THE DISTRIBUTION OF PLUTONIUM IN LIQUID WASTE DISPOSAL AREAS AT LOS ALAMOS

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Abstract

This paper describes an ecological investigation of plutonium in the Los Alamos Scientific Laboratory environs. Data are presented on the distribution of plutonium in the alluvial sediments, water, vegetation and rodents from Mortandad Canyon, an area which has been used for liquid waste disposal for 10 years.

Introduction

A survey was initiated during 1972 to determine the concentrations of liquid effluent-associated radionuclides in the alluvial sediments, water, and some of the natural biota in waste discharge areas. This paper will summarize preliminary findings on the distribution of ^{238}Pu and ^{239}Pu in Mortandad Canyon, an area which has been used as a liquid effluent disposal area since 1963. The data were obtained during a one-week sampling period in October 1972.

Methods and Materials

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 at an elevation of about 1700 meters.

Radionuclide-bearing liquid effluents from a waste treatment plant located on a mesa adjacent to Mortandad Canyon enter the stream channel at an elevation of 2200 meters 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 (post-outfall). The effluent disappears into the alluvium and the remainder of the stream channel is dry at distances beyond 1300 meters post outfall, where the canyon and stream channel widen with a corresponding increase in alluvium depth (>30 cm).

It was estimated that about 40 mCi of $^{238,\ 239}Pu$ was released into Mortandad Canyon from 1963 to 1973; and furthermore, that since 1970 at least 80 percent of the plutonium activity was ^{238}Pu ($^{238}Pu/^{239}Pu = 4$).

A permanent sampling network was established in the canyon during the summer of 1972 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, 2560, 5120, and 10, 240 m below the outfall (post-outfall).

Surface and ground water, sediment, vegetation and rodents were collected from the stream channel area in the canyon using collection techniques which have been described elsewhere.[1]

Samples were subjected to a hydrofluoric-nitric acid leach, an ion exchange separation, electrodeposition, and alpha-ray spectroscopy corrected for yield by use of tracer quantities of ^{242}Pu and ^{243}Am to quantify the plutonium content.

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 (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 greater than 30 percent. The minimum detectable amount of ^{238}Pu and ^{239}Pu based on a 23 hour count was 0.03 pCi/sample ($\alpha = 0.05$).

Results and Discussion

The ^{238}Pu 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 Fig. 1. The data for ^{239}Pu which behaved similar to that shown for ^{238}Pu in Fig. 1, can be inferred from $^{238}Pu/^{239}Pu$ activity ratios presented later in Table 2. The data for vegetation were grouped according to growth form (grasses, shrubs, and trees) and the grouped data were plotted as a function of distance post-outfall. All of the grass samples anayzed were of the genus *Poa* with the exception of the 5, 120 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 Fig. 1 that the chronic input of low level radioactive liquid wastes into Mortandad Canyon over the last 10 years has resulted in ^{238}Pu (and ^{239}Pu) concentrations in some post-outfall samples which are two to three orders of magnitude higher than corresponding pre-outfall samples. Maximum concentrations of plutonium in all samples occurred within 160 meters post-outfall and concentrations then declined steadily with distance to near pre-outfall levels at the 5, 120 and 10, 240 meter post-outfall sampling stations.

