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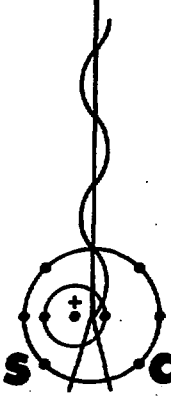

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TEMPORAL CHANGES IN THE DISTRIBUTION OF ^{137}Cs IN ALLUVIAL SOILS AT LOS ALAMOS

by

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ABSTRACT

The alluvial soils of three liquid-effluent receiving areas at Los Alamos were sampled to determine ^{137}Cs temporal distributional relationships. Soil radionuclide concentrations were determined as a function of soil depth and distance from the waste outfall, and discussed relative to runoff transport of ^{137}Cs-contaminated alluvium. The inventories of soil ^{137}Cs in various segments of each effluent-receiving area were calculated for two sampling periods and compared with amounts of ^{137}Cs added to the canyons in the liquid wastes. The distribution patterns of soil cesium were compared with the waste-use history of the three study areas and the hydrologic characteristics of the canyons.

I. INTRODUCTION

Fallout ^{137}Cs has resulted in the largest tissue doses received by human populations (Fredriksson et al., 1966), second only to ^{90}Sr. Besides worldwide fallout additions of ^{137}Cs to the environment, this isotope is commonly discharged to rivers and streams in treated low-level wastes from fuel reprocessing and nuclear power plants. The ^{137}Cs released to aquatic and terrestrial environs is readily adsorbed and retained by soils, which contain the largest portion of the ^{137}Cs inventory in these ecosystems.

Field studies of the behavior of ^{137}Cs have been made in a variety of locations and environments (Brisbin et al., 1974; Cline and Rickard, 1973; Davis, 1973; Francis and Tamura, 1973; Gallegos et al., 1970; Gustafson, 1969; Gustafson et al., 1966; Hakonson and Whicker, 1975; Howells et al., 1970; Kneip et al., 1970; Lentsch et al., 1970; Lerman and Taniguchi, 1973; Nelson and Whicker, 1969; Parker et al., 1965; Pendleton and Hanson, 1958; Pickering, 1969; Reynolds and Gloyna, 1964; Rogowski and Tamura, 1965 and 1970; Romney et al., 1973; Sharitz et al., 1975; and Wrenn et al., 1973). The laboratory and field experiments related to factors important in cesium transport in aquatic ecosystems in Texas have been summarized by Gromiea and Gloyna (1973). Although some of these studies involved describing temporal changes in soil ^{137}Cs distributions, the behavior of ^{137}Cs in intermittent, ephemeral streams, such as those adjacent to the Los Alamos Scientific Laboratory, has only recently been studied (Hakonson et al., 1973; Hakonson and Bostick, 1976; Nyhan et al., 1976).

The objective of the investigation described here was to characterize the horizontal and vertical distribution of ^{137}Cs in the alluvial soils of three liquid-effluent receiving areas at Los Alamos, and to compare temporal differences in these distributions. Changes in the distribution patterns of ^{137}Cs were also evaluated in terms of the waste-use histories and hydrologic characterstics of the three study areas.

II. SITE DESCRIPTIONS AND USE HISTORIES

The three study areas which have received low-level (<10% of Maximum Permissible Concentrations in water), treated, radioactive liquid wastes are located in Effluent-Mortandad, DP-Los Alamos, and Acid-Pueblo Canyons, near Los Alamos, New Mexico (Fig. 1). The alluvium of the intermittent streams in Effluent-Mortandad, DP, and Acid Canyons was formed from the rhyolitic volcanic formations of the Bandelier Tuff. The alluvial soils in Pueblo and Los Alamos Canyons were derived from the latite and quartz latite rocks of the Tschicoma Formation, intermixed with tuff particles from Acid and DP Canyons. The alluvium in the lower reaches of Pueblo Canyon also contains basaltic materials. The upper reaches of these canyons have stream channel widths of less than 1 m and alluvium depths of less than 0.15 m. The stream beds gradually increase in width and depth in the lower portion of the canyons, averaging about 3 m wide and up to 30 m deep. Although Griggs (1964) characterized the hydrology and geology of these areas, little is known about the physical-chemical properties of these alluvial soils.

