Distribution of Radionuclides in Northern Rio Grande Fluvial Deposits near Los Alamos National Laboratory, New Mexico

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2007

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April 2007

Acknowledgements

The New Mexico Environment Department gratefully acknowledges the following government agencies, groups and individuals for their time, effort and contribution to the completion of this report. The Santa Clara Pueblo Office of Environmental Affairs, the DOE Los Alamos Site Office, Doug Stavert and Bruce Gallaher of Los Alamos National Laboratory, John Young of NMED's Hazardous Waste Bureau, Cheryl Overstreet and Rich Mayer of USEPA Region 6, and Chris Hanlon-Meyer, Steve Yanicak, and Ralph Ford-Schmid of NMED's DOE Oversight Bureau. Review and comments from Gene Turner from the DOE Los Alamos Site Office, and Rich Mirenda, Steven Reneau, David Rogers, and Mike McNaughton from the Los Alamos National Laboratory improved our report.

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Abstract

The New Mexico Environment Department's Department of Energy Oversight Bureau conducted a five year study to identify radionuclide contamination originating from the Los Alamos National Laboratory (LANL) in abandoned channels, old flood plains, and other fluvial deposits along the Rio Grande. The highest proportions of LANL contaminants in sediments are nearest to the discharge sources. Sediment sorting by fluvial processes throughout the past 60 years contributed unexpected contaminant concentrations in White Rock Canyon.

We collected sediments from multiple depth intervals in cores and outcrops at five sites along the Rio Grande and from within the Rio Grande Channel at 8 sites. We evaluated radiochemical measurements statistically, by comparing the data to historical background values, by investigating grain size distribution and contaminant concentration relationships, and using plutonium atom ratios to identify contaminant sources.

We selected a site at Santa Clara Pueblo ~12 km upstream of the Los Alamos Canyon and Rio Grande confluence at Otowi Bridge to demonstrate background conditions. The remaining four sites are downstream of the Otowi Bridge. Cañada Ancha is ~five km downstream, while the Pajarito and Water canyon sites are about 11 and 14 km below the bridge, respectively. The Frijoles Site, farthest downstream, is approximately 19 km below the bridge.

Eight channel sediments were collected from the Rio Grande including locations 61 km (38 miles) north of the Otowi Bridge at Pilar to 134 km (83 miles) south of the Otowi Bridge to Albuquerque. Three Rio Grande channel locations were selected upstream of the Los Alamos Canyon and Rio Grande confluence. They reflect the upper Rio Grande watershed. Five sites are downstream of the Otowi Bridge and reflect areas downstream of the Laboratory.

Most of the LANL legacy contaminants in Rio Grande bank sediments were derived from the Los Alamos watershed. We found that $^{239/240}$plutonium was the most persistent radionuclide found in terraces downstream of LANL. By far, the largest concentrations were found at the Cañada Ancha site followed by the Frijoles site, and then the Water Canyon site. Elevated ^{137}cesium and uranium isotope concentrations were also found at Cañada Ancha, followed by the Frijoles Site. Strontium-90 was found to be elevated at the Cañada Ancha site and ^{241}americium was elevated at the Frijoles site. Contaminant measurements at the Pajarito site were all indistinguishable from background, although we identified legacy contaminants at levels diluted below the existing background references.

Rio Grande channel sediments contain plutonium and ^{137}cesium radionuclides with concentrations similar to global fallout levels of these contaminants observed in the upper Rio Grand watershed.

Introduction

From 1998 to 2004, the New Mexico Environment Department (NMED) DOE Oversight Bureau (DOE OB) conducted a study to evaluate historical Los Alamos National Laboratory (LANL, or the Laboratory) impacts to the Rio Grande. Figure 1 shows the Laboratory and the study area between Santa Clara Pueblo and Frijoles Canyon. We collected sediment samples from post-1940s deposits located within White Rock Canyon and had them analyzed for radiochemical contaminants. We evaluated the potential Laboratory impacts to White Rock Canyon by: 1) comparing individual radiochemical concentrations in sediments to regional background reference values, 2) statistically comparing up-gradient and down-gradient cases (groups of measurements), and 3) identifying the origin of plutonium in sediments using thermal ionization mass spectrometric (TIMS) analysis.

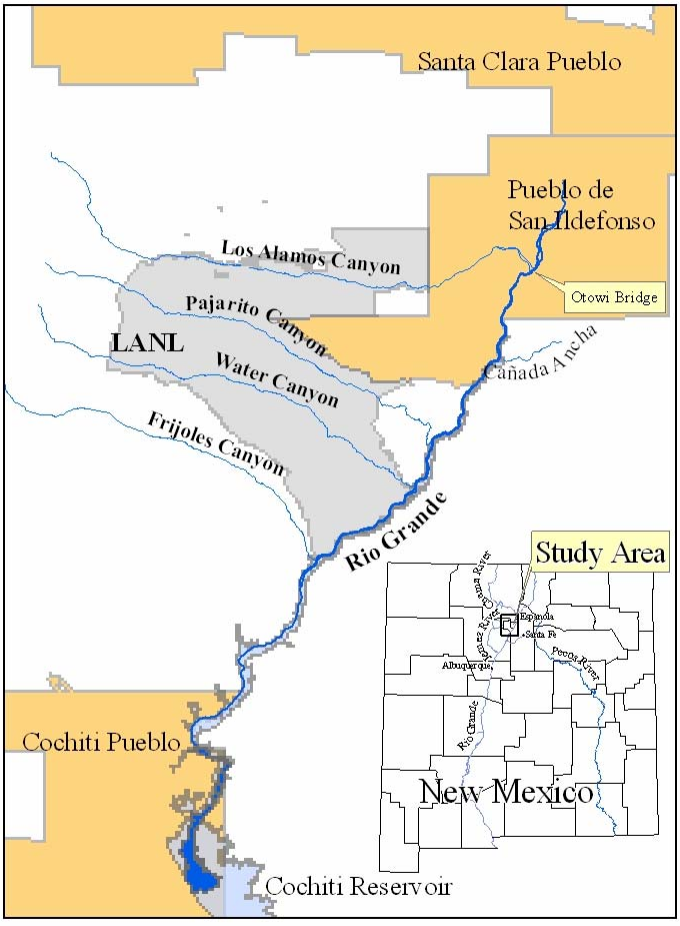

Figure 1. Study area along the Rio Grande

During the Manhattan Project through the Cold War years, the Laboratory discharged radioactive-liquid effluent derived from nuclear research into canyons within the facility. Acid and DP canyons, within the Los Alamos watershed, received much of these wastes. The Laboratory is situated on the Pajarito plateau, dissected by east-to-west oriented canyons that drain to the Rio Grande in White Rock Canyon.

From 1943 to 1986, radioactive-liquid effluent derived from Laboratory operations was discharged into DP and Acid canyons, tributaries to Los Alamos and Pueblo canyons, respectively. These canyons make up approximately 37% of the Los Alamos watershed. Guaje Canyon and its tributaries make up the remaining 62% of the watershed. Los Alamos and Pueblo canyons drain ephemerally to the east across the Laboratory to the Rio Grande. Los Alamos Canyon joins the Rio Grande at the Otowi Bridge about 6.4 km (4 miles) downstream or east of the Laboratory boundary.

The effluent discharges from LANL operations contained man-made radionuclide, heavy metal, and organic contaminants. Through time, storm runoff carried contaminants downstream to Pueblo and Los Alamos canyons and the Rio Grande, where they are stored in sediment deposits. Contaminant concentrations diminished with time and distance from the discharge sources, although relatively large radionuclide inventories remain in these canyons. These sediment deposits, within the Pueblo and Los Alamos drainages, provide the current contaminant sources available for transport.

Prior to completion of the Cochiti Dam in December 1973, sediments containing these wastes were transported, mixed with clean sediments, and deposited within the Los Alamos watershed channels and floodplains, as well as along the Rio Grande from the Otowi Bridge to Elephant Butte Reservoir. Elephant Butte Reservoir is located approximately 280 kilometers (174 miles) south of Los Alamos. After the close of the dam, deposition was primarily restricted between the Cochiti Reservoir and the upstream contaminant sources.

Insoluble contaminants, including plutonium, in waste discharges tend to adhere to soil and sediment particles, and are transported in suspended load and bedload during runoff. With each runoff event, contaminated sediments are carried progressively further downstream from the source term. These sedimentary deposits vary in age, physical character, spatial distribution, contaminant inventory and concentration, and susceptibility to remobilization. Susceptibility to re-mobilization is controlled by many factors including flood magnitude and frequency, and stream-channel stability.

In May 2000, the Cerro Grande fire severely burned the upper regions of the Los Alamos watershed as well as many other watersheds draining the Pajarito Plateau. In terms of contaminant source and transport, three significant conditions developed: 1) the ash contained elevated concentrations of man-made radionuclides as well as heavy metals, 2) stormwater runoff mobilized and transported the ash downstream, and 3) an increase in the frequency and magnitude of flooding destabilized channels draining the burned watershed and accelerated the erosion of older post-1945 contaminant-laden sediment

deposits downstream of the outfall source areas. These deposits were relatively stable prior to the fire.

Sedimentary deposits along White Rock Canyon below Otowi Bridge contain man-made radionuclides from three sources: 1) Laboratory legacy waste, 2) global fallout, and 3) contaminants concentrated in ash that may contain both Laboratory and fallout contaminants. Sediments deposited along the Rio Grande upstream of Otowi Bridge and after atmospheric testing of nuclear devices contain radionuclides derived from global fallout. Reference background values were derived for these contaminants from historical measurements of sediments and soils collected in Northern New Mexico.

Laboratory impacts are often determined by comparing the measured contaminant concentrations found in environmental samples to background reference values for radionuclides attributed to global fallout. Global fallout contaminants are associated with above ground nuclear weapons testing, re-entry and burn-up of plutonium-fueled satellites, and nuclear reactor accidents. In 2002, McLin and Lyons determined background references through statistical analyses of regional monitoring data collected from the 1974's to 1997 (McLin and Lyons, 2002).

Monitoring of radionuclides in runoff water and sediment entering the Rio Grande from the Los Alamos watershed by the Laboratory first began in 1958 and continues today. Data collected prior to the Cerro Grande fire show that contaminant concentrations in runoff had decreased through time, especially after effluent discharges were terminated in 1986.

William Graf (1993) published a report describing plutonium inputs and deposition in the Northern Rio Grande fluvial system. Through analysis of aerial photography and field investigations, he mapped sedimentary units that were deposited during the years of maximum plutonium contributions to the Rio Grande. He determined the age and spatial distribution of historical deposits, and whether they were channel or floodplain sediments. We identified post-1945 deposits using his maps.

During the mid- to late-1990s, the Laboratory performed detailed characterizations of sedimentary deposits in the Los Alamos watershed. They identified contaminant concentrations, distributions, and inventories of plutonium and other radionuclides along representative reaches in Los Alamos and Pueblo canyons. The characterization shows contaminant concentrations and distributions vary as a function of, but are not limited to, the distance downstream of the source, and the age and type of deposit. Higher contaminant concentrations tend to be associated with fine-grained flood-plain deposits near the contaminant source areas that were deposited when Laboratory operational releases were occurring (Reneau and others, 1998 a, b, and c).

The United States Geological Survey (USGS) sampled and analyzed Cochiti Lake sediments in 1996. Wilson and Van Metre (2000) presented these data and discussed their conclusions in a report completed in 2000. The USGS data set for man-made radionuclides for $^{239/240}$plutonium ($^{239/240}$Pu), tend to decrease upward in the sedimentary

sequence; meaning that older deposits in the lake have higher activities than younger shallower deposits. Their study also shows that ^{238}plutonium (^{238}Pu) and $^{239/240}$Pu were detectable at levels greater than background for Northern Rio Grande reservoir sediments (Wilson and Van Metre, 2000 in McLin and Lyons, 2002).

Additional studies by Gallaher and others (1999) and Gallaher and Efurd (2002) identified the source of plutonium and uranium (U) in sediments collected from reservoirs in the Rio Grande watershed, in the Rio Grande, and in drainages of the Pajarito Plateau. The studies used the analytical technique TIMS that discerned Laboratory derived plutonium from global fallout and natural uranium from enriched or depleted uranium. They showed that: 1) measurable amounts of Laboratory derived plutonium exist in stream and reservoir sediments of the Rio Grande downstream of the Laboratory, 2) approximately one-half of the plutonium in Cochiti Lake is from the Laboratory; and 3) on few occasions, enriched or depleted uranium was measured in stream or reservoir sediments downstream of the Laboratory. Natural uranium was commonly used and discharged during Laboratory operations, and is not usually discernable from natural uranium in the environment.

The objectives of this study are to determine if:

1) activities of the man-made radionuclides: ^{241}Am, ^{137}Cs, $^{234, 235, \text{and } 238}$ uranium (^{234}U, ^{235}U, and ^{238}U), ^{90}Sr, ^{238}Pu, and $^{239/240}$Pu in Rio Grande sediments located along the White Rock Canyon exceed background reference values;
2) plutonium and other radionuclide concentrations in sediments downstream of Otowi Bridge are statistically different than in upstream sediments;
3) the Laboratory contributed plutonium to these sediment deposits, and if so, estimate its contribution; and
4) the cumulative radiological risk is different between each sample location, and whether risk evaluations can identify LANL radiological contributions to the Rio Grande fluvial system.

These objectives were met by: a) comparing each measurement in our data set to regional background values published by McLin and Lyons (2002); b) statistically comparing our downstream data to our upstream data, and to McLin and Lyon's background data sets; c) conducting TIMS analysis on select samples; and d) utilizing the Environmental Protection Agency's (EPA) Superfund Preliminary Remediation Goal (PRG) for Radionuclide Risk Calculator.

Preliminary data assessments show that $^{239/240}$Pu concentrations are associated with ^{241} americium (^{241}Am), ^{137}cesium (^{137}Cs), and uranium isotopes concentrations. The correlation coefficients, R^2, range from 0.53 to 0.75. Strontium-90 (^{90}Sr) and ^{137}Cs showed a correlation of R^2 equal to 0.67. These correlations suggest that many of the contaminations discussed in this report may be co-located. We also used the opportunity to measure plutonium through different analytical laboratories using alternative analytical methods to provide additional quality control for our data. These plutonium measurements also allowed us to investigate contaminant affinities to grain size

variations. Although plutonium is the predominant contaminant found in the Rio Grande fluvial deposits described in this report, we found it contributed less than 1% of the cumulative health risk at one of the sites in this study found to be the most contaminated.

Sampling Sites

Sediments were collected at multiple depths along the Rio Grande banks and from the channel bottom surface in the Rio Grande to identify Laboratory contaminants entrained in floodplain, terrace, and channel sediments downstream of Los Alamos Canyon. Analytical data for sediments, collected for this study, from downstream of the Laboratory were compared to data from upstream sites. We also looked at historical sediment data to evaluate contaminant distribution in the northern Rio Grande Basin.

Thirty-two samples, as well as two duplicate samples, were collected from one outcrop and six coring sites during 1998 to 2001. Analytical laboratories analyzed the samples for ^{241}Am, ^{137}Cs, ^{243}Neptunium (^{243}Np), ^{238}Pu, $^{239/240}$Pu, ^{90}Sr, and ^{234}U, ^{235}U, and ^{238}U, and grain size. Eight fine grained slack-water condition samples were collected from the Rio Grande channel bottom and analyzed for ^{238}Pu, $^{239/240}$Pu, ^{137}Cs, and grain size (NMED, 2004). The analytical data are presented in Appendix A. The terrace and floodplain sites are located along the Rio Grande from the Santa Clara Pueblo to Frijoles Canyon. Figure 2 identifies the regional setting of the terrace and floodplain sites. We selected the locations based on the geomorphology and depositional ages described in a 1993 report written by William Graf (1993). Figure 3 identifies the channel sediment locations for this study. We collected channel fine grained sediments within the Rio Grande and Rio Chama at sites that demonstrated slack-water conditions.

Three coring sites are located on abandoned floodplains approximately 12.1 km (7.5 miles) upstream of the Laboratory at Santa Clara Pueblo, and are designated as the "background site or Santa Clara site". The three cores collected at the background site are located within about 400 meters of each other. See Figure 4 for the local setting along the Rio Grande at Santa Clara. This site includes pre-1941 sediments deposited before the Laboratory was established and nuclear atmospheric testing began, and post-1941 deposits that might contain only global fallout. One of the three cores represents a depositional period between 1941 and 1968; the other two cores represent deposition prior to 1941 (Graf, 1993).

Four sites are located downstream of the Laboratory, below the contaminant entry point at the Los Alamos Canyon and Rio Grande confluence – these sites are designated in this report as the "downstream sites". The sites are located on terraces and floodplains near west to east trending canyons that drain the Pajarito Plateau to the Rio Grande. From north to south, the sites are located near the mouths of Cañada Ancha, and Pajarito, Water, and Frijoles canyons. Figures 5 through 11 identify the sites.

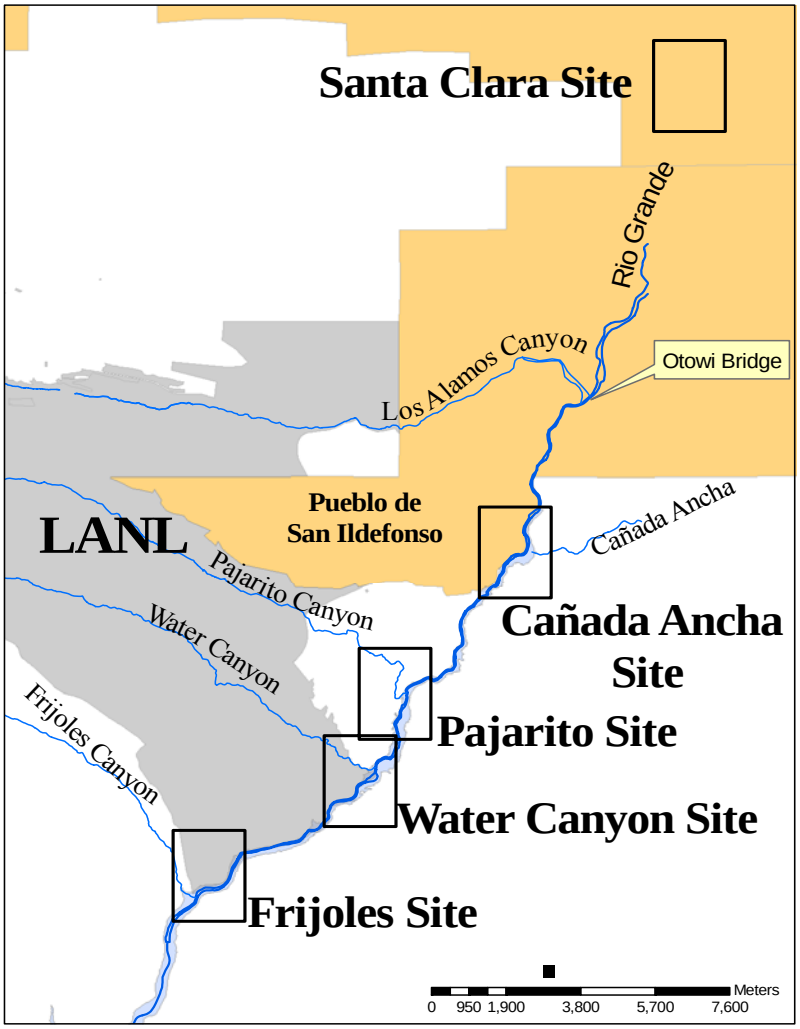

Figure 2. Core and outcrop sites along the Rio Grande

The site near Cañada Ancha is the most upstream location below the Los Alamos confluence with the Rio Grande. It is located on a floodplain that Graf (1993) had described as originating from a slough feature active during 1940 to 1958. A slough is a relatively inactive channel except during flooding. The slough was completely isolated from the main Rio Grande channel and filled with sediments during a 1967 flood (Graf, 1993).

The Frijoles site is located on a terrace that was pre-1950's floodplains, later buried by Cochiti Reservoir lacustrine deposits. This site is the most distant downstream location sampled for this project. Cochiti Reservoir construction was completed in 1973, and the maximum water levels were achieved between 1985 and 1988. Since then the water elevations have been reduced to current levels (Langman and Nolan, 2005). Maximum

water levels were as much as 15 meters higher than the present river level in the area, adding up to 1.5 meters of sediments.

The sites near Pajarito and Water canyons were field correlated to areas Graf (1993) described as floodplain and bar surfaces active until 1981. These sites are between the Ancha and Frijoles sites. The furthermost reach of Cochiti Reservoir may have extended to the Pajarito site during the mid 1980's. Downstream of Pajarito Canyon, at the Water Canyon site, lacustrine deposits are evident, indicating Cochiti Reservoir extended at least to this point.

Figure 3 shows the eight channel bottom sample sites along a 195 km (121 mile) reach of the Rio Grande. The samples were collected in slack-water conditions in order to select fine grained sediments transported in the river. The farthest north location is at Pilar, 61 km (38 miles) north of the Los Alamos Canyon confluence with the Rio Grande. A site was collected within the Rio Chama 5 km (3 miles) upstream of its confluence with the Rio Grande and 24 km (15 miles) above Otowi Bridge. The remaining sample upstream of the Los Alamos Canyon entrance to the Rio Grande is at the Rio Pojuaque. It is 3.8 km (2.4 miles) north.

The remaining five sites are downstream of the Rio Grande / Los Alamos confluence. Two sites are in the White Rock Canyon and the remaining three are below the Cochiti Reservoir. The White Rock Canyon locations are at Los Alamos Canyon 0.8 km (0.5 miles) and at Ancho Canyon 15 km (9 miles) downstream of Otowi Bridge. The locations below the Cochiti Reservoir are at Peña Blanca, San Angustura, and Albuquerque, 45, 73, and 133.6 km (28, 46, and 83 miles) south of Otowi Bridge, respectively.

These sample locations were generally selected behind large obstructions or in pools where water eddies and low energy flow systems allow fine grained sediments to selectively collect. The samples were collected from the surface of the channel bottom and represent active sediment deposition.

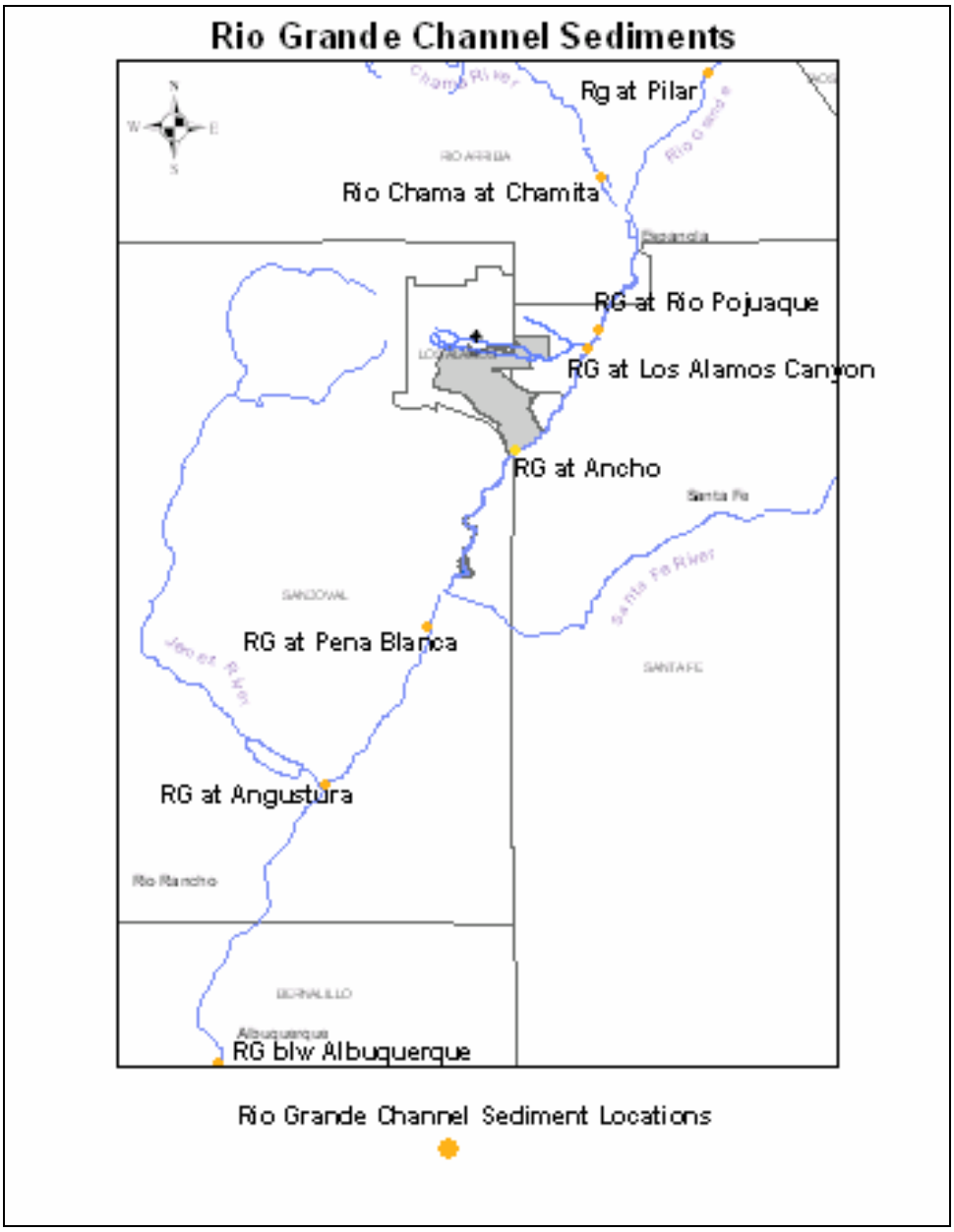

Figure 3. Channel sediments collected from the Rio Chama and Rio Grande

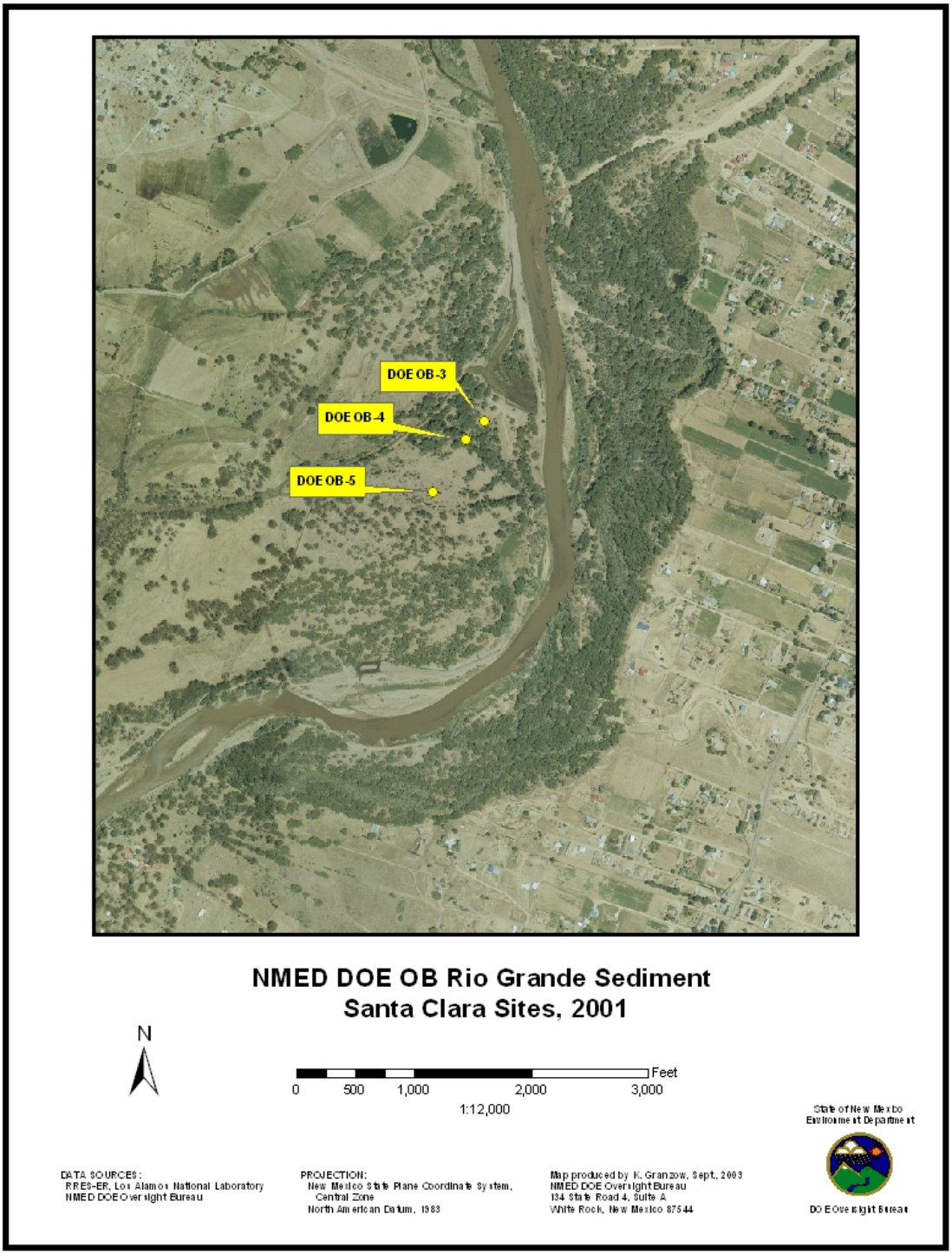

Figure 4. Coring sites at Santa Clara Pueblo

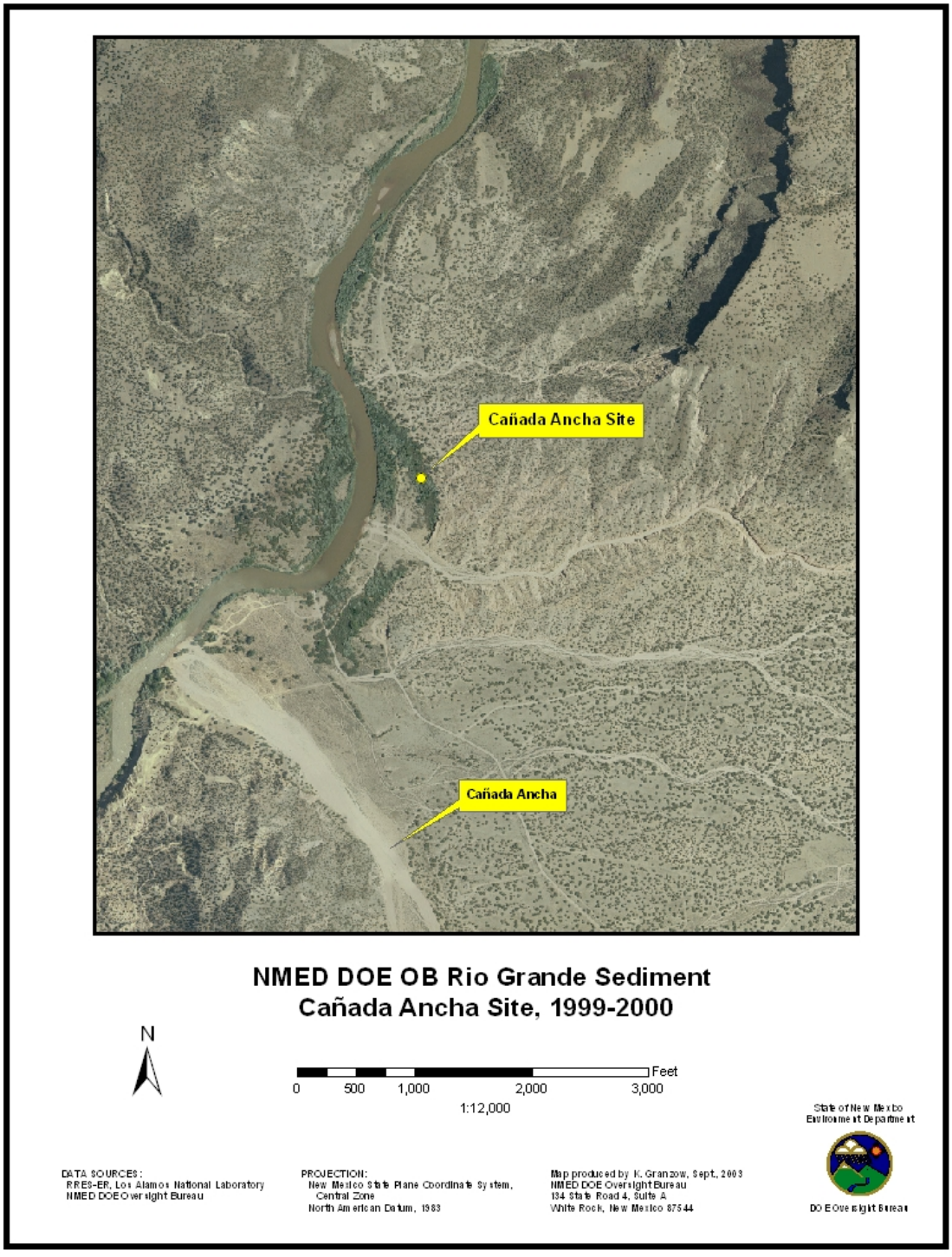

Figure 5. Core site at Cañada Ancha

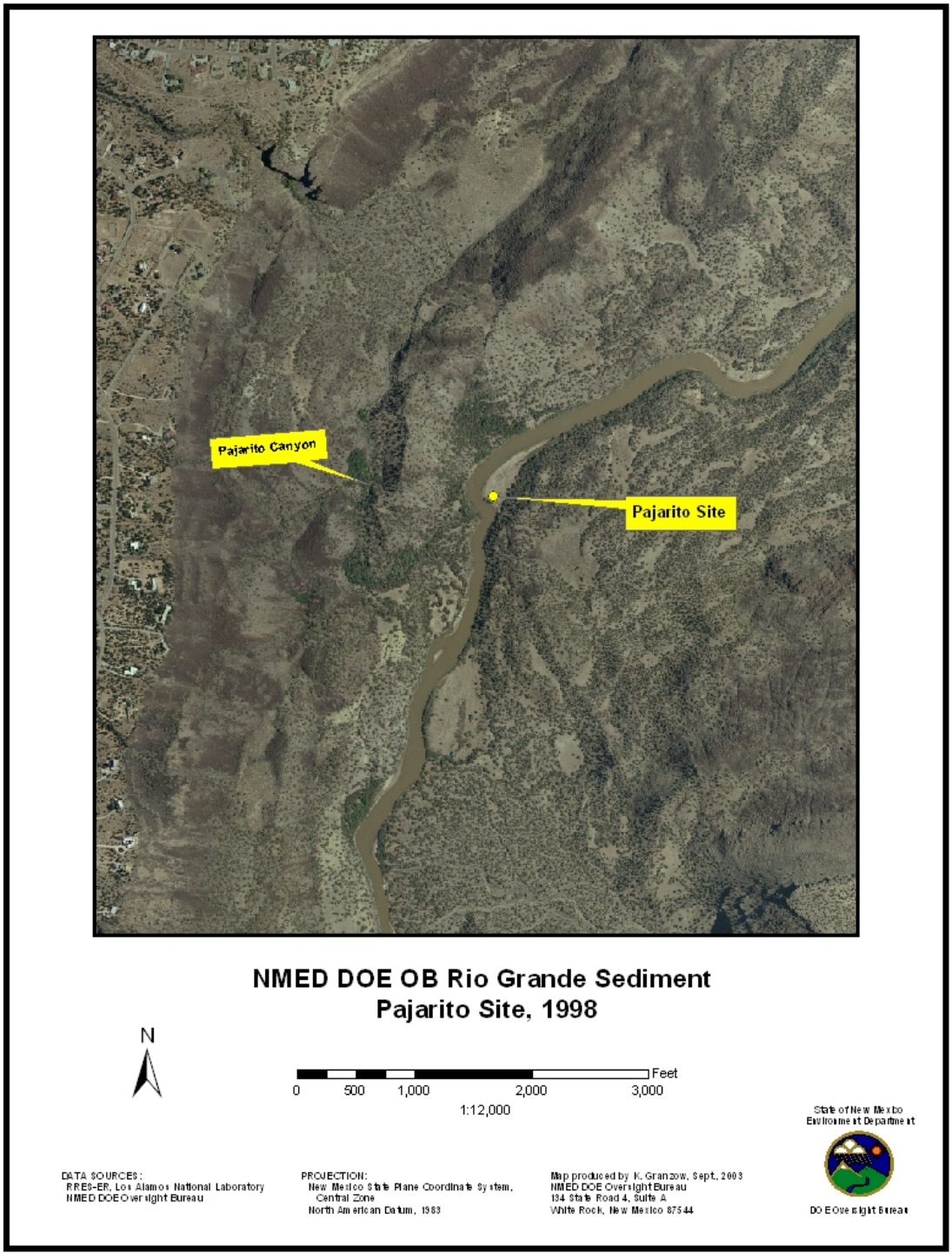

Figure 6. Coring Site at Pajarito Canyon

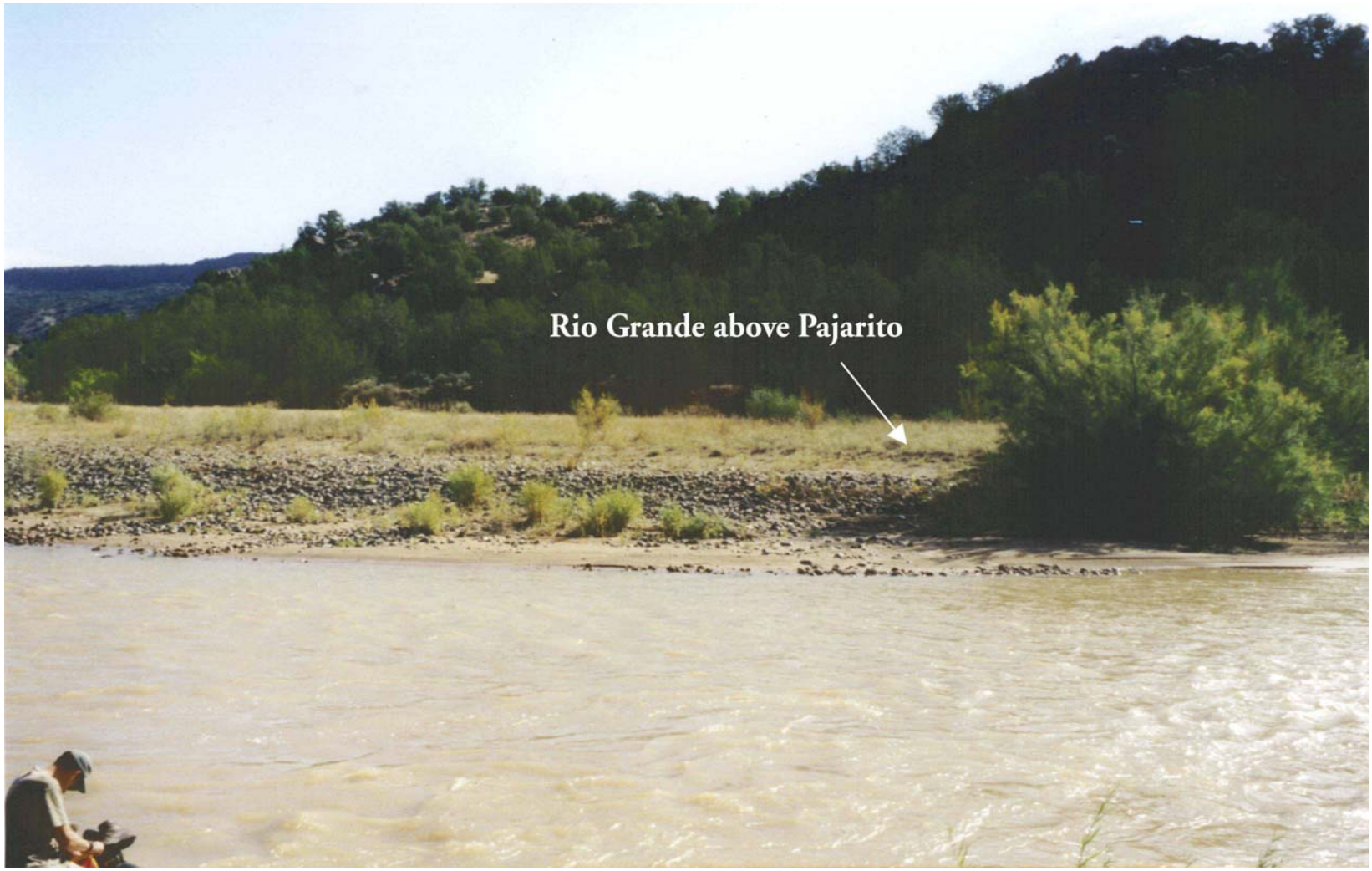

Figure 7. Coring Site on Rio Grande Near Pajarito Canyon

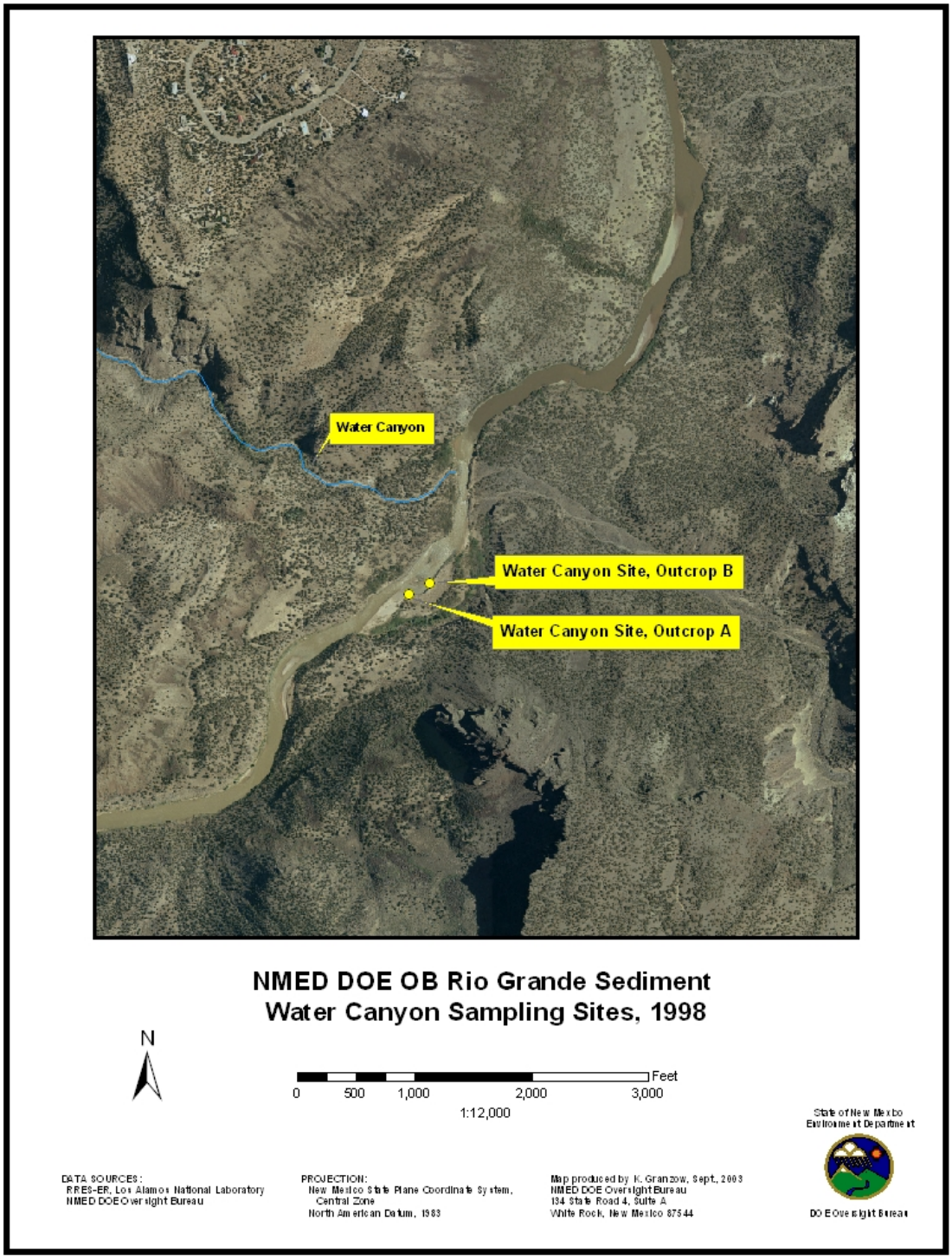

Figure 8. Coring sites at Water Canyon

Figure 9. Outcrop A on the Rio Grande near Water Canyon

Figure 10. Outcrop B on Rio Grande near Water Canyon

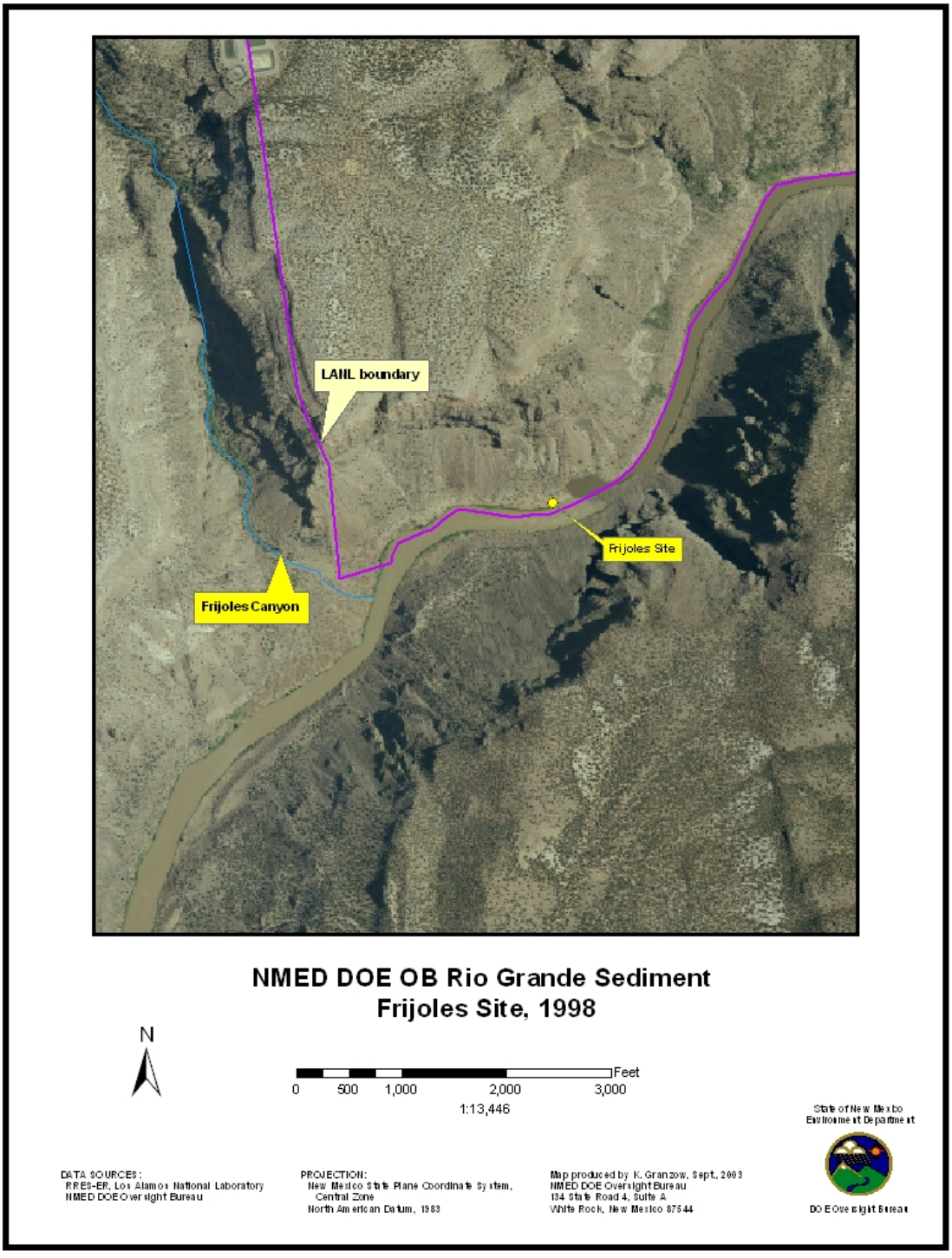

Figure 11. Core site at Frijoles Canyon

Data Evaluation

The following section describes the methods used to evaluate the data and discusses contaminant distribution along the Rio Grande. We compared individual radiochemical measurements to reference values that reflect background. In this case background refers to an environment not impacted from current anthropogenic activities. Then we made statistical comparisons of background data sets to our data set. We used TIMS to identify the source of plutonium measured in Rio Grande terrace sediments. Grain-size analysis and the correlations from those measurements and $^{239/240}$Pu activity concentrations provided additional insights regarding contaminant sources along the Rio Grande.

We found that $^{239/240}$Pu was the most persistent legacy radionuclide found in terraces downstream of LANL, although the distribution is not uniform. By far, the largest concentrations were found at the Cañada Ancha core site followed by the Frijoles site, and then the Water Canyon and Pajarito sites. Cesium-137 was also elevated by the greatest amount at Cañada Ancha, also followed by the Frijoles site. Strontium-90 was found to be elevated at the Cañada Ancha site and ^{241}Am was elevated at the Frijoles site. Anomalous uranium measurements were made at the Cañada Ancha and Frijoles sites as well. We identified sediments deposited prior to global fallout from atmospheric testing, followed by episodic deposition of LANL legacy waste in the Rio Grande, and then covered by floodplain materials containing mostly global fallout contaminants.

In the Rio Grande active channel, we found ^{238}Pu, $^{239/240}$Pu, and ^{137}Cs measurements in sediments to be similar to those in the upper Rio Grande watershed.

Appendix A contains all analytical results for this study.

Comparisons to Reference Values

We evaluated the potential Laboratory impacts in White Rock Canyon by comparing individual radiochemical concentrations in terrace sediments to LANL derived upper limits for background (BGULs) (McLin and Lyons, 2002). These reference values were derived from historical measurements of sediments collected in Northern New Mexico far beyond potential LANL impacts. If an individual measurement in an environmental sample is reported above its sample specific detection level and exceeds its reference value, assumptions are made that the contaminant is LANL derived.

A commercial analytical laboratory reported 32 measurements for ^{238}Pu, $^{239/240}$Pu, ^{241}Am, ^{237}Np, ^{234}U, ^{235}U, and ^{238}U, ^{90}Sr, and ^{137}Cs in terrace sediments discussed in this report. The Los Alamos Clean Chemistry and Mass Spectrometry Laboratory reported TIMS measurements for a subset of 10 terrace samples. The eight channel sediment samples were only measured for ^{238}Pu, $^{239/240}$Pu, and ^{137}Cs. Reference values do not exist for ^{237}Np, and uranium isotope measurements were compared to concentrations derived from the total mass reference reported as background.

We compared the measurements in our data set, excluding ^{237}Np, to LANL BGULs published in "*Background Radioactivity in River and Reservoir Sediments near Los Alamos, New Mexico*" (McLin and Lyons, 2002). McLin refined background estimates from a previous study by Purtyman and others (1987) to reflect upstream reservior sediments and upstream and downstream river sediments. These references were derived from measurements aquired between 1974 to 1997, collected in areas well beyond the potential influence of the Laboratory. They reflect upper tolerance levels, or the probable largest value that might be measured in background. The reference value for each parameter was determined at the 0.95 (two-tail) quantile of Mclin's data set.

All of the ^{238}Pu, $^{239/240}$Pu, and ^{137}Cs measurements in channel bottom sediments collected in the Rio Grande were reported below their specific BGUL.

The measurements in our remaining data set originate from multiple samples at different depth intervals collected at each of the five sites identified in the methods section. These sites include the upstream background site at Santa Clara, and four sites downstream of possible LANL impacts near Cañada Ancha, and Pajarito, Water and Frijoles canyons. Our evaluation identified potential LANL impacts at two sites, at the Ancha and Frijoles sites.

Our evaluation showed that $^{239/240}$Pu, uranium, and ^{137}Cs from more than one horizon at the Ancha site exceeded the LANL background values. It also showed that $^{239/240}$Pu and possibly uranium measurements from multiple horizons at the Frijoles site exceeded LANL background values. The exceptional values are presented in Table 1 adjacent to the reference values they exceed.

Six of 12 $^{239/240}$Pu alpha spectroscopy measurements at the Ancha site were greater than LANL's 0.013 pCi/g BGUL, and three of six measurements at the Frijoles site were greater. The exceptional values measured at Ancha site originate from horizons between 101 to 229 centimeters. Those at Frijoles site originate from three horizons between the surface and 152 centimeters.

Ten TIMS measurements were made on a subset of the preceding samples. TIMS measures the relative abundance of atoms for the plutonium isotopes, from which the activity concentrations can be calculated. Those measurements confirmed that four of the elevated plutonium measurements originally identified by alpha spectroscopy were greater than the BGUL, five of the values identified as being less than the BGUL were in fact less, and one sample at the Frijoles site at the 122 to 152 cm horizon may have been misidentified as an exceedance by alpha spectroscopy. That value was measured at 0.016 pCi/g by alpha spectroscopy and 0.008 pCi/g by TIMS. Further discussion of the TIMS measurements will be presented in a later section.

A number of uranium isotopes and ^{137}Cs measurements were also greater than their BGUL values at the Ancha site, although only three ^{235}U measurements exceeded its derived reference value at the Frijoles site. Isotopic uranium BGULs were derived from the total uranium mass referenced in Mclin and Lyons (2002) report, the relative.

