Quercus gambelli, Prunus virginiana, Salix spp. and Rhus trilobata. Tree samples included Acer negundo, Juniperus monosperma, Pinus ponderosa, Pinus flexilis and Pseudotsuga taxifolia.

It is apparent from the data in Figures 2 and 3 that the chronic input of low level radioactive liquid wastes into Mortandad Canyon over the last 10 years has resulted in plutonium concentrations in some post-outfall samples which are two to three orders of magnitude higher than corresponding pre-outfall samples. The maximum concentrations of plutonium in all samples were observed within 160 meters post-outfall and thereafter concentrations declined steadily with distance. Concentrations of both isotopes in all samples had dropped to near pre-outfall levels at the 5120 and 10,240 meter post-outfall sampling stations.

It appears that the movement of plutonium down the canyon over the last 10 years has been a slow process because the elevated levels of plutonium in all sample types were confined to the area between the effluent outfall and about 2500 meters post-outfall. This contamination pattern is not surprising when one considers the physical characteristics of the canyon in this area. Recall that the stream is dry below 1200 meters post-outfall and that the canyon

Table 1. The vertical concentration gradients of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  in sediments from Mortandad Canyon in October 1972.

Meters From Effluent Outfall		$^{238}\text{Pu}$ (pCi/g dry)				$^{239}\text{Pu}$ (pCi/g dry)			
		Increment (cm)				Increment (cm)			
		0-2.5	2.5-7.5	7.5-12.5	> 12.5	0-2.5	2.5-7.5	7.5-12.5	>12.5
100	pre-outfall	0.075	0.036	0.026	None	0.52	1.8	0.36	None
200	"	0.26	0.04	0.021	0.014	0.31	0.21	0.27	0.23
0	post-outfall	190.	49.	None	None	49.	None	None	None
20	"	144.	205.	9.9	None	314.	54.	0.96	None
40	"	144.	138.	124.	None	38.	62.	66.	None
80	"	106.	85.	19.	0.86	20.	25.	23.	2.6
160	"	178.		142.	328.	216.		28.	29.
320	"	15.	24.	24.	27.	4.4	4.4	4.4	3.3
640	"	20.	18.	15.	17.	5.0	6.9	4.2	4.7
1280	"	9.7	23.	11.	5.3	2.2	2.7	3.7	2.8
2560	"	3.4	9.8	7.8	0.45	0.83	2.7	2.2	0.49
5120	"	0.38	ND	0.15	0.016	0.27	ND	0.098	0.023
10240	"	0.18	0.028	0.020		0.13	0.023	0.0090	

(and stream channel) widen in this area and the alluvial deposits deepen. The capacity of the alluvium in the wider portion of the canyon apparently is sufficient not only to absorb the effluent water but also to absorb most of the large quantities of runoff which occurs after heavy rainfall. The plutonium in samples from the 2560 meter station likely was carried there from the upper part of the canyon by runoff but other plutonium redistribution mechanisms may be operative (e.g., wind resuspension).

Stream channel sediments clearly are the major reservoir of the waste plutonium (Figs. 2 and 3 and Table 1). Levels of both isotopes increased from less than 0.5 pCi/g (dry) at the pre-outfall stations to over 300 pCi/g in post-outfall samples. The concentrations of both  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  were relatively uniform to sampling depths of 30 cm from the effluent outfall to the 1280 meter post-outfall sampling station where surface water exists for at least part of the year.

The degree of vertical mixing of plutonium in the alluvial sediments in the canyon appears to be associated with the presence or absence of surface water. Whether the water physically mixes the sediments or acts as a medium for diffusion of plutonium is unknown at this time.

In arid ecosystems, the downward migration of plutonium is limited and in most cases the inventory is confined to the upper 2.5 cm (Mork, 1970; Olafson et al., 1957). There is some evidence, as reviewed by Francis (1973) that  $^{239}\text{Pu}$

does migrate downward in soils after extended exposure to the natural environment and consequently may become more available to vegetation with time because of an enhanced root contact with the isotope.

There is an interesting distributional relationship between the  $^{137}\text{Cs}$  and plutonium content of the alluvial soils. A ln-ln plot of the  $^{137}\text{Cs}$  concentrations in the 0-2.5 cm layer of post-outfall sediments versus the corresponding data for  $^{238}\text{Pu}$  (and  $^{239}\text{Pu}$ ) were linearly related. The correlation coefficients ( $r$ ) for the respective linear regressions were 0.97 ( $^{137}\text{Cs}$  vs  $^{238}\text{Pu}$ ) and 0.88 ( $^{137}\text{Cs}$  vs  $^{239}\text{Pu}$ ). The interpretation of this observation is not clear at this time, but may indicate that the distributive mechanism for these two radionuclides in sediments may be similar.