These canyons have been used as discharge areas for low-level radioactive liquid wastes for varying lengths of time. Acid-Pueblo Canyon received untreated liquid wastes from 1944 to 1951, and treated waste products from 1951-1963. Treated wastes have been discharged into Effluent-Mortandad Canyon since 1963 and into DP-Los Alamos Canyon since 1953. Long-term records of the amounts of ^{137}Cs in these liquid wastes are not available. Records do show that estimated annual inputs of 220 μCi and 1.5 μCi of ^{137}Cs were added to Effluent-Mortandad and DP-Los Alamos Canyons, respectively, from 1973 to 1974 (Schiager and Apt, 1974).

III. SAMPLING AND ANALYTICAL METHODS

Soil samples were collected in the canyons 100 m above the waste outfalls and at distances of 0, 20, 40, 80, 160, 320, 640, 1280, 2560, 5120, and 10 240 m below the outfalls, along a sampling network (Fig. 1) described by Hakonson et al. (1973). Individual cores were taken with a 30-cm polyvinyl chloride tube (2.4-cm i.d., schedule-89 pipe with a sharpened end), which was placed in a plastic bag, frozen, and cut into segments corresponding to 0-2.5, 2.5-7.5, 7.5-12.5, and 12.5-30-cm soil depths. When the alluvium was less than 12.5 or 30 cm deep, a partial core section was obtained and its length also measured. Each station was sampled during October-November 1972, at the center of and at two lateral positions in the stream channel, for a total of 3 cores per station. The core samples from the center of the stream channel were the only cores cut into segments to study the vertical distribution of ^{137}Cs.

In 1973, soil cores were collected during May-July from each sampling station, at 30-cm intervals along the center of the stream channel. Soil samples from all soil depths (0-2.5, 2.5-7.5, 7.5-12.5, and 12.5-30 cm) of the 10 cores collected 0, 40, 640, and 2560 m from the waste outfalls were analyzed for ^{137}Cs to determine the variation of ^{137}Cs levels at these four locations. The 5 soil cores collected 20, 80, 160, 320, 1280, and 5120 m from the waste outfall were sectioned into similar depth profiles and were composited, by depth, at each sampling location.

The soil samples were analyzed for ^{137}Cs on a 7.6 by 7.6-cm NaI(Tl) scintillation detector. Some samples contained measurable ^{134}Cs and ^{40}K, and the channels-ratio method was used to correct the ^{137}Cs photopeak area for these contributions.

The inventory of soil ^{137}Cs in each liquid effluent-receiving area was calculated as:

$$I = C\ L\ W,$$

where

I = mCi soil ^{137}Cs in stream-channel segment L,
C = average soil ^{137}Cs concentration (mCi/m^2) in segment L,
L = length of stream-channel segment (m) over which C applied, and
W = average width of stream channel (m) in segment L.

Levels of soil ^{137}Cs below the detection limit of 0.08 pCi ^{137}Cs/g ($P \leq 0.05$) were set equal to zero in this formula, resulting in minimal inventory estimates for such stream channel segments. The length of the

