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Transport and deposition of plutonium-contaminated sediments by fluvial processes, Los Alamos Canyon, New Mexico

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ABSTRACT

Between 1945 and 1952 the development of nuclear weapons at Los Alamos National Laboratory, New Mexico, resulted in the disposal of plutonium into the alluvium of nearby Acid and (to a lesser degree) DP Canyons. Previous research has identified some of this material in the Rio Grande and defined its distribution in the regional river system. The purpose of this paper is to explore the connection between the disposal sites and the main river, a 20 km link formed by the fluvial system of Acid, Pueblo, DP, and Los Alamos Canyons. Empirical data from 15 yr of annual sediment sampling throughout the canyon system has produced 458 observations of plutonium concentration in fluvial sediments. These data show that, overall, mean plutonium concentrations in fluvial sediment decline from 10 000 fCi/g near the disposal area to 100 fCi/g at the confluence of the canyon system and the Rio Grande. In finer detail, the concentrations fluctuate with downstream distance depending on the trap efficiency of various reaches, as controlled by hydraulic conditions. Temporal data from sites repeatedly sampled show the passage of waves of contaminated sediment through the canyon system. Field mapping identified 108 deposits of sediment, including active bed load, flood plains, bars, channel fills, and slack-water deposits. Volumes of sediment in these deposits (calculated from field measurements of the dimensions of the features), combined with the mean concentration values, produced a first approximation of the amount of plutonium in each deposit. The geographic distribution of deposits and plutonium is clustered: of the 1000 mCi of plutonium in the canyon system, 78% is in lower Pueblo Canyon, 18% is in lower Los Alamos Canyon, and the remainder is in the upper reaches of the system. Simulations using a computer model for water, sediment, and plutonium routing in the canyon system show that discharges as large as the 25 yr event would fail to develop enough transport capacity to completely remove the contaminated sediments from Pueblo Canyon. Lesser flows would move some materials to the Rio Grande by remobilization of stored sediments. The simulations also show that the deposits and their contaminants have a predictable geography because they occur where stream power is low, hydraulic resistance is high, and the geologic and/or geomorphic conditions provide enough space for storage.

INTRODUCTION

During World War II, the United States and its allies undertook a massive industrial and military effort to construct the first nuclear weapons. Referred to as the Manhattan Project, the effort included the construction of industrial installations, laboratories, and bomb manufacturing sites in several locations across the nation, with the final machining and assembly point at Los Alamos, New Mexico (Gosling, 1994). The accelerated construction of nuclear weapons successfully produced large numbers of weapons for the United States arsenal, but it led to a host of unforeseen issues related to risk of environmental contamination. During World War II and the first few years of the Cold War, managers suspected the potential hazards, but the perceived emergencies, the need for rapid production, and a lack of environmental knowledge fostered little attention to the issue of the risk of contamination (Groves, 1962). Each location in the far-flung industrial network developed its own set of contamination issues, but at Los Alamos National Laboratory the primary mechanism by which plutonium moved away from the immediate area of the installation was through mobility of contaminated fluvial sediments.

Shortly after the war, general concern arose about this route of environmental contamination: "The ultimate disposal of contaminated waste—sub-surface, surface and air-borne—needs much more thorough study. Even the simplest of such data—recorded periodic measurements of stream pollution below plants—are almost wholly lacking. . . . The disposal of contaminated waste in present quantities and by present methods, if continued for decades, presents the gravest of problems" (U.S. Atomic Energy Commission, 1948, p. 64). In the four and a half decades since that statement, workers at Los Alamos National Laboratory have collected data on stream pollution (summarized for sediments by Purtymun et al., 1987), but there has heretofore been little use of those data in a search for explanation and prediction based on the principles of earth science. This situation contrasts sharply with the extensive research in other topical areas of radiation safety such as human health and the fate of contaminants in biotic systems (Hacker, 1987).

Because soils and sediments contain strongly adsorbed plutonium in the vicinity of Los Alamos and because that plutonium constitutes almost all of the plutonium in the environment (Dahlman et al., 1980), geomorphic techniques that outline the mobility of the sediments are keys to understanding the fate of the element. Once adsorbed onto sedimentary particles, the plutonium is relatively immobile from a chemical standpoint until it is transported into an environment favorable for phase changes. Physical processes in this issue are therefore at least as important as the chemical ones. Other commentators have reached the same conclusion: "Studies of geomorphology . . . are basic to a clear understanding of the geochemistry of landscapes" (Fortescue, 1980, p. 249). "Water flow . . . can and does act as an important agent of radionuclide concentration and redistribution. . . . Most radionuclides quickly adsorb to sediment, plankton, and algae. . . . The movement of these organic and inorganic materials determines the fate of the radioactivity" (Whicker and Schultz, 1982, volume 1, p. 115). In the

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study of plutonium, "physical transport mechanisms become significant when compared with chemical transport mechanisms" (Coughtrey et al., 1984, p. 12). Hence, the present research emphasizes geomorphic processes in its investigation of the movement and storage of plutonium in the Los Alamos Canyon system.

RESEARCH QUESTIONS

The general issue of determining and explaining the amounts and location of plutonium in the physical environment of the Los Alamos Canyon system leads to four specific research questions addressed in this paper: (1) What are the present distribution and volume of sedimentary deposits in Los Alamos Canyon? (2) What is the present distribution of plutonium in the system? (3) Why are the sediments distributed in their present configuration? (4) Why are the components of the plutonium inventory in their present arrangements?

Previous workers have estimated sediment and plutonium inventories in the Los Alamos Canyon system, but this study is the first to map and measure specific individual deposits. This study represents a first approximation of a geographically accurate plutonium inventory, with a geomorphologic explanation. Whereas some investigations have used contaminants as tracers to better understand geomorphic processes, this study adopts the opposite approach in which principles of geomorphology explain the fate of the contaminants. The significance of this work is that if the reasons for the observed distributions can be outlined and explained, prediction of the near-term future of the hazardous materials will be enhanced, and cleanup efforts will be more likely to be successful. This paper complements but does not duplicate previous research into the geomorphology of plutonium in the Rio Grande (Graf, 1994) through investigating the connection between the source of pollution and the main river by way of Los Alamos Canyon.

THE GENERAL ISSUES IN LOS ALAMOS CANYON

During the weapons-construction process at Los Alamos, cleaning and machining of plutonium metal produced hazardous waste material in the form of plutonium dissolved in acid. Between 1945 and 1952, a ceramic sewer system conducted this liquid waste from buildings on the Pajarito Plateau, the site of the installation, to the edge of Acid Canyon (Kingsley, 1947), where it poured untreated from the sewer pipes down the canyon side and into alluvium on the canyon floor (Figs. 1 and 2). After 1952, the establishment of a waste-treatment plant eliminated the hazardous releases into Acid Canyon (Stoker et al., 1981, p. 10–11). During the 1960s, workers removed contaminated soil and building materials from the release point on the plateau, as well as rock from the canyon side and an unknown amount of alluvium from the canyon floor. These contaminated substances were stored in isolated burial sites. Acid Canyon, the primary source of plutonium contamination, is a small tributary to Pueblo Canyon, and DP Canyon (a small tributary of upper Los Alamos Canyon) also carried lesser amounts of plutonium from waste materials.

