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Subject: Pueblo Canyon Paper

Enclosed is a paper entitled "Plutonium in Pueblo Canyon, New Mexico: Geomorphic Controls on Contaminant Distribution". This paper summarizes the results of six years of field investigation by the Los Alamos National Laboratory's Environmental Restoration Project, building on and supplementing prior work on this topic. It is hoped that this paper will be of value both from a local perspective, by contributing to a better understanding of contamination in this important watershed, and from a broader scientific and environmental perspective, by helping to elucidate the primary importance of fluvial processes and recent geomorphic history in controlling contaminant distribution in sediments.

This paper has been submitted to a peer-reviewed journal for publication, and in the interim is citable as Los Alamos National Laboratory report LA-UR-03-1654. Questions concerning this work are welcome, as well as comments that might help us improve ongoing and future work in Pueblo Canyon and other canyons at Los Alamos.

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PLUTONIUM IN PUEBLO CANYON, NEW MEXICO: GEOMORPHIC CONTROLS ON CONTAMINANT DISTRIBUTION

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ABSTRACT

Plutonium was discharged in liquid effluent into the Pueblo Canyon watershed, an ephemeral stream system in New Mexico, from 1945 until 1964. This study evaluates the distribution of plutonium in the watershed after 55 years of sediment transport, deposition, and exchange by flash floods. Concentrations of plutonium in fluvial deposits vary over five orders of magnitude, and these variations can be attributed to three primary factors: time since contaminant releases, sorting of different sized particles, 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 active effluent releases, and concentrations are less in younger deposits, in coarser-grained deposits, and in deposits farther downstream. An estimated 1.1 Ci of plutonium is contained within about 360,000 m^3 of sediment, 60% within fine-grained deposits and 40% within coarse-grained deposits. The spatial distribution of plutonium is strongly affected by longitudinal variations in the size of sediment deposits of different age, reflecting the unique geomorphic history of the canyon. 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. A larger amount of plutonium associated with suspended sediment in flash floods is inferred to have been transported out of the watershed. An understanding of the geomorphic controls on contaminant distribution provides a foundation for evaluating potential present day risks to human health and ecosystems, for evaluating the effects of continued contaminant redistribution, and for evaluating remediation alternatives.

INTRODUCTION

Pollutants enter fluvial systems by a variety of processes including direct discharges in liquid effluent, surface runoff from mine tailings or non-point sources such as roads, 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. Once mobilized by runoff, sediments and associated contaminants can be incrementally transported downstream and deposited 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). These deposits of contaminated sediments can present potential environmental risks far downstream from the original source, and also constitute secondary contaminant sources for continued transport. Understanding the distribution and variability of contaminants within fluvial systems is critical 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.

This paper presents an evaluation of the distribution of plutonium in the Pueblo Canyon watershed, an ephemeral fluvial system in New Mexico that is partially within the Los Alamos National Laboratory (LANL). Sediments in the watershed contain a variety of contaminants that include radionuclides, metals, and organic chemicals (Reneau *et al.*, 1998b). In this paper we focus on plutonium because it is widely distributed at concentrations above atmospheric fallout

levels, and because it provides a unique tracer for evaluating interactions between contaminants and fluvial processes since 1945. It is also a contaminant that has generated concern from a regional perspective (Graf, 1994). The plutonium at LANL is largely bound to sediment particles, with only minor amounts being present in surface water or alluvial ground water (e.g., Stoker *et al.*, 1981; Rogers *et al.*, 2001). The primary isotope of concern, ^{239}Pu, has a half-life of 24,100 years, and, in human time frames, will be a permanent part of the landscape until removed by erosion or excavation.

The primary goal of this paper is to explore the relationship between contaminant distribution and fluvial processes. The work presented here included establishing a detailed watershed-scale estimate of plutonium inventory, defining variations in plutonium concentration, and identifying primary geomorphic controls on plutonium distribution. This work pertains to conditions as of the year 2000, prior to excavation of sediment deposits with the highest levels of plutonium in 2001 (Reneau *et al.*, 2002) and prior to an increase in flood magnitude and frequency following the Cerro Grande fire of May 2000 (Shaull *et al.*, 2002). This work builds on previous research by Stoker *et al.* (1981), Graf (1996), and others, providing a greatly enlarged data base of plutonium analyses and an improved geomorphic and sedimentologic framework for samples.

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}Pu). The analytical method used in this investigation (alpha spectroscopy) cannot distinguish ^{239}Pu from ^{240}Pu, although thermal ionization mass spectrometry (TIMS) analyses show that the ratio of ^{240}Pu to ^{239}Pu here is about 0.015 (Gallaher and Efurd, 2002) and the results are therefore dominated by ^{239}Pu. ^{238}Pu is also widespread in Pueblo Canyon sediments, but it is collocated with ^{239,240}Pu and averages only 0.5% of the concentration of these isotopes (Reneau *et al.*, 1998b).

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

Geography and Geology

Pueblo Canyon is located on the Pajarito Plateau of northern New Mexico (Figures 1 and 2) and is a major tributary to Los Alamos Canyon, which drains into the Rio Grande. The Pueblo Canyon watershed has a drainage area of 21.7 km^2 and a basin length of 16 km. Elevations range from 2790 m on the eastern flank of the Jemez Mountains to 1915 m at the confluence with Los Alamos Canyon. The watershed is underlain by rocks of the Jemez volcanic field, including dacitic lavas of the Pliocene and Miocene Tschicoma Formation in the headwaters, ignimbrites of the early Pleistocene Bandelier Tuff on the plateau, and Pliocene fanglomerates of the Puye Formation in the lower canyon (Smith *et al.*, 1970). 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 from laboratory outfalls, and Walnut Canyon, Graduation Canyon, and Kwage Canyon farther downstream (Figure 1).

Climate and Hydrology

The climate in the Pueblo Canyon watershed is semiarid, with annual precipitation ranging from about 65 cm a^{-1} in the headwaters to about 35 cm a^{-1} in the downstream reaches (Bowen, 1996). Almost half of the precipitation falls during summer thunderstorms, and these intense rains produce flashy runoff and are responsible for most of the sediment transport on the Pajarito Plateau (e.g., Reid *et al.*, 1999; Malmon, 2002). Surface flow in Pueblo Canyon is ephemeral except downstream from the Los Alamos County wastewater treatment plant. The watershed has a short gauged record (1992-present), and the maximum recorded flood peak in the lower canyon prior to the Cerro Grande fire was 0.3 m^3 s^{-1} in July 1999 (Shaull *et al.*, 2002). High water lines from an August 1991 flood in lower Pueblo Canyon were similar to that from a post-fire flood in October 2000 that had an estimated peak discharge of 4.2 m^3 s^{-1}. There is a high degree of spatial variability in summer rainfall and resulting floods in this area, and nearby canyons of similar size had reported floods up to 15 m^3 s^{-1} during the 1990s. In comparison, a post-fire flood in July 2001 had an estimated peak discharge of 41 m^3 s^{-1} in lower Pueblo Canyon (Shaull *et al.*, 2002).

Contaminant Release History

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. After World War II, weapons research and other work continued as part of the Los Alamos Scientific Laboratory and later LANL. Untreated effluents from radiochemical laboratories in the original Los Alamos technical area (TA-1) were discharged from late 1943 or early 1944 until April 1951 into the South Fork of Acid Canyon, a short first-order drainage. Following development of technologies to reduce radionuclide concentrations in wastewater, treated effluent was discharged into the South Fork of Acid Canyon from 1951 to 1964 from a facility at TA-45. The initial 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). The total amount of plutonium discharged into Acid Canyon is not known. Using archival data, Stoker *et al.* (1981, p. 26) estimated releases of about 0.18 Ci, but estimates of plutonium inventory in sediments in the Pueblo Canyon watershed (0.3 to 1.1 Ci; Stoker *et al.*, 1981; Graf, 1996; this study) indicate that the actual amounts released were much higher, and that the archival records are incomplete. These releases into Acid Canyon constitute the primary source of plutonium in the Los Alamos Canyon watershed, and the primary source of plutonium that has reached the Rio Grande from LANL sources; significant amounts of plutonium have also reached the river from redistribution of atmospheric fallout (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) (LANL, 2000; Figure 1).