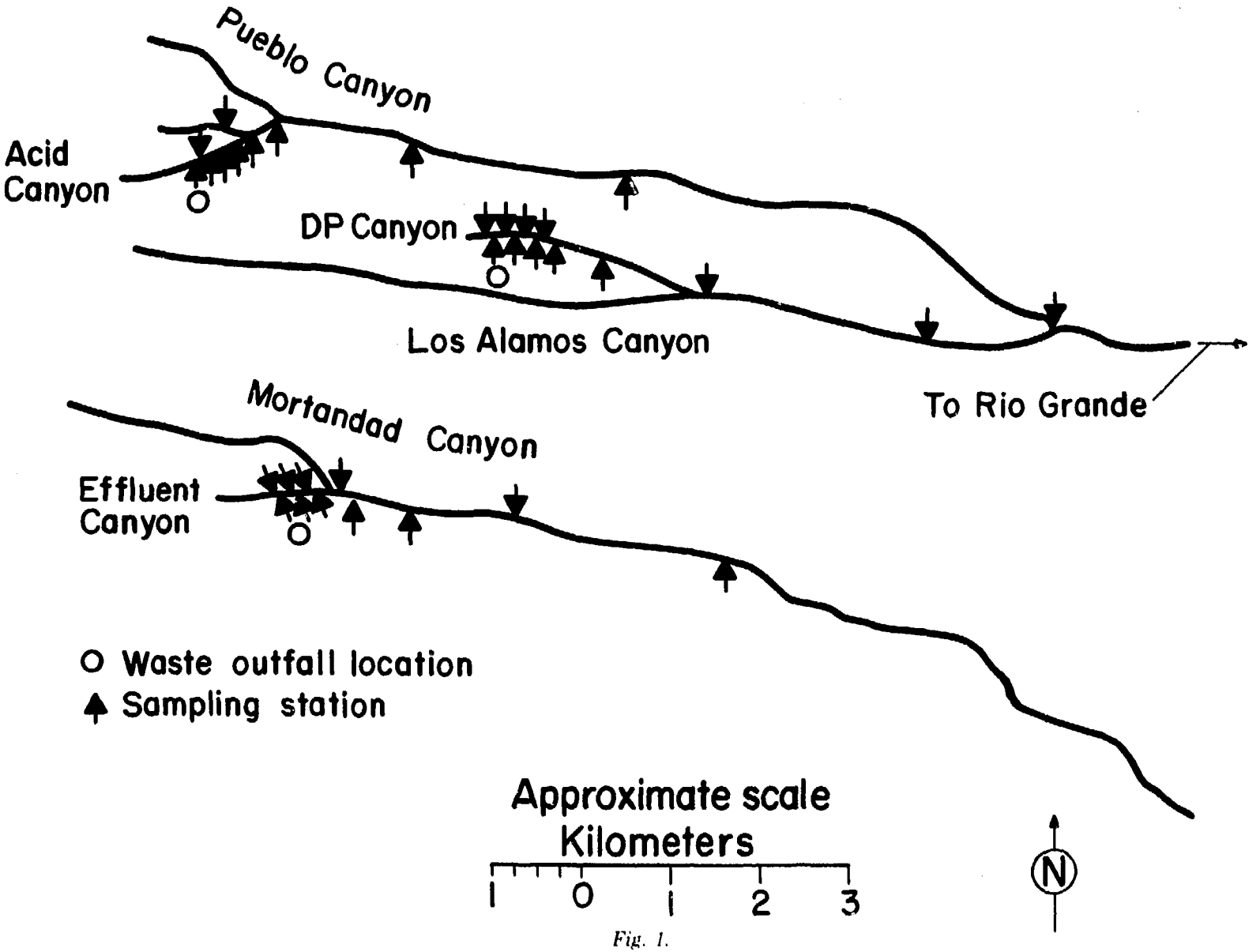

Fig. 1.
Location of sampling stations in canyon study areas.

segment (L) where C applied was taken as half the distance to the last upstream sampling station plus half of the distance to the next downstream sampling station. The total ^{137}Cs inventory for each canyon was calculated from the sum of the ^{137}Cs activity (I) found in all of the channel segments from each canyon.

IV. RESULTS

A. ^{137}Cs Spatial Relationships

Maximum concentrations of soil ^{137}Cs were detected during both sampling periods within 160 m below the waste outfalls, and ^{137}Cs levels gradually decreased with distance downstream (Table I). Elevated ^{137}Cs concentrations were measured throughout DP-Los Alamos Canyon and to post-outfall distances of 2.56 km in Effluent-Mortandad Canyon, whereas concentrations in Acid-Pueblo Canyon soils were generally less than worldwide fallout levels (up to 1 pCi ^{137}Cs/g) previously reported for New Mexico soils (Herceg, 1972 and 1973; Shiager and Apt, 1974).

The elevated levels of soil ^{137}Cs detected in the lower reaches of DP-Los Alamos Canyon (>1.5 km downstream from the waste outfall) and Effluent-Mortandad Canyon (1.5-2.56 km downstream from the waste outfall) showed that ^{137}Cs movement has occurred into portions of the stream channel which are normally dry even during a discharge of liquid wastes. The only significant quantities of surface water which occur at distances farther than 1.5 km from the outfalls consist of runoff resulting from snowmelt or rainstorms. Thus, these data support the findings of recent studies at Los Alamos which suggest hydrologic transport of contaminated soils by storm runoff as a transport vector in the downstream movement of radioactivity in these canyons (Purtymun, 1974 and Hakonson et al., 1975).

