

GEOMORPHIC CONTROLS ON CONTAMINANT DISTRIBUTION ALONG AN EPHEMERAL STREAM

STEVEN L. RENEAU,^{1*} PAUL G. DRAKOS,² DANNY KATZMAN,¹ DANIEL V. MALMON,³
ERIC V. MCDONALD⁴ AND RANDALL T. RYTT⁵

¹ Earth and Environmental Sciences Division, EES-9, MS D462, Los Alamos National Laboratory, Los Alamos, NM 87545, USA

² Glorieta Geoscience, Inc., 1723 Second Street, Santa Fe, NM 87505, USA

³ Swanson Hydrology and Geomorphology, 115 Limekiln Street, Santa Cruz, CA 95060, USA

⁴ Desert Research Institute, 2215 Raggio Parkway, Reno, NV 89512, USA

⁵ Neptune and Company, 1505 15th Street, Los Alamos, NM 87544, USA

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ABSTRACT

Sediment-borne contamination in a watershed can be highly variable as a result of fluvial processes operating over a range of time scales. This study presents a detailed analysis of the distribution of one contaminant along an ephemeral stream after 55 years of sediment transport, deposition, and exchange by flash floods. Wastewater containing plutonium was discharged into the Pueblo Canyon watershed from 1945 until 1964, and plutonium concentrations in fluvial deposits vary over five orders of magnitude. These variations can be attributed to three primary factors: time since contaminant releases, particle-size sorting, and mixing of sediment from different sources. The highest concentrations occur in fine-grained sediment deposits near the source that date to the period of effluent releases, and concentrations are lower in younger deposits, in coarser-grained deposits, and in deposits farther downstream. The spatial distribution of plutonium is strongly affected by longitudinal variations in the size of sediment deposits of different age. A major aggradation–degradation cycle in the lower canyon overlapped with the period of active effluent releases, and a significant portion of the total plutonium inventory is contained within large coarse-grained deposits below fill terraces that post-date 1945. The spatial pattern of contamination is thus determined by the specific geomorphic history of the watershed, in addition to processes of mixing and sorting during transport that occur in all fluvial systems. Copyright © 2004 John Wiley & Sons, Ltd.

KEY WORDS: sediment contamination; plutonium; aggradation–degradation cycle; sediment transport; ephemeral stream

INTRODUCTION

Pollutants enter fluvial systems by a variety of processes including direct discharges in wastewater, surface runoff from mine tailings or non-point sources, redistribution of atmospheric fallout, and inputs of contaminated groundwater. Many contaminants either originate as solid particles or have a strong affinity for particles, and their subsequent fate is strongly influenced by fluvial processes. Following mobilization by runoff, sediments and associated contaminants can be redeposited in a variety of settings including channels, floodplains, lakes, and estuaries. Along many streams and rivers, floodplains constitute significant storage sites for contaminants (e.g. Lewin and Macklin, 1986; Marron, 1992; Rowan *et al.*, 1995; Miller *et al.*, 1998; Marcus *et al.*, 2001). Contaminated sediment deposits can present potential environmental risks far downstream from the original source, and constitute secondary contaminant sources for continued transport. Understanding the distribution and variability of contaminants within fluvial systems is necessary for evaluating risk and potential remediation alternatives (e.g. Miller *et al.*, 1996). Defining the controls on contaminant distribution is also essential for understanding contaminant transport and for developing defensible contaminant transport predictions.

Pueblo Canyon, an ephemeral fluvial system in New Mexico that is partially within the Los Alamos National Laboratory (LANL), provides an excellent opportunity for evaluating how geomorphic processes acting over decadal time frames have redistributed contaminants in the environment. Plutonium was released into the

* Correspondence to: S. L. Reneau, Earth and Environmental Sciences Division, EES-9, MS D462, Los Alamos National Laboratory, Los Alamos, NM 87545, USA. E-mail: sreneau@lanl.gov

watershed beginning in 1945, associated with development of the first atomic bomb (Graf, 1994), and has since been widely distributed by floods. Concerns over potential environmental impacts have led to detailed investigations of plutonium and other contaminants, resulting in an extensive set of analytical data collected from within a well-defined geomorphic and sedimentologic context. The work presented here includes establishing a detailed watershed-scale estimate of plutonium inventory, defining spatial and temporal variations in plutonium concentration, and identifying primary geomorphic controls on plutonium distribution, building on previous research by Stoker *et al.* (1981), Graf (1996), and others. This work pertains to conditions as of the year 2000, prior to an increase in flood magnitude and frequency following the Cerro Grande fire of May 2000 and prior to excavation of sediment deposits with the highest levels of plutonium in 2001.

Note on terminology and units

Several isotopes of plutonium are present in the Pueblo Canyon watershed, and in this paper we use the term 'plutonium' synonymously with the combination of ^{239}Pu and ^{240}Pu ($^{239,240}\text{Pu}$). The analytical method used in this investigation (alpha spectroscopy) cannot distinguish ^{239}Pu from ^{240}Pu , but thermal ionization mass spectrometry analyses show that the results are dominated by ^{239}Pu ($^{240}\text{Pu}/^{239}\text{Pu}$ ratio = 0.015; Gallaher and Efurud, 2002). ^{238}Pu is also widespread, but it averages only 0.5 per cent of the concentration of $^{239,240}\text{Pu}$ (Reneau *et al.*, 1998).

The non-metric unit curie (Ci) is used in this paper to describe concentrations and amounts of plutonium, instead of the metric unit becquerel (Bq), because analytical laboratories in the United States typically report concentrations of radionuclides in picocuries per gram (pCi g^{-1}), and to be consistent with previous studies (e.g. Graf, 1994, 1996). One pCi g^{-1} equals 0.0367 Bq g^{-1} .

STUDY AREA

Pueblo Canyon is located on the Pajarito Plateau in northern New Mexico (Figures 1 and 2) and is a major tributary to Los Alamos Canyon, which drains into the Rio Grande. The watershed has a drainage area of 21.7 km^2 and a basin length of 16 km, and is underlain by rocks of the Jemez volcanic field (Smith *et al.*, 1970). Elevations range from 2790 to 1915 m. Several short tributary canyons feed into Pueblo Canyon on the plateau and are important as sources of either plutonium or uncontaminated sediment. These tributaries include Acid Canyon, which received discharges of radioactive effluent, and Walnut, Graduation, and Kwaga Canyons farther downstream (Figure 1).

Pueblo Canyon has a semiarid climate, with mean annual precipitation ranging from 650 mm a^{-1} in the headwaters to 350 mm a^{-1} to the east (Bowen, 1996). Almost half of the precipitation falls during summer thunderstorms, and these rains can produce flashy runoff that is responsible for most of the sediment transport on the Pajarito Plateau (e.g. Reid *et al.*, 1999; Malmon, 2002). Surface flow is ephemeral except downstream from the Los Alamos County wastewater treatment plant.