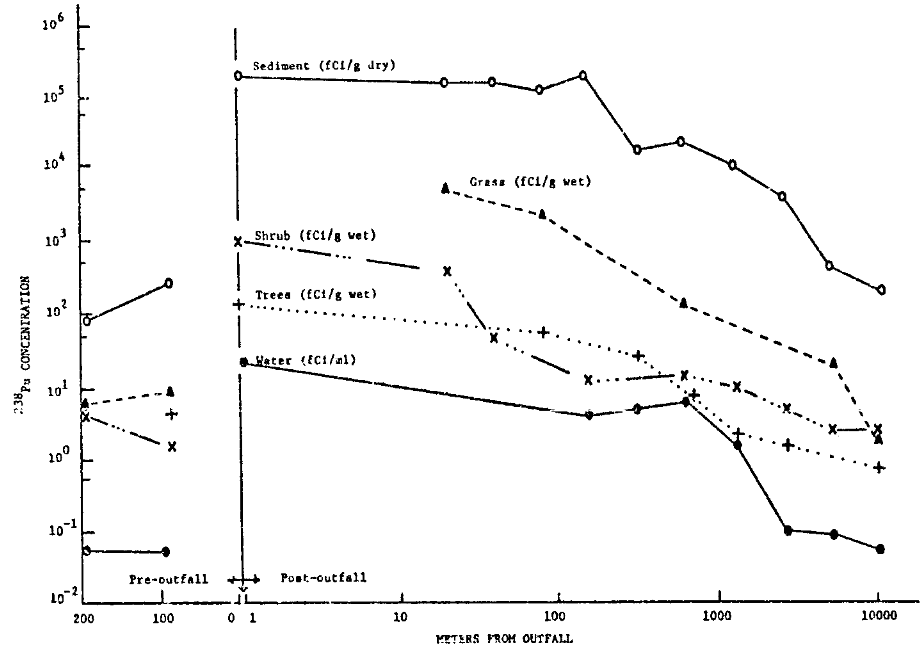

Figure 1. The ^{238}Pu concentrations in sediment (0-2.5 cm layer), vegetation and water from Mortandad Canyon in October 1972.

Stream channel sediments clearly are the major reservoir of the waste plutonium (Fig. 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 ^{238}Pu and ^{239}Pu were relatively uniform to sampling depths of 30 cm from the effluent outfall to the 1,280 meter post-outfall sampling station, where surface water exists for at least part of the year. At distances greater than 1,280 meters post-outfall, the plutonium was increasingly concentrated in the top 2.5 cm of sediment.

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.

There is some evidence, as reviewed by Francis[2], that plutonium does migrate downward in soils after extended exposure to the natural environment and may become more available to vegetation with time because of an enhanced root contact with the isotope. Studies at Trinity Site, which are described in a paper at this symposium demonstrated that there has been a marked penetration of Pu into the soils since 1950, when similar measurements were last made.

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

The plutonium concentration in surface and ground water in the stream channel paralleled the data for sediments but at a much lower level (Fig. 1). Maxima of 29 fCi ^{238}Pu/ml and about 1 fCi ^{239}Pu/ml were observed in surface water at the effluent outfall and concentrations of both isotopes decreased to less than 0.1 fCi/ml in ground water at the 5,120 m post-outfall sampling station.

There appeared to be a relationship between the proximity of vegetation to the 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 ^{238}Pu and ^{239}Pu, whereas shrubs and trees (>1 m tall) contained relatively moderate to low amounts of plutonium. Perhaps the rooting zone of the grasses contained higher Pu concentrations than the shrubs or trees or possibly the low growth form of the grasses increased their susceptability to exterior surface contamination by Pu contaminated materials.

If the plutonium measured in vegetation was contained within the plant, then calculated grass/sediment activity ratios of 2.3×10^{-2} and 7.8×10^{-3} (or about 8×10^{-2} and 3×10^{-2} on a dry weight basis) for ^{238}Pu and ^{239}Pu were relatively high compared to values of 10^{-6} - 10^{-3} reported by other investigators for the root uptake of Pu from plant-soil systems.[3,4,5,6] However, whether the plutonium was distributed within, or on, the plant is unknown at this time.

The plutonium concentrations in the liver, lungs, hide, and carcass of rodents (Peromyscus maniculatus, P. truei, Reithrodontomys megalotis) collected on the stream channel in Mortandad Canyon (Table 1) varied by as much as three orders of magnitude in samples from the same collection location. Some of this variation was undoubtedly due to the large uncertainties associated with the counting data and to species variation. There were insufficient samples at each station to permit any species comparison.

Mean plutonium concentrations were highest in the lung and hide samples from each collection location which suggested that inhalation of resuspended sediments may be the main route of Pu entry into these small, ground-dwelling rodents. Post-outfall concentrations varied from a maximum of about 8000 fCi ^{238}Pu/g wet in the lungs of one rodent down to levels which were indistinguishable from background. Concentrations of ^{238}Pu and ^{239}Pu in rodent tissues from pre-outfall and other areas[1] on site measured 10 fCi/g or less. Liver and carcass samples, in general averaged from 0.1-0.01 times the plutonium concentrations of hide and lungs.