Previous Sediment Investigations

Contaminants associated with sediments in Pueblo Canyon and Acid Canyon have been investigated in many studies since the initial radiochemical research in the watershed during World War II. The first sediment sampling, in 1946, indicated the presence of plutonium along the full length of Pueblo Canyon downstream from Acid Canyon, documenting rapid transport for a distance of at least 11 km from the source (Kingsley, 1947). Subsequent work has included repeated sediment sampling at a series of stations since 1970 as part of LANL annual environmental surveillance activities (e.g., Rogers *et al.*, 2001) and more intensive studies in the 1970s (e.g., Hakonson and Bostick, 1976; Nyhan *et al.*, 1976a, 1976b, 1982; Stoker *et al.*, 1981). More recently, existing data on plutonium in sediments were combined with geomorphic mapping of Pueblo Canyon to provide a revised estimate of the inventory of plutonium in the canyon (Graf, 1996). Findings of prior work included: 1) an overall downstream decrease in plutonium concentrations from the source, although much local variability may be present; 2) higher concentrations of plutonium in the fine-grained fraction of channel 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. Notably, sampling of sediment deposits prior to this study focused primarily on active channel sediments, and only limited data were available on plutonium concentrations in overbank deposits, where concentrations are the highest. Of the >200 plutonium analyses obtained from Acid Canyon and Pueblo Canyon prior to this investigation, only 18 samples were reported to be from well-vegetated surfaces adjacent to the channel, and 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 (Reneau *et al.*, 1998b, 2002; LANL, 2000; and unpublished work), 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 1). Field investigations in each reach included detailed geomorphic mapping at a scale of 1:200 to 1:1200. The mapping was focused on delineating discrete geomorphic units with varying deposit characteristics and/or age. 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, field measurements of alpha radiation were used to help map the areas with highest plutonium concentration. Identification of the base of post-1942 sediment utilized several methods, including: determining the depth of burial of trees and associated buried soils; noting the presence or absence of exotic materials imported to the watershed after 1942 (e.g., quartzite gravel, coal); and using laboratory analyses to determine the depth of plutonium.

Sediment sampling occurred in two to four phases in each reach, and the results from initial phases were used to help guide subsequent sampling and revise the geomorphic maps, where

appropriate. Selection of sites for sediment sampling included a stratified sample allocation process (Gilbert, 1987, p. 45-57) designed to reduce uncertainties in the plutonium inventory in each reach. Samples were preferentially allocated to units and sediment facies with either high variability in plutonium concentrations or a large portion of the total inventory. Additional samples were collected from sites that helped define the horizontal and vertical extent of contaminated sediments, and that provided information on variations in plutonium concentration as related to the age and grain size of the deposit. In total, 376 sediment layers were sampled for plutonium analyses at 155 sites downstream from TA-45. Samples for plutonium analyses were sieved to remove particles larger than 2 mm. Particle size analyses were also obtained on all samples.

Plutonium analyses within each reach were subdivided into data from fine-grained facies and coarse-grained facies 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 particle 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 sand to very coarse sand, which is typical of bed load sediment in active channels. 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, extrapolating to unsampled areas.

Ages of sampled sediment deposits were evaluated using a variety of methods. The best age control was obtained in lower Pueblo Canyon, where examination of sequential aerial photographs allowed determination of the approximate age of a series of channel deposits and times of inundation of some floodplain areas. Dendrochronological analyses provided supplemental age information for some sediment deposits. Additional information on temporal variations in radionuclide concentration is provided by analyses of sediment samples collected annually from active stream channels since 1970 (e.g., Purtymun, 1971; Hakonson and Bostick, 1976; Stoker *et al.*, 1981; Rogers *et al.*, 2001). These surveillance samples are inferred to be largely of the coarse-grained sediment that dominates the stream bed in Pueblo and Acid canyons. However, no particle size data or field descriptions are available and the effect of varying percentages of fine sediment in the surveillance samples on inter-annual variability in radionuclide concentration cannot be evaluated.

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 about 2 to 180 m in width and from less than 5 cm to at least 2 m in depth. The area of post-1942 sediment includes an active channel with a coarse to very coarse sand and gravel bed, and adjacent abandoned channels and floodplains that are typically vegetated. As used in this study, the term "abandoned channel" refers to areas occupied by the main stream channel since 1942 but now abandoned following channel migration and/or channel incision; included here are point bars, attachment bars, cut-off meanders, braid channels, and fill terraces. These surfaces are typically vegetated and include coarse-grained sediment containing exotic material that is overlain by 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 overlain by fine-grained sediment. These surfaces are commonly higher in elevation than adjacent abandoned channel surfaces and therefore inundated less frequently. An example of the distribution of geomorphic units in one study reach is shown in Figure 3.

Sediment texture and plutonium concentrations can be similar on abandoned channels and floodplains in a reach, but sediment thickness is generally greater on the abandoned channels. Median particle size in fine facies deposits on abandoned channels and floodplains is typically very fine sand or fine sand, with an average of 30% to 50% silt and clay. These deposits are locally dominated by coarse silt. Coarse facies deposits in both active and abandoned channels have a small but pervasive component of silt and clay, typically averaging 3% to 12% in the <2 mm fraction. These fine sediment particles in the coarse facies may largely represent either suspended sediment that infiltrated into the stream bed, associated with transmission losses, or thin layers of fine sediment deposited during the recessional portion of floods. Coarse facies sediment in abandoned channel units often has higher silt and clay percentages than the active channel, which is believed to record the downward translocation of fines from infiltrating floodwaters after abandonment of the surfaces. Both coarse and fine facies are coarser in the relatively steep and narrow western canyon than in the gentler, broader eastern canyon (Figure 4), consistent with a higher energy regime to the west.

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 5a). Abandoned channels occupy areas either 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 stored in each reach also tends to increase 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 5b). A total of about 360,000 m^3 of post-1942, plutonium-bearing sediment is estimated to be present downstream of the outfalls into the South Fork of Acid Canyon; about 80% are coarse sediment deposits and 20% are fine sediment deposits.

Geomorphic processes in all reaches since 1942 have included overbank flooding and associated sediment deposition, channel scour and fill, and bank erosion associated with lateral channel migration. The cut-off of meander bends and the abandonment of braid channels has occurred in some reaches. About 40 to 60 m of lateral channel migration occurred at one meander bend between 1954 and 1991, although bank erosion rates elsewhere are typically much lower. Downstream reaches have also experienced significant vertical changes in channel elevation associated with historic aggradation and degradation.

Field observations indicate that 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 small percentage of these units that contain relatively high plutonium concentrations and are inferred to date to the early 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 to document the characteristics of a historic aggradation-degradation cycle that strongly influences the present

distribution of plutonium. Over a roughly 30-year period from the early 1950s to the mid 1980s, the channel first aggraded then incised, with the peak of aggradation and subsequent incision progressing downstream. In reach P-4 West, a large bare sand lobe is visible on a 1954 photograph, suggesting deposition in a recent flood (Figure 6). The summer of 1952 was the wettest on record in Los Alamos, with 33 cm of rain recorded in August, suggesting that this sand lobe was deposited at that time. By 1965, the channel had incised below this level and evidence for aggradation was visible 300 m downstream, where the channel had widened and overtopped floodplains (Figure 6). By the early 1970s, the channel had incised through the length of reach P-4 West, and aggradation and overtopping of floodplains was evidenced downstream in reach P-4 East. Between 1981 and 1986, the entire length of reach P-4 East had been incised.

Examination of stratigraphic sections indicates that at least 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 m^3 of coarse sediment that was 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 (Figure 7), 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 Puye Formation fanglomerates, forming knickpoints that have migrated upstream about 85 m since 1972 (Figure 6).

Evidence for both aggradation and incision since 1942 is also present upstream in reaches P-2 East, P-3 West, and P-3 East (Figure 1), with estimated changes in channel elevation of at least 1.5 to 2 m. In contrast to the recent incision in downstream reaches, these reaches apparently experienced net aggradation in the last several decades. Field observations in 1997, 1998, and 1999 indicated that small floods completely infiltrated into the stream bed by the east end of reach P-3 West, causing their sediment load to be deposited. This demonstrates that high transmission losses can contribute to channel aggradation in Pueblo Canyon. Channel aggradation at that time began near the confluence of Pueblo and Kwage canyons, and is believed to have been associated with a high sediment flux from Kwage Canyon over several decades that was derived from rapidly eroding colluvial slopes about 0.5 km upstream.

PLUTONIUM DISTRIBUTION

Spatial Variations in Plutonium Concentration

Concentrations of plutonium in young sediments in the Pueblo Canyon watershed vary over five orders of magnitude, ranging from <0.07 pCi g^{-1} at sites upstream from Acid Canyon that primarily reflect 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, with maximum concentrations of plutonium being two orders of magnitude lower than in reach ACS.