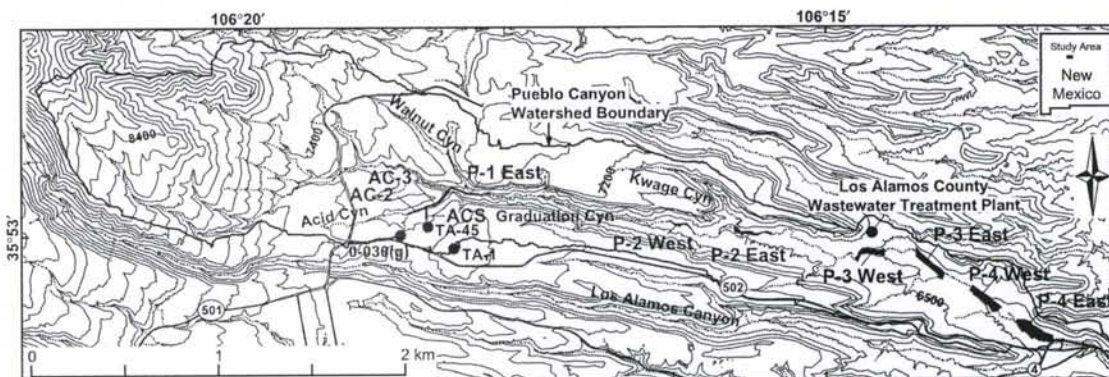


Figure 1. Map showing Pueblo Canyon watershed and locations of study reaches

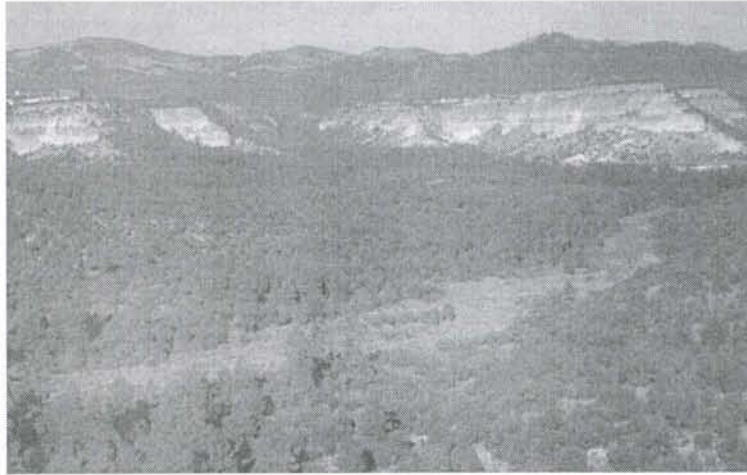


Figure 2. Photograph of Pueblo Canyon, looking west. Reach P-4 West is in foreground, and headwaters are in Jemez Mountains in the distance. Early Pleistocene ignimbrites of Bandelier Tuff are exposed in cliffs and canyon walls

Contaminant release history and behaviour

The Pueblo Canyon watershed included some of the original facilities of the Manhattan Project, established in 1943 for the purpose of developing the first nuclear weapons. Untreated radioactive effluents from the original Los Alamos technical area (TA-1) were discharged from late 1943 or early 1944 until 1951 into the South Fork of Acid Canyon, a short first-order drainage. Treated effluent was subsequently discharged into this drainage from 1951 to 1964 from a facility at TA-45. Releases of plutonium probably began in 1945, and most of the plutonium was probably discharged between 1945 and 1951 (Stoker *et al.*, 1981; Graf, 1994). Much smaller amounts of plutonium were discharged in the 1940s farther upstream in Acid Canyon from a septic tank outfall designated 0-030 (g) (Figure 1). The total amount of plutonium discharged into Acid Canyon is not known due to incomplete archival records.

Environmental sampling has demonstrated that the plutonium is largely bound to sediment particles, with only minor amounts being present as a dissolved or colloidal phase in surface water or shallow ground water (e.g. Stoker *et al.*, 1981; Rogers *et al.*, 2001). This is consistent with the expected geochemical behaviour of plutonium in the surface environment at near-neutral pH (Watters *et al.*, 1983). (Note the name 'Acid Canyon' is derived from 'acid sewers' that received effluent from radiochemical facilities, although waters here were not strongly acidic.)

Previous sediment investigations

The first sediment sampling in the watershed, in 1946, indicated the presence of plutonium along the full length of Pueblo Canyon downstream from Acid Canyon, documenting rapid transport to a distance of at least 11 km from the source (Kingsley, 1947). Subsequent work has included annual sediment sampling at a series of channel-bed stations since 1970 as part of environmental surveillance activities (e.g. Rogers *et al.*, 2001) and more detailed studies in the 1970s (e.g. Hakonson and Bostick, 1976; Nyhan *et al.*, 1976, 1982; Stoker *et al.*, 1981). More recently, existing data on plutonium in sediments were combined with geomorphic mapping to provide a revised estimate of the inventory of plutonium in the watershed (Graf, 1996). Findings of prior work included: (1) an overall downstream decrease in plutonium concentrations, although much local variability may be present; (2) higher concentrations of plutonium in the fine-grained fraction of channel-bed sediment than in the coarse-grained fraction; (3) higher concentrations of plutonium in inactive channels and 'banks' than in the active channel; and (4) the presence of a large fraction of the total plutonium inventory in the lower part of the canyon. Sediment sampling prior to this study focused on active channel sediments, and only limited data were available on plutonium concentrations in other settings. Of the >200 plutonium analyses from Acid and Pueblo

Table I. Characteristics of study reaches

Reach	Reach length (km)	Distance from Acid-Pueblo confluence (km)*	Drainage area (km ²)†	Average channel gradient (m m ⁻¹)	Notes
0-030(g)	0.24	-0.94	<0.1	0.062	First-order drainage below septic tank outfall
AC-2	0.10	-0.70	0.7	0.042	Relatively low gradient reach in tributary
ACS	0.29	-0.70	<0.1	0.096	First-order drainage below main outfalls
AC-3	0.41	-0.41	1.3	0.095	Steep bouldery reach in tributary
P-1 East	0.51	0	10.0	0.030	Relatively steep reach below Acid Canyon confluence
P-2 West	0.51	3.20	14.3	0.015	Canyon bottom broadens and flattens
P-2 East	0.46	4.49	16.5	0.015	Aggraded reach below Kwage Canyon confluence
P-3 West	0.51	6.41	18.3	0.010	Aggraded reach, lowest gradient in canyon
P-3 East	0.54	7.37	18.8	0.016	Aggraded reach, gradient steepens
P-4 West	0.52	8.31	20.0	0.018	Incised reach
P-4 East	0.60	9.16	21.1	0.019	Incised reach

* Distances from confluence of Acid and Pueblo Canyons to upstream end of reach are measured along the stream channel as depicted on 1:1200 scale maps with 0.61 m (2 feet) contour intervals.

