CONF-960312--72

LA-UR- 96-633

Title: MIGRATION OF Sr-20, Cs-137, AND Pu-239/240 IN CANYON BELOW LOS ALAMOS OUTFALL

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Submitted to: Waste Management '96, Tucson, AZ, February, 1996

Los Alamos
NATIONAL LABORATORY

Form No. 836 R5
ST 2629 10/91

Migration of Sr-90, Cs-137, and Pu-239/240 in Canyon Below Los Alamos Outfall

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Waste Management '96
Tuscon, Arizona
February 25-29.1996

Abstract

Technical Area-21 (TA-21) of Los Alamos National Laboratory (LANL) is on a mesa bordered by two canyons DP Canyon and Los Alamos (LA) Canyon. DP Canyon is a small semi-arid watershed with a well defined channel system where the stream flow is ephemeral. TA-21 has had a complex history of waste disposal as research to determine the chemical and metallurgical properties of nuclear materials occurred here from 1945-1978. Due to these operations, the TA-21 mesa top and bordering canyons have been monitored and characterized by the LANL Environmental Restoration Program. Results identify radionuclide values at outfall 21-011(k) which exceed Screening Action Levels, and points along DP Canyon which exceed regional background levels.

The radiocontaminants considered in this study are strontium-90, cesium-137, and plutonium-239 . This research examines sediment transport and speciation of radionuclide contaminant migration from a source term named SWMU 21-011(k) down DP Canyon. Three dimensional surface plots of data from 1977-1994 are used to portray the transport and redistribution of radioactive contaminants in an alluvial stream channel. An overall decrease in contamination concentration since 1983 has been observed which could be due to more stringent laboratory controls and also to the removal of main plutonium processing laboratories to another site.

Work supported by LANL ER Program, Field Unit-1 project leader-Garry Allen

INTRODUCTION

Technical Area-21(TA-21) of Los Alamos National Laboratory (LANL) lies on the northern section of the Laboratory, at an elevation of 7140 ft. It is located on the Pajarito Plateau, midway between the steep slopes of the Jemez mountains to the west and the Rio Grande river to the east. TA-21 lies on DP Mesa, immediately east-southeast of the Los Alamos town site. TA-21 was a plutonium processing facility for nuclear weapons research and development from 1945 to 1975. Liquid radioactive waste was treated at site treatment plant from 1969 to 1984. TA-21 is currently being decontaminated and decommissioned.

The Liquid Radioactive Waste Treatment Plant processed aqueous effluent from TA-21 chemical laboratories. The effluent included the isotopes plutonium-239, cesium-137, and strontium-90, among other radionuclides. The effluent was discharged into DP Canyon via an outfall referred to as 21-011(k). A map of TA-21 and DP canyon is shown in figure 1. DP Canyon flanks TA-21 to the north, has east-west orientation, has steep rocky walls and has ephemeral flow from

midway between the steep slopes of the Jemez mountains to the west and the Rio Grande river to the east. TA-21 lies on DP Mesa, immediately east-southeast of the Los Alamos town site. TA-21 was the plutonium processing facility for nuclear weapons research and development from 1945 to 1978. After 1978, operations were scaled back dramatically but not stopped completely. Liquid radioactive waste was treated at a site treatment plant from 1969 to 1995. TA-21 is currently being decontaminated and decommissioned.

The Liquid Radioactive Waste Treatment Plant processed aqueous effluent from TA-21 chemical laboratories. The effluent included the isotopes plutonium-239/240, cesium-137, and strontium-90, among other radionuclides. After processing, the effluent was discharged into DP Canyon via an outfall now referred to as SWMU 21-011(k). A map of TA-21 and DP canyon is shown in Figure 1. DP Canyon is the small east-west oriented canyon which flanks TA-21 on the north. The walls of the canyon are steep and rocky. The stream channel on the floor of the canyon receives ephemeral flow from precipitation and runoff (summer storms and winter snowmelt). Sediments in the stream channel consist of sand and illitic clay. DP Canyon joins Los Alamos Canyon approximately 1.5 km downstream from SWMU 21-011(k).

Place Figure 1 here.