Soil ^{137}Cs concentrations in Effluent-Mortandad and DP-Los Alamos Canyons did not exhibit a consistent relationship with soil depth at all the sampling locations. In some cases, cesium concentrations decreased steadily with depth and at other locations they exhibited just the opposite pattern. These data reflect considerable vertical mixing of soil ^{137}Cs by hydrologic transport processes in the canyons currently receiving treated effluents. Levels of ^{137}Cs in the soils of Acid-Pueblo Canyon increased abruptly with depth in some locations (i.e., 80 m from the waste outfall) where soil had accumulated, but ^{137}Cs concentrations were generally near worldwide fallout levels at most of the sampling locations. Although the quantity of ^{137}Cs released to Acid-Pueblo Canyon was never recorded, the elevated levels of soil ^{137}Cs measured in some of the sampling locations indicated that this isotope was a component of the waste entering the canyon. Consequently, the ^{137}Cs distributions observed in 1972 and 1973 (Table I) indicate that dilution and loss of ^{137}Cs-contaminated soils have occurred in this canyon which has not received effluent additions for the past 10 yr.

Soils from eight different locations in Effluent-Mortandad and in DP-Los Alamos Canyons were sampled during 1973 to determine the horizontal and vertical variation of ^{137}Cs concentrations in the alluvium (Table II). The coefficients of variation (standard deviation divided by the mean) of soil ^{137}Cs concentrations ranged from 0.16 to 1.2, and generally were less than 0.40. The coefficients of variation for levels of soil ^{137}Cs were generally consistent with distance from the outfall and soil depth, although some sampling locations exhibited larger coefficients of variation with increased depth in the soil profile. Since the variation due to counting was small (coefficients of variation less than 0.01), these data probably reflect a nonuniform penetration of ^{137}Cs into the alluvium.

B. Soil ^{137}Cs Inventories

The amount of soil ^{137}Cs found in successive stream channel segments in 1972 and 1973 was calculated from Effluent-Mortandad (Fig. 2), DP-Los Alamos (Fig. 3), and Acid-Pueblo (Fig. 4) Canyons. The total soil ^{137}Cs inventory found in each effluent-receiving area in 1972 and 1973 was calculated and compared with estimated amounts of ^{137}Cs added to the canyons in the liquid effluents during this time (Table III).

Inventory estimates of soil ^{137}Cs in all three effluent-receiving areas were higher in 1973 than in 1972 (Table III). These increases reflect waste additions between sampling periods in Effluent-Mortandad and DP-Los Alamos Canyons, which are currently receiving treated effluents. However, the increased ^{137}Cs inventory in Acid-Pueblo Canyon

TABLE I

^{137}Cs IN ALLUVIAL SOILS (FROM CENTER OF STREAM CHANNEL) FROM LIQUID WASTE DISPOSAL AREAS AT LOS ALAMOS IN 1972 AND 1973