Table 1. Data Table Highlighting Values that Exceed Background Reference Values

Station Description	^{238}Pu		$^{239/240}$Pu		^{241}Am		^{234}U		^{235}U		^{238}U		Total U		^{90}Sr		^{137}Cs	
	(pCi/g)	> 0.0087	(pCi/g)	> 0.013	(pCi/g)	>0.076	(pCi/g)	> 1.4	(pCi/g)	>0.07	(pCi/g)	>1.49	(mg/kg)	>4.49	(pCi/g)	>1.02	(pCi/g)	>0.56
Number of Stations that Exceed Reference		**none**		**9**		**none**		**2**		**6**		**2**		**2**		**none**		**4**
Santa Clara Sites																		
DOEOB 3																		
(abandoned floodplain, active 1941-1968)2B																		
5.18 - 30.5 cm (hand augered)	0.0006		0.0051		0.003		0.90		0.039		0.68		2.04		-0.01		0.102	
DOEOB 4																		
(abandoned flood plain, active prior to 1941)3B																		
33.5 - 39.6 cm (hand augered)	-0.0016		0.0008		0.003		0.90		0.044		0.91		2.73		0.12		<0.036	
73.2 - 88.4 cm (hand augered)	0.001		-0.00062		0.010		0.380		0.017		0.388		1.16		0.22		<0.027	
DOEOB 5																		
(abandoned floodplain, active prior to 1941)3B																		
24.4 - 39.6 cm (hand augered)	0.0007		-0.00022		0.008		1.14		0.060		0.98		2.94		0.07		<0.036	
57.9 - 73.2 cm (hand augered)	0.00000		0.0006		0.006		0.86		0.059		0.86		2.59		0.07		<0.033	
116 - 158 cm (hand augered)	0.00000		0.00000		-0.003		1.15		0.053		1.09		3.27		0.01		<0.041	
Cañada Ancha Site																		
(slough area during 1940-1958 or 1967) unit 3B																		
0 - 30.5 cm (hand augered)	0.0029		0.0022		0.0027		0.68		0.036		0.69		2.07		0.07		<0.093	
30.5 - 61 cm (hand augered)	-0.0013		0.0000		-0.0049		0.69		0.044		0.75		2.25		0.29		<0.13	
61 - 91.4 cm (hand augered)	0.0007		0.0017				0.78		0.036		0.78		2.34		0.36		<0.070	
91.4 - 101 cm (hand augered)	-0.0007		0.0033		0.0142		1.13		0.053		1.06		3.18		0.27		0.24	
101 - 110 cm (hand augered)	0.0056		0.067	**> 0.013**	0.026		1.96	**>1.4**	0.126	**>0.07**	1.99	**>1.49**	5.98	**>4.49**	0.36		0.60	**> 0.56**
110 - 125 cm (hand augered)	0.0013		0.0121		0.0041		1.25		0.070		1.20		3.60		0.31		0.27	
125 - 137 cm (hand augered)	0.0018		0.0262	**> 0.013**	0.016		1.28		0.071	**>0.07**	1.26		3.78		0.40		0.71	**> 0.56**
137 - 162 cm (hand augered)	0.0007		0.0430	**> 0.013**	0.019		1.41	**>1.4**	0.097	**>0.07**	1.55	**>1.49**	4.66	**>4.49**	0.64		0.98	**> 0.56**
162 - 186 cm (hand augered)	-0.0020		0.066	**> 0.013**	0.020		1.13		0.055		1.33		3.98		0.40		0.70	**> 0.56**
186 - 213 cm (hand augered)	0.00082		0.0144	**> 0.013**	0.0008		NA		NA		NA		NA		0.17		0.091	
213 - 229 cm (hand augered)	0.00122		0.0337	**> 0.013**	0.015		NA		NA		NA		NA		-0.06		<0.050	
323 - 335 cm (hand augered)	0.00131		0.0029		0.00030		NA		NA		NA		NA		0.11		<0.028	

Background Reference Used: McLin and Lyons, 2002 (BGULs)

Table 1 (continued). Data Table Highlighting Values that Exceed Background Reference Values

Station Description	^{238}Pu (pCi/g)		$^{239/240}$Pu (pCi/g)		^{241}Am (pCi/g)		^{234}U (pCi/g)		^{235}U (pCi/g)		^{238}U (pCi/g)		Total U (mg/kg)		^{90}Sr (pCi/g)		^{137}Cs (pCi/g)	
		> 0.0087		> 0.013		>0.076		> 1.4		>0.07		>1.49		>4.49		>1.02		>0.56
Pajarito Site																		
(active floodplain and bar surface)1B																		
0 - 30.5 cm (hand augered)	-0.0025		0.0029		0.0055		0.444		0.020		0.432		1.30		<0.17		<0.059	
30.5 - 45.7 cm (hand augered)	0.0017		0.0076		0.0100		0.73		0.063		0.72		2.17		<0.16		0.144	
Water Canyon Site																		
(active floodplain and bar surface)1B																		
0 - 30.5 cm (outcrop A)	-0.0013		0.0032		0.0030		0.63		0.042		0.65		1.95		<0.16		0.052	
30.5 - 61 cm (outcrop A)	-0.0032		0.0061		0.0062		0.60		0.028		0.53		1.59		<0.16		<0.064	
61 - 91.4 cm (outcrop A)	0.0006		0.0082		0.0108		0.73		0.033		0.76		2.28		<0.17		<0.12	
91.4 - 101 cm (outcrop A)	0.0025		0.0080		0.018		0.82		0.036		0.86		2.58		<0.15		0.132	
101 - 116 cm (outcrop A)	0.0015		0.0021		0.0002		0.72		0.040		0.73		2.19		<0.16		<0.064	
91.4 - 101 cm (outcrop B)	0.0015		0.0017		0.0130		0.75		0.046		0.85		2.55		<0.15		<0.096	
Frijoles Site																		
(pre-1950 floodplain deposits and reservoir sedimentation)																		
0 - 30.5 cm (hand augered)	-0.0012		0.0217	**> 0.013**	0.0184		1.22		0.075	**>0.07**	1.12		3.37		<0.15		0.128	
30.5 - 61 cm (hand augered)	0.0026		0.0179	**> 0.013**	0.022		1.31		0.091	**>0.07**	1.28		3.85		<0.23		0.301	
61 - 91.4 cm (hand augered)	0.0005		0.0091		0.0062		0.63		0.057		0.71		2.14		<0.22		0.098	
91.4 - 122 cm (hand augered)	0.0018		0.0053		0.0037		0.81		0.027		0.68		2.04		<0.21		0.101	
122 - 152 cm (hand augered)	0.0033		0.0162	**> 0.013**	0.0086		1.25		0.083	**>0.07**	1.26		3.79		<0.20		0.172	
152 - 183 cm (hand augered)	0.0005		0.0082		0.0114		1.12		0.040		1.07		3.20		<0.23		0.22	

Background Reference Used: McLin and Lyons, 2002 (BGULs)

Table 1 (continued). Data Table Highlighting Values that Exceed Background Reference Values

Station	Date	Pu^{238} (pCi/g)	> 0.0087	$Pu^{239/240}$ (pCi/g)	>0.013	Cs^{137} (pCi/g)	>0.56
Number of stations that exceed reference			none		none		none
Rio Grande Channel Sediments							
Rio Grande at Pilar	3/11/2003	0.0006		0.0067		0.163	
Chama River at Chamita	3/25/2003	-0.0004		0.0043		0.118	
Rio Grande at Rio Pojoaque	4/4/2003	-0.0008		0.0033		0.027	
Rio Grande below Los Alamos	4/4/2003	0.0000		0.009		0.138	
Rio Grande above Ancho	4/1/2003	-0.0011		0.0046		0.073	
Rio Grande at Pena Blanca	3/24/2003	0.0003		0.0033		0	
San Angostura Diversion	3/18/2003	-0.0004		0.010		0.2	
Rio Grande below Albuquerque	6/10/2003	-0.0003		0.0013		0.043	
Mean		-0.0003		0.0053		0.0953	
McLin, et al, 2002, River BGUL		0.0087		0.013		0.56	
Purtymun, et al, 1987 (mean)		0.0000		0.005		0.18	
Purtymun, et al, 1987 [upper limit background (mean + 2 sigma)]		0.0060		0.023		0.44	

abundance of each isotope in naturally occurring uranium, and by calculating the ^{234}U, ^{235}U, and ^{238}U isotope activity concentrations from their relative masses and specific activities. We found that ^{234}U background concentration was equal to 1.40 pCi/g, ^{235}U was equal to 0.07 pCi/g, and ^{238}U was equal to 1.49 pCi/g.

Two horizons at Ancha site, between 101 and 162 cm, contained ^{234}U, ^{235}U, and ^{238}U that exceeded the BGUL. An additional horizon contained only ^{235}U greater than the BGUL. Three horizons at the Frijoles site between the surface and 152 cm, contained only ^{235}U concentrations greater than the BGUL.

Four Ancha site horizons, between 101 and 286 cm, contained ^{137}Cs exceeding its 0.56 pCi/g BGUL. All uranium and cesium measurements identifying horizons containing exceptional levels of the LANL contaminants were corroborated by the plutonium measurements.

Based on an evaluation that includes only comparisons of individual measurements to BGUL values, LANL impacts were identified at Ancha site between 101 to 229 cm in depth, and at Frijoles site between the surface and 152 cm. This conclusion is based on multiple measurements of $^{239/240}Pu$, uranium isotopes, and ^{137}Cs that exceed the BGULs. We did not observe a LANL impact on the sediments in the Rio Grande River channel bottom.

Statistical Comparison of Mean Values

The following section describes the statistical comparison of our mean values at each site to the mean values obtained from the Santa Clara site and from the McLin and Lyons (2002) data set. A detailed description can be found in Appendix B. Statistics Description.

A statistical evaluation of radionuclide measurements in bank sediments collected from terrace strata along the Rio Grande below Los Alamos Canyon indicate $^{239/240}Pu$, ^{137}Cs,

^{90}Sr, and ^{241}Am are elevated when compared to reference conditions upstream from LANL.

Radiochemical data for Rio Grande sediments from terrace deposits downstream of Los Alamos Canyon were compared to two sources of background reference data; 1) data from terrace deposits above Los Alamos Canyon, and 2) data from historic background river sediments collected by LANL. The reference terrace sites are located at Santa Clara Pueblo, 12.1 km (7.5 miles) upstream of Los Alamos Canyon.

We made statistical comparisons between these reference conditions and four locations downstream of Los Alamos Canyon. Multiple sediment horizons were sampled and analyzed for radionuclides at the Ancha, Pajarito, Water, and Frijoles sites, 5.1, 10.9, 14.0, and 19.3 km (or 3.2, 6.8, 8.7, 12.0 miles) downstream of Los Alamos Canyon, respectively.

We evaluated these data by testing for normality and then running parametric and nonparametric tests to determine what relations existed between the reference data sets and those downstream of LANL. The Student's t-test was used to compare means of parametric or normally distributed data, and the Mann-Whitney U test was used to compare non-parametric or distribution free data. Statistical significance was determined at $p \leq 0.05$.

Based on these statistical comparisons, we found that $^{239/240}$Pu was the most persistent radionuclide found in terraces downstream of LANL. By far, the largest concentrations were found at the Ancha site followed by the Frijoles site, and then the Water site. Cesium-137 was also elevated by the greatest amount at Ancha, also followed by the Frijoles site. Strontium-90 was found to be elevated at the Ancha site and ^{241}Am was elevated at the Frijoles site. The differences for all constituents between the Pajarito site and reference conditions were statistically insignificant.

Values reported less than analytical capabilities (non-detects) were included in these evaluations. The potential exists that the descriptive statistics may contain a low bias and the following relationships that we describe are approximations. Radionuclide measurements were most elevated at the Ancha site, approximately 5.1 km (3.2 miles) downstream of Los Alamos Canyon. The $^{239/240}$Pu mean was 24 times greater than at Santa Clara and 7.6 times greater than the LANL background mean. The ^{137}Cs mean was 7.2 times greater than at the Santa Clara site and 2.2 times greater than the LANL background mean. The ^{90}Sr mean was 3.5 times greater than the mean at the Santa Clara site and 1.6 times greater than the LANL background mean.

We expected the contaminant concentrations and differences to diminish downstream with distance; instead we found the next greatest levels were present in the farthest downstream location, 19.3 km (12.0 miles) downstream of Los Alamos Canyon at the Frijoles site. The $^{239/240}$Pu mean was 13.9 times greater than at the Santa Clara site and 0.4 times greater than the LANL background mean. The ^{137}Cs mean was greater than at Santa Clara, and ^{241}Am measurements were significantly different and greater than the

LANL background mean. The mean value for ^{241}Am at the Frijoles site was 2.6 and 2.0 times greater than the Santa Clara site and background mean, respectively.

The Water Canyon site, 14.0 km (8.7 miles) downstream of Los Alamos Canyon, demonstrated $^{239/240}$Pu measurements 5.2 times greater than the Santa Clara site. The Pajarito site contained the lowest level of contaminants, at levels similar to the regional reference levels that describe global fallout. This site is 10.9 km (6.8 miles) downstream of Los Alamos Canyon and only 5.8 km (3.6 miles) downstream of the Ancha site.

An explanation for this phenomenon may be due to the Cochiti Reservoir closure in 1973. The lake level advanced to the vicinity of Pajarito Canyon. Grain size distribution analysis, discussed later in this report, indicates the lacustrine sediment deposits contained a higher percent of fines in the sediment matrix as distance increased downstream. The larger amounts of fines tend to increase contaminant concentrations in sediments relative to samples otherwise similar.

Thermal Ionization Mass Spectrometric Analysis Evaluation

This section describes the results and the method used for identifying the origin of plutonium at the study sites. Plutonium origins are defined as global fallout or due to Laboratory operations. The TIMS method, described here, allowed us to determine the origin of plutonium by quantifying the isotopic ratio of ^{240}Pu to ^{239}Pu in terrace deposits along the Rio Grande. While the more common alpha spectroscopy method is useful in characterizing plutonium concentrations, potential LANL impacts, and health and safety concerns, TIMS is a much better tool for identifying the sources of plutonium in the environment. The procedure for TIMS analysis of plutonium was developed by the Los Alamos Clean Chemistry and Mass Spectrometry Laboratory and is described in detail in Efurd and others, (1993). Data generated by TIMS for this study was compared and interpreted in part by review of a previous LANL report "*Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley*" (Gallaher and Efurd, 2002). More details regarding the methods and reference values we used to interpret the TIMS data can be found in Appendix C.

The TIMS evaluation methodology contrasts with using statistical reference levels derived from alpha spectroscopy measurements, traditionally used to identify potential LANL impacts. Gallaher and Efurd, (2002) found that the plutonium 240:239 atom ratio for global fallout in Northern New Mexico ranges between 0.13 and 0.21 at a 99.7% confidence level. Ratio values between 0.13 and 0.16 indicate a possible LANL impact, or some mixture of Laboratory and global fallout plutonium. Plutonium 240:239 atom ratios less than 0.13 indicate a LANL contribution of plutonium. The atom ratios that identify plutonium sources in Northern New Mexico are compiled in Table 2. In general, atom ratios less than 0.16 indicate a LANL component of plutonium exists, and values greater than 0.16 are indicative of plutonium that originates from global fallout.

Table 2. Plutonium 240:239 atom ratio ranges used to identify potential sources

Pu 240 : Pu 239 Atom Ratio	Likely Plutonium Source
0.16 - 0.21	Global fallout
0.13 - 0.16	LANL influence possible and/or mixture
Less than 0.13	LANL influence likely and/or mixture

The distribution of legacy plutonium buried in terraces along White Rock Canyon is not uniform. Older sediments deposited after 1943 contain a greater fraction of LANL derived plutonium, covered by more recent sediments containing only global fallout plutonium. We also found that lacustrine deposits originating from closure of Cochiti Reservoir contained a component of LANL plutonium. The average fraction of LANL plutonium in White Rock Canyon sediments was 34%. The greatest LANL influence occurs at the Ancha site containing legacy plutonium as great as 99%; note that the shallowest sample at the Ancha site did not contain LANL plutonium. The remaining sites downstream of the Ancha site appear to be affected to a lesser degree. The Laboratory plutonium in those sediments ranges from 8% to 47%.

While $^{239/240}$Pu concentration comparisons to the River BGUL suggest that LANL impacts exists only at the Ancha and Frijoles sites, almost all of the plutonium atom ratios indicate that there is some component of LANL derived plutonium at all sites. Most, but not all of the $^{239/240}$Pu concentrations for sediments at the Ancha and Frijoles sites shown in Table 3 below are greater than 0.013 pCi/g, and all but one of the plutonium 240:239 atom ratios obtained for this report are less than 0.16, ranging from 0.08 to 0.21. Keep in mind, plutonium concentrations greater than 0.013 pCi/g or 240:239 atom ratios less than 0.16 suggests a LANL impact.

Table 3 summarizes the TIMS and alpha spectroscopy measurements for $^{239/240}$Pu in the sediments we collected along the Rio Grande. The first column includes a general description of the site as well as depths from which the samples were collected. The following two columns show the activity measurements of the ^{239}Pu and ^{240}Pu isotopes. These values are derived from TIMS measurements of atoms of plutonium per gram in each sample multiplied by the specific activity for the appropriate isotope. The fourth column is the sum of the activities derived from the two isotopes. The fifth column is the atom-ratio values of ^{240}Pu to ^{239}Pu measured by TIMS.

Above Los Alamos Canyon at the Santa Clara site, as well as for the Pajarito and Water sites below Los Alamos Canyon, the activities suggest that LANL plutonium is not associated with those deposits. These values are less than the 0.013 pCi/g sediment BGUL reference value. Yet the plutonium atom ratios indicate there is a LANL influence at the Pajarito and Water Canyon sites. The ratios are between 0.11 and 0.15. Although less than 0.16, the values are relatively larger than those obtained for samples from the Ancha and Frijoles sites. This suggests a smaller content of LANL plutonium exists in the Pajarito and Water Canyon sites than at the other areas, particularly at the Cañada Ancha site.

Table 3. Plutonium measurements along the Rio Grande using both alpha spectroscopy and thermal mass spectroscopy analysis

Station Description	Thermal Ionization Mass Spectrometric Analysis				Alpha Spec	Duplicate Error Ratio
	^{239}Pu Activity (pCi/g)	^{240}Pu Activity (pCi/g)	^{239}Pu + ^{240}Pu Activity (pCi/g)	^{240}Pu : ^{239}Pu Atom Ratio	$^{239/240}$Pu (pCi/g)	DER
Santa Clara Site						
abandoned floodplain, active 1941-1968						
5 to 31 cm	0.0027	0.0016	0.0043	0.157	0.005	0.25
Cañada Ancha Site						
floodplain, overflow area flooded in 1958 or 1967 flood						
0 to 31 cm	0.0004	0.0003	0.0007	0.208	0.002	0.75
101 to 110 cm	0.0172	0.0059	0.0231	0.093	0.067	3.65
162 to 186 cm	0.0451	0.0157	0.0608	0.094	0.066	0.44
213 to 229 cm	0.0397	0.0023	0.0420	0.016	0.034	1.52
Pajarito Site						
active floodplain and bar surface						
31 to 46 cm	0.0016	0.0007	0.0023	0.118	0.008	0.95
Water Canyon Site						
active floodplain and bar surface						
0 to 31 cm	0.0021	0.0008	0.0029	0.110	0.003	0.07
91 to 101 cm	0.0026	0.0014	0.0040	0.151	0.008	0.76
Frijoles Site						
pre-1950 floodplain deposits and reservoir sedimentation						
31 to 61 cm	0.0106	0.0032	0.0138	0.081	0.018	0.49
122 to 152 cm	0.0057	0.0027	0.0084	0.131	0.016	1.07

With the exception of the surface sample, values at the Ancha site indicate a LANL input of plutonium. Both TIMS (0.0007 pCi/g) and alpha spectroscopy (0.002 pCi/g) plutonium activity measurements for the surface sample are less than the BGUL. The surface sample also has a large atom ratio of 0.21, indicating global fallout plutonium. These values clearly indicate a lack of LANL plutonium. The samples at depth tell a different story. The activity measurements are two to five times above the BGUL, and the small atom ratios, ranging from 0.02 to 0.09, demonstrate a large component of LANL plutonium.

At the Frijoles site, the picture is not as clear. Most of the plutonium measurements do suggest a LANL influence, but are near the reference level. The activity values of 0.0084 to 0.018 pCi/g are only slightly above or just below the BGUL for plutonium in sediments. Although one atom ratio value of 0.08 is a fairly clear indicator of a LANL impact, the second value of 0.13 only suggests a LANL impact is possible.

Based on the atom ratios, proportions or percentages of LANL derived plutonium can be calculated. We calculated the proportion of the LANL plutonium mixed with the plutonium from global fallout using the following equation. This equation is a modified form of a method originally described by Hardy and others, (1972) from Gallaher and

Efurd, (2002). The ^{240}plutonium to ^{239}plutonium ratios and the calculated proportion of LANL plutonium in our samples collected along the Rio Grande are compiled in Table 4.

$$\frac{[PuActivity]_L}{[PuActivity]_F} = \frac{(R_F - R_S)(1 + 3.67R_L)}{(R_S - R_L)(1 + 3.67R_F)}$$

where:

$[Pu\ Activity]_L$ = plutonium activity in LANL component,
$[Pu\ Activity]_F$ = plutonium activity in global fallout component,
R_S = Pu 240:239 atom ratio measured in the sediment sample,
R_L = 0.015, the Pu 240:239 atom ratio of plutonium released by LANL,
R_F = 0.169, the Pu 240:239 atom ratio of global fallout in Northern New Mexico, and
3.67 = ratio of half-lives of ^{239}Plutonium to ^{240}Plutonium.

Table 4. Percentage of LANL plutonium in sediments collected along the Rio Grande

Station Description	^{240}Pu : ^{239}Pu Atom Ratio (R_S)	LANL Component %
Santa Clara Site		
5 - 31 cm (hand augured)	0.16	5
Cañada Ancha Site		
0 - 31 cm (hand augured)	0.21	0
101 -110 cm (hand augured)	0.09	39
162 - 186 cm (hand augured)	0.09	38
213 - 229 cm (hand augured)	0.02	99
Pajarito Site		
31 - 46 cm (hand augured)	0.12	24
Water Canyon Site		
0 - 31 cm (outcrop A)	0.11	29
91 - 101 cm (outcrop A)	0.15	8
Frijoles Site		
31 - 61 cm (hand augured)	0.08	47
122 - 152 (hand augured)	0.13	18

We found the average fraction of LANL plutonium in White Rock Canyon sediments was 34%. The greatest LANL influence occurs at the Cañada Ancha site where LANL plutonium percentages range as high as 99% and as low as 0%, with an average of 44%. The greatest proportions of LANL plutonium occurs at depths reflecting episodic deposition during 1950's and 1960's floods. The Frijoles site, farthest downstream, appears to be affected to a lesser degree. The plutonium in the sediment is probably

derived from LANL, containing 18% and 47% LANL plutonium, averaging 33%. Based on the silt loam sediment texture, we suggest they were deposited while the Cochiti reservoir was filled above these horizons. Although plutonium concentrations at the Pajarito and Water sites were measured below the BGUL, the atom ratios suggest that from 8% to 29%, averaging 21%, of the plutonium in sediments are LANL derived.

The ^{240}Pu to ^{239}Pu ratios and LANL proportion of plutonium in our samples collected along the Rio Grande are shown in Figure 12. The vertical axis represents the ^{240}Pu to ^{239}Pu ratio scale, and the sample site and depth horizons are identified along the horizontal scale. The numerical figure at each shape identifies the proportion of LANL plutonium in the associated sample. The colored horizontal lines are the 0.16 and 0.13 references that identify the potential presence of LANL plutonium.

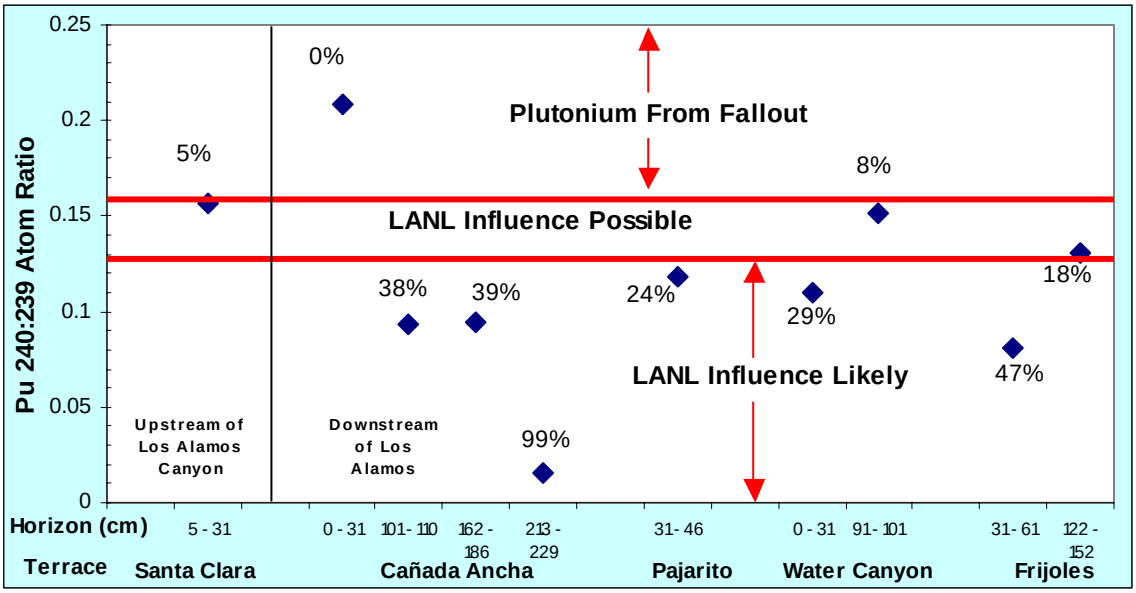

Figure 12. Percentage of LANL plutonium and the Pu 240:239 atom ratios for samples collected along the Rio Grande

The estimates of 0% LANL derived plutonium at the Ancha site surface horizon, and a 5% component of LANL plutonium at Santa Clara were unanticipated. It is possible that the surface at Ancha site is comprised of recent sediments containing only global fallout plutonium, while the Santa Clara site surface contains atmospheric impacts from LANL. It is also possible that at these low levels the analytical and sampling uncertainties may lead to anomalous conclusions. Most of the remaining samples at Cañada Ancha, Pajarito, Water Canyon, and Frijoles sites show more definite LANL influence, containing from 24% to 99% LANL derived plutonium. Two sample points, one at Water Canyon and one at Frijoles, exhibit possible LANL influence. Their Pu 240:239 ratios are 0.15 and 0.13 demonstrating 8% and 18% LANL content.

Figure 13, below, shows the relationships between the atom ratios and the plutonium concentrations obtained for samples reviewed in this study. The vertical axis represents the ^{240}Pu to ^{239}Pu ratios, while the horizontal axis represents the $^{239/240}$Pu concentrations.

The colored lines within the chart are reference values from historical data: the vertical line indicates the 0.013 pCi/g BGUL and the horizontal line indicates the 0.16 plutonium atom reference that defines a LANL impact.

Figure 13 demonstrates that, generally, samples containing plutonium concentrations less than the reference BGUL exhibit both fallout and LANL plutonium influences - not all samples containing plutonium concentrations below the reference value are free of LANL plutonium. The figure also shows that all samples containing plutonium concentrations greater than the BGUL have atom ratios that clearly express an existing component of LANL.

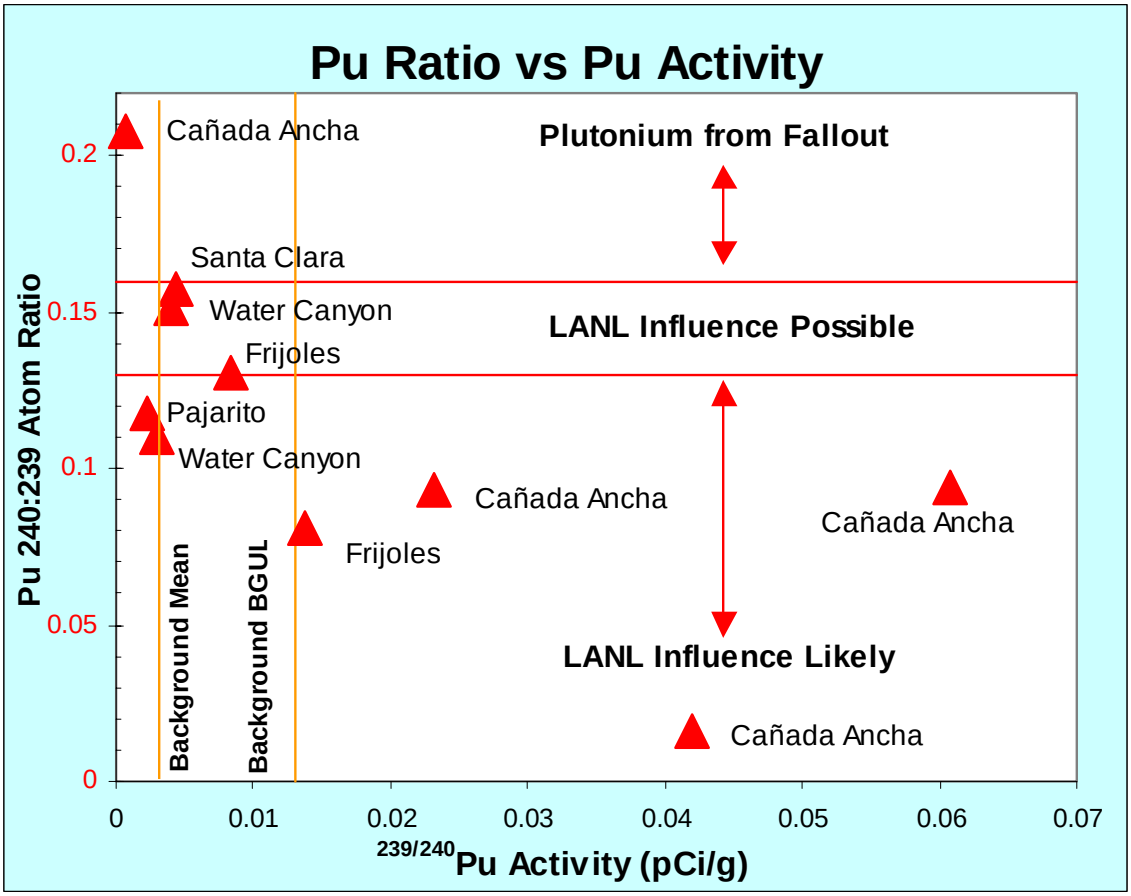

Figure 13. Plutonium 240:239 atom ratios and $^{239/240}$Pu concentrations measured in sediments along the Rio Grande, and the relation between them and the background references for river sediments

For example, the Cañada Ancha sample, in the upper left corner of the chart, is within the area outlined for global fallout. This area is defined by atom-ratio values greater than 0.16 and plutonium concentrations below the 0.013 pCi/g BGUL. This sample was collected at the surface and both the concentration and the atom ratio clearly demonstrate that plutonium is derived from atmospheric fallout. The other samples from Cañada Ancha and one from the Frijoles site, seen in the "LANL Influence Likely" section of the chart, clearly define the plutonium source. All four contain concentrations exceeding the BGUL and have atom ratios that demonstrate high percentages of LANL plutonium.

Those atom ratios are less than 0.16. The remaining samples are not as clearly defined and include samples from the Water Canyon, Santa Clara, Frijoles, and Pajarito sites. They contain plutonium concentrations less than the background reference, yet the atom ratios are less than 0.16, which identifies a portion of the plutonium as originating from LANL.

The Santa Clara sample, located in the upper left portion of the chart, contains a plutonium concentration below the BGUL, yet the atom ratio identifies the samples as containing a possible LANL influence. Santa Clara Pueblo is within the Laboratory wind-shed and historic operational releases may be the source of the LANL component in this sample. While the LANL impact at the Santa Clara site appears small, additional investigations may be warranted to clearly define the extent of LANL impacts.

To summarize, we found that there is LANL plutonium buried in terraces along White Rock Canyon, and that the distribution is not uniform. We found that older sediments deposited after 1943 contain larger proportions of LANL derived plutonium, covered by more recent sediments containing only global fallout plutonium. We also found that lacustrine deposits originating from closure of Cochiti Reservoir contained a component of LANL plutonium.

The greatest LANL influence was at the Cañada Ancha site, closest to Los Alamos Canyon. These influences were seen buried at depths reflecting episodic flood deposition that occurred during the 1950's and 1960's. Shallower deposits there contained smaller concentrations of plutonium and little or no component of Laboratory plutonium. We also found that LANL impacts occurred further down White Rock Canyon. Lacustrine sediments containing LANL plutonium were deposited at the mouths of Frijoles, Water, and possibly Pajarito canyons during a period in the 1970's and 1980's when Cochiti Reservoir was filled to a level greater than the present.

We also expect the LANL component of plutonium in Cochiti Reservoir is currently increasing due to geomorphic changes on the Pajarito Plateau resulting from the Cerro Grande fire. Increased magnitudes and frequencies of stormwater flows have occurred in Pueblo and Los Alamos canyons since the fire. The stormwater flows are carrying LANL legacy plutonium stored in the Los Alamos watershed to the Rio Grande.

Grain Size Distribution Evaluation

Past studies have shown a correlation of grain size distributions to concentrations of heavy metals and radionuclides. These constituents naturally bind more readily to clay- and silt-sized particles than to sandy portions of a sediment sample. This phenomenon is generally related to strong van der Waals forces, higher cation exchange capacity, total organic carbon content, and higher surface area of clay and silt particles relative to coarser materials (McLin and Lyons, 2002; Graf, 1993). Radioactive concentration, source identification, and depositional history can be evaluated by the distribution of particle sizes in sediments and contaminant correlations to grain size fractions.

We used particle size distribution analysis and contaminant correlations to identify historic fluvial processes within the study area. We identified four main depositional categories: 1) sediments deposited prior to global fallout followed by 2) episodic deposition of sediments containing LANL legacy waste and 3) lacustrine deposits containing a mixture of global fallout and legacy materials, and then 4) covered by floodplain sediments from the upper Rio Grande containing mostly global fallout materials.

We also collected samples within the Rio Grande channel in 2003 and had them analyzed for ^{238}Pu, $^{239/240}$Pu, ^{137}Cs, and particle size distributions. The samples represent active channel sediments in transport. They were collected in slack-water, or low energy conditions behind large channel obstructions such as boulders or in pools.

Particle size distribution analysis, performed by Desert Research Institute (Reno, NV), identifies particle size in samples using sieve and pipette methods, and reports the weight percent of each class. Appendix D contains a description of the method and Appendix A contains the complete data set of particle size distribution analysis in our samples. Textural classifications are based on the percent weights of clays, silts, and sands, and combinations of each. We inferred the deposition history from these classifications.

A summary data set of the grain size analysis for the samples collected from the banks of and in the Rio Grande channel is shown in Table 5. The first column on the left contains the stations and core depths. The grain size classes, listed in the following four columns, are listed as combined total sand, total silt, total clay, and silt plus clay, and reported in weight percent. We also include the ^{240}Pu to ^{239}Pu atom ratios acquired from the TIMS analysis discussed in the previous section, and the alpha spectroscopy measurements of $^{239/240}$Pu in the sixth and seventh columns. The plutonium measurements and their relationship to particle distributions were instrumental for evaluating depositional history.

The eighth column includes the activity concentrations of $^{239/240}$Pu normalized to the clay and silt content in each sample. These values will be discussed in greater detail later in this section. Conceptually, they can be used to identify LANL contaminant content regardless of dilution from coarse-grained sediments.

The last two columns are the textural classification, and the deposition interpretation. A number of grain size studies, including stormwater, Rio Grande channel samples from other projects, and work by other authors were considered in this report to identify deposition features (Ford-Schmid and Englert, 2004; Malmon and others, 2004).

Table 5. Summary data set of the grain-sized distribution analysis

Station Description	Total Sand % wt	Total Silt % wt	Clay % wt	Silt + Clay	Atom Ratio from TIMS (240:239)	Pu 239/240 (pCi/g) from Alpha Spec	Normalized pCi/g / silt + clay	Texture Description	Deposition Interpretation
Santa Clara Sites									
DOEOB 3									
(abandoned floodplain, active 1941-1968)2B									
5.18 - 30.5 cm (hand augered)	56.3	39.4	4.3	43.7	0.16	0.0051	0.0120	loam	Flood Plain
DOEOB 4									
(abandoned floodplain, active prior to 1941)3B									
33.5 - 39.6 cm (hand augered)	22.3	70	7.7	77.7	NA	0.0008	0.0010	silt loam	Flood Plain
73.2 - 88.4 cm (hand augered)	95.7	1.4	2.9	4.3	NA	-0.00062	-0.0140	sand	Channel
DOEOB 5									
(abandoned floodplain, active prior to 1941)3B									
24.4 - 39.6 cm (hand augered)	44.2	45.8	10	55.8	NA	-0.00022	0.0000	loam	Flood Plain
57.9 - 73.2 cm (hand augered)	20.2	71	8.9	79.8	NA	0.0006	0.0010	silt loam	Flood Plain
116 - 158 cm (hand augered)	24.4	65.7	9.8	75.6	NA	0	0.0000	silt loam	Flood Plain
Cañada Ancha Site									
(slough area during 1940-1958 or 1967) unit 3B									
0 - 30.5 cm (hand augered)	74.2	15.4	10.4	25.8	0.21	0.0022	0.0090	sandy loam	Channel
30.5 - 61 cm (hand augered)	68.2	19.2	12.5	31.8	NA	0	0.0000	sandy loam	Channel
61 - 91.4 cm (hand augered)	65.9	22.8	11.3	34.1	NA	0.0017	0.0050	sandy loam	Channel
91.4 - 101 cm (hand augered)	65	21	14	35	NA	0.0033	0.0090	sandy loam	Channel
101 - 110 cm (hand augered)	20.2	52.5	27.3	79.8	0.09	0.067	0.0840	clay loam	Flood Plain
110 - 125 cm (hand augered)	44.5	34.6	20.9	55.5	NA	0.0121	0.0220	loam	Flood Plain
125 - 137 cm (hand augered)	39.8	35.6	24.6	60.2	NA	0.0262	0.0440	loam	Flood Plain
137 - 162 cm (hand augered)	16	43.7	40.4	84	NA	0.043	0.0510	silty clay	Flood Plain
162 - 186 cm (hand augered)	8.7	42.8	48.5	91.3	0.09	0.066	0.0720	silty clay	Flood Plain
186 - 213 cm (hand augered)	17.7	45.2	37.1	82.3	NA	0.0144	0.0180	silty clay loam	Flood Plain
213 - 229 cm (hand augered)	2.2	47	50.9	97.8	0.02	0.0337	0.0340	silty clay	Flood Plain
323 - 335 cm (hand augered)	95.3	3.1	1.5	4.7	NA	0.0029	0.0620	sand	Channel
Pajarito Site									
(active floodplain and bar surface)1B									
0 - 30.5 cm (hand augered)	96.5	1.8	1.7	3.5	NA	0.0029	0.0830	sand	Channel
30.5 - 45.7 cm (hand augered)	87.4	9.2	3.5	12.6	0.12	0.0076	0.0600	sand	Channel
Water Canyon Site									
(active floodplain and bar surface)1B									
0 - 30.5 cm (outcrop A)	76.2	19.5	4.3	23.8	0.11	0.0032	0.0130	loamy sand	Lacustrine*
30.5 - 61 cm (outcrop A)	91.3	5.8	2.9	8.7	NA	0.0061	0.0700	sand	Lacustrine*
61 - 91.4 cm (outcrop A)	75.1	20.6	4.3	24.9	NA	0.0082	0.0330	loamy sand	Lacustrine*
91.4 - 101 cm (outcrop A)	69.1	27	3.9	30.9	0.15	0.008	0.0260	sandy loam	Lacustrine*
101 - 116 cm (outcrop A)	86.8	10.1	3.1	13.2	NA	0.0021	0.0160	loamy sand	Lacustrine*
91.4 - 101 cm (outcrop B)	38.4	55.7	5.9	61.6	NA	0.0017	0.0030	silt loam	Flood Plain
Frijoles Site									
(pre-1950 floodplain deposits and reservoir sedimentation)									
0 - 30.5 cm (hand augered)	29.2	60.8	10	70.8	NA	0.0217	0.0310	silt loam	Lacustrine*
30.5 - 61 cm (hand augered)	35.7	48.6	15.7	64.3	0.08	0.0179	0.0280	loam	Lacustrine*
61 - 91.4 cm (hand augered)	71	21.7	7.3	29	NA	0.0091	0.0310	sandy loam	Lacustrine*
91.4 - 122 cm (hand augered)	80.1	15.8	4.1	19.9	NA	0.0053	0.0270	loamy sand	Lacustrine*
122 - 152 cm (hand augered)	31.4	58.4	10.2	68.6	0.13	0.0162	0.0240	silt loam	Lacustrine*
152 - 183 cm (hand augered)	38.3	50.9	10.8	61.7	NA	0.0082	0.0130	silt loam	Lacustrine*
2003 RG Channel, slack water									
Rio Grande at Pilar	24.1	59.5	16.4	75.9	NA	0.0067	0.0088	silt loam	Channel*
Chama River at Chamita	11.5	65.8	22.7	88.5	NA	0.0043	0.0049	silty clay loam	Channel*
Rio Grande at Rio Pojoaque	35.3	49.0	15.7	64.8	NA	0.0033	0.0051	silt loam	Channel*
Rio Grande below Los Alamos	19.1	67.3	13.6	80.9	NA	0.0090	0.0111	silt loam	Channel*
Rio Grande above Ancho	64.9	29.3	5.8	35.1	NA	0.0046	0.0131	sandy loam	Channel*
Rio Grande at Pena Blanca	67.5	14.5	18.1	32.5	NA	0.0033	0.0101	sandy loam	Channel*
San Angostura Diversion	5.7	65.7	28.6	94.3	NA	0.0100	0.0106	silt loam	Channel*
Rio Grande below Albuquerque	67.0	27.0	5.9	33.0	NA	0.0013	0.0039	sandy loam	Channel*

Data provided by Desert Research Institute, Reno, NV
***Upper Reach of Reservoir**
***Collected from active slack water channel**

The following chart in Figure 14 illustrates the texture classification and relative content of sand, and silt and clay, for samples at each site. The textures range from sand to silty clay. The lower horizontal axis contains the texture class; the headings at the top identify the sample sites. At each site from left to right, the samples are arranged from the shallowest horizon to the deepest, or in the case of the channel sediments from the upstream to downstream locations. The vertical axis describes the percent weight of sands, and silts and clays.

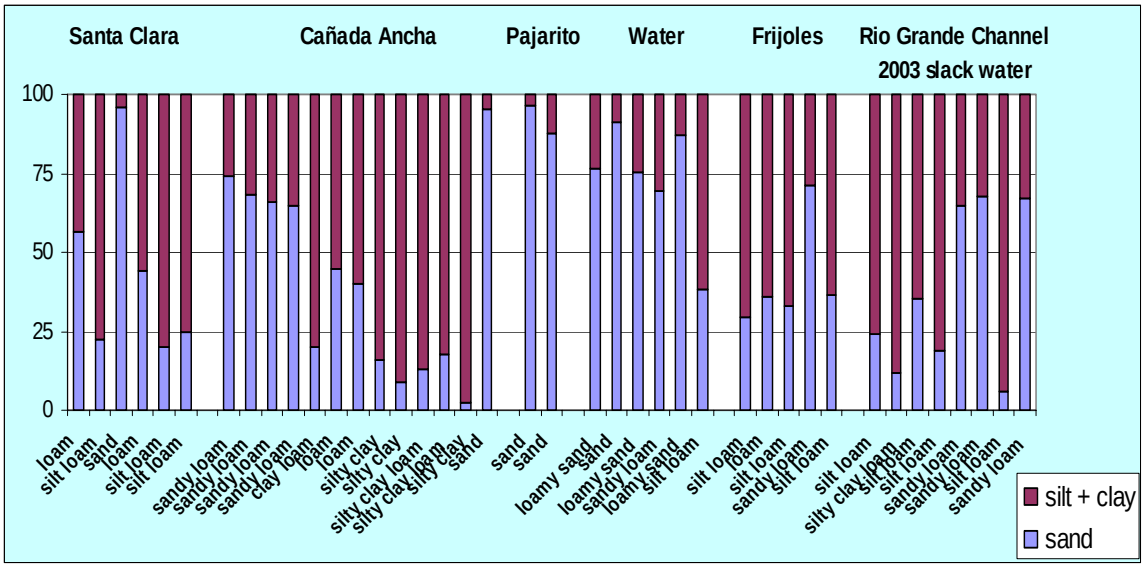

Figure 14. Texture classification and relative content of sand, and silt and clay

Sediment horizons at the Santa Clara, Frijoles, and Cañada Ancha sites are mostly silt to clay loam materials, with some exceptions. The samples collected from horizons at the Pajarito and Water Canyon sites, one horizon at the Santa Clara site, two horizons at the Frijoles site, and the shallowest and deepest horizons at the Cañada Ancha site are predominantly sand to sandy loam materials. Although not shown on Figure 14, the Cañada Ancha samples also contained a large fraction of gravel-size fragments, field identified as pumice fragments. Pumice is buoyant and locally available from the Los Alamos Canyon watershed. Sediments collected within the Rio Grande channel range from silty clay loam to sandy loams. They do not reflect bedload sediments and contain a larger content of fine grained particles than might be expected. The samples were selected to provide as fine grained sediment sample as possible.

The identification of channel and near channel deposits can be complex. The deposits originate from alternating fluvial processes; on-going channel and floodplain degradation and aggradation are constantly reworking landforms and deposits. River channels often change size, shape, pattern, or location in response to hydraulic controls and in doing so they create suites of floodplain features that include abandoned channels, terraces, active and inactive floodplains, and a variety of bars inside and outside the present active channels. (Gregory, 1977 in Graff, 1993)

Generally, fine material in sediment deposits originate in low energy systems such as suspended load deposition in receding floodwaters. High-energy systems transport coarse-grained bedload sediments found within channel bottoms and transported along the water-channel interface. Lacustrine deposits may vary depending on position relative to distance from the river entry to lakes. Coarser materials preferentially settle at the entrance. The lacustrine deposits contain increasing proportions of finer sediments as distance increases and flow velocity decreases.

To identify deposition history of samples in bank sediments along the Rio Grande, we also compared grain size distribution in samples from the Rio Grande terraces to grain size distribution studies from other projects. These studies include LANL and NMED on-going projects being performed in Pueblo and Los Alamos canyons. The following chart in Figure 15 demonstrates the comparison of average grain size classes for sediments found in channels, floodplains, and stormwater from Los Alamos, Pueblo, and White Rock canyons. Grain size fractions are identified along the bottom axis, ranging from coarse sediments containing particle sizes greater than 2.0 mm to clays that are less than 0.625 mm. The vertical axis describes the percent fraction of each class. The sediment sources are identified in the text box on the right and reflect high energy systems, color-coded in shades of red, to low energy deposition, color coded in shades of green. The samples collected from cores in this study along the Rio Grande banks are black and those from the Rio Grande river channel are blue. Individual columns represent the average percent weight of each grain size fraction measured in samples, and the combinations reflect the sediment source.

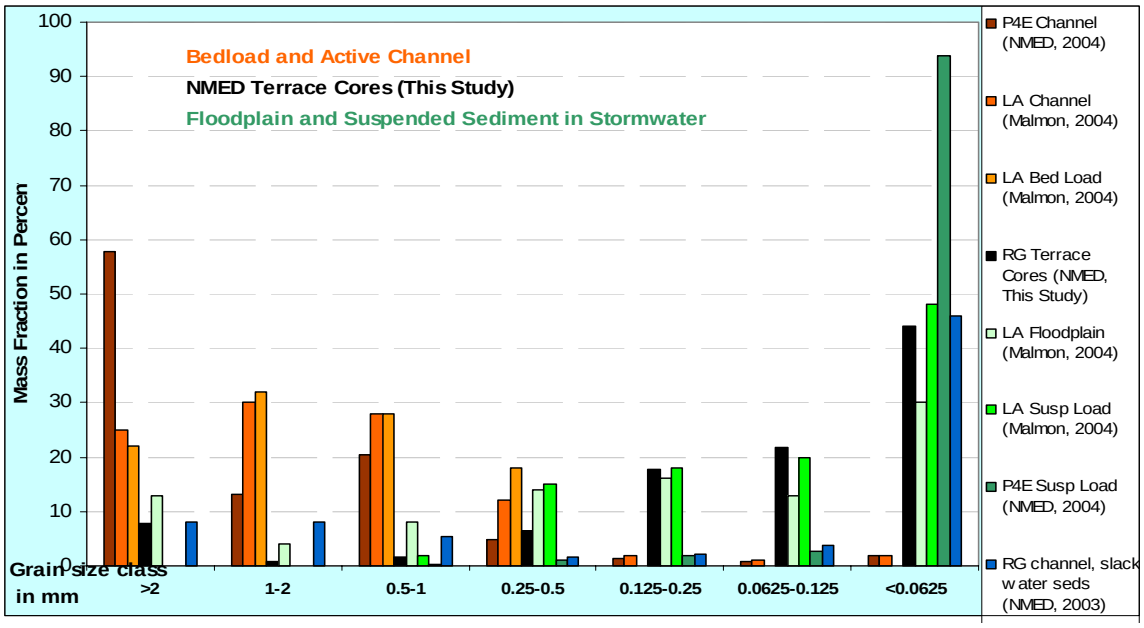

Figure 15. Grain size distribution in Rio Grande terrace deposits compared to other deposition features and surface water transport mechanisms

We compared the sediment classes measured in the Rio Grande terrace cores, reflected by the black columns in the chart, to those sediments from channels, floodplains, and storm-water from Los Alamos and Pueblo canyons. In sediments collected from sites we selected along the Rio Grande, most are similar to suspended sediments found in storm-water and floodplain materials derived from Los Alamos and Pueblo canyons, reflected by the green columns. The greatest particle size proportions in sediments from the Rio Grande terrace deposits are found in the fine fractions less than 0.25 mm. The channel sediments contain a large proportion of fines reflecting low energy deposition or slack-water conditions in the river channel.

Although most horizons from the core sites are fine-grained floodplain materials, some exhibit sandy textures similar to active channel deposits or encroaching lake boundary deposit conditions.

We made multiple comparisons of $^{239/240}$Pu content to grain size in multiple sample arrays from this study and found a consistent trend, although the correlations are often not strong, R^2 ranging from 0.36 to 0.65, where R^2 is the correlation coefficient. The correlations show that as the content of the silt and clay fractions increase, the plutonium concentrations increase. The samples originate from; areas that reflect background sources far upstream from the Laboratory, areas containing large volumes of LANL legacy waste, and areas containing mixtures of both. Comparisons made for sediments that originate from single source areas such as in Pueblo or Los Alamos canyons demonstrate the strongest correlations. Those samples that originate from mixtures, or comparisons that include background samples and contaminated samples, develop the least strong correlations.

The chart in Figure 16 demonstrates the correlation of sand, and silt and clay grain size fractions to $^{239/240}$Pu concentrations in the sediments collected for this study. These sediments may have been derived from multiple source areas; sediments containing only background, sediments containing relatively large components of LANL legacy $^{239/240}$Pu, and sediments containing mixtures of background and legacy $^{239/240}$Pu. The horizontal axis contains the plutonium concentration scale, while the vertical axis contains the percent weight of the grain fractions. The dark squares and trend line represent the silt and clay correlation to plutonium concentrations, while the light triangles and trend line represents an inverse correlation of the sand fraction to plutonium. The R^2 value of 0.36 reflects the plutonium / grain size correlation in all sediments collected for this study.

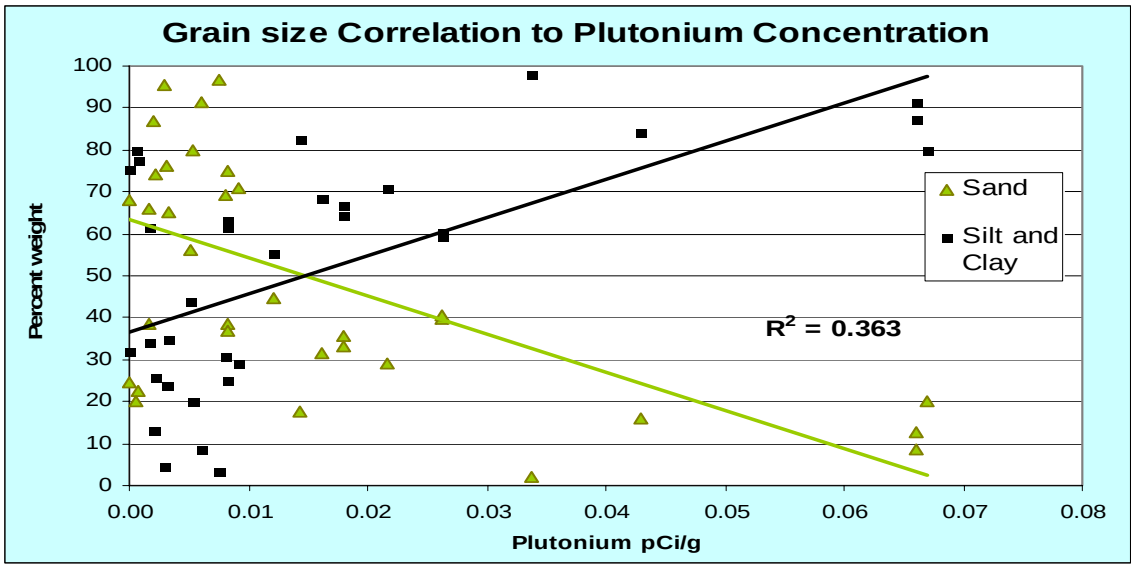

Figure 16. Grain size correlations to 239/240 plutonium concentrations

Los Alamos National Laboratory assumes that if a particular radioactivity value from a river or reservoir sediment sample exceeds the computed upper limit background value, then it probably came from a LANL effluent source. If the observed radioactivity is less than the computed upper limit for background, they likewise assume that it originated from fallout (McLin and Lyons, 2002). This report identifies LANL contaminants in sediments measured below the background reference. Contaminants in sediments become diluted with time and distance as they mix with sediments containing background constituents. Concentrations also vary in regard to the particle size distribution in sediments; samples containing a large fraction of coarse-grained materials demonstrate lower contaminant concentrations, often below reference values used to identify Laboratory impacts.

By normalizing the plutonium concentrations to the silt and clay fraction (dividing the concentration by the percent weight silt and clay / total weight) we can reduce the variability found in the plutonium concentration introduced by different grain size content. This method provides a relatively inexpensive procedure for gaining an insight into the source of plutonium measured in individual sediment samples, particularly those that contain concentrations of plutonium below the computed upper limits for background. We found that many samples containing concentrations of plutonium below the computed background reference levels include legacy plutonium from the Laboratory. These observations were supported by the TIMS measurements of plutonium content in samples collected in the terraces along the Rio Grande.

The following chart in Figure 17 demonstrates the relationship between $^{239/240}$Pu concentrations and their associated normalized values. The horizontal axis categorizes the samples we collected at five sites in terraces along and eight sites in the Rio Grande. The samples were collected at horizons of increasing depth, or in the case of the channel sites from upstream to downstream, illustrated from left to right. The black columns reflect plutonium activity measured by alpha spectroscopy. Its paired value is colored light blue, and reflects the normalized plutonium activity. The left vertical axis is the $^{239/240}$Pu activity scale in pCi/g (total), and the right axis is the normalized scale value, $^{239/240}$Pu activity in pCi/g (fines).

The horizontal dashed line represents the computed upper limit background value of 0.013 pCi/g. The small diamonds and numbers above the columns represent the 10 TIMS measurements in this study. They reflect the calculated component of Laboratory $^{239/240}$Pu by percent in the associated sample.

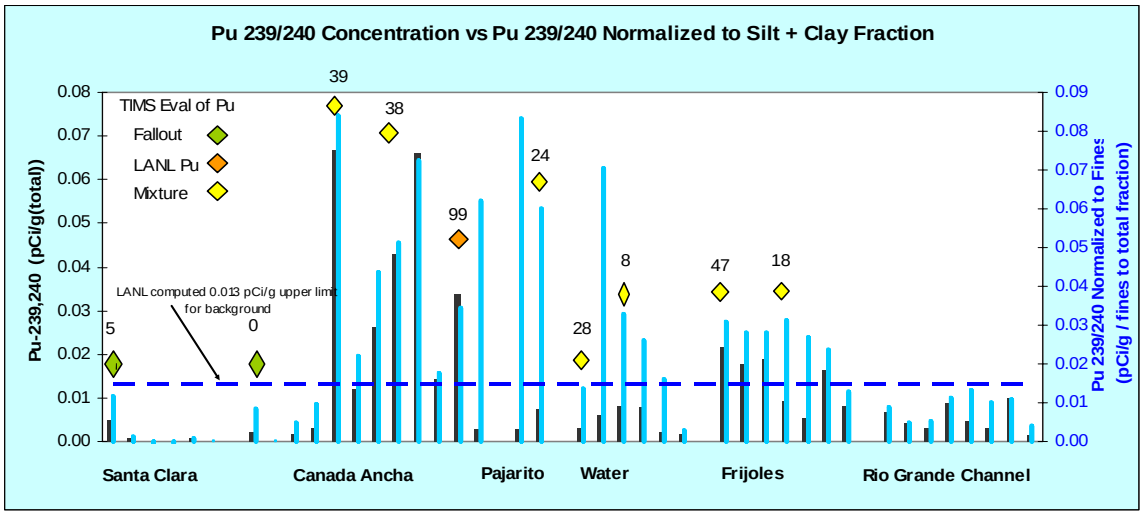

Figure 17. Plutonium 239/240 concentration to normalized value and legacy contaminant proportion relation in samples

Using particle size distribution analysis and plutonium correlations, we identified depositional mechanism and time, as well as provided supporting evidence for conclusions made from other evaluation techniques. We identified global fallout plutonium or pre-nuclear age conditions in sediments at the Santa Clara site, surface sediments at the Cañada Ancha site, and the deepest sampled sediments at Water and Frijoles sites. We also demonstrated legacy plutonium exists in sediments downstream of Los Alamos Canyon originating from episodic flooding and Cochiti Reservoir lacustrine deposition.