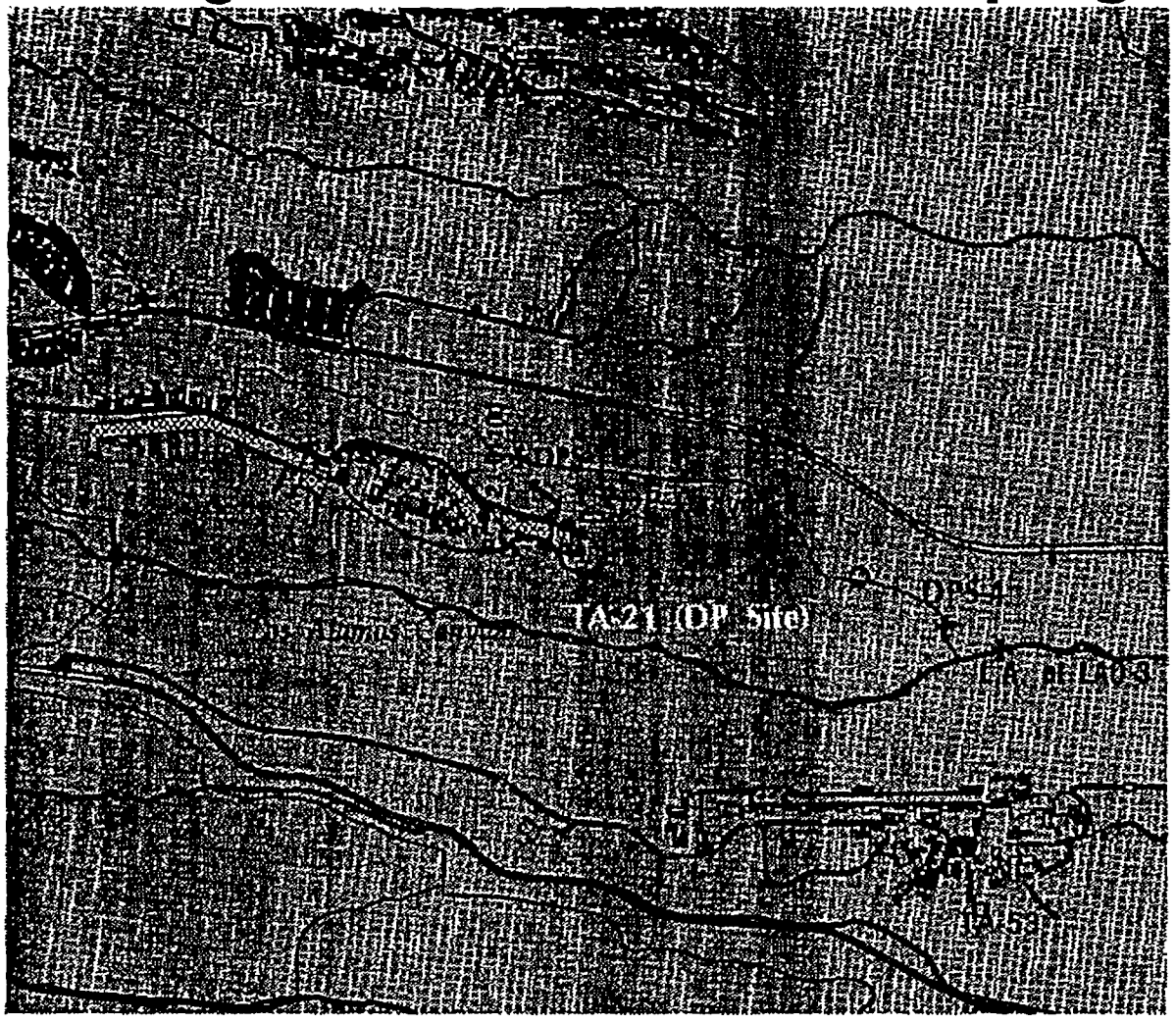
SWMU 21-011(k) received processed waste effluent streams from 1969 until 1978. From 1978 until 1984, it received chemical waste containing various radionuclides. A 400 L aqueous spill of untreated effluent probably containing various contaminants, occurred at the outfall in 1991.[1] The clay pipe at 21-011(k) released effluent into a precipitous rock strewn slope with minimal soil depth.