When the liquid plutonium waste reached the alluvium on the canyon floor, the relatively high pH conditions (8.0 or above) caused the plutonium to precipitate onto sedimentary particles in the form of plutonium oxide (for a general review of the radioecology and geochemistry of plutonium, see Coughtrey et al., 1984). Plutonium in this form is relatively insoluble, and it remains firmly adsorbed onto particles. As a result, >99% of the plutonium in the Los Alamos environment is associated with sediment (Schultz, 1977). Some downward movement of the material occurred in the surface sediment, but most of the plutonium remained near the surface where fluvial processes entrained it. Unconsolidated material from the waste-disposal site moved under natural stream processes out of Acid Canyon, down Pueblo Canyon, and through lower Los Alamos Canyon to the Rio Grande. In the Rio Grande, the waste plutonium joined plutonium from atmospheric fallout and became part of the channel and flood-plain

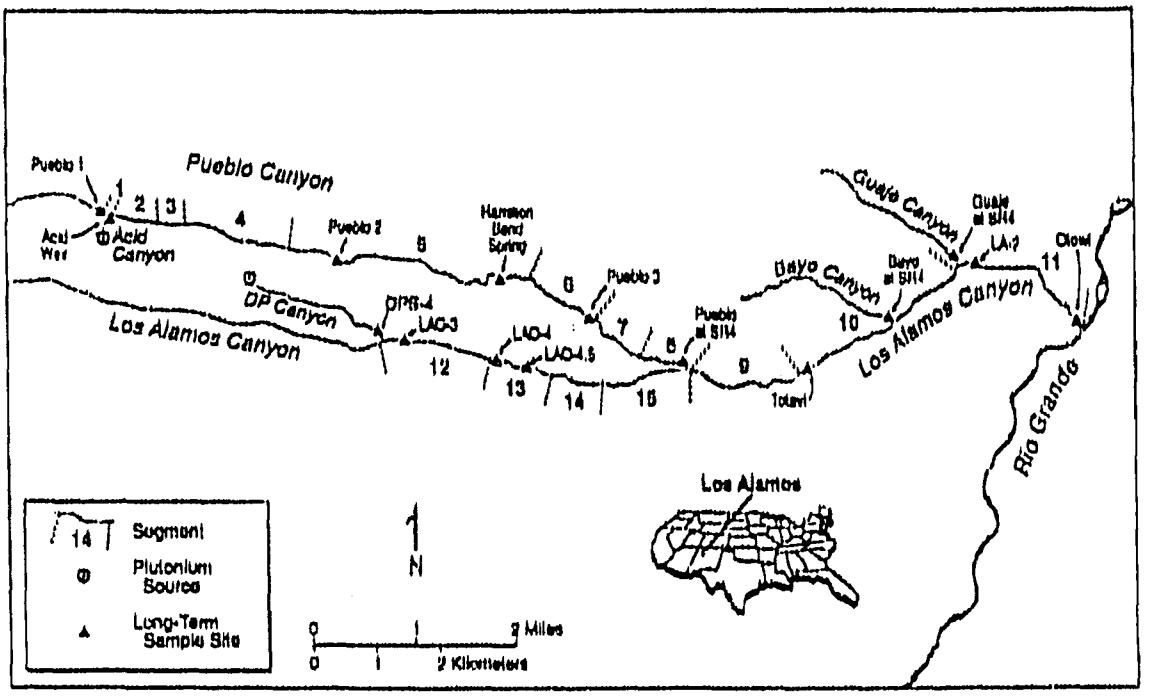

Figure 1. Map showing the Los Alamos Canyon system.

Figure 2. View of Pueblo Canyon looking northwest from the south wall of the canyon near the east gate of Los Alamos National Laboratory, with the Jemez Mountains in the background. The Bandelier Tuff forms the vertical walls. (W. L. Graf photograph 121-5, August 1994)

sediments of the regional river system. Previous investigations have shown that in the Northern Rio Grande system in the vicinity of Los Alamos, ≈10% of the plutonium is directly from Los Alamos Canyon, but in some sediments dominated by contributions from Los Alamos, it accounts for >80% (Graf, 1994). Along the main river, the concentrations of plutonium are not hazardous, given our present understanding of health physics, but the element is appearing in increasing concentrations in the sediments of Cochiti Reservoir ≈40 km downstream from Los Alamos.

The work reported on the following pages has its scientific roots in several previous investigations of the issue of plutonium contam[illegible] and movement. During the Cold War, some documents outlining [illegible]e of the plutonium were classified; however, researchers at Los Alamos National Laboratory studied many of the human health aspects of the element, and there were many examinations of the general behavior of plutonium in the natural environment. Beginning in the late 1970s, the laboratory published annual summaries of data reporting the concentrations of plutonium found in river waters and sediment in the vicinity of Los Alamos, though there were no extensive research projects to interpret this monitoring information. By the 1980s, relaxation of security restrictions and new interest in environmental quality stimulated an extensive unclassified review of the release of plutonium into the Los Alamos system (Stoker et al., 1981), and data summaries became generally available (Purtymun et al., 1987, 1990).

The obvious importance of stream processes in entraining, moving, and storing contaminated sediment (e.g., Carpenter, 1924; Wolfenden and Lewin, 1977; Imhoff et al., 1980; Graf, 1990) led to research into the hydrology of the canyon system and reassessments of the mobility of the contaminated materials (Lane et al., 1985; McLin, 1992). The dynamics of plutonium in the larger fluvial system of the Northern Rio Grande were the subject of an extensive investigation that provided a regional context for processes near Los Alamos (Graf, 1994). Taken together, these studies showed that although stream processes have transported some plutonium many kilometres from the original release point in Acid Canyon, most of the contamination remains in the Los Alamos Canyon system, where concentrations and amounts are as much as several orders of magnitude higher than in other locations. Although the inquiries produced improved understanding of plutonium in the region (summarized by Purtymun et al., 1987, 1990), the research reported here is an attempt to fill significant gaps in the understanding about how much plutonium is in the canyon system, where it is located, and why it has its present distribution.

GEOLOGIC AND HYDROLOGIC SETTING

Study Area

Los Alamos is situated on the surface of the Pajarito Plateau, the dissected flank of the immense caldera forming the Jemez Mountains of northwestern New Mexico (Smith et al., 1970). The canyons that score the surface of the plateau drain eastward from the high mountain surfaces to the Rio Grande, the master regional stream. The canyons, many >20 km long, have nearly vertical sides that connect them to the gently sloping plateau surfaces above. The canyon floors are variable in width and gradient. Some steep reaches are choked with large boulders from rock falls or debris flows; other reaches with relatively shallow gradients accommodate broad deposition zones for fine sediments. The Los Alamos Canyon system has two major upstream branches, upper Los Alamos Canyon and Pueblo Canyon, which join to form lower Los Alamos Canyon.

The Los Alamos Canyon system owes its morphology partly to the geologic materials into which it is excavated. Because of its position on the flank of the Valles and Toledo calderas of the Jemez Mountains, the area is underlain by volcanic and sedimentary rocks of Tertiary and later ages (Griggs and Hem, 1964; Smith et al., 1970). The principal formations include the Santa Fe, Polvadera, and Tewa Groups. The most important geologic unit for the present study is the early Pleistocene Bandelier Tuff, an 80–100-m-thick cap of eruption products on the Pajarito Plateau. It appears on the plateau surface and canyon walls as massive pumiceous tuff breccia and nonwelded tuff derived from ash flows and ash falls. These materials are variable in porosity and permeability, depending in part on their degree of welding, and they yield weathering products, soils, and stream sediments that are dominated by sand-sized particles (Nyhan et al., 1978).