The Rio Grande channel samples represent current sediment transport in 2003. The plutonium measurements reflect transport of global fallout materials similar to that found in the upper Rio Grande watershed. Contaminants that may be added at the Los Alamos Canyon confluence are attenuated or may only be recognized during episodic flooding from the Pajarito Plateau canyons.

Overall, we found that any evaluation of contaminants in sediments should always include consideration of grain size content. We identified coarse grained sediments containing legacy wastes that might normally be considered free of them. Samples from the lowest sediment horizon at the Cañada Ancha site as well as those from the Pajarito site show the dilution effects of coarse grain material in the samples. Although the plutonium concentrations measured in the samples were below the background reference value, other evaluation techniques identified the existence of a LANL plutonium component.

Site Discussions

This section describes the conditions at each site considered in this report and is based on the data and methods described above, including: 1) individual contaminant measurements compared to background reference values, 2) statistical comparisons of

data populations from background sites and sites downstream of the Laboratory, 3) TIMS analytical method, developed to specifically identify plutonium sources, and 4) grain size distribution evaluations and correlations to contaminant measurements.

Santa Clara Site

The sites at Santa Clara, 12.1 km (7.5 miles) upstream of the Los Alamos Canyon and Rio Grande confluence, represent background conditions. We collected samples from six horizons ranging in depth from 5 to 158 cm. The sediment units are abandoned floodplains that were active from 1941 to 1968, and prior to 1941. A loamy floodplain sample from 5 to 30 cm reflects sediments deposited from 1941 to 1968, during peak fallout loading. The remaining samples are silt loam to loam floodplain sediments deposited prior to 1941, except a sand unit that is probably an earlier abandoned channel or sand bar buried by the floodplain units. These samples were collected from 34 to 158 cm depths.

All of the $^{239/240}$Pu concentrations, measured by alpha spectroscopy, are less than the 0.013 pCi/g upper limit background reference level. The plutonium measurement for the shallow sediments deposited during 1941 to 1968 is 0.0051 pCi/g. The remaining values range from -0.0006 to 0.0008 pCi/g. These measurements, as well as the other radionuclide measurements obtained for this report suggest there is not a Laboratory impact to these sediments.

The normalized values also identify low to no plutonium content in the silts and clays at the Santa Clara site, representing a background source. However, the TIMS measurement suggests that up to 5% of the plutonium mixture is from Los Alamos. This may be from air emissions during early operations at the Laboratory, fallout primarily from U.S. atmospheric testing, or simply the uncertainty associated with the plutonium evaluation using the TIMS methods.

The sediments represent deposition during the pre-nuclear age as well as during periods of maximum atmospheric fallout.

Cañada Ancha Site

The Cañada Ancha site, 5.1 km (3.2 miles) downstream of the Los Alamos Canyon and Rio Grande confluence, reflect Laboratory impacts as well as background conditions. The site was an abandoned channel, or slough, except during floods. During 1940 through 1958 it was a relatively inactive channel except during flood periods that gradually filled it with sediment, and after a 1967 flood, the slough remnant was completely filled with sediment from flood slack water (Graf, 1993).

We collected samples from 12 horizons ranging in depth from the surface to 335 cm. The surface to 101 cm units, are sandy loam sediments reflecting the 1967 flood or later deposits. The deeper sediment units, from 101 to 229 cm, are silty clay to loam floodplain units that were active from 1940 to 1958 or 1967. A sand horizon, 323 to 335 cm deep, underlies these floodplain sediments and is probably an abandoned channel or sand bar unit.

The $^{239/240}$Pu measurements by alpha spectroscopy range from 0.000 to 0.067 pCi/g. The shallow units contain the lowest values, ranging from 0.000 to 0.0033 pCi/g and are lower than the upper limit background reference. They reflect the most recent sediments, deposited after 1967, during over-bank river flows leaving sandy slack-water deposits. These measurements suggest there is not a Laboratory impact in the shallow, recent sediment deposits. The normalized values, as well as a single TIMS measurement, also identify low to no plutonium content in the sandy loam sediments. Although these sediments were deposited after operations began at the Laboratory, the 1967 flood and later deposits did not originate from Los Alamos Canyon.

The highest $^{239/240}$Pu measurements by alpha spectroscopy in this study occurred in the deeper horizons at the Cañada Ancha site. The plutonium concentrations ranged from 0.021 to 0.067 pCi/g in 8 samples collected from the 101 to 229 cm deep sediments. They reflect the silty clay to loam floodplain deposits that were active from 1940 to 1958 or 1967. Most of these values are greater than the 0.013 pCi/g background level and indicate a Laboratory impact. Three TIMS evaluations for these units indicate 38 to 99% of the plutonium is Laboratory derived. The normalized values derived for these deeper sediments also support this evaluation.

The deepest sampled horizon, 323 to 335 cm, is a sandy channel deposit containing a $^{239/240}$Pu concentration of 0.0029 pCi/g. This value is less than the upper limit background level of 0.013 pCi/g and suggests there was not a Laboratory impact during the deposition of these sediments. The normalized value does suggest a substantial Laboratory impact, which indicates the coarse grained sand sediments diluted the plutonium concentration in the sample. A TIMS measurement was not made for this sample.

The plutonium measurements by alpha spectroscopy, the normalized plutonium values, and the TIMS evaluation of plutonium for the Cañada Ancha sediments reflect post nuclear-age background conditions as well as significant Laboratory impacts. The shallowest sediments are recent deposits that originate from the upper Rio Grande and contain little to no Laboratory plutonium. The deeper units may have been deposited during a period of maximum discharge into the Rio Grande from Los Alamos Canyon. These sediments contained the highest levels of plutonium. The deepest unit has a low plutonium concentration in the sediments, but the normalized value suggests the Laboratory impact may have been as great as in the upper horizons. The volume of coarse-grained materials in the sample diluted the plutonium concentration.

The statistical evaluations and individual measurements compared to background references provided evidence that Laboratory derived Cs^{137}, Sr^{90}, and uranium exist in Cañada Ancha sediments.

Pajarito Site

The Pajarito site is 10.9 km (6.8 miles) downstream of the Los Alamos Canyon and Rio Grande confluence, 29.1 km (18.1 miles) upstream from the Cochiti Reservoir dam or

15.5 km (9.6 miles) from the lake extent near the mouth of Alamo Canyon. The Pajarito site may have been inundated at the upper boundaries of Cochiti Reservoir during its maximum fill elevation during the mid 1980s.

We collected samples from two horizons ranging in depth from 34 to 88 cm. The sediments are texturally coarser than those at the Cañada Ancha and Frijoles sites (see Figure 14). These sand units may be lacustrine and result from deposition of bedload as the river-current velocity diminished while entering the lake. Finer material is progressively deposited in larger degrees downstream as the distance from the inlet to the reservoir increases.

The two $^{239/240}$Pu alpha spectroscopy measurements are 0.0029 and 0.0076 pCi/g. Both values suggest no Laboratory impact when compared to the background reference value. On the other hand, the normalized values suggest the Laboratory impact may have been as great here as at the Cañada Ancha site. A single TIMS measurement for the lowest horizon indicates as much as 24% of the plutonium in the sample is derived from the Laboratory.

The sediments may have been deposited during the mid 1980s at the maximum fill stage of Cochiti Reservoir. The site was at the upper limits of the lake where river current velocity quickly diminished, allowing deposition of the coarse materials carried in the sediment load.

Water Canyon Site

The Water Canyon site is 14.0 km (8.7 miles) downstream of the Los Alamos Canyon and Rio Grande confluence and 12.4 km (7.7 miles) from the present lake extent at Alamo Canyon. Cochiti Reservoir, during its maximum fill elevation during the mid 1980s, inundated or flooded this area by up to four meters.

We collected samples from two sites, in close proximity of each other, representing the same stratigraphy. The horizons range in depth from the surface to 116 cm. At the first site, lacustrine loamy-sands lie over an abandoned channel gravel unit. A sand unit, between 51 and 61 cm, may reflect an alternating advance and recession of the lake boundary. At the second site, a silt loam floodplain unit at 91 to 101 cm exists within the gravel unit. The gravel and floodplain units were deposited prior to filling of the Cochiti Reservoir. The remaining horizons are texturally finer than those at the Pajarito site and coarser than those at the Frijoles site (see Figure 14). This may reflect the progressive fining of lake deposits as current velocities diminish and distance from the lake entrance increases.

The plutonium concentrations, ranging from 0.0017 to 0.0082 pCi/g, are below the computed background level, and are similar to those found at the Pajarito site. However, the TIMS results suggest that some (8 to 28%) of the plutonium is derived from the Laboratory. The normalized plutonium concentrations also suggest a Laboratory impact, except in the deepest unit. That unit demonstrates measurements similar to upper Rio Grande background conditions or pre-nuclear age deposits.

The deepest units at this site were probably deposited on floodplains prior the 1940's. The shallower lacustrine sediments were deposited during the mid 1980s at the maximum fill stage of the Cochiti Reservoir, and contain plutonium originating from the Laboratory.

Frijoles Site

The Frijoles site is 19.3 km (12.0 miles) downstream of the mouth of Los Alamos Canyon and 7.1 km (4.4 miles) above the present lake extent at Alamo Canyon. The affects of the dam are plainly evident here; multiple stream channels, mud flats, and high water remnants exist, and increase further downstream towards the reservoir. Cochiti Reservoir, during its maximum fill elevation during the mid 1980s inundated this area by up to 15 meters. In the late 1980's the lake level declined and receded from this area.

We collected samples from six horizons ranging in depth from the surface to 183 cm. The sediments are texturally finer than at the Water Canyon or Pajarito sites, reflecting the greater distance from the river entrance to the lake. The sediments are lacustrine silty to sandy loam, and were deposited during the maximum fill stage of Cochiti Reservoir during the mid 1980s. The two horizons between 61 and 122 cm contained sediments of a more coarse texture, sandy loam and loamy sand. These may have originated from the advancing and receding nature of the lake.

Six $^{239/240}$plutonium measurements by alpha spectroscopy show concentrations similar to those found in Cochiti Lake. An additional quality control measurement was made on the sediments from the 31 to 61 cm horizon. McLin and Lyons (2002) reported an average of 0.0191 pCi/g in Cochiti Reservoir. Our measurements at the Frijoles site average 0.0139 pCi/g and range from 0.0052 to 0.0217 pCi/g. Four of the seven values are greater than the background reference, including the duplicate. The TIMS evaluations in two of those samples identify 18% and 47% of the plutonium is Laboratory derived. These values are similar to findings in previous Laboratory reports. Gallaher and Efurd (2002) reported that on a depth-weight basis, approximately 40% of the plutonium activity in the sediments near the dam is LANL derived.

The normalized data also indicate that the plutonium in all but the deepest horizon is Laboratory derived, including two horizons that contain alpha spectrometry measurements below the background reference. In the deepest horizon, between 152 and 182 cm, the plutonium measurements as well as the normalized values indicate the sediments originated from background sources.

In summary, the plutonium measurements by alpha spectroscopy suggest the lacustrine deposits at Frijoles Canyon originate from both background as well as Laboratory sediment sources. The normalized plutonium values support the conclusions that sediments containing plutonium with concentrations greater than the background reference were Laboratory derived. The TIMS evaluations also support this conclusion. The normalized data suggest that measurements in two horizons containing coarser materials and plutonium measurements less than background were in fact derived from

the Laboratory. The floodplain sediments in the deepest horizon, below 152 cm, were identified as originating from upper Rio Grande background sources.

The statistical evaluations and individual measurements compared to background references provided evidence that Laboratory derived ^{137}Cs, ^{241}Am, and uranium exist at the Frijoles site.

Rio Grande Channel Sites

Eight sediment samples reflecting low energy or slack-water conditions within the Rio Grande channel were collected in 2003. The upper Rio Grande watershed is represented by sites in the Rio Chama at Chamita, the Rio Grande at Pilar, and in the Rio Grande at the Rio Pojuaque, upstream of the Los Alamos Canyon / Rio Grande confluence, 61, 24, and 3.8 km (38, 15, and 2.4 miles), respectively. The sites in the Rio Grande downstream of the Laboratory are located in White Rock Canyon below Los Alamos Canyon and at Ancho Canyon, 0.8 and 15 km (0.5 and 9 miles) south of the Otowi Bridge. Three downstream sites are south of Cochiti Reservoir at Peña Blanca, San Angustura, and Albuquerque, 45, 73, and 134 km (28, 46, and 83 miles), respectively.

Sediments were collected in the Rio Grande channel at areas that demonstrated low energy flows. The stations were in slack-water conditions behind large obstructions such as boulders or in pools. These sediments are predominantly fine grained and in transit.

Plutonium-238, $^{239/240}Pu$, and ^{137}Cs measurements, as well as the normalized plutonium values are all less than their specific BGULs.

These measurements identified the contaminant source as global fallout. Laboratory contaminants could not be identified above or below the Los Alamos Canyon / Rio Grande confluence.

Data Evaluation Conclusions

We reached our conclusions by: 1) comparing individual laboratory measurements to established background references, 2) statistically comparing downstream data sets to background data sets, 3) analyzing a subset of samples with TIMS and evaluating the results by methods described by Gallaher in *"Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley"* (2002) and, 4) correlating particle size distributions and contaminant concentrations measured in the sediment samples.

We identified four main depositional categories: 1) sediments deposited prior to global fallout followed by 2) episodic deposition of sediments containing LANL legacy waste and 3) lacustrine deposits containing a mixture of global fallout and legacy materials, and then 4) covered by floodplain sediments from the upper Rio Grande containing mostly global fallout materials.

We found that the most persistent legacy radionuclide found in terraces downstream of LANL is $^{239/240}$Pu. The largest contaminant concentrations are found at the Ancha site and diminish with distance downstream of the Los Alamos Canyon confluence with the Rio Grande, except where lacustrine deposits from the Cochiti Reservoir in the lower reaches of White Rock Canyon are found. From north to south: the Cañada Ancha site contains the highest concentrations of $^{239/240}$Pu, ^{137}Cs, ^{90}Sr, and uranium isotopes. Contaminant measurements at the Pajarito site are all indistinguishable from background, although we identified potential legacy contaminants at levels diluted below the existing background references. The lowest elevated $^{239/240}$Pu concentrations are found at the Water Canyon site. The $^{239/240}$Pu activities at the Frijoles site are lower than the Ancha site but higher than the Water Canyon site; ^{137}Cs concentrations are elevated but lower than the Cañada Ancha site; and ^{241}Am and ^{235}U are elevated as well.

Plutonium-238, $^{239/240}$Pu, and ^{137}Cs measurements, as well as the normalized plutonium values in Rio Grande channel sediments are all less than their specific BGUL and are indistinguishable from global fallout levels found in the upper Rio Grande watershed. These sediments were sampled in 2003 and not correlated to storm flows from canyons in the Pajarito Plateau. Contaminants introduced by storm flow from Los Alamos canyon may be attenuated by upper Rio Grande watershed contributions of sediment.

Contaminant Sources

The largest proportion of contaminants discharged into the Rio Grande from Los Alamos Canyon occurred during 1951 to 1969 (Graf, 1993). The greatest $^{239/240}$Pu mass, 44 mCi, was transported from Los Alamos Canyon into the Rio Grande in 1957, followed by 22 mCi in 1968, and 18 and 17 mCi in 1952 and 1951. Several years of 10 mCi per year transport rates occurred intermittently during this period. These were all associated with large annual flow volumes or flood flow rates.

With the exception to flood events following the Cerro Grande fire, plutonium transport rates on this order have not been documented since the 1950's and 1960's. Since the fire, we estimate 55 mCi, 24 mCi, and 8 mCi of $^{239/240}$Pu was moved beyond the E060 gage in lower Pueblo Canyon into Los Alamos Canyon and beyond during 2001, 2002, and 2000 respectively (Englert, Ford-Schmid, 2004).

Estimates by Graf were based on undocumented modeled values for plutonium concentrations in bedload derived from ES samples post dating years of maximum plutonium measurements in the upper Los Alamos Canyon watershed. The flood sizes may also have been underestimated. Flood sizes were modeled on rainfall from a single precipitation station in Los Alamos. Laboratory scientists suspect that the accelerated post Cerro Grande fire transport of sediments and plutonium is dwarfed by that prior to 1965 (personal communication S., Reneau, 2007).

The origin of man-made radionuclides in the Rio Grande along White Rock Canyon and downstream includes:

1) untreated and treated radionuclide liquid waste discharged by the Laboratory at three former outfalls;
2) contaminated sediment deposits downstream of the outfalls;
3) ash generated during the 2000 Cerro Grande fire;
4) airborne release and re-suspension from Laboratory operations; and
5) global fallout from above ground nuclear testing, reactor accidents, and satellite re-entry.

Radioactive Liquid-Waste Outfalls

The greatest contribution of the Laboratory legacy radionuclides found in White Rock Canyon, originated from three radioactive liquid waste outfalls. Two outfalls were located in South Fork Acid Canyon, a tributary to Acid Canyon. The first outfall discharged untreated radioactive effluent from early operations at Technical Area (TA)-1. It was replaced by a second outfall from a radioactive liquid waste treatment facility at TA-45. The third outfall, located in DP Canyon, discharged from a second treatment facility at TA-21. DP Canyon is a small tributary canyon to Los Alamos Canyon. Figure 18 shows the location of the TAs and canyons impacted by them.

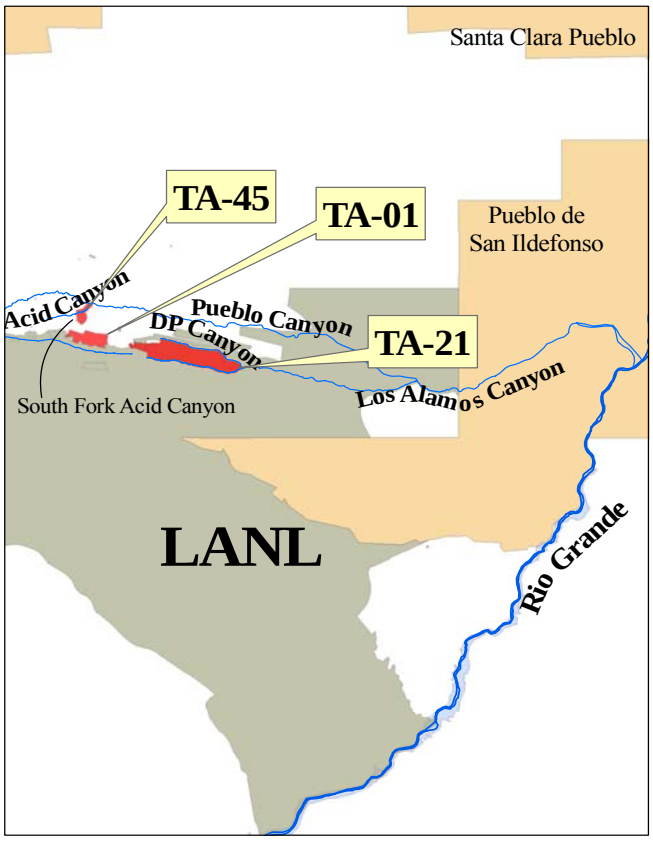

Figure 18. Radioactive liquid waste treatment facilities and canyons they discharge to

Technical Areas-1 and -45

The Laboratory discharged untreated radioactive liquid wastes from TA-1 operations into Acid Canyon from 1943 through April 1951. In 1951, they completed construction of a wastewater treatment plant at TA-45, and began discharge of treated radioactive wastewater into Acid Canyon until June of 1964 (Stoker, 1981). These releases contained a variety of radionuclides, the most predominant being $^{239/240}$Pu, ^{90}Sr, and tritium (^{3}H). Early estimates of the discharge volumes and contaminant inventories discharged into Acid Canyon are compiled in Table 6.

Table 6. Estimate of historical effluent and contaminant release from TAs-01, -45, and -21

Outfall	Location	Years of Operation	Total Volume Released (m^3 / *gal*)	^{241}Am (mCi)	^{137}Cs (mCi)	^{238}Pu (mCi)	$^{239/240}$Pu (mCi)	^{90}Sr (mCi)
TAs - 01 & -45	Acid Canyon	1943 - 1964	629,371 / *165,000,000*	NP	NP	NP	170	94
TA-21	DP Canyon	1952 - 1986	272,896 / *72,100,000*	8.9	28.4	2.6	34.1	9.5

NP - not provided or not available

Data Sources: Stoker et al., 1981; SAIC, 1998; LANL, 1999

Early release estimates indicated 170 mCi of plutonium was discharged into Acid Canyon (Stoker, 1981). In contrast to the effluent-release estimates, Stoker (1981) demonstrated that the total inventory of plutonium in sediments downstream of the outfall was 630 +/- 300 milicurie (mCi), about three times the total release estimates. Recent sediment transport studies and inventory assessments by Lane and others (1985) and Malmon and others (2004) suggest that these earlier estimates may be incomplete and unreliable. They found, based on initial inputs for plutonium-routing calculations, the range of plutonium in the effluent discharge could be up to 3 curries.

Technical Area 21

The third outfall was associated with the plutonium processing facility at TA-21. Treated radioactive liquid waste was discharged into DP Canyon, a small northwest trending tributary that enters the middle reach of Los Alamos Canyon. The effluent releases at TA-21 occurred from 1952 through 1986. Americium-241, ^{137}Cs, $^{239/240}$Pu, ^{90}Sr, and ^{3}H were major constituents in these effluents. The early estimates of contaminant inventories and discharge volumes from TA-21 are presented in Table 6. Recent inventory evaluations as mentioned earlier, suggest that these early estimates may be incomplete and unreliable.

Contaminated Sediment Deposits

Contaminants discharged into the canyons adsorb to sediment particles, and normal fluvial processes distribute and store them downstream in valley-floor sediments. These deposits vary in age, physical character, distribution, contaminant inventory and

concentration, and susceptibility to remobilization. Some deposits are stable for many years while others are quickly remobilized. These sediments are the current sources for radionuclide transport to the Rio Grande and the contaminants stored by them are referred to as the LANL legacy wastes.

The Environmental Protection Agency's Resource Conservation and Recovery Act (RCRA) required the Laboratory to characterize these deposits in the Los Alamos watershed during the 1990s through 2001. The primary objectives of the characterization efforts were to quantify the presence and distribution of contaminants, determine human health and ecological risk, and determine potential cleanup requirements.

Generally, these data show that activity concentrations decrease with distance downstream from the outfall sources, and deposits with higher proportions of finer grained sediments tend to have higher activity concentrations. They found that the highest levels of contamination are associated with fine-grain deposits near the source areas. Table 7 provides contaminant ranges and averages for sediment deposits in DP Canyon, Los Alamos Canyon below the DP Canyon confluence, and Acid and Pueblo canyons. The data in Table 7 was obtained from LANL reports (Reneau and others, 1998a, b, and c).

By 2003, LANL estimated that before the Cerro Grande fire, up to 1,300 mCi remained in Pueblo Canyon from the Acid and Pueblo canyons confluence downstream to the Pueblo and Los Alamos canyons confluence (Reneau and others, 1998). NMED estimated that during 2000 to 2002, stormwater had transported 87 mCi of plutonium, approximately 7% of the inventory, out of Pueblo Canyon (Englert and Ford-Schmid, 2004). NMED's latest estimates, from 2000 through 2006, indicate 199 mCi plutonium 239/240 has been transported from Pueblo Canyon by storm water (report in press). These transport rates may be underestimated due to the complexities associated with multiple surges in flood stage resulting from the random nature of rainfall and contributions of flow from adjacent canyons

In the 1960s and early 1980s, two remediation efforts focused predominantly on the mesa top, were conducted at the TAs-1 and -45 Acid Canyon outfall areas to remove hot-spot residual contamination. Prior to remediation, the highest $^{239/240}$Pu activity concentration found at the outfall areas was 163,000 pCi/g (LANL, 1992). In 1999, NMED collected and analyzed two canyon bottom sediment samples located approximately 213 m (700 feet) below the former outfall areas. The $^{239/240}$Pu results prompted the Laboratory to perform a more detailed characterization. They showed that cleanup was necessary and in 2001 sediments with $^{239/240}$Pu levels greater than approximately 280 pCi/g were removed (Reneau and others, 2002). The highest pre-cleanup activity concentration in the sediments was 7,780 pCi/g.

Prior to two remediation efforts at the TA-21 outfall area in 1996 and 2002, initial site characterization identified radiochemical concentrations as high as 2,600 pCi/g for ^{241}Am, 3,226 pCi/g for ^{137}Cs, 46,000 pCi/g for $^{239/240}$Pu, 2,200 pCi/g for ^{238}Pu, and 1,800 pCi/g for ^{90}Sr (LANL, 1994). After the Cerro Grande fire, LANL also removed

Table 7. Summary of radionuclide characterization data for sediments collected in the Los Alamos watershed

	^{241}Am (pCi/g)			^{137}Cs (pCi/g)			^{238}Pu (pCi/g)			$^{239/240}$Pu (pCi/g)			^{90}Sr (pCi/g)		
	Range (DT/ND)	Avg	1 STD	Range (DT/ND)	Avg	1 STD	Range (DT/ND)	Avg	1 STD	Range (DT/ND)	Avg	1 STD	Range (DT/ND)	Avg	1 STD
TA-21															
DP Canyon[1]	-0.22 - 71 (73/NA)	6.60	10.70	0.27 - 442 (73/NA)	40.80	65.00	0.0127 - 2.79 (55/NA)	0.46	0.56	0.0272 - 48 (55/NA)	4.00	6.80	0.09 - 32.8 (62)	6.20	7.90
Los Alamos Canyon[2]	0.044 - 28 (79/105)	3.30	5.60	0.075 - 230 (152/32)	12.60	33.00	0.015 - 2.01 (68/94)	0.17	0.30	0.034 - 13.8 (140/20)	1.98	2.72	0.84 - 39.56 (39/41)	8.26	10.65
TA-45															
Acid Canyon[3]	0.433 - 101 (30/7)	19.24	29.04	0.118 - 148 (28/11)	10.53	28.75	0.045 - 37.3 (92/21)	2.20	4.84	2.3 - 7780 (111/0)	473.18	1145.86	0.19 - 80 (21/12)	13.20	24.70
Pueblo Canyon	0.11 - 11.5 (20/17)	1.31	2.61	0.42 - 1.53 (22/15)	0.42	0.34	0.014 - 2.08 (101/184)	0.15	0.28	0.005 - 502 (280/3)	11.97	38.56	0.95 - 1.4 (3/11)	1.10	0.24

[1] - Detected versus non-detected values were not differentiated in the DP Canyon data set.

[2] - Data derived from sampling data collected in Los Alamos Canyon below its confluence with DP Canyon.

[3] - Includes data collected in Acid Canyon and tributary that received effluent.

Range - Determined from detections only.

(DT/ND) - Number of detections/Number of non-detections.

Avg - Average calculated from detections only.

1 STD - One standard deviation calculated from detections only.

NA - Not available or not determined.

Data Sources: LANL LA-UR-98-3324, 98-3974, 98-3975, 99-4238, and 02-5785.

contaminated sediment units in Los Alamos Canyon just below the DP Canyon confluence. They had previously identified a large inventory of cesium in these sediments. As a precautionary measure, they removed them before predicted floodwaters transported them downstream.

Local and Regional Fallout

Both local and regional fallout contribute to the radionuclide inventory in the Rio Grande fluvial system. Local fallout is generated by LANL operational releases, including stack emissions, open-detonation testing, and wind re-suspension of contaminated soils. Local fallout began in the early 1940s as part of fission research to produce an atomic weapon. Current operational controls have reduced the contaminant emissions into the local environment.

Above-ground nuclear weapons testing, reactor accidents, and re-entry of plutonium-fueled satellites dispersed radionuclides into the atmosphere that eventually fell back to earth. Five nations detonated 484 nuclear devices in the atmosphere from 1945 to 1986. The largest number of atmospheric tests in a year, 96, occurred in 1958. This was followed by 81 open air detonations in 1952. The Partial Test Ban Treaty became effective in 1963 and the U.S., Soviet Union, and the United Kingdom confined further tests to underground. Perkins and Thomas (1980) estimated 363 kCi of $^{239/240}$Pu was distributed onto the earth's surface by 1980 (Graf, 1993).

Atmospheric fallout occurs within days to years of detonation. Larger particles ranging in size from 100 to 200 microns fall to the earth locally within days. Particles smaller than 100 microns, are ejected to greater altitudes, distributed more widely, and take months to fall back to earth. Particles less than 10 microns take years before deposition and are distributed throughout the entire hemisphere. Early fallout from open air testing at the Nevada Test Site (NTS) probably reached the San Juan Mountains and the Rio Grande watershed – northwest New Mexico and Southwest Colorado. The maximum concentrations from the NTS are distributed in the northern hemisphere within a band between latitudes 40 and 50 degrees north. Contaminant loading to the Northern Rio Grande system was probably greatest in the 1960's.

Cerro Grande fire

As a result of the 2000 Cerro Grande fire, the watersheds from Santa Clara Canyon to Water Canyon were burned, especially the upper portions along the mountain front. Previous studies show that ash derived from the fire contains higher amounts of radioactivity than do the local soil and sediment. For example, Kraig and others (2002) documented that the ash contains about eight times as much ^{137}Cs as does the local background soils.

After the fire, we collected ash that represented burned overstory and understory components of the forest. We found contaminant concentrations were statisticaly different and larger than contaminants in background soil. For example, we derived a 0.6

pCi/g reference level that reflected the probable largest value of $^{239/240}$Pu in ash. This level was 30 times greater than the LANL 0.02 pCi/g regional reference value for soils.

We also studied $^{239/240}$Pu measurements in ash-laden sediments in stream channels and on channel banks. The sediments were collected from burned upper-watershed areas downstream to the banks of the Rio Grande including 15 ash-laden bank deposit samples collected near or along the Rio Grande. Those measurements indicated the plutonium concentrations were diminishing with time and distance from the areas impacted by the fire. The plutonium in ash became diluted as the it mixed with clean soils and sediments.

Twenty-two ash and ash-laden sediment samples collected soon after the Cerro Grande fire from the burned forest floor area and in drainages downstream of the burn area were used to develop the 0.6 pCi/g $^{239/240}$Pu in ash reference. This group did not include the 15 ash-laden bank deposits near or along the Rio Grande. The bank samples near the Rio Grande demonstrated significant dilution and the average $^{239/240}$Pu value, 0.06 pCi/g, was only slighty above the local LANL background (Ryti and others 1998).

Hopkins (2001) and Katzman and others (2001) also showed that the ash from the Cerro Grande fire contained elevated levels of $^{239/240}$Pu, and other radionuclides, compared to that of ash samples collected in the 2000 Viveash fire area. The Viveash fire occurred shortly after the Cerro Grande fire. This fire was located in the Sangre de Cristo Mountains, approximately 90 kilometers to the east of Los Alamos, and is separated from LANL by the Rio Grande basin. A portion of the plutonium in the Cerro Grande fire ash may have been from historic air stack emissions at the Laboratory (Gallaher and Koch, 2004, LANL Environmental Surveillance at Los Alamos during 2000, Katzman, Ryti, and Reneau, 2001). These findings indicate that the ash-laden sediments transported from the burn areas contained elevated levels of radionuclides and became a short term source of contaminants to the Rio Grande. See Appendix E for additional information about transport since the Cerro Grande fire.

Contaminant Transport

Fluvial processes in Los Alamos and Pueblo canyons are the primary mechanisms for legacy radionuclide transport to the Rio Grande. Surface water flow in the watershed is ephemeral and/or intermittent with one short (0.8 to 1.6 km) spring-fed perennial reach in the upper portion of Los Alamos Canyon. Surface water runoff usually occurs during the spring snowmelt and summer monsoon storm season. Snowmelt often starts in March and ends in May, transporting low suspended sediment loads ranging from 0.1 to 1.0 gram per liter (g/L). Snowmelt runoff to the Rio Grande frequently occurs in Los Alamos Canyon but not Pueblo Canyon. Summer thunderstorm runoff events in Los Alamos and Pueblo canyons occur episodically with durations lasting several hours. Storm runoff carries suspended loads up to five orders of magnitude greater than snowmelt runoff. However, over time, the total sediment inventory transport has been reported to be greater during the relatively long spring runoff periods than from short summer storm flows.

The Laboratory collected runoff data during the late 1960's and mid 1970's and showed that 1) on a per unit weight basis, radionuclide activities are higher in the suspended sediment than that of bed sediments (Purtyman, 1990); and 2) the radionuclide inventory transported down stream is greater for bed sediments, especially during snowmelt runoff (Stoker, 1981). This phenomenon is due to annual snowmelt flow durations being longer and transporting far greater amounts of sediment than infrequent, short duration stormwater flows.

William Graf evaluated early stormwater runoff rates and plutonium concentrations in lower Los Alamos canyon and reported his conclusions in a report published in 1993 (Graf, 1993). He estimated 188 mCi of plutonium was transported from Los Alamos Canyon into the Rio Grande by stormwater runoff from 1944 to 1986. Graf suggested that the contribution to the plutonium budget from Los Alamos Canyon is associated with relatively coarse sediment, which often behaves as bedload. Infusions of these materials into the Rio Grande were largest in 1951, 1952, 1957, and 1968. He suggested that the Los Alamos Canyon contribution to the entire plutonium budget in the Rio Grande watershed is relatively small (~10%) in relation to global fallout contributed from the upper Rio Grande watershed. However, in these four critical years it constituted 71 – 86 percent of the plutonium in bedload immediately down stream from Otowi Bridge (Graf, 1993). These transport values may be largely underestimated. Graf reported modeled data based on limited information and may have underestimated transport by an order of magnitude or more (S. Reneau, personal communication, 2007).

The Laboratory continued to periodically monitor snowmelt and stormwater transport of $^{230/240}$Pu and ^{238}Pu during the 1970s to the mid 1990s. The data sets are small and were collected at three locations downstream of contaminant sources in the Los Alamos watershed. The collection sites are in Los Alamos Canyon at NM State Road 4 above its junction with Pueblo Canyon, in Pueblo Canyon at NM State Road 502 above its junction with Los Alamos Canyon, and in Los Alamos Canyon at the Rio Grande. Figure 19 shows these locations and only partly represents the current stormwater monitoring by NMED and the Laboratory in the Los Alamos watershed.

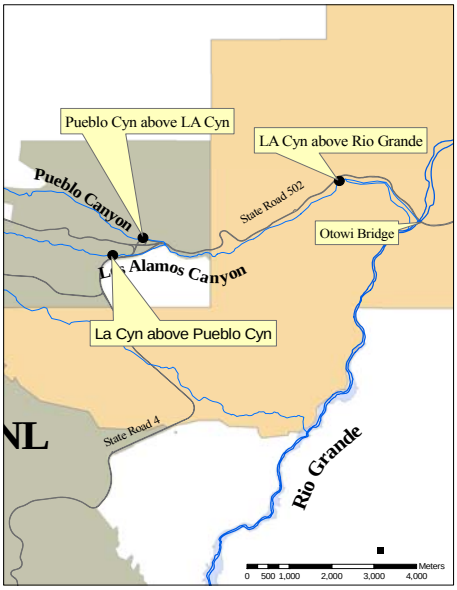

Figure 19. Historical storm water monitoring locations

We did not recognize concentration trends in the data sets, possibly due to the small amount of temporal and spatial data. However, inventory transport to the Rio Grande diminished during the 1970s through the 1990s due to climatic changes. Few snowmelt or storm flows reached the Rio Grande through Los Alamos Canyon.

In the snowmelt samples, $^{239/240}$Pu concentrations in suspended sediments range from 0.0 to 31.1 pCi/g. The mean concentration in suspended

sediments from Los Alamos Canyon at State Road 4 is 3.32 pCi/g. The mean plutonium concentration at the Pueblo Canyon State Road 502 site is 4.33 pCi/g, and at the Los Alamos Canyon / Rio Grande station it is 1.62 pCi/g. These references suggest higher concentrations of plutonium originate in Pueblo Canyon and upper Los Alamos Canyon but dilute as sediments are transported downstream to the Rio Grande.

After the Cerro Grande fire in May of 2000, the Laboratory and NMED increased stormwater monitoring efforts from watersheds affected by the fire. After the 1977 La Mesa fire and the 1996 St. Peters Dome fire, Jack Veenhuis showed that the magnitude and frequency of floods after forest fires could dramatically increase (Veenhuis, 2002). These fires occurred in the Jemez Mountains south and southwest of the area burned by the 2000 Cerro Grande fire. Chemical analysis of ash generated during the Cerro Grande fire demonstrated elevated levels of radionuclides, heavy metals, and other contaminants. These measurements, as well as the potential of greater flood magnitudes and frequencies, suggest an additional contaminant source would become available for transport to the Rio Grande.

NMED found that most of the ash and associated contaminants were transported to the Rio Grande within two years after the fire. More significantly, we also found that the increased flooding destabilized the canyon channels, and legacy contaminants stored there were being transported to the Rio Grande. NMED showed that by 2002, 87 mCi of $^{239/240}$Pu had been transported beyond Pueblo Canyon in stormwater suspended sediments alone (Englert and Ford-Schmid, 2004). Our latest estimates, from 2000 through 2006, indicate 199 mCi plutonium 239/240 has been transported from Pueblo Canyon by storm water (report in press). Transport rates of legacy wastes stored in Pueblo and Los Alamos canyons have not been as great as these since the 1950's and 1960's when plutonium concentrations in the canyons were much greater.

Most of the Laboratory contaminants that we measured in the White Rock Canyon sediments were probably transported to the Rio Grande from the Los Alamos watershed. The Cerro Grande fire occurred after the beginning of this study and was not part of the original considerations. However, the repercussions of the Cerro Grande fire have caused a substantial increase of legacy contaminant transport to the Rio Grande. Legacy contaminant transport rates currently being measured in Los Alamos watershed storm water have not been so large since the early years of Laboratory operation when they discharged untreated effluent to the canyons and seasonal flood events were more frequent.

Additional discussion regarding historical contaminant transport and stormwater transport since the Cerro Grande fire can be found in Appendix E Historical Contaminant Transport and Transport since the Cerro Grande Fire.

To summarize, we considered the following to be the primary processes and mechanisms for contaminant transport to the Rio Grande:

1. Contaminant transport occurs by spring snowmelt, primarily in Los Alamos Canyon, and summer-storm runoff in both, Los Alamos and Pueblo canyons.
2. Longer intervals of spring snowmelt deliver a significant amount of bedload sediment containing smaller concentrations of contaminants to the Rio Grande.
3. Storm runoff carries suspended sediment loads up to five orders of magnitude greater than snowmelt.
4. Most radionuclides and metals tend to adhere to the finer-grained suspended sediment fraction at higher concentrations than coarse grained sediments.

Methodology

The methods we used to evaluate the Laboratory contaminant distribution in White Rock Canyon terrace and Rio Grande channel sediments are described in this section. This section describes how we collected samples, determined the appropriate analytical services, evaluated the chemical data, and provided the quality control necessary to complete the assessment of Laboratory contaminants in White Rock Canyon sediments.

Core and Outcrop Sampling

Two boreholes were excavated at each site, using a 6.35-cm diameter by 30.5 cm length stainless-steel sand auger with varying lengths of steel extensions. The boreholes were augured to a depth of refusal, usually when cobble and gravel channel beds were encountered. The auger bucket was inserted to the bottom of the borehole and turned down its length to obtain sequential 30.5 cm cores. It was extracted and the cores placed in sequence on a long board or length of plastic sheeting marked in metric increments, where the sample composition was identified and described. The auger was then re-inserted to acquire additional increments of core sample until it was no longer able to penetrate the substrate.

The first core provided stratigraphic information, or lithologic correlations, later used to determine the sampling intervals - the second core provided the specific samples. The sampling intervals were selected based on textural characteristics such as grain size and mineralogy. We collected samples from homogenous layers that represent alternating and different depositional events; for example, coarse grain units might reflect high-energy flood events, while fine grain units might reflect low-energy slack water depositional characteristics. The auger was pre-cleaned by scrubbing with a detergent (Alconox™) solution and rinsed with de-ionized water prior to re-inserting it into the borehole for each sample increment.

Samples at the Water Canyon site were collected from an outcrop and were also selected based on lithology. The outcrop face was scraped with a shovel to expose fresh stratigraphy. The lithologic units were described and sampled in place. Like the core samples, homogenous units that reflect alternating deposition were sampled with a pre-cleaned stainless-steel scoop.

All samples were placed in dedicated polyethylene bottles, labeled, chilled and kept on ice at 4 +/- 2^{o} Celsius, and shipped to a contract analytical laboratory for chemical analyses. All field data such as sample depth and lithologic descriptions were documented in logbooks at the time of sampling and can be reviewed in Appendix F.

Rio Grande Channel Sampling

Three sites represent the upper Rio Grande watershed which includes the San Juan Mountains in southern Colorado and the Rio Chama in northern New Mexico. The sites are distant from the Laboratory, the potential of an impact from the facility is small, and they reflect global fallout from atmospheric testing of nuclear weapons. The remaining five sites are downstream of Laboratory, two in White Rock Canyon and three below the Cochiti Reservoir. These sites have the potential to exhibit legacy contaminant transport from the Laboratory.

At each site, we used a Petite Ponar dredge to collect sediments from areas that appeared to have the lowest stream velocities, such as in pools or eddies behind large natural or man-made obstructions like boulders and stream gage or grade control structures. These areas provided fine grained sediments from the active channel of the Rio Grande and represent current sediment transport.

The Petite Ponar dredge was pre-cleaned by scrubbing with a detergent (Alconox™) solution and rinsed with de-ionized water prior to sample collection at each site. The dredge was lowered into the channel from catwalks at stream gages or irrigation diversions and allowed to accelerate from the water surface to the channel bottom penetrating the water sediment interface. At some locations a sampler waded into the stream channel to access the best locations. After the device was embedded into the channel bottom, a messenger was slipped down a rope activating the closure of the spring loaded dredge. The sample in the dredge was then hauled to the surface and poured into a stainless steel bowl. These steps were repeated until a sufficient amount of sediments were acquired.

The sediments in the stainless steel bowl were thoroughly mixed and then placed in dedicated polyethylene bottles, labeled, chilled and kept on ice at 4 +/- 2^{o} Celsius, and shipped to a contract analytical laboratory for chemical analyses.

Sample Analysis

The sediment samples were analyzed at multiple analytical laboratories. The laboratories used routine analytical methods prescribed by DOE and EPA federal agencies, or equivalent methods described by professional organizations, and developed by individual laboratories.

Desert Research Institute conducted the Particle Size Distribution Analysis or grain-size analysis for our sediment samples. Their procedure is based on the combination of pipette and dry sieving procedures to determine particle size distribution of sand, silt, and clay in soil and sediment samples as specified by the USGS and the Soil Science Society

of America and reported by Day (1965), Jackson (1969), Janitzky (1986), and Gee and Bauder (1986).

Paragon Analytics, Inc (PAI) and the Laboratory's Los Alamos Clean Chemistry and Mass Spectrometry Laboratory conducted the radiochemical analyses. PAI utilized alpha spectrometry methods that meet or exceed the requirements referenced by DOE/EML 4.5.2.1. used to identify and quantify alpha-emitting radionuclides in solids. The radionuclides include ^{241}Am, ^{237}Np, ^{238}Pu, $^{239/240}$Pu, ^{234}U, ^{235}U, and ^{238}U. PAI used low-background gas-flow proportional counting methods to measure ^{90}Sr. These methods meet the calibration, data collection, and analysis requirements of EPA method 900.0. They measured ^{137}Cs and other gamma emitters such as ^{60}Cobalt using gamma spectroscopy methods that are equivalent or exceed EPA Procedure 901.1 and DOE/EML Procedure 4.5.2.3.

The Mass Spectrometry Lab developed an analytical technique titled TIMS, capable of plutonium isotope speciation. Plutonium-239 and -240 isotopes are indistinguishable by alpha spectroscopy because they have very similar emission energies at approximately 5.2 MeV. The TIMS technique provides very precise and accurate atomic mass measurements. A reliable evaluation of plutonium sources can be made using this method. This information is acquired by comparing the atomic ratio of ^{239}Pu and ^{240}Pu isotopes. For example, plutonium released at the Laboratory has a distinct atomic ratio (^{240}Pu/^{239}Pu) different than plutonium derived from above-ground testing at the Nevada Test Site.

See Appendix D for a more detailed description of the analytical methods used for this report**.**

Quality Control

Evaluation of duplicate measurements is part of a quality control process used in determining accuracy and precision of environmental data. We evaluated blind field duplicates as well as laboratory duplicates as prescribed in our Program's standard operation procedures. In this case, field and laboratory duplicates were measured for multiple parameters and evaluated by their duplicate error ratios or DERs. DERs quantify the difference between duplicate measurements, particularly those that result in low values containing relatively large uncertainty. Radiochemical measurements in environmental samples commonly result in low values containing uncertainty that is equal or exceeds the measurement.

Sixty-one duplicate measurements were made: 17 field and 44 laboratory duplicate evaluations. These evaluations were made for two blind field duplicates and 12 laboratory duplicate samples. The analyses include ^{241}Am, ^{137}Cs, ^{237}Np, ^{238}Pu, $^{239/240}$Pu, ^{90}Sr, ^{234}U, ^{235}U, and ^{238}U.

A field duplicate is a single homogeneous sample prepared on-site and divided into two sub-samples before being submitted to a laboratory and analyzed using identical analysis.

These sub-samples are commonly referred to as split samples. Analytical duplicates are similar except laboratory technicians split a field sample into duplicate sub-samples at the laboratory. The laboratory DER is the common calculation made to measure repeatability, quantifying accuracy and precision of analytical techniques. The results, as well as the measurement uncertainty, are evaluated to determine the validity of the duplicate measurements. The DER is defined below:

$$DER = \left(\frac{|S - D|}{2 * \sqrt{\sigma_S^2 + \sigma_D^2}} \right)$$

Where:

$|S - D|$ = the absolute value of the difference between a sample measurement (S) and a duplicate measurement (D).
σ_S^2 = the square of the total propagated uncertainty (1 sigma) of the sample measurement
σ_D^2 = the square of the total propagated uncertainty (1 sigma) of the duplicate measurement

The DER gives the degree to which the sample and duplicate measurements are comparable. A DER less than or equal to 1.42 indicates the results are statistically equivalent to a 95% confidence level. A DER greater than 1.42 and less than 2.13 places the results in a warning range, and greater than 2.13 places the results outside a 3σ uncertainty, or 99.7% confidence level.

All duplicate measurements and the DERs are compiled in Table 8. A narrative summary of the laboratory quality control evaluation is found in Appendix G. The narrative includes steps that were taken to rectify analytical problems, if necessary, to provide data that meets normal laboratory quality specifications.

We found that all DER evaluations for our data set were less than 2.13. The average DER for all measurements is 0.03, and the maximum value is 0.19. All duplicate measurements fall within an acceptable range and suggest the analyses for this project are accurate and precise.

We also had 10 duplicate samples analyzed for $^{239/240}$Pu activity concentrations using different analytical techniques. Sample duplicates were submitted to both the Los Alamos Clean Chemistry and Mass Spectrometry Laboratory, and Paragon Inc., a commercial analytical laboratory. The Los Alamos Clean Chemistry and Mass Spectrometry Laboratory measured the samples by TIMS, and Paragon measured the samples using alpha spectroscopy methods. The plutonium mass, reported for ^{240}Pu and ^{240}Pu by TIMS, were converted to activity concentrations (pCi/g) and compared to the activity concentrations reported by alpha spectroscopy measurements reported by Paragon.

Table 8. Duplicate error ratios between duplicate samples

Description	^{238}Pu (pCi/g)	unc	DER
sample	0.0006	0.0015	
(lab duplicate)	0.0002	0.0011	0.00
sample	0.0007	0.0015	
(lab duplicate)	0.001	0.0023	0.00
sample	-0.0020	0.0032	
(lab duplicate)	0.0084	0.0042	0.06
(field duplicate)	0.0023	0.0029	0.03
sample	0.0026	0.0038	
(field duplicate)	0.0011	0.0036	0.01
sample	0.0033	0.0037	
(lab duplicate)	-0.0041	0.0043	0.04
sample	0.0005	0.0026	
(lab duplicate)	-0.0051	0.0043	0.03
		ave	0.02
		min	0.00
		max	0.06

Description	$^{239/240}$Pu (pCi/g)	unc	DER
sample	0.0051	0.0031	
(lab duplicate)	0.0033	0.0019	0.01
sample	0.0017	0.0015	
(lab duplicate)	0.0028	0.0024	0.01
sample	0.066	0.012	
(lab duplicate)	0.068	0.013	0.01
(field duplicate)	0.069	0.013	0.01
sample	0.0179	0.0083	
(field duplicate)	0.0188	0.0061	0.00
sample	0.0162	0.0079	
(lab duplicate)	0.0094	0.0072	0.03
sample	0.0082	0.0051	
(lab duplicate)	0.0104	0.0073	0.01
		ave	0.01
		min	0.00
		max	0.03

Sample	^{90}Sr (pCi/g)	unc	DER
Sample	-0.01	0.25	
(lab duplicate)	0.14	0.25	0.10
Sample	0.07	0.09	
(lab duplicate)	0.06	0.09	0.01
Sample	0.40	0.12	
(field duplicate)	0.36	0.11	0.04
Sample	0.17	0.17	
(lab duplicate)	0.15	0.15	0.02
Sample	0.23	0.23	
(field duplicate)	0.12	0.09	0.11
		ave	0.06
		min	0.01
		max	0.11

Description	^{234}U (pCi/g)	unc	DER
Sample	0.90	0.16	
(lab duplicate)	0.92	0.16	0.02
Sample	0.76	0.11	
(lab duplicate)	0.78	0.12	0.02
Sample	1.13	0.16	
(field duplicate)	1.15	0.17	0.02
(lab/field duplicate)	1.14	0.16	0.01
Sample	0.444	0.091	
(lab duplicate)	0.441	0.087	0.00
Sample	0.73	0.13	
(lab duplicate)	0.59	0.11	0.14
Sample	1.31	0.2	
(field duplicate)	1.26	0.18	0.04
		ave	0.04
		min	0.00
		max	0.14

Description	^{235}U (pCi/g)	unc	DER
Sample	0.039	0.022	
(lab duplicate)	0.055	0.025	0.04
Sample	0.033	0.016	
(lab duplicate)	0.036	0.017	0.01
Sample	0.055	0.022	
(field duplicate)	0.079	0.028	0.05
(lab/field duplicate)	0.081	0.026	0.00
Sample	0.020	0.019	
(lab duplicate)	0.025	0.021	0.01
Sample	0.063	0.03	
(lab duplicate)	0.064	0.028	0.00
Sample	0.091	0.037	
(field duplicate)	0.079	0.027	0.03
		ave	0.02
		min	0.00
		max	0.05

Description	^{238}U (pCi/g)	unc	DER
Sample	0.68	0.13	
(lab duplicate)	0.71	0.13	0.03
Sample	0.80	0.12	
(lab duplicate)	0.78	0.12	0.02
Sample	1.33	0.19	
(field duplicate)	1.24	0.18	0.07
(lab duplicate)	1.14	0.16	0.08
Sample	0.432	0.089	
(lab duplicate)	0.479	0.092	0.05
Sample	0.72	0.13	
(lab duplicate)	0.66	0.12	0.06
Sample	1.28	0.2	
(field duplicate)	1.10	0.16	0.15
		ave	0.07
		min	0.02
		max	0.15

Description	^{241}Am (pCi/g)	unc	DER
Sample	0.003	0.011	
(lab duplicate)	0.008	0.012	0.02
Sample	0.0027	0.0078	
(lab duplicate)	0.0081	0.0084	0.02
Sample	0.020	0.011	
(field duplicate)	0.029	0.014	0.03
(lab/field duplicate)	0.027	0.014	0.01
Sample	0.0008	0.0011	
(lab duplicate)	0.0009	0.0013	0.00
Sample	0.022	0.01	
(field duplicate)	0.007	0.01	0.05
Sample	0.0086	0.009	
(lab duplicate)	0.0137	0.0084	0.02
Sample	0.0114	0.0071	
(lab duplicate)	0.0096	0.0076	0.01
		ave	0.02
		min	0.00
		max	0.05

Description	^{237}Np (pCi/g)	unc	DER
Sample	0.00579	0.01808	
(lab duplicate)	-0.0172	0.01744	0.06
Sample	-0.0029	0.01946	
(lab duplicate)	-0.0019	0.0206	0.00
Sample	0.00574	0.02168	
(field duplicate)	-0.0023	0.02292	0.02
Sample	-0.0009	0.00448	
(lab duplicate)	-0.0030	0.00752	0.01
Sample	-0.002	0.012	
(lab duplicate)	0.0025	0.0098	0.02
Sample	0.007	0.014	
(field duplicate)	-0.0060	0.02017	0.03
(lab/field duplicate)	-0.0028	0.01885	0.01
Sample	-0.004	0.011	
(lab duplicate)	-0.0007	0.0092	0.01
		ave	0.02
		min	0.00
		max	0.06

Description	^{137}Cs (pCi/g)	unc	DER
Sample	0.102	0.031	
(lab duplicate)	0.095	0.035	0.01
Sample	0.70	0.20	
(field duplicate)	0.78	0.30	0.05
(lab duplicate)	1.06	0.41	0.19
Sample	0.091	0.028	
(lab duplicate)	0.104	0.089	0.02
Sample	0.172	0.086	
(lab duplicate)	0.175	0.084	0.00
		ave	0.06
		min	0.00
		max	0.19

Table 3, in the "Thermal Ionization Mass Spectroscopy Evaluation" section discussed earlier in this report, summarizes the TIMS and alpha spectroscopy duplicate measurements for $^{239/240}$Pu in the sediments we collected along the Rio Grande. The first column includes a general description of the site as well as depths from which the samples were collected. The following two columns are the activity measurements of the ^{239}Pu and ^{240}Pu isotopes. They are derived from TIMS measurements of atoms of plutonium per gram in each sample multiplied by the specific activity for the appropriate isotope. The fourth column is the sum of the activities derived from the two isotopes. The sixth column contains the measurements made by alpha spectroscopy. Although the TIMS and alpha spectroscopy measurements are similar, the differences demonstrate the data variability. The variability includes random and systematic error associated with the

sampling and analytical procedures. The Duplicate Error Ratio, listed in the last column, is a measure of the similarities between the TIMS and alpha spectroscopy measurement of plutonium concentration in the samples.

We found that all but one DER evaluations were less than 2.13. One DER for a measurement at Cañada Ancha site was 3.65; outside the acceptable 2.13 duplicate range. Plutonium measurements for the 101 cm horizon at Cañada Ancha site were 0.0231 pCi/g, by TIMS, compared to 0.067 pCi/g, by alpha spectroscopy. Although the alpha measurement was almost three times the TIMS measurement, the difference did not change the conclusions made from these measurements.

Comparison of Radionuclide Health Risks

We asked the U.S. Environmental Protection Agency (EPA, Region 6 office in Dallas, Texas) to perform a hypothetical health risk evaluation at two sites along the Rio Grande. The objective of this exercise was to provide a comparison of relative risk associated with the cumulative effects of multiple radionuclides at each location. The assessment was performed using identical and overly conservative input parameters at each site, the upstream Santa Clara site and the downstream Ancha site. The Santa Clara site is accessible to the local community and has been used for agricultural purposes, while the Ancha site is isolated within the White Rock Canyon.

In evaluating the risk from radionuclides in sediments along the Rio Grande, EPA used the Superfund Preliminary Remediation Goal (PRG) for Radionuclides Risk Calculator that can be found at **http//epa-prgs.ornl.gov/radionuclides/prg search.shtml**. A residential scenario (long term occupancy) was used for each evaluation, and is based upon six years of exposure as a child and 24 years as an adult. This scenario includes direct ingestion of soil, inhalation of fugitive dusts, external exposure to radionuclides in the soil, and ingestion of homegrown produce. The risk calculator also includes the contribution of decay-chain products, if applicable. Table 9 contains the residential scenario parameters used in the Superfund PRG for Radionuclides Risk Calculator and provides the most conservative evaluation. The reader should be aware that risk assessments are normally site specific and input parameters are adjusted for different scenarios, for example recreational, agricultural, or industrial.