† Drainage area measured from upstream end of reach.

Canyons prior to this investigation, only 18 samples were reported to be from well-vegetated surfaces adjacent to the channel. An additional 13 samples were from 'inactive channels' consisting of sand and gravel with sparse vegetation.

METHODS

Plutonium in the sediment of Pueblo and Acid Canyons was evaluated in a series of study reaches extending from the contaminant sources to the confluence with Los Alamos Canyon, adopting the 'representative reach' approach of Graf (1994). The reaches were selected to encompass the variations in geomorphic characteristics and depositional history within these canyons, and include 4.4 km of the total 11.2 km of channel downstream from TA-45 (Figure 1, Table I). Field investigations in each reach included detailed geomorphic mapping, focused on delineating discrete geomorphic units with varying deposit characteristics and/or age (Reneau *et al.*, 1998). Stratigraphic descriptions at 60 to 140 sections in each reach were used to estimate the average thickness of sediment in each unit that post-dates initial Manhattan Project activities (post-1942 sediment), and to select representative sample layers for plutonium analyses. In reaches near the source with relatively high levels of plutonium, field alpha radiation measurements were useful in mapping variations in contamination.

Sediment sampling occurred in two to four phases in each reach, and included a stratified sample allocation process (Gilbert, 1987, pp. 45–57) designed to reduce uncertainties in plutonium inventory. In total, 376 sediment layers were sampled for plutonium analyses at 155 sites downstream from TA-45. Samples were sieved to remove particles larger than 2 mm. Particle-size distribution analyses were also obtained for all samples.

Plutonium results within each reach were subdivided into data from fine-grained samples and coarse-grained samples for purposes of estimating plutonium inventory and defining variations in average plutonium concentration. The fine facies includes deposits that have a range in median particle size from coarse silt to medium sand in the <2 mm fraction. This size range is typical of suspended sediment deposited on floodplains as shown by field observations, data from suspended-sediment samples collected during floods, and theoretical calculations (Malmon, 2002). The coarse facies has a median particle size that ranges from medium to very coarse sand, which is typical of bed load sediment. Where possible, plutonium analyses were also subdivided by deposit age. Inventory estimates combined data on the area, thickness, density, and average plutonium concentration in each geomorphic unit and sediment facies in each reach, with extrapolation to unsampled areas.

Constraints on sediment age were obtained using a variety of methods. In lower Pueblo Canyon, examination of sequential aerial photographs allowed determination of the approximate age of a series of channel deposits

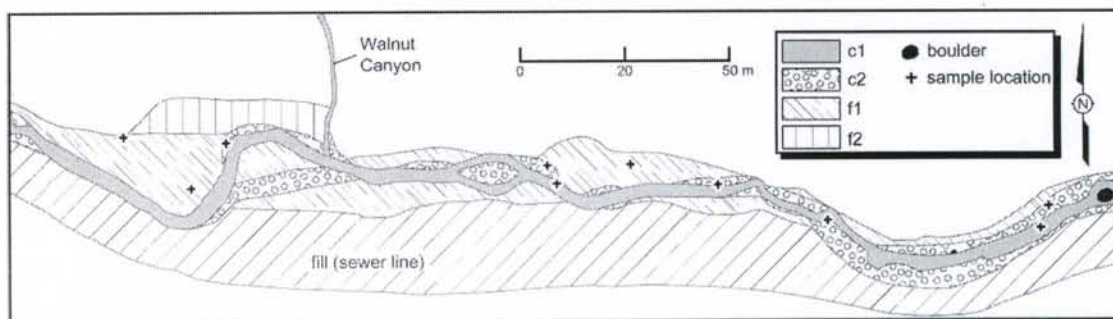


Figure 3. Geomorphic map of the eastern half of reach P-1 East, showing distribution of post-1942 units and sample locations. 'c1' indicates the active channel; 'c2' indicates abandoned post-1942 channels; 'f1' indicates post-1942 floodplains; 'f2' indicates possible post-1942 floodplains

and times of inundation of some floodplain areas. Dendrochronological analyses provided supplemental age information for some deposits. Additional information on temporal variations in plutonium concentration was provided by analyses of sediment samples collected annually from active channels since 1970 (e.g. Hakonson and Bostick, 1976; Stoker *et al.*, 1981; Rogers *et al.*, 2001). These samples are inferred to mostly consist of the coarse-grained sediment that dominates the stream bed, although some samples could be dominated by fine sediment.

GEOMORPHIC CHARACTERISTICS AND RECENT GEOMORPHIC HISTORY

Post-1942 sediment in Acid and Pueblo Canyons downstream from the outfalls occurs in a strip that varies from less than 1 m to 180 m in width and from less than 5 cm to at least 2 m in depth. This area includes an active channel with a coarse to very coarse sand and gravel bed, and adjacent abandoned channels and floodplains that are typically vegetated. The term 'abandoned channel' is used for areas occupied by the stream channel since 1942 but now abandoned following channel migration and/or incision, and these units typically include coarse-grained sediment underlying fine-grained sediment, although coarse and fine facies are often interstratified. The term 'floodplain' is used for pre-1943 surfaces that have been flooded since 1942 and that are predominantly capped by fine-grained sediment. The abandoned channels are typically lower than adjacent floodplains and contain thicker post-1942 sediment deposits. An example of the distribution of geomorphic units in one study reach is shown in Figure 3.

Median particle size in coarse facies deposits is typically coarse sand, with an average of 3 to 12 per cent silt and clay in the <2 mm fraction. Median particle size in fine facies deposits is typically very fine sand or fine sand, with an average of 30 to 50 per cent silt and clay. The fine sediment particles in the coarse facies may represent either suspended sediment that infiltrated into the stream bed, associated with transmission losses, or fine sediment deposited during the waning stages of floods. Both coarse and fine facies are coarser in the relatively steep and narrow upper canyon than in the gentler, broader lower canyon (Reneau *et al.*, 1998).

The width of the area flooded since 1942 generally increases downstream, and in most reaches over half of this area is occupied by active and abandoned channels (Figure 4a). Abandoned channels occupy areas similar to or much larger than the active channel in each reach, reflecting significant changes in channel location since 1942. The volume of post-1942 sediment in each reach also generally increases downstream. Volumes of coarse and fine sediment are similar in upstream reaches, but much larger volumes of coarse sediment are present in downstream reaches (Figure 4b). A total of about 360 000 m³ of post-1942, plutonium-bearing sediment is estimated to be present downstream of the outfalls into the South Fork of Acid Canyon, about 80 per cent in coarse sediment deposits and 20 per cent in fine sediment deposits.