The plutonium content of surface and ground water in the stream channel paralleled the data for sediments but at a much lower level (Figs. 1 and 2). Maximums of 29 fCi  $^{238}\text{Pu}/\text{ml}$  and about 1 fCi  $^{239}\text{Pu}/\text{ml}$  were observed in surface water at the effluent outfall. However concentrations of both isotopes had decreased to less than 0.1 fCi/ml in ground water at the 5120 m post-outfall sampling station. The ratio of  $^{238}\text{Pu}$  in the first three post-outfall surface water samples and the corresponding 0 - 2.5 cm layer of sediment (i.e., pCi  $^{238}\text{Pu}/\text{ml}$  : pCi  $^{238}\text{Pu}/\text{g}$  (dry)) averaged  $1.4 \times 10^{-4}$ . The corresponding value for  $^{239}\text{Pu}$  was  $3.3 \times 10^{-5}$ .

There have been major changes in the chemistry of the effluent water over the past 10 years. During the first seven years of effluent disposal into Mortandad Canyon when  $^{239}\text{Pu}$  was the major constituent the liquid wastes were released at about pH 11. Within the last three years when  $^{239}\text{Pu}$  was the major constituent, the pH of effluent water has dropped to 8-9. Sediment pH was measured at each sampling station in the present study and the values increased from 6.5 in pre-outfall sediments to 8.6 in the first 160 meters post-outfall. The pH of sediment at the 5120 and 10,240 meter stations had dropped to about 7. The literature is inconclusive about the influence of pH on plutonium discrimination factors (i.e. conc/g water or plant / conc/g soil for liquid-soil and plant-soil systems (Wilson and Cline, 1966; Rhodes, 1957; Rediske et al., 1955; Newbould, 1963).

There appears to be a relationship between the proximity of the vegetation to ground surface and the plutonium content of the plant material. In general, grass species which were <50 cm tall, contained the highest levels of both  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ , whereas shrubs and trees (>1 m tall) contained relatively moderate to low amounts of plutonium. If one assumes that the source of plutonium to the vegetation is the effluent water (i.e. root uptake of plutonium) then there would appear to be a concentration of both  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  in most vegetation samples. However, if the sediments were the source of plutonium to the vegetation

then root uptake factors or resuspension factors (i.e. depending on the contamination mechanism) were relatively low. The ratio of  $^{239}\text{Pu}$  in post-outfall vegetation and the 0 - 2.5 cm layer of the sediments (i.e.  $\text{pCi } ^{239}\text{Pu/g}$  vegetation (wet) /  $\text{pCi } ^{239}\text{Pu/g}$  sediment (dry)) averaged  $2.3 \times 10^{-3} \pm 1.6 \times 10^{-3}$ ,  $2.5 \times 10^{-3} \pm 2.4 \times 10^{-3}$  and  $9.9 \times 10^{-4} \pm 1.2 \times 10^{-3}$  for grasses, shrubs and trees, respectively. Corresponding values for  $^{239}\text{Pu}$  in grasses, shrubs and trees were  $7.8 \times 10^{-3} \pm 4.2 \times 10^{-3}$ ,  $4.0 \times 10^{-3} \pm 5.1 \times 10^{-3}$  and  $3.4 \times 10^{-3} \pm 5.3 \times 10^{-3}$ , respectively.

Grass/sediment ratios of  $2.3 \times 10^{-3}$  and  $7.3 \times 10^{-3}$  (or about  $8 \times 10^{-3}$  and  $3 \times 10^{-2}$  on a dry weight basis) for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  are relatively high compared to values of  $10^{-6} - 10^{-3}$  reported by other investigators for the root uptake of Pu from plant-soil systems. (Newbould, 1963; Cummings and Roberts, 1971; Rediske et al., 1955; Wilson and Cline, 1966). It is possible that the high ratios for grass/sediment in the present study have resulted from external contamination of the plant material and not from physiological uptake of plutonium.

