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The Distribution of Mercury, Cesium-137, and Plutonium in an Intermittent Stream at Los Alamos[1]

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ABSTRACT

This paper summarizes the results of a study on the distribution of Hg, ^{137}CS, ^{238}Pu, and ^{239,240}Pu in channel sediments and adjacent bank soils in an intermittent stream used for treated liquid effluent disposal since 1963.

Concentrations of the three radionuclides and Hg in stream bank soils were comparable to adjacent channel sediments demonstrating that the stream bank serves as a deposition site for chemicals released to the channel. This finding has important implications on the long-term behavior of effluent contaminants since other studies at Los Alamos have shown that the vegetated stream banks retard downstream movement of chemicals bound to soils and provide a pathway for transport of these materials to biota.

Concentrations of the radionuclides and mercury were more uniformly distributed with distance and depth in the channel sediments than in the bank soils. The action of periodic surface water in the channel partially explains those differences. Statistical analysis of the data revealed that 50 to 85% of the variability in contaminant concentrations in bank and channel locations was due to variation with distance while depth contributed relatively little to variability.

Additional Index Words: **environmental contaminants, liquid effluents, stream sediment, radionuclides.**

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Intermittent streams are a prominent feature of southwest U.S. watersheds and serve in the collection and transport of surface water runoff and associated chemicals. The role of intermittent streams in regulating the distribution and transport of soil contaminants is largely unknown despite the importance of these streams in the physiography of areas currently being developed for energy resources. Past environmental studies at Los Alamos Scientific Laboratory in northern New Mexico emphasized the distribution and transport of contaminants in three intermittent streams used for disposal of treated industrial liquid wastes over the last 36 years (5, 6, 10). The goal of that research was to characterize and describe the processes which result in the transport of some environmental contaminants in southwestern ecosystems.

This paper summarizes the results of a study on the distribution of Hg, ^{137}Cs, and Pu in stream bank soil and stream channel sediments from an intermittent stream used for treated liquid effluent disposal since 1963. Specific objectives of the study were to compare concentrations of those elements in bank soils and channel sediments to evaluate the potential of stream banks as a deposition site for liquid effluent contaminants. Additionally, data were collected to evaluate the relative imortance of sampling depth and distance in contributing to the variability in elemental concentrations.

Industrial liquid wastes generated by the Los Alamos Scientific Laboratory are collected at a central location, treated to reduce the levels of radioactive contaminants below federal guidelines for drinking water, and then they are released to the local environs (1, 10).

The composition of the effluent is complex and varies daily depending on the activities of facilities generating the waste. The effluent nearly always is alkaline due to chemicals used during waste treatment with pH varying from about 7 to 12 and averaging about 9 (10). The chelate content of the effluent also varies considerably as evidenced by fluctuations in radionuclide removal efficiencies during the treatment process. These factors make it difficult to speculate on the chemical form of the elements in the effluent. Changes in the chemical and physical association of the elements as the effluent contacts the environment are unknown.

Mortandad Canyon has been used for disposal of liquid wastes since 1963. Stable (nonradioactive) Hg is also a component of the waste released to Mortandad Canyon as a result of loss of the element from chemical laboratories into drain systems. Records maintained over the past few years show that a few tens to hundreds of g of Hg are released annually to this canyon (1, 10). The quantity of Pu and Cs released annually to the canyon averages about 10 and 100 mCi, respectively. The isotopic composition of the Pu in the waste has changed from predominately ^{239,240}Pu before 1968 to mostly ^{238}Pu at the present time.

About 2×10^5 liters of liquid effluent are discharged to Mortandad Canyon daily (5 days/week) in two pulse releases. The effluent fills the small (1 m wide by 0.5 m deep) channel and rapidly begins to infiltrate into the alluvium. The effluent stream is confined to the trough-shaped stream channel; however, runoff from summer rainstorms periodically overflows the channel and transports contaminants to the densely vegetated stream banks. An average of about six rainfall runoff events occur in Mortandad Canyon each year; an average of two events overflow the stream channel each year. Contaminants also may be transported to the bank by water infiltrating from the channel.

