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ACCUMULATION AND TRANSPORT OF SOIL PLUTONIUM IN LIQUID WASTE DISCHARGE AREAS AT LOS ALAMOS

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Abstract

ACCUMULATION AND TRANSPORT OF SOIL PLUTONIUM IN LIQUID WASTE DISCHARGE AREAS AT LOS ALAMOS.

Plutonium inventory estimates for the surface 12.5 cm of soil in Mortandad Canyon did not reflect all the plutonium added to the canyon during a 7 month interval. The methods used in this study indicated that about 2 mCi ^{238}Pu and 0 mCi ^{239,240}Pu were added to the canyon during the interval, compared with known additions of 5.5 mCi ^{238}Pu and 0.4 mCi ^{239,240}Pu. The discrepancy likely was the result of the large sampling variability, indicating that inventory changes in this order (i.e. up to 17%) are not detectable with any certainty. However, factors other than sampling variability may be involved, including losses of plutonium to depths exceeding 12.5 cm. The relative distribution of plutonium within the canyon demonstrates that transport has occurred beyond the extent of surface water and that runoff from summer rainstorms can serve as a radionuclide transport vector in landscapes exhibiting these hydrologic features. There was a highly significant relationship between suspended sediment concentrations and total amounts of radioactivity in water. The flow rates achieved during the runoff event play an important part in determining the total amount of sediment and thus radioactivity transported downstream. The storm runoff event sampled during this study resulted in the downstream transport of about 1-2% of the sediment inventories of plutonium.

The soils component of terrestrial ecosystems appears to be the major reservoir of plutonium originating from both weapons and industrial sources [1-3]. Data at present available indicate that transport of plutonium within the soils component and from soils to biotic components can occur by physical processes with biological and chemical processes playing a largely undefined role in the redistribution of this element. Relatively high concentrations of plutonium in native vegetation and in pelt and lung samples from small ground-dwelling rodents are usually attributed to resuspension processes, whereby the plutonium associated with the soil is redeposited on external surfaces or is inhaled [4-6].

The purpose of this study was to investigate the accumulation and transport of plutonium in the sediments of Mortandad Canyon in Los Alamos, New Mexico. This area has received plutonium in treated liquid effluents since 1963. Data are presented on the horizontal distribution of plutonium and total inventories in stream channel sediments for two sampling periods spanning a seven-month interval in 1972 and 1973. Data were also gathered on the importance of storm runoff in the downstream transport of plutonium contaminated sediment.

METHODS AND MATERIALS

Study area

The Los Alamos Scientific 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 a pronounced elevational gradient in the east-west direction, was formed by successive ash flows from a volcanic crater 16 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. Summer rainfall accounts for about 75% of the annual precipitation. Drainage from the 113 km^2 Laboratory site is via the many canyons that divide the plateau.

Nearly all of the liquid wastes generated by the Laboratory since its beginning in 1943 have been collected by industrial waste lines, treated (since 1951) and released into one of three semi-drainage canyons including Mortandad Canyon (Fig. 1).

Near the waste outfall in Mortandad Canyon, at an elevation of about 2200 m, the stream channel is narrow (< 1 m), rocky and contains relatively thin layers of sediment (< 0.3 m). The stream channel slope averages about 0.1 in the 2000 m segment immediately below the waste outfall. Further downstream the channel widens to 2-3 m and the alluvial depth increases to a maximum of 30 m.

The volume of treated effluent entering the canyon averages about 2×10^5 l/d and has not fluctuated significantly since the treatment plant became operational in 1963. The effluent from the treatment plant rapidly soaks into the alluvium below the waste outfall and disappears underground about 800-1200 m post-outfall, with maximum downstream movement occurring during the winter months when ground surfaces are frozen.

Waste treatment plant records were used in estimating the amounts of plutonium released to the canyon since 1963 [7]. The isotopic composition of the effluent (i.e. $^{238}Pu/^{239,240}Pu$) was not measured until 1972. Mortandad Canyon received $^{239,240}Pu$ contaminated waste prior to 1968, but since then has received primarily ^{238}Pu.

Relatively large surface water flows occur in the canyon during storm runoff events. The runoff also rapidly soaks into the thick alluvial deposits and reaches post-outfall distances of about 3000 m. None of the runoff with its associated radioactivity has ever left Laboratory boundaries [7].