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 using the geomorphic record of channel changes in reach P-4, discussed previously, supplemented by data from active channel samples collected since 1970. In reach P-4, multiple samples of coarse sediment were collected from deposits of five different ages that range from ca. 1952 to 1991, as determined using sequential aerial photographs. The highest concentrations of plutonium were measured in sediment deposited ca. 1952 and between 1954 and 1965, during the time of active releases of radioactive effluent, and sediment deposited after 1965 yielded consistently lower concentrations (Figure 8). Active channel samples collected since 1970 have similar concentrations, and no trends are apparent over the last 30 years.

Plutonium concentrations in coarse sediment in upstream reaches also show variations related to age, although age control is less precise and trends with time are less well defined than in reach P-4. In all cases, the highest concentrations are present in sediment that is inferred, based on geomorphic context, to pre-date 1965. Channel sediment data from surveillance stations upstream in Pueblo Canyon show no systematic changes since the 1970s, consistent with the data in Figure 8. In contrast, data from lower Acid Canyon suggest general decreases since 1970 (Figure 9). Average concentrations decrease from 14.5 pCi g^{-1} in the 1970s to 11.2 pCi g^{-1} in the 1980s and 7.8 pCi g^{-1} in the 1990s.

Within fine-grained sediment, clear temporal variations 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 10). In other cases these variations are seen where approximate ages can be determined by relating overbank sediment to adjacent dated channel deposits (e.g., reach P-4, Figure 6). Table 2 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 were probably deposited during a flood in August 1991 have much lower concentrations than sediments in reach P-4 East dating to ca. 1965-1985 (Table 2), suggesting continued declines in plutonium concentration in fine sediment that are not seen in coarse sediment.

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 2) were found in only 4% of the area of contaminated sediment.

Variations Within Reaches: Effects of Particle Size Distribution

Data from each reach shows that average concentrations of plutonium are always higher in fine-grained sediment than in coarse-grained sediment, as expected from the geochemical behavior of plutonium (Watters *et al.*, 1983). In more detail, the data also suggest general positive relationships between plutonium concentrations and percent clay or percent silt and clay,

although considerable scatter exists in these relationships that are at least partially caused by temporal variations in plutonium concentration within a reach, as discussed above.

Examples of relationships between plutonium and silt and clay content in two Pueblo Canyon reaches are shown in Figure 11, divided into general age groups based on geomorphic context. Within these groups, plutonium concentrations can vary by a factor of 10 between the coarsest sediment, with <5% silt and clay, and the finest sediment, with 50 to 80% silt and clay. In addition to the effects of including sediment of variable age in these plots, the mixing of variable amounts of sediment from contaminated sources with sediment from uncontaminated parts of the watershed in the sampled deposits could also contribute to the scatter in these relationships.

The best correlations between plutonium concentration and silt and clay content in Pueblo Canyon are found in both general age groups in reach P-4 (Figure 11a), with poorer correlations present upstream (Figure 11b). 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) in the upper canyon. Although the correlation is poorer upstream in reach P-1 East, the slope of the regression is similar to that in the younger sediments downstream and suggests the same general relation holds along the length of Pueblo Canyon.

Data from Acid Canyon suggest similar relations as seen in Pueblo Canyon. With the exception of three outliers, samples from reach AC-3 display a strong correlation between plutonium concentration and silt and clay content (Figure 12), with a slope of the regression very similar to that from reach P-4. The outliers are inferred to represent relatively old sediment layers, although field evidence is not conclusive.

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 are seen in both relatively old and relatively young sediment layers, and in both coarse-grained and fine-grained sediment (e.g., Figure 11, Table 2). Such decreases are consistent with the progressive mixing of non-contaminated sediment with plutonium-bearing sediment in floods, resulting in downstream dilution.

Variations in plutonium concentrations between reaches were also examined by calculating volume-weighted averages that combine average concentrations from each geomorphic unit with the estimated volume of that unit. Results for both coarse and fine sediment are shown in Figure 13, and reveal large geographic variations that are similar between these two facies. Average concentrations decrease from the source in the South Fork of Acid Canyon to a point about 4.5 km downstream from the confluence with Pueblo Canyon, remain low and fairly constant from 4.5 to 8.3 km, and then increase significantly. These variations are attributable to differences in the age distribution of sediment deposits in each reach, and also to variable sources of sediment. The area of high concentrations in lower Pueblo Canyon corresponds to the area that aggraded during the period of effluent releases, including a high percentage of deposits dating to the period when plutonium concentrations were highest. In contrast, the low concentrations begin at the confluence with Kwage Canyon and include reaches that have aggraded during the past several decades when plutonium concentrations were lowest. These data support the inference from morphologic changes along the channel that Kwage Canyon is a major source of clean sediment at present, 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. The relative amount stored in fine-grained versus coarse-grained sediment in each reach also displays significant longitudinal variations, as shown in Figure 14.

Upstream from Kwage Canyon (at west end of reach P-2 East), about 75 to 85% 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, and lowest in reach AC-3, with a steep bouldery channel (Figure 14). Inventories are similar in reaches AC-3, P-1 East, and P-2 West, with the downstream decrease in plutonium concentrations (Figure 13) offset by a downstream increase in sediment volume (Figure 5b).

Between Kwage Canyon and the head of incision in lower Pueblo Canyon (upstream end of reach P-4 West), total inventories are relatively low and roughly equal amounts of plutonium are stored in the fine facies and the coarse facies (Figure 14). 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, in reaches P-4 West and P-4 East, the amount of stored plutonium increases dramatically, and about 70% of the estimated inventory there is contained within the coarse facies (Figure 14). This increase is due to both the occurrence of relatively high 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.

An estimate of the total inventory of plutonium in the Pueblo Canyon watershed was made using the data presented in Figure 14, and extrapolating to unsampled reaches. An estimated 1.1 Ci of plutonium is present in the watershed, as summarized in Table 3 and Figure 15. Of this amount, 23% is contained within Acid Canyon, 33% within Pueblo Canyon upstream from Kwage Canyon, 16% between Kwage Canyon and the head of incision, and 28% in the lower 2.2 km of Pueblo Canyon. An estimated 61% is contained within fine-grained sediment and 39% within coarse-grained sediment. Half is present along the first 4.9 km of channel and half along the lower 6.8 km.

Figure 15 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 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 results of this investigation indicate that the present distribution of plutonium in Pueblo Canyon reflects the combined effects of 1) the timing of plutonium 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. The net result is a complex pattern of plutonium distribution that displays both general spatial trends as well as considerable local variability, consistent with the conclusions of Graf (1996).

In Pueblo Canyon, there are strong temporal variations in plutonium concentration in deposits of 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 contemporaneous with or soon after peak releases, from the mid-1940s to the mid-1960s. At present, these deposits have an irregular, discontinuous distribution both within reaches and downstream from the source due to spatial variability in deposition during this period and subsequent erosion. These results are consistent with studies in other regions 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 concentrations and the particle size distribution of sediment layers is also consistent with expectations based on the geochemical behavior of plutonium and other actinides (Watters *et al.*, 1983) as well as previous field investigations (e.g., Nyhan *et al.*, 1976b). Data from this investigation indicates that plutonium released into Acid Canyon quickly became associated with fine-grained sediment particles, despite the small percentage of fines below the stream bed. The highest concentrations of plutonium were found in deposits on floodplains within 250 m of the source that had 50-70% silt and clay, indicating that within very short distances significant segregation of plutonium onto fine sediment particles had occurred, and that these fines were being quickly suspended within floodwaters and deposited in overbank settings.

Preferential deposition of fine-grained sediment on abandoned channels and floodplains results in the highest concentrations and much of the inventory of plutonium being found in these settings. Preferential storage of contaminants on floodplains in other areas 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 may be in part related to high transmission losses in this ephemeral stream system, accompanying infiltration of suspended sediment into the stream bed. The quantity of plutonium in coarse sand deposits is also related to substantial historic aggradation that led to the deposition of large volumes of bed load sediment and 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 other 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 particularly characteristic of many such basins.

Because of these similarities, we infer that the historic aggradation-degradation cycle in lower Pueblo Canyon records a significant increase in sediment supply in the watershed during or before the early 1950s. Although the source of the sediment is not certain, 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 semi-arid and arid watersheds elsewhere (e.g., Thornes, 1979).