Stream sediments issuing from this terrain typically have <5% of their weight in particle sizes smaller than sand. Channel alluvium deposits in the source areas of the plutonium pollution have ≈0.5% of their bulk by weight

in the silt and clay fraction. Data from 38 samples drawn from the surface of channel beds throughout the Los Alamos, Pueblo, Acid, and DP Canyon systems contain 3.8% silt and clay (Stoker et al., 1981, p. 126, 130, and 133; Lane et al., 1985, p. 35; Graf, 1994, p. 237). Major tributary canyons to lower Los Alamos Canyon have similar coarse sediments in their stream channels. Bayo Canyon sediments contain 2.3% silt and clay, and those in Guaje Canyon contain 7.5% fines (Lane et al., 1985). Fills occupying abandoned channels have ≈25% fines, whereas flood-plain sediments contain 55% (Graf, 1994, p. 237).

The energy driving the fluvial system of Los Alamos Canyon comes from the precipitation climatology of the region. Total precipitation ranges from >50 cm/yr in the nearby Jemez Mountains to ≈45 cm at Los Alamos and <30 cm in the Rio Grande Valley (Bowen, 1990, p. 5). The maximum monthly rainfall occurs in August as the result of summer thunderstorm activity, whereas highly variable snowfall has a December maximum. The streams in the Los Alamos Canyon system are intermittent, except for the reach immediately downstream from a wastewater-treatment plant. Fluvial transport of sediment is directly related to spring snowmelt and summer flash floods (White and Wells, 1978). Under these conditions, from a 152 km^2 (58.7 mi^2) drainage area, the 100 yr design storm would produce at the mouth of Los Alamos Canyon a peak flow of 52 m^3/s (1845 ft^3/s) and a runoff volume of 1.2×10^6 m^3 (1010 acre-ft) (McLin, 1992, p. 24).

Vegetation also influences the fluvial processes in the study area. The mountain slopes that supply much of the runoff for the canyon system are covered by forests of ponderosa pine and Douglas fir, which protect snow packs from rapid spring melt (White and Wells, 1978). A pinyon pine and one-seed juniper community dominates the vegetation of the Pajarito Plateau (U.S. Department of Energy, 1979, p. 3/38–3/41; Brown et al., 1980). The canyon floors have ponderosa forests that cover some flood plains, especially in upper Los Alamos, upper Pueblo, Acid, and DP Canyons. Riparian vegetation throughout the canyon system includes grass covers on flood plains and willow, tamarisk, and cottonwood in the lowest reaches of the system, providing significant hydraulic roughness on flood-plain surfaces.

Functional Reaches of the Canyon System

The Los Alamos Canyon system is not a simple set of conduits for water and sediment with slowly changing conditions in the downstream direction. Rather, it is fragmented into several specific and definable segments with internally consistent processes and forms that are different from those of neighboring segments. Geomorphic properties (such as channel pattern and dimensions) provide a convenient basis for defining a series of functional segments in the system (Table 1). There are several particular features in the system that affect the transport and storage of sediment and its plutonium. For example, in middle Pueblo Canyon, segment 3 is a boulder-choked reach where only limited storage of sediment and plutonium is likely because there is no space for such storage on the canyon floor. A short distance downstream, however, are segments 5 and 6, a part of Pueblo Canyon with a wide floor and shallow gradient that defines a geomorphic condition conducive to deposition. The channel in segments 5 and 6 is small, has extensive flood plains, and is associated with sedimentary deposits that bury recent artifacts (Figs. 3, 4, and 5). This depositional zone ends in the downstream direction at a knickpoint several metres high that is slowly migrating headward. Repeated field observations since 1987 and historic aerial photography suggest a rate of recession of a few metres per year on average, though most erosion occurs episodically during flood events that occur only once every 5 yr. Because of its association with Acid Canyon and its extensive sediment storage areas, Pueblo Canyon is the most important geographic component of the plutonium inventory.

Pueblo Canyon is floored by Holocene alluvial deposits in many sections, but such deposits are small, scattered, and limited in number in upper Los Alamos Canyon (Griggs and Hem, 1964, Plate 1). The channel in upper Los Alamos Canyon is confined by colluvium and is entrenched in some areas. Only in the lowest reaches of the canyon does it appear as an unconfined stream, but even there it lacks extensive flood plains or overbank areas underlain by sediments active in the present regime of the stream. Sediment and plutonium storage is therefore much more limited in upper Los Alamos Canyon than elsewhere in the system.

On a larger scale in lower Los Alamos Canyon, a narrow gorge with a relatively steep gradient in segment 9 restricts the possibility for storage of sediment and plutonium. The gorge, eroded into Quaternary basalt of the Sante Fe Group, ends as the stream issues into a terrain dominated by more easily eroded sedimentary rocks, and the broadening channel has a shallower gradient with more horizontal space for deposits. The reaches formed by the lowest 4 km of Los Alamos Canyon are radically different from the reaches above. Bayo Canyon joins the system ≈4 km above Otowi (the termination of Los Alamos Canyon at the Rio Grande), and Guaje Canyon joins the system ≈2 km above Otowi. These tributaries contribute some water, and they appear to add at least as much sediment as is being brought down Los Alamos Canyon, as indicated by active deposits in and

TABLE 1. REACHES OF PUEBLO AND LOS ALAMOS CANYONS

Segment number	Reach name and description	Downstream distance to lower end (m)	Mean gradient (m/m)	Mean active channel width (m)	Mean active channel depth (m)	Mean channel roughness (Manning's n)	Bed sediment silt/clay by weight (%)	Total plutonium (mean) (fCi/g)	Total plutonium (standard deviation) (fCi/g)
Source	Acid Canyon							10151	3040
1	Upper Los Alamos Canyon	61	0.0197	1.8	1.1	0.040	0.8	10	16
2	Upper middle Pueblo Canyon	659	0.0336	1.8	1.1	0.050	2.6	N.D.	N.D.
3	Middle middle Pueblo Canyon	1280	0.0325	1.2	1.6	0.080	2.6	N.D.	N.D.
4	Lower middle Pueblo Canyon	3768	0.0203	1.8	1.2	0.035	3.0	N.D.	N.D.
5	Upper lower Pueblo Canyon	8338	0.0109	3.7	0.5	0.050	3.0	1048	1562
6	Middle lower Pueblo Canyon	9506	0.0151	6.1	0.5	0.025	11.0	278	747
7	Lower lower Pueblo Canyon	10214	0.0200	13.2	1.6	0.025	3.7	N.D.	N.D.
8	Junction segment of Pueblo Canyon	11750	0.0180	16.3	2.3	0.025	3.5	516	270
9	Narrows segment of Los Alamos Canyon	13007	0.0288	4.1	2.4	0.060	3.5	188	195
10	Totavi segment of Los Alamos Canyon	17043	0.0115	29.3	0.9	0.035	3.6	197	190
11	Otowi segment of Los Alamos Canyon	19702	0.0090	97.0	1.1	0.030	4.2	112	78.1

Notes: N.D.—not determined. See Figure 1 for map with corresponding segment numbers. Mean gradient is the overall decrease in elevation divided by the along-channel distance for each segment. Mean channel width and depth are from samples of 10 such measurements of the active channel for each reach. Hydraulic roughness is a composite of 10 field estimates for the active channel in each reach. Sediment data are from Stoker et al., 1981, p. 126, 130, 133; Lane et al., 1985, p. 35; and Graf, 1994, p. 237. Plutonium data are concentrations for active bed-load sediments from annual monitoring and surveillance reports published by Los Alamos National Laboratory.