The plutonium concentrations in the liver, lungs, hide and carcass of rodents collected on the stream channel in Mortandad Canyon (Table 2) varied by as much as three orders of magnitude in samples from the same collection location. Some of this variation is undoubtedly due to the large uncertainties associated with the counting data (i.e. low plutonium content and small sample masses) and to some

Table 2. The  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  content of rodents from Montedison Canyon in October 1972.

Location (meters)	$^{238}\text{Pu}$				$^{239}\text{Pu}$			
	Liver	Lung	Blood	Carcass	Liver	Lung	Blood	Carcass
100 and 200 (pre-outfall)	2.8 (2.8)	9.9 (17) <sup>a</sup> -37. (28)	-1.0 (1.4) -0.7 (0.7)	0.0 (3.0) -1.4 (1.4)	0.92 (2.8)	3.3 (6.6)	4.1 (1.7)	0.0 (3.0) 0.0 (1.6)
$\bar{x} \pm 1 \text{ S.D.}^{\text{ab}}$	$2.8 \pm 2.8$	$5 \pm 7$	$0.0 \pm 0.0$	$-0.7 \pm 1.0$	$0.92 \pm 2.8$	$5.7 \pm 3.4$	$9.6 \pm 7.1$	$0.0 \pm 0.0$
0 (post-outfall)	28 (30) 17. 45. (15) 22. (6.3) 5.4 (4.3)	148. (121) 63. 1119. (149) 24. (34) 11. (30)	1307. (126) 910. (70) 594. (65) 610 (70) 25. (8.2)	88. (3.9) 25. (4.1) 0.15 (0.22) 31. (4.1) 7.5 (1.6)	4.2 (3.1) 17. 21. (10) 4.2 (2.1) 3.2 (3.2)	74. (74) 63. 407. (102) 69. (60) 7.6 (15)	109. (28) 110. (70) 57. (15) 116. (25) 41. (8.2)	11. (12) 16. (1.8) 0.07 (0.22) 1.3 (2.0) 4.0 (1.2)
$\bar{x} \pm 1 \text{ S.D.}$	$75 \pm 16$	$326 \pm 533$	$609 \pm 471$	$43 \pm 43$	$8.2 \pm 8.6$	$137 \pm 182$	$103 \pm 58$	$6.5 \pm 6.8$
2560	19. (13) 7.9 (4.5) 0.03 (3.9) 15. (6.1) 0.0 (14) 0.0 (3.0) 2.2 (3.3) 7.1 (5.9)	3467. (1367) 410. (117) 19. (23) 67. (135) 16. (27) 18. (24) 32. (23) 53. (31)	309. (170) 26. (22) 18. (4.3) 24. (4.5) 3.8 (15) 15. (9.3) 0. (4.4) 9.7 (5.0)	0.06 (0.71) 2.0 (0.57) 3.4 (0.95) 1.6 (0.67) 0. (1.6) 13. (1.6) 0.81 (0.54) -1.3 (0.72)	7.4 (8.9) 3.4 (2.3) 0. (2.0) 3.5 (4.3) 14. (9.1) 0.0 (2.2) 4.5 (10) 1.2 (2.4)	501. (364) 111. (65) 0.0 (16) 27. (47) -9.1 (9.1) -6.0 (12) 0.0 (18) 4.4 (8.8)	198. (75) 4.3 (8.7) 4.7 (1.8) 3.1 (1.4) 7.6 (7.6) 5.6 (5.6) 4.0 (3.1) 6.1 (2.9)	1.0 (0.71) 0.29 (0.29) 1.8 (0.57) -0.34 (0.34) -0.46 (0.69) 2.0 (7.1) 0.40 (0.40) 0.0 (0.62)
$\bar{x} \pm 1 \text{ S.D.}$	$6.4 \pm 7.3$	$761 \pm 906$	$75 \pm 176$	$2.7 \pm 4.3$	$4.3 \pm 4.7$	$80 \pm 174$	$29 \pm 68$	$0.69 \pm 0.82$
10240	20. (11) 0. (10) 11. (19) 1.4 (2.7) -2.0 (2.8) -34. (34) 0. (8.8) 87. (104) 0.96 (1.9) 0. (3.5) -11. (5.6) 7.2 (9.5)	487. (133) 15. (19) 7794. (1790) 23. (29) 0. (17) 12. (20) 0. (29) 18. (31) 942. (604) -12. (36) 4.5 (9.3) 15. (29) 44. (30)	30. (22) 130. (100) 15. (31) 7.7 (7.7) 7.5 (5.3) 7.9 (3.8) 3.0 (4.8) 4.2 (5.5) 6.8 (5.2) -4.7 (5.3) 0.33 (1.3) 3.0 (4.1) 14. (11)	5.6 (1.9) 0.30 (0.77) 77. (6.8) 1.5 (0.65) -0.67 (1.9) 0.10 (0.20) 0.37 (0.37) 0.0 (1.8) 2.6 (1.1) 0.92 (0.57) 0.31 (0.31) 0.30 (0.36) 13. (7.6)	2.5 (3.8) 4.1 (7.1) 13. (9.7) 0.69 (2.1) 1.9 (1.9) 0.0 (34) 5.9 (5.9) 121. (52) 0.96 (1.9) 0.0 (2.8) 1.4 (4.2) 4.8 (7.2)	58. (50) 0.0 (3.7) 0.0 (1790) 6.4 (16) 22. (13) 0.0 (16) 21. (14) 4.4 (13) 172. (673) 16. (20) 23. (14) 51. (22) 0.0 (15)	0.0 (22) 6115. (1439) 8.7 (15) 12. (7.2) 0.0 (2.6) 4.3 (3.0) 4.0 (3.0) 26. (6.4) 10. (6.2) 4.1 (4.4) 6.2 (2.3) 3.3 (2.2) 9.7 (7.1)	0.93 (1.9) 0.77 (0.48) 15. (2.6) 0.0 (0.24) -0.05 (0.89) 1.3 (0.51) -0.37 (0.55) 1.8 (1.8) 2.6 (1.1) 1.3 (0.57) 0.31 (0.41) 0.51 (0.51) 0.0 (8.4)
$\bar{x} \pm 1 \text{ S.D.}$	$11 \pm 25$	$700 \pm 2228$	$22 \pm 49$	$7.9 \pm 21$	$13 \pm 34$	$136 \pm 1153$	$477 \pm 1694$	$1.9 \pm 4.0$