The geomorphic history of Pueblo Canyon since initial contaminant releases, specifically the location and timing of aggradation and incision, strongly influences the present contaminant distribution. Different patterns of contamination would have resulted if releases corresponded to different parts of an aggradation-degradation cycle. Effective modeling of long-term contaminant transport in this system needs to incorporate both channel-floodplain interactions (e.g., Malmon *et al.*, 2002) and cycles of aggradation and degradation associated with temporal variations in sediment input (e.g., Pickup *et al.*, 1983; Kelsey *et al.*, 1987). Understanding the recent geomorphic history of Pueblo Canyon helps in the development of models that incorporate relevant processes, although the predictive ability of such models will be inherently limited by difficulties in predicting the timing and exact 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, especially in fine sediment. The occurrence of a particularly wet summer in 1952, the year after releases of untreated radioactive effluent ceased, may have allowed a large part of the plutonium inventory which had accumulated in the stream bed to be rapidly distributed downstream. The large volume of coarse sediment deposited in lower Pueblo Canyon in the 1950s contains some of this plutonium, but correlative deposits of fine sediment are relatively small.

Although the relative amounts of fine and coarse sediment transport in flash floods in Pueblo Canyon have not been estimated, field measurements and sediment transport modeling in the adjacent watershed of upper Los Alamos Canyon indicate larger amounts of suspended load than bed load transport in a flood (Malmon, 2002). This work also indicates that less than 10% of the suspended sediment in a flood passing through a reach is deposited on adjacent floodplains. In combination, these lines of evidence suggest that much of the suspended sediments and associated plutonium from floods in the 1950s was transported out of Pueblo Canyon. Investigations in lower Los Alamos Canyon, downstream from Pueblo Canyon, have located only relatively small amounts of plutonium in young sediment deposits (~0.1 Ci; Reneau *et al.*, 1998a), in turn suggesting transport directly to the Rio Grande. Earlier modeling by Lane *et al.* (1985) in Pueblo Canyon led to a similar conclusion that a relatively small percentage of the sediment yield in a flood was deposited on floodplains, and that much more plutonium has been transported out of Pueblo Canyon than remains in storage.

These conclusions contrast with those of Graf (1994, p. 150), who estimated that only about 10% of the plutonium had reached the Rio Grande by 1985. His estimate of an average of 4.8 mCi per year entering the river from 1948 to 1985, or 182 mCi total, incorporated assumptions that suspended sediment transport of plutonium was not significant and that plutonium concentrations in active channel sediment since 1970 were representative of earlier years. Data from this investigation indicate that both of these assumptions should be revised and that larger amounts of plutonium have reached the Rio Grande. An improved estimate of the quantity of plutonium that has reached the river should incorporate both transport in suspended

sediment and higher concentrations in both bed load and suspended load sediment prior to 1965, and our data suggest that the estimate of Graf (1994) may be an order of magnitude too low.

CONCLUSION

Plutonium in the Pueblo Canyon watershed has a complex spatial distribution resulting from 55 years of fluvial transport subsequent to the initial releases of radioactive effluent in World War II. This study reinforces work in other fluvial systems that has elucidated the effects of geomorphic processes on contaminant distribution, including the effects of sediment mixing, particle size sorting, aggradation, and degradation. About 1.1 Ci of plutonium is contained with about 360,000 m^3 of sediment, 60% within fine-grained sediment deposits and 40% within coarse-grained sediment deposits. The highest concentrations of plutonium occur in fine-grained deposits near the source that date to the period of active effluent releases, and concentrations are less in younger deposits, in coarser-grained deposits, and in deposits farther downstream. Potential environmental impacts are also highest near the source and have declined over time as a result of the progressive redistribution of plutonium. The temporal relation of effluent releases to an aggradation-degradation cycle along the channel strongly influences the current spatial distribution of plutonium, and a significant portion of the plutonium inventory in the watershed is contained within coarse-grained deposits in historic fill terraces that have been abandoned following channel incision. A larger amount of plutonium associated with suspended sediment in flash floods is inferred to have been transported out of the watershed, although the total released into the watershed is unknown. An understanding of the geomorphic controls on contaminant distribution provides a foundation for evaluating potential present day risks to human health and ecosystems, for evaluating the effects of continued contaminant redistribution, and for evaluating remediation alternatives.

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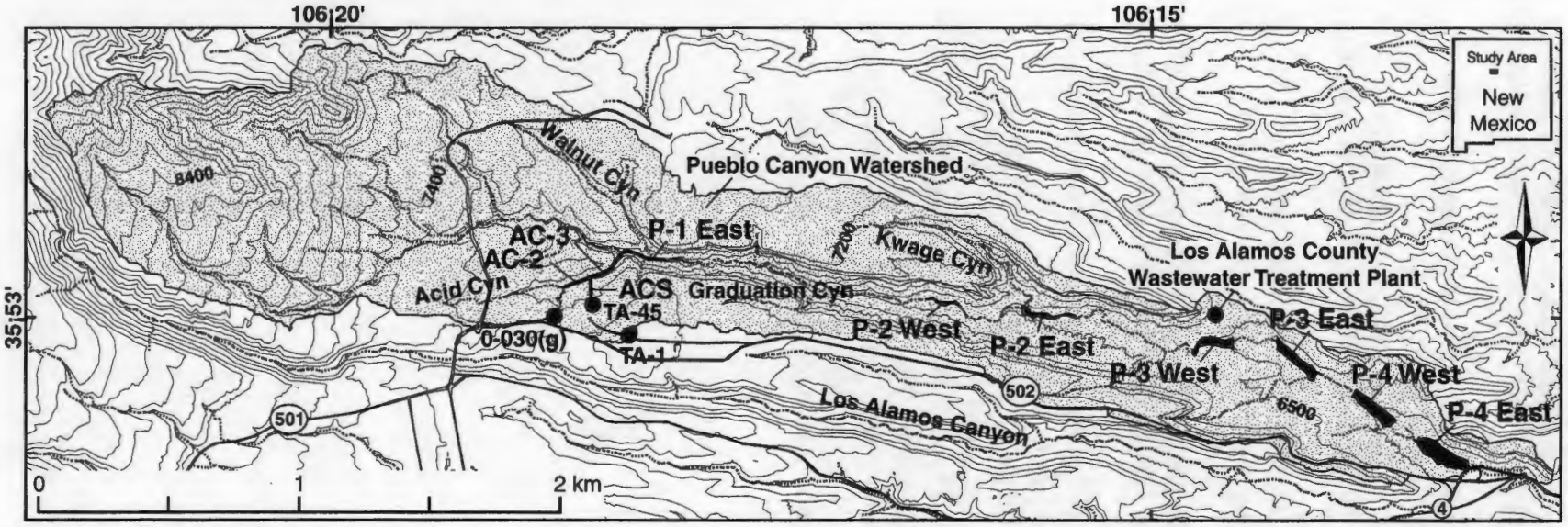

Figure 1. Map showing Pueblo Canyon watershed and locations of study reaches.

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

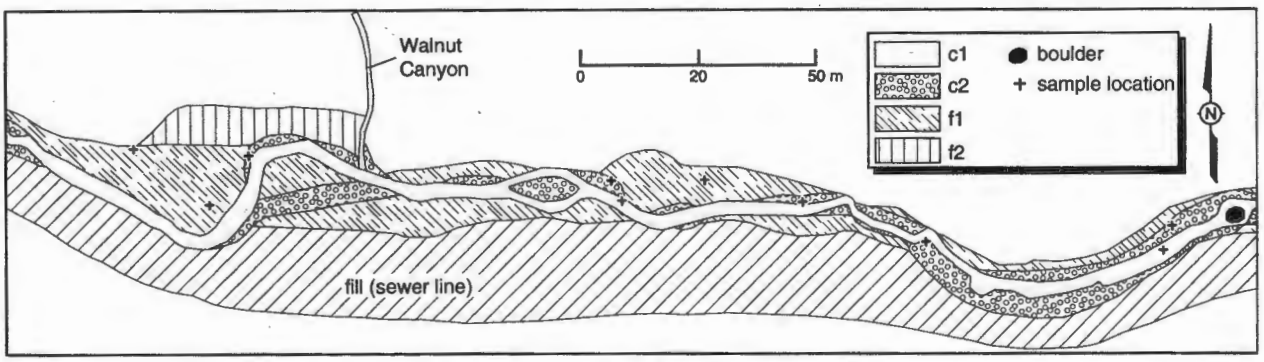

Figure 3. **Geomorphic map of the eastern half of reach P-1 East, showing distribution of post-1942 units and sample locations. "c1" indicates the active channel; "c2" indicates abandoned post-1942 channels; "f1" indicates post-1942 floodplains; "f2" indicates possible post-1942 floodplains.**