Figure 3. View of exposed cross section of flood-plain sediments in middle Pueblo Canyon showing the stratification of materials. The depositional history of the site is partly revealed by the trees and their root collars. The collapsed tree on the right side of the view has its root collar buried beneath 1.2 m of sediment near the level of the present channel in the foreground. The ponderosa trunk in the center of the view still stands in a similar but less exposed arrangement. Several younger, much smaller trees have rooted on the surface of the flood plain at the left. Ring counts of the trees and packaging litter suggest that the exposed sediments are post-1930. (W. L. Graf photograph 121-16, August 1994)

Figure 4. View of the channel in upper middle Pueblo Canyon showing a grass-covered point bar and flood-plain deposit to the right of the active channel. Note driftwood stranded during a recent flood resting on the boundary between the flood plain and the colluvial slope at the right. (W. L. Graf photograph 107-14, August 1993)

near their channels. The lowest reaches of lower Los Alamos Canyon, with their wide, braided channels and extensive flood plains, are relatively far from plutonium sources and are not as important in the plutonium inventory as Pueblo Canyon.

METHODS

Empirical Analysis

Questions concerning the distribution of sediments and plutonium are empirical problems, requiring a methodology that rests on directly observed evidence. The approach used in this work was to measure the volume of sediment in the various segments and then to calculate the amount of plutonium by multiplying the volumes by contaminant concentrations. For the distribution of the sediment, the scale of the canyon system is small enough to permit direct field mapping of the individual sedimentary deposits along the entire length of Acid and Pueblo Canyons (12 km), DP and upper Los Alamos Canyons (6 km), and lower Los Alamos Canyon (8 km). Field mapping on the ground throughout the entire canyon system provided basic information on the location, width, length, and depth of each deposit. Field measurements with tape and optical ranging devices provided distance and horizontal dimensions, whereas tape measurements of bank exposures and penetration with a soil probe or steel reinforcing bar driven to bedrock or large boulders revealed the depths of deposits. Active bed load on the channel floor, flood plains, point bars, mid-channel bars, attachment bars, and slack-water flood deposits provided direct locations and dimensions for each individual deposit.

Flood plains, defined as depositional areas for fine sediments outside the

Figure 5. Eroding flood plain in middle Pueblo Canyon, showing the remobilization of previously stored sediments deposited during the 1970s and 1980s as indicated by buried packaging litter (W. L. Graf photograph 108-30, August 1993).

banks of the channel, but actively receiving sediment during high flows in the present regime of the stream, occur as discontinuous features along the channel. They often occupy parts of the canyon floor that are wider than nearby areas. Because of their limited lengths along the channel and their discontinuous nature, their forms are best described as pocket flood plains, which are small depositional features, next to the active channel, that fill semicircular scallops cut into the otherwise inactive colluvial materials along the bank.

For some reaches in the upper canyon system, deposits were <5 m^2 in surface area and were thus too small to map individually. For these reaches, 10 sample cross sections provided average values of bed-load and overbank deposits per unit length of the channel, usually 0.25–1.00 m^3 of bed-load storage per metre of channel length and 0.5–2.0 m^3 of overbank material per metre of channel length. Mapping from aerial photography provided horizontal dimensions of the relatively large deposits of the lowest 4 km of Los Alamos Canyon. Aerial photography used in this study was high-quality, color, vertical imagery at a scale of 1:7200, taken in September 1991. The imagery scale was large enough to permit identification of individual trees and boulders during the field investigation. The base map for field mapping was a 1:4800 scale map with a 0.6 m contour interval; map data was managed with an ArcInfo geographic information system. In total, all the methods produced data for 108 deposits.

The field evidence, maps, and aerial photography provided measures for the length, width, and thickness of each deposit, which, when multiplied together, produced the volume of each deposit. The location of each deposit was designated as the downstream distance of the center of the deposit measured from an arbitrary starting point. The reliability of the field data is high for 1993, the period of field mapping, but subsequent hydrology events will alter some measures. Horizontal measurements and thickness measurements made in the field are likely to be accurate to within ≈1%, and distance measurements are accurate within ≈2%; some measures of reach lengths made by digitizer from the base map are likely to be accurate to within 5%.

The empirical data for plutonium concentrations used in this study have been previously published by Los Alamos National Laboratory in a series of annually released environmental surveillance reports. Data are available from 458 samples taken from a set of sample sites throughout the Los Alamos Canyon system that laboratory personnel have sampled on an annual basis for 15 yr or more. The unit of measure used in the Los Alamos radiochemistry data for concentration of plutonium in sediments is the curie (Ci), equal to the number of disintegrations (≈37×10^9) per second measured from 1 g of pure radium. Concentrations commonly encountered in sediment samples are much smaller than 1 Ci; they are usually measured in picocuries (pCi, or 10^{-12} Ci) or femtocuries (fCi, or 10^{-15} Ci). The correlative unit in the International System is the becquerel (Bq), equal to one disintegration per second.

Sediment samples from active stream sediments are from deposition zones downstream from boulders or cobbles. Samples from dry stream beds are composite accumulations created by scooping a trench ≈2 cm deep across the entire channel and mixing the resulting extract. On near-channel surfaces, the composite samples were created from sites that yielded five plugs 75 mm in diameter and 50 mm deep collected from the center and four corners of a 10 m square area. Variation of plutonium concentrations with depth remains a significant unknown because of lack of sampling from buried strata. Gamma-ray spectrometry on an HPGe detector with a shielded chamber provided concentration measurements with errors expressed as one standard deviation about the mean radiation count, usually 0.2 fCi/g for ^{238}Pu and 0.2–10 fCi/g for ^{239}Pu and ^{240}Pu (≈5%). The largest errors are usually associated with the highest concentrations (Environmental Protection Group, 1994, p. VIII/9).

An approximation of the plutonium inventory in the Los Alamos Canyon system is possible by multiplying the volumes of sediment measured in the field by the plutonium concentrations determined by laboratory analysis of each deposit and summing the results. Basic data and interim calculations are in supplementary tables 1–3 and supplementary maps[1]. Concentrations of plutonium for near-channel deposits were from group averages as reported by Lane et al. (1985, p. 45), because analyses of individual deposits are not available. Although it would be preferable to have assays of each de-

[1]GSA Data Repository Item 9655, supplementary tables and maps, is available on request from Documents Secretary, GSA, P.O. Box 9140, Boulder, CO 80301, E-mail editing@geosociety.org.

posit identified in this study, expense prohibits such an approach. Samples taken once each year from a few fixed, repeatedly sampled sites may be more valuable for establishing long-term averages and for establishing trends if they exist. By combining the errors in the landform survey, sediment measurement process, and radiochemical analysis (Environmental Protection Group, 1994), it is likely that the maximum error in the final estimates of the plutonium inventory for the canyon system is ≈10%.