Table 9. Superfund PRG residential parameters for radionuclide risk calculator

Target Risk (unit less) - 1.0 E-6	Total Exposure Duration - 30 yrs.
Adult Exposure Duration - 24 yrs.	Child Exposure Duration - 6 yrs.
Exposure Frequency - 350 days/yr	Adult Intake Rate - 100 mg/day
Child Intake Rate - 200 mg/day	Indoor Exposure Time Fraction - 0.683
Adult Inhalation Rate - 20 mg3/day	Child Inhalation Rate - 10 m3/day
Indoor Dilution Factor - 0.4 (unit less)	Area Correction Factor (unitless) - 0.9
Gamma Shielding Factor - 0.4 (m3/kg)	Contaminated Plant Fraction (unitless) - 0.25
Child Vegetable Consumption Rate (kg/yr) - 3.8	Q/C (g/m2-s per kg/m3) - 93.7
Child Fruit Consumption Rate (kg/yr) - 5.4	F(x) (unit less) - 0.194
Surface Area (acres) - 0.5	Fraction of Vegetative Cover - 0.5
Adult Vegetable Consumption Rate (kg/yr) - 10.4	Adult Fruit Consumption Rate (kg/yr) - 20.5
Particulate Emission Factor (m3/kg) - 1.36 E+09 (Albuquerque Climatic Zone)	Mean Annual Wind Speed (m/s) - 4.69
Equivalent Threshold Value of Wind speed at 7m (m/s) - 11.32	
Age-Adjusted Ingestion Factor (mg-yr/kg-day) - 120	
Age-Adjusted Inhalation Factor ((mg-yr/kg-day) - 18	
Age-Adjusted Inhalation Dilution Factor (unit less) - 0.4	
Age-Adjusted Vegetable Consumption Rate (kg/yr) - 9.08	
Age-Adjusted Fruit Consumption Rate (kg/yr) - 17.48	
Indoor Exposure Time Fraction - 0.683 (unit less)	
Outdoor Exposure Time Fraction - 0.073 (unit less)	

Q/C = Dispersion portion of the Particulate Emission Factor
F(x) = function dependent on mean annual wind- speed/equivalent threshold value of wind speed

The PRG calculator determines the concentration of each radionuclide that would contribute 1.0E-6 health risk (one excess cancer in one-million people exposed). The 1.06E-6 risk-concentrations were calculated for each radionuclide and are listed in Table 10. Dividing a site-specific radionuclide concentration by the 1.0E-6 risk-concentration (from Table 10) and then multiplying that result by 1.0E-06 determines the hypothetical risk for each radionuclide. For example, the 1.0E-6 risk concentration for ^{137}Cs, determined by the PRG calculator, is 0.0597 pCi/g. The ^{137}Cs concentration in surface sediments at Santa Clara is 0.102 pCi/g; the actual risk, 1.71 E-6, is then calculated by completing the equation:

$$(0.102 \text{ pCi/g} / 0.0597 \text{ pCi/g}) \text{ X } 1.0 \text{ E-6} = 1.71 \text{ E-6}.$$

Table 10. 1.0E-6 risk concentration numbers (pCi/g) calculated by PRG radionuclide calculator

Actinium-228 + decay chain = 732	Plutonium-238 + decay chain = 2.98
Americium-241 + decay chain = 1.87	Plutonium-239/240 + decay chain = 2.6
Bismuth-212 = 22,600	Sodium-22 + decay chain = 0.0865
Bismuth-214 = 8,190	Strontium-90 + decay chain = 0.231
Cesium-137 + decay chain = 0.0597	Thallium-208 = 22,600
Cobalt-60 + decay chain = 0.0361	Uranium-234 + decay chain = 4.02
Lead-212 = 3,640	Uranium-235 + decay chain = 0.195
Lead-214 = 46,300	Uranium-238 + decay chain = 0.743
Neptunium-237 + decay chain = 0.130	

We compared the hypothetic risks from three horizons at the Ancha site to the Santa Clara 5 to 31 cm surface horizon, to provide a context for the radionuclide health risks

calculated for this study. The risks calculated for the Ancha site originate from the: 1) 0 to 31 cm horizon, 2) 91 to 186 cm horizons, and 3) 186 to 335 cm horizons. The following narrative describes the hypothetical risks associated with each radionuclide measurement and the comparisons of the accumulative risks between the units described above. Appendix H shows the associated risk for all radionuclide measurements made for this study as well as the cumulative risk for each horizon.

The surface sample interval 5 to 31 cm at the Santa Clara site is from an abandoned floodplain. The floodplain had been active from 1941 to 1968, and contains only global fallout materials deposited during a period when fallout may have been the greatest. This site has been identified as a background reference site to which we compared the Ancha site risk evaluations.

Three intervals at the Ancha site were evaluated. They reflect post nuclear-age background conditions as well as Laboratory impacts. The shallowest sediments, from 0 to 91 cm, are recent deposits that originate from the upper Rio Grande and contain little to no Laboratory contaminants. The deeper units, from 91 to 186 cm, may have been deposited during a period of maximum discharge into the Rio Grande from Los Alamos Canyon and reflect episodic deposition of sediments containing the largest concentrations of legacy waste. The deepest units, from 186 to 335 cm, contain legacy contaminants but are diluted with greater amounts of coarse-grained sediments than other horizons.

Table 11 contains radionuclide concentrations and their associated risk for all four units, as well as the cumulative risk for each horizon. Seventeen radionuclides were assessed for these units except the lowest Ancha horizon. Isotopic uranium measurements were not made and the average risks calculated for the upper horizons were used. It should be noted that: 1) radionuclide concentrations reported as negative values and calculated risks less than 1E-8 were assessed as 0 risk in the cumulative risk, and 2) fallout, Laboratory legacy, and naturally occurring radionuclides contribute to each risk evaluation.

The first column identifies the radionuclides that were assessed, followed by the concentrations and risks for the Santa Clara and the Ancha sites. The last row lists the cumulative risk for each unit. Cumulative risk is the sum of risk contributed by each radionuclide.

Table 11. Hypothetic risks calculated for Santa Clara and Ancha sediment units

	Santa Clara Site (5.18 - 30.5 cm)		Cañada Ancha Site (0 - 30.5 cm)		Cañada Ancha Site (96-186 cm)		Cañada Ancha Site (186-335 cm)	
Radionuclides	Concentration (pCi/g)	RISK[1]	Concentration (pCi/g)	RISK	Average Concentration (pCi/g)	RISK	Average Concentration (pCi/g)	RISK
^{228}Ac	0.64	0	0.37	0	0.97	0	1.16	0
^{241}Am	0.003	0	0.0027	0	0.0166	0	0.0054	0
^{214}Bi	0.77	0	0.66	0	1.31	0	1.19	0
^{212}Bi	0.79	0	1.20	0	1.72	0	1.04	0
^{137}Cs	0.102	1.71E-06	0.093	1.56E-06	0.583	9.77E-06	0.056	9.44E-07
^{60}Co	0.05	1.39E-06	0.13	3.60E-06	0.19	5.21E-06	0.04	1.07E-06
^{22}Na	0.048	5.55E-07	0.14	1.62E-06	0.153	1.77E-06	0.044	5.09E-07
^{237}Np	0.001	0	-0.0029	0	0.0042	3.19E-08	0.0006	0
^{212}Pb	0.86	0	0.74	0	1.36	0	0.95	0
^{214}Pb	0.83	0	0.79	0	1.46	0	1.13	0
^{238}Pu	0.001	0	0.003	0	0.001	0	0.001	0
$^{239/240}$Pu	0.005	0	0.002	0	0.036	1.39E-08	0.017	0
^{90}Sr	-0.01	0	0.07	3.03E-07	0.40	1.72E-06	0.07	3.17E-07
^{208}Tl	0.264	0	0.073	0	0.372	0	0.417	0
^{234}U	0.90	2.24E-07	0.68	1.69E-07	1.36	3.38E-07	1.14	2.84E-07
^{235}U	0.039	2.00E-07	0.036	1.85E-07	0.079	4.03E-07	0.065	3.33E-07
^{238}U	0.68	9.15E-07	0.69	9.29E-07	1.40	1.88E-06	1.18	1.59E-06
Cumulative Risk		**4.99E-06**		**8.36E-06**		**2.11E-05**		**5.05E-06**

[1]Risk calculations equal to 0 or less than E-8 assigned risk = 0

Risk Assessment Discussion

The total risk at the Santa Clara site, used to reflect background, was 4.99E-06. The risk results at the Ancha site, located downstream of the Laboratory, are 8.36E-06, 2.11E-05, and 5.05E-06. The hypothetic risk for the 91 to 186 cm horizon, 2.11E-05, is above the 1.0E-05 risk level normally considered acceptable by NMED and four times greater than the background site. EPA uses a target risk range for clean-up of 10E-04 to 10E-06. Be aware that this exercise was done for comparison purposes and not to determine acceptance or rejection of contamination levels.

The surface horizon, 0 to 30 cm, at Ancha contains radionuclides from global fallout and reflects the same risk calculated at the background reference Santa Clara site. The deepest horizon at Ancha, 186 to 335 cm, contains legacy contaminants, but diluted to concentration levels that are similar to background conditions and reflects similar risks.

It should be noted here that the risk analysis performed on both locations included very conservative assumptions. The objective of this exercise was to evaluate the different cumulative effects of multiple radionuclides at separate location and horizons. The assessment was performed using identical and overly conservative input parameters at each site - the upstream Santa Clara site and the downstream Ancha site.

First, the calculation included radionuclides that are found naturally in the environment and the "fallout" radionuclides from atmospheric testing of nuclear weapons. Second, if

an activity concentration was reported less than a detection limit, then the reported value (the statistically most probable value) was used in the risk assessment. If a reported value was not available, then the detection limit was used. In a typical risk assessment, half of the detection limit value is used. Third, in most radionuclide risk assessments, only surface intervals from 0 to 15 cm would be used. At the Ancha site, the shallower samples (0 - 91.4 cm) reflected background conditions and had much lower concentrations of radionuclides than the deeper sampling intervals (91.4 - 186 cm). We used the surface (0 - 31 cm) interval from the shallow Ancha samples for comparisons, because clean surface sediments shield human receptors and limit the exposure duration. Fourth, all values for ^{60}Co and ^{22}Na were non-detects and detection limits were used in the risk calculations. Detection limits are sample specific and ^{60}Co and ^{22}Na measurements at the Ancha site yielded detection limits up to three times greater than measured at the Santa Clara site. The ^{60}Co and ^{22}Na cancer risk for the mid-interval Ancha site samples was calculated to be 6.98E-06, 33% of the total risk there.

An assessment was also made using only values reported above their sample specific detection level. Table 12 contains the summary results for this assessment. Appendix H also contains the risk values for each radionuclide measurement reported above its sample specific detection limit. When only values reported above detection limits were used, the calculated hypothetical risk at each site is reduced on average by 49%. The resulting risk values at Santa Clara reduce from 4.99E-06 to 3.05E-06, the surface Ancha value reduces from 8.36E-06 to 1.28E-06, at the mid-interval Ancha group, it reduces from 2.11E-05 to 1.41E-05 and at the deepest Ancha interval, and the risk reduces from 5.05E-06 to 2.72E-06. The hypothetic risk for the Ancha mid interval, 1.41E-05, is still above the 1.0E-05 risk level normally considered acceptable by NMED and about five times greater than the background site.

Uranium contributes the largest portion of hypothetical risk in samples that demonstrate conditions unaffected by fallout contaminants. For example, 100% of the calculated risk in the pre-1941 samples from Santa Clara is from uranium (see Table 2 Appendix H). At the Santa Clara surface interval, uranium and ^{137}Cs contributes 44% and 56% of the hypothetical risk, respectively. This sample demonstrates post-1941 global fallout conditions. While at the Ancha mid-interval, ^{137}Cs and ^{90}Sr are the largest contributors to the hypothetical risk and contribute approximately 79% of the total risk, 66% for ^{137}Cs and 13% for ^{90}Sr. Uranium contributes approximately 20% and $^{239/240}$plutonium contributes less than 1% of the hypothetical risk.

Table 12. Hypothetic risks calculated for Santa Clara and Ancha sediment units using only reported values above detection levels

Radionuclides	Santa Clara (5 - 31 cm)		Ancha (0 - 31 cm)		Ancha (91 - 186 cm)		Ancha (186 - 335 cm)	
	Concentration pCi/g	Risk	Concentration pCi/g	Risk	Average Concentration pCi/g	Risk	Average Concentration pCi/g	Risk
^{228}Ac	0.64	0	0.37	RV BDL	0.97	0	1.16	0
^{241}Am	0.003	RV BDL	0.0027	RV BDL	0.0166	0	0.005	0
^{214}Bi	0.77	0	0.66	0	1.31	0	1.19	0
^{212}Bi	0.79	0	1.2	0	1.72	0	1.04	0
^{137}Cs	0.102	1.71E-06	0.093	RV BDL	0.583	9.77E-06	0.056	5.08E-07
^{60}Co	0.05	RV BDL	0.13	RV BDL	0.19	RV BDL	0.04	RV BDL
^{22}Na	0.048	RV BDL	0.140	RV BDL	0.153	RV BDL	0.044	RV BDL
^{237}Np	0.00098	RV BDL	-0.0029	RV BDL	0.00415	RV BDL	0.00059	RV BDL
^{212}Pb	0.86	0	0.74	0	1.36	0	0.95	0
^{214}Pb	0.83	0	0.79	0	1.46	0	1.13	0
^{238}Pu	0.0006	RV BDL	0.0029	RV BDL	0.0011	0	0.0011	0
$^{239/240}$Pu	0.0051	0	0.0022	RV BDL	0.0363	0	0.017	0
^{90}Sr	-0.01	RV BDL	0.07	RV BDL	0.40	1.72E-06	0.07	RV BDL
^{208}Tl	0.264	0	0.073	RV BDL	0.372	0	0.417	0
^{234}U	0.90	2.24E-07	0.68	1.69E-07	1.36	3.38E-07	1.143*	2.84E-07
^{235}U	0.039	2.00E-07	0.036	1.85E-07	0.079	4.03E-07	0.065*	3.33E-07
^{238}U	0.68	9.15E-07	0.69	9.29E-07	1.40	1.88E-06	1.18*	1.59E-06
Cumulative Risk		3.05E-06		1.28E-06		1.41E-05		2.72E-06

RV BDL - Reported values below detection limit

*Mean Ancha Values

0 values less than risk<E-8

Summary

The New Mexico Environment Department DOE Oversight Bureau identified global fallout and Los Alamos National Laboratory legacy radionuclides along the Rio Grande. The highest proportions of LANL contaminants in sediments are nearest to the historical Laboratory discharge sources, although sediment sorting by fluvial processes contributed unexpected concentrations downstream.

We collected sediments from multiple horizons in cores and outcrops at five sites along the Rio Grande. Data were evaluated by: 1) statistical analysis of radiochemical results; 2) comparing the data to historical background values; 3) analyzing grain-size distribution and contaminant concentration relationships; and 4) using plutonium atom ratios to identify contaminant sources.

Most of the legacy contaminants in Rio Grande sediments are in the White Rock Canyon to Cochiti Lake reach, and were derived from the Los Alamos Canyon watershed. To determine background conditions, we selected a site at Santa Clara Pueblo, upstream of the Los Alamos Canyon and Rio Grande confluence. This confluence is near the Otowi Bridge on New Mexico State Road 4. The remaining four sites are downstream of the Otowi Bridge. The Ancha site is five km downstream, while the Pajarito site and Water Canyon site are 11 and 14 km below the bridge, respectively. The Frijoles site, farthest downstream, is 19 km below the bridge.

We found that $^{239/240}$Pu was the most persistent radionuclide found in terraces downstream of LANL. By far, the largest concentrations were found at the Ancha site followed by the Frijoles site, and then the Water Canyon site. Cesium-137 was also elevated by the greatest amount at Ancha site, also followed by Frijoles site. Anomalous uranium levels were also identified at the Ancha and Frijoles sites. Strontium-90 was found to be elevated at the Ancha site and ^{241}Am was elevated at the Frijoles site. Contaminant measurements at the Pajarito site were all indistinguishable from background, although we identified potential legacy contaminants at levels diluted below the existing background references.

An EPA risk evaluation exercise was prepared to relate the cumulative risks from all radionuclide measurements in terrace deposits samples for this study. It was made using a conservative exposure scenario based upon a long term residential occupancy scenario. It was noted that risk assessments are normally site specific, and the Santa Clara site is an agricultural area while the Ancha site is within an isolated area used for recreation. Both scenarios would include only surface soils for exposure pathways and reduced exposure duration parameters. The evaluation did not identify a difference in a health risk between the downstream Ancha and the upstream Santa Clara surface intervals. However the exercise did identify a hypothetical risk to be four to five times greater at deeper intervals from the Ancha site than at the surface sediments from the Santa Clara site. This risk exercise assumed that the mid-level Ancha sediments (96-186 cm) are located as surface sediments.

We included eight Rio Grande channel sediment samples in this evaluation. The samples were collected in 2003 from slack-water, low energy areas in the river, are predominantly fine-grained sediments, and reflected current transport. We found that the analytical measurements in the river channel sediments reflect global fallout constituents similar to that found in the upper Rio Grande watershed.

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APPENDIX A. Analytical Data

Table 1 - Plutonium analytical results from alpha spectroscopy and TIMS for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico.

Station Description	Date Collected	TIMS Results						Alpha Spec Results			
		^{239}Pu Activity (pCi/g)	1 STD	^{239}Pu + ^{240}Pu Activity (pCi/g)	1 STD	^{240}Pu/^{239}Pu Atom Ratio	1 STD	^{238}Pu (pCi/g)	unc/DL	$^{239/240}$Pu (pCi/g)	unc/DL
Santa Clara Sites											
DOEOB 3											
(abandoned flood plain, active 1941-1968)2B											
5.18 - 30.5 cm (hand augered)	4/24/01	0.00273	0.00010	0.00431	0.00015	0.157	0.013	0.0006	0.0015/0.0035	0.0051	0.0031/0.0015
(lab duplicate)								0.0002	0.0011/NA	0.0033	0.0019/NA
DOEOB 4											
(abandoned flood plain, active prior to 1941)3B											
33.5 - 39.6 cm (hand augered)	4/24/01	NA	-	NA	-	NA	-	-0.0016	0.0014/0.0037	0.0008	0.0011/0.0021
73.2 - 88.4 cm (hand augered)	4/24/01	NA	-	NA	-	NA	-	0.001	0.0014/0.0026	-0.00062	0.00091/0.0028
DOEOB 5											
(abandoned flood plain, active prior to 1941)3B											
24.4 - 39.6 cm (hand augered)	4/25/01	NA	-	NA	-	NA	-	0.0007	0.0013/0.0028	-0.00022	0.00099/0.0028
57.9 - 73.2 cm (hand augered)	4/25/01	NA	-	NA	-	NA	-	0.00000	0.00095/0.0024	0.0006	0.00085/0.00084
116 - 158 cm (hand augered)	4/25/01	NA	-	NA	-	NA	-	0.00000	0.0010/0.0025	0.00000	0.00100/0.00100

Table 1 - Plutonium analytical results from alpha spectroscopy and TIMS for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico. (continued)

Station Description	Date Collected	TIMS Results						Alpha Spec Results			
		^{239}Pu Activity (pCi/g)	1 STD	^{239}Pu + ^{240}Pu Activity (pCi/g)	1 STD	^{240}Pu/^{239}Pu Atom Ratio	1 STD	^{238}Pu (pCi/g)	unc/DL	$^{239/240}$Pu (pCi/g)	unc/DL
Cañada Ancha Site (slough area during 1940-1958 or 1967) unit 3B											
0 - 30.5 cm (hand augered)	2/16/99	0.00040	0.00002	0.00070	0.00004	0.208	0.028	0.0029	0.0027/0.0044	0.0022	0.0020/0.0032
30.5 - 61 cm (hand augered)	2/16/99	NA	-	NA	-	NA	-	-0.0013	0.0024/0.0059	0.0000	0.0016/0.0040
(field duplicate)											
61 - 91.4 cm (hand augered)	2/16/99	NA	-	NA	-	NA	-	0.0007	0.0015/0.0015	0.0017	0.0015/0.0015
(lab duplicate)								0.001	0.0023/NA	0.0028	0.0024/NA
91.4 - 101 cm (hand augered)	2/16/99	NA	-	NA	-	NA	-	-0.0007	0.0035/0.0074	0.0033	0.0031/0.0052
101 - 110 cm (hand augered)	2/16/99	0.01721	0.00033	0.02310	0.00051	0.093	0.006	0.0056	0.0030/0.0031	0.067	0.012/0.0038
110 - 125 cm (hand augered)	2/12/99	NA	-	NA	-	NA	-	0.0013	0.0030/0.0059	0.0121	0.0044/0.0040
125 - 137 cm (hand augered)	2/12/99	NA	-	NA	-	NA	-	0.0018	0.0016/0.0015	0.0262	0.0067/0.0015
137 - 162 cm (hand augered)	2/12/99	NA	-	NA	-	NA	-	0.0007	0.0021/0.0045	0.0430	0.0090/0.0036
162 - 186 cm (hand augered)	2/12/99	0.04511	0.00028	0.06076	0.00035	0.094	0.001	-0.0020	0.0032/0.0075	0.066	0.012/0.0052
(lab duplicate)								0.0084	0.0042/NA	0.068	0.013/NA
(field duplicate)		NA	-	NA	-	NA	-	0.0023	0.0029/0.0052	0.069	0.013/0.0042
186 - 213 cm (hand augered)	4/10/00	NA	-	NA	-	NA	-	0.00082	0.00093/0.00074	0.0144	0.0043/0.00074
213 - 229 cm (hand augered)	4/10/00	0.03971	0.00038	0.04199	0.00044	0.016	0.002	0.00122	0.00073/0.00069	0.0337	0.0054/0.00069
323 - 335 cm (hand augered)	4/10/00	NA	-	NA	-	NA	-	0.00131	0.00079/0.00087	0.0029	0.0011/0.00087

Table 1 - Plutonium analytical results from alpha spectroscopy and TIMS for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico. (continued)

Station Description	Date Collected	TIMS Results						Alpha Spec Results			
		^{239}Pu Activity (pCi/g)	1 STD	^{239}Pu + ^{240}Pu Activity (pCi/g)	1 STD	^{240}Pu/^{239}Pu Atom Ratio	1 STD	^{238}Pu (pCi/g)	unc/DL	$^{239/240}$Pu (pCi/g)	unc/DL
Pajarito Site (active flood plain and bar surface)1B											
0 - 30.5 cm (hand augered)	9/28/98	NA	-	NA	-	NA	-	-0.0025	0.0029/0.0097	0.0029	0.0037/0.0064
30.5 - 45.7 cm (hand augered)	9/28/98	0.00158	0.00002	0.00226	0.00004	0.118	0.007	0.0017	0.0034/0.0075	0.0076	0.0056/0.0067
Water Canyon Site (active flood plain and bar surface)1B											
0 - 30.5 cm (outcrop A)	9/29/98	0.00208	0.00002	0.00292	0.00005	0.110	0.005	-0.0013	0.0030/0.0095	0.0032	0.0043/0.0082
30.5 - 61 cm (outcrop A)	9/29/98	NA	-	NA	-	NA	-	-0.0032	0.0031/0.012	0.0061	0.0055/0.0084
61 - 91.4 cm (outcrop A)	9/29/98	NA	-	NA	-	NA	-	0.0006	0.0031/0.0057	0.0082	0.0057/0.0057
91.4 - 101 cm (outcrop A)	9/29/98	0.00256	0.00010	0.00398	0.00014	0.151	0.012	0.0025	0.0037/0.0071	0.0080	0.0053/0.0027
101 - 116 cm (outcrop A)	9/29/98	NA	-	NA	-	NA	-	0.0015	0.0031/0.0057	0.0021	0.0035/0.0069
91.4 - 101 cm (outcrop B)	9/29/98	NA	-	NA	-	NA	-	0.0015	0.0031/0.0056	0.0017	0.0035/0.0077

Table 1 - Plutonium analytical results from alpha spectroscopy and TIMS for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico. (continued)

Station Description	Date Collected	TIMS Results: ^{239}Pu Activity (pCi/g)	1 STD	$^{239}Pu + ^{240}Pu$ Activity (pCi/g)	1 STD	$^{240}Pu/^{239}Pu$ Atom Ratio	1 STD	Alpha Spec Results: ^{238}Pu (pCi/g)	unc/DL	$^{239/240}Pu$ (pCi/g)	unc/DL
Frijoles Site											
(pre-1950 flood-plain deposits and reservoir sedimentation)											
0 - 30.5 cm (hand augered)	**9/30/98**	NA	-	NA	-	NA	-	-0.0012	0.0056/0.015	0.0217	0.0094/0.0057
30.5 - 61 cm (hand augered)	**9/30/98**	0.01063	0.00013	0.01379	0.00018	0.081	0.003	0.0026	0.0038/0.0074	0.0179	0.0083/0.0054
(field duplicate)		NA	-	NA	-	NA	-	0.0011	0.0036/0.0072	0.0188	0.0061/0.0054
61 - 91.4 cm (hand augered)	**9/30/98**	NA	-	NA	-	NA	-	0.0005	0.0034/0.0088	0.0091	0.0057/0.0051
91.4 - 122 cm (hand augered)	**9/30/98**	NA	-	NA	-	NA	-	0.0018	0.0028/0.0026	0.0053	0.0042/0.0026
122 - 152 cm (hand augered)	**9/30/98**	0.00566	0.00011	0.00837	0.00020	0.131	0.008	0.0033	0.0037/0.0055	0.0162	0.0079/0.0055
(lab duplicate)								-0.0041	0.0043/NA	0.0094	0.0072/NA
152 - 183 cm (hand augered)	**9/30/98**	NA	-	NA	-	NA	-	0.0005	0.0026/0.0048	0.0082	0.0051/0.0024
(lab duplicate)								-0.0051	0.0043/NA	0.0104	0.0073/NA
Gallaher et al., : Global fallout						0.16 to 0.21					
Gallaher et al., : Global fallout or mixture of fallout and LANL possible						0.13 to 0.16					
Gallaher et al., : LANL or mixture of LANL and global fallout						0.07 to 0.13					
Gallaher et al., : Modern LANL PU Range						0.05 to 0.07					
Gallaher et al., : Pre-1960s LANL PU Range						0.01 to 0.03					
McLin et al., 2002, River BGUL				0.013				0.0087		0.013	
Purtymun et al., 1987 (mean):				0.005				0.000		0.005	
Purtymun et al., 1987 [upper limit background (mean + 2sigma)]:				0.023				0.006		0.023	
Mean LANL ER Specific Background (Ryti et al., 1998)				0.025				0.0021		0.025	
LANL Specific ER UTL (Ryti et al., 1998)				0.068				0.006		0.068	
Method Used	Acid leaching method (Efurd,2002)							PAI714R4(alpha spec)		PAI714R4(alpha spec)	

BGUL - Upper limit for background = 0.95 (two-tailed) quantile.
DL - Detection limit, miniumun detectable concentration and/or activity.
NA - not applicable or not analyzed.
unc - Total propagated uncertainty (2 sigma).
NP - Not provided.

Table 2 - Additional radionuclide results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico.

Station Description	Date Collected	234U (pCi/g)	unc/DL	235U (pCi/g)	unc/DL	238U (pCi/g)	unc/DL	Total U** (mg/kg)	90Sr (pCi/g)	unc/DL	241Am (pCi/g)	unc/DL	237Np (pCi/g)	unc/DL
Santa Clara Sites														
DOEOB 3														
(abandoned flood plain, active 1941-1968)2B														
5.18 - 30.5 cm (hand augered)	**4/24/2001**	0.90	0.16/0.025	0.039	0.022/0.024	0.68	0.13/0.029	2.04	-0.01	0.25/0.43	0.003	0.011/0.028	0.001	0.02346/0.03357
(lab duplicate)		0.92	0.16/NA	0.055	0.025/NA	0.71	0.13/NA	2.14	0.14	0.25/0.41	0.008	0.012/NA		
DOEOB 4														
(abandoned flood plain, active prior to 1941)3B														
33.5 - 39.6 cm (hand augered)	**4/24/2001**	0.90	0.15/0.023	0.044	0.023/0.026	0.91	0.16/0.026	2.73	0.12	0.24/0.40	0.003	0.011/0.027	0.0069	0.02140/0.04139
73.2 - 88.4 cm (hand augered)	**4/24/2001**	0.380	0.081/0.025	0.017	0.014/0.016	0.388	0.082/0.019	1.16	0.22	0.25/0.40	0.010	0.014/0.028	0.0058	0.01808/0.03547
(lab duplicate)													-0.0172	0.01744/0.04610
DOEOB 5														
(abandoned flood plain, active prior to 1941)3B														
24.4 - 39.6 cm (hand augered)	**4/25/2001**	1.14	0.19/0.028	0.060	0.028/0.024	0.98	0.17/0.022	2.94	0.07	0.23/0.38	0.008	0.012/0.025	-0.0005	0.01276/0.02074
57.9 - 73.2 cm (hand augered)	**4/25/2001**	0.86	0.15/0.019	0.059	0.027/0.023	0.86	0.15/0.023	2.59	0.07	0.21/0.36	0.006	0.012/0.022	0.0011	0.01498/0.03197
116 - 158 cm (hand augered)	**4/25/2001**	1.15	0.19/0.020	0.053	0.025/0.017	1.09	0.18/0.023	3.27	0.01	0.22/0.38	-0.003	0.013/0.038	0.005	0.01341/0.02175

Table 2 - Additional radionuclide results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico. (continued)

Station Description	Date Collected	234U (pCi/g)	unc/DL	235U (pCi/g)	unc/DL	238U (pCi/g)	unc/DL	Total U** (mg/kg)	90Sr (pCi/g)	unc/DL	241Am (pCi/g)	unc/DL	237Np (pCi/g)	unc/DL
Cañada Ancha Site														
(slough area during 1940-1958 or 1967) unit 3B														
0 - 30.5 cm (hand augered)	2/16/1999	0.68	0.11/0.013	0.036	0.016/0.0093	0.69	0.11/0.0093	2.07	0.07	0.09/0.15	0.0027	0.0078/0.018	-0.0029	0.01946/0.02697
(lab duplicate)									0.06	0.09/0.15	0.0081	0.0084/NA	-0.0019	0.02060/0.03556
30.5 - 61 cm (hand augered)	2/16/1999	0.69	0.11/0.012	0.044	0.019/0.015	0.75	0.11/0.012	2.25	0.29	0.10/0.14	-0.0049	0.066/0.023	-0.0029	0.02049/0.03727
61 - 91.4 cm (hand augered)	2/16/1999	0.76	0.11/0.015	0.033	0.016/0.016	0.80	0.12/0.014	2.40	0.36	0.11/0.14	0.014	0.011/0.017	0.00364	0.01735/0.00978
(lab duplicate)		0.78	0.12/NA	0.036	0.017/NA	0.78	0.12/NA	2.34						
91.4 - 101 cm (hand augered)	2/16/1999	1.13	0.16/0.0096	0.053	0.021/0.012	1.06	0.15/0.012	3.18	0.27	0.10/0.15	0.0142	0.0100/0.006:	0.00115	0.01911/0.02650
101 - 110 cm (hand augered)	2/16/1999	1.96	0.26/0.017	0.126	0.034/0.0097	1.99	0.26/0.014	5.98	0.36	0.11/0.14	0.026	0.014/0.015	0.00853	0.02013/0.03038
110 - 125 cm (hand augered)	2/12/1999	1.25	0.17/0.011	0.070	0.024/0.012	1.20	0.17/0.012	3.60	0.31	0.11/0.14	0.0041	0.0082/0.018	-0.0050	0.02017/0.03275
125 - 137 cm (hand augered)	2/12/1999	1.28	0.18/0.013	0.071	0.024/0.0099	1.26	0.18/0.013	3.78	0.40	0.12/0.14	0.016	0.010/0.0061	0.01426	0.02480/0.04391
137 - 162 cm (hand augered)	2/12/1999	1.41	0.19/0.021	0.097	0.029/0.015	1.55	0.21/0.014	4.66	0.64	0.15/0.14	0.019	0.012/0.016	0.00023	0.02286/0.03459
162 - 186 cm (hand augered)	2/12/1999	1.13	0.16/0.021	0.055	0.022/0.012	1.33	0.19/0.016	3.98	0.40	0.12/0.14	0.020	0.011/0.0057	0.00574	0.02168/0.03002
(field duplicate)		1.15	0.17/0.020	0.079	0.028/0.018	1.24	0.18/0.018	3.73	0.36	0.11/0.14	0.029	0.014/0.013	-0.0023	0.02292/0.02832
(lab duplicate of field duplicate)		1.14	0.16/NA	0.081	0.026/NA	1.14	0.16/NA	3.43			0.027	0.014/NA		
186 - 213 cm (hand augered)	4/10/2000	NA		NA		NA		NA	0.17	0.15/0.24	0.0008	0.0011/0.0011	0.00358	0.00659/0.01172
(lab duplicate)											0.0009	0.0013/NA		
213 - 229 cm (hand augered)	4/10/2000	NA		NA		NA		NA	-0.06	0.15/0.25	0.015	0.011/0.012	-0.0009	0.00430/0.00659
323 - 335 cm (hand augered)	4/10/2000	NA		NA		NA		NA	0.11	0.14/0.24	0.00030	.00059/0.001	-0.0009	0.00448/0.01004
(lab duplicate)													-0.0030	0.00752/0.01646

Table 2 - Additional radionuclide results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico. (continued)

Station Description	Date Collected	234U (pCi/g)	unc/DL	235U (pCi/g)	unc/DL	238U (pCi/g)	unc/DL	Total U** (mg/kg)	90Sr (pCi/g)	unc/DL	241Am (pCi/g)	unc/DL	237Np (pCi/g)	unc/DL
Pajarito Site														
(active flood plain and bar surface)1B														
0 - 30.5 cm (hand augered)	9/28/1998	0.444	0.091/0.031	0.020	0.019/0.028	0.432	0.089/0.0096	1.30	<0.17	NA/0.17	0.0055	).0049/0.005(	0.0024	0.0095/0.017
(lab duplicate)		0.441	0.087/NA	0.025	0.021/NA	0.479	0.092/NA	1.44	<0.15	NA/0.15				
30.5 - 45.7 cm (hand augered)	9/28/1998	0.73	0.13/0.025	0.063	0.030/0.027	0.72	0.13/0.025	2.17	<0.16	NA/0.16	0.0100	).0063/0.003(	0.010	0.012/0.016
(lab duplicate)		0.59	0.11/NA	0.064	0.028/NA	0.66	0.12/NA	1.99						
Water Canyon Site														
(active flood plain and bar surface)1B														
0 - 30.5 cm (outcrop A)	9/29/1998	0.63	0.12/0.030	0.042	0.027/0.034	0.65	0.12/0.024	1.95	<0.16	NA/0.16	0.0030	).0047/0.009(	0.001	0.001/0.023
30.5 - 61 cm (outcrop A)	9/29/1998	0.60	0.11/0.025	0.028	0.021/0.027	0.53	0.10/0.018	1.59	<0.16	NA/0.16	0.0062	).0050/0.003'	-0.003	0.011/0.026
61 - 91.4 cm (outcrop A)	9/29/1998	0.73	0.13/0.0096	0.033	0.021/0.023	0.76	0.13/0.023	2.28	<0.17	NA/0.17	0.0108	0.0083/0.013	0.009	0.012/0.018
91.4 - 101 cm (outcrop A)	9/29/1998	0.82	0.14/0.033	0.036	0.025/0.035	0.86	0.14/0.023	2.58	<0.15	NA/0.15	0.018	0.012/0.016	-0.001	0.010/0.020
101 - 116 cm (outcrop A)	9/29/1998	0.72	0.13/0.037	0.040	0.027/0.035	0.73	0.13/0.037	2.19	<0.16	NA/0.16	0.0002	0.0049/0.013	-0.002	0.012/0.026
(lab duplicate)													0.0025	0.0098/NA
91.4 - 101 cm (outcrop B)	9/29/1998	0.75	0.13/0.025	0.046	0.026/0.025	0.85	0.14/0.030	2.55	<0.15	NA/0.15	0.0130	).0081/0.008(	-0.003	0.012/0.027

Table 2 - Additional radionuclide results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon, New Mexico. (continued)

Station Description	Date Collected	234U (pCi/g)	unc/DL	235U (pCi/g)	unc/DL	238U (pCi/g)	unc/DL	Total U** (mg/kg)	90Sr (pCi/g)	unc/DL	241Am (pCi/g)	unc/DL	237Np (pCi/g)	unc/DL
Frijoles Site														
(pre-1950 flood-plain deposits and reservoir sedimentation)														
0 - 30.5 cm (hand augered)	**9/30/1998**	1.22	0.19/0.018	0.075	0.031/0.021	1.12	0.17/0.026	3.37	<0.15	NA/0.15	0.0184	0.0095/0.008	-0.001	0.014/0.021
30.5 - 61 cm (hand augered)	**9/30/1998**	1.31	0.20/0.030	0.091	0.037/0.027	1.28	0.20/0.036	3.85	<0.23	NA/0.23	0.022	0.010/0.0066	0.007	0.014/0.029
(field duplicate)		1.26	0.18/0.018	0.079	0.027/0.016	1.10	0.16/0.0063	3.31	0.12	0.09/0.14	0.007	0.010/0.020	-0.0060	0.02017/0.03477
(lab duplicate of field duplicate)													-0.0028	0.01885/0.02613
61 - 91.4 cm (hand augered)	**9/30/1998**	0.63	0.11/0.029	0.057	0.026/0.019	0.71	0.12/0.019	2.14	<0.22	NA/0.22	0.0062	0.0062/0.010	-0.001	0.011/0.021
91.4 - 122 cm (hand augered)	**9/30/1998**	0.81	0.13/0.042	0.027	0.021/0.032	0.68	0.12/0.030	2.04	<0.21	NA/0.21	0.0037	0.0047/0.008	0.0028	0.0085/0.0085
122 - 152 cm (hand augered)	**9/30/1998**	1.25	0.19/0.031	0.083	0.033/0.018	1.26	0.19/0.018	3.79	<0.20	NA/0.20	0.0086	0.0090/0.016	0.000	0.011/0.011
(lab duplicate)											0.0137	0.0084/NA		
152 - 183 cm (hand augered)	**9/30/1998**	1.12	0.17/0.021	0.040	0.022/0.087	1.07	0.17/0.021	3.20	<0.23	NA/0.23	0.0114	0.0071/0.007	-0.004	0.011/0.030
(lab duplicate)											0.0096	0.0076/NA	-0.0007	0.0092/NA
McLin et al., 2002, River BGUL		NP		NP		NP		4.49	1.02		0.076		NP	
Purtymun et al., 1987 (mean):		NP		NP		NP		2.6	0.23		NP		NP	
Purtymun et al., 1987 [upper limit background (mean + 2sigma)]:		NP		NP		NP		4.4	0.87		NP		NP	
Mean LANL ER Specific Background (Ryti et al., 1998)		1.40		0.087		1.22		3.76	0.229		0.026		NP	
LANL Specific ER UTL (Ryti et al., 1998)		2.59		0.20		2.29		6.99	1.04		0.04		NP	
Method Used		PAI714R4(alpha spec)		PAI714R4(alpha spec)		PAI714R4(alpha spec)			ASTMD5811-95(GPC)		Alpha Spec		Alpha Spec	

BGUL - Upper limit for background = 0.95 (two-tailed) quantile.
DL - Detection limit, minimum detectable concentration and/or activity.
NA - not applicable or not analyzed.
unc - Total propagated uncertainty (2 sigma).
NP - Not provided.
** - Total uranium concentrations calculated from the isotope-specific activity measurements.

Table 3 - Gamma spectroscopy analytical results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon

Station Description	Date Collected	241Am (pCi/g)	unc	137Cs (pCi/g)	unc	60Co (pCi/g)	unc	212Pb (pCi/g)	unc	214Pb (pCi/g)	unc	228Ac (pCi/g)	unc	208Tl (pCi/g)	unc	214Bi (pCi/g)	unc	212Bi (pCi/g)	unc	22Na (pCi/g)	unc	235U (pCi/g)	unc
Santa Clara Sites																							
DOEOB 3 (abandoned flood plain, active 1941-1968)2B																							
5.18 - 30.5 cm (hand augered)	**4/24/2001**	<0.057	BDL	0.102	0.031	<0.050	BDL	0.86	0.15	0.83	0.15	0.64	0.13	0.264	0.055	0.77	0.15	0.79	0.30	<0.048	BDL	<0.27	BDL
(lab duplicate)		<0.034	BDL	0.095	0.035	<0.049	BDL	0.92	0.15.17	0.89	0.16	0.83	0.17	0.251	0.056	0.77	0.15	0.87	0.34	<0.060	BDL	<0.19	BDL
DOEOB 4 (abandoned flood plain, active prior to 1941)3B																							
33.5 - 39.6 cm (hand augered)	**4/24/2001**	<0.059	BDL	<0.036	BDL	<0.038	BDL	1.01	0.18	1.01	0.18	0.86	0.16	0.315	0.062	0.91	0.17	1.10	0.35	<0.043	BDL	<0.15	BDL
73.2 - 88.4 cm (hand augered)	**4/24/2001**	<0.22	BDL	<0.027	BDL	<0.037	BDL	0.467	0.088	0.430	0.083	0.396	0.090	0.148	0.035	0.407	0.085	0.44	0.24	<0.042	BDL	<0.13	BDL
DOEOB 5 (abandoned flood plain, active prior to 1941)3B																							
24.4 - 39.6 cm (hand augered)	**4/25/2001**	<0.058	BDL	<0.036	BDL	<0.043	BDL	0.81	0.14	0.78	0.14	0.72	0.14	0.288	0.058	0.76	0.15	0.76	0.32	<0.050	BDL	<0.29	BDL
57.9 - 73.2 cm (hand augered)	**4/25/2001**	<0.061	BDL	<0.033	BDL	<0.036	BDL	0.92	0.16	0.92	0.16	0.81	0.16	0.256	0.052	0.78	0.23	0.97	0.34	<0.039	BDL	<0.14	BDL
116 - 158 cm (hand augered)	**4/25/2001**	<0.31	BDL	<0.041	BDL	<0.043	BDL	1.10	0.19	1.08	0.19	0.83	0.16	0.350	0.069	0.88	0.25	1.02	0.34	<0.050	BDL	<0.14	BDL

Table 3 - Gamma spectroscopy analytical results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon. (continued)

Station Description	Date Collected	241Am (pCi/g)	unc	137Cs (pCi/g)	unc	60Co (pCi/g)	unc	212Pb (pCi/g)	unc	214Pb (pCi/g)	unc	228Ac (pCi/g)	unc	208Tl (pCi/g)	unc	214Bi (pCi/g)	unc	212Bi (pCi/g)	unc	22Na (pCi/g)	unc	235U (pCi/g)	unc
Cañada Ancha Site (slough area during 1940-1958 or 1967) unit 3B																							
0 - 30.5 cm (hand augered)	**2/16/1999**	<0.38	BDL	<0.093	BDL	<0.13	BDL	0.74	0.20	0.79	0.22	<0.37	BDL	<0.073	BDL	0.66	0.23	1.2	1.1	<0.14	BDL	<0.30	BDL
30.5 - 61 cm (hand augered)	**2/16/1999**	<0.52	BDL	<0.13	BDL	<0.11	BDL	0.84	0.24	0.86	0.24	0.79	0.33	<0.12	BDL	0.95	0.28	0.84	0.24	<0.13	BDL	<0.43	BDL
(lab duplicate)		<0.32	BDL	<0.078	BDL	<0.067	BDL	0.78	0.19	0.99	0.22	0.65	0.23	0.293	0.091	0.94	0.22	<0.80	BDL	<0.066	BDL	<0.39	BDL
61 - 91.4 cm (hand augered)	**2/16/1999**	<0.39	BDL	<0.070	BDL	<0.092	BDL	0.77	0.20	0.98	0.24	0.50	0.22	<0.079	BDL	0.98	0.24	0.85	0.86	<0.077	BDL	<0.40	BDL
91.4 - 101 cm (hand augered)	**2/16/1999**	<0.57	BDL	0.24	0.12	<0.14	BDL	0.81	0.24	0.91	0.26	<0.51	BDL	<0.15	BDL	0.92	0.32	<1.5	BDL	<0.17	BDL	<0.44	BDL
101 - 110 cm (hand augered)	**2/12/1999**	<1.5	DDL	0.60	0.30	<0.41	DDL	1.85	0.62	2.56	0.76	<1.3	DDL	0.41	0.30	1.85	0.04	<4.1	DDL	<0.29	DDL	<1.2	DDL
110 - 125 cm (hand augered)	**2/12/1999**	<0.45	BDL	0.27	0.13	<0.18	BDL	1.33	0.32	1.21	0.34	0.95	0.38	0.46	0.15	1.21	0.34	<0.98	BDL	0.15	BDL	<0.39	BDL
125 - 137 cm (hand augered)	**2/12/1999**	<0.40	BDL	0.71	0.16	<0.088	BDL	1.10	0.24	1.41	0.29	1.05	0.28	0.45	0.12	1.18	0.28	1.13	0.86	<0.072	BDL	<0.41	BDL
137 - 162 cm (hand augered)	**2/12/1999**	<0.71	BDL	0.98	0.26	<0.16	BDL	1.65	0.38	1.44	0.36	1.09	0.40	<0.16	BDL	1.36	0.39	1.1	1.2	<0.16	BDL	<0.56	BDL
162 - 186 cm (hand augered)	**2/12/1999**	<0.50	BDL	0.70	0.20	<0.15	BDL	1.42	0.33	1.24	0.33	0.93	0.39	0.60	0.18	1.32	0.36	1.5	1.5	<0.076	BDL	<0.41	BDL
(field duplicate)		<0.95	BDL	0.78	0.30	<0.25	BDL	1.19	0.48	1.63	0.52	<0.84	BDL	<0.24	BDL	1.07	0.61	<2.8	BDL	<0.20	BDL	<1.0	BDL
(lab duplicate of field duplicate)		<1.3	BDL	1.06	0.41	<0.54	BDL	1.12	0.54	1.81	0.63	<1.3	BDL	<0.26	BDL	2.11	0.71	4.2	3.3	<0.44	BDL	<1.2	BDL
186 - 213 cm (hand augered)	**4/10/2000**	0.59*	0.12	0.091	0.028	<0.041	BDL	0.74	0.14	1.33	0.27	1.30	0.25	0.48	0.11	1.45	0.33	1.03	0.60	<0.045	BDL	0.29*	0.14
(lab duplicate)		<0.78	BDL	0.104	0.089	<0.050	BDL	1.38	0.24	1.35	0.26	1.36	0.34	0.449	0.085	1.27	0.29	0.98	0.35	<0.060	BDL	<0.32	BDL
213 - 229 cm (hand augered)	**4/10/2000**	<0.74	BDL	<0.050	BDL	<0.049	BDL	1.38	0.23	1.42	0.26	1.65	0.35	0.57	0.12	1.47	0.31	1.46	0.64	<0.056	BDL	<0.30	BDL
323 - 335 cm (hand augered)	**4/10/2000**	<0.12	BDL	<0.028	BDL	<0.026	BDL	0.74	0.15	0.65	0.13	0.54	0.13	0.202	0.049	0.64	0.20	0.63	0.27	<0.031	BDL	<0.16	BDL

Table 3 - Gamma spectroscopy analytical results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon. (continued)

Station Description	Date Collected	241Am (pCi/g)	unc	137Cs (pCi/g)	unc	60Co (pCi/g)	unc	212Pb (pCi/g)	unc	214Pb (pCi/g)	unc	228Ac (pCi/g)	unc	208Tl (pCi/g)	unc	214Bi (pCi/g)	unc	212Bi (pCi/g)	unc	22Na (pCi/g)	unc	235U (pCi/g)	unc
Pajarito Site (active flood plain and bar surface)1B																							
0 - 30.5 cm (hand augered)	9/28/1998	<0.27	BDL	<0.059	BDL	<0.075	BDL	0.42	0.13	0.44	0.12	0.43	0.16	<0.060	BDL	0.44	0.15	<0.59	BDL	<0.058	BDL	<0.28	BDL
30.5 - 45.7 cm (hand augered)	9/28/1998	<0.43	BDL	0.144	0.087	<0.12	BDL	0.99	0.23	1.02	0.24	0.83	0.29	<0.10	BDL	0.85	0.27	1.1	1.0	<0.12	BDL	<0.40	BDL
Water Canyon Site (active flood plain and bar surface)1B																							
0 - 30.5 cm (outcrop A)	9/29/1998	<0.17	BDL	0.052	0.055	<0.082	BDL	0.67	0.16	0.86	0.19	0.75	0.21	0.231	0.077	0.58	0.18	0.82	0.64	<0.081	BDL	<0.25	BDL
30.5 - 61 cm (outcrop A)	9/29/1998	<0.30	BDL	<0.064	BDL	<0.059	BDL	0.57	0.14	0.49	0.14	<0.24	BDL	0.222	0.074	0.67	0.18	<0.73	BDL	<0.066	BDL	<0.31	BDL
(lab duplicate)		<0.32	BDL	<0.074	BDL	<0.11	BDL	0.7	0.18	0.60	0.18	0.59	0.24	0.205	0.100	0.80	0.22	<0.62	BDL	<0.093	BDL	<0.25	BDL
61 - 91.4 cm (outcrop A)	9/29/1998	<0.42	BDL	<0.12	BDL	<0.097	BDL	0.76	0.20	0.81	0.22	<0.54	BDL	<0.10	BDL	0.78	0.23	0.9	1.1	<0.12	BDL	<0.38	BDL
91.4 - 101 cm (outcrop A)	9/29/1998	<0.37	BDL	0.132	0.093	<0.078	BDL	0.88	0.22	1.00	0.24	0.99	0.31	0.33	0.12	0.95	0.25	1.3	1.0	<0.11	BDL	<0.30	BDL
101 - 116 cm (outcrop A)	9/29/1998	<0.17	BDL	<0.064	BDL	<0.072	BDL	0.94	0.20	1.07	0.22	0.99	0.24	0.385	0.097	0.84	0.21	1.13	0.71	<0.083	BDL	<0.28	BDL
91.4 - 101 cm (outcrop B)	9/29/1998	<0.64	BDL	<0.096	BDL	<0.13	BDL	1.02	0.25	1.18	0.28	1.08	0.35	<0.091	BDL	1.06	0.29	1.4	1.0	<0.12	BDL	<0.32	BDL

Table 3 - Gamma spectroscopy analytical results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon. (continued)

Station Description	Date Collected	241Am (pCi/g)	unc	137Cs (pCi/g)	unc	60Co (pCi/g)	unc	212Pb (pCi/g)	unc	214Pb (pCi/g)	unc	228Ac (pCi/g)	unc	208Tl (pCi/g)	unc	214Bi (pCi/g)	unc	212Bi (pCi/g)	unc	22Na (pCi/g)	unc	235U (pCi/g)	unc
Frijoles Site																							
(pre-1950 flood-plain deposits and reservoir sedimentation)																							
0 - 30.5 cm (hand augered)	**9/30/1998**	<0.23	BDL	0.128	0.094	<0.10	BDL	1.29	0.28	1.16	0.27	1.22	0.33	0.44	0.12	1.11	0.28	<0.84	BDL	<0.13	BDL	<0.38	BDL
30.5 - 61 cm (hand augered)	**9/30/1998**	<0.36	BDL	0.301	0.097	<0.085	BDL	1.27	0.27	0.99	0.23	1.12	0.30	0.38	0.11	1.01	0.25	1.44	0.83	<0.086	BDL	<0.42	BDL
(field duplicate)		<0.74	BDL	<0.23	BDL	<0.25	BDL	1.28	0.44	1.45	0.45	<0.71	BDL	0.55	0.25	0.78	0.50	<2.6	BDL	<0.18	BDL	<0.56	BDL
61 - 91.4 cm (hand augered)	**9/30/1998**	<0.18	BDL	0.098	0.065	<0.063	BDL	0.98	0.21	0.89	0.20	0.85	0.23	0.259	0.082	0.77	0.20	1.09	0.82	<0.10	BDL	<0.26	BDL
91.4 - 122 cm (hand augered)	**9/30/1998**	<0.35	BDL	0.101	0.089	<0.091	BDL	0.86	0.21	0.90	0.22	<0.33	BDL	<0.078	BDL	0.64	0.22	<0.93	BDL	<0.13	BDL	<0.29	BDL
122 - 152 cm (hand augered)	**9/30/1998**	<0.20	BDL	0.172	0.086	<0.076	BDL	1.23	0.25	1.15	0.25	0.87	0.27	0.31	0.10	0.95	0.25	1.18	0.84	<0.072	BDL	<0.31	BDL
(lab duplicate)		<0.21	BDL	0.175	0.084	<0.087	BDL	1.25	0.26	1.40	0.29	0.96	0.28	0.37	0.12	1.31	0.29	<1.0	BDL	<0.11	BDL	<0.33	BDL
152 - 183 cm (hand augered)	**9/30/1998**	<0.52	BDL	0.22	0.11	<0.16	BDL	1.09	0.28	1.00	0.26	0.99	0.33	0.41	0.14	1.11	0.31	1.4	1.2	<0.14	BDL	<0.48	BDL
McLin et al., 2002, River BGUL		0.076		0.56		NP		NP		NP		NP		NP		NP		NP		NP		NP	
Purtymun et al., 1987 (mean):		NP		0.18		NP		NP		NP		NP		NP		NP		NP		NP		NP	
Purtymun et al., 1987 [upper limit background (mean + 2sigm		NP		0.44		NP		NP		NP		NP		NP		NP		NP		NP		NP	
Mean LANL ER Specific Background (Ryti et al., 1998)		0.026		0.211		NP		NP		NP		NP		NP		NP		NP		NP		0.087	
LANL Specific ER UTL (Ryti et al., 1998)		0.040		0.90		NP		NP		NP		NP		NP		NP		NP		NP		0.20	
Method Used				**901.1 Modified**																			

BGUL - Upper limit for background = 0.95 (two-tailed) quantile.
DL - Detection limit, miniumun detectable concentration and/or activity.
NA - not applicable or not analyzed.
unc - Total propagated uncertainty (2 sigma).
NP - Not provided.
* - Nuclide identification tentative

Table 4. Particle size distribution analytical results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon.