Sampling

A sampling transect was established in the canyon during October 1972 with stations (Fig. 1) at two locations above the waste discharge outfall to serve as control areas and also at 0, 20, 40, 80, 160, 320, 640, 1280, 2560, 5120 and 10240 m below the outfall. 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 driven into the sediment to a maximum depth of 30 cm. Each station was

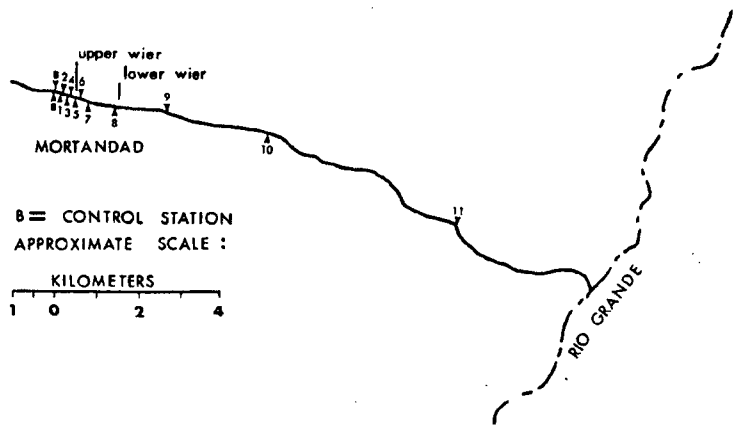

FIG. 1. Plutonium and ^{137}Cs sampling locations and water gauging stations in the three canyons receiving liquid effluents.

sampled at the centre and two lateral positions in the stream channel for a total of three cores per station. The sampling method resulted in about a 10% compaction of the contained core; a correction was not made for this error. 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 depths were about equivalent. Individual sediment samples were sealed in a plastic bag to prevent cross-contamination and were frozen until the radionuclide analyses could be initiated. The frozen core sample from the centre of the stream channel was cut into sections of 0-2.5, 2.5-7.5, 7.5-12.5 and > 12.5 cm to provide data on the vertical distribution of ^{137}Cs and plutonium. In May 1973, or about seven months later, the transect was resampled using the same techniques but with increased emphasis on replication [8].

The plutonium inventory was calculated for several segments of stream channel during both sampling periods using the sediment concentration data. The inventory estimates were calculated from:

$$I = C\,L\,W \qquad (1)$$

where I = plutonium in stream segment L (mCi);
C = average plutonium concentrations (mCi/m^2) at a particular sampling location;
L = length (m) of stream channel segment over which C applied; and
W = average width of stream (m) in segment L.

The length (L) of each segment about a sampling location was taken as half of the distance to the preceding station upstream plus half of the distance to the next station downstream. The plutonium inventory for the canyon was calculated from the sum of the activities (I) found in the respective channel segments.

A follow-up study was conducted on 15 September 1974 in Mortandad Canyon to determine the importance of storm runoff in the downstream transport of plutonium and ^{137}Cs with stream channel sediments. The runoff from one rainstorm occurring on the upper watershed was sampled frequently to determine the association of each radionuclide with the suspended sediment and liquid fractions and the total activities transported by the event.

Flow characteristics of the runoff were measured at two stream gauging stations during the event (Fig. 1). The gauging stations were equipped with a clock-driven water stage recorder, set up over a 0.152 m modified Parshall flume for low flow and a 1.22 m wier for large runoff events.

Samples of water and suspended sediments were taken during a 4.25-hour period at the lower gauging station, which was located 1200 m below the waste outfall and about 1000 m below the upper gauging station. Sample collections were begun 5 minutes after the leading edge of the runoff passed through the lower gauging station.

A DH-48 stream flow sampler was used to collect duplicate water samples in the centre of the stream about 6 cm below the surface and 6 cm above the bottom of the flow. Subsequent reference to samples from the two depth zones will be as surface and bottom samples. The sampler collected suspended particulates up to 6 mm (0.25 in) in diameter. Samples were collected in 500 ml glass containers, sealed and refrigerated at 1°C until sample processing was initiated. Individual samples were prefiltered through Whatman paper (No. 40) prior to filtration through a 0.45 μm Millipore membrane filter. The filters were oven-dried at 100°C for 24 hours prior to wet ashing with concentrated HNO_3 and HF. Water samples were acidified with concentrated HNO_3 immediately after the filtration step to reduce radionuclide plating on the container surface.