Model Simulation

Although the empirical data are valuable in establishing the present situation, they provide little insight into the functioning of the system as it changes through time. Explanations of the dynamics of the plutonium in the stream sediments of the system depend on the construction of a model that can simulate the movement of water, sediment, and plutonium through the system. Such a model need not be particularly complicated, but it must account for spatial and temporal variation of the processes. The riverine accounting and transport (RAT) model, designed in an earlier study to simulate water, sediment, and radionuclide routing on the Rio Grande, was adapted to the more limited Los Alamos Canyon system to provide explanations for the observed distributions of sediment and plutonium.

The RAT model, a computer program of ≈1400 lines of QBasic program language, replicates fluvial processes in a series of stream segments over a simulated period of several hours or days (see Graf, 1994, p. 203–210, 279–280, for a detailed description). The input for the model consists of initial boundary conditions describing the basic morphology of the channel system (such measures as the channel width, depth, gradient, and hydraulic roughness), plus information on flood events and injections of plutonium contamination from a point source. The model routes water, sediment, and radionuclides through the system, accounting for input, transport, internal storage, and output, calculating the mixing and downstream dispersion of contaminants, and estimating their concentrations in the resulting sedimentary deposits. In the Acid Canyon–Los Alamos Canyon case, there are 11 reaches with an average length of <2 km (mapped in Fig. 1).

The model is physically based, iterative, self-updating, and spatially variable. The basis of the model operations is repeated calculation of the hydrologic characteristics of flow using ordinary equations of continuity and Manning's equation combined with initial boundary conditions defining channel geometry and roughness. Calculations using Bagnold's (1966) sediment-transport functions describe the movement of sediment by using the flow depths produced by the hydrologic calculations. The Bagnold approach is useful because it closely approximates sediment transport in dryland streams (Graf, 1988, p. 149). Simple accounting functions keep track of the amounts of sediment and radionuclides in various parts of the stream system to determine contaminant concentrations in deposits. The model is iterative because it calculates the movement of sediment and radionuclides through the system for simulated periods (usually 1 hr), updates the initial boundary conditions to account for erosion and sedimentation, and then begins the calculations again for the next period. The RAT model is spatially variable because its boundary conditions are segments of the stream system, with each segment defined by its own geometry and roughness. The model calculates changes within each segment, and so erosion may occur in one place while deposition is occurring elsewhere. In the operation of the model, contaminated sediment enters from a point source (the confluence of Acid Canyon with Pueblo Canyon, for example), but in all subsequent iterations, contaminated sediments that had been stored temporarily along the stream might be remobilized through local erosion, converting the stream to a multiple-source system. The model is not particularly sophisticated, but it represents a first approximation to the results of fluvial processes on a segment-by-segment basis.

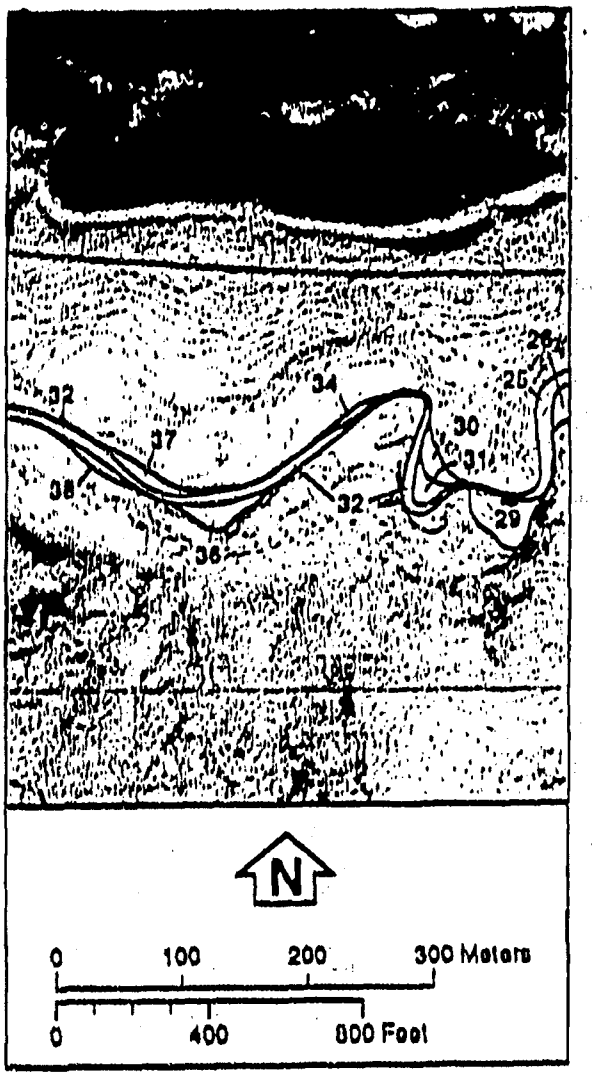

Figure 6. Example map showing the distribution and dimensions of deposits defined by field measurements in a section of Pueblo Canyon.

Experiments with the RAT model in this study used the 25 yr design storm, which creates a flow in Pueblo Canyon through a 7 hr period with a maximum discharge of ≈6 m^3/s (McLin, 1992, p. 10). The 25 yr storm is useful for simulation because it is large enough to cause considerable sediment transport, and so it is informative about system changes. In most places it exceeds bankfull capacity. The event is similar in magnitude to several actual events recorded in or estimated for the canyon system (Lane et al., 1985). The reliability of the model is difficult to assess, but it is probably more useful for establishing trends and changes over space and time rather than for making highly accurate predictions of volumes of sediment or plutonium storage. The model predicts plutonium concentrations that are within ≈20% of the measured values at sites sampled by the laboratory.

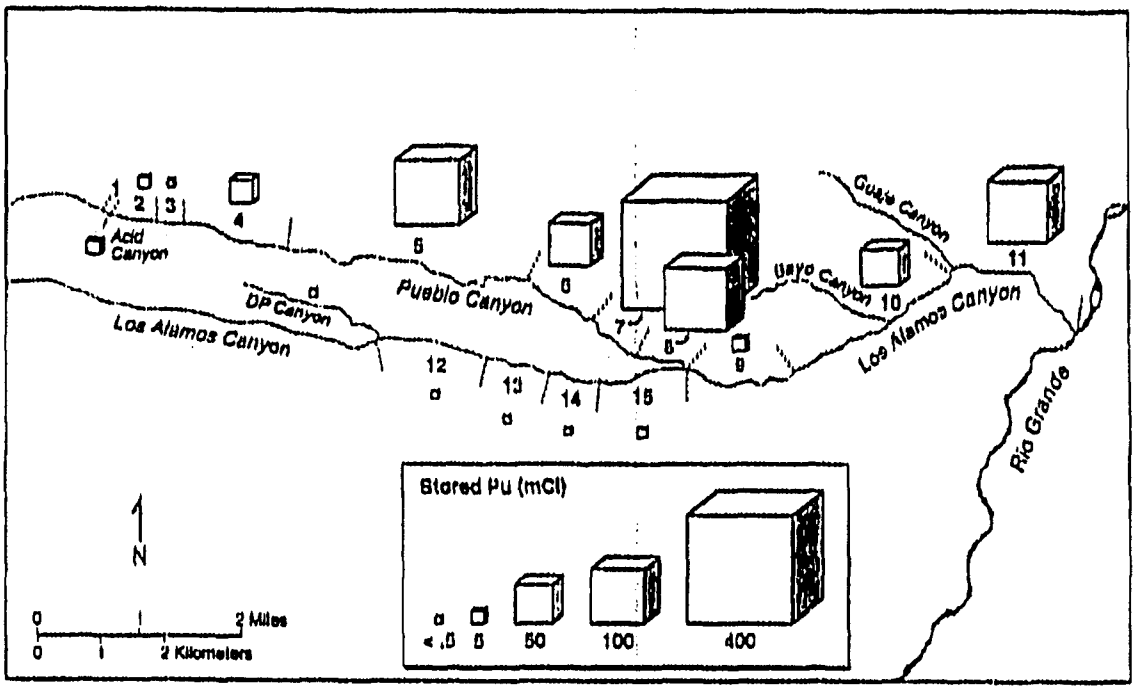

Figure 7. Map showing the general distribution of the stored plutonium in sediments of the Los Alamos Canyon system. Proportional cubes represent the amount of plutonium stored within each segment. The total amount of plutonium in the system is ≈1000 mCi.