<sup>a</sup> (CI/g wet  $\pm 1 \text{ S.D.}$  based on counting statistics.

<sup>ab</sup> The mean was calculated by assigning a 0.0 to any negative value and by disregarding any less than (<) value.



degree due to species variation. There were insufficient samples at each station to permit any species comparison.

Relatively high concentrations of about 4 - 7 pCi/g (wet) were observed in the lungs and hide of four rodents (all four were Peromyscus)(spp), one which was collected from the 2560 meter post-outfall station and three from the 10,240 meter station. Most of the remaining plutonium concentrations measured 1 pCi/g or less. Concentrations of  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  in pre-outfall samples and from other locations on site measure 10 fCi/g wet or less (Herczeg, 1972).

The mean plutonium concentration for each tissue type and collection location was highest for the lung and hide and lead one to speculate that inhalation may be the main route of entry to these small, ground dwelling rodents.

Recall that the carcass sample was comprised of all the bone and skeletal muscle from each rodent. Since about 90 percent of gut absorbed plutonium is associated with the skeleton (Katz et al., 1955; Weeks et al., 1956), the concentrations in rodent skeletons could have been a factor of four higher than those listed for the whole carcass, assuming that the skeleton comprised 20 percent of the carcass weight.

If the four high plutonium concentrations mentioned earlier are not included in the calculation of the means, then the mean plutonium concentration in all rodent tissues as a function of distance post-outfall closely follow the patterns which were observed in sediment, water and vegeta-

tion. Mean values were highest at the outfall and then decreased with distance post-outfall.

It is of interest to calculate the  $^{238}\text{Pu}/^{239}\text{Pu}$  ratio in the various sample types since these ratios in vegetation and rodents may reflect the source of contamination. It is apparent from the ratios presented in Table 3 that the effluent water contains a preponderance of  $^{239}\text{Pu}$ , a fact which was mentioned earlier. Not only does the effluent water reflect the greater  $^{239}\text{Pu}$  inputs but so do all the remaining sample types.

The fact that the  $^{238}\text{Pu}/^{239}\text{Pu}$  ratio exceeds 1 in some of the post-outfall remainder sediment core sections (i.e. from depth  $>17.5$  cm) indicates that the complete vertical mixing of  $^{238}\text{Pu}$  has occurred over the last three years. Recall that there is surface water in the canyon down to about 1200 meters post-outfall.

The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios greater than unity in sediments from the 5120 and 10,240 meter stations may indicate that some of the plutonium in the canyon has moved a considerable distance down Mortandad Canyon, despite the low sediment concentrations measured at these sites. Another possible explanation is that plutonium associated with atmospheric effluents from a Laboratory facility are modifying the  $^{238}\text{Pu}/^{239}\text{Pu}$  ratio upward.