Analytical techniques

All samples were analysed for ^{137}Cs on a 7.6 × 7.6 cm NaI(Tl) detector coupled to a 1024-channel pulse-height analyser. The minimum detectable ^{137}Cs, based on an 83 minute count time, was 5 pCi/g for a 1 g sediment sample and 30 pCi/l for a 500 ml water sample.

After gamma counting, the samples were further digested in concentrated HNO_3 and HF as an initial step in the plutonium chemistry [9]. The solutions were passed through an ion-exchange column to isolate plutonium from the sample. Plutonium was electrodeposited on stainless-steel discs and counted for alpha-particle emissions on a surface-barrier detector for 8×10^4 s to measure the ^{238}Pu and $^{239,240}Pu$ content. The minimum detectable activity ($P \leq 0.05$) was 0.05 pCi/sample based on variations in counts due to background and tracer and on a 50% ^{242}Pu tracer recovery.

Results and discussion

Various aspects of the soil plutonium and ^{137}Cs data were discussed in previous papers [4, 8]. Findings pertinent to the present study will be summarized briefly.

Concentrations of both isotopes of plutonium in alluvial sediments from the sampling transect averaged about 200 pCi/g dry weight in 1972 and 1973.

Highest levels were measured within 160 m of the waste outfall and decreased steadily with distance downstream, although not in a linear fashion. Fall-out levels of plutonium were measured at the 5120 and 10 280 m sampling stations, demonstrating that downstream transport of the plutonium had not occurred to these distances.

The plutonium concentration patterns were not consistent with depth; at some locations highest concentrations were measured in the lowest profile (i.e. 12.5-30 cm), while at other locations the surface profile (i.e. 0-2.5 cm) contained highest concentrations. Evidence that considerable vertical mixing had occurred in a short period of time was noted in $^{238}Pu/^{239,240}Pu$ ratios, which averaged 3.6 at all depths at the first nine post-outfall sample locations in 1972. Recall that ^{238}Pu was not added to this canyon until 1968.

Plutonium concentrations were highly variable in triplicate samples from the same sampling station; the coefficient of variation (standard deviation/mean) ranged from 0.06-1.4 and averaged about 0.8.

The parameters used in Eq. (1) to calculate plutonium inventories are presented in Table I, along with the estimated inventories and relative distribution of plutonium in 10 stream channel segments extending from the liquid effluent outfall to 7680 m post-outfall. The inventory estimates apply only to the 0-12.5 cm soil depth due to sampling depth differences between locations and years.

The calculated inventory in 1972 was about 21 mCi ^{238}Pu and 6.5 mCi $^{239,240}Pu$, corresponding to estimates of 23 mCi ^{238}Pu and 5.5 mCi $^{239,240}Pu$ in 1973 (Table I). During the 7-month interval between sample collections the treatment plant released about 5.5 mCi ^{238}Pu and 0.4 mCi $^{239,240}Pu$ to the canyon. The difference in inventory estimates for two periods reflected about 2 mCi ^{238}Pu of the 5 mCi added during the interval and none of the 0.4 mCi $^{239,240}Pu$.

The similarity in the two inventory estimates is surprising in the light of the observed variability in plutonium concentrations. The 2 mCi increase in ^{238}Pu may be indicative of the 5 mCi added to the canyon, although additional data are needed through time to verify that such increases are real.

Treatment plant release records and observed $^{238}Pu/^{239,240}Pu$ activity ratios that averaged 3.6 indicated that a total of about 8.8 mCi $^{239,240}Pu$ and 32 mCi ^{238}Pu was released to the canyon as of October 1972. The addition of 0.4 mCi $^{239,240}Pu$ during the 7-month period represented an increase in inventory of about 4% while the 5.5 mCi ^{238}Pu represented a 17% increase.