Sediment residence times outside the active channel are spatially variable, influenced both by rates of bank erosion and by the width of geomorphic units. In the narrower upstream reaches, sediment in abandoned channel settings appears to have average residence times much shorter than 50 years. This interpretation is based on the

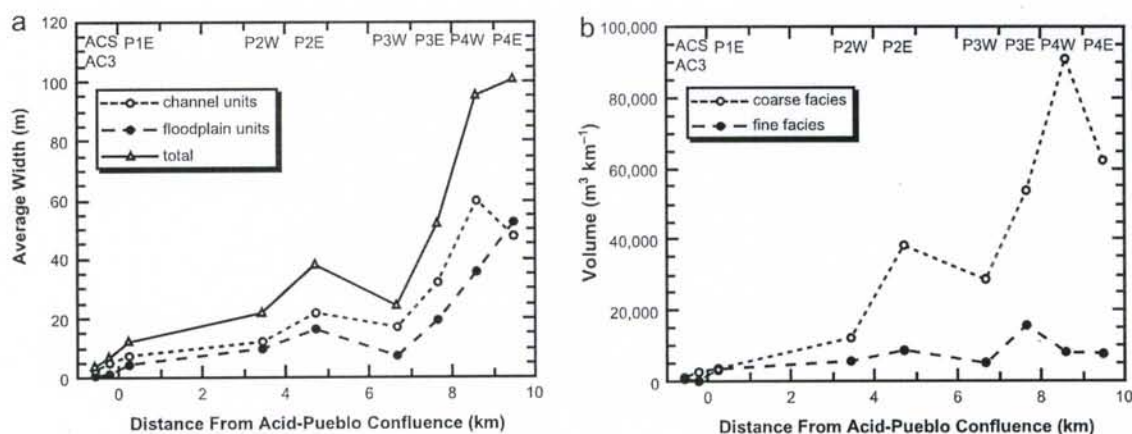


Figure 4. Width and volume of post-1942 geomorphic units and sediment facies in Acid and Pueblo Canyon reaches downstream from the primary outfalls. (a) Average width of channel and floodplain units, calculated as unit area divided by reach length. (b) Estimated volume of coarse facies and fine facies sediment, normalized by reach length and expressed in units of cubic metres per kilometre of channel

small percentage of these units that contain relatively high plutonium concentrations and are inferred to date to the period of effluent releases (discussed in a later section). In contrast, large deposits of sediment in the eastern reaches date to the period of effluent releases, and indicate much longer residence times in the lower part of the watershed.

Sequential aerial photographs dating back to 1935, combined with geomorphic mapping, have been used to reconstruct channel changes in lower Pueblo Canyon and document the characteristics of a historic aggradation-degradation cycle that strongly influences the present distribution of plutonium. Over a 30 to 35 year period from the early 1950s to the early-to-mid-1980s, the channel first aggraded then incised, with the peak of aggradation and subsequent incision progressing downstream. In reach P-4 West, the peak of aggradation is represented by a large sand lobe that was not vegetated in 1954 (Figure 5). The summer of 1952 is the wettest on record in Los Alamos, with 33 cm of rain in August, suggesting that this sand lobe was deposited in associated floods. By 1965, the channel had incised below this sand lobe and evidence for aggradation was visible 300 m downstream, where the channel had widened and overtopped floodplains (Figure 5). By the early 1970s, the channel had incised through the length of reach P-4 West, and aggradation and overtopping of floodplains was occurring downstream in reach P-4 East. The entire length of reach P-4 East was subsequently incised between 1981 and 1986.

Examination of stratigraphic sections indicates that up to 2 m of coarse sand was deposited in this aggradation cycle, and subsequent incision has been up to 3.4 m in reach P-4 West, decreasing to about 1.5 m downstream in reach P-4 East. Over 85 000 m³ of coarse sediment deposited in this aggradation cycle is estimated to remain in the lower 2.2 km of Pueblo Canyon beneath historic fill terraces. Surveyed cross-sections, combined with aerial photograph examination, indicate that incision rates averaged over decadal time scales in P-4 West were roughly constant between the 1950s and 2001, demonstrating continued adjustment of the channel. Upstream, incision ends abruptly at the west end of reach P-4 West where the incising channel encountered resistant Pliocene fanglomerates, forming knickpoints that have migrated upstream about 85 m since 1972 (Figure 5).

Evidence for a more recent period of aggradation over the last several decades is present upstream in reaches P-2 East, P-3 West, and P-3 East (Figure 1), with the channel bed raised at least 1.5 to 2 m by deposition of coarse sediment. Small floods in 1997 to 1999 completely infiltrated into the stream bed by the east end of reach P-3 West, indicating that transmission losses contributed to channel aggradation. This aggradation is believed to have been associated with high sediment discharge from Kwage Canyon that was supplied from rapidly eroding colluvial slopes 0.5 km upstream. Larger floods following the May 2000 Cerro Grande fire have subsequently incised the area near Kwage Canyon while accelerating aggradation downstream in reach P-3 West (Lyman *et al.*, 2002).