METHODS AND MATERIALS

Soil cores were collected from 10 channel and 10 bank sampling sites randomly selected along a 100-m segment (Site 1) of Mortandad Canyon about 500 m below the effluent outfall. The size of the watershed upstream from the sampling location was about 1.3 km^2. Ten cores were collected in the channel while 40 cores (four per location) were collected on the stream bank. The rectangular (or trough) shape of the stream channel was used to distinguish between channel and bank locations. The four bank soil cores were collected systematically

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at each location at the corners of a 0.25-m^2 (0.25 m lateral × 1.0 m parallel to the channel) template placed along the boundary between the alluvial stream bed and the vegetated stream bank. Thus, bank soil core data represent averages for the zone 0 to 25 cm lateral to the stream channel.

The core samples were frozen and sectioned into 0- to 2.5-, 2.5- to 7.5-, and 7.5- to 30-cm segments; 5-g aliquots of the blended sample were then taken for Hg analysis by flameless atomic absorption (3). The remaining sample was oven-dried and counted for ^{137}Cs on a 10 × 20 cm NaI detector coupled to a 2048 multichannel analyzer. Sample aliquots were analyzed for ^{238}Pu, ^{239,240}Pu, and Hg using wet chemistry followed by instrumental analysis (3, 10). Elemental concentrations in all cases were sufficient to limit instrumental uncertainties to <10% (p <0.05). Soil particle size determinations were made on one core sample from each of the 10 stream bank locations and on every core sample from the channel location.

RESULTS AND DISCUSSION

SOIL CHARACTERISTICS

The particles constituting the alluvium in Mortandad Canyon were derived from weathering of acid volcanic rocks. Tuff, pumice, latite, and rhyolite are the major constituents of the alluvium with minor amounts of quartz and sanidine crystals. Sand-sized particles consist of quartz and sanidine crystals or crystal fragments while the silt-clay fraction is mainly montmorillonite and illite.

Over 97% (by wt) of the channel sediments consists of sand-size particles >53 μm (Table 1) and coarse sands (>500 μm) comprise over 70% of the mass of the channel sediments. Bank soil particle size distributions are generally similar to those in channel sediments although silt-clays comprise a significantly higher (p <0.05) percentage of the bank soil mass. The organic carbon content of channel sediment averages about 0.5% by wt (8). Organic carbon was not measured in bank soils but is probably slightly higher (1 to 5%) than in the channel.

CONCENTRATION DATA

Concentrations of Hg, ^{137}Cs, and Pu in bank soils were comparable to those in channel sediments (Fig. 1), demonstrating that the effluent contaminants were transported at least 25 cm into the bank area. The data (Fig. 1) also illustrate the much greater range and skewness of concentrations observed in the bank soil, although median and arithmetic mean concentrations between locations were generally within a factor of two for all contaminants.

Table 1—Relative particle size distribution in bank soils and channel sediments from sampling site I in Mortandad Canyon.

		Percent of soil mass in size range				
Location	Depth, cm	<53 μm	53-106 μm	106-300 μm	300-500 μm	>500 μm
Bank	0-2.5	11(7.3)†	19(7.1)	28(5.1)	17(5.1)	25(9.3)
	2.5-7.5	14(5.9)	15(6.7)	23(7.1)	16(5.2)	32(23)
	7.5-30	6.8(4.2)	10(7.8)	18(11)	13(5.6)	53(25)
Channel	0-2.5	0.82(0.39)	1.1(0.37)	5.1(2.5)	10(6.3)	83(8.6)
	2.5-7.5	1.4(1.2)	1.0(0.71)	5.6(2.7)	11(4.9)	81(7.4)
	7.5-30	3.0(0.53)	3.0(2.6)	10(4.5)	12(6.3)	72(12)

† Mean % of soil wt (1 standard deviation); based on sample size of 10.