Station Description	Date Collected	Soil Moisture (g/g)	% wt. Gravel	Sand Fractions 2.0-1.0 mm %wt.	1.0-0.5 mm %wt.	0.5-0.25 mm %wt.	0.25-0.125 mm %wt.	0.125-0.0625 mm %wt.	Silt Fractions Fine Silt %wt.	Co. Silt %wt.	Total Sand %wt.	Total Silt %wt.	Clay %wt.	%Error: Sand	%Error: All
Santa Clara Sites															
DOEOB 3 (abandoned flood plain, active 1941-1968)2B															
5.18 - 30.5 cm (hand augered)	**4/24/2001**	0.17	0.0	0.2	1.1	2.6	15.2	36.9	5.7	33.7	56.3	39.4	4.3	0.21	0.21
DOEOB 4 (abandoned flood plain, active prior to 1941)3B															
33.5 - 39.6 cm (hand augered)	**4/24/2001**	0.06	0.0	0.1	0.2	1.2	4.2	16.5	11.7	58.3	22.3	70.0	7.7	0.30	0.30
73.2 - 88.4 cm (hand augered)	**4/24/2001**	0.01	0.0	0.3	5.9	19.4	59.7	10.3	-0.9	2.2	95.7	1.4	2.9	0.10	0.10
(lab duplicate)		*NA*	*NA*	*0.2*	*5.5*	*19.7*	*55.9*	*14.5*	*0.3*	*1.4*	*95.9*	*1.7*	*2.4*	*0.10*	*0.10*
DOEOB 5 (abandoned flood plain, active prior to 1941)3B															
24.4 - 39.6 cm (hand augered)	**4/25/2001**	0.11	0.0	0.0	0.1	0.7	8.2	35.1	6.3	39.5	44.2	45.8	10.0	0.13	0.13
57.9 - 73.2 cm (hand augered)	**4/25/2001**	0.20	0.0	0.0	0.3	1.8	4.0	14.1	13.6	57.3	20.2	71.0	8.9	0.06	0.06
116 - 158 cm (hand augered)	**4/25/2001**	0.28	0.0	0.7	1.9	3.0	5.4	13.2	10.2	55.5	24.4	65.7	9.8	0.25	0.25
Cañada Ancha Site (slough area during 1940-1958 or 1967) unit 3B															
0 - 30.5 cm (hand augered)	**2/16/1999**	0.02	0.0	3.2	3.2	8.7	32.3	26.8	5.1	10.3	74.2	15.4	10.4	0.00	0.00
30.5 - 61 cm (hand augered)	**2/16/1999**	0.04	6.2	1.7	3.8	7.8	30.0	24.7	5.2	14.0	68.2	19.2	12.5	0.29	0.29
61 - 91.4 cm (hand augered)	**2/16/1999**	0.05	4.2	0.8	2.5	9.8	22.7	30.0	5.0	17.9	65.9	22.8	11.3	0.17	0.17
91.4 - 101 cm (hand augered)	**2/16/1999**	0.03	2.9	0.5	1.9	7.3	24.4	30.7	7.9	13.2	65.0	21.0	14.0	0.20	0.20
101 - 110 cm (hand augered)	**2/16/1999**	0.14	36.4	0.4	0.4	1.1	3.7	13.9	24.0	28.5	20.2	52.5	27.3	0.60	0.60
110 - 125 cm (hand augered)	**2/12/1999**	0.04	3.5	0.3	0.8	2.2	12.7	28.4	11.6	23.0	44.5	34.6	20.9	0.05	0.05
125 - 137 cm (hand augered)	**2/12/1999**	0.07	15.9	0.8	0.8	2.1	8.2	27.5	14.1	21.5	39.8	35.6	24.6	0.39	0.39
(laboratory duplicate)		*NA*	*NA*	*0.7*	*0.9*	*2.0*	*8.8*	*27.9*	*15.3*	*20.1*	*40.6*	*35.4*	*24.1*	*0.31*	*0.31*
137 - 162 cm (hand augered)	**2/12/1999**	0.19	36.1	0.5	1.1	1.3	3.3	9.9	18.8	24.8	16.0	43.7	40.4	-0.06	-0.06
162 - 186 cm (hand augered)	**2/12/1999**	0.29	50.4	0.1	0.3	0.6	2.0	5.5	27.0	15.8	8.7	42.8	48.5	0.13	0.13
(field duplicate)		0.06	32.4	0.4	0.8	1.5	3.1	6.7	28.7	19.7	12.7	48.4	38.9	0.17	0.17
186 - 213 cm (hand augered)	**4/10/2000**	0.35	10.6	0.8	0.9	1.6	3.9	10.5	24.7	20.4	17.7	45.2	37.1	0.08	0.08
213 - 229 cm (hand augered)	**4/10/2000**	0.71	67.6	0.0	0.2	0.3	0.5	1.2	25.8	21.1	2.2	47.0	50.9	0.00	0.00
323 - 335 cm (hand augered)	**4/10/2000**	0.06	29.4	13.3	12.5	31.9	30.8	6.7	0.7	2.5	95.3	3.1	1.5	0.18	0.18

Table 4. Particle size distribution analytical results for sediment-core/outcrop samples collected along the Rio Grande, White Rock Canyon. (continued)

Station Description	Date Collected	Soil Moisture (g/g)	% wt. Gravel	Sand Fractions: 2.0-1.0 mm %wt.	Sand Fractions: 1.0-0.5 mm %wt.	Sand Fractions: 0.5-0.25 mm %wt.	Sand Fractions: 0.25-0.125 mm %wt.	Sand Fractions: 0.125-0.0625 mm %wt.	Silt Fractions: Fine Silt %wt.	Silt Fractions: Co. Silt %wt.	Total Sand %wt.	Total Silt %wt.	Clay %wt.	%Error: Sand	%Error: All
Pajarito Site															
(active flood plain and bar surface)1B															
0 - 30.5 cm (hand augered)	9/28/1998	0.00	0.0	1.2	7.2	38.0	37.3	12.8	-0.9	2.7	96.5	1.8	1.7	0.20	0.20
30.5 - 45.7 cm (hand augered)	9/28/1998	0.00	0.9	0.2	0.9	10.3	44.1	31.8	-0.7	9.9	87.4	9.2	3.5	0.14	0.14
(laboratory duplicate)		*NA*	*NA*	*0.4*	*1.0*	*10.2*	*40.0*	*34.8*	*0.0*	*11.7*	*86.5*	*11.7*	*1.7*	*0.11*	*0.11*
Water Canyon Site															
(active flood plain and bar surface)1B															
0 - 30.5 cm (outcrop A)	9/29/1998	0.01	2.6	0.7	1.0	10.1	27.2	37.3	2.4	17.1	76.2	19.5	4.3	-0.05	-0.05
30.5 - 61 cm (outcrop A)	9/29/1998	0.00	2.7	0.3	1.0	19.1	55.5	15.3	0.7	5.1	91.3	5.8	2.9	0.20	0.20
61 - 91.4 cm (outcrop A)	9/29/1998	0.01	0.0	0.0	0.2	3.1	34.0	37.7	2.8	17.8	75.1	20.6	4.3	0.15	0.15
91.4 - 101 cm (outcrop A)	9/29/1998	0.01	0.0	0.0	0.4	1.8	20.4	46.4	3.0	24.0	69.1	27.0	3.9	0.14	0.14
101 - 116 cm (outcrop A)	9/29/1998	0.00	8.3	1.0	1.3	10.4	41.9	32.2	1.4	8.7	86.8	10.1	3.1	0.00	0.00
91.4 - 101 cm (outcrop B)	9/29/1998	0.01	2.0	1.2	1.7	3.0	6.7	25.2	7.6	48.0	38.4	55.7	5.9	0.62	0.62
Frijoles Site															
(pre-1950 flood-plain deposits and reservoir sedimentation)															
0 - 30.5 cm (hand augered)	9/30/1998	0.02	0.0	0.0	0.4	0.7	2.2	25.5	13.7	47.1	29.2	60.8	10.0	0.32	0.32
30.5 - 61 cm (hand augered)	9/30/1998	0.05	3.4	0.0	0.3	1.4	3.5	30.2	17.1	31.5	35.7	48.6	15.7	0.35	0.35
(field duplicate)		0.03	9.5	0.0	0.3	1.7	4.2	26.8	18.8	32.7	33.1	51.5	15.3	0.20	0.20
61 - 91.4 cm (hand augered)	9/30/1998	0.02	4.9	0.0	0.3	9.9	42.1	18.6	4.7	17.0	71.0	21.7	7.3	0.10	0.10
(field duplicate)															
91.4 - 122 cm (hand augered)	9/30/1998	0.01	0.3	0.0	0.3	16.4	35.5	27.8	1.9	13.9	80.1	15.8	4.1	0.09	0.09
122 - 152 cm (hand augered)	9/30/1998	0.02	0.1	0.1	0.3	1.9	4.4	24.6	13.2	45.2	31.4	58.4	10.2	0.16	0.16
152 - 183 cm (hand augered)	9/30/1998	0.02	0.1	0.0	0.1	0.6	2.4	34.8	10.3	40.6	38.3	50.9	10.8	0.37	0.37
(laboratory duplicate)		*NA*	*NA*	*0.0*	*0.1*	*0.5*	*1.9*	*34.1*	*10.4*	*42.9*	*36.8*	*53.3*	*10.0*	*0.20*	*0.20*

Data provided by Desert Research Institute
Methods used: pipette method for particle size analysis specified by the U.S. Geological Survey and the Soil Science of America (Day, 1965;Jackson, 1969; Janitzky, 1986; and Gee and Bauder, 1992)

Table 5 - Radionuclide results (2003) for slack water channel sediment samples collected in the Rio Grande and Rio Chama

Station	Date	Pu^{238}				$Pu^{239/240}$				Cs^{137}		
		(pCi/g)	Unc	MDC	Qual	(pCi/g)	Unc	MDC	Qual	(pCi/g)	Unc	MDC
Rio Grande at Pilar	3/11/2003	0.0006	0.0029	0.0018	U,Y2	0.0067	0.0044	0.0047	Y2	0.163	0.097	0.14
Chama River at Chamita	3/25/2003	-0.0004	0.0021	0.0041	U	0.0043	0.0027	0.0013	LT	0.118	0.08	0.11
Rio Grande at Rio Pojoaque	4/4/2003	-0.0008	0.002	0.004	U	0.0033	0.0026	0.0032	LT	0.027	0.096	0.17
Rio Grande below Los Alamos	4/4/2003	0.0000	0.0017	0.0027	U	0.009	0.0037	0.001		0.138	0.061	0.079
Rio Grande above Ancho	4/1/2003	-0.0011	0.0017	0.0039	U	0.0046	0.0026	0.0011	LT	0.073	0.05	0.08
Rio Grande at Pena Blanca	3/24/2003	0.0003	0.0016	0.001	U	0.0033	0.0023	0.0025	LT	0	0.13	0.23
San Angostura Diversion	3/18/2003	-0.0004	0.0019	0.0044	U	0.01	0.0045	0.003		0.2	0.26	0.42
Rio Grande below Albuquerque	6/10/2003	-0.0003	0.0017	0.004	U	0.0013	0.0016	0.00098	LT	0.043	0.031	0.42
McLin, et al, 2002, River BGUL		0.0087				0.013				0.56		
Purtymun, et al, 1987 (mean)		0.0000				0.005				0.18		
Purtymun, et al, 1987 [upper limit background (mean + 2 sigma)]		0.0060				0.023				0.44		

U - result is less than the sample specific MDC

Y2 - Chemical yield outside default limits

LT - Result is less than requested MDC, greater than sample specific MDC

Unc - Tottal propagated uncertainty (2 sigma)

MDC - Minimum detectable concentration

BGUL - Upper limit for background = 0.95 (two-tailed) quantile

Table 6 - Particle size distribution analytical results for slack water channel samples collected in the Rio Grande and Rio Chama (2003)

Station	Date	Sand Fractions						Silt Fractions						
		Gravel % wt.	2.0-1.0 mm %wt.	1.0-0.5 mm %wt.	0.5-0.25 mm %wt.	0.25-0.125 mm %wt.	0.125-0.0625 mm %wt.	Co. Silt %wt.	Fine Silt %wt.	Total Sand %wt.	Total Silt %wt.	Clay %wt.	Total Silt+Clay	Normalized Pu/Silt+Clay
Rio Grande at Pilar	3/11/2003	8.68	1.6	2.6	1.8	8.5	9.5	40	19.5	24.1	59.5	16.4	75.9	0.00883
Chama River at Chamita	3/25/2003	0.03	0	0.05	0.39	0.96	10.03	50.19	15.58	11.54	65.77	22.69	88.46	0.00486
Rio Grande at Rio Pojoaque	4/4/2003	0.25	0	0.03	0.29	3.2	31.66	38.72	10.3	35.25	49.02	15.73	64.75	0.00510
Rio Grande below Los Alamos	4/4/2003	0.18	0.06	0.13	0.19	0.84	17.96	55.17	12.17	19.09	67.34	13.57	80.91	0.01112
Rio Grande above Ancho	4/1/2003	0.12	0	0.14	3.02	25.36	36.28	24.79	4.5	64.89	29.29	5.82	35.11	0.01310
Rio Grande at Pena Blanca	3/24/2003	25.06	11.54	28.75	19.16	4.8	3.31	5.92	8.55	67.48	14.47	18.06	32.53	0.01014
San Angostura Diversion	3/18/2003	0	0.1	0.25	0.94	1.63	2.76	38.41	27.25	5.72	65.66	28.62	94.28	0.01061
Rio Grande below Albuquerque	6/10/2003	0.03	0	1.61	11.17	24.3	29.89	23.2	3.85	67.03	27.04	5.93	32.97	0.00394

APPENDIX B. Statistics Description

Introduction

We evaluated the data in this report using several methods; 1) by comparing individual measurements to background reference levels, 2) investigating grain size correlations to plutonium, 3) identifying plutonium sources using isotopic relationships, 4) calculating and comparing hypothetical risks, 5) and statistically comparing data cases representing each site. This section describes how the Oversight Bureau statistically evaluated the radionuclide measurements from core and outcrop samples collected from terrace deposits representing abandoned channels and floodplains along the Rio Grande. We used statistical methods to determine what relationships exist between terraces downstream of LANL and an upstream reference site. The evaluation also included comparisons to LANL background data for river sediments in northern New Mexico.

We selected five locations, a reference site that is unlikely to demonstrate an environmental impact and four sites that may have been impacted by Los Alamos National Laboratory contaminants. The reference site is 12.1 km (7.5 miles) upstream of Los Alamos Canyon, the most upstream canyon that emanates from Laboratory property. The terrace is a Rio Grande channel that was abandoned and filled prior to 1941. It also contains one sample from an abandoned flood plain, active between 1941 and 1968. The remaining four sites are downstream of Los Alamos Canyon and are active floodplains, or have been active since the creation of the Laboratory.

Data compiled and evaluated by McLin and others (2002) from the Laboratory were also used to demonstrate background conditions. LANL background measurements were compiled into cases that reflect the deposition and dispersion of global fallout materials from nuclear atmospheric testing in river sediments.

Commercial analytical laboratories measured the sediments for ^{238}Pu, $^{239/240}$Pu, ^{90}Sr, uranium isotopes -234, -235, and -238, ^{241}Am, ^{237}Np, and ^{137}Cs. We compiled multiple measurements of each constituent into cases, or data pools, reflecting sediments from cores drilled in terraces along the Rio Grande. These data populations reflect contaminant conditions in flood plains and channels abandoned and filled during episodic flood events before and after the 1940's.

Plutonium-239/240, ^{137}Cs, ^{90}Sr, and ^{241}Am appear to be statistically elevated in sediments downstream of Los Alamos Canyon when compared against the reference sites. By far, the largest concentrations were found at the Cañada Ancha core site, 5.1 km (3.2 miles) downstream of Los Alamos Canyon and Rio Grande confluence.

Methods

We compiled analytical measurements from individual sites into statistical samples or cases (pools of data). Multiple horizons were selected from cores drilled up to 3.4 m (11 feet) deep for radiochemical analysis. These cases reflect the contaminant

characterization that might be expected at those sites. The data distributions were determined and then the site data cases were compared.

We used the Shapiro-Wilk W test for normality to determine the distribution of each data set. Probability plots were then constructed to help evaluate the extent the distribution of the measurements follows the normal distribution. We compared the data sets using the Student's t and Mann-Whitney U tests to evaluate what relationships exist between them, whether there was a significant difference. The t-test is used to evaluate normally distributed data, while the U test is used to evaluate data free of a definable distribution. When evidence was obtained that a difference exists between the data sets, descriptive statistics and box plots were evaluated to further assess the differences.

The reliability of a relation between variables observed in our samples can be quantitatively estimated and represented using a standard measure called p-value or statistical significance level. The *p*-level represents the probability of error involved in accepting our research hypothesis about the existence of a difference. Technically, this is the probability of error associated with rejecting the hypothesis of no difference between the two categories of observations, when the hypothesis is true. We used $p < 0.05$ to determine statistical significance.

We first analyzed the terrace deposit data sets. The Santa Clara site upstream of Los Alamos Canyon was compared to the sites downstream of Los Alamos Canyon. Then the terrace data sets were compared to the LANL background data set for northern New Mexico river sediments.

Analytical Detection Discussion

Analytic laboratories often report results that are below their measurement capabilities as non-detects, or less than a value determined as their detection level. Whenever the measurement technique permits, we reported and evaluated those values, as well as the uncertainty value, and minimum detection value for each sample. This follows recommendations by Environmental Protection Agency (1980, chapter 6), American society of Testing Materials (1984), and others as found in Gilbert (1987).

There were many measurements reported below their sample specific detection limit. Including these points into a data pool could generate a bias to the descriptive statistics for that group of data. Most of these cases or sediment intervals, with multiple non-detects, were eventually interpreted to originate from background conditions, including the Santa Clara deposits and the shallow sediment deposits at Cañada Ancha.

Multiple $^{239/240}$Pu and ^{137}Cs measurements were reported below their respective detection limits. Generally, the measurements reported below their detection limit were in the pre-1941 Santa Clara sediments, the shallow sediment intervals at Cañada Ancha, at Pajarito, and four of six measurements at the Water Canyon site. Alternatively, $^{239/240}$Pu and ^{137}Cs were detected in the post-1941 Santa Clara deposits, the mid to deep Ancha intervals, and

in all of the Frijoles sediment samples. A mixture of detections and non-detects were observed in the Pajarito and Water sites.

Americium-241 measurements demonstrated a similar pattern, in that most of the non-detects were observed in background conditions, at Santa Clara and the most shallow and deepest Ancha intervals. A mixture of detects and non-detects were observed in the sediment deposits in the downstream sites.

Almost all ^{238}Pu and most ^{90}Sr values were reported as non-detects or below their sample specific detection limit, except in the mid to deep sediment intervals at the Ancha site.

There were no ^{237}Np measurements reported above their respective detection limits.

Twenty-nine samples were measured for uranium isotopes -234, -235, and -240. All were reported above their sample specific detection limits except three ^{235}uranium values.

Normality Tests

The Shapiro-Wilk W test is used in testing for normality. If the W statistic is significant, provides $p < 0.05$, then the hypothesis that the respective distribution is normal should be rejected. The Shapiro-Wilk W test is the preferred test of normality because of its good power properties as compared to a wide range of alternative tests (Shapiro, Wilk, & Chen, 1968).

Table B1 identifies the data pools evaluated for normality using the Shapiro-Wilk's normality test. The data pools include LANL background river sediment data from McLin's (2002) report and the data acquired for this project. The radionuclides are identified in the first column and the number of measurements evaluated in the second. The p values are listed in the fourth column and are highlighted with red if $p < 0.05$. The distribution, whether it is normal or distribution free, is identified in the last column.

Table B1. Normality tests for radionuclides in background river sediments and in terrace deposits

Shapiro-Wilk W Tests for Normality				
LANL Background River Sediments				
	N	W statistic	p < 0.05 significance	Distribution
^{238}Pu	143	0.7063	0.0000	Distribution Free
$^{239/240}$Pu	142	0.6482	0.0000	Distribution Free
^{90}Sr	74	0.7656	0.0000	Distribution Free
^{241}Am	61	0.3129	0.0000	Distribution Free
^{137}Cs	131	0.8332	0.0000	Distribution Free
Rio Grande Terrace Deposits				
	N	W statistic	p < 0.05 significance	Distribution
^{238}Pu	32	0.9692	0.4764	Normal
$^{239/240}$Pu	32	0.6967	0.0000	Distribution Free
^{90}Sr	32	0.9338	0.0499	Distribution Free
^{234}U	29	0.9349	0.0738	Normal
^{235}U	29	0.9304	0.0564	Normal
^{238}U	29	0.9344	0.0717	Normal
^{241}Am	32	0.9783	0.7487	Normal
^{237}Np	32	0.9419	0.0847	Normal
^{137}Cs	32	0.6609	0.0000	Distribution Free

All radionuclide measurements from the LANL background river sediments are distribution free. The Shapiro-Wilk W tests did provide evidence that ^{238}Pu, uranium isotopes -234, -235, and -238, ^{241}Am, and ^{237}Np measurements made on the terrace deposits are from normally distributed populations. The Shapiro-Wilk W tests were not able to support that $^{239/240}$ Pu, ^{90}Sr, and ^{137}Cs measurements on terrace sediments are from normally distributed populations.

Normal Probability Plots

This type of graph is used to evaluate the normality of the variable distribution, that is, whether and to what extent the distribution of the variable follows the normal distribution. The radionuclide measurements were plotted in a scatter plot against the values "expected from the normal distribution."

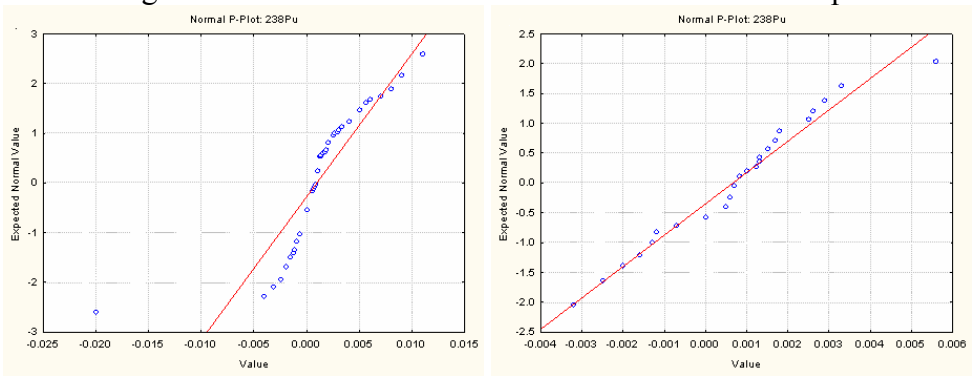
The standard normal probability plot is constructed as follows. First, the deviations from the mean, called residuals are rank ordered. From these ranks, z values or the standardized values of the normal distribution are computed based on the assumption that the data come from a normal distribution. These z values are plotted on the *Y*-axis in the plot. If the observed residuals, which are plotted on the *X*-axis, are normally distributed, then all values should fall onto a straight line. If the residuals are not normally

distributed, then they will deviate from the line. Outliers may also become evident in this plot.

Probability plots as well as normality tests were completed and presented below for each radionuclide data set established in this report. Radionuclides that demonstrated a normal distribution were tested using parametric tests, those that were not normally distributed, were tested using parametric as well as non-parametric tests.

Plutonium-238

Background Sediments | Rio Grande Terrace Deposits



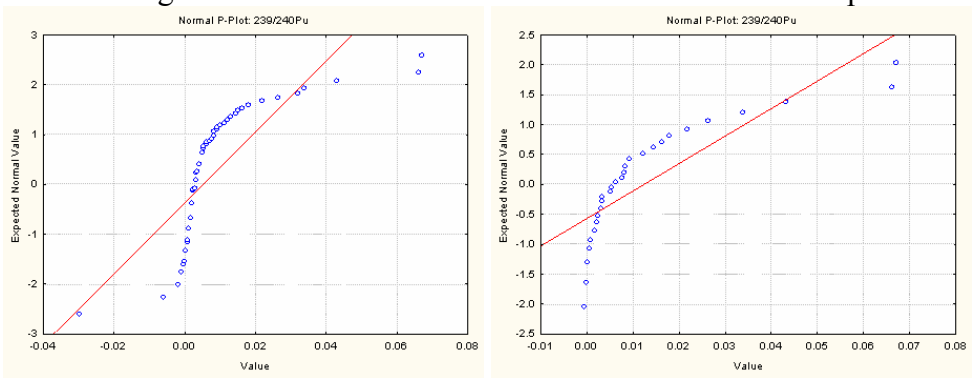
Non Normal Distribution | Normal Distribution

Figure B1. Probability plots for Plutonium-238 in background sediments and terrace deposits

Plutonium-238 measurements do not have a normal distribution in background sediments ($p = 0.0000$), while they do have a normal distribution in the Rio Grande terrace deposits ($p = 0.4764$). Both the parametric Student's t-tests and nonparametric alternative Mann-Whitney U tests were used to determine if sample groups are from the same population.

Plutonium-239/240

Background Sediments | Rio Grande Terrace Deposits



Non Normal Distribution | Non Normal Distribution

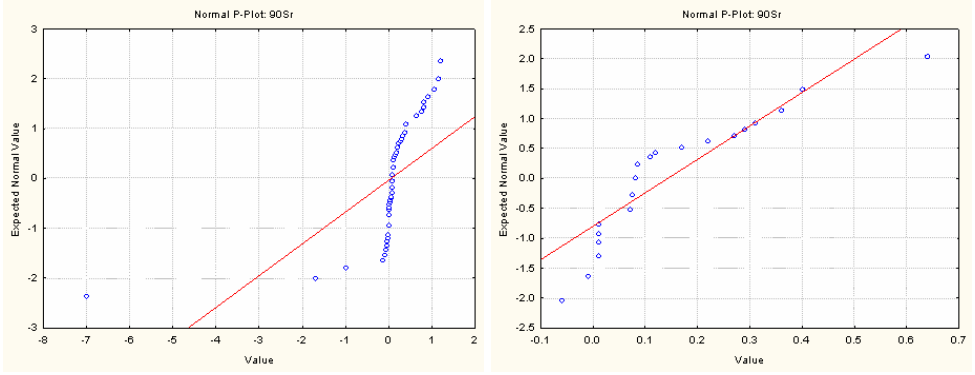
Figure B2. Probability plots for Plutonium-239/240 in background sediments and terrace deposits

Plutonium-239/240 measurements do not have a normal distribution in background sediments ($p = 0.0000$), nor do they have a normal distribution in the Rio Grande terrace

deposits (p = 0.0000). The Mann-Whitney U tests were used to determine if nonparametric sample groups are from the same population.

Strontium-90

Background Sediments | Rio Grande Terrace Deposits

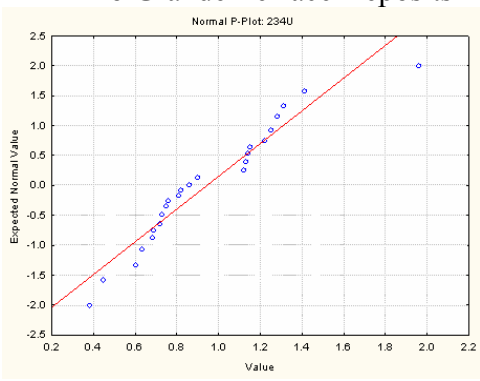


Non Normal Distribution | Non Normal Distribution

Figure B3. Probability plots for Strontium-90 in background sediments and terrace deposits

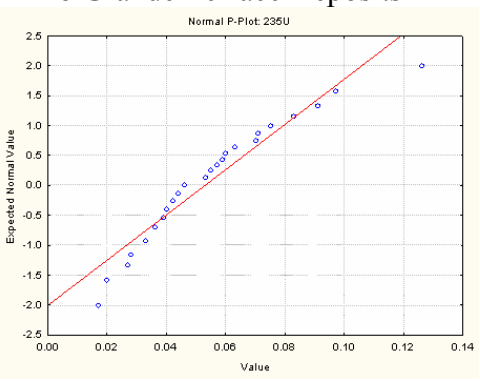
Strontium-90 measurements do not have a normal distribution in background sediments (p = 0.0000), nor do they have a normal distribution in the Rio Grande terrace deposits (p = 0.0499). The Mann-Whitney U tests were used to determine if nonparametric sample groups are from the same population.

Uranium-234
Rio Grande Terrace Deposits



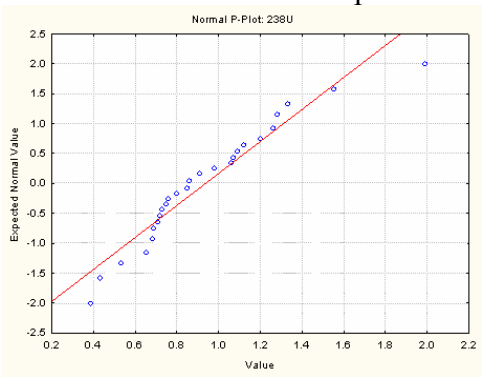
Normal Distribution

Uranium-235
Rio Grande Terrace Deposits



Normal Distribution

Uranium-238
Rio Grande Terrace Deposits



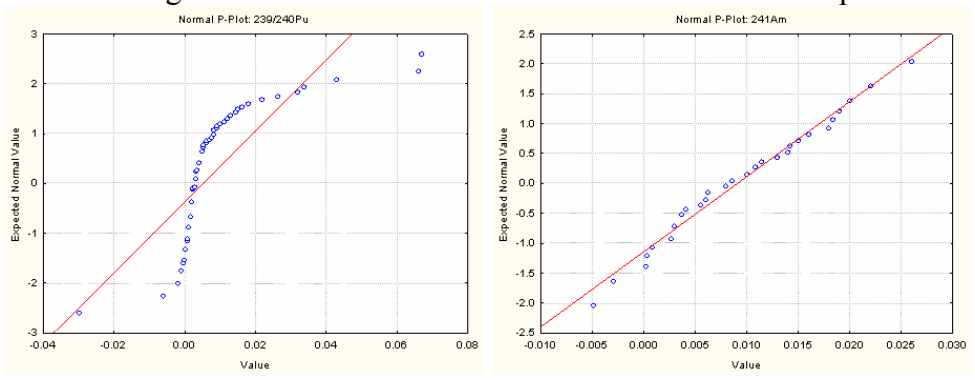
Normal Distribution

Figure B4. Probability plots for Uranium Isotopes-234, -235, and -238 in terrace deposits

Uranium-234, -235, and -238 isotope measurements have a normal distribution in the Rio Grande terrace deposits (p = 0.0738, 0.0564, 0.0717 respectively). The Student's t-tests were used to determine if parametric sample groups within the terrace deposits are from the same population. Isotopic Uranium measurements were not made for the Rio Grande background sediment.

Americium-241

Background Sediments | Rio Grande Terrace Deposits



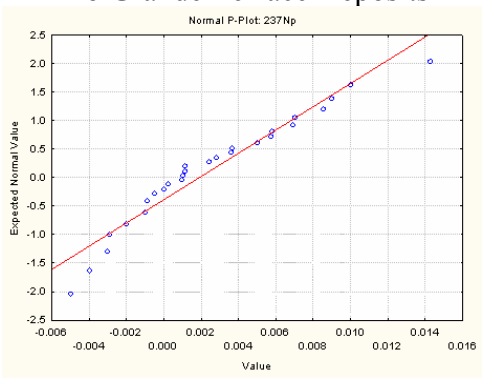
Non Normal Distribution | Normal Distribution

Figure B5. Probability plots for Americium-241 in background sediments and terrace deposits

Americium-241 measurements do not have a normal distribution in background sediments (p = 0.0000), while they do have a normal distribution in the Rio Grande terrace deposits (p = 0.7487). Both the parametric Student's t and the alternative Mann-Whitney U nonparametric tests were used to determine if sample groups are from the same population.

Neptunium-237

Rio Grande Terrace Deposits



Normal Distribution

Figure B6. Probability plots for Neptunium-237 in terrace deposits

Neptunium-237 measurements have a normal distribution in the Rio Grande terrace deposits (p = 0.0847). The Student's t-tests were used to determine if parametric sample groups within the terrace deposits are from the same population. Neptunium-237 measurements were not made for the Rio Grande background sediment.

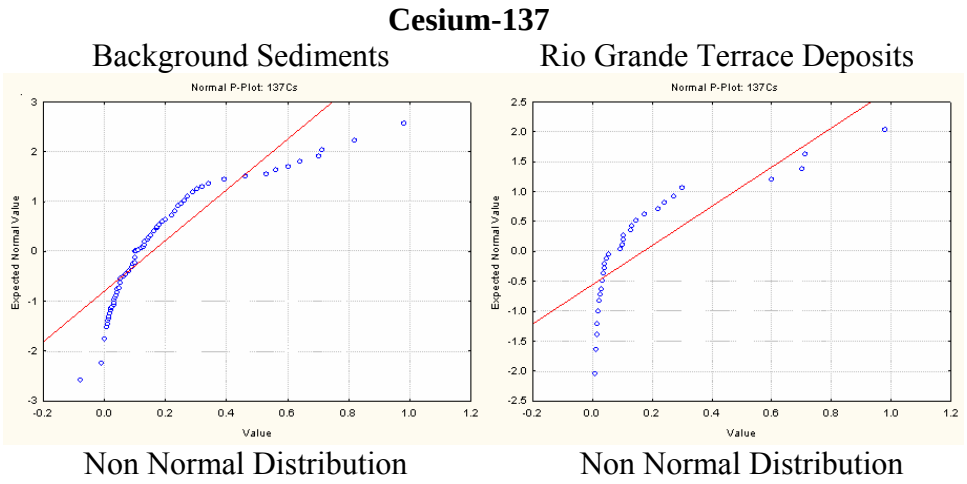


Figure B7. Probability plots for Cesium-137 in background sediments and terrace deposits

Cesium-137 measurements do not have a normal distribution in background sediments ($p = 0.0000$), nor do they have a normal distribution in the Rio Grande terrace deposits ($p = 0.0000$). The Mann-Whitney U tests were used to determine if nonparametric sample groups are from the same population.

Comparative Statistics

Student's t-test and the Mann-Whitney U test statistical methods were used to compare the sample sets described in this report and determine the relation between them, whether radionuclide concentrations in sediments collected along the Rio Grande are from the same sources. If evidence can be obtained from statistical tests that samples downstream of Los Alamos Canyon are different from the background reference samples, conclusions can be made that radionuclides originate from the Los Alamos National Laboratory.

Student's t-test is the most commonly used method to evaluate the differences in means between two groups. Theoretically, the t-test can be used even if sample sizes are very small, as long as the variables are normally distributed within each group and the variation of measurements in the two groups is not reliably different. As mentioned before, the normality assumption can be evaluated by looking at the distribution of the data or by performing a normality test. If these conditions are not met, then you can evaluate the differences in means between two groups using the nonparametric alternative Mann-Whitney U test.

Upstream Reference Deposits Compared to Downstream Deposits

The following tables summarize the comparative and descriptive test statistics for measurements of radionuclides in sediments collected from terrace cores and banks along the Rio Grande. The means and standard deviations, the Student's t-test and Mann-

Whitney U test statistics, as well as the derived p value for each sample are listed in the tables. The tests were marked significant at $p < 0.050$.

The cases described below include radionuclide measurements in strata collected from terraces along the Rio Grande, upstream of Los Alamos Canyon, and at four sites in White Rock Canyon downstream of Los Alamos Canyon. The case reflecting the site upstream of Los Alamos Canyon was collected along the Rio Grande at the Santa Clara pueblo and represents a non-impacted reference site. Deposition of sediments collected from these core samples occurred prior to 1943, well before atmospheric testing of nuclear weapons began. It does contain 1 sample deposited after 1941. Sediment samples collected from sites below Los Alamos Canyon were deposited after creation of the Los Alamos National Laboratory and may contain higher concentrations of radionuclides than the reference sites.

The reference sediment sample, represented by columns under the Santa Clara headings in each of the following four tables, reflect radionuclide concentrations that developed in channels and flood plains before and after nuclear atmospheric testing began. The remaining core samples, represented by the columns under individual site names in each table, consist of radionuclides measured in abandoned channels and old floodplains deposited after 1943, downstream of Los Alamos Canyon. The first table (Table B2) reflects cores collected from terraces at the Santa Clara site compared to the core collected from Cañada Ancha site. The remaining three tables reflect the Santa Clara cores compared to sediments collected in White Rock Canyon from terrace materials at Pajarito, Water, and Frijoles canyons. The p values are listed under the Student's t and Mann-Whitney U test columns at the far right side of the tables and are highlighted with red if $p < 0.05$.

Table B2. Statistical comparison of Santa Clara and Cañada Ancha terrace deposits

Santa Clara and Canada Ancha Comparisons								Student's t-		Mann-Whitney U Test Marked tests are significant at p <.050		
	Valid N		df	Mean		Std.Dev.		t-value	p	U	Z adjusted	p
	Santa Clara	Cañada Ancha		Santa Clara	Cañada Ancha	Santa Clara	Cañada Ancha					
238Pu	6	12	16	0.0001	0.0010	0.0009	0.0020	-1.0608	0.3045	20	-1.5024	0.1330
239/240Pu	6	12	16	0.0009	0.0227	0.0021	0.0247	-2.1223	0.0498	7.5	-2.6707	0.0076
90Sr	6	12	16	0.08	0.2767	0.0830	0.1841	-2.4654	0.0254	12	-2.2548	0.0241
234U	6	9	13	0.8883	1.1433	0.2797	0.4086	-1.3275	0.2072	19	-0.9445	0.3449
235U	6	9	13	0.0453	0.0650	0.0162	0.0303	-1.4456	0.1720	18	-1.0626	0.2880
238U	6	9	13	0.818	1.1811	0.2508	0.4192	-1.8938	0.0807	13	-1.6499	0.0990
241Am	6	12	16	0.0045	0.0106	0.0046	0.0096	-1.4571	0.1644	24	-1.1245	0.2608
237Np	6	12	16	0.0032	0.0020	0.0031	0.0055	0.4828	0.6358	26	0.9376	0.3485
137Cs	6	12	16	0.0305	0.3302	0.0351	0.3274	-1.9777	0.0654	9	-2.5301	0.0114

Plutonium-239/240 and ^{90}Sr measurements for the Santa Clara and Cañada Ancha terrace deposits are from different populations based on both the Student's t-test as well as the nonparametric Mann-Whitney U test. The Mann-Whitney U test also establishes evidence that ^{137}Cs measurements are from different populations. Plutonium-239/240, ^{90}Sr, and ^{137}Cs means for Cañada Ancha terrace deposits are approximately 25, 3, and 7 times greater than the Santa Clara means. These conclusions suggest a LANL impact to the Cañada Ancha terrace deposits.

Table B3. Statistical comparison of Santa Clara and Pajarito terrace deposits

Santa Clara and Pajarito Comparisons	Valid N		df	Mean		Std.Dev.		Student t-Test t-value	Student t-Test p	Mann-Whitney U Test (Marked tests are significant at p <.050) U	Mann-Whitney U Test Z adjusted	Mann-Whitney U Test p
	Santa Clara	Pajarito		Santa Clara	Pajarito	Santa Clara	Pajarito					
238Pu	6	2	6	0.00012	-0.00040	0.00093	0.00297	0.42746	0.68396	6.0	0.0000	1.0000
239/240Pu	6	2	6	0.00094	0.00525	0.00210	0.00332	-2.24404	0.06598	1.0	-1.6667	0.0956
90Sr	6	2	6	0.08	0.08250	0.08295	0.00354	-0.04043	0.96906	4.0	-0.6707	0.5024
234U	6	2	6	0.88833	0.58700	0.27974	0.20223	1.37510	0.21824	2.0	1.3413	0.1798
235U	6	2	6	0.04533	0.04150	0.01616	0.03041	0.24354	0.81570	5.0	-0.3333	0.7389
238U	6	2	6	0.818	0.57600	0.25076	0.20365	1.21700	0.26930	3.0	1.0000	0.3173
241Am	6	2	6	0.00450	0.00775	0.00459	0.00318	-0.90673	0.39950	3.5	-0.8434	0.3990
237Np	6	2	6	0.00322	0.00620	0.00305	0.00537	-1.03059	0.34247	3.0	-1.0000	0.3173
137Cs	6	2	6	0.03058	0.08675	0.03514	0.08096	-1.49344	0.18593	1.0	-1.6767	0.0936

Although the means for all radionuclide measurements at the Pajarito terrace deposits, with the exception of ^{238}Pu, are slightly larger than those for the Santa Clara terrace deposits, the differences are statistically insignificant. These small differences do not provide evidence that there is a LANL impact at this site.

Table B4. Statistical comparison of Santa Clara and Water Canyon terrace deposits

Sanat Clara and Water Comparisons	Valid N		df	Mean		Std.Dev.		Student's t- t-value	Student's t- p	Mann-Whitney U Test (Marked tests are significant at p <.050) U	Mann-Whitney U Test Z adjusted	Mann-Whitney U Test p
	Santa Clara	Water		Santa Clara	Water	Santa Clara	Water					
238Pu	6	6	10	0.00012	0.00027	0.00093	0.00213	-0.15828	0.87738	13.5	-0.7244	0.4688
239/240Pu	6	6	10	0.00094	0.00488	0.00210	0.00293	-2.67646	0.02323	3.0	-2.4019	0.0163
90Sr	6	6	10	0.080	0.07917	0.08295	0.00376	0.02458	0.98087	12.0	-0.9710	0.3315
234U	6	6	10	0.88833	0.70833	0.27974	0.08085	1.51414	0.16094	6.0	1.9249	0.0542
235U	6	6	10	0.04533	0.03750	0.01616	0.00650	1.10164	0.29643	10.0	1.2810	0.2002
238U	6	6	10	0.818	0.73000	0.25076	0.12538	0.76886	0.45975	10.5	1.2031	0.2290
241Am	6	6	10	0.00450	0.00853	0.00459	0.00664	-1.22407	0.24898	11.0	-1.1288	0.2590
237Np	6	6	10	0.00322	0.00017	0.00305	0.00458	1.35669	0.20471	8.0	1.6041	0.1087
137Cs	6	6	10	0.03058	0.05033	0.03514	0.04316	-0.86916	0.40514	10.0	-1.2855	0.1986

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Although the means for some of the radionuclide measurements in the Water Canyon terrace deposits are slightly larger than Santa Clara values, only $^{239/240}$Pu is significantly different. The plutonium mean is approximately five times greater than that for Santa Clara mean. These conclusions suggest a LANL impact.

Table B5. Statistical comparison of Santa Clara and Frijoles terrace deposits

Sanat Clara and Frijoles Comparisons	Valid N		df	Mean		Std.Dev.		Student's t-test (Marked tests are significant at p <.050) t-value	p	Mann-Whitney U Test U	Z adjusted	p
	Santa Clara	Frijoles		Santa Clara	Frijoles	Santa Clara	Frijoles					
238Pu	6	6	10	0.00012	0.00125	0.00093	0.00164	-1.47201	0.17178	11.0	-1.1248	0.2607
239/240Pu	6	6	10	0.00094	0.01307	0.00210	0.00644	-4.38266	0.00137	0.0	-2.8823	0.0039
90Sr	6	6	10	0.08	0.02158	0.08295	0.02618	1.64514	0.13097	13.5	0.7244	0.4688
234U	6	6	10	0.88833	1.05667	0.27974	0.27391	-1.05317	0.31705	12.0	-0.9625	0.3358
235U	6	6	10	0.04533	0.06217	0.01616	0.02524	-1.37594	0.19887	11.0	-1.1209	0.2623
238U	6	6	10	0.818	1.02000	0.25076	0.26435	-1.35798	0.20432	9.5	-1.3635	0.1727
241Am	6	6	10	0.00450	0.01172	0.00459	0.00714	-2.08203	0.06398	6.0	-1.9249	0.0542
237Np	6	6	10	0.00322	0.00063	0.00305	0.00381	1.29595	0.22411	10.0	1.2833	0.1994
137Cs	6	6	10	0.03058	0.17000	0.03514	0.07921	-3.94074	0.00277	2.0	-2.5665	0.0103

The means for most radioactive measurements at the Frijoles site are only slightly larger than at the Santa Clara site, with the exception of $^{239/240}$Pu and ^{137}Cs. The plutonium-239/240 and ^{137}Cs means are both significantly different than those at the Santa Clara, approximately 14 and six times greater, respectively. These conclusions suggest a LANL impact.

Box Plots

Box and Whisker plots were created to evaluate and "intuitively visualize" the ranges of radionuclide measurements within a sample population, or case, and the relationships between cases. The box plots demonstrate the medians, the 25th and 75th quartiles, the minimum and maximum typical values, and the mild and extreme outliers for analytical measurements from each case.

In these plots a box, representing the 25th and 75th quartile, is placed around the median. Whiskers, a line with a serif on both ends, reflect the typical minimum and maximum value. Mild and extreme outliers, which are atypical, infrequent observations, are represented by open circles and # symbols. Outlier definitions are subjective, and in these cases, the mild outliers are measurements greater than 1.5 times the difference between the 25th and 75th quartiles, while the extreme outliers are greater than 3 times the quartile difference.

As mentioned above, outliers are atypical, infrequent measurements that might represent random error in field sampling or the analytical measurements. It should also be noted that these measurements might be indicative of an environmental phenomenon that is qualitatively different than the typical observations, for example from a detrimental impact to the environment. These departures from typical measurements observed within a population were individually evaluated.

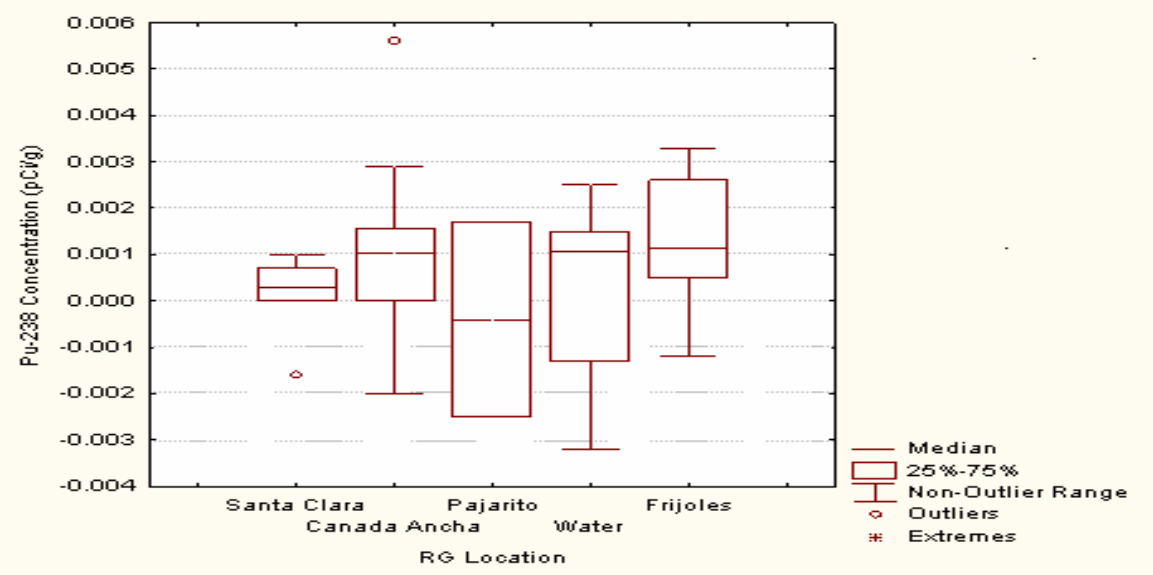
Box Plots For Upstream Reference Deposits and Downstream Deposits

The box plots presented in this section describe the radionuclide measurements in strata collected from terraces along the Rio Grande, upstream of Los Alamos Canyon, and at four sites in White Rock Canyon below Los Alamos Canyon. Recall that the case

reflecting the site upstream of Los Alamos Canyon is data from sediments collected along the Rio Grande at the Santa Clara site and represents non-impacted reference sediments. Deposition of buried sediments collected from this core sample occurred prior to and after atmospheric testing of nuclear weapons began. Sediment samples collected from sites below Los Alamos Canyon were deposited after creation of the Los Alamos National Laboratory and may contain higher concentrations of radionuclides than the reference sites.

The box plot at the left side of each chart is the Santa Clara case, while the remaining box plots reflect sediment strata from terraces along the Rio Grande near Cañada Ancha, Pajarito, Water, and Frijoles canyons. The box plots represent relatively small data sets, from two to 12 measurements, including outliers. These outliers or unusual measurements could reflect potential analytical or sampling error, such as cross contamination, as well as concentration variability associated with episodic deposition. Nine charts were made, one for each of the following radionuclides; ^{238}Pu, $^{239/240}$Pu, ^{90}Sr, uranium isotopes -234, -235, -238, ^{241}Am, ^{237}Np, and ^{137}Cs.

Plutonium-238

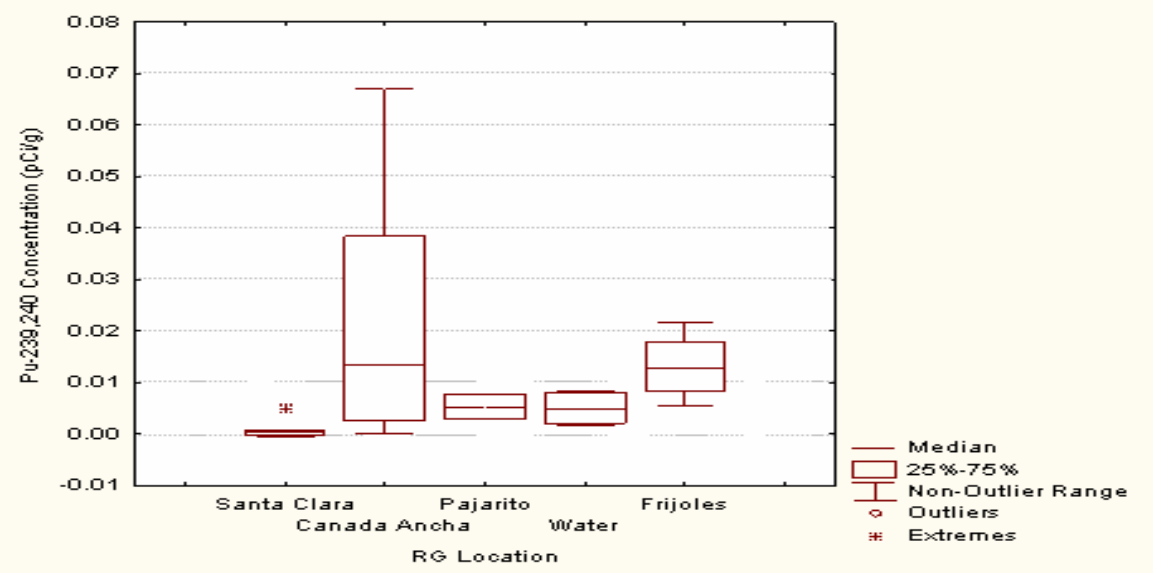


Pu^{238}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.0003	0.0010	-0.0004	0.0011	0.0012
25th	0.0000	0.0000	-0.0025	-0.0013	0.0005
75th	0.0007	0.0016	0.0017	0.0015	0.0026
Min	-0.0016	-0.0020	-0.0025	-0.0032	-0.0012
Max	0.0010	0.0056	0.0017	0.0025	0.0033

Figure B8. Plutonium-238 box plots for the Santa Clara reference site compared to downstream terrace deposits

These box plots demonstrate that the differences between Pu^{238} measurements in the terrace groups are insignificant; all the p values are greater than 0.050. A single outlier in

the Cañada Ancha sample suggests a potential environmental impact from LANL. The evidence is inconclusive.

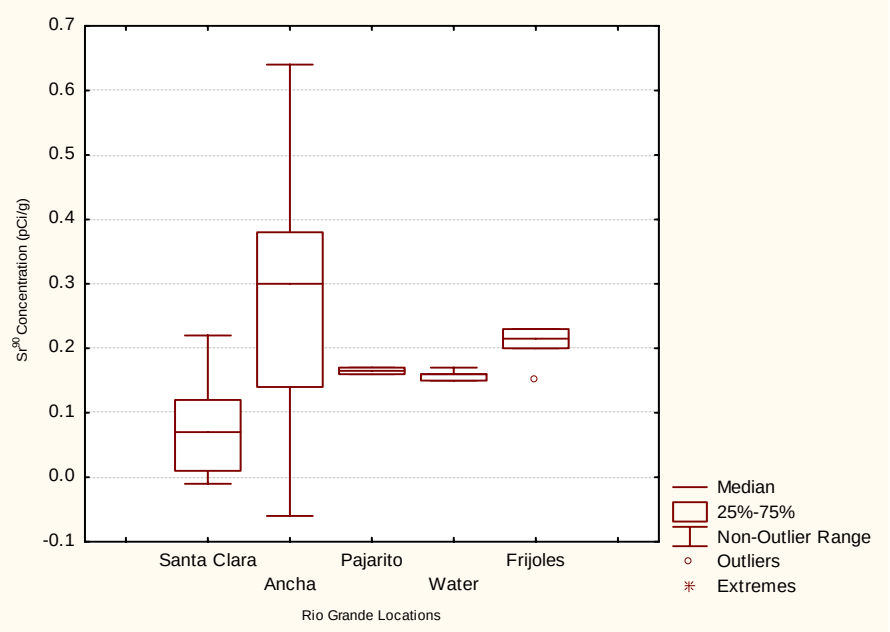


$Pu^{239/240}$	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.0003	0.0133	0.0053	0.0047	0.0127
25th	-0.0002	0.0026	0.0029	0.0021	0.0082
75th	0.0008	0.0384	0.0017	0.0080	0.0179
Min	-0.0006	0.0000	0.0029	0.0017	0.0053
Max	0.0051	0.0670	0.0076	0.0082	0.0217

Figure B9. Plutonium-239/240 box plots for the Santa Clara reference site compared to downstream terrace deposits

These box plots demonstrate significant differences exist between Santa Clara and the Cañada Ancha, Water, and Frijoles sites with respect to $^{239/240}$Pu. The medians and upper values are substantially larger than those in Santa Clara. The difference at Pajarito was determined not significant at $p = 0.066$, all other p values are less than 0.050. The maximum value in the Santa Clara case was the surface value of 0.005 pCi/g. Although a low level, this outlier is atypical of the much lower values found at the Santa Clara site.

A relatively larger range of measurements in the Cañada Ancha terrace, 0.000 to 0.067 pCi/g, might reflect episodic deposition. Grain size distribution analysis indicated large variability as well. The variability in these measurements, contaminant as well as grain size, may be indicative of episodic deposition, ranging from channel to floodplain like deposits originating from background to LANL inputs.

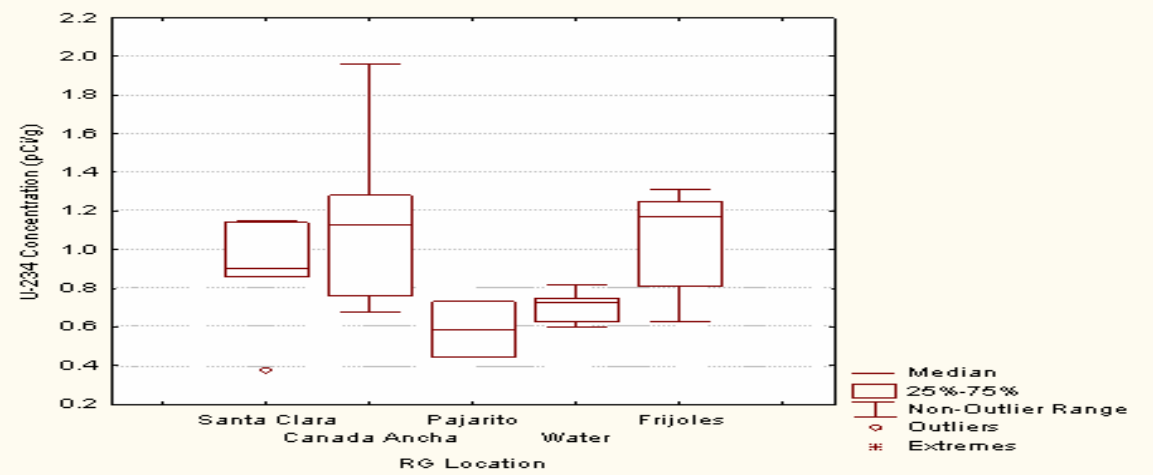


Sr^{90}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.07	0.30	0.17	0.16	0.22
25th	0.01	0.14	0.16	0.15	0.20
75th	0.12	0.38	0.17	0.16	0.23
Min	-0.01	-0.06	0.16	0.15	0.15
Max	0.22	0.64	0.17	0.17	0.23

Figure B10. Strontium-90 box plots for the Santa Clara reference site compared to downstream terrace deposits

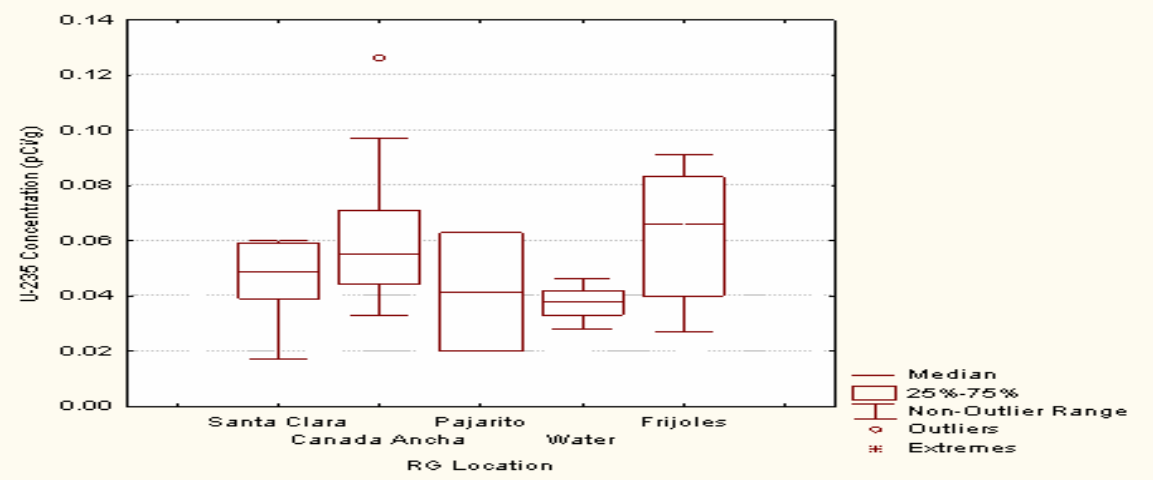
The box plots for ^{90}Sr demonstrate the only significant differences between Santa Clara and the other cases was at Cañada Ancha, p value = 0.025. The other terrace locations demonstrate little difference to Santa Clara, the p values were all greater than 0.050. The relatively large range of strontium values, -0.06 to 0.64 pCi/g, at the Cañada Ancha site may also reflect episodic deposition.