Vegetation and rodent tissue seem to reflect the enhanced  $^{239}\text{Pu}$  content of water and sediment in the canyon but not in any readily identifiable pattern.

Table 3. The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios in water, sediment, vegetation, and rodents from Mortandad Canyon in October 1972.

Distance From Outfall	$^{238}\text{Pu}/^{239}\text{Pu}$											
	Water	Sediment				Vegetation			Rodents			
		0-2.5	2.5-7.5	7.5-12.5	>12.5	Grass	Shrub	Trees	Liver	Lung	Hide	Carcass
200		0.14	0.02	0.07	—	4.6	2.1					
100		0.83	0.15	0.08	0.06		1.7	1.3	3.0	3.0		
0	29	3.9	—	—	—		4.2	1.6	3.5	1.4	7.0	7.8
20		0.46	3.8	10.	—	14.	7.0					
40		3.8	2.2	1.9	—		2.2					
80		5.3	3.4	0.83	0.33	10.		8.3				
160	8.4	0.82	—	5.1	11.3		1.1					
320	12.	3.4	5.1	5.1	8.1			1.4				
640	9.5	4.0	2.6	3.6	3.6	2.8	1.9	1.1				
1280	8.2	4.4	8.5	3.0	1.9		1.3	3.3				
2560	1.8	4.1	3.6	3.5	0.92		1.9	1.2	2.3	6.8	3.1	3.2
5120	>1.6	1.4	—	1.5	0.70	5.7	2.6					
10240		1.4	1.2	2.2	—	1.8	1.0	0.32	2.3	4.5	6.5	3.8

## CONCLUSIONS

The release of low level plutonium wastes in liquid effluents has resulted in elevated plutonium concentrations in several of Mortandad Canyons ecological components. The alluvial soils in the mezic portion of the canyon, contains nearly all of the  $^{238}\text{Pu}$ - $^{239}\text{Pu}$  which has been released over the last 10 years. It is speculated that the major vector in the transport of sediment bound plutonium into the xeric portion of the canyon is runoff which occurs after heavy rains. However, the morphology of Mortandad Canyon is such that it would likely take an extremely heavy rain to move any significant quantities of plutonium down the entire length of the stream channel. The generally wet nature of the stream channel in the areas of relatively high plutonium contamination coupled with the dense vegetative cover along the stream bank would seem to preclude wind redeposition of plutonium.

The presence of water in the stream channel appears to be correlated with the rate and degree of vertical mixing of plutonium in the alluvial sediments. The mechanisms involved are not understood but may include the mixing action of the flowing water and/or the water may serve as a medium for the vertical diffusion of plutonium.

The effect that complete vertical mixing of the plutonium in sediments (i.e. down to bedrock or 30 cm) has on the availability of this radionuclide to vegetation is unknown. Our

data on low growing grass species show that the plutonium concentration ratios for plant/sediment are in the order of  $3 \times 10^{-2}$  -  $8 \times 10^{-2}$ , which is about an order of magnitude higher than that reported by others for root uptake of plutonium from soils. However, in the present study we cannot rule out the possibility of externally deposited plutonium on the plant materials. There does appear to be a relationship between growth form and the plutonium content of the plant. Lower growth forms contained higher plutonium concentrations than larger forms. The change in plutonium concentration in plant samples as a function of distance post-outfall closely follows the pattern observed in water and sediments.

The highest mean  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  concentrations in the lung and hide of rodents from the canyon suggest that resuspension of sediment bound plutonium may be a prime mechanism in the contamination of rodents. The fact that the species of rodents sampled in the present study dwell underground and, of course, are in intimate contact with the ground surface probably indicates that the resuspension process occurs on a micro-scale.

The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios calculated from the data in the present study had some utility in assessing the vertical and horizontal movement of the effluent associated plutonium. It appears that in the mezie portion of the canyon, the vertical mixing to the depths sampled was completed within a three year period. The  $^{238}\text{Pu}/^{239}\text{Pu}$  ratios in vegetation

and rodents reflected the ratio in water and sediments but in a manner which has yet to be defined.

The tremendous spread in the plutonium data for rodent tissues indicate that the contamination of the small mammal populations living near the stream channel is heterogeneous with many individuals receiving minute quantities of plutonium and others receiving relatively large amounts.

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