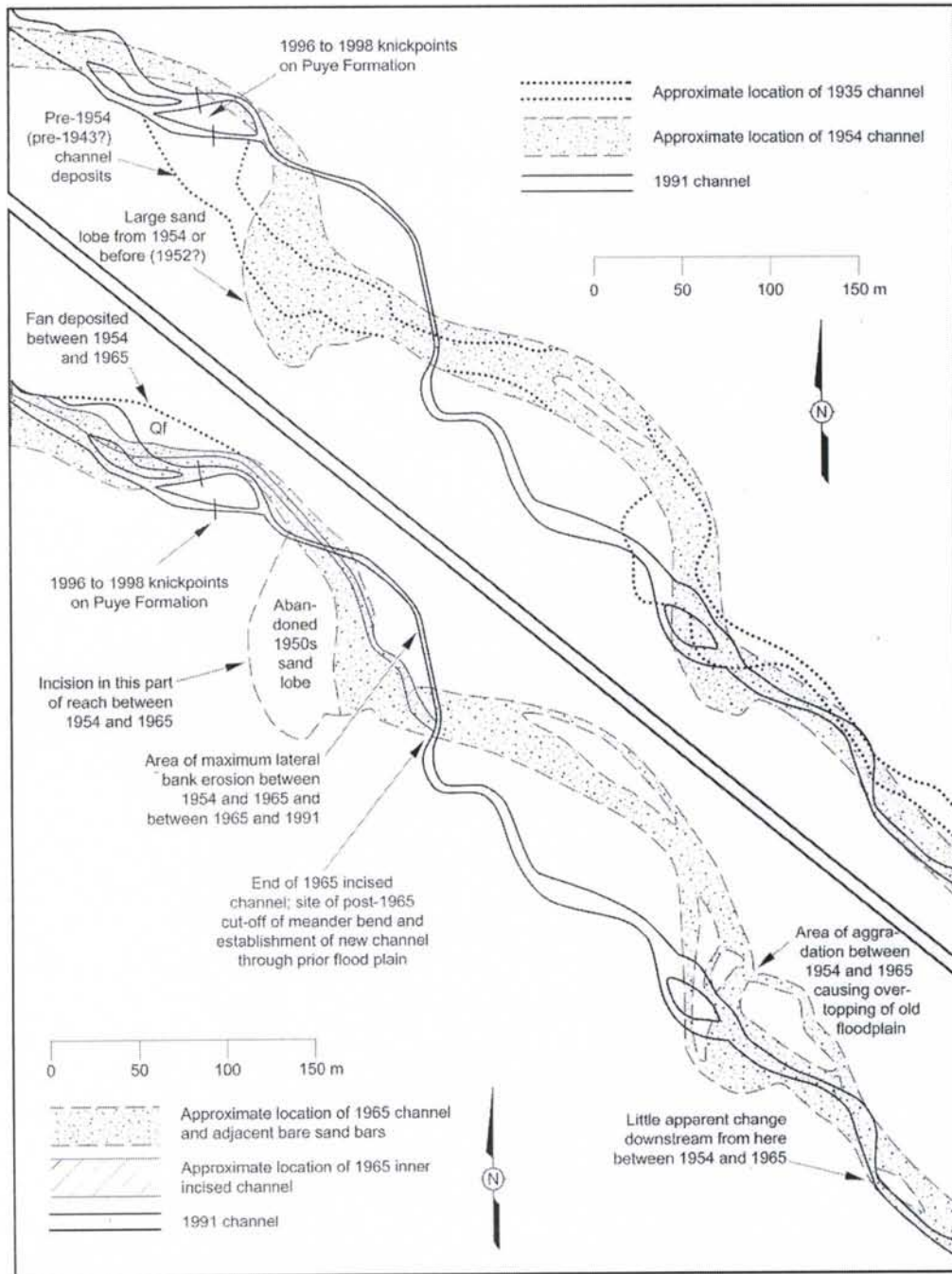


Figure 5. Maps of reach P-4 West, based on aerial photographs and field observations, illustrating key aspects of channel changes since 1935. Upper half of figure highlights channel in 1954, and shows channel location in 1935 and 1991 for comparison. Lower half of figure highlights channel in 1965, and shows channel location in 1991 for comparison

PLUTONIUM DISTRIBUTION

Spatial variations in plutonium concentration

Concentrations of plutonium in post-1942 sediments in the Pueblo Canyon watershed vary over five orders of magnitude, ranging from $<0.07 \text{ pCi g}^{-1}$ at sites upstream from Acid Canyon (primarily reflecting global atmospheric fallout from nuclear weapons testing), to about 8000 pCi g^{-1} close to the outfalls. Although a general pattern of decreasing concentrations downstream from the source is evident, consistent with mixing and dilution by uncontaminated sediments (e.g. Marcus, 1987), concentrations within each reach typically vary by several orders of magnitude related to the age and particle-size distribution of individual sediment layers. Down-canyon variability is also present due to differences in the timing of significant deposition between reaches. These factors are discussed below for the South Fork of Acid Canyon (reach ACS) and downstream reaches. Upstream reaches in Acid Canyon (AC-2, 0–030 (g)) are not discussed here because they are not significant from a watershed perspective.

Variations within reaches: effects of sediment age. Temporal variations in plutonium concentrations in channel sediments of lower Pueblo Canyon over the past 50 years were reconstructed by sampling coarse sediment from deposits of five different ages in reach P-4 (c. 1952 to 1991), supplemented by data from active channel samples (1970–2000). The highest concentrations of plutonium were measured in sediment deposited c. 1952 and between 1954 and 1965, during the time of active releases of radioactive effluent (Figure 6a). Sediment deposited after 1965 yielded consistently lower concentrations, and no trends are apparent over the last 35 years.

Plutonium concentrations in coarse sediment in upstream reaches also show variations related to age, with the highest concentrations measured in sediment that is inferred from geomorphic context to pre-date 1965. Channel sediment data from all surveillance stations in Pueblo Canyon also show no systematic changes since the 1970s, consistent with the data in Figure 6a. In contrast, data from lower Acid Canyon suggest general decreases since 1970 (Figure 6b), with average concentrations decreasing from 14.5 pCi g^{-1} in the 1970s to 7.8 pCi g^{-1} in the 1990s.

Within fine-grained sediment, clear age trends are also present in plutonium concentrations. In some cases, these variations are seen within individual stratigraphic sections, where sediment with relatively low plutonium concentrations overlies older, texturally similar sediment with much higher concentrations (Figure 7). In other cases, these variations are seen where approximate ages can be determined by relating overbank sediment to adjacent dated channel deposits. Table II summarizes data on plutonium concentrations in three areas where distinctions could be made between relatively old and relatively young fine-grained sediment. In all areas, maximum and average concentrations in the older sediments are an order of magnitude higher than in the younger sediments. Notably, sediments in reach P-3 East that probably date to 1991 have much lower concentrations

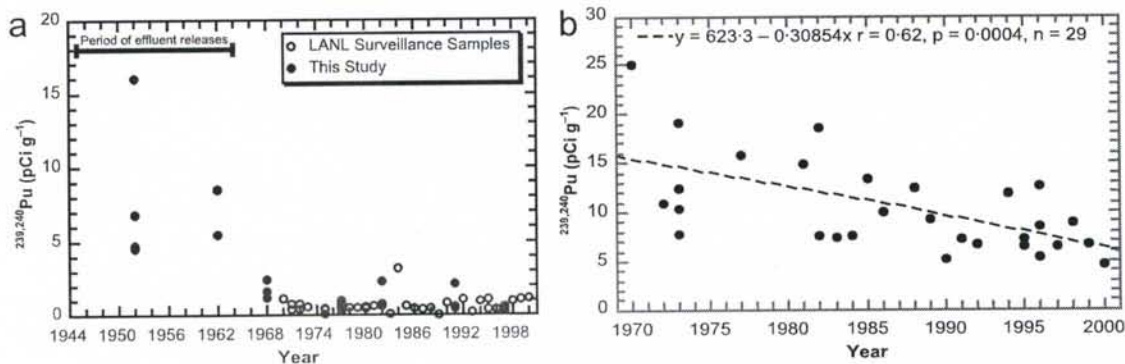


Figure 6. Temporal variations in $^{239,240}\text{Pu}$ concentration in channel samples from (a) lower Pueblo Canyon (reach P-4), 1952–2000, and (b) lower Acid Canyon (reach AC-3), 1970–2000. Data from LANL environmental surveillance reports (e.g. Rogers *et al.*, 2001), Stoker *et al.* (1981), and this study. Ages of reach P-4 samples between 1952 and 1991 in this study are estimated from examination of sequential aerial photographs. Three anomalous values $<0.2 \text{ pCi g}^{-1}$ from Acid Canyon are not included in plot (1987, 1993, 2000)