The total plutonium inventories in the 0-12.5 cm profile, calculated from Eq. (1), represent about one-half of the plutonium released to the canyon. Inventory estimates for 1972 based upon the entire cores (i.e. to depths up to 30 cm) were 8.3 mCi $^{239,240}Pu$ and 30 mCi ^{238}Pu, which correspond very well to total estimated additions of about 41 mCi to this date. These results indicate that the discrepancy between the inventory estimate in 1972 and the total input of the element was accountable in the lower depth profile.

Conversely, the inventory estimate based on the entire cores from 1973 was about 24 mCi ^{238}Pu and 6.6 mCi $^{239,240}Pu$, which falls about 30% short of the documented input of 46 mCi. Some of this discrepancy is due to consistently shallower sampling depths in 1973 than in 1972.

The distribution of plutonium in the stream channel was very similar at both sampling periods, indicating that little downstream transport had occurred during the 7-month period. There was slightly more plutonium in

TABLE I. PARAMETERS USED IN CALCULATING THE PLUTONIUM INVENTORY IN MORTANDAD CANYON ALONG WITH ESTIMATES OF THE INVENTORIES IN SEVERAL STREAM CHANNEL SEGMENTS

		1972				1973			
W(m)[a]	L(m)	C(mCi/m²)	I(mCi)	%	Accum.%	C(mCi/m²)	I(mCi)	%	Accum.%
					^{238}Pu				
1	10	0.033	0.33	1.5	1.5	0.043	0.43	1.9	1.9
1	20	0.027	0.55	2.6	4.1	0.023	0.46	2.0	3.9
1	30	0.026	0.78	3.6	7.7	0.023	0.69	3.0	6.9
1	60	0.014	0.81	3.8	12	0.008	0.51	2.2	9.1
1	120	0.02	2.4	11	23	0.011	1.3	5.6	15
1	240	0.005	1.1	5.1	28	0.009	2.2	9.4	24
1	480	0.004	1.7	7.9	36	0.006	3.0	13	37
1	960	0.003	2.6	12	48	0.003	2.7	12	49
4	1920	0.002	11	51	99	0.002	12	52	101
2	3840	0.0004	0.10	0.47	99.5	0.00001	0.03	0.13	101.2
	TOTAL		21.4				23.3		
					$^{239,240}Pu$				
1	10	0.009	0.09	1.4	1.4	0.013	0.13	2.4	2.4
1	20	0.023	0.45	6.9	8.3	0.008	0.15	2.7	5.1
1	30	0.012	0.35	5.4	14	0.014	0.43	7.8	13
1	60	0.005	0.29	4.5	18	0.0009	0.06	1.1	14
1	120	0.006	0.72	11	29	0.002	0.23	4.2	18
1	240	0.001	0.22	3.4	33	0.002	0.46	8.4	27
1	480	0.001	0.53	8.2	41	0.001	0.55	10	37
1	960	0.0006	0.56	8.6	49	0.0005	0.49	8.9	46
4	1920	0.0004	3.2	49	98.4	0.0004	3.0	55	101
2	3840	0.00003	0.08	1.2	99.6	0.00001	0.03	0.55	101
	TOTAL		6.5				5.5		

[a] L, W, and C represent the parameters used in Eq.(1) to calculate plutonium inventory (I); W is the average width of stream channel segment L over which the average plutonium concentration (C) applies.

the initial 240 m post-outfall stream segment in 1972 than in 1973. The inventory in the 240 m post-outfall was 23% of the total stream bed ^{238}Pu compared to 15% in 1973. In 1972 29% of the $^{239,240}Pu$ inventory was within 240 m of the outfall, while only 18% was there in 1973. Some snowmelt had occurred prior to the sampling in 1973, possible resulting in limited downstream transport of sediment or ice-bound plutonium.

About 35-40% of the calculated plutonium inventory during both years was present in the 1000-m segment immediately below the waste outfall,

which corresponds to the area containing permanent surface water. Over one-half of the plutonium was present in the lower portion of the canyon where the stream channel is normally dry, indicating that considerable downstream transport had occurred by a process involving factors other than the liquid effluent.

Rainstorm runoff was identified as a potential vector in the transport of radioactivity at Los Alamos nearly 30 years ago [10]. More recent work was begun to characterize and quantify the importance of runoff in plutonium transport [11, 12].