LOCATION, DEPTH, AND DISTANCE RELATIONSHIPS

The relationships between contaminant concentrations in soil and sampling depth and distance were assessed using the following ANOVA model:

$$Y_{ijk} = \mu + \alpha_i + \beta_j + (\alpha\beta)_{ij} + \epsilon_{ijk}$$

where μ is the parametric mean of the population; α_i is the fixed treatment effect for a given depth i (i = 1, 2, 3); β_i is the fixed treatment effect of the jth distance (j = 1, 2, ..., 10); $(\alpha\beta)_{ij}$ is the interaction term in the subgroup representing the ith depth and the jth distance (not included in the model for channel data because there is only one observation per subgroup); Y_{ijk} is the kth log-transformed concentration in subgroup ij; and ϵ_{ijk} is the term of the kth item in subgroup ij. Significant differences (p <0.05) in contaminant concentrations were observed with sample depth for all elements in the stream bank location; concentrations decreased with depth, following a pattern that is typical of contaminants initially deposited on the soil surface (2, 6, 7). Although concentrations of the elements differ with depth, it is apparent (Table 2) that there has been considerable transport of all four contaminants into the lowest depth profile.

In contrast to the bank data, contaminant concentration did not change significantly with depth (p <0.05) in the stream channel. This result indicates that concentrations of the elements are more uniform with depth in the channel than in the bank.

In all cases the concentrations of contaminants in bank soils changed significantly (p <0.05) with distance following the pattern indicated for ^{137}Cs (Fig. 2). In no case was the change in concentration linear with distance.

Contaminant concentrations in channel sediments, as represented by the ^{137}Cs data (Fig. 2), were much more consistent with distance than in bank soils. In addition, concentrations of all four nuclides generally decreased with distance in contrast to the fluctuating pattern observed in bank soils. Concentrations were significantly

Table 2—Relationship of elements with sampling depth in bank soils and channel sediments from sampling site I in Mortandad Canyon.

	Depth profile, cm			
Element	0-2.5	2.5-7.5	7.5-30	Significance, p†
		Channel		
Hg (ppb)	99 (1.4)‡	72 (0.63)	70 (0.70)	0.735
^{137}Cs (pCi/g)	359 (0.29)	325 (0.30)	422 (0.39)	0.062
^{238}Pu (pCi/g)	25 (0.27)	24 (0.28)	29 (0.38)	0.472
^{239,240}Pu (pCi/g)	7.2 (1.9)	3.9 (0.72)	4.6 (0.63)	0.867
		Bank		
Hg (ppb)	995 (1.5)	165 (1.4)	123 (1.9)	<0.001
^{137}Cs (pCi/g)	225 (1.0)	186 (1.8)	182 (2.3)	<0.001
^{238}Pu (pCi/g)	24 (1.4)	27 (2.0)	18 (2.4)	<0.001
^{239,240}Pu (pCi/g)	6.3 (0.87)	5.6 (1.7)	5.6 (2.3)	<0.001

† p derived from ANOVA (F-tests) performed on log-transformed data; p is the probability of observing an F value as large or larger than that observed given that the null hypothesis is true.

‡ Arithmetic mean (coefficient of variation, c = SD/$\bar{x}$); means based on n of 40 for bank data and n of 10 for channel data.