Uranium-234



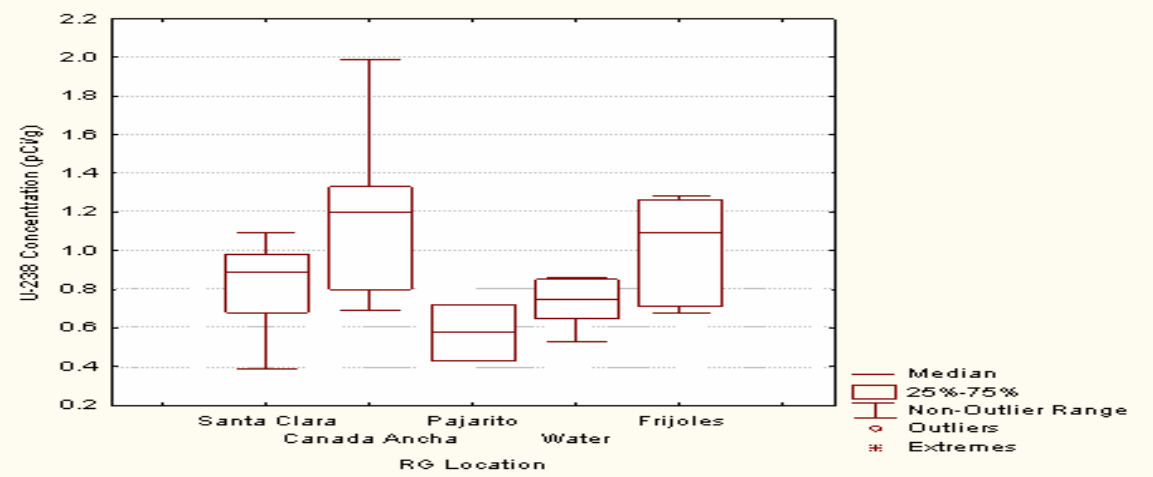
U^{234}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.90	1.13	0.59	0.73	1.17
25th	0.86	0.76	0.44	0.63	0.81
75th	1.14	1.28	0.73	0.75	1.25
Min	0.38	0.68	0.44	0.60	0.63
Max	1.15	1.96	0.73	0.82	1.31

Uranium-235



U^{235}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.049	0.055	0.042	0.038	0.066
25th	0.039	0.040	0.020	0.033	0.040
75th	0.059	0.083	0.063	0.042	0.083
Min	0.017	0.027	0.020	0.028	0.027
Max	0.060	0.091	0.063	0.046	0.091

Uranium-238

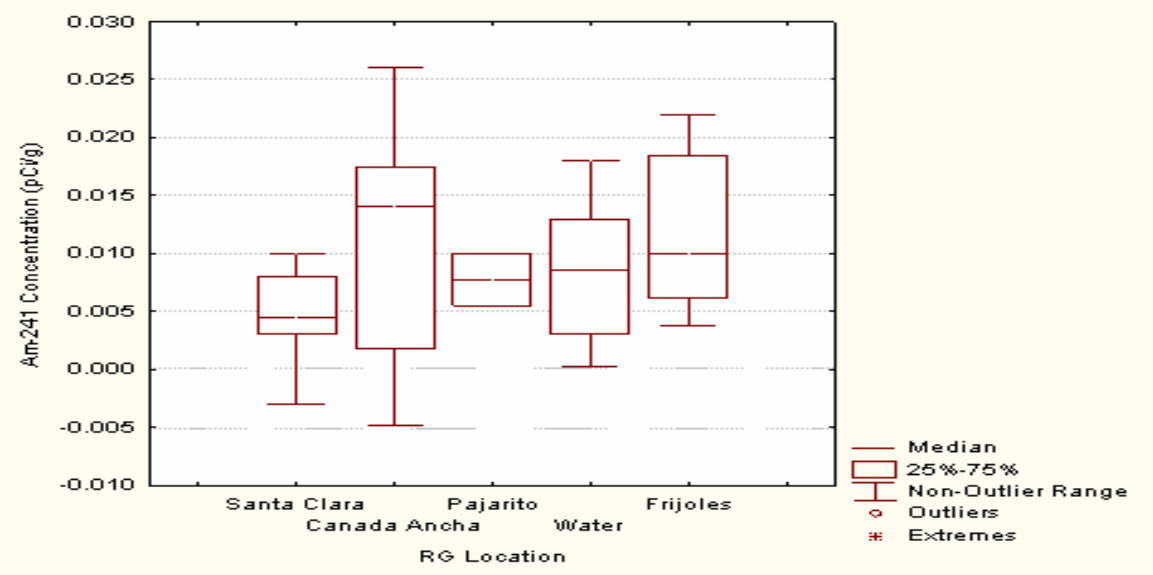


U^{238}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.885	1.200	0.576	0.745	1.095
25th	0.680	0.800	0.432	0.650	0.710
75th	0.980	1.330	0.720	0.850	1.260
Min	0.388	0.690	0.432	0.530	0.680
Max	1.090	1.990	0.720	0.860	1.280

Figure B11. Uranium Isotopes-234, -235, and -238 box plots for the Santa Clara reference site compared to downstream terrace deposits

Box plots for uranium-234, -235, and -238 isotopes demonstrate little difference in measurements between the Santa Clara reference site and the remaining terrace locations below the Laboratory.

Americium-241

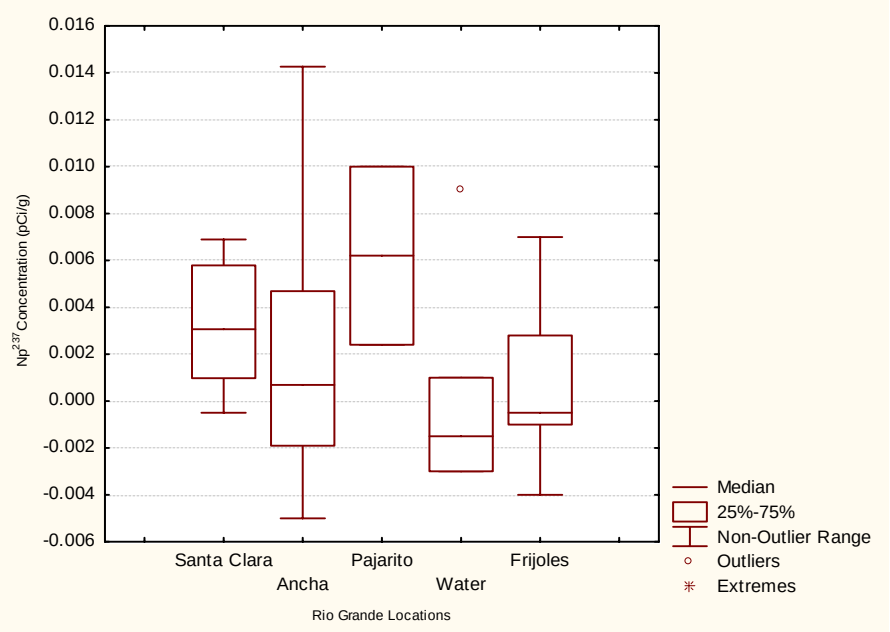


Am^{241}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.0045	0.0141	0.0078	0.0085	0.0100
25th	0.0030	0.0018	0.0055	0.0030	0.0062
75th	0.0080	0.0175	0.0100	0.0130	0.0184
Min	-0.0030	-0.0049	0.0055	0.0002	0.0037
Max	0.0100	0.0260	0.0100	0.0180	0.0220

Figure B12. Americium-241 Box Plots for Santa Clara, the Reference Conditions, Compared to Downstream Terrace Deposits

The box plots for ^{241}Am demonstrate little difference in measurements between the Santa Clara reference site and the remaining terrace locations below the Laboratory. The wide range of data at the Cañada Ancha site might reflect episodic deposition, although the evidence is inconclusive.

Neptunium-237

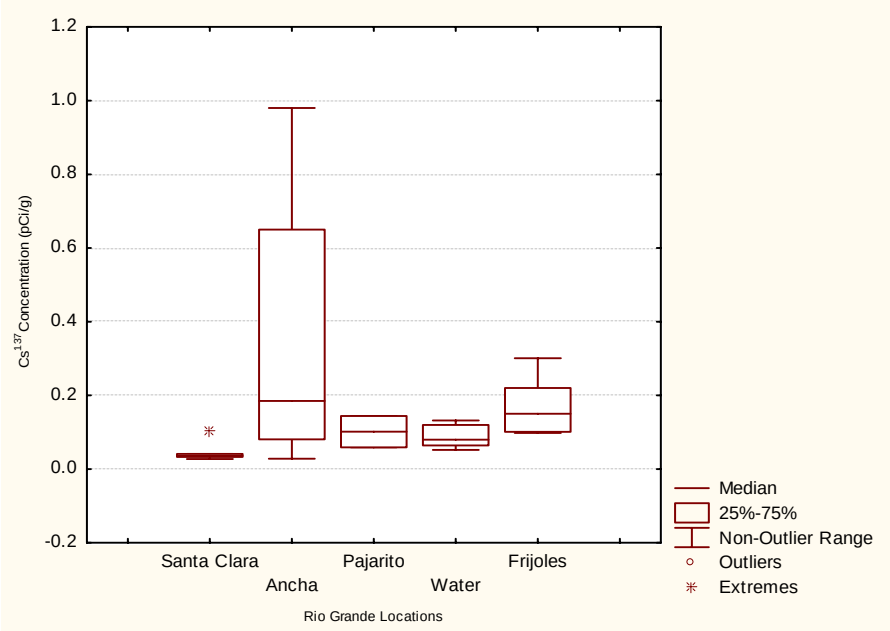


Np^{237}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.0031	0.0007	0.0062	-0.0015	-0.0005
25th	0.0010	-0.0019	0.0024	-0.0030	-0.0010
75th	0.0058	0.0047	0.0100	0.0010	0.0028
Min	-0.0005	-0.0050	0.0024	-0.0030	-0.0040
Max	0.0069	0.0143	0.0100	0.0090	0.0070

Figure B13. Neptunium-237 box plots for the Santa Clara reference site compared to downstream terrace deposits

The box plots for ^{237}Np demonstrate little difference in measurements between the Santa Clara reference site and the remaining terrace locations below the Laboratory. All measurements were reported as non-detects. The only conclusion that can be made is ^{237}Np was not found above detection limits upstream or downstream from the Laboratory.

Cesium-137



Cs^{137}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles
Median	0.036	0.185	0.102	0.080	0.150
25th	0.033	0.081	0.059	0.064	0.101
75th	0.041	0.650	0.144	0.120	0.220
Min	0.027	0.028	0.059	0.052	0.098
Max	0.102	0.980	0.144	0.132	0.301

Figure B14. Cesium-137 box plots for the Santa Clara reference site compared to downstream terrace deposits

The box plots demonstrate significant differences in ^{137}Cs measurements between the Santa Clara reference case and the Cañada Ancha ($p = 0.011$) and Frijoles ($p = 0.01$) sites below the laboratory. All other p values were greater than 0.050. The relatively large range of measurements at the Cañada Ancha site may reflect episodic deposition, while the smaller range at the Frijoles site may suggest deposition was uniformly distributed during flooding of Cochiti Reservoir.

Background Reference River Sediments Compared to Terrace Deposits

The following tables summarize the comparative and descriptive test statistics for measurements of radionuclides in sediments collected from terrace cores and background sediments collected by LANL. The means and standard deviations, the Student's t-test and Mann-Whitney U test statistics, as well as the derived p value for each sample are listed in the tables. The tests were marked significant at $p < 0.050$.

We collected sediments from cores drilled into terraces above and below Los Alamos Canyon to examine historic depositional variances. The LANL Environmental Surveillance Program has collected bank sediments since the 1970's to establish reference conditions. These sediments were collected in river channels not affected by the Laboratory. The data can be found in the LANL report "Background Radioactivity in Rivers and Reservoir Sediments near Los Alamos, New Mexico" (McLin and others, 2002).

The reference sediment case, represented by columns under the Rio Grande Background (RG Bkgnd) headings, reflects radionuclide concentrations derived from the dispersion and transport of global fallout materials in northern New Mexico. The sample includes from 29 to over 100 measurements. The core samples, represented by the columns under individual site names in each table, consist of a relatively small number of samples collected from abandoned channels and old floodplains. The first table reflects cores collected from terraces at the Santa Clara site, upstream of Los Alamos Canyon. The remaining four tables reflect cores collected in White Rock Canyon from terrace materials deposited after the Los Alamos National Laboratory was created.

Table B6. Statistical comparison of background sediments and Santa Clara terrace deposits

Rio Grande Background and Santa Clara Comparisons								Student's t-		Mann-Whitney U Test		
								Marked tests are significant at p <.050				
	Valid N		df	Mean		Std.Dev.		t-value	p	U	Z adjusted	p
	RG Bkgnd	Santa Clara	df	RG Bkgnd	Santa Clara	RG Bkgnd	Santa Clara					
238Pu	111	6	115	0.0011	0.0001	0.0032	0.0009	0.7371	0.4626	242.5	1.1523	0.2492
239/240Pu	110	6	114	0.0031	0.0009	0.0052	0.0021	0.9868	0.3258	145.0	2.3307	0.0198
90Sr	42	6	46	-0.0138	0.0800	1.2104	0.0829	-0.1880	0.8517	121.0	0.1562	0.8758
241Am	29	6	33	-0.1005	0.0045	0.6111	0.0046	-0.4158	0.6802	67.0	-0.8845	0.3764
137Cs	99	6	103	0.1527	0.0306	0.1400	0.0351	2.1239	0.0361	93.0	2.8194	0.0048

Plutonium-239/240 measurements for the Santa Clara terrace deposits and background sediments are from different populations based on the Mann-Whitney U test for distribution free data populations. Evidence that ^{137}Cs is from different populations exists from both the Student's t-test and the Mann-Whitney U test. Plutonium-239/240 and ^{137}Cs means for background deposits are approximately three and five times greater than the Santa Clara means. These conclusions support the hypothesis that most of the Santa Clara deposits were deposited prior to nuclear atmospheric testing.

Table B7. Statistical comparison of background sediments and Cañada Ancha terrace deposits

Rio Grande Background and Cañada Ancha Comparisons								Student's t-		Mann-Whitney U Test		
								Marked tests are significant at p <.050				
	Valid N		df	Mean		Std.Dev.		t-value	p	U	Z adjusted	p
	RG Bkgnd	Cañada Ancha	df	RG Bkgnd	Cañada Ancha	RG Bkgnd	Cañada Ancha					
238Pu	111	12	121	0.0011	0.0010	0.0032	0.0020	0.0460	0.9634	632.0	-0.2964	0.7670
239/240Pu	110	12	120	0.0031	0.0227	0.0052	0.0247	-7.1940	0.0000	334.0	-2.8282	0.0047
90Sr	42	12	52	-0.0138	0.2767	1.2104	0.1841	-0.8230	0.4143	155.0	-2.0218	0.0432
241Am	29	12	39	-0.1005	0.0106	0.6111	0.0096	-0.6249	0.5357	136.0	-1.0934	0.2742
137Cs	99	12	109	0.1527	0.3120	0.1400	0.3425	-3.0366	0.0030	532.0	-0.5894	0.5556

Plutonium-239/240, ^{90}Sr, and ^{137}Cs measurements for the background sediments and the Cañada Ancha terrace deposits are from different populations. A $^{239/240}$Pu difference was based on both the Student's t-test as well as the Mann-Whitney U test alternative. Evidence that ^{90}Sr is from different populations exists from the Mann-Whitney U test, and the ^{137}Cs difference is determined from the Student's t-test. Plutonium-239/240 and ^{137}Cs means for Cañada Ancha terrace deposits are approximately seven and two times greater than the background means. However the interpretation that a cesium difference exists is inconclusive when based on the parametric t-test value derived for nonparametric data. The ^{90}Sr mean in Cañada Ancha sediments is approximately 0.28 pCi/g while the background value is negative. These conclusions suggest a LANL impact to the Cañada Ancha terrace deposits.

Table B8. Statistical comparison of background sediments and Pajarito terrace deposits

Rio Grande Background and Pajarito Comparisons	Valid N		df	Mean		Std.Dev.		Student t-Test t-value	Student t-Test p	Mann-Whitney U Test (Marked tests are significant at p <.050) U	Mann-Whitney U Test Z adjusted	Mann-Whitney U Test p
	RG Bkgnd	Pajarito		RG Bkgnd	Pajarito	RG Bkgnd	Pajarito					
238Pu	111	2	111	0.0011	-0.0004	0.0032	0.0030	0.6542	0.5144	83.0	0.6277	0.5302
239/240Pu	110	2	110	0.0031	0.0053	0.0052	0.0033	-0.5884	0.5575	65.0	-1.0003	0.3172
90Sr	42	2	42	-0.0138	0.0825	1.2104	0.0035	-0.1112	0.9120	39.5	0.1413	0.8877
241Am	29	2	29	-0.1005	0.0078	0.6111	0.0032	-0.2465	0.8070	11.0	-1.4616	0.1439
137Cs	99	2	99	0.1527	0.0868	0.1400	0.0810	0.6620	0.5095	68.0	0.7566	0.4493

Statistical tests of radionuclide measurements in background sediments and those at the Pajarito terrace deposits do not indicate a significant difference in values.

Table B9. Statistical comparison of background sediments and Water Canyon terrace deposits

Rio Grande Background and Water Comparisons	Valid N		df	Mean		Std.Dev.		Student's t-test t-value	Student's t-test p	Mann-Whitney U Test (Marked tests are significant at p <.050) U	Mann-Whitney U Test Z adjusted	Mann-Whitney U Test p
	RG Bkgnd	Water		RG Bkgnd	Water	RG Bkgnd	Water					
238Pu	111	6	115	0.0011	0.0003	0.0032	0.0021	0.6163	0.5389	314.0	0.2410	0.8096
239/240Pu	110	6	114	0.0031	0.0049	0.0052	0.0029	-0.8440	0.4005	202.5	-1.6063	0.1082
90Sr	42	6	46	-0.0138	0.0792	1.2104	0.0038	-0.1863	0.8530	116.5	0.2969	0.7665
241Am	29	6	33	-0.1005	0.0085	0.6111	0.0066	-0.4318	0.6687	58.0	-1.2798	0.2006
137Cs	99	6	103	0.1527	0.0503	0.1400	0.0432	1.7790	0.0782	125.0	2.3772	0.0174

The Mann-Whitney U tests suggest there is a significant difference in the ^{137}Cs measurements between background and terrace deposits at the Water Canyon site. The background mean value is three times greater than found at the Water Canyon site. This suggests no evidence of a LANL impact.

Table B10. Statistical comparison of background sediments and Frijoles terrace deposits

Rio Grande Background and Frijoles Comparisons								Student's t-		Mann-Whitney U Test		
								Marked tests are significant at p <.050				
	Valid N		df	Mean		Std.Dev.		t-value	p	U	Z adjusted	p
	RG Bkgnd	Frijoles	df	RG Bkgnd	Frijoles	RG Bkgnd	Frijoles					
238Pu	111	6	115	0.0011	0.0013	0.0032	0.0016	-0.1367	0.8915	289.0	-0.5581	0.5768
239/240Pu	110	6	114	0.0031	0.0131	0.0052	0.0064	-4.5177	0.0000	33.0	-3.7416	0.0002
90Sr	42	6	46	-0.0138	0.0216	1.2104	0.0262	-0.0709	0.9438	93.0	1.0312	0.3025
241Am	29	6	33	-0.1005	0.0117	0.6111	0.0071	-0.4444	0.6597	35.0	-2.2914	0.0219
137Cs	99	6	103	0.1527	0.1700	0.1400	0.0792	-0.2984	0.7660	228.0	-0.9536	0.3403

The Student's t-tests and the Mann-Whitney U tests suggest there is a significant difference between the $^{239/240}$Pu measurements from background and the Frijoles Canyon site. The $^{239/240}$Pu mean value at the Frijoles site is four times greater than background. The Mann-Whitney U test suggests that ^{241}Am is different in background from the Frijoles site. The mean value for ^{241}Am at the Frijoles site is 0.012 pCi/g, while it is negative in the background sediments. This evidence suggests there is a LANL impact.

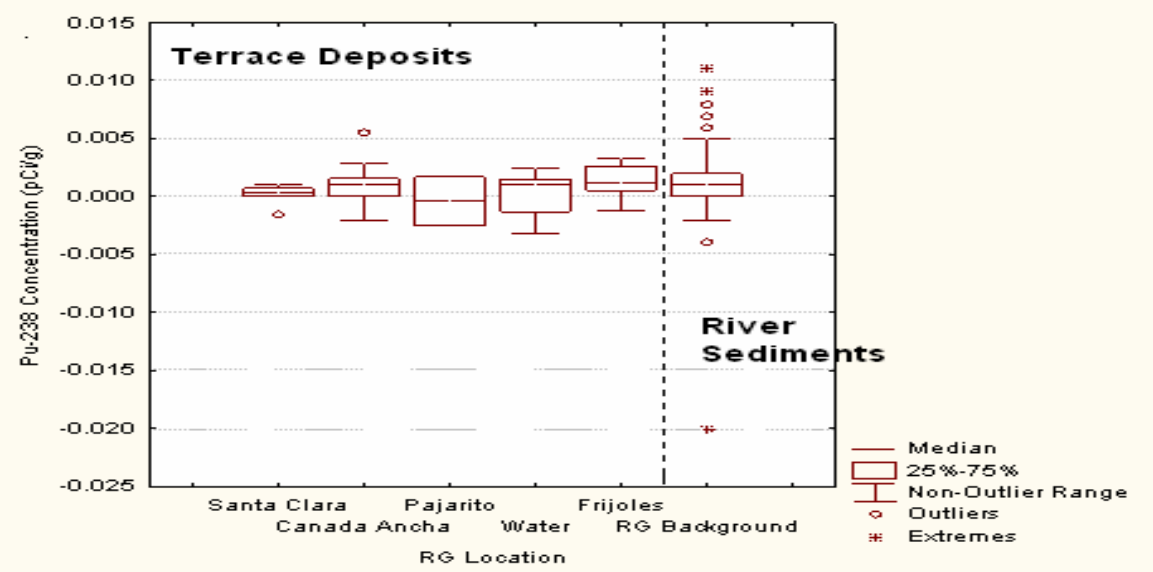
Box Plots For Background Reference River Sediments and Terrace Deposits

The following box plots show measurements of radionuclides in sediments collected from terrace cores along the Rio Grande and background sediments collected by LANL. We collected sediments from cores drilled into terraces above and below Los Alamos Canyon to examine historical deposition variances. The LANL Environmental Surveillance Program has collected bank sediments since the 1970's to establish reference conditions.

The reference sediment sample, represented by the box plots on the far right of each chart, reflects radionuclide concentrations that develop from the dispersion and transport of global fallout materials in northern New Mexico. The sample includes 29 to over 100 measurements as well as outliers and extreme measurements. These unusual measurements could reflect potential analytical or sampling error. The core samples, represented by the preceding five box plots in each chart, consist of a relatively small number of samples collected from abandoned channels and floodplains. The far left box plot reflects cores collected from terraces at the Santa Clara site, upstream of Los Alamos Canyon. The remaining four box plots reflect cores collected in White Rock Canyon from terrace materials deposited after Los Alamos National Laboratory was created.

Charts were created for ^{238}Pu, $^{239/240}$Pu, ^{90}Sr, ^{241}Am, and ^{137}Cs. Neptunium-237 and uranium isotopes were not analyzed for the background river sediments.

Plutonium-238

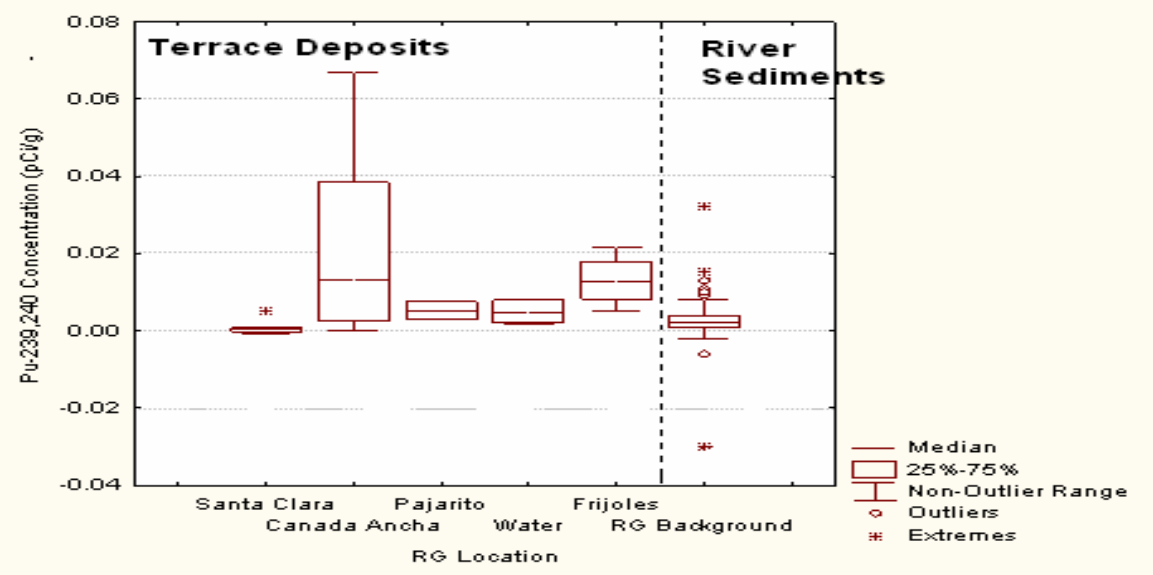


Pu^{238}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles	River Sediments
Median	0.0003	0.0010	-0.0004	0.0011	0.0012	0.0010
25th	0.0000	0.0000	-0.0025	-0.0013	0.0005	0.0000
75th	0.0007	0.0016	0.0017	0.0015	0.0026	0.0020
Min	-0.0016	-0.0020	-0.0025	-0.0032	-0.0012	-0.0200
Max	0.0010	0.0056	0.0017	0.0025	0.0033	0.0110

Figure B15. Plutonium-238 box plots for background reference river sediments compared to terrace deposits

These box plots demonstrate the differences in ^{238}Pu measurements between the terrace groups and background sediments are insignificant; all p values are greater than 0.050. A single outlier in the Cañada Ancha sample suggests a potential environmental impact from LANL. The evidence is inconclusive of a LANL impact.

Plutonium-239/240



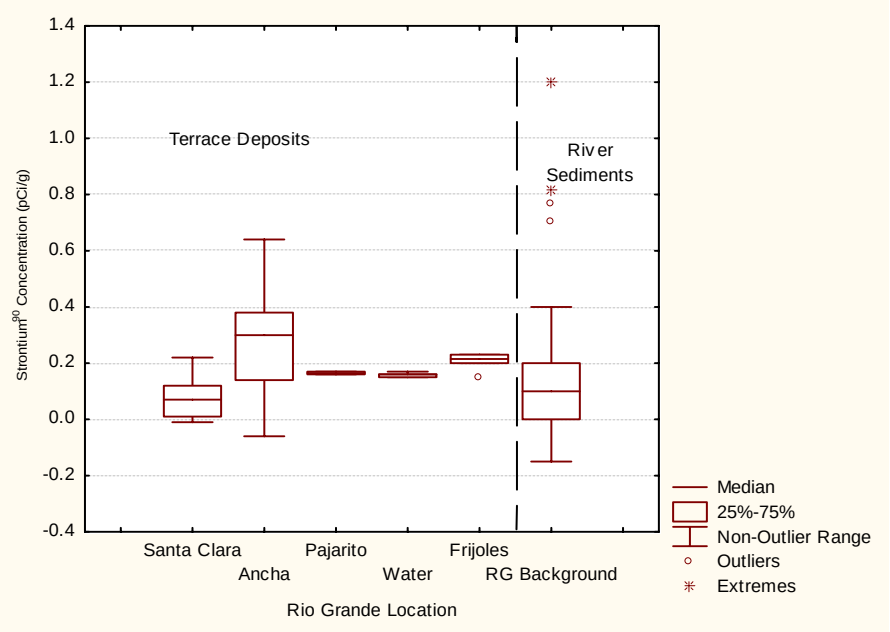
$Pu^{239/240}$	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles	River Sediments
Median	0.0003	0.0133	0.0053	0.0047	0.0127	0.0020
25th	-0.0002	0.0026	0.0029	0.0021	0.0082	0.0010
75th	0.0008	0.0384	0.0017	0.0080	0.0179	0.0040
Min	-0.0006	0.0000	0.0029	0.0017	0.0053	-0.0300
Max	0.0051	0.0670	0.0076	0.0082	0.0217	0.0320

Figure B16. Plutonium-239/240 box plots for background reference river sediments compared to terrace deposits

These box plots demonstrate significant differences between the background sediments and Santa Clara ($p = 0.020$), Cañada Ancha ($p = 0.0047$), and Frijoles ($p = 0.000$) cases. The medians and upper values in Santa Clara are less than those in background. The medians and upper values at the Cañada Ancha and Frijoles sites are substantially larger than those in background.

This evidence suggests that buried Santa Clara sediments are relatively free of $^{239/240}$Pu from global fallout. The maximum value at Santa Clara is 0.0008 pCi/g, notwithstanding the 0.005 pCi/g surface value. The Cañada Ancha and Frijoles sediments, on the other hand, have $^{239/240}$Pu concentrations much greater than background. Plutonium-239/240 concentrations are up to six times greater in the Cañada Ancha and Frijoles sites, suggesting a LANL impact.

Strontium-90

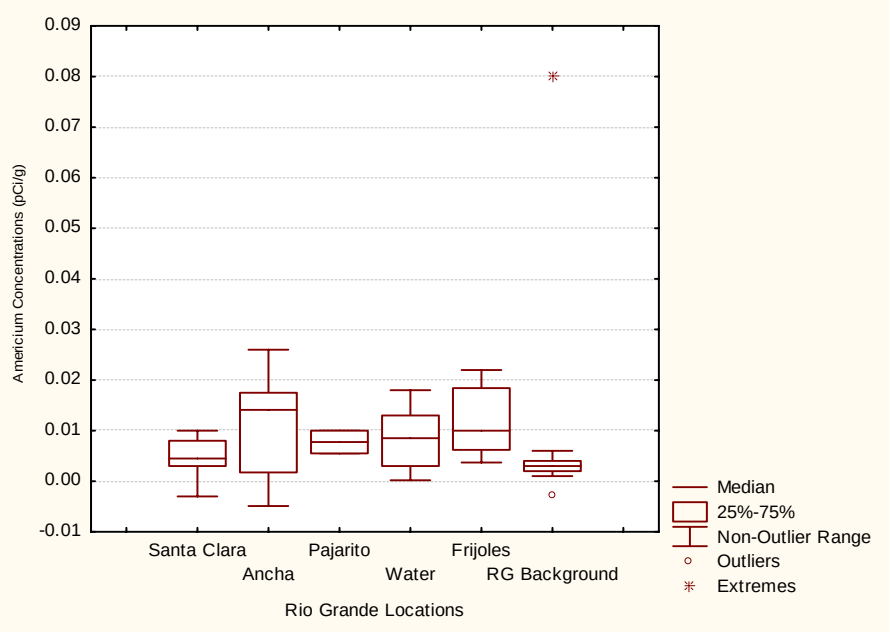


Sr^{90}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles	River Sediments
Median	0.07	0.30	0.17	0.16	0.22	0.10
25th	0.01	0.14	0.16	0.15	0.20	0.00
75th	0.12	0.38	0.17	0.16	0.23	0.20
Min	-0.01	-0.06	0.16	0.15	0.15	-0.15
Max	0.22	0.64	0.17	0.17	0.23	1.20

Figure B17. Strontium-90 box plots for background reference river sediments compared to terrace deposits

The box plots for ^{90}Sr demonstrate the only significant difference between background sediments and the other cases was at Cañada Ancha, p value = 0.043. The p values for the other sites were all greater than 0.050. The ^{90}Sr median is three times the background value, suggesting a LANL impact. The relatively large range of strontium values, -0.06 to 0.64 pCi/g, in Cañada Ancha may also reflect episodic deposition.

Americium-241



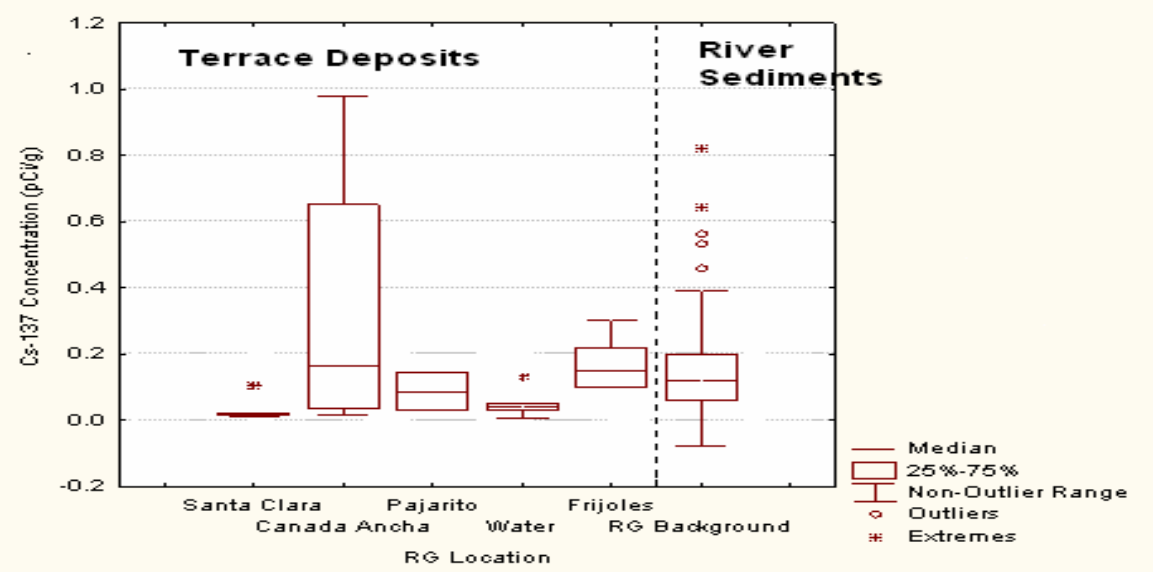
Am^{241}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles	River Sediments
Median	0.0045	0.0141	0.0078	0.0085	0.0100	0.0030
25th	0.0030	0.0018	0.0055	0.0030	0.0062	0.0020
75th	0.0080	0.0175	0.0100	0.0130	0.0184	0.0040
Min	-0.0030	-0.0049	0.0055	0.0002	0.0037	-0.0030
Max	0.0100	0.0260	0.0100	0.0180	0.0220	0.0800

Figure B18. Americium-241 box plots for background reference river sediments compared to terrace deposits

The box plots for ^{241}Am demonstrate a difference in measurements between the background sediments and at the Frijoles site (U-test p = 0.022, t-test p = 06597). While the background data pool is distribution free, the Frijoles case is normally distributed. The p values for the remaining sites are greater than 0.050. Americium values are up to three times those from background. This evidence may be inconclusive but suggests a LANL impact.

A minimum value of -3.26 pCi/g was measured for background in 1987, and is unlikely. The measurement is probably a laboratory error and was not used to estimate the descriptive statistics.

Cesium-137



Cs^{137}	Santa Clara	Canada Ancha	Pajarito	Water	Frijoles	River Sediments
Median	0.036	0.185	0.102	0.080	0.150	0.120
25th	0.033	0.081	0.059	0.064	0.101	0.060
75th	0.041	0.650	0.144	0.120	0.220	0.200
Min	0.027	0.028	0.059	0.052	0.098	-0.080
Max	0.102	0.980	0.144	0.132	0.301	0.820

Figure B19. Cesium-137 box plots for background reference river sediments compared to terrace deposits

The box plots demonstrate significant differences in ^{137}Cs measurements between background conditions and Santa Clara ($p = 0.0048$). The smaller values at Santa Clara than background suggest global fallout influence is less below the surface there.

The box plots identify a potential difference between ^{137}Cs measurements at the Cañada Ancha site (t-test $p = 0.003$, U-test $p = 0.5556$) and LANL background measurements. The ^{137}Cs distributions were nonparametric in the background as well as the Cañada Ancha site. The Man-Whitney U-test did not identify a significant difference while the parametric t-test suggested there is one. The difference in the Cañada Ancha data (median is 1.5 times greater) and the relatively large range of measurements suggests that a LANL impact exists but does not provide conclusive evidence.

Summary

An evaluation of radionuclide measurements in sediments collected from terrace strata along the Rio Grande below Los Alamos Canyon, indicate $^{239/240}$Pu, ^{137}Cs, ^{90}Sr, and ^{241}Am are statistically elevated when compared to reference conditions upstream from LANL.

Sediments from abandoned channels and floodplains filled during episodic flooding in the Rio Grande were collected downstream of Los Alamos Canyon and compared to terrace deposits above Los Alamos Canyon and historic background river sediments references developed by LANL. The terrace sites included a location at Santa Clara Pueblo, 12.1 km (7.5 miles) upstream of Los Alamos Canyon, and four sites downstream. Those four locations were on terraces near Cañada Ancha, Pajarito, Water, and Frijoles canyons.

Commercial analytical laboratories analyzed these sediments for ^{238}Pu, $^{239/240}$Pu, ^{90}Sr, uranium isotopes -234, -235, and -238, ^{241}Am, ^{237}Np, and ^{137}Cs. We evaluated the data by testing the data sets for normality and then running parametric and nonparametric tests to determine what relations existed between the reference data sets and those downstream of LANL.

Based on statistical comparisons, we found that $^{239/240}$Pu was the most persistent radionuclide found in terraces downstream of LANL. By far, the largest concentrations were found at the Cañada Ancha core site followed by the Frijoles site, and then the Water Canyon site. Cesium-137 was also elevated by the greatest amount at the Cañada Ancha site, also followed by Frijoles site. Strontium-90 at the Cañada Ancha site and ^{241}Am at the Frijoles site were found to be elevated. The differences for all constituents between Pajarito and reference conditions were statistically insignificant.

Radionuclide measurements were most elevated at the Cañada Ancha site, approximately 5 km (4 miles) downstream of Los Alamos Canyon. The $^{239/240}$Pu mean was 25 times greater than at Santa Clara and seven times greater than the LANL background mean. The ^{137}Cs mean was 10 times greater than at the Santa Clara site and two times greater than the LANL background mean. The ^{90}Sr mean was 3.5 times greater than the Santa Clara site. The ^{90}Sr mean for LANL's background mean was negative and 0.28 pCi/g at the Cañada Ancha site.

We expected the contaminant concentrations and differences to diminish downstream with distance; instead we found the next greatest levels were present in the farthest downstream location, 19 km (12.5 miles) downstream of Los Alamos Canyon at Frijoles Canyon. The $^{239/240}$Pu mean was 14 times greater than at the Santa Clara site and four times greater than the LANL background mean. The ^{137}Cs mean was greater than at Santa Clara, and ^{241}Am measurements were significantly different and greater than the LANL background.

The Pajarito site contained the smallest level of contaminants, at levels similar to the regional reference levels that describe global fallout. This site is 11 km (7 miles) downstream of Los Alamos Canyon and only 7 km (3 miles) downstream of the Cañada Ancha site. The Water Canyon site, 14 km (8.5 miles) downstream of Los Alamos Canyon, exhibited $^{239/240}$Pu measurements five times greater than the Santa Clara site.

This phenomenon may be due to the Cochiti Reservoir closure in 1973. The lake waters backed up to just below or at Pajarito Canyon. Grain size distribution analysis, discussed

in the main body of this report, indicates the lacustrine sediment deposits contained a higher proportion of fines as distance increased downstream. Larger proportions of fine particles in sediments tend to increase contaminant concentrations relative to samples otherwise similar.

APPENDIX C. Thermal Ionization Mass Spectroscopy Methods Description

Introduction

This section identifies the origin of plutonium in terrace deposits along the Rio Grande. We collected sediments at varying depths from cores and outcrops along the Rio Grande and used both, Alpha Spectroscopy and Thermal Ionization Mass Spectroscopy to measure the concentration of $Pu^{239/240}$ and identify the origin of plutonium in the sediments. Thermal Ionization Mass Spectroscopy analytical methods are helpful in distinguishing between LANL and global fallout plutonium sources using isotopic ratios. Statistical reference levels, derived from Alpha Spectroscopy measurements, have long been used to identify potential LANL impacts.

We collected sediment samples from cores and outcrops at five sites along the Rio Grande to determine plutonium distribution and to identify the origin. The site above LANL is 12.3 km (7.7 miles) upstream of the Los Alamos Canyon and Rio Grande confluence at the Santa Clara Pueblo, a site we believed to be free of LANL impacts. Four sites downstream of Los Alamos Canyon were sampled to investigate the potential impact from the Laboratory in White Rock Canyon. These sites include terraces along the Rio Grande at Cañada Ancha, Pajarito, Water, and Frijoles canyons. Los Alamos Canyon is the major source of legacy plutonium in Rio Grande sediments. It contains both Acid and DP canyons that received radioactive wastewater discharges during 1943 to 1963, the early days of Laboratory operations.

We determined LANL impacts by comparing plutonium concentration as well as isotope ratio data from samples in White Rock Canyon to data from the Santa Clara site and reference data derived from the LANL environmental surveillance program. We also used the data and methods described in the LANL report "*Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley*" (Gallaher and Efurd, 2002) to reach the conclusions described in this section.

We found that there are measurable concentrations of LANL plutonium buried in terraces along White Rock Canyon, and that the distribution is not uniform. We hypothesized that as distance from the Laboratory plutonium sources increased, the concentrations and proportion of LANL derived plutonium in contaminated sediments would decrease. We also hypothesized that younger sediments, deposited after 1943, contained diminishing concentrations and smaller proportions of LANL derived plutonium.

The greatest LANL influence was at a Cañada Ancha, closest to Los Alamos Canyon. These influences were seen buried at depths reflecting episodic flood deposition that occurred during the 1950's and 1960's. We also found that LANL impacts occurred further down stream, in Frijoles Canyon. Lacustrine sediments containing LANL plutonium were deposited in Frijoles Canyon during a period in the mid 1980's when the Cochiti Reservoir was filled to a level greater than the present.

We also suspect the LANL component of plutonium in the Cochiti Reservoir is currently increasing due to changes at Los Alamos resulting from the Cerro Grande fire. Increased

magnitudes and frequencies of storm water flows have occurred in Pueblo and Los Alamos canyons since the fire (Englert and Ford-Schmid, 2004).

We also found that while the alpha spectroscopy method is useful in characterizing plutonium concentrations, potential LANL impacts, and health and safety concerns, Thermal Ionization Mass Spectroscopy is a much better tool for identifying the sources of plutonium in the environment.

Analytical Methods

The Thermal Ionization Mass Spectroscopy (TIMS) procedure allows for the quantification of the isotopic composition of the plutonium in environmental samples by measuring the relative abundance of atoms for the isotopes Plutonium 240 and Plutonium 239. Determination of the Plutonium 240:239 atom ratio can be used to distinguish the components of global fallout and Laboratory plutonium, and quantify the mixture. Mass spectrums are obtained by converting a small amount of a purified sample into rapidly moving ions and resolving them on the basis of their mass to charge ratio. A thermal ionization mass spectrometer produces ions by heating a chemically separated and purified aliquot of a sample. The ions are resolved into discrete spectral peaks, (their mass to charge ratio) by mass analyzers, and quantified by measuring the peak heights and comparing the spectra with reference standards. The procedures for TIMS analysis of plutonium were developed by the Los Alamos Clean Chemistry and Mass Spectroscopy Laboratory and are described in detail by Efurd and others (1993) and Gallaher and Efurd (2002).

The LANL environmental surveillance program has used alpha spectroscopy to measure plutonium and determine potential Laboratory impacts since 1974. The Laboratory developed a statistical value from multiple samples during the following years to represent a reference level for the highest likely background measurement. Alpha spectroscopy measurements of plutonium are obtained after a sediment sample is digested, chemically separated, purified, and fixed onto a planchet. Alpha spectroscopy measures the number of alpha particles emitted by radionuclide isotopes in environmental samples. Radionuclide isotopes emit alpha particles at discrete energy groups and analytical laboratories use these energy levels to identify individual isotopes. Measuring the rate of the alpha interactions quantifies the concentration in a sample. Although alpha spectroscopy is the primary method for measuring plutonium, the resolution achieved by this method is not adequate to resolve between plutonium 239 and 240 isotopes. Combined plutonium 239 and plutonium 240 alpha emissions are measured at approximately 5,155 keV (Plutonium 239 occurs at 5,105 keV and Plutonium 240 occurs at 5,123 keV).

Reference Values

In 2002, LANL released a report "*Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley*" by Bruce Gallaher and others (2002) who used the Thermal Ionization Mass Spectroscopy analytical methods to identify the origin of plutonium in northern New Mexico Rio Grande sediments. The study extended from 1991 through 1998 and used isotopic ratio techniques to fingerprint

plutonium sources. Over 100 samples from the Rio Grande watershed were measured by the TIMS method, and used to identify LANL derived plutonium. Results of these measurements were also used to resolve reference values for global fallout in northern New Mexico.

Gallaher found that the plutonium 240:239 isotope ratio for global fallout in northern New Mexico ranges between 0.13 and 0.21 at a 99.7% confidence level. He demonstrated that plutonium measurements by TIMS having 240:239 ratios greater than 0.16 reflect background conditions and values less than 0.13 indicate a probable LANL contribution of plutonium. Values between 0.16 and 0.13 suggest a possible LANL input or some mixture from LANL and global fallout plutonium. These isotope ratio ranges that identify plutonium sources in northern New Mexico are compiled in Table C1.

Table C1. Plutonium 240:239 Atom Ratio Ranges Used to Identify Potential Sources

Pu 240 : Pu 239 atom ratio	Likely Plutonium Source
0.16 - 0.21	Global fallout
0.13 - 0.16	LANL influence possible and/or mixture
Less than 0.13	LANL influence likely and/or mixture

Plutonium Concentration References

Also in 2002, LANL revised the background reference concentration they use for $Pu^{239/240}$. Other radionuclide and chemical reference values derived by their program were revised, but only $Pu^{239/240}$ is discussed here. A background reference concentration, referred to as the upper tolerance level or UTL, is a statistical value reflecting the highest probable concentration that would be measured in a background population. LANL published this revision in a report, "*Background Radioactivity in River and Reservoir Sediments near Los Alamos, New Mexico*", by S. McLinn and D. Lyons (2002). The $Pu^{239/240}$ UTL was based on measurements made from 1974 to 1997 for Rio Grande sediments from background sites using alpha spectroscopy analytical methods. Before the revision, the reference statistic was calculated by adding the mean and two standard deviations of the background sediment population that included a blended group of reservoir and river populations.

The revisions were based on a re-evaluation of the background population and an alternate descriptive statistic. LANL excluded sample locations downstream of their facility as well as developing references for both river and reservoir sediments. They also chose to use the 0.95 percentile statistic (0.95, 0.95) rather than the upper tolerance level previously described, the mean plus two times the standard deviation of the historical background population. These reference values are now referred to as the Upper Limits for Background, or BGULs.

Reference values describing $Pu^{239/240}$ concentrations and isotope ratios in northern New Mexico river and lake sediments are compiled in Table C2. LANL reported a $Pu^{239/240}$ upper limit for background river sediments as 0.013 pCi/g and a background value for

reservoir sediments as 0.020 pCi/g. The 95th percentile statistic we derived for data reported in Gallaher's 2002 report was supportive of the BGULs reported by McLin.

Table C2. Plutonium$^{239/240}$ concentration and 240:239 isotope ratios for river and reservoir sediments in northern New Mexico

Data from McLin Report 2002	Plutonium 239/240 (Mean) (pCi/g)	Plutonium 239/240 (95th Percentile) (pCi/g)	Plutonium 240:239 Atom Ratio (Mean) (Unitless)
River Sediments	0.003	0.013	-
Reservoir Sediments	0.008	0.020	-
Data From Gallaher Report 2002			
Reference Reservoirs	0.011	0.022	0.166
Chama Reservoirs	0.006	0.009	0.170
Upper Rio Grande Reservoirs	0.017	0.022	0.162
NMED Data			
Terraces downstream of LANL	0.018	0.040	0.111

Reference Reservoirs include Lowe, Rio Grande, Abiqui, and Heron reservoirs
Chama River Reservoirs include Abiqui and Heron Reservoirs
Upper Rio Grande Reservoirs include Lowe and Rio Grande Reservoirs

For this table, we recompiled data from Gallaher's report to reflect background populations similar to that reported by McLin. The population describing the Upstream River Sediments reference value was compiled from samples collected on the upper Rio Grande, the Jemez River, the Chama River, as well as samples from Frijoles Canyon at the Bandelier Monument headquarters, and from the Rio Grande at Pena Blanca. The measurements reflecting the Upstream Reservoir reference value are from Lowe and Rio Grande Reservoirs on the upper Rio Grande, and Heron and Abiqui Reservoirs on the Chama River. In addition, we computed separate references for reservoirs on the Chama River and the upper Rio Grande.

The River Sediment background values computed from Alpha Spectroscopy in McLin's report and the TIMS methods in Gallaher's report are similar, the means are both 0.003 pCi/g and the 95th percentile statistics are 0.013 pCi/g and 0.008 pCi/g for Alpha and TIMS methods, respectively. The Reservoir values are also similar; the means are equal to 0.008 pCi/g and 0.011 pCi/g, establishing 0.020 pCi/g and 0.022 pCi/g upper limits (0.95, 0.95) by Alpha and TIMS methods, respectively. The Regional value derived for the reservoirs on the upper Rio Grande is over two times greater than found for reservoirs on the Chama River. They are 0.022 pCi/g compared to 0.009 pCi/g, respectively. This reflects the global fallout variability associated with differences in latitude, elevation, and climate.

We compared individual measurements on sediments from terraces along the Rio Grande to these reference values to reach the conclusions made in this report. The average $Pu^{239/240}$ concentration we found in terrace sediments downstream of the Los Alamos Canyon and Rio Grande confluence is 0.018 pCi/g, and the 95th percentile of this group is 0.040 pCi/g. The average plutonium 240:239 isotope ratio value is 0.111.

Sample Locations

We collected 10 sediment samples from 10 horizons in cores and outcrops at 5 sites along the Rio Grande, upstream of LANL and in White Rock Canyon downstream of LANL, to determine plutonium distribution and to identify the origin. The sites are located at Santa Clara Pueblo, and in or near canyons Cañada Ancha, Pajarito, Water, and Frijoles. The Santa Clara site is 12.1 river kilometers (7.5 miles) upstream of the Otowi Bridge. The bridge is at the confluence of Los Alamos Canyon and the Rio Grande and will be a reference location for this report. The Santa Clara site is in an area not affected by the closure of the Cochiti Dam and not impacted by potential inputs of plutonium from LANL. Sandy loam sediments were collected from a 5 to 31 cm horizon representing an abandoned floodplain, active from 1941 to 1968.

The remaining sites are downstream of Los Alamos Canyon, in White Rock Canyon, and were active fluvial depositional sites after 1943. LANL discharged untreated and treated radioactive industrial wastewater into Acid Canyon from 1943 to 1963. Treated radioactive wastewater was discharged into DP Canyon from the LANL plutonium processing facility at TA-21 from 1952 to 1986. Acid and DP canyons are tributaries in the Los Alamos watershed and the main source of legacy contaminants being transported to the Rio Grande. See Appendix E for additional discussion of historical and current contaminant transport from the Los Alamos Canyon system.

The Cañada Ancha site is 5.1 km (3.2 miles) downstream of Los Alamos Canyon, and a primary site for accumulation of LANL contaminants. It was an abandoned channel, or slough, except during floods. During 1940 through 1958 it gradually filled with sediment, and after a 1967 flood, it was completely filled with sediment. Sandy loam sediments in the upper meter, grading to a finer textural silty clay loam with depth, were collected from 4 horizons to a 2.3 m depth. Possibly, a sand horizon representing the original channel bottom was found between 2.3 m and 3.4 m.

The Pajarito and Water canyon sites are 10.9 km (6.8 miles) and 14.0 km (8.7 miles) downstream of Los Alamos Canyon. Sediment samples are texturally coarser than those at Cañada Ancha and Frijoles, sand at Pajarito Canyon and sand to loamy sand at Water Canyon. We collected sediments from a single horizon in a 40 cm core drilled into the Pajarito terrace, and two horizons in 1 m of an outcrop in Water Canyon. The sediments appear to reflect a bank-channel interface, reflecting a lateral movement of the Rio Grande channel or deposits developing from progressing slack water conditions as the Cochiti Reservoir stage changed.

The Frijoles Canyon site is 19.3 km (12 miles) downstream of Los Alamos Canyon and 20.7 km (12.9 miles) upstream of the Cochiti Dam, although affects of the dam are evident here. Multiple stream channels, mud flats, and high water remnants are evident; increasing as downstream distance increases, and demonstrate that this area was inundated up to 15 m by the maximum stage height of Cochiti reservoir. By 1988 the lake level declined and abandoned this area. Two silt loam sediment samples were collected from two horizons in a 1.8 m core. We expected lower concentrations of

plutonium at this site due to greater distance from the Los Alamos confluence, and were uncertain of whether the source might be LANL or global fallout.

Results

We collected 10 sediment samples from terrace deposits along the Rio Grande. Sample duplicates were submitted to both the Los Alamos Clean Chemistry and Mass Spectrometry Laboratory, and Paragon Inc., a commercial analytical laboratory. The Los Alamos Clean Chemistry and Mass Spectrometry Laboratory measured the samples by TIMS, and Paragon measured the samples using alpha spectroscopy methods. The activities per gram measurements for both plutonium 240 and 239 isotopes were derived from the atom measurements, acquired by the TIMS methods, and the specific activity for each isotope. The plutonium isotope ratios are based on the relative abundance of atoms for each isotope. The combined 239 and 240 activities, derived from the TIMS analysis, as well as the alpha spectroscopy activities were reported for each sample.