Distance from Waste Outfall	^{137}Cs Concentration (pCi/g) at Each Soil Depth							
	0 - 2.5 cm		2.5 - 7.5 cm		7.5 - 12.5 cm		Remainder[a]	
	1972	1973	1972	1973	1972	1973	1972	1973
Effluent-Mortandad Canyon								
−100 m[b]	ND[c]	1.9	1.6	2.1	14	0.76	0.3	
0 m	3100	1740		2010		2530		140
20 m	1090	1820	180	2410	22	74		
40 m	720	2660	1350	1400	1070	650		310
80 m	920	2180	1300	2710	770	1240	22	470
160 m	3360	3260	1230	800	---[d]	1270	1960	1120
320 m	210	1740	640	720	1240	530	290	110
640 m	300	1040	370	710	410	800	470	790
1.28 km	130	230	180	240	110	160	68	76
2.56 km	67	65	150	120	88	70	18	23
5.12 km	1.3	1.2	ND	1.6	ND	0.8	0.1	0.9
DP-Los Alamos Canyon								
−100 m	ND	0.25	ND	0.33	---	0.45	ND	0.50
0 m	820	960	570	1020	100	750	52	600
20 m	440	800	560	1130	210	340	250	
40 m	28	430	2.3	430	3.2	300	2.9	13
80 m	750	880	160	620	2.3	81	5.2	17
160 m	15	10	21	17	19	33	45	28
320 m	11	12	13	23	10	39	20	28
640 m	42	0.3	91	35	59	38	42	42
1.28 km		0.5		53		55		33
2.56 km	12	1.8	14	1.9	18	2.4	15	
5.12 km	1.1	2.3	1.7	11	3.2	40	2.8	
Acid-Pueblo Canyon								
−100 m	ND	0.20	ND	0.26				
0 m	ND	4.3	ND	0.3		0.6		2.1
20 m	3.9	0.6		0.3				
40 m	0.4	0.5	1.4	0.7	ND	1.1	0.1	2.9
80 m	ND	0.3	5.4	1.3	29	1.3	20	22
160 m	0.3	1.2	0.5	3.4	1.7	3.0	1.2	2.9
320 m	ND	0.29	1.0	1.2	1.8	1.0	0.9	1.1
640 m	0.3	1.9	0.8	1.6	ND	1.2	0.7	2.2
2.56 km	ND	1.6	---	0.6	---	0.8	ND	
5.12 km	ND	2.5	ND	1.7	ND	0.8	ND	0.7
10.2 km	ND	0.44	ND	0.29	ND	0.44	0.2	0.8

[a]The remainder section was comprised of soil from the 12.5-cm depth to a maximum of 30 cm.
[b]Negative values represent distances upstream from the waste outfalls.
[c]ND = less than minimum sensitivity (95% confidence level) of analysis (0.08 pCi ^{137}Cs/g).
[d]Missing data.

TABLE II

COEFFICIENT OF VARIATION OF ^{137}Cs CONCENTRATIONS IN SOIL AT SELECTED LOCATIONS IN EFFLUENT-MORTANDAD AND DP-LOS ALAMOS CANYONS

Distance from Waste Outfall	Coefficient of Variation of Soil ^{137}Cs Concentrations at Each Soil Depth			
	0-2.5 cm	2.5-7.5 cm	7.5-12.5 cm	Remainder[a]
Effluent-Mortandad Canyon				
0 m	0.30 (10)[b]	0.25 (10)	0.49 (6)	
40 m	0.28 (10)	0.43 (10)	1.0 (6)	
640 m	0.27 (10)	0.24 (10)	0.16 (10)	0.28 (8)
2.56 m	0.20 (10)	0.23 (10)	0.35 (10)	0.27 (9)
DP-Los Alamos Canyon				
0 m	0.50 (10)	0.91 (8)		
40 m	0.24 (10)	0.18 (10)	0.54 (10)	1.2 (10)
640 m	0.19 (10)	0.16 (10)	0.33 (10)	0.16 (10)
2.56 km	0.33 (10)	0.34 (9)		

[a]The remainder section was comprised of soil from the 12.5-cm depth to a maximum of 30 cm.
[b]Numbers in parentheses represent number of core segment samples assayed for ^{137}Cs.