The rainstorm investigated in this study occurred from 12:00-15:00 hours on 15 September 1974, during which time about 2.9 cm of precipitation was deposited on the upper Mortandad Canyon watershed. The resulting runoff began flowing through the upper gauging station (Fig. 1) at 12:45 hours and reached the lower gauging station, 1100 m downstream, at 14:25 hours for an average transit velocity of 0.15 m/s. Peak flow through the upper gauging station occurred about 1 hour after the runoff passed through the weir. Peak flow through the lower gauging station occurred within 10 minutes after arrival of the runoff water (Table II).

The maximum flow rate through the lower gauging station was 0.3 m^3/s, corresponding to a velocity of 1.5 m/s (4.8 ft/s) and a water depth of 31 cm in the centre of the stream channel. The maximum surface width of the flow was 1.1 m. Flow rates generally decreased following the initial peak but not a linear fashion.

There appeared to be four distinct phases to the flow rate data during the runoff event: (1) a rapid increase to peak flow at 5 min; (2) a period of relatively rapid decline in flow rate immediately following the peak to about 60 minutes into the event; and (3) a period of stable flow from 60-185 minutes. Another decline occurred from 185 min to the end of the 270 min observation period.

Maximum suspended sediment concentrations were measured 15 minutes after the event began. Concentrations of 17 ± 0.5 (S.D.) g/l (17000 ppm) and 74 ± 12 g/l were measured at this time in samples from the surface and bottom of the flow respectively. Higher sediment concentrations were consistently measured in the bottom samples, reflecting the presence of suspended sand size fractions in the sample that probably resulted from turbulent flow. The relative particle size distribution in suspended sediment was not determined, although inspection of the samples confirmed that coarse materials comprised a higher percentage of the bottom sample than in surface samples. Sediment concentrations in all samples decreased at a relatively rapid rate from 15 to 55 minutes after the event began, corresponding to the rapid decrease in flow rate. Concentrations in surface samples decreased by about a factor of 2 while the corresponding decrease in bottom samples was a factor of 4 to 5.

Sediment concentrations in surface and bottom samples became increasingly similar as coarse materials settled out of the runoff water. Top and bottom samples, per se, were not obtained after 185 minutes due to the shallowness of the runoff stream (< 15 cm).

Total runoff passing through the lower gauging station during the 270 min observation period was estimated as 3540 m^3. During the first half of the event (i.e. 135 min) about 65% of the total runoff volume had passed through the weir.

TABLE II. CHARACTERISTICS OF A STORM RUNOFF EVENT AT THE LOWER WIER IN MORTANDAD CANYON ON 15 SEPTEMBER 1974

Elapsed time (min)	Flow rate (m^3/s)	Suspended sediment load: Top[a] (g/l)	Suspended sediment load: Bottom (g/l)	Cumulative runoff (m^3)	Cumulative runoff (%)
5	0.30	14	56	90	2.5
10	0.30				
15		17 (0.50)[b]	74 (12)	234	6.6
18	0.29			303	8.6
22	0.30			537	15
35	0.27				
40		13 (0.21)	33 (15)	697	20
45	0.26			775	22
50	0.26			853	24
55	0.24	8.9 (0.64)	12 (1.5)	925	26
60	0.26				
120		4.5 (0.04)	23 (20)	1930	54
125	0.26			2160	61
140	0.24			2380	67
155	0.26				
160		4.8 (2.5)	7.3 (5.5)	2540	72
165	0.24			2610	74
170	0.24			2830	80
185	0.23	3.3 (0.09)	6.6 (0.98)	2970	84
195	0.18			3080	87
205	0.18			3130	88
210	0.14	3.3[c]		3220	91
220	0.11			3280	93
230	0.10				
233		1.7		3370	95
245	0.09				
250		2.5 (0.14)		3430	97
255	0.09			3450	98
260	0.08	2.3 (0.28)		3500	99
270	0.07			3540	100

[a] Samples designated as top were collected about 6 cm below the surface of the flow, while bottom samples were collected about 6 cm above the stream bed.
[b] 1 S.D. based upon duplicate samples.
[c] Top and bottom samples, per se, were not collected after 185 min due to the shallow flow (i.e. < 15 cm).