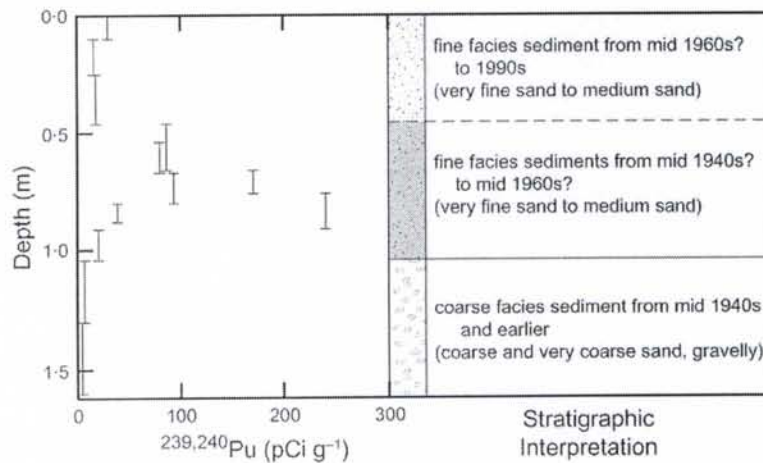


Figure 7. Depth variations in plutonium concentration below geomorphic surface in reach P-1 East with relatively high levels of plutonium (c2b unit), showing simplified stratigraphy. Subsurface layer with highest plutonium concentrations is shaded

Table II. Plutonium concentrations in fine-grained sediment

Sampling reach	Estimated age	Maximum $^{239,240}\text{Pu}$ (pCi g $^{-1}$)	Average $^{239,240}\text{Pu}$ (pCi g $^{-1}$)	Standard deviation (pCi g $^{-1}$)	Median particle size class §
ACS	1945–1964*	7780	1120	1830	vfs
ACS	1965–2000*	205	97	61	vfs
P-1 East	1945–1964 \dagger	502	189	150	fs
P-1 East	1965–1997 \dagger	39	18	11	fs
P-4 West	1945–1964 \ddagger	171	35	56	vfs
P-4 East	1965–1985 \ddagger	19	6.6	3.9	csi
P-3 East	1991 \ddagger	1.7	1.6	0.1	vfs

* Sediments with highest $^{239,240}\text{Pu}$ concentrations are from geomorphic units that are inferred to be largely contemporaneous with effluent releases, and sediments in other units are inferred to largely post-date effluent releases.

\dagger Sediments with highest $^{239,240}\text{Pu}$ concentrations are from subsurface layers in one geomorphic unit that are inferred to be contemporaneous with effluent releases; stratigraphically higher layers in this unit and sediments with similar $^{239,240}\text{Pu}$ concentrations in nearby units are inferred to largely post-date effluent releases.

\ddagger Ages estimated from spatial relations with channel deposits dated using sequential aerial photographs.

\S Determined from averages of multiple analyses: csi, coarse silt; fs, fine sand; vfs, very fine sand.

than sediments in reach P-4 East dating to *c.* 1965–1985 (Table II), suggesting continued declines in plutonium concentration in fine sediments that are not seen in coarse sediments.

The areas with highest concentrations of plutonium are typically restricted to relatively small portions of each reach, reflecting the limited area occupied by pre-1965 sediment. For example, in reach P-1 East, the highest plutonium concentrations (average of 189 pCi g $^{-1}$; Table II) were found in only 4 per cent of the area of contaminated sediment.

Variations within reaches: effects of particle size distribution. Average concentrations of plutonium in each reach are consistently higher in fine-grained sediment than in coarse-grained sediment, as expected from the geochemical behaviour of plutonium (Watters *et al.*, 1983). When the data are divided into general age groups to reduce variability related to temporal trends in contaminant concentration, they indicate statistically significant relationships ($p < 0.05$) between plutonium concentrations and percentage clay or percentage silt and clay (Figure 8). Within these groups, plutonium concentrations can vary by a factor of 10 between the coarsest sediment, with <5 per cent silt and clay, and the finest sediment, with 50 to 80 per cent silt and clay. The best correlations in Pueblo Canyon are found in reach P-4 (Figure 8a), with greater scatter in the relationships

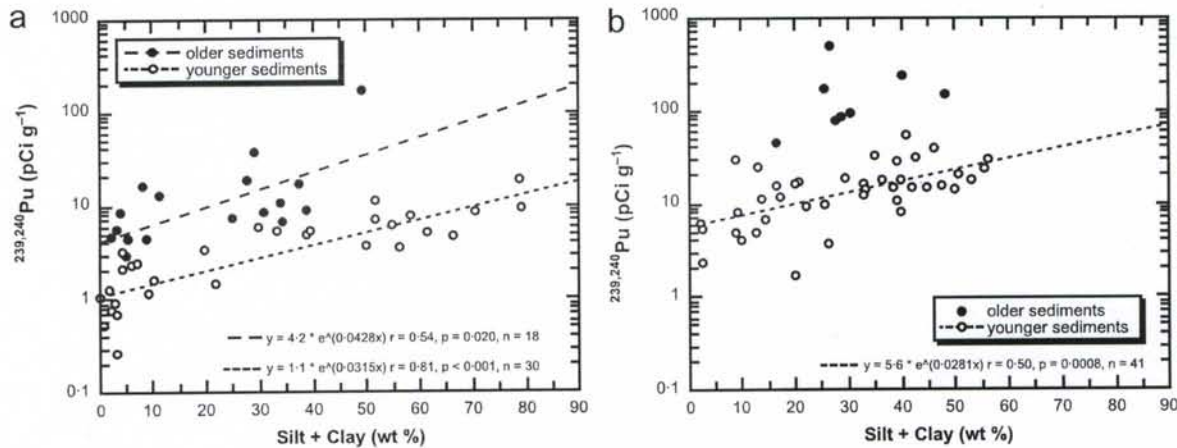


Figure 8. Scatter plots showing relations of plutonium concentration to percentage silt and clay in Pueblo Canyon. (a) Reach P-4. (b) Reach P-1 East. Samples are separated into general age groups, with the older samples believed to date to the period of effluent releases and the younger samples post-dating this period, although some overlap in age may exist between these subsets. Samples that are inferred to pre-date peak contamination in each reach or to be dominated by sediments from other sources (e.g. tributary drainages) are excluded from these plots. No linear regression is plotted for the older sediments in reach P-1 East because of the small sample size