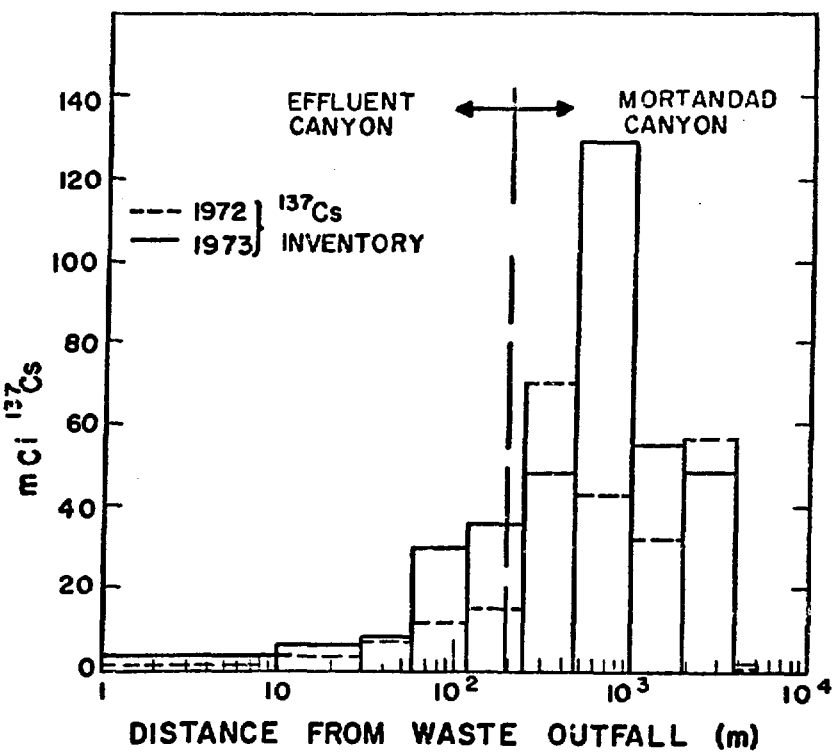

Fig. 2.
Soil ^{137}Cs inventory as a function of distance from the Effluent-Mortandad Canyon waste outfall in 1972 and 1973.

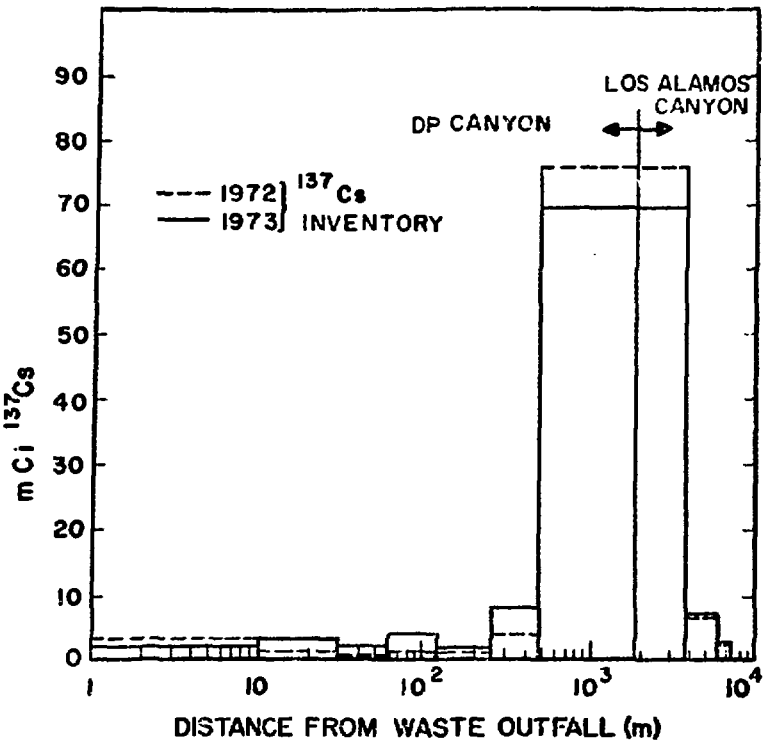

Fig. 3.

Soil ^{137}Cs inventory as a function of distance from the DP-Los Alamos Canyon waste outfall in 1972 and 1973.