The $^{238}Pu/^{239}Pu$ ratios for the various sample types, which are presented in Table 2, demonstrate that nearly all of the samples contained a preponderance of ^{238}Pu on an activity basis. The $^{238}Pu/^{239}Pu$ ratios which exceed one in some of the post-outfall remainder (>12.5 cm depths) sediment core sections evidence the fact that complete vertical mixing of ^{238}Pu has occurred over the last three years.

The $^{238}Pu/^{239}Pu$ ratios greater than unity in sediments from the 5,120 and 10,240 meter stations may indicate that some of the plutonium has moved a considerable distance down Mortandad Canyon, despite the low sediment concentrations measured at these sites.

Table 1. The ^{238}Pu and ^{239}Pu content of rodents from Mortandad Canyon in October 1972.

Location (meters)	^{238}Pu Liver	^{238}Pu Lung	^{238}Pu Hide	^{238}Pu Carcass	^{239}Pu Liver	^{239}Pu Lung	^{239}Pu Hide	^{239}Pu Carcass
100 and 200 (pre-outfall)		9.9 (17)*	-1.0 (1.4)	0.0 (3.0)		3.3 (6.6)	4.1 (1.7)	0.0 (3.0)
	2.8 (2.8)	-37. (28)	-8.7 (8.7)	-1.4 (1.4)	0.92 (2.8)	8.1 (16)	15. (5.8)	0.0 (1.6)
$\bar{X} \pm 1$ S.D.**	2.8	5 ± 7	0.0	0.0	0.92	5.7 ± 3.4	9.6 ± 7.1	0.0
0 (post-outfall)	28 (10)	148. (111)	1307. (126)	88. (3.9)	4.2 (3.1)	74. (74)	189. (28)	11. (12)
	<17.	<63.	910. (70)	88. (4.1)	<17.	<63.	110. (20)	16. (1.8)
	45. (15)	1119. (169)	594. (65)	0.15 (0.22)	21. (10)	407. (102)	57. (15)	0.07 (0.22)
	22. (6.3)	24. (54)	610 (70)	31. (4.1)	4.2 (2.1)	60. (60)	116. (25)	1.3 (2.0)
	5.4 (4.3)	11. (30)	25. (8.2)	7.5 (1.6)	3.2 (3.2)	7.6 (15)	41. (8.2)	4.0 (1.2)
$\bar{X} \pm 1$ S.D.	25 ± 16	326 ± 533	689 ± 471	43 ± 43	8.2 ± 8.6	137 ± 182	103 ± 58	6.5 ± 6.8
2560	19. (13)	5467. (1367)	509. (170)	0.86 (0.71)	7.4 (8.9)	501. (364)	198. (85)	1.0 (0.71)
	7.9 (4.5)	410. (117)	26. (22)	2.0 (0.57)	3.4 (2.3)	111. (65)	4.3 (8.7)	0.29 (0.29)
	0.03 (3.9)	19. (23)	18. (4.3)	3.4 (0.95)	0. (2.0)	0.0 (16)	4.7 (1.8)	1.8 (0.57)
	15. (6.1)	67. (135)	24. (4.5)	1.6 (0.67)	3.5 (4.3)	27. (47)	3.1 (1.4)	-0.34 (0.34)
	0.0 (14)	18. (27)	3.8 (15)	0. (1.6)	14. (9.1)	-9.1 (9.1)	7.6 (7.6)	-0.46 (0.69)
	0.0 (3.0)	18. (24)	12. (9.3)	13. (1.6)	0.0 (2.2)	-6.0 (12)	5.6 (5.6)	2.0 (7.1)
	2.2 (3.3)	32. (23)	0. (4.4)	0.81 (0.54)	4.5 (18)	0.0 (18)	4.0 (3.1)	0.40 (0.40)
	7.1 (5.9)	53. (31)	9.7 (5.0)	-1.3 (0.72)	1.2 (2.4)	4.4 (8.8)	6.1 (2.9)	0.0 (0.62)
$\bar{X} \pm 1$ S.D.	6.4 ± 7.3	761 ± 906	75 ± 176	2.7 ± 4.3	4.3 ± 4.7	80 ± 174	29 ± 68	0.69 ± 0.82
10240	20. (11)	487. (133)	38. (22)	5.6 (1.9)	2.5 (3.8)	58. (58)	0.0 (22)	0.93 (1.9)
	0. (10)	15. (19)	180. (108)	0.30 (0.77)	4.1 (7.1)	0.0 (3.7)	6115. (1439)	0.77 (0.48)
	11. (19)	7794. (1798)	13. (31)	77. (6.8)	13. (9.7)	0.0 (1798)	8.7 (13)	15. (2.6)
	1.4 (2.7)	23. (29)	7.7 (7.7)	1.5 (0.65)	0.69 (2.1)	6.4 (16)	12. (7.2)	0.0 (0.24)
		0. (17)	7.5 (5.3)	-0.67 (1.9)		22. (13)	0.0 (2.6)	-0.05 (0.89)
	2.8 (2.6)	12. (20)	7.9 (3.8)	0.10 (0.20)	1.9 (1.9)	0.0 (16)	4.3 (3.0)	1.3 (0.51)
	-34. (34)	0. (29)	3.0 (4.8)	0.37 (0.37)	0.0 (34)	21. (14)	4.8 (3.0)	-0.37 (0.55)
	0. (8.8)	18. (31)	4.2 (5.5)	0.0 (1.8)	5.9 (5.9)	4.4 (13)	26. (6.4)	1.8 (1.8)
	87. (104)	942. (404)	6.8 (5.2)	2.6 (1.1)	121. (52)	4172. (673)	10. (6.2)	2.6 (1.1)
	0.96 (1.9)	-12. (36)	-4.7 (5.3)	0.92 (0.57)	0.96 (1.9)	16. (20)	4.1 (4.4)	1.3 (0.57)
	0. (3.5)	4.5 (9.1)	0.33 (1.3)	0.31 (0.31)	0.0 (2.8)	23. (14)	6.2 (2.3)	0.31 (0.41)
	-11. (5.6)	15. (29)	3.0 (4.1)	0.38 (0.36)	1.4 (4.2)	51. (22)	3.3 (2.2)	0.51 (0.51)
	7.2 (9.5)	44. (30)	14. (11)	13. (7.6)	4.8 (7.2)	0.0 (15)	9.7 (7.1)	0.0 (8.4)
$\bar{X} \pm 1$ S.D.	11 ± 25	780 ± 2228	22 ± 49	7.9 ± 21	13 ± 34	336 ± 1153	477 ± 1694	1.9 ± 4.0