TABLE III. MEAN $^{239,240}Pu$ CONCENTRATIONS IN RUNOFF SAMPLES COLLECTED FROM MORTANDAD CANYON ON 15 SEPTEMBER 1974

Elapsed time (min)	Water (pCi/l)[a]	Suspended sediment[a] Top (pCi/g)	Bottom	Top (pCi/l)	Bottom	Mean total activity (pCi/l)
5	0.25 (0.10)[b]	3.4	1.3	48	73	61 (18)
15	0.67 (0.76)	2.8 (0)	1.9 (1.4)	47 (1.4)	131 (84)	90 (69)
40	0.39 (0.29)	4.4 (1.5)	2.9 (2.1)	56 (20)	79 (26)	68 (23)
55	0.67 (0.64)	5.7 (0)	4.2 (1.8)	51 (3.5)	51 (29)	51 (16)
120	0.56 (0.38)	5.6 (1.3)	2.9 (2.3)	25 (5.7)	45 (5.7)	36 (13)
160	0.51 (0.34)	6.2 (3.2)	5.1 (1.6)	26 (0)	33 (16)	30 (9.7)
185	0.17 (0.11)	8.3 (0.1)	5.6 (1.0)	28 (0.71)	37 (0.71)	32 (5.2)
210	0.21	4.9	--	16	--	16
233	0.22	13	--	22	--	22
245	0.40	8.8 (0.35)	--	22 (0.71)	--	22 (0.71)
260	1.3 (1.3)	7.9 (1.7)	--	18 (1.4)	--	19 (2.8)

[a] Averages for water based on 4 samples (2 top and 2 bottom), water filtered through 0.45 μm membrane; averages for suspended sediment based on 2 samples.
[b] Parenthetic value ± 1 S.D.

TABLE IV. MEAN ^{238}Pu CONCENTRATIONS IN RUNOFF WATER COLLECTED FROM MORTANDAD CANYON ON 15 SEPTEMBER 1974

Elapsed time (min)	Water (pCi/l)[a]	Suspended sediment[a] Top (pCi/g)	Bottom (pCi/g)	Top (pCi/l)	Bottom (pCi/l)	Mean total activity (pCi/l)
5	2.5 (0.14)[b]	25	7.3	350	410	380 (40)
15	4.9 (5.5)	19 (0.71)	12 (9.3)	310 (2.1)	790 (550)	550 (430)
40	3.1 (1.6)	30 (11)	18 (15)	390 (150)	460 (220)	430 (160)
55	5.1 (2.9)	41 (2.1)	37 (23)	360 (7.1)	460 (340)	410 (200)
120	3.3 (1.3)	41 (17)	18 (15)	190 (80)	270 (20)	320 (170)
160	3.9 (3.2)	44 (21)	33 (9.2)	180 (7.8)	210 (110)	200 (66)
185	1.9 (1.0)	59 (5.0)	36 (3.5)	190 (11)	230 (11)	210 (24)
210	3.0	36	--	120	--	120
233	2.4	91	--	152	--	154
250	3.2	67 (0.71)	--	160 (9.9)	--	160 (9.9)
260	6.2 (3.6)	64 (9.2)	--	150 (2.8)	--	150 (6.4)

[a] Averages for water based on 4 samples (2 top and 2 bottom), water filtered through 0.45 μm membrane; averages for suspended sediment based on 2 samples.
[b] Parenthetic value ± 1 S.D.

TABLE V. ^{137}Cs CONCENTRATIONS IN RUNOFF WATER COLLECTED FROM MORTANDAD CANYON ON 15 SEPTEMBER 1974

Elapsed time (min)	Water (pCi/l)[a]	Suspended sediment[a] Top (pCi/g)	Bottom	Top (pCi/l)	Bottom	Mean total activity (pCi/l)
5	88 (10)[b]	330	150	4600	8400	6600 (2700)
15	26 (28)	220 (9.0)	110 (32)	3700 (280)	7900 (1100)	5800 (2500)
40	44 (4.4)	200 (83)	160 (67)	2600 (1100)	4700 (190)	3700 (1400)
55	54 (12)	240 (12)	240 (3.5)	2200 (260)	2800 (420)	2500 (480)
120	44 (19)	300 (65)	170 (65)	1400 (300)	3300 (1900)	2300 (1600)
160	32 (9.6)	500 (75)	200 (8.5)	2500 (1600)	1500 (1200)	2000 (1300)
185	66 (34)	360 (190)	250 (37)	1200 (650)	1700 (7.1)	1500 (460)
210	56	340	--	1100	--	1200
233	34	650	--	1100	--	1100
250	80 (0)	390 (53)	--	960 (80)	--	980 (110)
260	44 (12.)	310 (44)	--	730 (190)	--	750 (190)