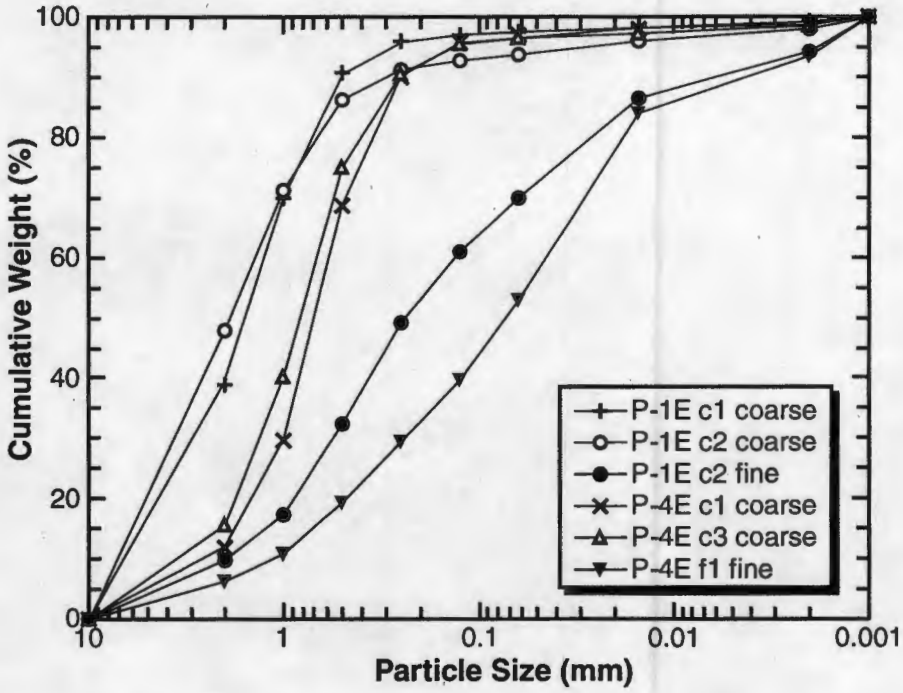

Figure 4. Examples of particle size distribution in coarse and fine facies sediment in Pueblo Canyon, illustrating differences between the steep, narrow western canyon (reach P-1 East) and the broader, gentler eastern canyon (reach P-4 East). Data represent averages of multiple samples in each reach, and individual samples are better sorted. No size distributions were obtained for the >2 mm fraction, and the coarse end of each distribution is arbitrarily set at 10 mm.

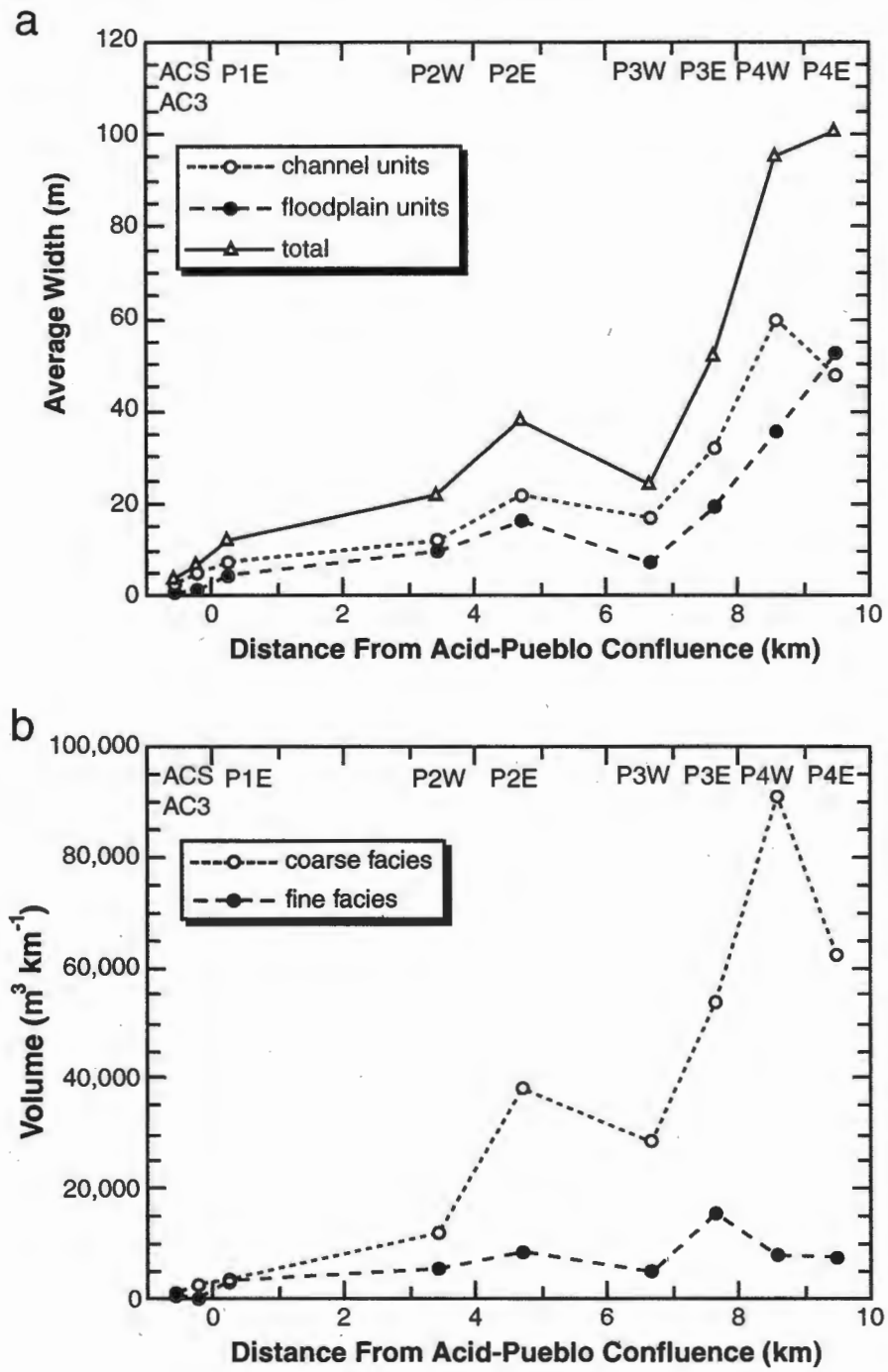

Figure 5. **Width and volume of post-1942 geomorphic units and sediment facies in Acid Canyon and Pueblo Canyon reaches downstream from the primary outfalls. a) Average width of channel and floodplain units, calculated as unit area divided by reach length. b) Estimated volume of coarse facies and fine facies sediment, normalized by reach length and expressed in units of m^3 per km of channel.**