Sediment Sampling Technique

This investigation examined temporal trends in available data for radionuclides at sediment sampling stations in DP Canyon. The sampling locations included the SWMU site, two locations (DPS-1 and DPS-4) in DP Canyon, and the site of well LAO-3, near the confluence of DP and Los Alamos Canyons (See Figure 1). The first site was sampled in the summers of 1992 and 1993.[2] The last three sites have been sampled repeatedly by the Environmental Protection Group.[3].

Stream bed samples were taken from sampling stations along DP Canyon shown on Figure 1. A consistent sampling procedure for the sediments was repeated annually to ensure comparability of the results. Samples from the bed of DP stream were collected by digging a line of uniform depth in an undisturbed area across the main channel.[4] Using a trowel, 500 ml of sediment was collected at a depth of one-half to three-fourths inch. Soil from SMWU 21-011(k), in contrast with sediment, samples were taken at a depth of one to six inches and six to eighteen inches in the soil. Care was taken to collect the clay-rich fraction with which the radionuclides are commonly associated.[5] The sediment sample is mixed prior to analysis and analyzed at the LANL *Environmental Measurements Lab* and each year the field samples were taken within a period from late April through May.[5]

Fig. 1 TA-21 Sediment Sampling Stations



Drainages
Springs
Env. Surv. - Soil Sampling Stations
Structures - Perm Buildings
Material Disposal Areas
Transportation - Paved Roads
Boundaries - Operable Units

J.Murphy, CST-7, LANL

[illegible]	[illegible]	[illegible]
DPS-1	1633325.1943	1774858.4216
DPS-4	1637502.6961	1773289.9210
LosAlamos At Bridge	1618259.7538	1775612.8922
LosAlamos At LAO-1	1629407.3780	1773946.5074
LosAlamos At GS-1	1648151.1161	1770889.4199
LosAlamos At LAO-3	1638047.6964	1771074.5209
LosAlamos At LAO-4.5	1643654.3653	1772135.8231
LosAlamos At SR4	1651895.1870	1771535.9279

1500 0 1500 3000 Feet

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The Environmental Chemistry Group at LANL used a documented Quality Assurance and Sampling Procedure for the sample analysis and data verification. The analyses of radioactive constituent concentrations were determined by gamma-ray spectrometry.

Table I portrays the regional background concentration, the Screening Action Level (SAL), and half life of the three elements.

Place Table I here.

A SAL is set on potential contaminants in various media based on conservative calculations of human health risk. Soil SALs for radionuclides are calculated using a residential scenario at an annual dose limit of 10 millirem (10 mrem/a) above regional background.

Plutonium-239 decays by alpha- and gamma-emission with a half life of 24,100 years (a). Plutonium-240 is also an alpha- and gamma-emitter, with a half-life of 6560 a. Cesium decays by beta-and gamma-emission, with a half life of 30.17 a. Strontium decays by beta-emission, with a half-life of 28 a.

Clay minerals are very effective at retaining these radionuclides. As a monovalent alkali cation, cesium readily exchanges with potassium, and is commonly strongly bound to surface and interlayer sites of clay particles.[6] Thus, contaminant cesium is expected to be associated with the finer fractions (especially the <0.053 mm fraction) in soils and sediments.[7] Transport of cesium commonly occurs in association with soil and sediment erosion.[7] There is evidence of substantial drainage erosion from the SWMU 21-011(k) clay pipe down the slope to DP stream in the form of two drainage channels.

Strontium is a divalent alkali earth cation which readily substitutes for calcium in mineral lattices and on surface sites. Although transport of strontium will also be largely through sediment transport, sorption on mineral grains is generally less strong for strontium, and hence, transport in runoff may also be significant.