RESULTS

Empirical Results

Field identification of those deposits likely to have been active since the 1945 introduction of plutonium to the Los Alamos environment is not especially difficult, because there are radical differences between recently active and inactive landforms, flood-plain or bar sediments, and channel or near-channel surfaces. Those geomorphic features that have been active in the past half-century have no ponderosa pine >50 yr old, though they may have riparian species such as willow, tamarisk, or grasses and sedges of younger age. The recent features have deposits that are obviously fluvial in origin, with laminar stratification in all but a few point-bar deposits. Recent sandy deposits bury the root collars of mature trees and artifacts such as food wrappers and construction materials of recent origin (Fig. 3). Those features that have not been active in the past half century have mature, stable forest vegetation of mature ponderosa pine (or pinyon and/or juniper in lower Los Alamos Canyon). The inactive features in the canyon system are mostly fluvial terraces, debris flows, and colluvial deposits with mass-movement characteristics.

The most common active features that store sediment and plutonium along the canyon are bed load on the channel floor, flood plains, attachment bars, points bars, mid-channel bars, and slack-water flood deposits. A common type of sediment and plutonium storage feature in Pueblo and upper Los Alamos Canyons is the pocket flood plain. These small flood plains, often <40 m wide, 100 m long, and 1 m thick have distinctive riparian vegetation covers that are different from nearby inactive forms with their upland vegetation. Pueblo Canyon has 63 mapped deposits of various dimensions, upper Los Alamos Canyon has 32, and lower Los Alamos Canyon has 15. Figure 6 shows an example of the distribution along a representative reach of Pueblo Canyon.

Figures 7 and 8 show the general distribution of plutonium in the Los Alamos Canyon system based on mapping of the sedimentary deposits and long-term surveillance data for concentrations. The lower reaches of Pueblo Canyon, where extensive deposits of relatively high concentrations of the element occur, are the most important locations for stored plutonium. Much lesser amounts of plutonium and sediment appear in the upper reaches of Pueblo Canyon and upper Los Alamos Canyon. Sediment volumes are large in the lowest reaches of lower Los Alamos Canyon, but the concentrations of plutonium in these sediments are low (but still a order of magnitude higher than regional background levels) because of dilution by sediment issuing from Bayo and Guaje Canyons, which drain uncontaminated areas.

The distribution of plutonium in Pueblo Canyon, the most important part of the entire system for plutonium storage, shows the detailed geography of the storage, a complexity that is otherwise hidden if only mean values for the entire system are reported. Figure 9 shows the distribution of plutonium in individual deposits scattered through the entire length of Pueblo Canyon (Table 1 describes the segments). Because of geomorphic conditions that control the amount of available space for deposition on the canyon floor, some segments have no substantial storage, whereas other reaches have numerous deposits. The canyon floor immediately downstream from the confluence with Acid Canyon, the contamination source, stores some plutonium because of extensive flood-plain areas where the canyon floor widens, and because of high concentrations of plutonium that occur near the source. Two deposits near the boundary between segments 6 and 7 (shown in Fig. 9) are prominent in the plutonium distribution. Both of these deposits are large attachment bars with flood deposits along the channel sides; each bar is large (with surface area of >5000 m^2 and depths of ≈3 m), containing large volumes of sediment, and each occurs in the part of the canyon with relatively high concentrations of plutonium. The large volumes combined with high concentrations produce large amounts of plutonium in storage.

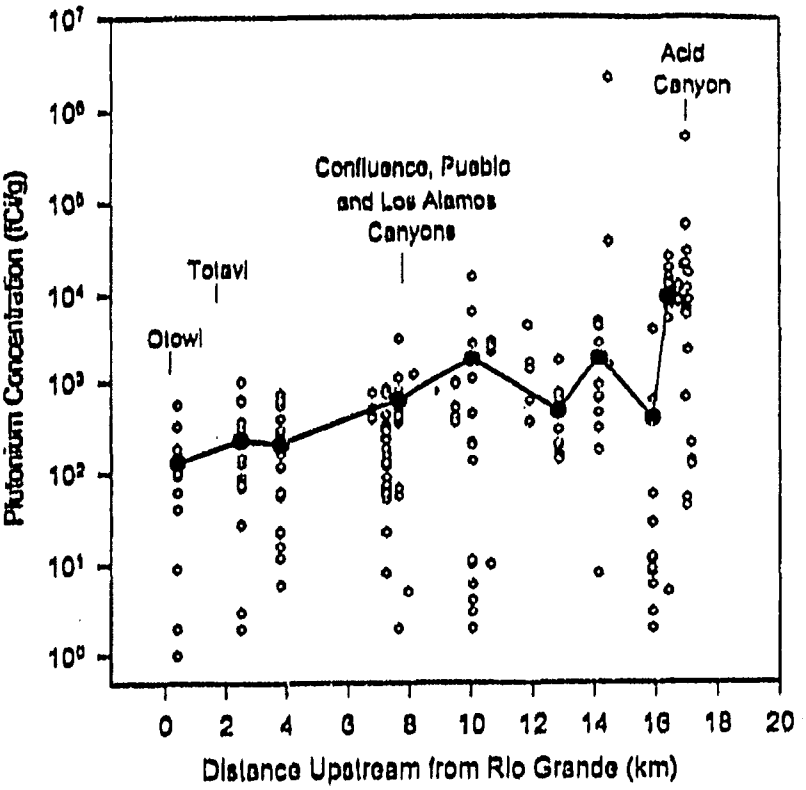

Figure 8. The relationship between downstream distance and plutonium concentration for bed-load sediments throughout the Pueblo and Los Alamos Canyon system based on 230 samples, generally taken from 1977 to 1992 and published annually in the surveillance reports of Los Alamos National Laboratory. The open diamonds represent individual sample values. The filled circles connected by the line represent mean values at various locations, calculated by using all the data available at that location except the highest and lowest values to eliminate unusual values with inordinate impacts on the mean.