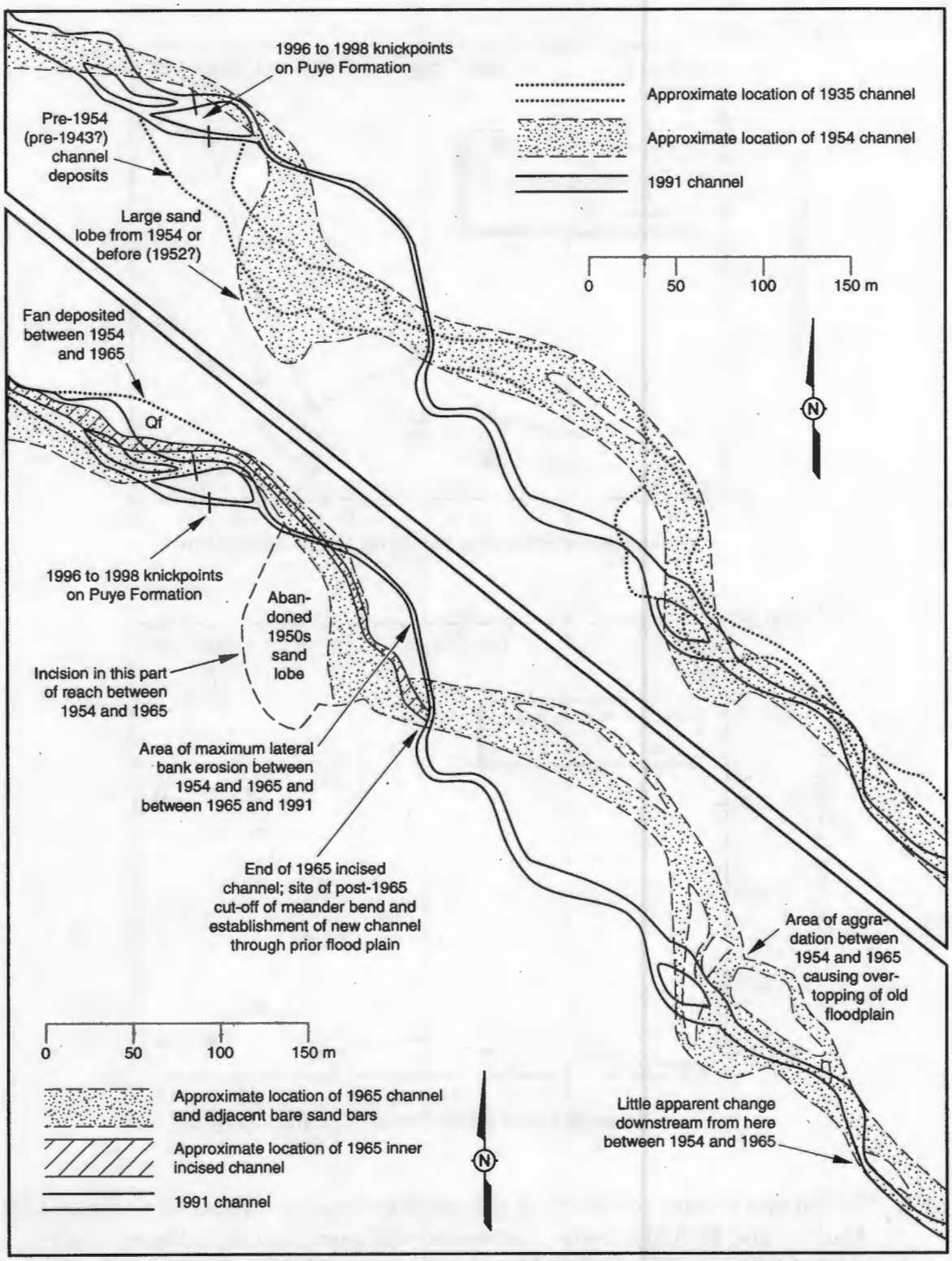

Figure 6. Maps of reach P-4 West, based on aerial photographs and field observations, illustrating key aspects of channel changes since 1935. Upper half of figure highlights channel in 1954, and shows channel location in 1935 and 1991 for comparison. Lower half of figure highlights channel in 1965, and shows channel location in 1991 for comparison.

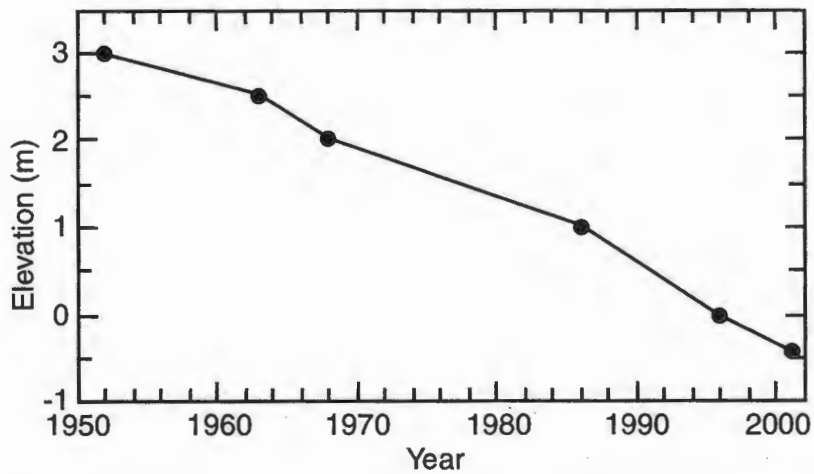

Figure 7. **Plot of elevation of channel in different years vs. approximate age in reach P-4 West, relative to channel elevation in 1996. Elevations from 1952 to 1986 are estimated from 1996 field survey of surfaces identified on aerial photographs. A re-survey in 2001 after large post-fire floods (Lyman *et al.*, 2002) indicates similar average incision rates as before the Cerro Grande fire.**

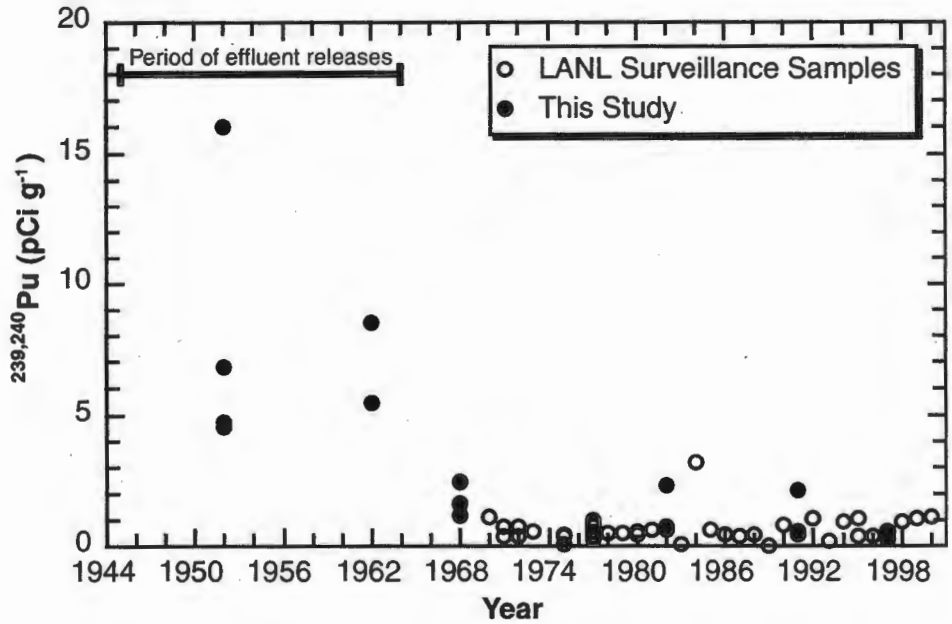

Figure 8. **Temporal variations in ^{239,240}Pu concentration in channel samples from lower Pueblo Canyon (reach P-4), 1952-2000. Data from LANL environmental surveillance reports (e.g., Rogers *et al.*, 2001), Stoker *et al.* (1981), and this study. Ages of samples between 1952 and 1991 in this study are approximate and are estimated from examination of sequential aerial photographs.**

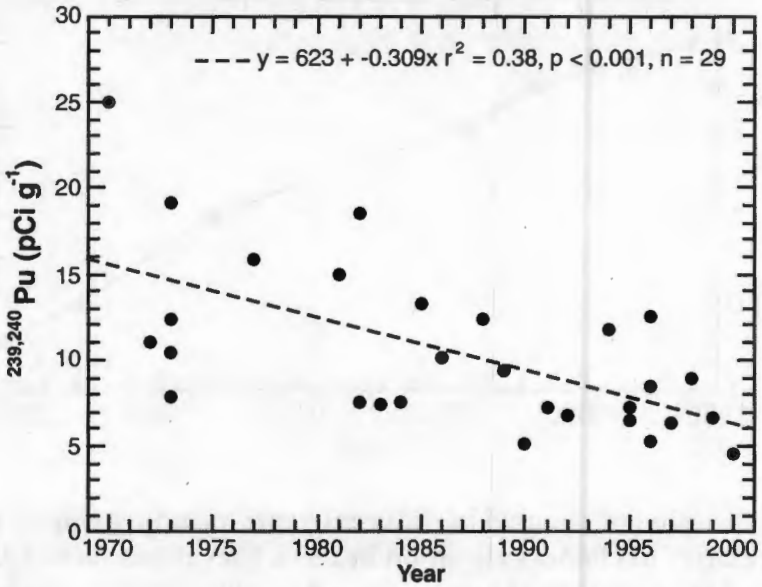

Figure 9. Temporal variations in $^{239,240}Pu$ concentration in active channel samples from lower Acid Canyon (reach AC-3), 1970-2000. Data from LANL environmental surveillance reports (e.g., Rogers *et al.*, 2001), Stoker *et al.* (1981), and this study. Three anomalous values <0.2 pCi g^{-1} are not included in plot (1987, 1993, 2000).