Because of their similarity to potassium and calcium, respectively, cesium and strontium are also bioactive (readily absorbed by plants, soil microbes, and animals). Plants and microbes whose growth is limited by calcium and/or potassium will tend to take up larger amounts of strontium and/or cesium.[8]

RESULTS AND DISCUSSION

This section discusses the present concentrations in soil and sediment at SWMU 21-011(k), and the variations with time of radionuclide activities at the sediment sampling stations downstream. The three radionuclides discussed are Pu-239/240, Sr-90, and Cs-137. (Note that the standard measurements for plutonium do not distinguish the two isotopes, so the value reported is for the sum). At the source [SWMU 21-011(k)], Pu-239/240 activities range from 7.2 to 46,000 pCi/g,

Table I
Properties of Radionuclides

Radionuclide	Background * pCi/g	SAL pCi/g	Half-Life years
Strontium-90	0.25	4.4	29.1
Cesium-137	0.35	5.1	30.2
Plutonium-239	0.009	24	24,100

* Fresquez, P.R., et al. Radionuclides and Radioactivity in Soils Within and Around Los Alamos National Laboratory: 1974 to 1994. LA-UR-95-3671. 1995.

Cs-137 activities ranged from 8.23 to 2675 pCi/g, and Sr-90 activities ranged from 1.96 to 1800 pCi/g. Figures 2,3, and 4 show the variations in activities as a function of time and location in DP Canyon. Table II summarizes the data on radionuclides activities in DP Canyon.

The plutonium chart (Figure 2) portrays the maximum activities occuring at DPS-1, with values at DPS-4 and LAO-3 consistently below 1.0 pCi/g. After 1986, the DPS-1 activities are likewise below 1.0 pCi/g. The outfall clearly displayed high activities (1900 times the soil SAL) after the decline in levels at DPS-1, so it would appear that transport of plutonium was facilitated by the effluent from the outfall. Without the continued effluent, relatively small degrees of transport occurred to the stream channel, and the remaining contamination at DPS-1 was moved downstream and mixed with less contaminated soil by runoff.

Place Table II here.

Place Figure 2 here

Plutonium does not appear to be migrating to DPS-1 or DPS-4 at levels above the SAL. Cesium historically has had high levels at DPS-1, and DPS-4 but currently the levels are well below the SAL. The strontium data is similar. There is an evident decrease in the three radionuclides downstream.

The cesium data (figure 3) indicate that concentrations were above the SAL at all three stations until the late 1980s, but have subsequently declined to activities at or near background levels. The high levels seen at SWMU 21-011(k) are not found in the stream channel, so dispersion and mixing of contaminated sediment appears to have diluted the concentration of cesium. In addition, the cessation of effluent release appears to have permitted contamination levels in the channel to decline through transport and/or dilution. However, the activities remained higher for cesium than for plutonium, and cesium is the predominant radionuclide contaminant in DP Canyon. This conclusion is corroborated by the EG&G aerial radiological gamma surveys in 1975 and 1982.[9] These surveys show that cesium is the dominant anthropogenic radionuclide found outside of TA-21, and that it had migrated approximately 600 meters down DP Canyon.

Place Figure 3 here.

The strontium data (Figure 4) show a pattern similar to that for cesium, with high values at the source, values above the SAL at DPS-1 until the mid-1980s, and values below the SALs, at all stations since then. The values are lower than those for cesium, which may reflect the composition of the original effluent, but may also represent the lower sorptivity of strontium. Strontium may have been mobilized into the surface water and alluvial ground water, and transported deeper, or further downstream.

Place figure 4 here

Table III shows sorption coefficients for all three elements. Strontium is less strongly sorbed onto clay mineral surfaces than plutonium and cesium, but appears to have behaved in a manner

Table II.
Radionuclide Concentrations in DP Canyon (pCi/g)

Element	SAL	21-011(k)	DPS-1 maximum	DPS-1 1994	DPS-4 maximum	DPS-4 1994
Pu-239	24	46,000*	8.11	0.16	0.25	0.1
Cs-137	5.1	2675*	23.9	1.9	21	1.9
Sr-90	4.4	1800*	14	1	4.6	4

*1) FIMAD, Oracle database, Los Alamos National Laboratory (1995)
2) ENVIRONMENTAL PROTECTION GROUP, "Environmental Surveillance at Los Alamos Reports 1977-1994", Los Alamos National Laboratory (1977-1994).