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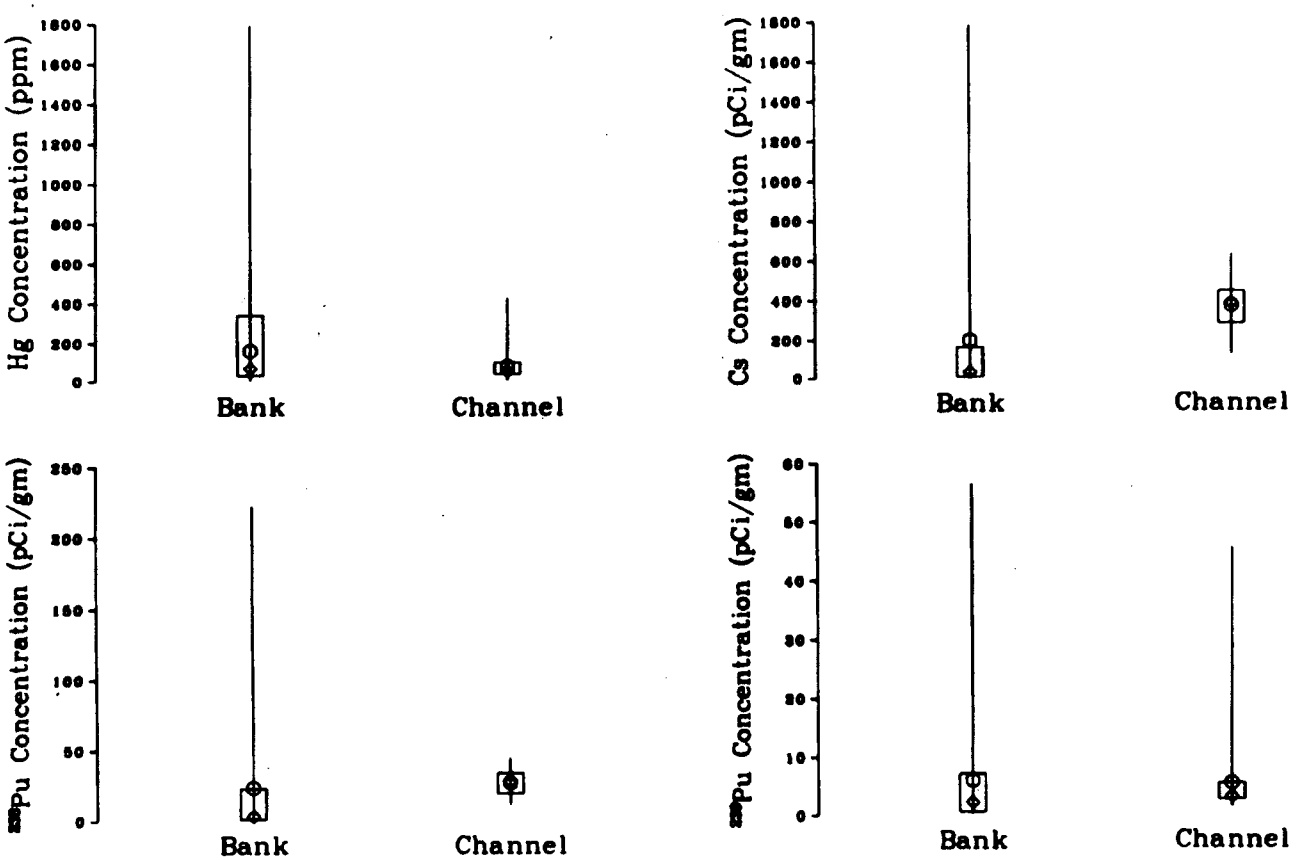

Fig. 1—Concentrations of Hg, ^{137}Cs, ^{238}Pu, and ^{239,240}Pu in stream bank soils and channel sediments displayed as box plots. Arithmetic mean = diamond; and box enclosed middle 50% of the data (i.e., from first to third quartile).

different ($p < 0.05$) with distance for ^{137}Cs and Hg, however, they were not significant for ^{238}Pu and ^{239,240}Pu. Previous studies in this canyon have shown that concentrations of ^{137}Cs, ^{238}Pu, and ^{239,240}Pu decrease with distance downstream (4), reflecting the point source of the contaminants and the gradient produced by periodic surface water flows.

CONCENTRATION VARIABILITY

The relative importance of depth and distance in contributing to the observed variability in contaminant concentrations was evaluated with the ANOVA model described previously. The results, as inferred from the percent reduction in sums of squares, show the dominating effect of distance in contributing to the observed concentration variability in both sampling locations (Table 3). In stream bank samples, the distance variable accounted for 54 to 64% of the total variability while depth accounted for <15%. A similar pattern was observed in channel sediments where distance accounted for 50 to 85% of the variability and depth accounted for <6%. In combination (including interaction term for bank samples), distance and depth accounted for 66 to 74% of the concentration variability in bank soils and about 51 to 88% of the concentration variability in channel sediments.