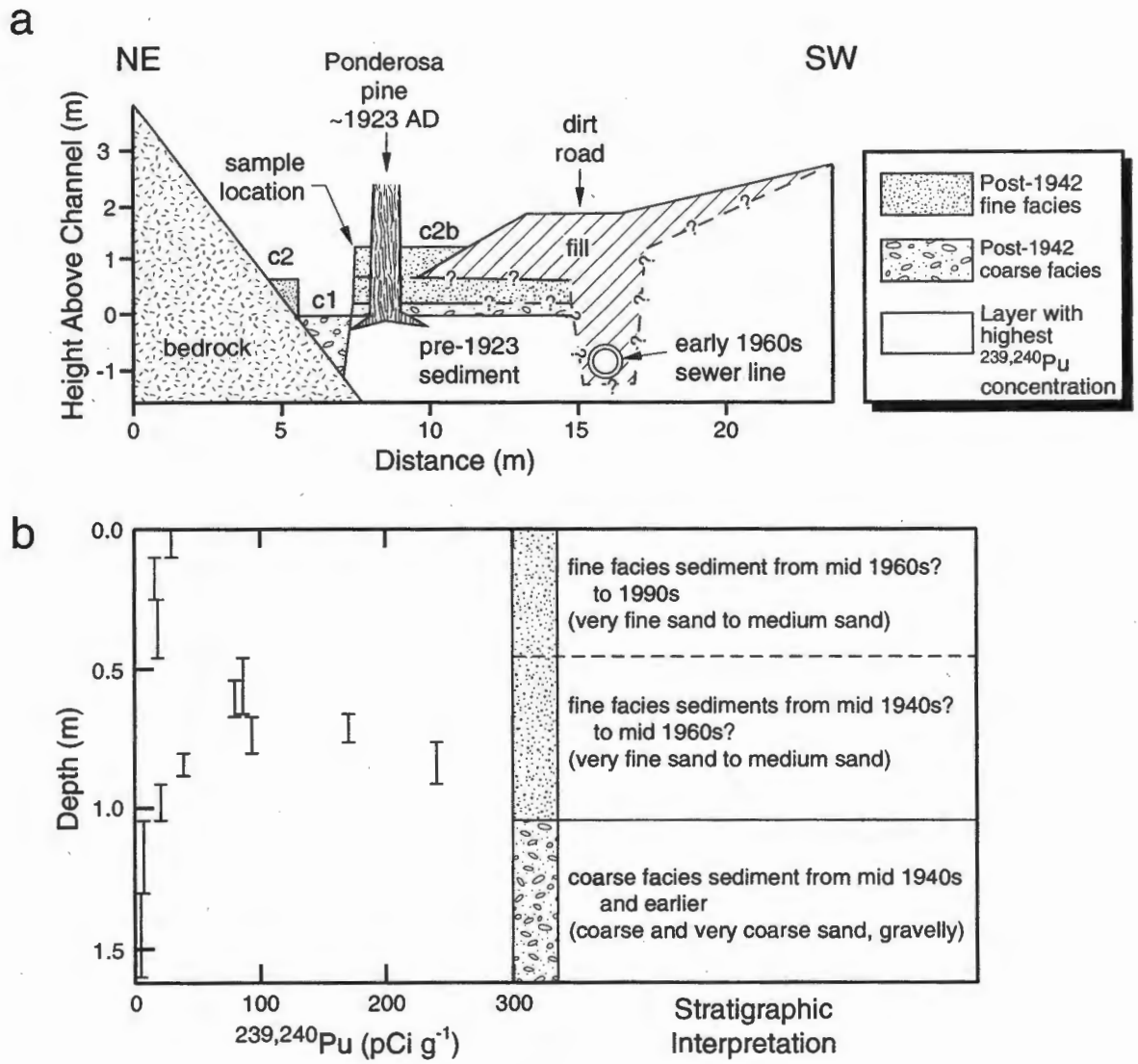

Figure 10. a) Schematic cross section across geomorphic surface in reach P-1 East with relatively high levels of plutonium (c2b unit). Subsurface layer with highest plutonium concentrations is highlighted. b) Depth variations in plutonium concentration below same surface, showing simplified stratigraphy.

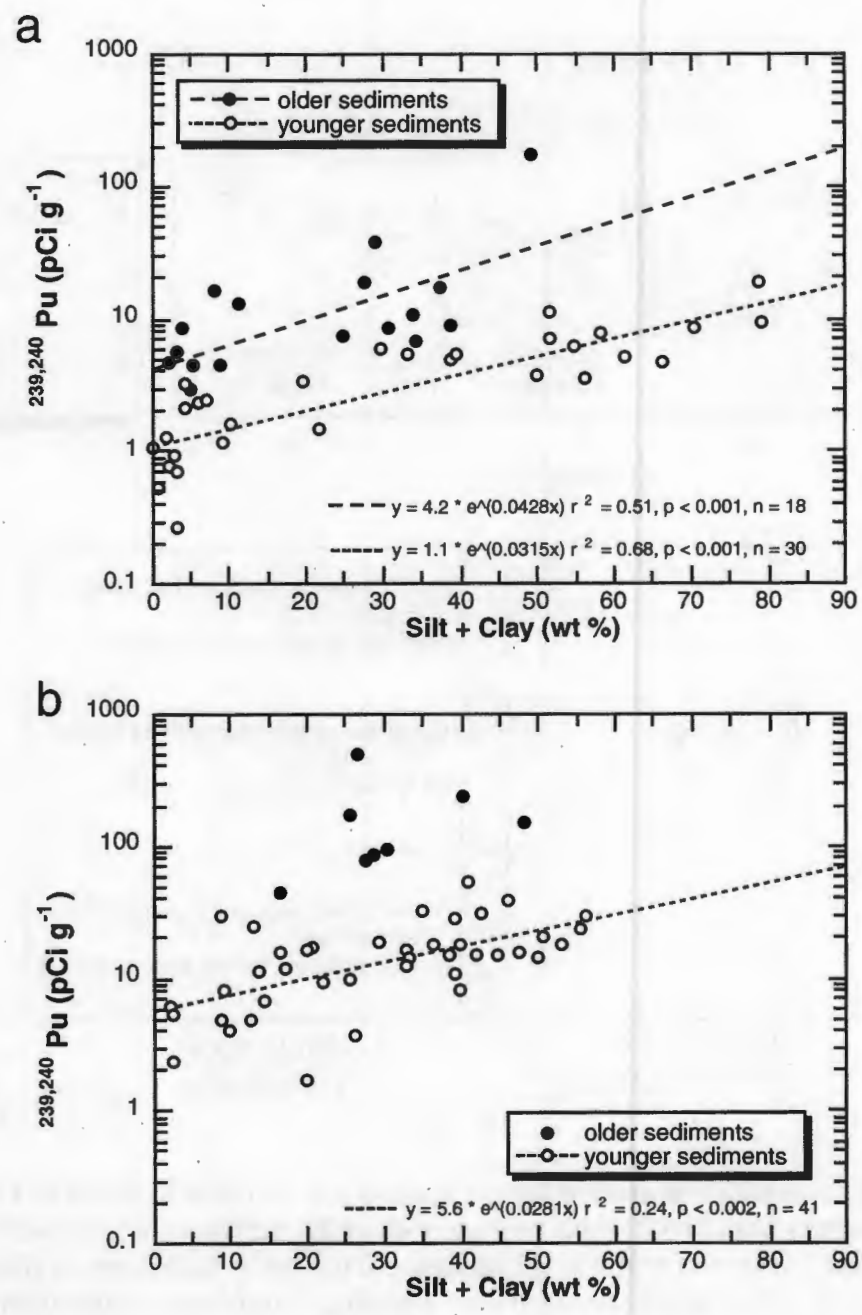

Figure 11. Scatter plots showing relations of plutonium concentration to percent silt and clay in Pueblo Canyon. a) Reach P-4. b) Reach P-1 East. Samples are separated into general age groups, with the older samples believed to date to the period of effluent releases and the younger samples post-dating 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. Linear regressions are not plotted for the older sediments in reach P-1 East because of the small sample size.