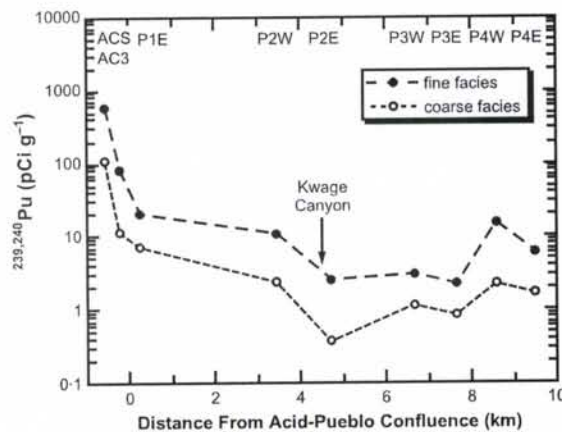


Figure 9. Estimated average plutonium concentration in coarse and fine facies sediment in each reach. Values are volume-weighted averages that combine data on average plutonium concentrations in each geomorphic unit in each reach with the estimated volume of that unit. Reaches upstream from reach ACS not shown

existing upstream (Figure 8b). These differences may be due to a combination of having more reliable age estimates of deposits in reach P-4, and less thorough mixing of sediment from different sources (e.g. Acid Canyon and upstream parts of Pueblo Canyon) and sorting in the upper canyon. Although the correlation is poorer upstream, the slope of the regression is similar to that obtained downstream and suggests that the same general relation holds along the length of Pueblo Canyon.

Variations between reaches. General downstream decreases in plutonium concentration between the outfall in the South Fork of Acid Canyon (reach ACS) and lower Pueblo Canyon exist in both relatively old and relatively young sediment layers, and in both coarse-grained and fine-grained sediment (e.g. Figure 8, Table II). These trends are consistent with the progressive mixing of uncontaminated sediment with plutonium-bearing sediment in floods, resulting in downstream dilution. However, deviations from this overall trend are seen using volume-weighted data (Figure 9). Average concentrations decrease from the source to a point about 4.5 km downstream from the Acid–Pueblo confluence, remain low and fairly constant from 4.5 to 8.3 km, and then

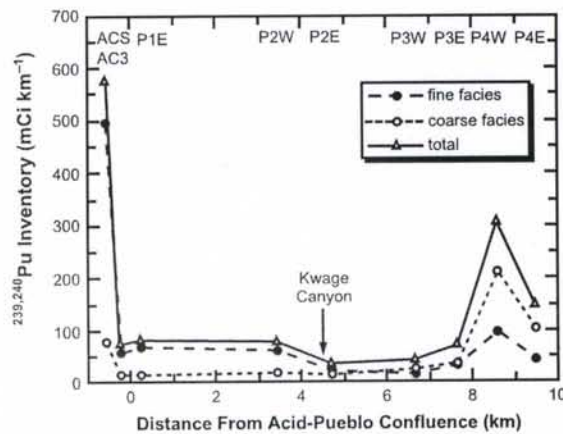


Figure 10. Estimated plutonium inventory in each reach, showing the portions in coarse and fine facies sediment. Values are normalized by reach length and shown in units of milli-curies per kilometre of channel (mCi km^{-1}). Reaches upstream from reach ACS not shown

increase significantly. This pattern can be attributed to differences in the age distribution and source of sediment in each reach. The area of high concentrations in lower Pueblo Canyon corresponds to the area that aggraded during the period of effluent releases, when measured plutonium concentrations were highest. In contrast, the low concentrations begin at the confluence with Kwage Canyon, a source of uncontaminated sediment, and include reaches that aggraded after effluent releases ceased. These data support the inference from morphologic changes along the channel that Kwage Canyon has been a major source of uncontaminated sediment, causing significant dilution of plutonium transported from upstream and contributing to channel aggradation downstream.

Spatial variations in plutonium inventory

The amount of plutonium stored in each reach, or the plutonium inventory, shows significant spatial variations between the source in the South Fork of Acid Canyon and the confluence with Los Alamos Canyon (Figure 10). The relative amount stored in fine-grained versus coarse-grained sediment in each reach also displays significant longitudinal variations.

Upstream from Kwage Canyon, about 75 to 85 per cent of the inventory in each reach is contained within fine-grained sediment deposits. The normalized inventory (units of mCi km^{-1}) is highest in reach ACS, a first-order drainage adjacent to the source (Figure 10). Inventories are similar in reaches AC-3, P-1 East, and P-2 West, with the downstream decrease in plutonium concentrations (Figure 9) offset by a downstream increase in sediment volume (Figure 4b).

Between Kwage Canyon and reach P-4 West, total inventories are relatively low and roughly equal amounts of plutonium are stored in the fine and coarse facies (Figure 10). However, normalized inventories in coarse sediment in reaches P-3 West and P-3 East are higher than in upstream reaches, associated with large volumes of coarse-grained sediments.

In the lowest part of Pueblo Canyon, the amount of stored plutonium increases dramatically, and about 70 per cent of the estimated inventory in reaches P-4 West and P-4 East is contained within coarse facies sediment (Figure 10). This increase is due to the occurrence of higher concentrations of plutonium, associated with deposits contemporaneous with effluent releases, and the occurrence of large volumes of coarse sediment, associated with the period of aggradation that extended from the 1950s into the 1980s in different parts of these reaches.

The total inventory of plutonium in the Pueblo Canyon watershed was estimated using the data presented in Figure 10, extrapolating to unsampled reaches (Table III and Figure 11). Of the estimated 1.1 Ci of plutonium in the watershed, 61 per cent is contained within fine-grained sediment and 39 per cent within coarse-grained sediment. Half is present along the first 4.9 km of channel and half along the lower 6.8 km.