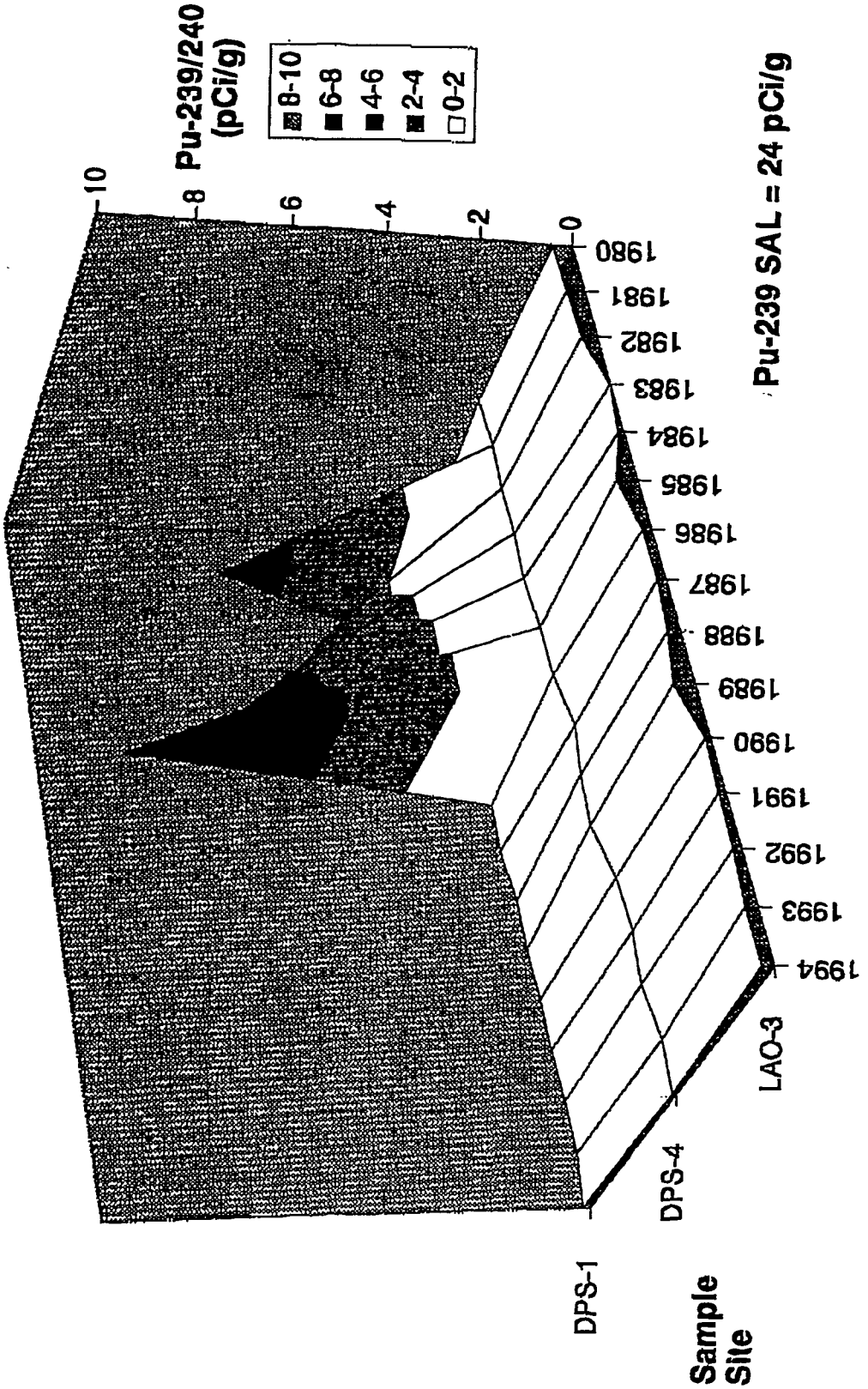

Figure 2 Plutonium-239/240 in DP Canyon Sediment

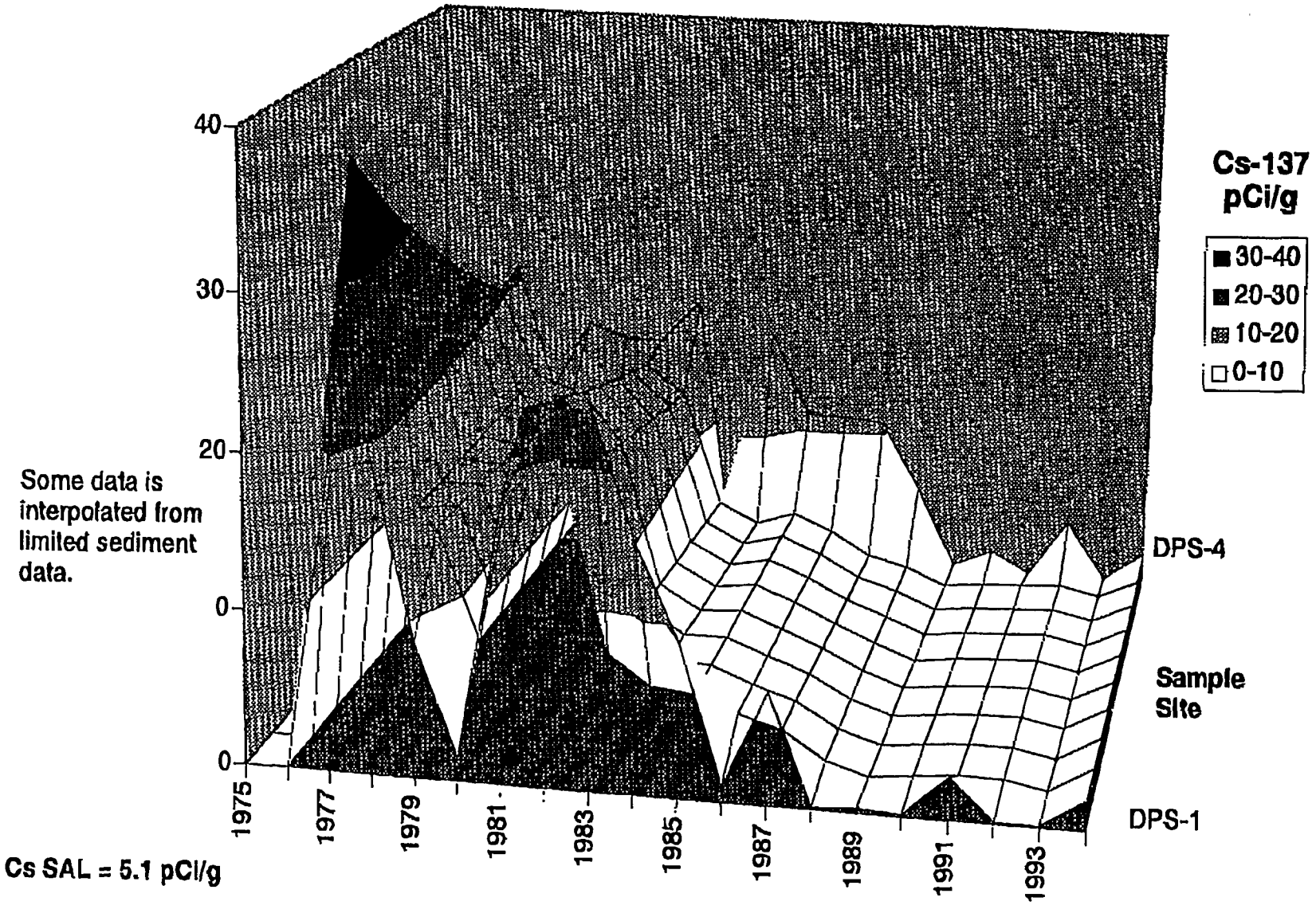

Figure 3. Cesium-137 in DP Canyon Sediment

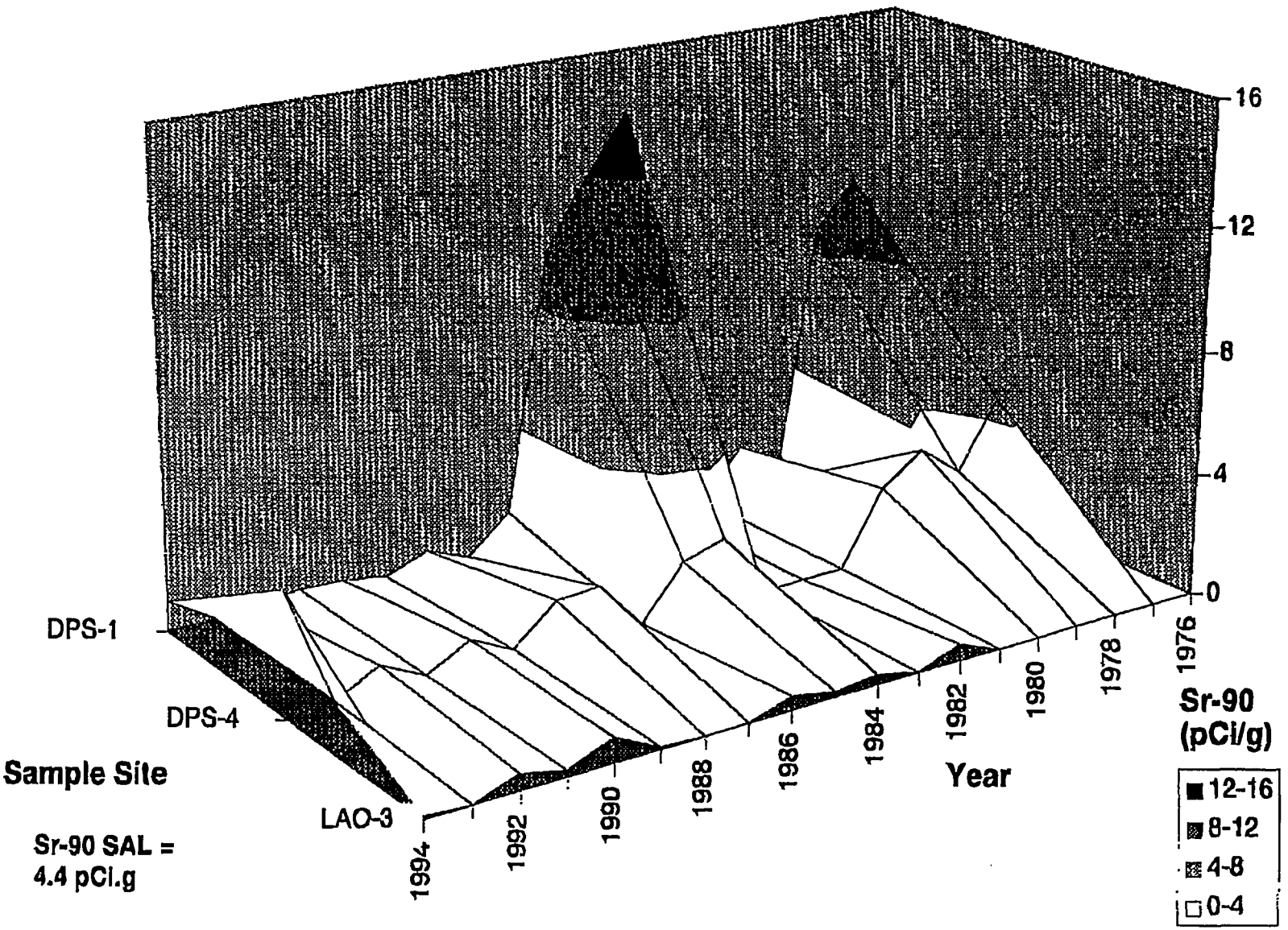

Figure 4. Strontium-90 in DP Canyon sediment.

Table III
Sorption Coefficients

ELEMENT	Mean Soil (Kd)	Mean Sediment (Kd)
Pu-239	≤500*	≤500*
Cs-137	438.87	430.04
Sr-90	35.7	21.4

* 1) C.F.V. Mason and N.Lu, estimated, LANL, 1996.
2) S. KUNG, "Sorption and Desorption of Cesium and Strontium on TA-2 and TA-41 Soils", Progress Report, Los Alamos National Laboratory (1995).

similar to cesium in the sediment of DP Canyon. The pattern for plutonium appears unusual in the light of its estimated sorption coefficient, as it is expected to sorb slightly less strongly than cesium, yet it appears hardly to have been transported at all. It is conceivable that plutonium in the effluent was sorbed onto or precipitated in extremely fine colloidal particles which percolated into the soil to some depth. The cesium and strontium, which sorbed rapidly onto clay or silt particles at the surface may have been more susceptible to transport in surface runoff. Further analysis is underway to test this concept.