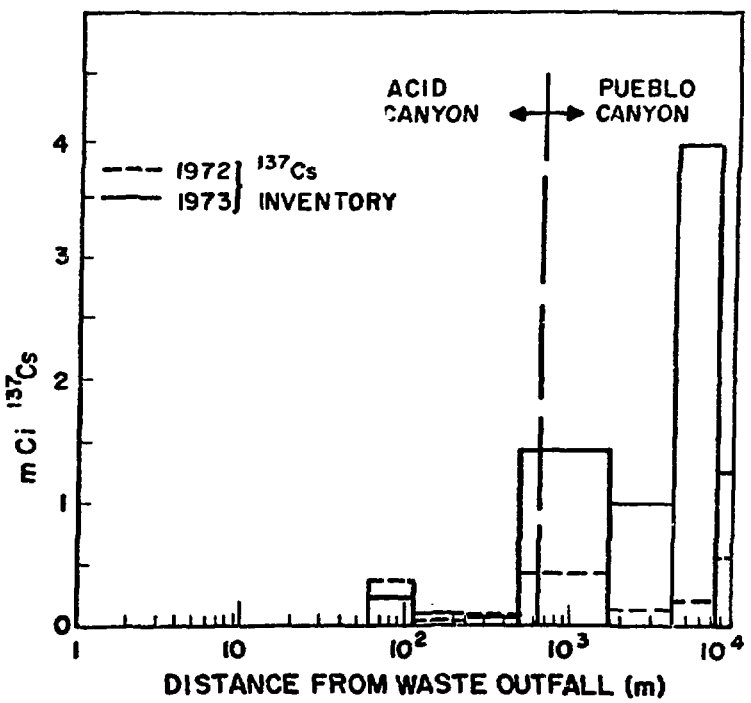

Fig. 4.

Soil ^{137}Cs inventory as a function of distance from the Acid-Pueblo Canyon waste outfall in 1972 and 1973.

TABLE III

TOTAL ^{137}Cs INVENTORIES IN ALLUVIUM OF LIQUID WASTE DISCHARGE AREAS

Liquid Waste Discharge Area	Total Observed ^{137}Cs Inventory (mCi) in Soils			Estimate of ^{137}Cs Added to Canyons in Liquid Wastes (mCi)[a]
	1972 Sampling Period	1973 Sampling Period	Increase in Inventory	
Effluent-Mortandad Canyon	237	363	126	127
DP-Los Alamos Canyon	95.4	100	4.60	1.00
Acid-Pueblo Canyon	1.94	8.19	6.25	0.0

[a]Estimate based on average annual input of ^{137}Cs in liquid wastes to canyons (1973-4 data) and the length of time between soil sampling periods.

cannot be attributed to waste additions, since this canyon has not received effluents since 1963. The ^{137}Cs inventory based on soil radionuclide data collected in Effluent-Mortandad Canyon increased by an estimated 126 mCi between October 1972 and May 1973, corresponding closely to an effluent addition of 127 mCi ^{137}Cs according to the waste treatment plant's records (Table III). All of the increased ^{137}Cs inventory was measured within 1.5 km of the effluent outfall, which corresponds to the extent of surface flow of the liquid effluents in this canyon. The additional 50-60 mCi ^{137}Cs found 2-5 km from the waste outfall represents ^{137}Cs which was translocated during runoff events from snowmelt and late summer rainstorms.

Only two stream channel segments in Effluent-Mortandad Canyon had estimated ^{137}Cs inventories that were lower in 1973 than in 1972 (Fig. 2). Losses from the 240-480 segment may be due to enhanced scouring processes which occur in this portion of the effluent pathway, resulting in a higher ^{137}Cs inventory in the 480-960 m segment in 1973 than in 1972. The decrease in the radionuclide inventory in the 240-480 m canyon segment probably occurred because Mortandad Canyon is a larger watershed than Effluent Canyon, resulting in large amounts of soil and runoff water entering the waste discharge pathway at the junction of Effluent and Mortandad Canyons. The observed losses from the 1920-3840 m segment were not statistically significant changes in ^{137}Cs inventories, i.e., the 1972 and 1973 inventory estimates were both within 30% of the mean amounts of ^{137}Cs found in this area for either sampling period (Table II).

The general distribution of ^{137}Cs in DP-Los Alamos Canyon soils is consistent with our knowledge of hydrologic and soil transport processes in this intermittent stream (Purtymun, 1974). In DP-Los Alamos Canyon soils, all of the estimated 4.6 mCi increase in ^{137}Cs inventory from 1972 to 1973 occurred within 120 m of the waste outfall. However, 89-94% of the ^{137}Cs inventory occurred in the lower portion of this canyon system (i.e., 120-6680 m below the waste outfall) for both sampling periods (Fig. 3). Since this segment of the stream channel is normally dry except during periods of storm runoff, the horizontal distribution of ^{137}Cs in this canyon reflects periods of intense runoff from late summer rainstorms, which move soil and ^{137}Cs considerable distances downstream.