Figure 11 also compares the estimated plutonium inventory from this study with that of previous work by Stoker *et al.* (1981) and Graf (1996). All three studies identified the large abandoned channel deposits in lower

Table III. Estimated plutonium inventory in Pueblo Canyon watershed

Area	Channel length (km)	Estimated fine facies inventory (mCi)	Estimated coarse facies inventory (mCi)	Total plutonium inventory (mCi)
Acid Canyon watershed upstream from South Fork	0.53	9	2	11
South Fork of Acid Canyon	0.29	158	23	181
Acid Canyon downstream from South Fork	0.41	24	7	31
Pueblo Canyon between Acid and Kwage Canyons	4.49	286	76	362
Pueblo Canyon between Kwage Canyon and reach P-4 West	3.82	80	99	179
Pueblo Canyon below west end of reach P-4 West	2.18	97	214	311
Total	11.72	654	421	1075

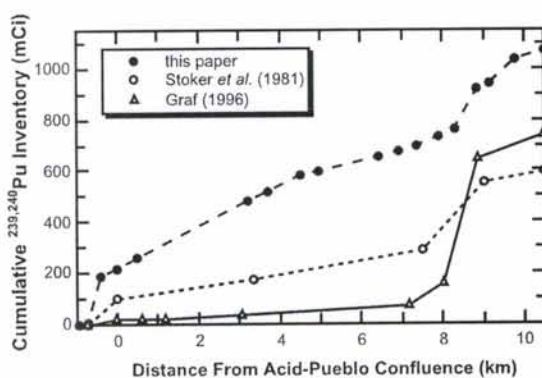


Figure 11. Plots of cumulative plutonium inventory versus distance along channel, comparing estimates in Stoker *et al.* (1981), Graf (1996), and this study

Pueblo Canyon as being important storage sites for plutonium, and the reaches immediately upstream as having much less plutonium. However, data obtained in this investigation indicate significantly more plutonium in reaches farther upstream than previously recognized. These differences result from a combination of more intensive sampling of fine-grained sediment deposits in this study and revisions to sediment volumes.

DISCUSSION

The distribution of plutonium in the Pueblo Canyon watershed reflects the interplay of (1) the timing of contaminant releases, (2) relations between particle size and plutonium concentration, (3) downstream transport and mixing, and (4) sediment deposition and subsequent remobilization during floods, including the influence of alternating aggradation and degradation along parts of the channel.

Systematic temporal variations exist in plutonium concentration in both fine-grained and coarse-grained sediment that are directly linked to the history of effluent releases in the watershed. These temporal variations are a primary factor controlling present spatial variations in plutonium concentration. Sediment with the highest levels of plutonium in each reach were probably deposited contemporaneously with, or soon after, peak releases, from the mid-1940s to the mid-1960s. These deposits have an irregular, discontinuous distribution due to spatial variability in deposition during this period and subsequent erosion. This finding is consistent with other studies which have reported the highest levels of contamination in geographically restricted sediment deposits dating to the general time of contaminant releases (e.g. Lewin *et al.*, 1977; Rowan *et al.*, 1995).

The general inverse relation between plutonium concentration and particle size is also consistent with the geochemical behaviour of plutonium and other actinides (Watters *et al.*, 1983) as well as previous field

investigations (e.g. Nyhan *et al.*, 1976). Data from this investigation indicate that much of the plutonium released into Acid Canyon quickly sorbed to fine-grained sediment particles, despite the small percentage of fines in the stream bed. The highest concentrations of plutonium were found in floodplain deposits within 250 m of the source that have 50–70 per cent silt and clay, indicating that within very short distances significant segregation of plutonium onto fine sediment particles had occurred, followed by suspension during floods and redeposition in overbank settings.

Due to preferential deposition of fine-grained sediment on abandoned channels and floodplains, the highest concentrations and much of the plutonium inventory occurs in these settings. Preferential storage of contaminants on floodplains is well documented in the literature (e.g. Lewin and Macklin, 1986; Marron, 1992; Rowan *et al.*, 1995). The concurrent storage of a significant fraction of the plutonium inventory in coarse sand deposits is related to substantial historic aggradation that led to the deposition of large volumes of bed load sediment, and to subsequent abandonment of these surfaces.

The cycle of aggradation and degradation documented since the 1950s in lower Pueblo Canyon is similar to that occurring in areas which have experienced large, transient increases in sediment supply. Examples include channels that received large volumes of mining debris (Gilbert, 1917; Lewin *et al.*, 1983; Knighton, 1989; Macklin and Lewin, 1989; James, 1993; Rowan *et al.*, 1995), and watersheds that had major increases in hillslope erosion following fires (Germanoski and Harvey, 1993) or logging (Madej and Ozaki, 1996). The downstream progression in the locus of aggradation and subsequent incision over periods of decades, causing abandonment of former channels and creation of historic fill terraces, seems characteristic of many systems with large increases in sediment input.

Because of these similarities, we infer that the historic aggradation–degradation cycle in lower Pueblo Canyon records a significant increase in sediment supply during or before the early 1950s. The period of initial aggradation corresponds to a time of pronounced drought which has been associated with vegetation changes and accelerated erosion elsewhere on the Pajarito Plateau (Allen and Breshears, 1998). This suggests a possible link between drought-induced landscape changes and channel aggradation. Development of the Los Alamos town site could also have increased sediment input. Regardless of possible effects of recent climate variability or land use change on sediment supply, episodic aggradation and degradation may also be a natural characteristic of the Pueblo Canyon channel related to high variability in discharge, as inferred for ephemeral channels in semiarid and arid watersheds elsewhere (e.g. Thornes, 1979).

The geomorphic history of Pueblo Canyon since initial contaminant releases, including the location and timing of aggradation and incision, strongly influenced contaminant distribution. Different patterns of contamination would have resulted if releases corresponded to different parts of an aggradation–degradation cycle. This study indicates that models to predict long-term contaminant transport in many systems should incorporate both channel–floodplain interactions (e.g. Malmon *et al.*, 2002, 2003) and cycles of aggradation and degradation associated with temporal variations in sediment input (e.g. Pickup *et al.*, 1983; Kelsey *et al.*, 1987), along with corresponding uncertainties in the characteristics of these cycles.

The timing of floods in relation to contaminant releases may also have had a strong influence on the present distribution of plutonium. A particularly wet summer occurred in 1952, the year after releases of untreated radioactive effluent ceased. Associated floods should have been effective at mobilizing much of the plutonium inventory that had accumulated in the stream bed and rapidly dispersing it downstream. This interpretation is consistent with the deposition of large volumes of coarse sediment and associated plutonium in lower Pueblo Canyon in the 1950s. Based on the small size of correlative deposits of fine sediment and modelling results from the adjacent watershed of upper Los Alamos Canyon (Malmon, 2002), we infer that much of the suspended sediment and associated plutonium from floods in the 1950s was transported out of Pueblo Canyon, although the amount that has left the canyon has not been reliably quantified.

CONCLUSION

The distribution of sediment-borne contamination can be highly variable as a result of both the history of contaminant releases and geomorphic processes acting over decadal time frames. This variability contains systematic trends with distance, particle size, and age, related to downstream mixing of contaminated and

uncontaminated sediment, particle sorting, and temporal variations in contaminant input. Superimposed on such trends can be irregular longitudinal patterns in the size of sediment deposits of different age, reflecting the specific geomorphic history of a watershed. Important in this local history are the size and sequencing of floods and sediment input following contaminant releases, and the timing and magnitude of erosion and deposition at different sites. Establishing a detailed, site-specific geomorphic framework for contaminant distribution can lead to improvements in addressing a variety of problems related to the environmental impacts of sediment contamination.

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