Table C3 summarizes the TIMS and alpha spectroscopy measurements for $Pu^{239/240}$ in the sediments we collected along the Rio Grande. The first column includes a general description of the site as well as depths from which the samples were collected. The following two columns are the activity measurements of the plutonium 239 and 240 isotopes. They are derived from TIMS measurements of atoms of plutonium per gram in each sample multiplied by the specific activity for the appropriate isotope. The fourth column is the sum of the activities derived from the two isotopes. The fifth column is the ratio values of plutonium 240 to plutonium 239 isotopes measured by TIMS. The sixth column contains the measurements made by alpha spectroscopy (Paragon Analytics, Inc.). Although the TIMS and Alpha Spectroscopy measurements are similar, the differences demonstrate the data variability. The variability includes random and systematic error associated with the sampling and analytical procedures. The Duplicate Error Ratio, listed in the last column, is a measure of the similarities between the TIMS and Alpha Spectroscopy measurement of plutonium concentration in the samples.

Table C3. Plutonium$^{239/240}$ measurements along the Rio Grande using both Alpha Spectroscopy and Thermal Ionization Mass Spectroscopy methods

	Thermal Ionization Mass Spectroscopy				Alpha Spectroscopy	Duplicate Error Ratio
Station Description	^{239}Pu Activity (pCi/g)	^{240}Pu Activity (pCi/g)	$^{239}Pu + {}^{240}Pu$ Activity (pCi/g)	$^{240}Pu : {}^{239}Pu$ Atom Ratio	$^{239/240}Pu$ (pCi/g)	DER
Rio Grande below Santa Clara						
abandoned flood plain, active 1941-1968						
5 to 31 cm	0.0027	0.0016	0.0043	0.16	0.005	0.25
Rio Grande above Canada Ancha						
flood plain, overflow area flooded in 1958 or 1967 flood						
0 to 31 cm	0.0004	0.0003	0.0007	0.21	0.002	0.75
101 to 110 cm	0.0172	0.0059	0.0231	0.09	0.067	3.65
162 to 186 cm	0.0451	0.0157	0.0608	0.09	0.066	0.44
213 to 229 cm	0.0397	0.0023	0.0420	0.02	0.034	1.52
Rio Grande above Pajarito Canyon						
active flood plain and bar surface						
31 to 46 cm	0.0016	0.0007	0.0023	0.12	0.008	0.95
Rio Grande below Water Canyon						
active flood plain and bar surface						
0 to 31 cm	0.0021	0.0008	0.0029	0.11	0.003	0.07
91 to 101 cm	0.0026	0.0014	0.0040	0.15	0.008	0.76
Rio Grande above Frijoles						
pre-1950 flood-plain deposits and reservoir sedimentation						
31 to 61 cm	0.0106	0.0032	0.0138	0.08	0.018	0.49
122 to 152 cm	0.0057	0.0027	0.0084	0.13	0.016	1.07

Evaluation of duplicate measurements is part of the quality control process used in determining accuracy and precision of environmental data. In this case, we divided a single homogeneous sample into two sub-samples, submitted them to different laboratories, and had the samples analyzed using different analytical techniques. These sub-samples are commonly referred to as split samples. Duplicate Error Ratio, or DER, is the common calculation made to measure repeatability, the accuracy and precision of analytical techniques. The results, as well as the measurement uncertainty, are evaluated to determine the validity of the duplicate measurements. The DER is defined below:

$$DER = \left(\frac{|S - D|}{2 * \sqrt{\sigma_S^2 + \sigma_D^2}} \right)$$

Where:
$|S - D|$ = the absolute value of the difference between a sample measurement (S) and a duplicate measurement (D).
σ_S^2 = the square of the total propagated uncertainty (1 sigma) of the sample measurement
σ_D^2 = the square of the total propagated uncertainty (1 sigma) of the duplicate measurement

The DER gives the degree to which the sample and duplicate measurements are comparable. A DER less than or equal to 1.42 indicates the results are statistically equivalent to a 95% confidence level. A DER greater than 1.42 and less than 2.13 places the results in a warning range, and greater than 2.13 places the results outside a 3σ uncertainty, or 99.7% confidence level.

We found that all but one DER evaluations were less than 2.13. One DER for a measurement at Cañada Ancha was 3.65; outside the acceptable 2.13 duplicate range. Plutonium measurements for the 101 cm horizon at Cañada Ancha were 0.0231 pCi/g, by TIMS, compared to 0.067 pCi/g, by alpha spectroscopy. Although the alpha measurement was almost three times the TIMS measurement, the difference did not change the conclusions made from these measurements. For example, both values are well above the background reference, 0.013 pCi/g.

While comparisons to the River BGUL suggest that LANL impacts exists only at Cañada Ancha and Frijoles canyons, almost all of the plutonium isotope ratios indicate that there is some component of LANL derived plutonium at all sites. Most, but not all of the $Pu^{239/240}$ concentrations for sediments at Cañada Ancha and Frijoles are greater than 0.013 pCi/g BGUL, and all but one of the plutonium 240:239 isotope ratios obtained for this report are less than 0.16, ranging from 0.08 to 0.21. Recall that, plutonium concentrations greater than 0.013 pCi/g or 240:239 isotope ratios less than 0.16 suggests a LANL impact.

The plutonium activity measurements for Santa Clara, above Los Alamos Canyon, as well as for Pajarito and Water canyons, below Los Alamos Canyon, suggest that LANL plutonium is not associated with those sites. These values are less than the 0.013 pCi/g BGUL sediment reference value. Yet the plutonium isotope ratios derived from the TIMS methods indicate there is a LANL influence at Pajarito and Water canyons. The ratios are between 0.11 and 0.15. Although less than 0.16, the values are larger than measured for samples from Cañada Ancha and Frijoles. This suggests a smaller content of LANL plutonium exists in Pajarito and Water canyons than the other sites, particularly in Cañada Ancha.

Except for the surface sample, the values at Cañada Ancha indicate a LANL input of plutonium at this location. Both the TIMS (0.0007 pCi/g) and alpha spectroscopy (0.002 pCi/g) plutonium measurements for the surface sample, are less than 0.013 pCi/g. It also has a large isotope ratio, 0.21, indicating global fallout plutonium. These values clearly indicate a lack of LANL plutonium. The samples at depth are a different story. The activity measurements are 2 to 5 times background, and the small isotope ratios, ranging from 0.02 to 0.09, demonstrate a large component of LANL plutonium.

At the Frijoles site, the interpretation is not as clear. Most of the plutonium measurements do suggest a LANL influence, but are near the reference level. The activity values are only slightly above the upper tolerance level for plutonium in sediments, 0.0084 pCi/g to 0.018 pCi/g. Although the isotope ratio value, 0.08, is a fairly clear indicator of a LANL impact, the second value of 0.13 only suggests a LANL impact is possible.

Based on the isotope ratios, proportions or percentages of LANL derived plutonium can be calculated. We calculated the proportion of the LANL plutonium mixed with the plutonium from global fallout using the following equation. This equation is a modified form of a method originally described by Hardy and others (1972) from Gallaher and others (2002). The LANL proportion of plutonium in our samples collected along the Rio Grande are compiled in Table C4 and shown in Figure C1.

$$\frac{[PuActivity]_L}{[PuActivity]_F} = \frac{(R_F - R_S)(1 + 3.67R_L)}{(R_S - R_L)(1 + 3.67R_F)}$$

where:

(*PuActivity*) = plutonium activity in LANL component,
(*PuActivity*) = plutonium activity in global fallout component,
R_S = Pu 240:239 atom ratio measured in the sediment sample,
R_L = 0.015, the Pu 240:239 atom ratio of plutonium released by LANL,
R_F = 0.169, the Pu 240:239 atom ratio of global fallout in northern New Mexico, and
3.67 = ratio of half-lives of Plutonium 239 to Plutonium 240.

Table C4. Proportion of LANL plutonium in sediments collected along the Rio Grande

Station Description	(R_s) ^{240}Pu : ^{239}Pu Atom Ratio	LANL Component %
Santa Clara		
5 - 31 cm (hand augured)	0.16	5
Canada Ancha		
0 - 31 cm (hand augured)	0.21	0
101 -110 cm (hand augured)	0.09	39
162 - 186 cm (hand augured)	0.09	38
213 - 229 cm (hand augured)	0.02	99
Pajarito		
31 - 46 cm (hand augured)	0.12	24
Water		
0 - 31 cm (outcrop A)	0.11	29
91 - 101 cm (outcrop A)	0.15	8
Frijoles		
31 - 61 cm (hand augured)	0.08	47
122 - 152 (hand augured)	0.13	18

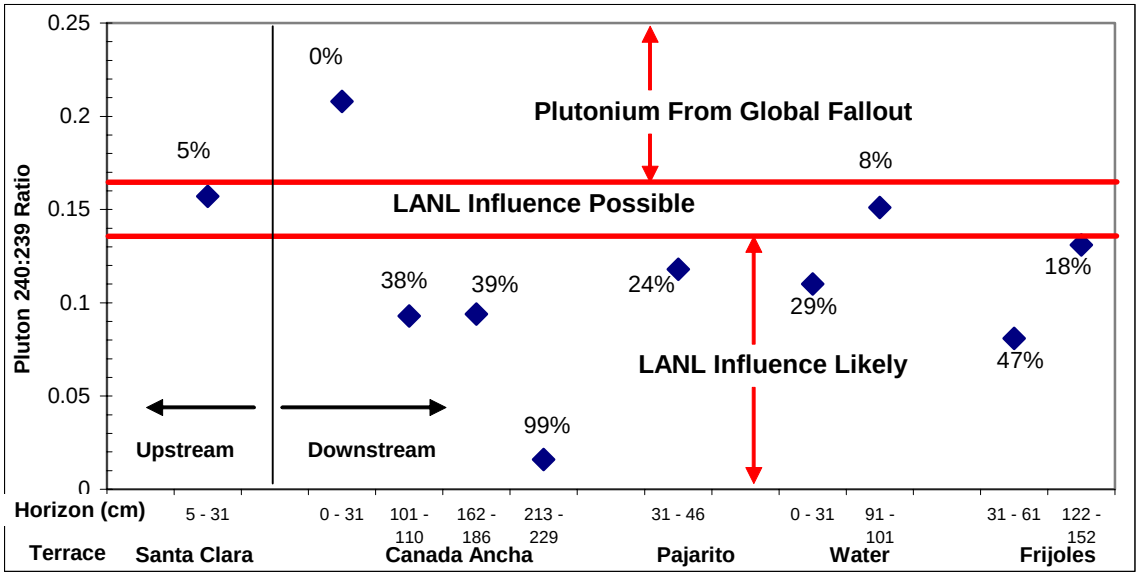


Figure C1. Proportion of LANL plutonium and the 240:239 isotopic ratios for samples collected along the Rio Grande

We found the average fraction of LANL plutonium in White Rock Canyon sediments was 34%, ranging as high as 99% and as low as 0%. The greatest LANL influence occurs at the Cañada Ancha site 5.1 km (3.2 miles) downstream of Los Alamos Canyon and at depths reflecting episodic deposition during 1950's and 1960's floods. The

Frijoles site most downstream appeared to be affected to a lesser degree. The plutonium in two sediment samples appear to be derived from LANL, they contain 18% and 47% LANL plutonium. Based on the silty loam sediment texture, we suggest they were deposited while the Cochiti reservoir was filled above these horizons. Although plutonium concentrations at the Pajarito and Water canyon sites were measured below the BGUL, the isotopic ratios suggest that from 8% to 29% of the plutonium in sediments are LANL derived.

While the proportions of 0% LANL derived plutonium at the Cañada Ancha surface horizon, and a 5% component of LANL plutonium at Santa Clara were unanticipated. It's possible that the surface at Cañada Ancha is comprised of recent sediments from the upper Rio Grande containing only global fallout plutonium, while the Santa Clara surface contains atmospheric impacts from LANL. We find it more probable that at these low levels the analytical and sampling uncertainties may lead to incorrect conclusions. The remaining samples at Cañada Ancha, Pajarito, Water, and Frijoles show more definite LANL influences, containing from 18% to 99% LANL derived plutonium.

The following figures, Figure C2 and Figure C3, show the relationships between the isotopic ratios and the plutonium concentrations obtained for the samples reviewed in this report, and the reference levels that define the origin of plutonium. Figure C2 demonstrates these relationships for our samples collected along the Rio Grande. Figure C3 includes the data from Galaher's 2002 report. Both figures generally demonstrate that, samples containing plutonium concentrations less than the background reference also include isotope ratios that express global fallout plutonium. These figures also show that samples containing plutonium concentrations greater than the background reference include isotope ratios that express an existing component of LANL. An important observation that should be recognized is that not all samples containing plutonium concentrations below the reference value, for example the LANL BGUL or UTL, are free of LANL plutonium.

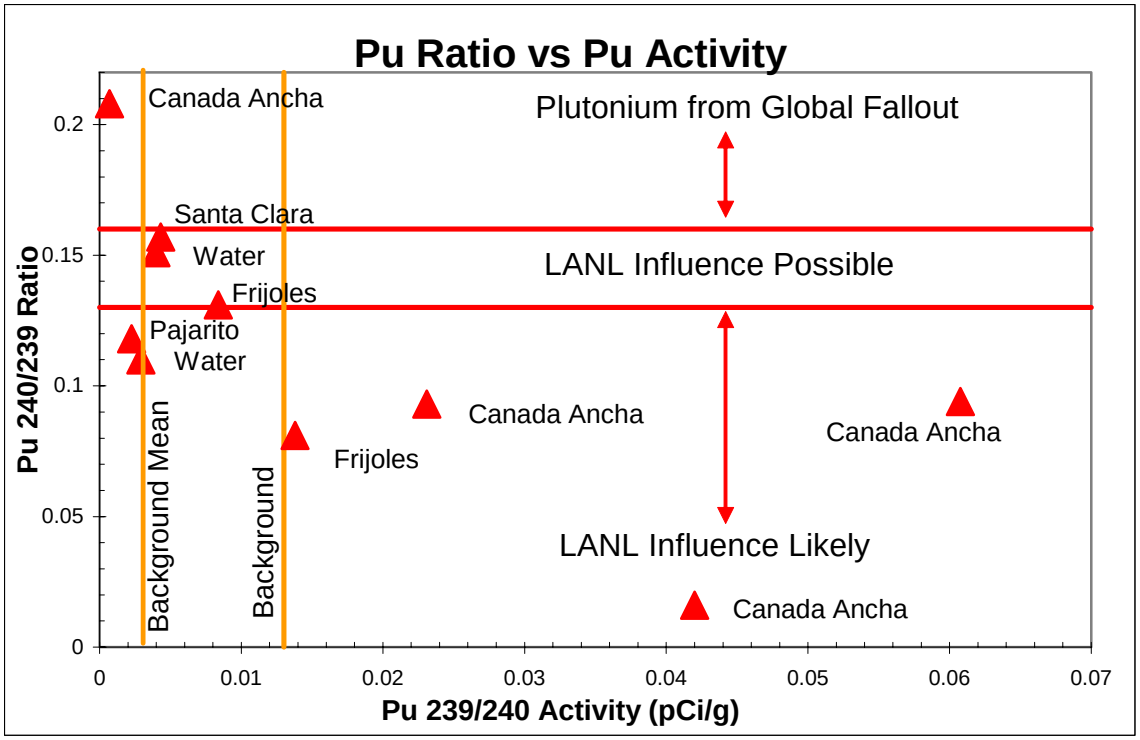


Figure C2. Plutonium 240:239 isotope ratios and $Pu^{239/240}$ concentrations measured in sediments along the Rio Grande, and the relation between them and the background references for river sediments.

For example, the Cañada Ancha sample, in the upper left corner of the chart, is within the area outlined for global fallout. This area is defined by isotope values greater than 0.16 and plutonium concentrations below the 0.013 pCi/g BGUL. This Cañada Ancha sample was collected at the surface and the concentration and isotope values clearly demonstrate that plutonium in this sample is from atmospheric fallout. Other samples from Cañada Ancha and one from Frijoles, seen in the lower right section of the chart, also clearly define the plutonium source. All four contain concentration values that exceed the BGUL and have isotope ratios that demonstrate high percentages of LANL plutonium. Those isotope ratios are less than 0.13. Samples that are not as clearly defined include two samples from Water, a Frijoles sample, and a Pajarito sample. They contain plutonium concentrations less than the background reference, yet the isotope ratios are less than 0.16, which identifies the plutonium source as possibly originating from LANL.

The Santa Clara sample, located in the upper left portion of the chart, contains a plutonium concentration below the BGUL, yet the isotope ratio identifies the samples as containing a possible LANL influence. The likelihood of a LANL impact at Santa Clara is small, but additional investigations to clearly define the extent of LANL impacts here are outside the scope of this report.

Figure C3 demonstrates the relationships between our samples and those collected by LANL and reported in Gallaher's report "Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley" (2002). LANL

collected over 100 samples within the Rio Grande watershed and around the Laboratory to identify the origin of plutonium in northern New Mexico. This chart is from the 2002 report although adjustments have been made to clarify the sample populations. It also includes data for reservoir samples, and the 0.0201 pCi/g reservoir BGUL is represented in the chart rather than the 0.013 pCi/g river background reference.

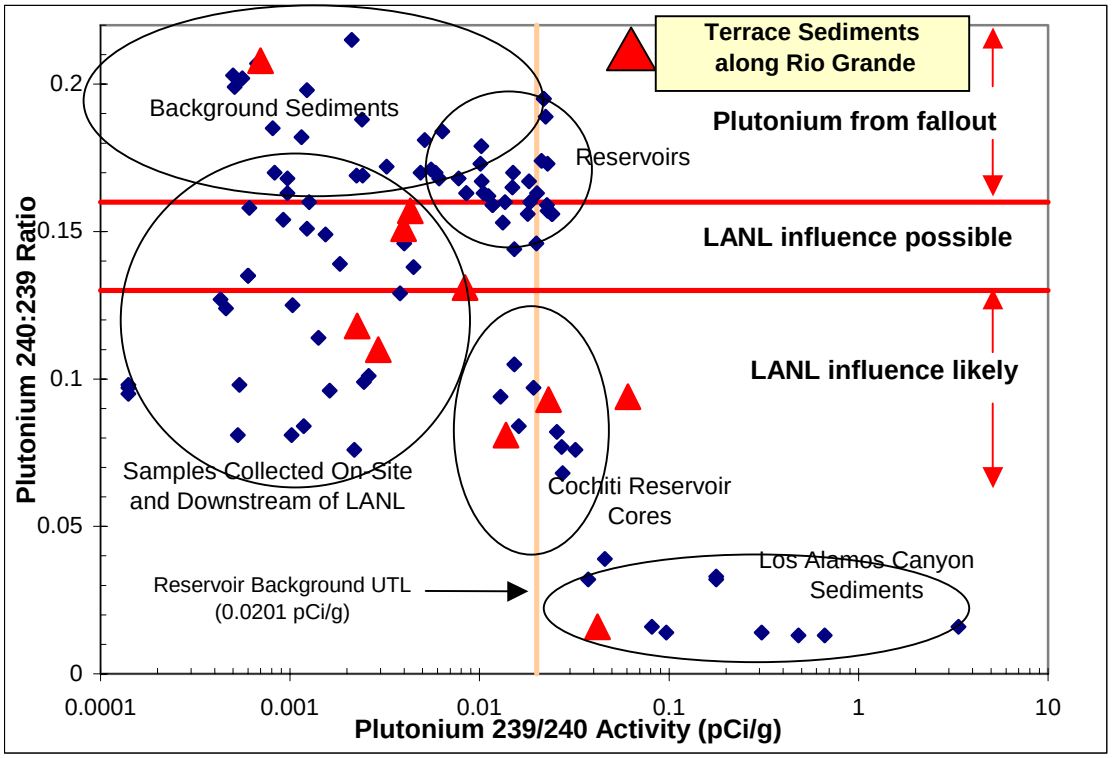


Figure C3. Plutonium ratio and concentration measurements in sediments along the Rio Grande from this report, as well as those made by LANL in Gallaher's report "Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley" (2002)

Sample populations are circled and labeled according to a general reference location, for example, Background Sediments, Reservoirs, Cochiti Reservoir Cores, Los Alamos Canyon Sediments, and Samples Collected On-Site and Downstream of LANL. "Background Sediment" values demonstrate data for samples from river reference sites, or locations distant from the Laboratory that demonstrate LANL impacts are unlikely. These sites include sediments from the Chama River, and the Rio Grande just above and below the confluence with the Chama River. The background reference also includes samples from the Jemez River, Frijoles Canyon at the Bandelier Monument headquarters, and on the Rio Grande below Cochiti Reservoir at Pena Blanca. Values for these samples are less than the background reference level for river sediments, 0.013 pCi/g $Pu^{239/240}$ concentration, and plutonium 240 to 239 isotope ratios greater than the 0.16. All measurements indicate the plutonium is from global fallout. This area is in the upper left portion of the chart.

Values within the group identified as "Los Alamos Canyon Sediments" include measurements for sediments collected from Pueblo Canyon below Acid Canyon, and from Los Alamos Canyon below DP Canyon. Both Acid Canyon and DP Canyon contained radioactive wastewater outfalls and are the major source of plutonium in Rio Grande sediments. Sediments from these areas contain plutonium concentrations greater than the River BGUL, 0.013 pCi/g, and have isotope ratios less than 0.13. This area is delineated in the lower right portion of the chart and clearly describes LANL influence. The remaining groups fall between this area and that delineated as "Background Sediments".

The values within the "Reservoir" group are from 6 reservoirs. They include the Heron and Abiqui reservoirs on the Chama River, the Lowe and Rio Grande reservoirs from the farthest northern reaches of the Rio Grande, surface samples from the bottom of Cochiti Reservoir, and core samples from Elephant Butte Reservoir. These values are at, slightly above and below, the 0.0201 Reservoir BGUL. Most of these values are less than the BGUL and greater than the 0.16 definitive isotope ratio for background. From those "Reservoir" values within the area defined as having a possible LANL influence, isotope ratios less than 0.16, most are from the Cochiti reservoir and should be excluded as a background reference.

The values delineated by the "Cochiti Reservoir Core" group reflect sediments transported after 1973 when the reservoir was completed. It contains re-suspension of sediments deposited during 1950's and 1960's, a period of the greatest legacy contribution to the Rio Grande fluvial system, and mixing with sediments containing fallout contaminants. Values for core samples from the Cochiti Reservoir are delineated separately and demonstrate significant legacy content. They are greater than or at the Reservoir BGUL, and all but 1 are within the area on the chart that is delineated as having a likely LANL influence. LANL plutonium in Cochiti Reservoir has been estimated to be 40% on a depth-weight basis (Gallaher and Efurd, 2002.).

The largest group of values is delineated by the group "Samples Collected On-Site and Downstream of LANL". This group includes measurements of samples collected in canyons within LANL boundaries, at the perimeter, and downstream into White Rock Canyon. All of the plutonium concentrations, measured in sediments from this group are less than the BGUL. This group also includes many plutonium 240:239 isotope ratio values which show that a portion of plutonium in these sediments originate from LANL. As found in Figure C2, this also demonstrates that Laboratory plutonium may be associated with sediments containing plutonium concentrations that fall below the values established for background.

The values for samples described in this study are designated by the red triangles and are superimposed onto this chart for reference. They extend from areas on the chart delineating background to areas indicating a LANL influence. Those values, including concentrations that exhibit background conditions and isotope ratios that indicate a LANL input of plutonium, generally demonstrate that sediments transported from LANL are mixing with 'clean' sediments. We expected that as distance from the Laboratory

increases, the concentrations and proportion of LANL derived plutonium in contaminated sediments would decrease. We found other factors contribute to concentrations of legacy contaminants in downstream sediments, such as grain size, climate, and hydrologic conditions of upstream channels.

Figure C3 also demonstrates a difference in plutonium concentrations between river and reservoir sediments and the necessity for separate BGULs. The reservoir BGUL, 0.020 pCi/g, is slightly larger than the river BGUL, 0.013 pCi/g. The difference may be explained by the differences in grain size. Sediment texture in reservoirs is finer than that found on channel bottoms, and finer sediments provide a greater surface area and greater cation exchange capacity for binding plutonium to sediment particles. Therefore, plutonium concentrations found in reservoirs, originating from fallout and legacy sources, could be expected to be greater than in rivers. Obviously, contaminated sediments transported from Los Alamos Canyon contain greater plutonium concentrations regardless of texture.

The samples depicted in the Los Alamos Canyon group, from Gallaher's report, are primarily bed load material, and plutonium transport may be greater than implied here. Suspended sediments in storm flow are much finer, and potentially come from sloughing banks and re-suspended flood plain materials. Plutonium concentrations measured in Pueblo Canyon are as great as 502 pCi/g (Reneau, and others, 1998), and have been measured as much as 16 times greater in Acid Canyon. Since the Cerro Grande fire, the magnitude and frequency of storm flow and the associated plutonium transport from Pueblo Canyon has increased (Englert and Ford-Schmid, 2002).

Recent changes in the Los Alamos watershed, caused by the Cerro Grande fire, may have increased the rate of plutonium transport to levels not seen since the 1950's and 1960's. The New Mexico Environment Department estimated 87 mCi of $Pu^{239/240}$ was transported out of Pueblo Canyon during 2000 to 2002 (Englert and Ford-Schmid 2002). Our latest estimates, from 2000 through 2006, indicate 199 mCi plutonium 239/240 has been transported from Pueblo canyon (report in press). These transport rates may be underestimated due to the complexities associated with multiple surges in flood stage resulting from the random nature of rainfall and contributions of flow from adjacent canyons. They only include flows greater than 10 cfs, and do not include bedload transport. The largest reported movement of plutonium prior to this was in 1957 when 43.95 mCi was transported into the Rio Grande. The second largest movement occurred in 1968 when 21.82 mCi of plutonium was transported (Graf, 1993). Graf's estimates were based on modeled values based on bedload measurements and may have greatly underestimated the transport mass. The LANL component of plutonium in Cochiti Reservoir is expected to increase in the near future.

Plutonium Concentration Variability

The following three charts demonstrate the relative plutonium concentrations in river, reservoir, and terrace sediments discussed in this report. Figure C4 demonstrates plutonium concentrations for sediments collected in rivers, reservoirs, and terraces, as well as the river and reservoir statistical reference levels calculated by LANL. They were

further separated into subgroups representing regional or upstream locations relative to locations downstream of LANL. The terrace core sediments were collected along the Rio Grande downstream of Los Alamos Canyon.

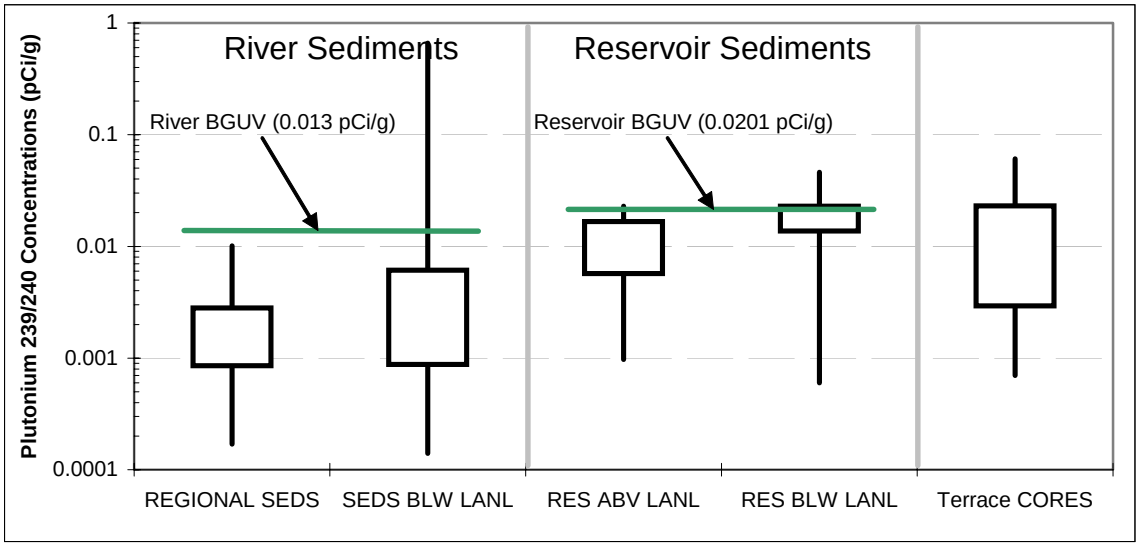


Figure C4. Relationship of plutonium concentrations in river, reservoir, and terrace sediments and LANL upper tolerance levels for river and lake sediments.

Figure C4 demonstrates that reservoir sediments contain a larger concentration of plutonium than river sediments, probably due to grain size, and that sample locations downstream of LANL also contain larger and more variable concentrations than found upstream. The terrace sample locations, subject of this report, include upstream and downstream locations as well as channel, floodplain, and lacustrine sediments.

LANL originally developed the background reference values from measurements made from 10 stations sampled in northern New Mexico from 1979 to 1997. The upper 95% confidence level or upper tolerance level (UTL), for this group was calculated by adding its mean and two standard deviations. In 2002, LANL separated their regional sediment population into river and reservoir groups, and eliminated stations that were downstream of the Laboratory. They then calculated new background reference values for river and reservoir sediments in northern New Mexico using the upper 0.95 quantile (McLinn, 2002). These background sites are generally from the same locations described in this report for regional sediments.

Between 20% and 30% of the measurements in the stations downstream of LANL, eliminated from the background population, were greater than the revised reference values, while their isotope ratios described by Gallaher (2002) and this report identify a greater occurrence of LANL plutonium.

Figure C5 represents a further breakdown of the river sediment subgroups. These subgroups include samples from background locations, from Los Alamos and Pueblo canyons, from sites along the perimeter of LANL (including three on-site samples in

Sandia Canyon and seven samples in Mortandad Canyon downstream of the LANL boundary), and from White Rock Canyon. The regional locations were sampled at the Chama River, at the Rio Grand above and below the confluence with the Chama, at the Rio Grande below Cochiti reservoir at Pena Blanca, at the Jemez River, and at Frijoles Canyon close to the Bandelier National Monument headquarters. The second subset of samples was collected in Pueblo Canyon from below Acid Canyon to Los Alamos Canyon and in Los Alamos Canyon from below DP Canyon to the Rio Grande. The perimeter subset of samples was collected in canyons along the downstream (eastern) boundary of LANL. The White Rock Canyon samples were collected along the Rio Grande downstream of Los Alamos Canyon.

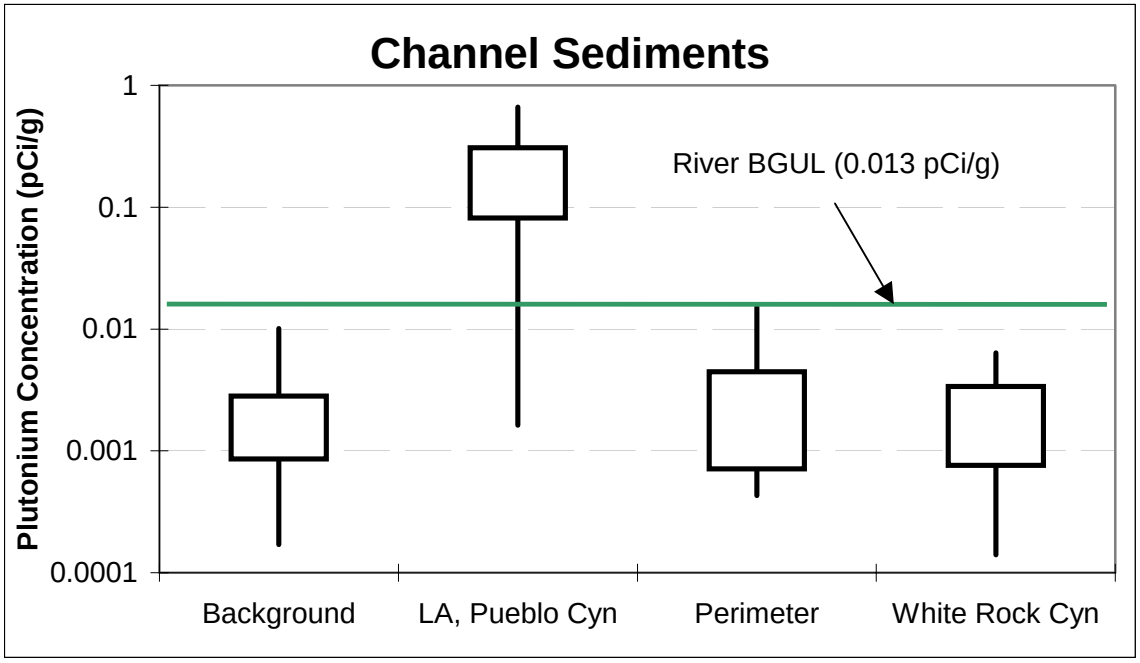


Figure C5. Plutonium concentrations measured in river sediments

None of the background stations, perimeter, or White Rock Canyon samples discussed in this report exceeded the 0.013 pCi/g BGUL. Eight of the nine measurements collected in the Los Alamos Canyon system exceeded the BGUL. The maximum value was 0.662 pCi/g, measured in upper Pueblo Canyon. This supports the conclusion that the majority of LANL plutonium was from the Los Alamos Canyon watershed. Although values for samples collected at perimeter and White Rock Canyon stations are less than the BGUL, the isotopic ratios discussed in the preceding sections indicate that there is also a LANL component of plutonium in those samples.

Figure C6 represents a further breakdown of the reservoir sediment subgroups from Figure C4. These subgroups include samples from Heron and Abiqui Reservoirs on the Chama, the Rio Grande and Lowe reservoirs on the upper Rio Grande, the Cochiti and Elephant reservoirs on the lower Rio Grande, and core samples collected within Cochiti Reservoir.

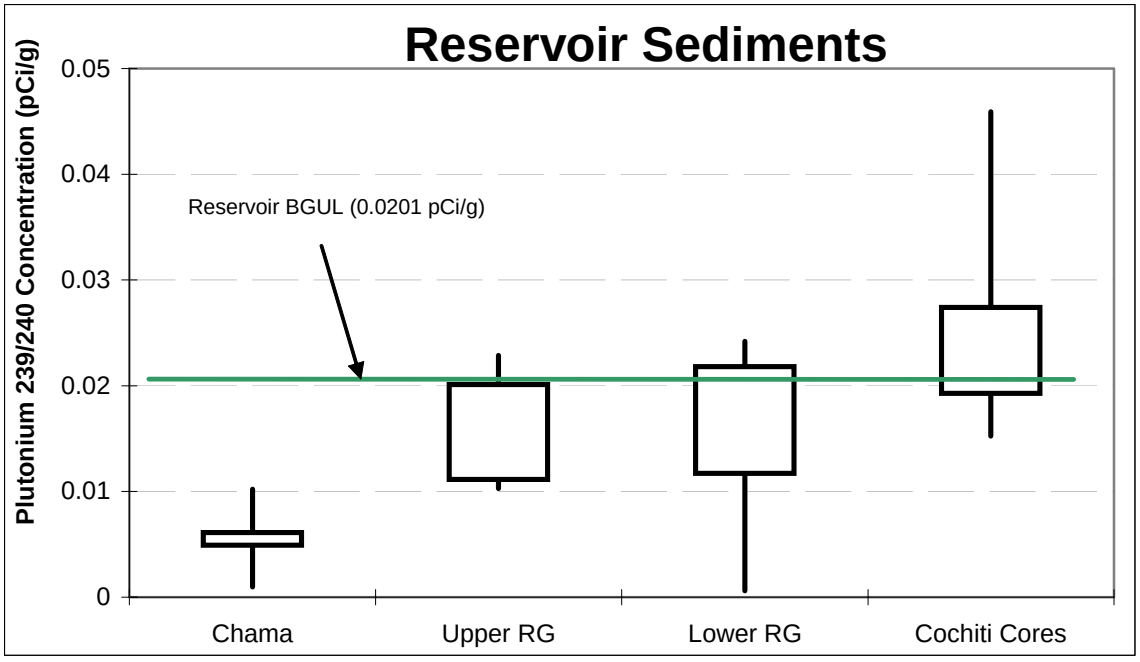


Figure C6. Plutonium concentrations measured in reservoir sediments

The Chama and upper Rio Grande subgroups reflect regional conditions, although values for reservoirs on the upper Rio Grande, averaging 0.017 pCi/g, are almost three times more than reservoirs on the Chama, which average 0.006 pCi/g. This probably reflects a difference in global fallout rates in these two regions. The upper Rio Grande samples were collected at higher latitudes, in southern Colorado, and receive sediments from watersheds below the continental divide and elevations that exceed 13,000 feet. We suggest they are in greater proximity to the Nevada Test Site wind-shed and contain greater concentrations of fallout.

A river station on the Chama, Chama at Chamita from McLin's 2002 report, supports this suggestion. The average of 26 plutonium measurements in river sediments at Chamita from 1974 to 1997 is 0.001 pCi/g, similar to the 0.007 pCi/g average from 73 samples collected in reservoirs on the Chama during the same period. The average value for reservoir data on the Chama also averages 0.006 pCi/g, from data in Gallaher and others (2002). All of these values are significantly smaller than the average plutonium value, 0.017 pCi/g, calculated for the upper Rio Grande reservoirs. We can conclude that the upper Rio Grande supplies the greatest portion of fallout plutonium and suspended sediments to the river system. The upper Rio Grande watershed is larger than the Rio Chama watershed and contributes more water as well as sediments to the reach below their confluence.

The concentration values in the lower Rio Grande subgroup suggest potential LANL impacts exist. The pooled concentrations in the lower Rio Grande and in the Cochiti Reservoir appear to be slightly larger. Most of the plutonium measurements are at the

BGUL, while seven of the 20 measurements are slightly over it. The samples were collected from Cochiti and Elephant Butte Reservoirs.

At Elephant Butte, half the values were slightly over the BGUL, yet all isotope ratios exceeded 0.016. This indicates the plutonium source is from global fallout. Meanwhile fewer measurements at Cochiti, three of 12, exceeded the BGUL, yet all of the isotope ratios indicated a LANL influence was more than likely.

The Cochiti core values for plutonium concentrations further support the likelihood of a LANL impact to the Rio Grande. Almost all of the concentration values as well as the isotope ratios indicated that a portion of the plutonium in Cochiti sediments is LANL derived.

Much of this section is a review of data and methods published in LANL reports by Gallaher and others (2002) and McLinn and others (2002) and their relationships to the measurements made for this study. The review was helpful in; 1) defining the background reference for plutonium, 2) identifying that not all measurements below reference values are free of legacy materials, 3) identifying that not all measurements above the reference values contain legacy materials, and 4) background conditions contribute a large variable related to sediment texture and origination, i.e. fine sediments generally contain relatively larger concentrations than coarser materials, and sediments contributed to the Rio Grande system from areas higher in elevation and latitude contribute larger concentrations of fallout materials.

APPENDIX D. Analytical Methods

Introduction

Analytical laboratories often use combinations of both chemical and instrument techniques to quantify low contaminant levels found in environmental samples. Analytical procedures consist of several parts assembled in laboratory SOPs for specific projects or sample types. These parts describe the chemical processes that isolate and purify, and then measure a constituent. The SOPs usually include:

laboratory sample preparation
sample dissolution
sample purification
preparation for counting
counting
data reduction

Laboratories are capable of adjusting their methodologies to achieve various detection limits. Some customers only require levels to satisfy regulatory standards. Our bureau normally request analytical methods capable of measuring contaminants below most environmental background reference levels. Detection limits of 10 to 50% of these values should be the target. Analytical methods adjusted to achieve lower detection limits often require a substantial increase in laboratory efforts.

Laboratories normally state detection limits based on ideal or optimistic situations and may not be achievable under actual measurement conditions. These levels could be considered their advertised limits. Detection limits for individual measurements are quite variable and are subject to variation between samples, instruments, and procedures. Sample size and geometry, element or compound abundance, self-absorption, and matrix interferences, instrument efficiency, ambient laboratory background, chemical recovery, and counting times contribute to these variations.

The term background above is used to describe two contexts. Environmental background references describe the highest probable levels normally measured in the environment. Ambient backgrounds are low, ubiquitous levels in the laboratory subtracted from sample measurements. Both uses are based on multiple or continuous measurements, are statistically developed, and used to establish accurate and precise evaluations of constituents in the environment.

Routine analytical methods have been issued by federal or state agencies, described by professional organizations, published in refereed journals, or developed by individual laboratories. Non-routine methods continue to be developed to address situations with unusual or problematic matrices, improve detection limits, or identify new parameters. Non-routine methods include adjustments to routine methods, and new method developments published in refereed literature.

Performance characteristics, such as detection limits, precision, and accuracy are routinely documented with each analysis. These characteristics should be evaluated as closely as the reported measured values.

The following descriptions are of the major classes of radionuclide analysis normally requested by this bureau and particle-size distribution analysis.

Alpha Emitting Radionuclides measured by alpha spectroscopy

Commercial laboratories use alpha spectrometry to accurately identify and quantify alpha-emitting radionuclides in soil, water, air filters, and other sample matrix. Alpha spectrometry can identify and quantify uranium-234, -235, and -238, thorium-228, -230, and -232, plutonium-238, -239, and -240, ^{210}Po, ^{237}Np, and ^{241}Am by counting their alpha emissions at specific energies. Each radionuclide emits alpha particles at distinct energies. A spectrum is constructed that includes the rate of emissions at those energies. The laboratory uses this spectrum to identify and quantify each analyte.

Laboratory sample preparation involves drying a sample and grinding the remaining matrix to a fine-grain, homogeneous aliquot. This aliquot could be heated to high temperatures in a furnace or treated with strong oxidizers to eliminate organic materials. A small portion of the homogenized sample is usually all that is required for the individual analysis.

Once the sample has been prepared, a 2 to 10 gram aliquot is dissolved or leached to provide a clear solution containing the radionuclide of interest. Often the sample is completely dissolved using combinations of nitric, hydrochloric, and hydrofluoric acids. In some cases, leaching with strong acids can consistently provide greater than 80% recovery of a radionuclide and may be acceptable for certain applications. Tracers of known activity and of similar chemical properties to the radionuclides of interest are added before separation. The laboratories measure the tracers to calibrate the instruments and determine the overall analyte recovery during the chemical procedures. Plutonium-242, uranium-232, and americium-243 are examples of tracers that could be used.

After dissolution, the sample is purified using a variety of chemical reactions to remove bulk chemical and radionuclide impurities. Ferric-hydroxide co-precipitation is commonly performed to remove constituents that do not form insoluble hydroxides. The precipitate containing the analytes of interest is re-dissolved and the solution is passed through multiple ion exchange columns. These columns are used to sequentially retain the analytes of interest. The columns are eluted with acid washes, re-dried, re-dissolved, and the solutions co-precipitated with lanthanum fluoride. These steps provide a chemically and radiologically pure sample having very little mass. Other examples of purification techniques include liquid-liquid extraction, distillation, and electro-deposition.

After the sample is purified, it is prepared for counting by analyte deposition onto a small planchet. Because of the limited penetrating power of alpha particles, the preparation for counting is often a critical step. Most methods require that sample aliquots be prepared as a virtually weightless mount in vacuum chambers at fixed distances from the alpha detectors. Although electro-deposition onto a stainless steel planchet is the traditional

method for preparing samples for counting, precipitation of the radionuclide of interest onto the surface of a filter planchet is often used to prepare samples for alpha spectroscopy. While this technique generally produces a spectrum with lower resolution, the preparation time is relatively short compared to electro-deposition.

The radionuclides on these planchets are then measured by alpha spectroscopy using detectors housed in a light-tight vacuum chamber. The alpha detectors consist of high-resolution ion implanted silicon diodes. Alpha particle emissions that strike the diodes create voltage pulses proportional to specific energies. Alpha spectroscopy produces a spectrum of alpha particles detected at different energies and displayed by a histogram of the number of counts versus the alpha energy. Peaks associated with specific alpha energies are used to identify the radionuclides, and the activities of the sample are quantified by the disintegration rates. Because the sample is purified prior to counting, all of the alpha particles come from radionuclides of a single element simplifying the process of associating each peak with a specific radionuclide.

Energy levels of alpha particles emitted by radionuclides commonly monitored by our bureau include ^{241}Am at a 5485 keV (kilo electron volt) energy level, ^{238}Pu at 5499 keV, ^{239}Pu at 5155 keV, ^{234}U at 4775 keV, ^{235}U at 4396 keV, and ^{238}U particle emissions at a 4196 keV energy level. Plutonium-239 and ^{240}Pu emissions are at similar energies and are indistinguishable using alpha spectroscopy. Another method, Thermal Ionization Mass Spectroscopy, is used to quantify these isotopes. This method is described in the next section.

Two system calibrations are necessary to characterize the analysis performance. A source with at least two known alpha energies is counted to correlate the voltage pulses with alpha energy. A standard source of known activity is analyzed to determine the system efficiency for detecting alphas. Since the sample and detector are in a vacuum, most commonly encountered alpha energies will be detected with approximately the same efficiency

For environmental detection levels, typically below 0.004 Bq/g or 0.1 pCi/g, samples are counted for 1000 minutes or more. To achieve lower detection limits, laboratories may count larger sample aliquots for longer periods. In addition to the reported concentration value, the laboratories must document performance characteristics of the analysis, such as the analyte and tracer net counts and count time, background counts and background count time, detector efficiencies, and total uncertainty.

These commercial laboratory methods for alpha spectroscopy meet or exceed the requirements referenced by DOE/EML 4.5.2.1.

Thermal Ionization Mass Spectroscopy

The Thermal Ionization Mass Spectroscopy (TIMS) procedure allows for the quantification of the isotopic composition of the plutonium in environmental samples by measuring the relative abundance of atoms for the isotopes ^{240}Pu and ^{239}Pu. Determination of the Plutonium 240:239 atom ratio can be used to distinguish the components of global fallout and Laboratory plutonium, and quantify the mixture. Mass spectrums are obtained by converting a small amount of a purified sample into rapidly moving ions and resolving them on the basis of their mass-to-charge ratio. A thermal ionization mass spectrometer produces ions by heating a chemically separated and purified aliquot of a sample. The ions are resolved into discrete spectral peaks, (their mass-to-charge ratio) by mass analyzers, and quantified by measuring the peak heights and comparing the spectra with reference standards. The procedures for TIMS analysis of plutonium were developed by the Los Alamos Clean Chemistry and Mass Spectroscopy Laboratory and are described in detail in Efurd and others (1993). (In Gallaher and others 2002).

The LANL environmental surveillance program has used alpha spectroscopy to measure plutonium and determine potential Laboratory impacts since 1974. The Laboratory developed a statistical value from multiple samples during the following years to represent a reference level for the highest likely background measurement. If a sample measurement is less than this value, an assumption is made that the plutonium originates from global fallout and not the Laboratory. TIMS measurements can identify Laboratory and global fallout components of plutonium in samples that might normally be measured below the background reference.

Alpha spectroscopy measurements of plutonium are obtained after a sediment sample is digested, chemically separated, purified, and fixed onto a planchet. Alpha spectroscopy measures the number of alpha particles emitted by radionuclide isotopes in environmental samples. Radionuclide isotopes emit alpha particles at discrete energy groups and analytical laboratories use these energy levels to identify individual isotopes. Measuring the rate of the alpha interactions quantifies the concentration in a sample. Although alpha spectroscopy is the primary method for measuring plutonium, the resolution achieved by this method is not adequate to resolve between ^{239}Pu and ^{240}Pu isotopes. Combined ^{239}Pu and ^{240}Pu alpha emissions are measured at approximately 5,155 keV (^{239}Pu occurs at 5,105 keV and ^{240}Pu occurs at 5,123 keV).

The TIMS method is capable of accurately and precisely resolving the ^{239}Pu and ^{240}Pu isotopes. However, the method is not routine or currently available at commercial laboratories.

Beta Emitting Radionuclides (Strontium-90)

Commercial laboratories use low-background gas flow proportional counters to count beta emissions from yttrium-90 (^{90}Y), the daughter product of ^{90}Sr decay. Although, this method accurately identifies and quantifies ^{89}Sr and ^{90}Sr radionuclides in soil, water, air filters, and other sample matrices, it does not resolve the individual isotope's emission energies. Rigorous separation of the yttrium isotope is required from which the strontium isotopes can be quantified.

Laboratory sample preparation, dissolution, and purification procedures for beta emitting radionuclide analysis are similar to that for alpha emitting radionuclides. Heterogeneous samples are ground, mixed, and sieved to produce a fine-grained homogeneous aliquot. Two to 10 gram sample aliquots are spiked with a stable strontium carrier, dissolved with concentrated acids, and then physical and chemical treatments separate and purify the sample for measurement. Portions of the pre- and post-separation solutes are measured by inductively coupled plasma - atomic emission spectroscopy, ICP- AES, for non-radioactive strontium and gas flow proportional beta counts of blank spike samples to monitor chemical yields of the separation technique.

After dissolution, the sample undergoes a preliminary count that represents the total ^{89}Sr and ^{90}Sr activity plus a small fraction of ^{90}Y that has grown in by radioactive decay. The strontium sample is allowed to reach secular equilibrium with its ^{90}Y progeny, a period of approximately 14 days. The sample is then purified using a variety of chemical reactions to remove bulk chemical and radionuclide impurities. The aliquot solute is passed through an extraction column to separate yttrium from the sample solute. The radioisotope yttrium is then eluted from the column with diluted acids, evaporated or fixed onto a stainless steel planchet, and beta counted in a gas-flow proportional counter. Other examples of purification techniques include liquid-liquid extraction, distillation, electro-deposition, and filtration through specially prepared filters.

After the purified yttrium is fixed onto a planchet, the sample undergoes a final 3 to 8 hour beta count on a gas flow proportional counter. Beta particle emissions from the ^{90}Y progeny produce ionization in a gas-filled chamber, generating a small electronic pulse for each interaction. The pulse height is dependent upon the incident energy of the particle. The ^{90}Sr concentration is determined from the yttrium activity and the ^{89}Sr concentration by computing the difference between preliminary and final counts. Data reduction for beta emitting radionuclides is less complicated than that for photon emitting radionuclides. Since the beta detectors report total beta activity, the calculation to determine the concentration for the radionuclide of interest is straightforward. The counter provides raw counting information to computer and spreadsheet-based analysis programs, generating results in units of radioactivity per unit sample volume.

These procedures used to measure ^{90}Sr meet the calibration, data collection, and analysis requirements of EPA method 900.0.

Photon Emitting Radionuclides (Gamma Spectroscopy)

Commercial laboratories use gamma spectrometry to accurately identify and quantify multiple gamma-emitting radionuclides in soil, water, air filters, and other sample matrices. Gamma spectrometry methods, using high-resolution germanium or sodium iodide detectors, can measure mixed isotopes by counting their gamma emissions or photons at specific energies. Laboratories use a spectrum, constructed from the rate of emissions at specific energies, to identify and quantify each analyte

There is no special sample preparation required for gamma spectroscopy measurements beyond placing a sample in a known geometry around the detector. Efficiency calibration curves are developed for different geometries since the counting efficiency depends on the distance from the sample to the detector. Generally, dried, ground materials, sieved to produce homogeneous samples are used. Large volumes, 500 mL, are place into Marinelli beakers that fit around the detector and provide exceptional counting efficiencies. Small volumes can be placed into Petri dishes and placed on top of a detector for counting.

The samples in these geometries are typically counted for 1000 seconds to 1000 minutes on instruments using high-resolution germanium or sodium iodide semiconductors. Germanium detectors have better resolution and can identify radionuclides at lower concentrations, although sodium iodide semiconductors are more efficient. Gamma emissions from radionuclides detected by a semiconductor germanium crystal, provides a small electronic pulse for each gamma interaction that is proportional to the gamma incident energy. The intensity or count rate /energy spectrum is collected and displayed on a multi channel analyzer (MCA). Computers, generating results in units of radioactivity per unit sample volume, subsequently interprets the data collected by the MCA.

Data reduction is usually the critical step in measuring photon-emitting radionuclides. There are often several hundred individual gamma ray energies detected within a single sample. Computer software is usually used to identify the peaks, associate them with the proper energy, associate the energy with one or more radionuclides, correct for the efficiency of the detector and the geometry of the sample, and provide results in terms of concentrations with the associated uncertainty. It is important that the software be either a well-documented commercial package or thoroughly evaluated and documented before use. The counts in each peak or energy band, the sample weight, the efficiency calibration curve, and the isotope's decay scheme are factored together to give the sample concentration.

Although several hundred individual gamma ray energies can be detected by gamma spectroscopy, our bureau is commonly interested in ^{137}Cs. The energy level of gamma photons emitted by ^{137}Cs is 661.7 keV. Additional radionuclides and their energy levels that are measured by gamma spectroscopy include ^{210}Pb (45.6keV), ^{241}Am (59.5keV), ^{60}Co (1173.2 and 1332.5keV), and ^{40}K (1460.8keV).

This gamma spectroscopy procedure is equivalent or exceeds EPA Procedure 901.1 and DOE/EML Procedure 4.5.2.3.

Particle-Size Distribution Aanlysis

Analytical laboratories use a combination of pipette and dry sieving procedures to determine particle size distribution of sand, silt, and clay in soil and sediment samples. Pipette procedures are based on Stokes Law that spherical particles settle in a fluid at rates proportional to their radius and mass. This procedure is based on the pipette method for particle size analysis specified by the U.S. Geological Survey and the Soil Science Society of America and as reported by Day (1965), Jackson (1969), Janitzky (1986), and Gee and Bauder (1992).

Particle size distributions are reported as the representative percent weight fractions for each size component in a sample. The gravel fraction is reported as the percent weight from the entire sample. The remaining sand, silt, and clay components are weight percents from the size fractions less than 2 mm. The size classes are listed in the following table.

Major Class	**Minor Class**	**Minimum Size (mm)**
Gravel	Gravel	2.0
Sand	Very coarse sand	1.0
	Coarse sand	0.5
	Medium sand	0.25
	Fine sand	0.125
	Very fine sand	0.0625
Silt	Coarse silt	0.0015
	Fine silt	0.0002
Clay	Clay	<0.0002

A sample is initially dried and weighed, and then sieved to remove particles greater than 2 mm, including organic matter and gravel. The percent weight of gravel is determined from the weight of the gravel component divided by the initial weight of the sample.

The remaining particle sizes less than 2 mm, including the sand, silt, and clay fractions, are dispersed into a sodium pyrophosphate solution and wet sieved through a 0.063 mm sieve to separate the sand fraction from the silt and clay fractions. The sand fraction retained on the sieve is dried and re-sieved through stacked 1.0, 0.5, 0.25, 0.125, and 0.063 mm sieves. These fractions, including very coarse, coarse, medium, fine, and very fine sand components, are then individually weighed to determine the particle distribution within the sample.

The remaining particles less than 0.063 mm are measured to determine the silt and clay components. The silt and clay particle size distribution is determined by pipette

procedures based on settling rates of particles with different radius and mass. The settling rate for particles less than 0.0015 mm is determined for the ambient laboratory temperature. Based on this rate, a sample aliquot is drawn at the appropriate time to acquire sample material that is less than 0.0015 mm (0.2 μm). A settling rate is also determined for particles greater than 0.2 μm. The sample is shaken and an aliquot is drawn at the appropriate time to acquire a sample material less than 0.2 μm. The sample aliquots are dispersed into containers, dried, and weighed, and then weight percents for the silt and clay components are calculated.

Calculations are computed on laboratory computers from all the weights and sample information described above. Additional weights are recorded from blanks to achieve quality control performances. The following measurements are used to determine the particle size distribution in a soil or sediment sample:

Total weight of oven-dry sample
Weight of oven-dry sample fine-earth fraction (< 2 mm)
Weight of PSDA oven-dry sample
Total weight of sand after wet-sieving
Weight of all sand-size fractions and remaining silt fraction from dry sieving
Weight of clay-and silt+clay oven-dried aliquots
Weight of dispersant from blanks.