The estimated ^{137}Cs inventory found in Acid-Pueblo Canyon soils increased from 1.9 to 8.2 mCi from 1972 to 1973 (Table III), and most of this apparent increase occurred from 480 m to 10.2 km below the waste outfall (Fig. 4). Since this canyon system has not received liquid wastes for the last 10 yr, and generally contained worldwide fallout levels of soil ^{137}Cs, the increased inventory probably does not reflect statistically significant temporal differences in ^{137}Cs inventories. However, other casual factors for the increased ^{137}Cs inventory cannot be ruled out at this time, i.e., the calculated increase in ^{137}Cs inventory may represent release of ^{137}Cs from the stream bank soils or from layers of weathering volcanic tuff below the stream channel.

V. DISCUSSION

The use of soil ^{137}Cs data for estimating temporal changes in inventories should be further explored in view of current interest in environmental monitoring and radioecological research programs. The techniques used in the present study for calculating changes in the total inventory of ^{137}Cs in the soils of the effluent-receiving areas provided a good estimate of the ^{137}Cs additions to Effluent-Mortandad Canyon, where the total inventory increased by 54% over an 8-month period. However, when additions of ^{137}Cs were low relative to the variation in ^{137}Cs levels, such as in DP-Los Alamos Canyon, temporal changes in the radionuclide inventories could not be accurately estimated. The alluvium in Acid-Pueblo Canyon generally contained worldwide fallout levels of ^{137}Cs during both sampling periods, resulting in nonsignificant temporal differences in the inventories of ^{137}Cs added to this canyon from the waste treatment facility over 10 yr ago. A more intensified sampling scheme and a better definition of the ^{137}Cs source term may be needed to detect significant temporal inventory changes in DP-Los Alamos and Acid-Pueblo Canyons.

The physical transport of alluvium by snowmelt and rainstorm runoff is an important factor in understanding the distribution of ^{137}Cs (and other radionuclides) in the alluvial soils at Los Alamos, since it is generally recognized that ^{137}Cs is fixed by Los Alamos soils and tuff (Christenson et al., 1958) in a relatively immobile form. Gross-alpha and -beta radionuclide measurements in Effluent-Mortandad Canyon soils suggested a seasonal pattern of radionuclide distributions, with a buildup occurring from fall until spring (a period similar to that discussed in this report), followed by extensive losses of radioactivity during runoff events from July through September (Purtymun, 1967).

The inventory data presented in Figs. 2 and 3 support and further elucidate the fall-spring portion of the seasonal pattern of radionuclide distributions postulated by Purtymun (1967). The radionuclide inventory data for Effluent-Mortandad (Fig. 2) and DP-Los Alamos (Fig. 3) Canyons indicate that there was a gradual increase in the ^{137}Cs inventories close to the waste outfalls during the winter months. Although snowmelt runoff was recorded in the canyons between the 1972 and 1973 sampling periods, flows were not sufficient to cause major shifts in the inventories of alluvial soils. This is because, although runoff transport of radioactivity is a major transport mechanism in these effluent-receiving areas, runoff from snowmelt is usually not sufficient to cause turbulent flow in these intermittent streams. In addition, ice formation on the sides of the stream channels during a large portion of the winter months limits ^{137}Cs and soil movement. In contrast, the turbulent runoff from late summer rainstorms may transport about 2% of the ^{137}Cs inventory in the canyon into the normally dry portion of the stream during one relatively small storm (Hakonson et al., 1975).

Although we have determined the horizontal and vertical distribution of soil ^{137}Cs and the temporal changes in ^{137}Cs inventories in the effluent-receiving areas, much additional research is needed to fully evaluate ^{137}Cs seasonal patterns in the soils and biota of these ecosystems. The variations in soil ^{137}Cs concentrations merit further investigation, since sample variation has such an important bearing on the level of sampling effort required to detect statistically significant inventory changes with time. Solution and sediment transport of ^{137}Cs should be evaluated seasonally in these intermittent streams, in view of the temporal variation of turbulent flow processes and the potential role of these processes in radionuclide redistribution. The potential role of nonradioactive soils reacting with soluble ^{137}Cs in the stream channel should be evaluated, as well as soil-plant ^{137}Cs relationships. Judicious selection of sampling periods should also be initiated to provide useful information on the long-term behavior of ^{137}Cs in the environment.

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