Place Table III here

Strontium is less strongly sorbed onto clay mineral surfaces than plutonium and cesium, but appears to have behaved in a manner similar to plutonium in the sediment of DP Canyon. Strontium is likely to remain stored in the banks of the stream channel, but to a smaller extent.[10] Strontium sorbs to the LA Canyon sediments less strongly than cesium by a factor of 12. Batch strontium sorption experiments were conducted on 20 soil and 16 channel sediment samples collected in TA-21, and upper Los Alamos Canyon. Cesium sorption coefficients (Kd) ranged from 162.3 to 1444.3 ml/g in soil and 77.5 to 1034.1 ml/g in sediment.[11] Strontium sorption coefficients ranged from 15.8 to 67.7 ml/g, with a mean of 35.7 ml/g for soils. For strontium in LA Canyon channel sediments sorption coefficients ranged from 8.8 to 41.3 ml/g, with a mean of 21.4 ml/g.[11] These data suggest that strontium is a non-conservative solute that is partially removed from solution through cation exchange.

CONCLUSIONS

The outfall at SWMU 21-011(k) is a major source for radionuclide contamination in DP Canyon. Plutonium has not migrated from the source in concentrations sufficient to exceed the SAL, and activities declined very rapidly after effluent ceased to be released into the SWMU. Cesium and strontium have been transported in quantities sufficient to exceed the SALS, although only cesium activity exceeded SALS at station DPS-4 after 1977. Both declined in activity after effluent ceased to be released into the SWMU. Recent year samples have all shown activities below the SAL values. The systematic relationships among multiple contaminants are complex, involving interactions among surface and ground water hydrologic and geochemical processes, and understanding may also require detailed knowledge of the effluent discharge and surface water history.

ACKNOWLEDGMENTS

This work was funded by the US Department of Energy through the Los Alamos National Laboratory Environmental Restoration Project and Field Unit-1 Project leader- Garry Allen. The authors thank Robert K. Greene, Thane Hendrix, Ningping Lu, Maxwell N. Maes, Gary McMath, David B. Rogers, Ines Triay, William R.J. R. Turney, and Douglas Walther.

REFERENCES

1. D. SALAZAR, Personal communication. Los Alamos National Laboratory (July 1995).

2. ENVIRONMENTAL RESTORATION PROGRAM, "TA-21 Operable Unit RCRA Facility Investigation - Outfalls Investigation", Los Alamos National Laboratory (February 1994).

3. ENVIRONMENTAL PROTECTION GROUP, "Environmental Surveillance at Los Alamos Reports 1977-1994", Los Alamos National Laboratory (1977-1994).

4. ENVIRONMENTAL PROTECTION GROUP, "Environmental Surveillance at Los Alamos during 1993", LA-12973-ENV, Los Alamos National Laboratory (1993).

5. M.N. MAES, Personal communication, Los Alamos National Laboratory (October 1995).

6. T. TAMURA, D.G. JACOBS, "Structural Implications in Cesium Sorption", *Health Physics*, 2, 391-8 (1960).

7. N. LU, C.F.V. MASON, W.R.J.R. TURNEY, "Characterization and Immobilization of Cesium-137 in a Canyon Adjacent to the Site of the Plutonium R& D Facilities at Los Alamos National Laboratory", LA-UR-95-4126. Los Alamos National Laboratory (November 1995).

8. M. CRESSER et al, "Soil Chemistry" Environmental Chemistry Series, Cambridge University, Press Cambridge (1993).

9. A.E. FRITZSCHE, "An Aerial Radiological Survey of Technical Areas 2,21, and 53 and Surroundings-Los Alamos National Laboratory", EG&G Energy Measurements, EGG-10617-1030 (1990).

10. P.A. LONGMIRE, et al, "Hydrochemistry of Upper Los Alamos Canyon, Los Alamos, NM", New Mexico Geological Society Field Trip Guidebook (1996).

11. S. KUNG, "Sorption and Desorption of Cesium and Strontium on TA-2 and TA-41 Soils", Progress Report, Los Alamos National Laboratory (1995).

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