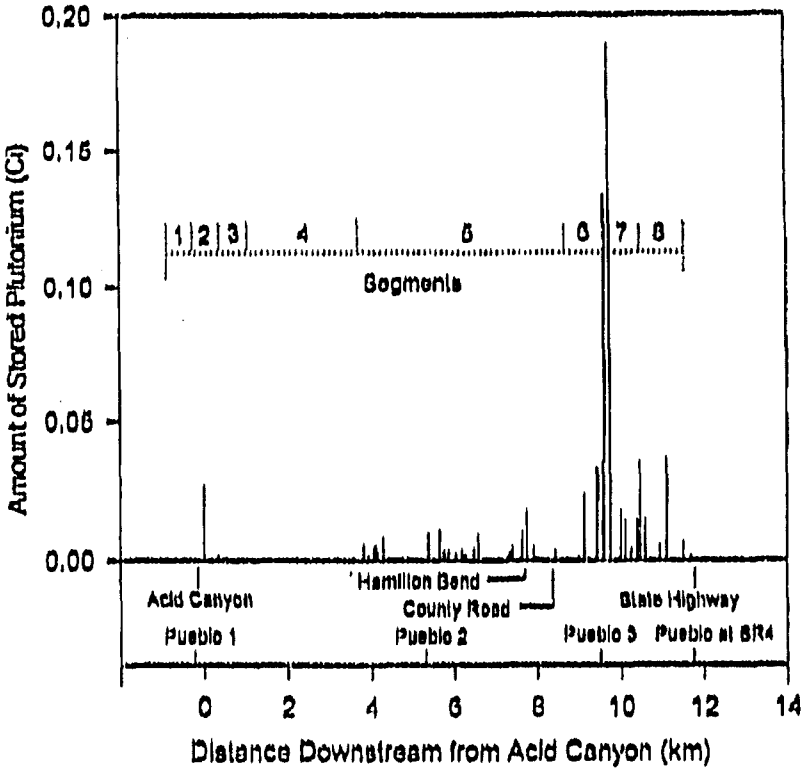

Figure 9. The spatial distribution of plutonium in deposits along Pueblo Canyon, with the amount of stored plutonium represented by the height of each bar. The numbered segments correspond to those shown in Figure 1. Los Alamos National Laboratory long-term sample sites shown across the bottom of the graph. Based on field data combined with published laboratory data.

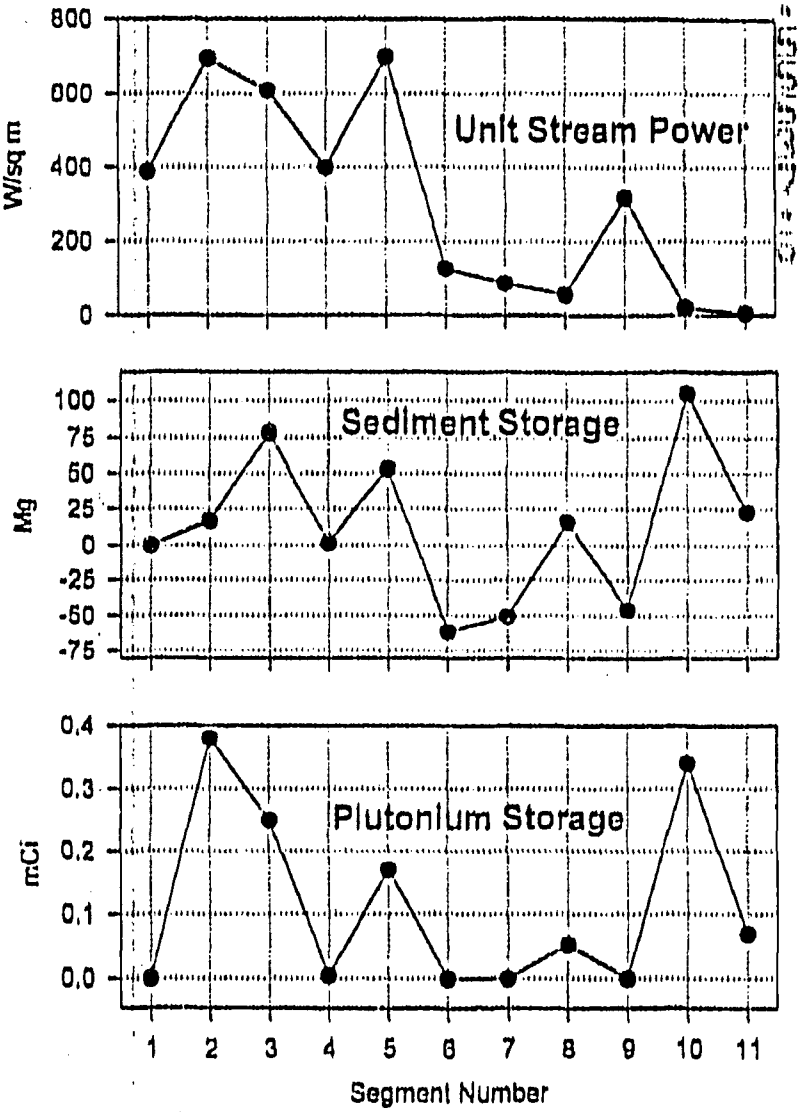

Figure 13. The results of simulations using the RAT model to predict the concentrations of plutonium in sediments in various segments of the Los Alamos Canyon system. Downstream changes in stream power, sediment storage, and plutonium storage are shown for data at one simulated hour at a peak discharge of 6 m^3/s, similar to the 25 yr flood from Pueblo Canyon with no additional flow from upper Los Alamos Canyon. The simulation assumed a release of 100 Mg of sediment from Acid Canyon with 10 000 fCi/g of plutonium. The data shown are for the channel only, so they do not reflect overbank storage that occurs in segments 6 and 7. In the lowest reaches, sediment and plutonium storage vary inversely with stream power, whereas in other reaches, local conditions allow erosion even at relatively low stream power. No plutonium storage occurs in those segments with erosion. Contamination enters the system in segment 2. The segments are represented solely by their numbers (locations are shown in Fig. 1) and are not represented to scale in this graph.

ment 2 of upper Pueblo Canyon would have concentrations greater, rather than less, than 1000 fCi/g. There are no long-term data available for this particular segment, but the nearest related sample site, Pueblo 2, in fact has values in the expected range, with a long-term average of 1645 fCi/g.

This set of simulations shows why plutonium contamination remains in Pueblo Canyon and is much less farther downstream in lower Los Alamos Canyon. The masses of material released from Acid Canyon were too large for the transport capacity of the stream in Pueblo Canyon to conduct the contaminants completely through the system, so much was stored within Pueblo Canyon. Events smaller than the 25 yr event moved minor amounts of contaminated material downstream, where they elevated concentrations of plutonium in the lower reaches of the system before being diluted by relatively clean sediments from Bayo and Guaje canyons. This selective, limited removal continues in smaller events through knickpoint migration and lateral channel erosion directly observed in the field between 1987 and 1995.

Simulated Spatial Variation

Simulation modeling also illustrates the reasons behind the geographic distribution of plutonium in Acid, Pueblo, and Los Alamos Canyons. Figure 13 shows the distribution of plutonium concentrations in channel sediments downstream through the first nine segments of Pueblo and Los Alamos Canyons below the injection point at Acid Canyon. Segment 1 is an initiating segment of Pueblo Canyon immediately upstream from the Acid Canyon confluence, so that the injection of contaminated material is into segment 2. The assumptions underlying the simulations were for the 25 yr hydrologic event with an injection from Acid Canyon of a variety of masses of sediment with 10 000 fCi/g of plutonium (a likely degree of contamination given the empirical data from Acid Weir). Although there is an overall decline in concentrations in the downstream direction, there is considerable geographic variation within that decline.