CONCLUSION

Radionuclide and Hg concentration data demonstrated that the stream banks within at least 25 cm of the channel become contaminated with chemicals released

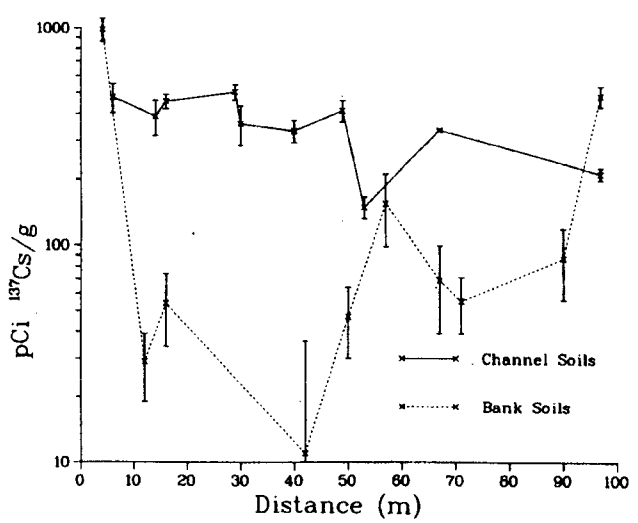

Fig. 2—Cesium-137 concentrations in bank soils and channel sediments as a function of distance within a 100-m segment of Mortandad Canyon. Vertical bars are ± 1 standard error.

Table 3—The contribution of distance and depth to soil contaminant concentration variability in sampling site I in Mortandad Canyon.

Parameter	Percent reduction in total sum of squares†			
	Hg	^{137}Cs	^{238}Pu	$^{239,240}Pu$
	Bank			
Depth	9	15	10	12
Distance	60	54	64	55
Combined	68	70	74	66
	Channel			
Depth	1	6	5	1
Distance	75	85	54	50
Combined	76	88	57	51

† Based on analysis of variance model:

$$Y_{ijk} = \mu + \alpha_i + \beta_j + (\alpha\beta)_{ij} + \epsilon_{ijk}$$

(see text for definitions).

in liquid effluents to an intermittent stream. These results have important implications on the long-term behavior of waste contaminants in Los Alamos intermittent streams because the stream banks, which are heavily vegetated, retard downstream movement of bank soils and associated chemicals. Comparison of radionuclide inventories in Los Alamos intermittent streams with different use histories demonstrates that the stream bank retains Pu and ^{137}Cs much more effectively than the highly erosive stream channel. For example, in a canyon that has not received liquid effluents since 1963, 65 to 100% of the $^{239,240}Pu$ and ^{137}Cs inventories in study plots were present in the stream bank.[5] Effluent radionuclides originally present in the stream channel have been transported to downstream areas primarily by storm runoff events.

The deposition of waste contaminants in stream bank soils also provides a source of the contaminants to streamside flora and associated fauna. Elevated concentrations of ^{137}Cs and Pu have been measured in plants and in a few animals from these areas (5), primarily as a result of physical suspension of soil particles to biological surfaces.

The distribution of the contaminants is generally much more uniform in channel sediments than in bank soils, as inferred from the concentration data and results of statistical analyses. Previous studies have demonstrated the importance of storm runoff in transporting contaminants to downstream areas in Mortandad Canyon (4). The unidirectional flow of the surface water tends to evenly distribute the contaminants in a continuous gradient along the channel. Although we do not understand the mechanisms of contaminant transport to the stream bank, we speculate that surface water overflow from the channel carries contaminated sediments to the bank. The highly erratic nature of this overflow and the uneven stream bank morphology likely result in the large variation in elemental concentrations that were observed in stream bank soils.

Statistical analysis of the data demonstrated the importance of distance as a variable contributing to contaminant concentration variability in both channel and bank locations. The point source of effluent and the concentration gradient produced by the flowing water at least partially accounts for this finding. Relatively little of the total variation in contaminant concentrations could be attributed to variation with sampling depth. About 10 to 50% of the total variability could not be explained by distance or depth variables and may be related to the chemical or physical association of the contaminants in the effluent.

Identification of important variables contributing to concentration variability is essential to design effective experiments or monitoring programs. For example, in designing monitoring programs to document changes in contaminant concentrations in intermittent streams such as at Los Alamos, some benefit in cost and sensitivity to changes may be obtained by designing a sampling program to reduce the variability due to distance.

[5] J. W. Nyhan, G. C. White, and G. Trujillo. 1979. Soil plutonium and cesium in stream channels and banks of Los Alamos liquid effluent receiving areas. In preparation.

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