* fCi/g wet ± 1 S.D. based on counting statistics.

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

Vegetation and rodent tissue reflected the enhanced ^{238}Pu content of water and sediment in the canyon but not in any readily identifiable pattern. There was a tendency for the ratio in grasses to exceed the ratio in shrubs and trees.

The release of low level plutonium wastes in liquid effluents has resulted in significant plutonium concentrations in several of Mortandad Canyons ecological components. The presence of water in the stream channel appeared

Table 2. The $^{238}Pu/^{239}Pu$ ratios in water, sediment, vegetation, and rodents from Mortandad Canyon in October 1972.

Distance From Outfall	Water	Sediment 0-2.5	Sediment 2.5-7.5	Sediment 7.5-12.5	Sediment >12.5	Vegetation Grass	Vegetation Shrub	Vegetation Trees	Rodents Liver	Rodents Lung	Rodents Hide	Rodents Carcass
200		0.14	0.02	0.07	--	4.6	2.1					
100		0.83	0.19	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

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. Data on low growing grass species showed that the plutonium concentration ratios for plant/sediment are on the order of 3×10^{-2} to 8×10^{-2}, which was 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 appeared to be a relationship between growth form and the plutonium content of the plant. Lower growth forms contained higher plutonium concentrations than higher growth forms.

The highest mean ^{238}Pu and ^{239}Pu concentrations in the lung and hide of rodents from the canyon suggested that resuspension of sediment-bound plutonium may be a prime mechanism in the contamination of rodents. The appreciable variation in the plutonium data for rodent tissues indicated 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.

The $^{238}Pu/^{239}Pu$ ratios calculated from the data in the present study provided assessment of the vertical and horizontal movement of the effluent-associated plutonium. It appeared that in the mezic portion of the canyon, the vertical mixing to the depths sampled was completed within a three year period. The $^{238}Pu/^{239}Pu$ ratios in vegetation and rodents reflected the ratio in water and sediments but in a manner which has yet to be defined.

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