Simulations showed that when releases of 50 or 75 Mg are considered, there is no storage in segments 2 and 4, because there is ample stream power to move them. With masses of 100 Mg or greater (as in Fig. 13), however, segments 2 and 4 store contaminated material. Segments 6, 7, and 9 do not store the channel sediments that are the subject of these simulations under any of the scenarios for geomorphic and hydraulic reasons. In segment 6, the channel is <2 m wide, so virtually all of the contaminated sediment spills out of the channel and is spread across a broad flood plain (consisting of alluvial fill and a sand splay). Segment 7 is a channel entrenched into an arroyo bounded on the upstream end by a prominent knickpoint. The channel is evacuating sediments from the segment; stream power is high and channel storage is low. Finally, segment 9 is the narrow canyon reach of lower Los Alamos Canyon immediately below the largest knickpoint in the entire system caused by a resistant ledge of basalt 75 m high. A steep gradient and narrow canyon floor preclude storage of contaminated sediments in appreciable quantities in the segment.

Those segments with high concentrations of plutonium in stored sediments have identifiable geomorphic and hydraulic explanations for such storage. Segment 3 has a high hydraulic roughness and traps small quantities of sediment among its boulders. Because segment 3 is relatively close to the source of contamination, the concentration of plutonium in this stored sediment is relatively high. In segment 5, the channel is broadly defined and braided in a wide section of the canyon; therefore, the segment stored large amounts of contaminated material with concentrations that are somewhat less than those found farther upstream. Pre-1945 uncontaminated sediments appear to have been remobilized and mixed with post-1945 contaminated sediments. The results reduced concentrations of plutonium in the contaminated sediments as they moved through the system. Segment 8 ends in a culvert under the state highway, and during and after flood events, channel storage of contaminated sediments results from backwater effects. Finally, storage in segment 10 results from a widening of the channel after it exits the steep, narrow segment 9. Simulations for segment 10 show that stream power declines rapidly because of divergence and decreased depth of flow, which lead to sediment deposition and storage. Downstream from segment 10, concentrations decrease as a result of mixing with sediments from Bayo and Guaje Canyons.

DISCUSSION AND CONCLUSIONS

In the absence of other considerations, it seems reasonable to expect that the concentrations of plutonium in sediments would decrease in the downstream direction away from the point source of contamination, because of mixing and dispersion. Such distance decay functions are common in many spatial distributions as a reflection of dilution and dispersion, and negative logarithmic or exponential functions are the typical mathematical expressions describing the decline in concentration or amount of a pollutant as distance from its source increases (Wertz, 1949; Glover, 1964; Lewin, 1978). For radionuclides, if all other aspects of the system are equal, "the distribution of transuranic elements from point sources at nuclear facilities typically produces decreasing concentrations with distance from the source" (Watters et al., 1983, p. 91).

In fluvial systems, however, all other things are not equal, as demonstrated in previous studies of the distributions of mercury and thorium (Graf, 1985, 1990). At least five primary attributes of the fluvial system work to complicate the generalization of smoothly defined decay with increasing distance. First, the hydraulic processes in open channel flow sort sedimentary materials, preferentially transporting the finer particles greater distances. Because metal contaminants, including plutonium, adsorb to greater concentrations on finer particles (Gibbs, 1973), the sorting that occurs in the fluvial transport process affects the final distribution of the element after deposition. Second, sediment does not move consistently and constantly through the channel system, but rather it moves as waves. This is especially true of dryland streams (such as those at Los Alamos), where almost all the sediment transport is during flood events of a few hours or days. Much more sediment is moved on the rising limb (Richards, 1982, p. 98) of the hydrograph than on the falling limb, creating additional variation in the rate of transport through any given reach. Because the sediment moves in waves or slugs, it is deposited unevenly as well, creating a discontinuous distribution of sediment and plutonium that does not precisely fit a smooth decay function with increasing distance. Third, once contaminated sediment enters a stream system from a point source, portions of the total come to rest part way through the system. When flood events remobilize these temporarily stored contaminants, the storage areas become additional sources scattered throughout the stream system, obfuscating the effect of a single point source.

Fourth, deposition of the sediment and associated plutonium is quite particular and occurs only in those places where stream power declines, for example, where the channel suddenly widens or, in an overbank situation, where a flood plain occurs (e.g., segments 9 and 10 in Fig. 13). The available space for these conditions on the canyon floor is a product of geomorphic factors that may not be related to the stream at all, but rather to changes in geologic substrate or to colluvial processes on the canyon sides. Fifth, tributary streams introduce uncontaminated sediments that mix with the contaminated materials at various irregular points along the system, causing abrupt declines in concentrations and equally abrupt changes in the total amount of sediment stored in the system. These factors conspire to produce the uneven downstream distribution of plutonium concentrations. Certainly, these concentrations show an overall decline between the source of contamination and the canyon outlet at the Rio Grande (Fig. 8), but in detail they reflect the imposition of local conditions on a general trend.

In summary, the empirical results of this investigation show that sediments are stored along the various reaches of the Los Alamos Canyon system in bed load, channel bars, slack-water deposits, and flood plains. An especially common storage feature is the pocket flood plain, an accumulation of stratified sediment in small reentrants in colluvium at the side of the channel. An approximation of the total plutonium inventory indicates that the canyon sediment system stores ≈1000 mCi. Highest concentrations of plutonium occur in the stored overbank deposits rather than the active channel materials. The arrangement of plutonium associated with the deposits takes on the discontinuous distribution of the stored sediment. Therefore, at the scale of a few kilometres or less, plutonium concentrations and amounts do not necessarily decline in the downstream direction. Most of the plutonium in the system is in lower Pueblo Canyon. When the plutonium moves through the channel system, it may do so in waves that are reflected in the long-term data from fixed, repetitively sampled sites, but perhaps not clearly indicated in the record of deposited sediments. When the plutonium reaches the Rio Grande, dilution of channel sediments by tributary contributions reduces the concentrations from ≈10 000 fCi/g to ≈100 fCi/g.

The simulations of water, sediment, and contaminant routing through the Acid, Pueblo, and Los Alamos Canyon system using the RAT model produced two important generalizations about the behavior the system. First, the masses of contaminated material released from Acid Canyon were apparently ≈100 Mg or more, and the sediments remained mostly within Pueblo Canyon because the stream has not developed enough transport capacity to move much of the material through the system. Therefore, concentrations of plutonium in stored channel sediments do not decline rapidly with the passage of time and associated discharge events smaller than the 25 yr design flood.

Second, at a scale of analysis that encompasses the entire canyon sys-

tem, concentrations of plutonium in stored channel sediments decline in the downstream direction. At finer scales of analysis there is considerable spatial variation in concentrations, and they do not decline smoothly in the downstream direction. This arrangement occurs because of geographic variation in the primary controls of the transport system. The stream power that drives the transport system is spatially variable in response to the influences of canyon geometry (particularly canyon width and gradient), which ultimately reflects geologic controls. The hydraulic resistance that counteracts the stream power is also spatially variable, with roughness elements having a distribution related to vegetation or to boulders that affect various stream segments to different degrees. The combination of stream power and resistance produces geomorphic work, in this case the transport and storage of sediment and associated contaminants. The results of research in the Los Alamos Canyon system suggest that the geographic variability of transport and storage is explainable by empirical and simulation approaches to the underlying mechanisms of stream power and resistance. Because the power and resistance are spatially variable, the resulting work of sediment and contaminant transport also has a distinctive and predictable geography.

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