[a] Averages for water based on 4 samples (2 top and 2 bottom), water filtered through 0.45 μm membrane; averages for suspended sediment based on 2 samples.
[b] Parenthetic value ± 1 S.D.

The concentrations of ^{137}Cs, ^{238}Pu and $^{239,240}Pu$ in runoff samples exhibited very similar patterns with time (Tables III-V). The filtered water contained low concentrations of all radionuclides; levels ranged from about 30-80 pCi ^{137}Cs/l, 2-5 pCi ^{238}Pu/l and 0.2-1 pCi $^{239,240}Pu$/l. Concentrations of the radionuclides in water were variable and did not exhibit a consistent pattern with time. On the other hand, levels of all three radionuclides in the suspended sediment fraction were relatively high and generally increased throughout the event. Concentrations in suspended sediments ranged from 100-600 pCi ^{137}Cs/g, 10-100 pCi ^{238}Pu/g and 1-10 pCi $^{239,240}Pu$/g.

Significantly higher ($P \leq 0.05$) concentrations of the radionuclides were measured in particulate samples collected near the surface of the flow compared to those from near the bottom. As previously mentioned, a greater proportion of fine particle sizes comprised the surface samples compared to those from near the bottom. Previous studies in this canyon have shown that particles less than 53 μm comprised only 2% of bed sediments by weight but contained about 15% of the radioactivity [13].

About 1% of the radionuclide inventory in each sample was present in the water fraction, whereas 99% was associated with the suspended particulates. More total radioactivity was associated with suspended sediments in bottom samples than with corresponding surface samples (Tables III-V), even though the latter exhibited significantly higher radionuclide concentrations. Previous work had demonstrated that about 80% of the radionuclide inventory in Mortandad Canyon bed sediments was associated with particle sizes greater than 105 μm, which comprised about 95% of the alluvium by weight.

The total amount of radioactivity associated with suspended sediments in each water sample (pCi/l) steadily decreased through the runoff event, even though the concentrations (pCi/g) generally increased.

There was a significant linear correlation ($P \leq 0.05$) between ^{137}Cs and the corresponding plutonium isotopes in individual suspended sediment samples. This relationship was observed previously in Mortandad Canyon sediments [4, 8] and indicates that the two elements are distributed similarly along the stream channel and in the various size fractions.

The relationship between flow rate and mean total radionuclide concentrations (and mean suspended sediment concentration) in water samples is presented in Fig. 2. The radionuclide and suspended sediment concentrations were relatively constant for flow rates in the range 0.07-0.25 m^3/s, possibly indicating that all available fine materials were in suspension at these flow rates. However, radionuclide and suspended sediment concentrations were directly correlated with increasing flow in the range 0.25-0.3 m^3/s.

It seems likely that rates in excess of about 0.25 m^3/s resulted in the suspension of coarse particles (> 105 μm) that contained over 95% of the sediment mass and 90% of the radionuclide inventory.

Suspended sediment concentrations were used in a power function relationship to predict total concentrations of the specific radionuclides in runoff water samples. The equations resulting from the least squares fit of the data were:

$$Y = 110X^{0.40}, \quad r^2 = 0.82, \quad n = 11 \tag{2}$$

$$Z = 14X^{0.47}, \quad r^2 = 0.87, \quad n = 11 \tag{3}$$

$$C = 580X^{0.62}, \quad r^2 = 0.94, \quad n = 11 \tag{4}$$

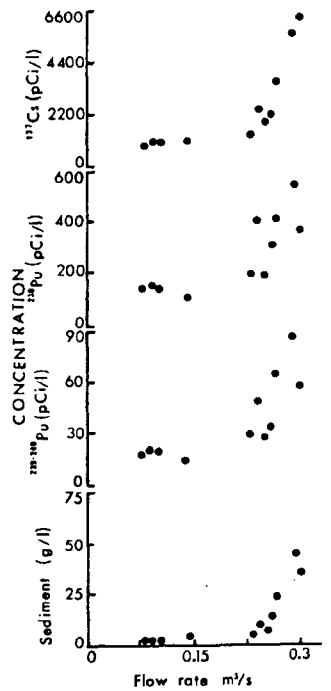

FIG.2. Concentration of sediment and radioactivity in unfiltered runoff water from Mortandad Canyon as a function of runoff flow rate.

where Y, Z and C are the average total ^{238}Pu, ^{239,240}Pu and ^{137}Cs concentrations in unfiltered runoff water (pCi/l) and X is the average suspended sediment concentration (g/l). The multiple correlations coefficients (r^2) were all highly significant ($P \leq 0.01$).

The total amount of radioactivity transported by the event was calculated from average total radionuclide concentrations and cumulative runoff through the event. The estimated transport was 8.5 mCi ^{137}Cs, 1.1 mCi ^{238}Pu and 0.2 mCi ^{239,240}Pu. In the case of plutonium this activity represented from 1-2% of the total added to the canyon as of September 1974. In the 11 years that Mortandad Canyon has received contaminated wastes over 50% of the radioactivity has been transported into the dry portion of the canyon by snowmelt and storm runoff, indicating that on an average the annual losses of radioactivity to the dry portion of the stream are also about 50%.

The most efficient portion of the storm runoff for transporting the radionuclides was near the beginning of the event when sediment concentrations were high as a result of high velocities and flow rates. Nearly 80% of the sediment and 70% of the radioactivity were transported during the first 120 min of the 270 min observation period.

CONCLUSIONS

Plutonium inventory estimates for the upper 12.5 cm of sediment in Mortandad Canyon did not reflect all the plutonium added to the canyon during a 7-month interval. The methods used in this study indicate that about 2 mCi ^{238}Pu and 0 mCi ^{239,240}Pu were added to the canyon during the interval, compared with known additions of 5.5 mCi ^{238}Pu and 0.4 mCi ^{239,240}Pu. The discrepancy was probably the result of the large sampling variability, indicating that inventory changes of this order (i.e. up to 17%) are not detectable with any certainty. However, factors other than sampling variability may be involved, including losses of plutonium to depths exceeding 12.5 cm.

The relative distribution of plutonium within the canyon demonstrates that transport has occurred beyond the extent of permanent surface water and that runoff from summer rainstorms can serve as a radionuclide transport vector in landscape exhibiting these hydrologic features. There appears to be a highly significant relationship between suspended sediment concentrations and total amounts of radioactivity in water. The flow rates occurring during the runoff event play an important part in determining the total amount of sediment and thus radioactivity transported downstream.

Although fine materials exhibited the highest concentrations, most of the radioactivity was associated with the more abundant coarser size fractions, which served as the most important component of sediment involved in radionuclide transport. The water fraction was relatively unimportant in radionuclide transfer, although the flowing water did serve as the transport vector.

Additional studies should be conducted to determine the radionuclide transport characteristics of runoff events that are either larger or smaller than the one examined in this study. Particle size determinations would be valuable in relating flow rates (or velocity) to types of suspended material in the runoff.

Storm runoff serves as a transport vector for sediment deposited radioactivity as does wind in arid terrestrial environments. The implications to humans are different in that other transfer pathways may become important.

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DISCUSSION

S.R. WRIGHT: Have you sampled game animals such as deer or rabbits living in the Los Alamos area?

T.E. HAKONSON: Yes, and very little activity was found – primarily because the animals don't spend much time in these canyons.

R.O. GILBERT: Were you able to gauge the precision of your estimates of ^{238}Pu and ^{239}Pu inventory in the stream channel of Mortandad Canyon? It appears that you have enough data to obtain at least rough approximations of these precisions.

T.E. HAKONSON: No; that is why I presented the information on the coefficients of variation for the concentrations. Obviously the error on the inventory estimate was at least as great as the errors on the concentrations that went into it.