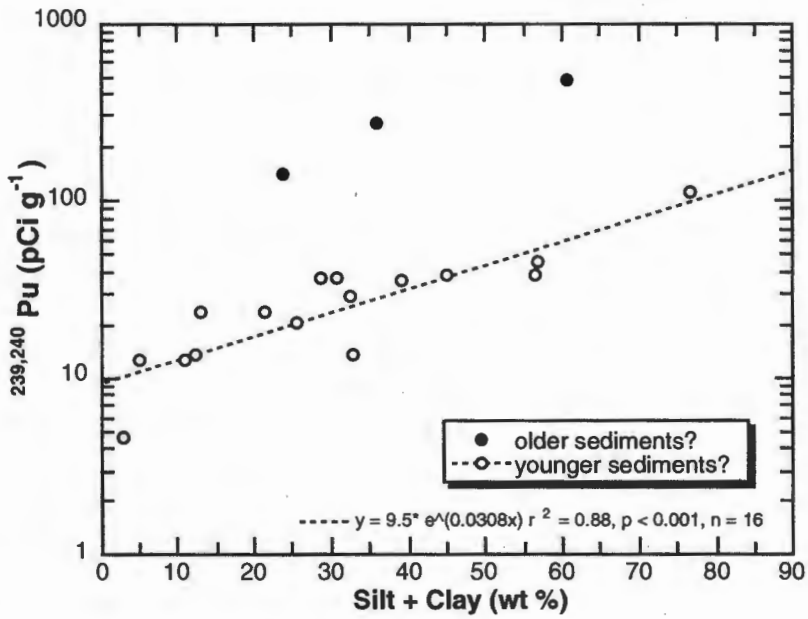

Figure 12. **Scatter plot showing relations of plutonium concentration to percent silt and clay in reach AC-3. Three outliers that are inferred to represent relatively old sediment layers are excluded from the regression. Samples that are inferred to pre-date peak contamination in this reach are excluded from the regression analysis.**

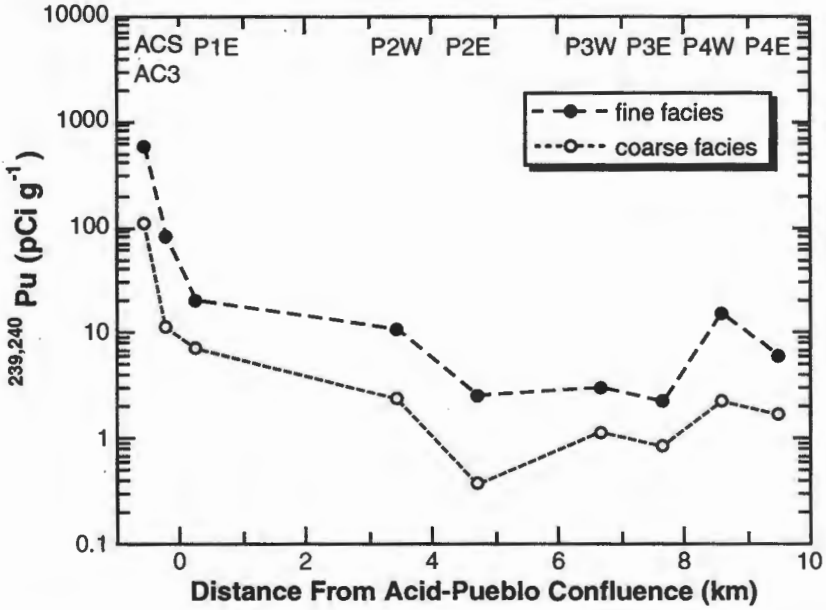

Figure 13. **Estimated average plutonium concentration in coarse and fine facies sediment in each reach. Values are volume-weighted averages that combine data on average plutonium concentrations in each geomorphic unit in each reach with the estimated volume of that unit. Reaches upstream from reach ACS not shown.**

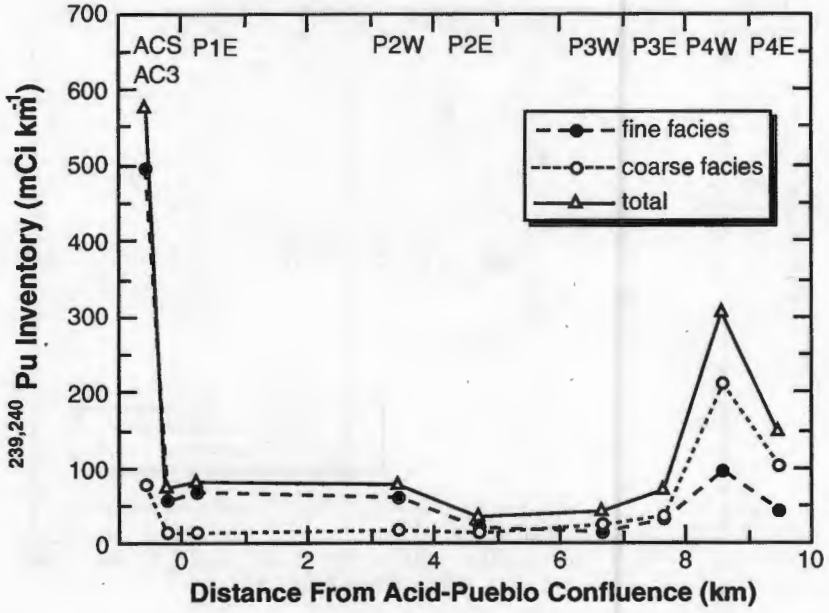

Figure 14. Estimated plutonium inventory in each reach, showing the portions in coarse and fine facies sediment. Values are normalized by reach length and shown in units of milli-curies per kilometer of channel (mCi km^{-1}). Reaches upstream from reach ACS not shown.

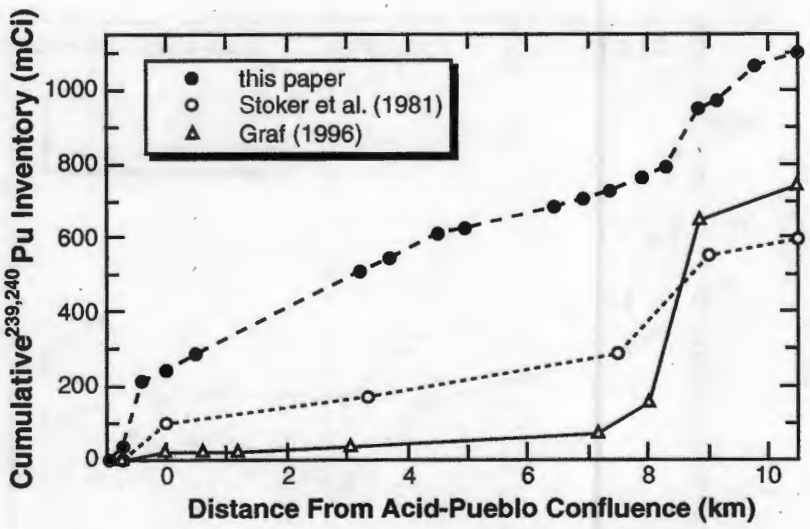

Figure 15. Plots of cumulative plutonium inventory vs. distance along channel, comparing estimates in Stoker *et al.* (1981), Graf (1996), and this study.

Table 1. Characteristics of Study Reaches

Reach	Reach Length (km)	Distance from Acid-Pueblo Confluence (km)[a]	Drainage Area (km^2)[b]	Average Channel Gradient ($m\ m^{-1}$)	Notes
0-030(g)	0.24	-0.94	<0.1	0.062	1st-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	1st-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

[a] 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 2 foot (0.61 m) contour intervals.

[b] Drainage area measured from upstream end of reach.

Table 2. Plutonium Concentrations in Fine-Grained Sediments

Sampling Reach	Estimated Age	Maximum $^{239,240}Pu$ (pCi g^{-1})	Average $^{239,240}Pu$ (pCi g^{-1})	Standard Deviation (pCi g^{-1})	Median Particle Size Class[a]
ACS	1945–1964[b]	7780	1120	1830	vfs
ACS	1965–2000[b]	205	97	61	vfs
P-1 East	1945–1964[c]	502	189	150	fs
P-1 East	1965–1997[c]	39	18	11	fs
P-4 West	1945–1964[d]	171	35	56	vfs
P-4 East	1965–1985[d]	19	6.6	3.9	csi
P-3 East	1991[d]	1.7	1.6	0.1	vfs

[a] Determined from averages of multiple analyses; csi: coarse silt; fs: fine sand; vfs: very fine sand.

[b] Sediments with highest $^{239,240}Pu$ concentrations are from geomorphic units that are inferred to be largely contemporaneous with effluent releases, and sediments in other units are inferred to largely postdate effluent releases.

[c] Sediments with highest $^{239,240}Pu$ 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 $^{239,240}Pu$ concentrations in nearby units are inferred to largely postdate effluent releases.

[d] Ages estimated from spatial relations with channel deposits dated using sequential aerial photographs.

Table 3. 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	29	6	35
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 head of incision	3.82	80	99	179
Pueblo Canyon below head of incision	2.18	97	214	311
Total	**11.72**	**674**	**425**	**1099**