APPENDIX E. Historical Contaminant Transport and Transport Since the Cerro Grande Fire

Contaminant Transport and Cerro Grande Ash

After the Cerro Grande fire, we expected increased runoff from watersheds impacted during the fire. We expanded our stormwater monitoring efforts to assess potential changes in hydrology, suspended sediment yield, and contaminant transport rates from the Pajarito plateau. The contaminants we were concerned with included fallout materials concentrated in the burned-forest biomass and LANL legacy wastes distributed in areas around the Laboratory. During a three-year period, we observed elevated $^{239/240}$Pu concentrations in Pueblo Canyon stormwater and began to focus our stormwater monitoring there. Concentrations of ^{90}Sr and ^{137}Cs associated with forest fire ash diminished each year as the ash was flushed from the mountain slopes.

We compared our measurements to a number of reference values in order to understand changes in the environment. The LANL regional background level for $^{239/240}$Pu in northern New Mexico soils is 0.02 pCi/g (mean plus two standard deviations), and the background level in river sediments is 0.01 pCi/g (mean plus two standard deviations). These reference levels and the methodology used to develop them are described in the LANL Environmental Surveillance (ES) reports (LANL, 2002). Their references were derived from soil and sediment samples measured over a period of years from samples collected well beyond the potential influence of the Laboratory, and include a mean of those values plus a measurement of variability. Those numbers reflect an upper tolerance level at the 95% confidence level, the mean plus two standard deviations. They reflect the most probable largest value that might be measured in areas beyond potential impact by the Laboratory. During 1999 we began a soil background study in the Jemez Mountains and established a similar reference level of 0.04 pCi/g for $^{239/240}$Pu in soils.

LANL's Environmental Restoration (RCRA/HSWA) group established additional reference values for soils and sediments at the Laboratory (Ryti, 1998). They used environmental results taken from ES regional stations, as well as samples they collected at LANL perimeter and on-site locations during 1992 through 1995. The statistical treatment used to establish the upper 95% confidence tolerance level was also slightly different than the ES derived regional background values. The ES and ER reference values can be found in Table E-1.

After the fire, we collected ash that represented materials burned during the fire, including overstory and understory components of the forest. The upper tolerance level, or value that we developed to reflect the probable largest value or activity of $^{239/240}$Pu in ash was 0.6 pCi/g. We also evaluated $^{239/240}$Pu measurements in ash-laden sediments in stream channels and on channel banks. These samples were collected from the upper burned watershed areas downstream to the banks of the Rio Grande along White Rock Canyon. These measurements indicated that plutonium activities were diminishing with time and distance from the areas impacted during the fire. The plutonium in ash became diluted as it mixed with clean soils and sediments.

Twenty-two ash and ash-laden sediment samples collected soon after the Cerro Grande fire from the burned forest floor area and in drainages near the burn area were used to develop the 0.6 pCi/g ash reference. This data set did not include 15 additional ash-laden bank deposits samples collected along the Rio Grande in White Rock. These samples demonstrated significant dilution with an average activity of 0.06 pCi/g, near our 0.04 pCi/g upper tolerance background level.

During the first stormwater season after the Cerro Grande fire, we established a stormwater monitoring program to assess contaminant transport with respect to ash and the increase in stormwater flow rates, durations, and frequencies as a result of the fire. Stormwater samples were collected based on opportunity and included 30 samples from several canyons impacted by the fire. We observed $^{239/240}$Pu measurements in suspended sediments in Los Alamos and Pueblo canyons that were substantially greater than the 0.6 pCi/g reference value we established for the Cerro Grande ash and greater than pre-fire. Over the following two years, we focused our monitoring in these areas, particularly Pueblo Canyon. These reference values and measurements made for plutonium and other contaminants in stormwater suspended sediments are summarized in Table E-1.

Table E-1. Plutonium-239/240 reference values and activities measured in stormwater suspended sediments

Reference Values		**pCi/g**
LANL Regional Background Soils (ES)	UTL[a] mean + 2sd	**0.02**
LANL Perimeter Soils Reference (ER)	Linear interpolation	**0.054**
LANL On-site Sediments Reference (ER)	Linear interpolation	**0.068**
NMED Jemez Mountain Soils	UTL[a] mean + 2sd	**0.04**
Cerro Grande Ash	UTL[a] mean + 2sd	**0.6**
Plutonium-239/240 in Suspended Sediments		
2000 – 2002 Pajarito Plateau Stormwater (without Pueblo and Los Alamos canyon)	Mean	**0.1**
2000 – 2002 Pueblo Canyon Stormwater	Mean	**3.4**

[a] Upper Tolerance Level = the sample population mean plus two times its standard deviation (sd)

The following charts represent the values we observed from samples collected during 2000 to 2002. Figure E-1 demostrates the $^{239/240}$Pu differences in Cerro Grande ash and suspended sediments in stormwater from three canyon groups during 2000. They include reference canyons dissecting the Pajarito Plateau, mid Los Alamos Canyon, and lower Pueblo Canyon. It reflects much larger plutonium concentrations in Los Alamos and Pueblo canyons.

The reference values shown in the chart represent $^{239/240}$Pu in suspended sediments from 28 stormwater samples collected in drainages below the burned forest area other than mid Los Alamos and lower Pueblo Canyons. The minimum, 25th percentile, 75th percentile, and maximum values for these reference canyons are 0.001, 0.06, 0.18, and 0.4 pCi/g, respectively. They include five samples collected in Pueblo and Acid canyons above areas impacted by post-1943 Laboratory discharges from TA-45, three in upper Los

Alamos Canyon above the DP Canyon confluence, and 20 in other canyons that dissect the Pajarito Plateau. The samples collected in the other canyons include one in Guaje Canyon, six in Pajarito Canyon, five in Water Canyon, three in Cañada del Buey, and five stormwater samples from the Rio Grande downstream of the Laboratory.

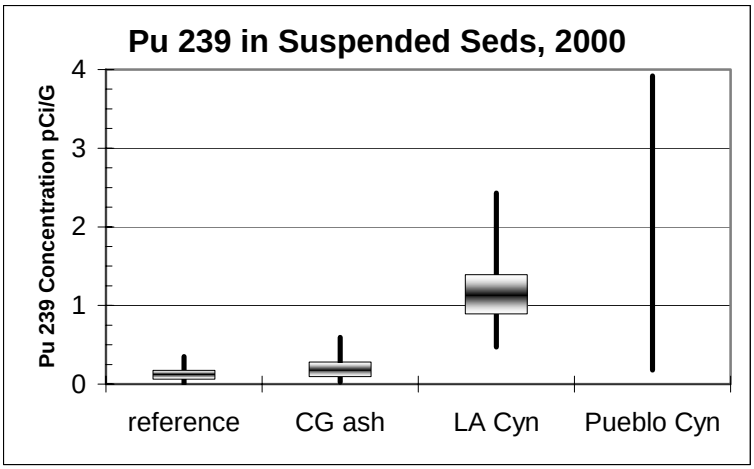


Figure E-1. 2000 Plutonium-239/240 in ash/ash-laden sediment (CG ash) from the Cerro Grande fire, in stormwater suspended sediments from reference canyons, Los Alamos (LA) Canyon, and Pueblo Canyon

The second group in the Figure E-1 chart reflects the values from 22 Cerro Grande ash and ash-laden sediments in or near the burned forest area. They reflect the ash contribution of plutonium expected in the solid phase of the stormwater samples. The minimum, 25th and 75th percentiles, and maximum values are 0.03, 0.09, 0.28, and 0.60 pCi/g, respectively. The Cerro Grande ash group is retained in the following two charts for reference.

Six stormwater samples collected in mid Los Alamos Canyon demonstrate potential transport of legacy materials. The samples were collected at the LANL E050 stormwater gage station below the retention structure at State Road 4, along the eastern boundary of the Laboratory. Those values ranged from 0.47 to 2.43 pCi/g, consistantly larger than the reference plutonium concentrations in stormwater derived from the other Pajarito sites and ash from the Cerro Grande fire.

Only two samples were collected in Pueblo Canyon. They were collected in lower Pueblo Canyon just above the Bayo Wastewater Treatment Plant and were measured at 0.18 and 3.92 pCi/g. Two stormwater samples were collected in the South Fork of Acid Canyon and contained $^{239/240}$Pu activity concentrations in suspended sediments at 107 and 38.1 pCi/g. They are not represented on the Figure E-1 chart.

From these data comparisons and the notable increased magnitude and frequency of stormwater runoff, we recognized that legacy contaminants from Acid, Pueblo, and Los Alamos canyons were potentially being moved at greater rates than before the fire. We began to focus stormwater monitoring in Pueblo and Los Alamos canyons.

Figure E-2 represents samples we collected in 2001. It also corroborates our observations made in 2000 that legacy plutonium transport rates in Pueblo Canyon were increasing. During 2001, we collected eight samples to reflect reference stormwater conditions. They were collected in drainages below the burned forest areas and include three samples in Pajarito Canyon and five in Water Canyon. The minimum, 25th percentile, 75th percentile, and maximum values of $^{239/240}$Pu in suspended sediments are 0.06, 0.07, 0.16, and 1.01 pCi/g, respectively. These data are similar to reference values collected from runoff in the Pajarito canyons during 2000.

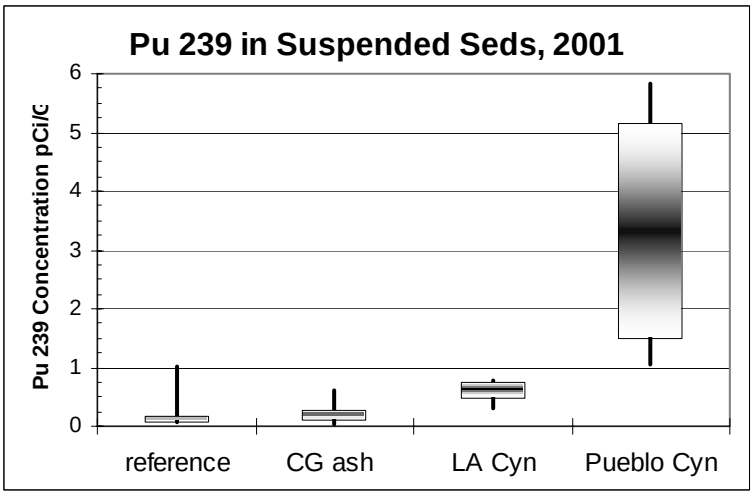


Figure E-2. 2001 Plutonium-239/240 in ash/ash-laden sediments from the Cerro Grande fire, and in stormwater suspended sediment from reference canyons, Los Alamos (LA) Canyon, and Pueblo Canvon

The Cerro Grande ash reference in Figure E-2 is from the same samples described for 2000. They are the ash and ash-laden samples collected shortly after the fire from the forest floor and stream channels in close proximity to the burned watershed.

Four samples were collected from Los Alamos Canyon 8 kilometers (5 miles) upstream of the Pueblo Canyon confluence. The minimum, 25th percentile, 75th percentile, and maximum values are 0.31, 0.47, 0.74, and 0.79 pCi/g, respectively. Most of the measurements are greater than the 0.6 pCi/g upper tolerance value used to describe plutonium in the Cerro Grande ash. These samples demonstrate potential transport of legacy contaminants, but at a smaller degree than in Pueblo Canyon.

Five samples were collected in lower Pueblo Canyon at stormwater gage E060. The minimum, 25th percentile, 75th percentile, and maximum values are 1.05, 1.50, 5.16, and 5.83 pCi/g, respectively. These values are up to 10 times greater than the Cerro Grande ash reference and reflect legacy contaminants.

Our evaluation indicated both Los Alamos and Pueblo canyons were contributing legacy plutonium to offsite transport. Pueblo Canyon was contributing more sediment at higher plutonium concentrations than Los Alamos Canyon. We also observed a greater frequency of floods at greater flow rates at the lower Pueblo Canyon gage station E060. We observed total $^{239/240}$Pu concentrations in water as high as 253 pCi/L. Stormwater retention structures and lower plutonium concentrations in Los Alamos Canyon suspended sediments, as well as the lack of stormwater controls and apparent greater

runoff potential in Pueblo Canyon led us to focus additional monitoring efforts in Pueblo Canyon to more fully characterize storm-runoff contaminant transport.

Figure E-3 represents samples collected during 2002, and continues to corroborate our findings from previous years that plutonium transport from Pueblo Canyon has increased. An evaluation of plutonium concentrations in reference canyons and samples collected in Pueblo Canyon above Acid Canyon suggests that increases of plutonium in stormwater from ash had diminished.

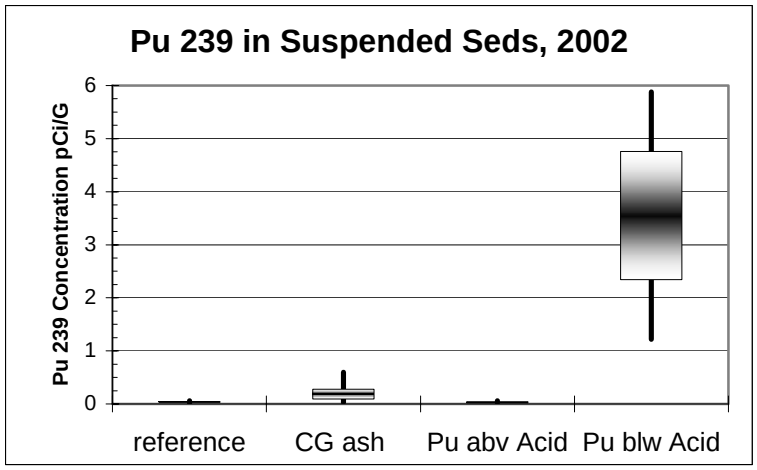


Figure E-3. 2002 Plutonium-239/240 concentrations in Cerro Grande ash, reference canyons, and Pueblo Canyon above and below Acid Canyon, the major contaminant source term

The same Cerro Grande ash samples were used for reference. Ash did not appear to have a long-term effect on contaminant transport from the Pajarito Plateau. Evaluations of other constituents such as ^{90}Sr and ^{137}Cs suggested the same.

A total of five reference samples were taken from Cañada del Buey, and Pajarito and Guaje canyons. The minimum, 25th percentile, 75th percentile, and maximum values are 0.01, 0.03, 0.04, and 0.06 pCi/g $^{239/240}$Pu in suspended sediments, respectively. Twelve samples in Pueblo Canyon above the Acid Canyon confluence also demonstrate reference conditions. The minimum, 25th percentile, 75th percentile, and maximum values are 0.02, 0.02, 0.04, and 0.06 pCi/g, respectively. These values are similar to background sediment measurements.

Most of twenty samples that reflect plutonium transport from Pueblo Canyon were collected in the lower reaches of the canyon at E060, near the Los Alamos Canyon confluence. The minimum, 25th, 75th percentiles, and maximum $^{239/240}$Pu concentrations from Pueblo Canyon stormwater samples are 1.22, 2.34, 4.76, and 5.88 pCi/g $^{239/240}$Pu in suspended sediments, respectively. Two additional samples in Acid Canyon, not represented in the Figure E-3 chart, were measured at 9.1 and 22.3 pCi/g.

The plutonium concentrations measured in Pueblo Canyon stormwater suspended sediments did not diminish during the three years described in this report. This condition also suggests an alternative source of plutonium in stormwater rather than from the Cerro Grande ash.

Our evaluation of ^{137}Cs measurements in ash and stormwater suspended sediments demonstrated similar conditions. Cesium-137, like $^{239/240}$Pu, concentrates in ash after fire

reduces the biomass of an organism. As time and distance increases from the source, in this case the burned forest areas, the ash mixes with clean soils and sediments, diluting the original concentrations. Figure E-4 shows ^{137}Cs concentrations diminish each year after the Cerro Grande fire until it approximates the LANL regional reference background level for soils at 0.51 pCi/g. The measurements in the Cerro Grande ash were more variable than seen in the Viveash ash. A storm event occurred before the Viveash samples were collected, and mixing with the underlying soil may have modified the variation as seen in the Cerro Grande ash.

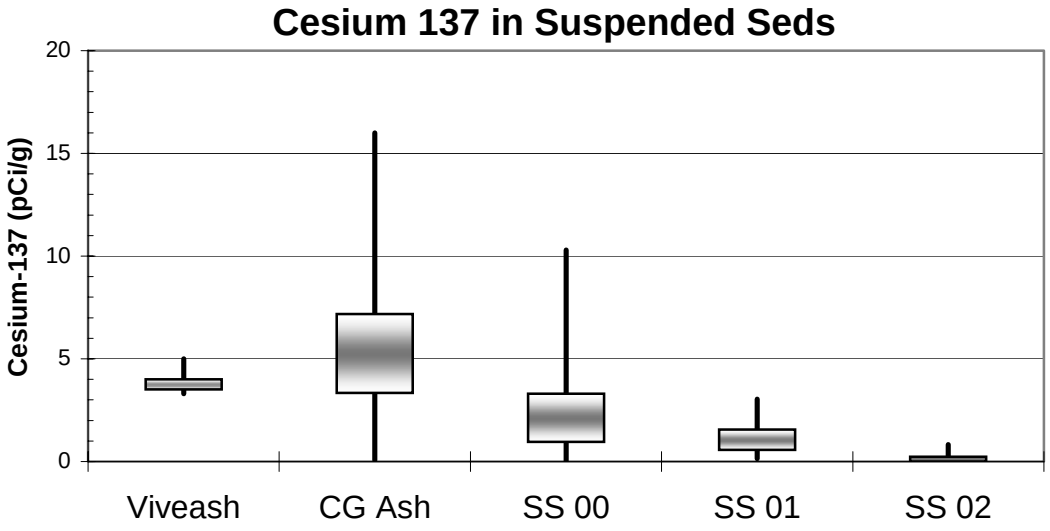


Figure E-4. Cesium-137 concentrations in Viveash and Cerro Grande ash/ash laden sediment (CG Ash), and in stormwater suspended sediment samples collected in 2000 (SS 00), 2001 (SS01), and 2002 (SS 02)

Five ash samples were collected in the Viveash area, approximately 72 kilometers (45 miles) east of the Cerro Grande fire. Twenty-eight ash and ash-laden samples were collected and analyzed for ^{137}Cs in the Cerro Grande burn area. Twenty-eight samples were also collected from stormwater runoff during 2000. In 2001 and 2002, 17 and 15 samples were collected from runoff, respectively. The ^{137}Cs activities in the Viveash ash, Cerro Grande ash, and the 2000, 2001, and 2002 stormwater runoff suspended sediments ranged from 3.3 to 5 pCi/g, 0.06 to 16 pCi/g, 0.0 to 10.3 pCi/g, 0.14 to 3.04 pCi/g, and –0.34 to 0.82 pCi/g, respectively. These values diminish at an approximate 50% rate from each preceding year. A Mortandad Canyon suspended sediment sample from 2000 was measured at 234 pCi/g and is not represented in Figure E-4.

Figure E-5 demonstrates similar characteristics for ^{90}Sr concentrations in ash and suspended sediments, although ^{90}Sr concentrations reached background reference levels, 0.71 pCi/g, by 2001. The Viveash samples were measured at levels near background. Strontium-90 is more soluble than $^{239/240}$Pu and ^{137}Cs and may have been removed more efficiently by the storm runoff.

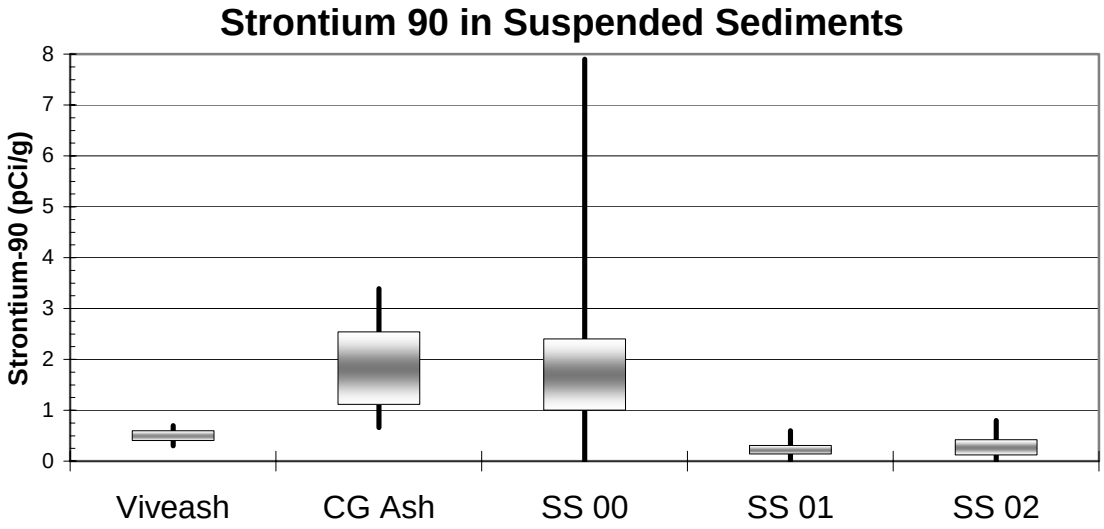


Figure E-5. Strontium-90 measurements in ash collected from the 2000 Viveash and Cerro Grande fires, and in suspended sediments collected from stormwater in 2000 (SS 00), 2001 (SS 01), and 2002 (SS 02)

These ^{90}Sr measurements were made on the same samples described above for ^{137}Cs. Five samples were collected from ash in the Viveash area, 28 ash and ash-laden samples were in the Cerro Grande area. Twenty-eight samples were collected from stormwater runoff during 2000, 17 samples in 2001, and 15 in 2002. The ^{90}Sr concentrations in the Viveash and Cerro Grande ash, and in the 2000, 2001, and 2002 storm runoff ranged from 0.3 to 0.7 pCi/g, 0.66 to 3.39 pCi/g, 0.0 to 7.9 pCi/g, -0.11 to 0.6 pCi/g, and 0.0 to 0.8 pCi/g, respectively.

The legacy contaminants described in this report refers to the discharges from LANL during the first 20 years of operations. LANL discharged untreated and treated radioactive industrial wastewater into Acid Canyon from 1943 to 1963. Early release estimates indicated 180 mCi of plutonium were discharged into the canyon (Stoker, 1981). Later inventory estimates of plutonium in Pueblo Canyon sediments indicated a larger release. In 1985, LANL estimated that 300 to 900 mCi of $^{239/240}$Pu existed in Pueblo Canyon, and up to 3 curies of plutonium could have been released into the canyon (Lane and others, 1985). By 2003, LANL estimated up to 1.3 curies existed in Pueblo Canyon from the Acid-Pueblo Canyon confluence downstream to the Pueblo-Los Alamos Canyon confluence (Reneau, 2003). Younger sediment deposits have replaced much of the older post-1943 more contaminated units. These younger units are comprised of cleaner background sediments mixed with the older, more contaminated sediments deposited during the early operations at the Laboratory when maximum discharges occurred.

Historical Plutonium Transport

In 1993, William Graf estimated 188 mCi of plutonium was transported from Los Alamos Canyon into the Rio Grande by storm runoff from 1944 to 1986. Graf (1993) suggested that the contribution to the plutonium budget from LANL is associated with relatively coarse sediment that often behaves as bed load in the Rio Grande. Infusions of these materials into the Rio Grande were largest in 1951, 1952, 1957, and 1968. Although the LANL contribution to the entire plutonium budget was relatively small (and may be largely underestimated), in these four years it constituted 71 to 86 percent of the plutonium in bed load immediately downstream from Otowi Bridge (Graf, 1993).

Graf developed his estimates from previous researchers' calculations for the probable sediment yield from the canyon into the Rio Grande. They used data from an intermittent storm water gage record for the (Los Alamos) stream and precipitation records at nearby locations (Graf, 1993). His evaluation was for plutonium contribution to the Rio Grande at the confluence with Los Alamos Canyon. Pueblo Canyon is a tributary to Los Alamos Canyon with its confluence located about 8 km (5 miles) upstream of the Rio Grande. Pueblo Canyon provides the majority of Laboratory contaminants to lower Los Alamos Canyon and the Rio Grande. Other sources include radioactive liquid-waste discharges into DP Canyon; a small tributary to Los Alamos Canyon located approximately 5.5 km upstream of the Los Alamos-Pueblo Canyon confluence. Analytical methods may also have been different than those we used to evaluate suspended sediments in storm water.

Estimates by Graf were likely underestimated. They were based on undocumented modeled values for plutonium concentrations in bedload derived from ES samples post-dating years of maximum plutonium measurements in the upper Los Alamos Canyon watershed. The flood sizes may also have been underestimated. Flood sizes were modeled on rainfall from a single precipitation station in Los Alamos. Laboratory scientists suspect that the accelerated post-Cerro Grande fire transport of sediments and plutonium is dwarfed by that prior to 1965 (personal communication, S., Reneau, 2007).

A summary of Graf's (1993) findings is provided below in Table E-2. We included additional information and data calculated from the Graf data in Table E-2. The average concentration of $^{239/240}$Pu was derived from plutonium mass transported per year measured in mCi, and the sediment yield per year measured in tons. The average suspended sediment concentration was derived from the water volume flow for each year measured in acre feet and the sediment yield per year measured in tons. We found these measurements similar to those measured in Pueblo Canyon stormwater during 2000 to 2002.

Table E-2. Estimates of plutonium-239/240 and sediment transport into the Rio Grande from Los Alamos Canyon (Graf, 1993)

From W. L. Graf, (1993) Geomorphology of Plutonium in the Northern Rio Grande

Water, Sediment, and Plutonium Data for Los Alamos Canyon						Additional Calc from table	
Year	Water (ac ft)	Flood (cfs)	Sediment (tons)	Pu Sum (mCi)	Pu (Yr) (mCi)	Pu ave conc. in SS pCi/g	ave SSC mg / l
1943	22	66	466		0		15578
1944	198	631	8393	3	2.798	0.37	31175
1945	0	0	61	3	0.03	0.54	0
1946	28	80	611	3	0.32	0.58	16049
1947	1	2	65	3	0.05	0.85	47805
1948	0	0	61	3	0.04	0.72	0
1949	0	0	61	3	0.05	0.90	0
1950	6	20	77	3	0.06	0.86	9438
1951	236	687	9814	20	16.9	1.90	30584
1952	209	386	6316	38	17.61	3.07	22226
1953	2	4	12	38	0.03	2.76	4413
1954	40	129	1006	41	2.86	3.13	18497
1955	91	283	2783	50	8.83	3.50	22492
1956	0	0	0	50	0		0
1957	433	649	16470	94	43.95	2.94	27975
1958	63	203	2002	101	7.36	4.05	23371
1959	33	59	532	103	1.74	3.61	11857
1960	0	0	154	103	0.75	5.37	0
1961	18	53	443	106	2.4	5.97	18101
1962	0	1	138	107	0.88	7.03	0
1963	88	283	2772	117	10.07	4.00	23167
1964	0	0	0	117	0	0	0
1965	124	233	3163	127	9.88	3.44	18760
1966	10	32	165	127	0.53	3.54	12135
1967	129	351	4197	137	10.24	2.69	23928
1968	287	924	14120	159	21.82	1.70	36184
1969	124	149	2899	164	4.96	1.89	17194
1970	0	0	0	164	0	0.00	0
1971	16	42	247	165	0.42	1.87	11354
1972	0	0	0	165	0	0	0
1973	109	349	3955	173	8.2099	2.29	26686
1974	6	20	129	173	0.3301	2.82	15812
1975	4	6	99	173	0.3	3.34	18203
1976	6	20	77	174	0.23	3.29	9438
1977	1	4	8	174	0.0299	4.12	5884
1978	108	293	3198	180	6.0101	2.07	21778
1979	10	312	426	181	1.4899	3.86	31331
1980	0	0	183	182	0.8001	4.82	0
1981	0		0	182	0	-	-
1982	0	-	0	182	0	-	-
1983	43	-	24357	185	2.78	0.13	416597
1984	0	-	0	185	0	0.00	0
1985	43	-	41461	187	2.08	0.06	709140
1986	3	-	2460	188	1.59	0.71	603079

Note: 1943-1980 data from calculation by J. L. Lane in support of Lane, Purtyman, and Becker (1985): 1981- 1986 data from Purtyman et. Al. (1990) using different techniques. The comparability of the two data sets is unknown.

Graf showed plutonium transport from Los Alamos Canyon into the Rio Grande at 44 mCi in 1957. Based on data in Table E-2, 86% of the plutonium transport inventory during 1943 to 1986 occurred during the 1950s and 1960s. Approximately 15% was transported during the remaining periods of his study, the 40's, 70's and 80's. The four greatest mass transport rates occurred in 1957, 1968, 1952, and 1951, where 44, 22, 18, and 17 mCi of plutonium transport occurred, respectively. Water volume that passed through lower Los Alamos during those years was 433, 287, 209 and 236 acre feet, with single maximum runoff rates of 649 (18.4 m3/s), 924 (26.2 m3/s), 386 (10.9 m3/s), and 687 (19.5 m3/s)cfs. These values for transport inventory, annual water volume, and single largest annual flow rate associations are demonstrated in Figures E-6, E-7, and E-8.

We observed similarities in the measurements presented by Graf, values derived from those measurements, and our measurements from lower Pueblo Canyon. Plutonium concentrations in sediment that we derived from his data range from 7.03 pCi/g to 0 pCi/g. The average plutonium concentration in sediments sampled from 1950 to 1981, is 3.4 pCi/g. The average suspended sediment concentration in stormwater for this period was calculated as 15,045 mg/L. The pre-1950 and post-1981 values reflected very low values and may suggest contaminant dispersion had not reached lower Los Alamos Canyon or storm flow rates were greatly diminished during these periods. During 2000 to 2002 we estimated that stormwater flows from Pueblo Canyon transported 87 mCi of $^{239/240}$Pu in 22,000 tons of sediment, averaging 4.5 pCi/g plutonium in 13,133 mg/L suspended sediments (Englert and Ford-Schmid, 2004). Preliminary estimation of transport since the 2000 Cerro Grande fire to 2006 indicates that a total 199 mCi $^{239/240}$Pu has been transported from Pueblo canyon (report in press). These transport rates may be underestimated due to the complexities associated with multiple surges in flood stage resulting from the random nature of rainfall and contributions of flow from adjacent canyons

Storm runoff and sediment transport is closely associated with annual precipitation and precipitation intensity. Figure E-6 demonstrates precipitation amounts for the years 1943 to 1986 during the months April through October. It also shows the long term 20 year average at 1986 for summer months to be 13.8 inches (35 cm). The total long term average, including precipitation during November through March, was 18.6 inches (47 cm). Precipitation during the four years of greatest plutonium transport inventory was 24 inches (61 cm) during 1957, 15 inches (38 cm) in 1968, 25 inches (64 cm) in 1952, and 13 inches (33 cm) in 1951. In 1957 and 1952, precipitation was almost double the summer rainfall average of 13.8 inches (35 cm). In 1968 and 1951, the rainfall was near the average. The two greatest floods occurred in 1968 and 1951, when 924 cfs (26.2 m^3/s) and 687cfs (19.5 m^3/s) flows occurred, respectively. Northern New Mexico was demonstrating drought conditions during the 1950s and is often the case, rainfall that did occur, originated during infrequent but intense rain events.

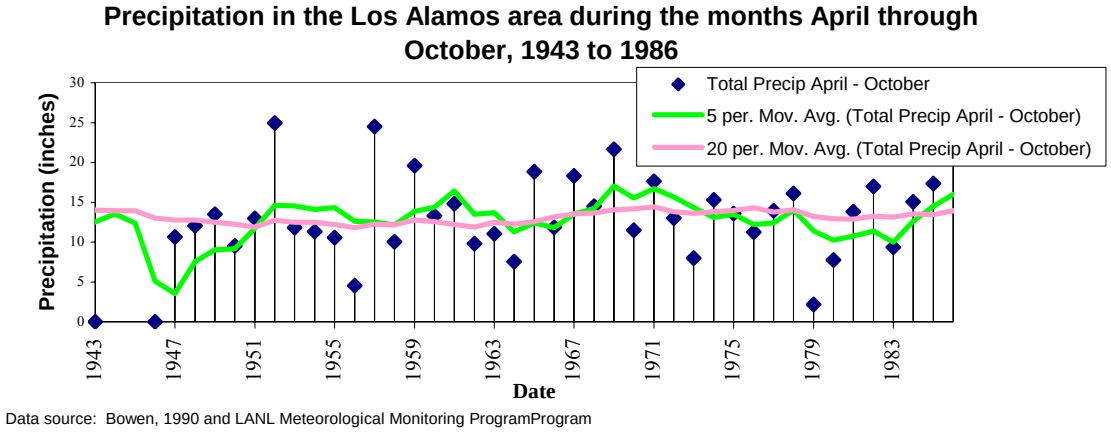


Figure E-6. Annual precipitation in Los Alamos area from 1943 to 1986

During the late 1990s and into the 2000s, New Mexico experienced a drought. During 2000, 2001, and 2002, the average annual precipitation between two rain gages in the upper Pueblo Canyon watershed was 13 (33 cm), 8 (20 cm), and 12 (30 cm) inches, respectively. The Cerro Grande fire exacerbated the water runoff volume to rainfall rates. We observed that the total volume of flow through Pueblo Canyon dramatically increased as total rainfall decreased. A normalized value provides a rough sketch of changes in annual flow to precipitation rates. Major changes in the runoff conditions can be observed and represented by dividing the annual flow, measured in acre feet, by annual precipitation, reported in inches per year. For example, the normalized value described above in Pueblo Canyon doubled after the Cerro Grande fire, from 15 to 36 (unit less). A similar relationship in lower Los Alamos Canyon occurred during the period as described by Graf (1993). In lower Los Alamos Canyon, the ratio between flow and precipitation was small, less than one, during times of minimal transport. During periods of greatest transport, the ratio increases ranged from five to as much as 15.

Annual water volumes as well as the single greatest flow rate per year are demonstrated in Figure E-7. The vertical bars demonstrate the relatively large flow volumes described above. For example, in 1951, 1952, 1957 and 1968, relatively large runoff volumes of 236, 209, 433, and 287 acre feet occurred, respectively, and were associated with rainfall amounts of 13 (33 cm), 25 (64 cm), 24 (61 cm), and 15 inches (38 cm), respectively. The flow volume to precipitation ratios described above, were 15, 7, 15, and 15, respectively. Individual flood rates were also greatest in each of the years with greatest inventory transport. During 1951, 1952, 1957, and 1968, the annual single greatest flow rates were 687 (19.5 m^3/s), 386 (10.9 m^3/s), 649 (18.4 m^3/s), and 924 cfs (26.2 m^3/s). In 1944, a large flow of 631 cfs (17.9 m^3/s) occurred, although the plutonium transport rate was relatively small. This was probably due to plutonium not being thoroughly distributed downstream of the source term.

After 1969, it appears flows through lower Los Alamos Canyon to the Rio Grande were nonexistent to infrequent. The high runoff rates during the early years may have been a

result of the destabilization of the mesa-top ground surface during the construction of the Laboratory. The Pajarito Plateau fluvial systems may have slowly stabilized after the main construction activities were completed and resulted in lower runoff rates.

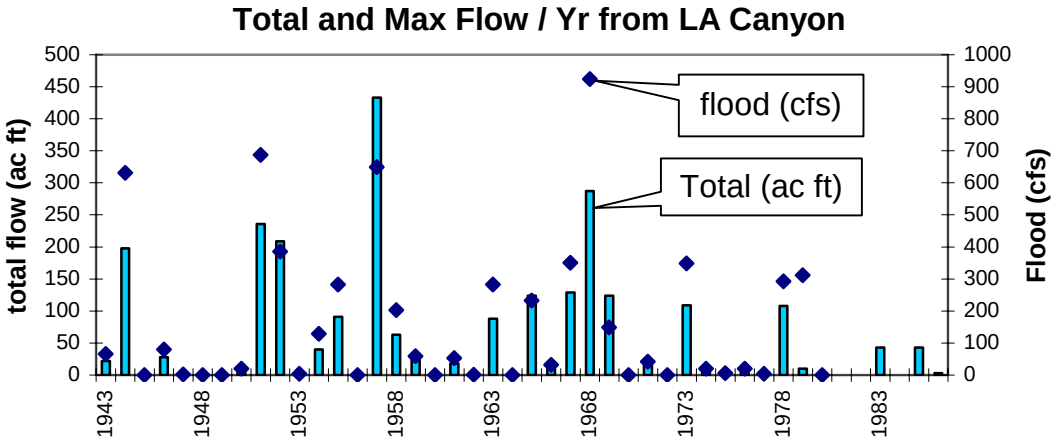


Figure E-7. Total annual flow in acre feet and the maximum flow for the single greatest runoff event per year in cfs.

Figure E-8 demonstrates the annual mass transport rates of plutonium estimated by Graf (1993). It shows the relative relationships of mass transport between years. For example, the greatest mass transport rate was 44 mCi per year in 1957, followed by 22 mCi in 1968, and 18 and 17 mCi in 1952 and 1951, respectively. Several years of 10 mCi per year transport rates occurred. These were all associated with large annual flow volumes or flood flow rates.

Since the Cerro Grande fire, we estimate 55 mCi, 24 mCi, and 8 mCi of $^{239/240}$Pu was moved beyond the E060 gage in lower Pueblo Canyon during 2001, 2002, and 2000, respectively (Englert and Ford-Schmid, 2004). Preliminary assessments of stormwater since 2002 to the end of 2006 indicates an additional 111 mCi of $^{239/240}$Pu has been transported out of Pueblo Canyon. Transport rates as large as these have not been seen since the 1950s and 1960s.

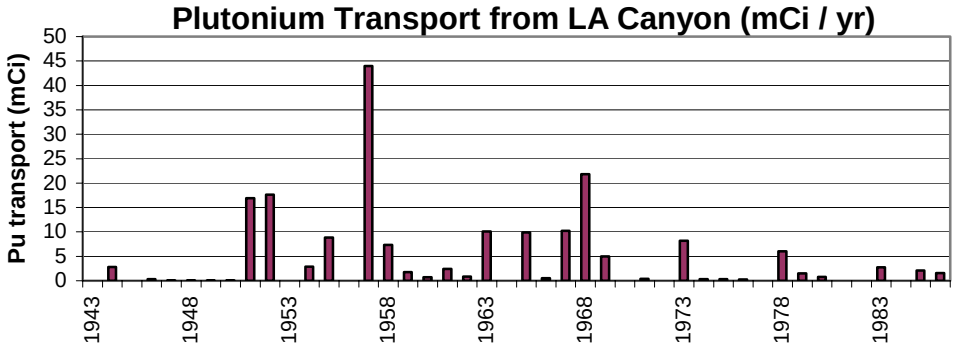


Figure E-8. Plutonium annual mass transport rates estimated by Graff

APPENDIX F. Field Notes

Terrace Core Field Notes
Santa Clara Site
Station DOE OB 3
April 24, 2001
Time: 12:55

Sample	Horizon	Description
1	0 – 14”	very fine grain sand, silt, clay
2	17” – 23”	medium grain sand
3	23”	sand with gravel

Santa Clara Site
Station DOE OB 4
April 24, 2001
Time: 12:00

Sample	Horizon	Description
1	0 – 9”	silt, clay, brown
2	9’ - 13”	clay dark brown, at 13” very fine sand, light brown
3	15” – 24”	fine sand, brown, clay horizon at 22”-23”
4	24” – 29”	fine sand, brown
5	29” – 35”	fine sand, brown – medium to coarse sand, brown at 31”

Santa Clara Site
Station DOE OB 5
April 24, 2001
Time: 12:00

Sample	Horizon	Description
1	0 – 9”	fine silt, clay, brown
2	9’ - 10”	very fine sand, clay
3	10” – 23”	fine silt, clay, brown
4	23” – 28.5”	increasing clay, silt, moist at bottom
5	28.5” – 30.5”	increasing clay, silt, moist
6	30.5”- 40”	clay to medium grain sand, coarsening downward, some pebbles
7	40” – 43”	clay to medium grain sand, coarsening downward, some pebbles
8	43” – 46”	very coarse grain sand, gravel, with cobbles
9	46” – 62”	saturated, clay at bottom of hole, top is gravel
10	62” – 68”	saturated clay

Cañada Ancha Site
Station
February 12, 16, 1999 and April 10, 2000
Time: 16:00

Sample	Horizon	Description
1	0 – 1	sandy soil, light orange brown – sandy loam
2	1 – 2	sandy soil with quartz grain sand, light brown – sandy loam
3	2 – 3	sandy soil with quartz grain sand, light brown – sandy loam
4	3 – 4	sandy soil, with quartz grain sand, pebbles, clay, dark brown – sandy loam to clay loam to loam
5	4 – 5	sandy soil, increasing clay, dark brown – loam
6	5 – 6	predominantly clay, dark brown, with sandy soil – silty clay loam
7	6 –6.5	clay, dark brown on top (2-3”), sandy soil, light brown – silty clay loam
8	6.5 – 7	sandy soil, brown, predominantly clay, dark brown at bottom 4 5 “, moist – silty clay loam
9	7 – 7.5	top 2 – 3 “, clay, dark brown to gray black, bottom, clay sand (water table), wet dark brown – coarse grain sand – silty clay
10	7.5 – 8	wet, coarse grain sand, brown – silty clay
11	8 – 8.5	wet, coarse grain sand, brown
12	8.5 – 9	wet, coarse grain sand with pebbles, brown
13	9 – 9.75	wet, coarse grain sand with pebbles, brown
14	9.75 – 10	wet, coarse grain sand with pebbles, brown, more pebbles at bottom
15	10 – 10.25	top - wet, coarse sand, brown, bottom – coarse sand with pebbles, brown - sand
16	10.25 – 10.5	top - wet, coarse sand, brown, bottom – coarse sand with pebbles, brown- sand
17	10.5 – 10.9	top - wet, coarse sand, brown, bottom – coarse sand with pebbles, brown, more gravel at bottom- sand
18	10.9 – 11	wet coarse sand with gravel- sand

Pajarito Canyon Site
Station 1A
September 28, 1998
Time: 16:15

Sample	Horizon	Description
1	0 – 11”	medium to coarse grained sand
2	11 – 12”	gravel (0.2 – 1.0 cm), gray,
3	18”	

Station 1B
September 28, 1998
Time: 17:00

Sample	Horizon	Description
1	0 – 1’	medium sand, tan to gray, possible eddy deposit

Water Canyon Site
Station 2
September 29, 1998
Time: 17:00

Sample	Horizon	Description
1	0 – 1’	medium sand
2	1 – 2’	medium sand
3	2 – 3’	fine sand
4	3 – 4’	large cobble

Water Canyon Site
Station 2B
September 29, 1998
Time: 17:00

Sample	Horizon	Description
1	3.0 – 3.3	brown, silt lens with bed structure, pinches out laterally

Frijoles Canyon Site
Station 3
September 30, 1998
Time: 12:00

Sample	Horizon	Description
1	0 – 1”	organic soil “O” horizon, dark brown, moist
2	1 – 12”	fine sand, silt, plus very little clay, brown
3	12” – 24”	fine sand, silt, clay, brown
4	24” – 36”	fine sand, silt, clay, brown
5	36” – 48”	fine sand, silt, clay, brown, *silt layer at 48” with visible red particles (FeO?), dry
6	48” – 60”	fine sand, silt, clay, brown, increasing fines to 60”, dry
7	60” – 72”	fine sand, silt, clay, dry

APPENDIX G. Laboratory Quality Control Narrative

Laboratory Quality Control Narrative

The following is a summary of the laboratory quality control results and steps taken to rectify measurements that did not meet quality standards. Commercial laboratories analyzed 35 sediment samples for ^{241}Am, ^{238}Pu and $^{239/240}$Pu, uranium-234, -235, and-238 by alpha spectroscopy, ^{90}Sr by gas flow proportional counting of beta emissions, ^{137}Cs by gamma spectroscopy, and particle size distribution by grain size sieve and pipette methods. Sixteen samples were collected and submitted for analysis during 1998, 10 samples during 1999, three samples during 2000 and six samples during 2001. All samples analyzed for grain size distributions were submitted in 2002 for analysis.

Americium-241

Paragon Analytics, Inc. used alpha spectrometry procedures described by their Standard Operating Procedures (PAI SOPs) 773, 778, 780, and 714 to measure ^{241}Am. These commercial laboratory methods meet or exceed the requirements referenced by DOE/EML 4.5.2.1.

No anomalous situations were encountered during the preparation or analysis of samples collected in 1998 and 2001 samples. All quality control criteria were met.

In 1999, sample results failed to meet internal data quality objectives using 5-gram aliquots. They were re-extracted using 2-gram aliquots. The ^{241}Am activity in the method blank was above the Method Detection Concentration and the results were compared to the blank activity to determine the validity of each measurement. An unidentified peak was also present in all samples at an energy level in the range of 6030 to 6050 keV but was not believed to affect the reported results. There were no further anomalous situations encountered during preparation or analysis of the samples and all quality control criteria were met.

Lower Method Detection Concentrations, 0.002 pCi/g, were requested for the 2000 samples, and 20-gram aliquots were prepared and analyzed by the laboratory. Potential interference from ^{228}Th and low tracer recoveries during the ^{241}Am analysis required the laboratory to put the samples through a thorium clean-up procedure. The minimum detection levels were not met and the best available results were submitted and compared to the method blank activity to validate the measurements. No further anomalous situations were encountered and all quality control criteria were met.

Isotopic Plutonium

Paragon Analytics, Inc. used alpha spectrometry procedures described by PAI SOPs 773, 778, and 714 to measure ^{238}Pu and $^{239/240}$Pu. These commercial laboratory methods meet or exceed the requirements referenced by DOE/EML 4.5.2.1.

No anomalous situations were encountered during the preparation or analysis of samples collected during 1998. All quality control criteria were met.

The duplicate error ratio for a ^{238}Pu analysis of a 1999 sample collected between 91 and 101 cm (3 and 3.3 feet) at Cañada Ancha and its laboratory duplicate was greater than the laboratory warning limits of 1.42. The DER was equal to 1.97 and was considered acceptable according to Paragon Analytics, Inc. data evaluation methods described in PAI SOP 715. No further anomalous situations were encountered during the preparation or analysis of these samples, and all remaining quality control criteria were met.

Lower Method Detection Concentrations, 0.002 pCi/g, were requested for the 2000 samples, and 20-gram aliquots were prepared and analyzed by the laboratory. Potential interference from ^{228}Th during the ^{238}Pu analysis in samples collected at Cañada Ancha between 213 and 229 cm (7.0 and 7.5 feet), as well as low tracer recoveries for samples collected at Cañada Ancha between 186 and 213 cm (6.1 and 7.0 feet) and between 323 and 335 cm (10.6 and 11.0 feet) required additional thorium clean up. Results were reported for the cleaned-up batch for samples with results less than 0.002 pCi/g. No further anomalous situations were encountered during the preparation or analysis of these samples. All quality control criteria were met.

In an effort to achieve 0.002 pCi/g detection levels for samples collected during 2001, 10-gram aliquots were measured for 1000 minutes. The minimum detection levels we requested were not met and the best available results were submitted. All quality control criteria were met.

Isotopic Uranium

Paragon Analytics**,** Inc. used alpha spectrometry procedures described by PAI SOPs 773, 778, and 714 to measure uranium isotopes, -234, -235, and-238. These commercial laboratory methods meet or exceed the requirements referenced by DOE/EML 4.5.2.1.

A technician error was discovered during initial sample preparation procedures in samples collected in 1998. The samples were re-extracted in a separate batch and re-analyzed. All quality control criteria were met from the second batch.

No anomalous situations were encountered during preparation and analysis of samples collected during 1999. All quality control criteria were met.

A potential high bias in ^{235}U results is reported for samples collected during 2001. This bias is due to the possible tailing of the ^{234}U peak into the ^{235}U region on the spectroscope. The results were submitted without further qualification. No other anomalous situations were encountered during the preparation and analysis of these samples and all quality control criteria were met.

Strontium-90

Paragon Analytics, Inc. used gas flow proportional counter spectrometry procedures described by PAI SOPs 707, and 724 to measure ^{90}Sr. These procedures meet the calibration, data collection, and analysis requirements of EPA method 900.0.

Total strontium is reported as ^{90}Sr. The presence of other radioisotopes of strontium may cause positive bias in the measured strontium concentration. The chemical yield observed for some samples sampled in 1998 and 1999 fell between 100% and 110%. To minimize the introduction of low bias, results were calculated conservatively assuming quantitative chemical yield (100%). The magnitude of the low bias was estimated to be less than 10% of the reported value. No further problems were encountered during the preparation and analysis of these samples or following samples collected in 2000 or 2001. All remaining quality control criteria were met.

Cesium-137

Paragon Analytics, Inc. used high-resolution germanium or sodium iodide detectors gamma spectrometry methods described by PAI SOPs 739, and 713 to measure gamma-emitting radioisotopes. Cesium-137 is the only gamma-emitting isotope evaluated for this project. This procedure is equivalent or exceeds EPA Procedure 901.1 and DOE/EML Procedure 4.5.2.3.

The Full Width Half Maximum (FWHM) instrument calibration had expired before analysis of samples collected in the 1998 data set. However, the FHWM is checked daily for each detector and would not be used if the calibration criteria were not within control limits. The data quality is believed to be unaffected. No further problems were encountered with these samples, samples collected in 1999, 2001 or their associated quality control samples. All remaining quality control criteria were met.

In 2000, the sample volumes were insufficient to prepare a duplicate. A duplicate analysis was performed on one sample in lieu of a preparation duplicate. No problems were encountered and all quality control criteria were met with the ^{137}Cs measurements.

Particle Size Distribution

Methods used by the Desert Research Institute Soil Characterization and Quaternary Pedology Laboratory for particle-size distribution analysis (PSDA) of soils and sediments follow standard QA/QC procedures and analytical methods commonly employed for the analysis of sediment and soils. Particle size analysis consists of determining representative percent weight fractions of the sand, silt, and clay fractions. Determination of PSD is by the pipette and dry sieving methods specified by the U.S. Geological Survey and the Soil Science Society of America.

Results of duplicate analysis for all the samples collected during 1998, 1999, 2000, and 2001 were within the 99.5% confidence level, with 1 exception, sample 0105085-3, which contained 95% sand. The remaining quality control criteria were met

APPENDIX H. Calculated Risk Tables

Table 1. Risk calculated for all radionuclide measurements

Station Description	Date Collected	234U (pCi/g)	RISK[1]	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
Santa Clara Sites																			
DOEOB 3 (abandoned flood plain, active 1941-1968)2B																			
5.18 - 30.5 cm (hand augered)	4/24/2001	0.90	2.24E-07	0.039	2E-07	0.68	9.15E-07	-0.01	0	0.003	0	0.001	0	0.001	0	0.005	0	0.102	1.71E-06
DOEOB 4 (abandoned flood plain, active prior to 1941)3B																			
33.5 - 39.6 cm (hand augered)	4/24/2001	0.90	2.24E-07	0.044	2.26E-07	0.91	1.22E-06	0.12	5.19E-07	0.003	0	0.007	5.3E-08	-0.002	0	0.001	0	0.036	6.03E-07
73.2 - 88.4 cm (hand augered)	4/24/2001	0.38	9.45E-08	0.017	8.72E-08	0.39	5.22E-07	0.22	9.52E-07	0.010	0	0.006	4.45E-08	0.001	0	-0.001	0	0.027	4.52E-07
DOEOB 5 (abandoned flood plain, active prior to 1941)3B																			
24.4 - 39.6 cm (hand augered)	4/25/2001	1.14	2.84E-07	0.060	3.08E-07	0.98	1.32E-06	0.07	3.03E-07	0.008	0	-0.001	0	0.001	0	0.000	0	0.036	6.03E-07
57.9 - 73.2 cm (hand augered)	4/25/2001	0.86	2.14E-07	0.059	3.03E-07	0.86	1.16E-06	0.07	3.03E-07	0.006	0	0.001	0	0.000	0	0.001	0	0.033	5.53E-07
116 - 158 cm (hand augered)	4/25/2001	1.15	2.86E-07	0.053	2.72E-07	1.09	1.47E-06	0.01	4.33E-08	-0.003	0	0.005	3.86E-08	0.000	0	0.000	0	0.041	6.87E-07

Table 1. Risk calculated for all radionuclide measurements, cumulative risk per horizon in last column

Station Description	Date Collected	60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
Santa Clara Sites																		
DOEOB 3 (abandoned flood plain, active 1941-1968)2B																		
5.18 - 30.5 cm (hand augered)	4/24/2001	0.05	1.39E-06	0.86	0	0.83	0	0.64	0	0.264	0	0.77	0	0.79	0	0.05	5.55E-07	**4.99E-06**
DOEOB 4 (abandoned flood plain, active prior to 1941)3B																		
33.5 - 39.6 cm (hand augered)	4/24/2001	0.038	1.05E-06	1.01	0	1.01	0	0.86	0	0.315	0	0.91	0	1.10	0	0.04	4.97E-07	4.4E-06
73.2 - 88.4 cm (hand augered)	4/24/2001	0.037	1.02E-06	0.47	0	0.43	0	0.40	0	0.148	0	0.41	0	0.44	0	0.04	4.86E-07	3.66E-06
DOEOB 5 (abandoned flood plain, active prior to 1941)3B																		
24.4 - 39.6 cm (hand augered)	4/25/2001	0.043	1.19E-06	0.81	0	0.78	0	0.72	0	0.288	0	0.76	0	0.76	0	0.05	5.78E-07	4.59E-06
57.9 - 73.2 cm (hand augered)	4/25/2001	0.036	9.97E-07	0.92	0	0.92	0	0.81	0	0.256	0	0.78	0	0.97	0	0.04	4.51E-07	3.98E-06
116 - 158 cm (hand augered)	4/25/2001	0.043	1.19E-06	1.10	0	1.08	0	0.83	0	0.350	0	0.88	0	1.02	0	0.05	5.78E-07	4.56E-06

[1]Calculated risk - negative or less than E-8 assigned 0 risk value

[2]Risk calculated for reported values or detection limits for measurements reported below sample specific detection, those values identified in Table 2 and risk recalculated

[3]Cells shaded in yellow, 2 to 6 times greater than 1941 to 1968 Santa Clara globbal fallout reference

Table 1. Risk calculated for all radionuclide measurements (continued)

Cañada Ancha Site (slough area during 1940-1958 or 1967) unit 3B		234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
0 - 30.5 cm (hand augered)	**2/16/1999**	0.68	1.69E-07	0.036	1.85E-07	0.69	9.29E-07	0.07	3.03E-07	0.003	0	-0.003	0	0.003	0	0.002	0	0.093	1.56E-06
30.5 - 61 cm (hand augered)	**2/16/1999**	0.69	1.72E-07	0.044	2.26E-07	0.75	1.01E-06	0.29	1.26E-06	-0.005	0	-0.003	0	-0.001	0	0.000	0	0.130	2.18E-06
61 - 91.4 cm (hand augered)	**2/16/1999**	0.76	1.89E-07	0.033	1.69E-07	0.80	1.08E-06	0.36	1.56E-06	0.014	0	0.004	2.8E-08	0.001	0	0.002	0	0.070	1.17E-06
91.4 - 101 cm (hand augered)	**2/16/1999**	1.13	2.81E-07	0.053	2.72E-07	1.06	1.43E-06	0.27	1.17E-06	0.014	0	0.001	0	-0.001	0	0.003	0	0.240	4.02E-06
101 - 110 cm (hand augered)	**2/16/1999**	1.96	4.88E-07	0.126	6.46E-07	1.99	2.68E-06	0.36	1.56E-06	0.026	1.39E-08	0.009	6.56E-08	0.006	0	0.067	2.58E-08	0.600	1.01E-05
110 - 125 cm (hand augered)	**2/12/1999**	1.25	3.11E-07	0.070	3.59E-07	1.20	1.62E-06	0.31	1.34E-06	0.004	0	-0.005	0	0.001	0	0.012	0	0.270	4.52E-06
125 - 137 cm (hand augered)	**2/12/1999**	1.28	3.18E-07	0.071	3.64E-07	1.26	1.7E-06	0.40	1.73E-06	0.016	0	0.014	1.1E-07	0.002	0	0.026	1.01E-08	0.710	1.19E-05
137 - 162 cm (hand augered)	**2/12/1999**	1.41	3.51E-07	0.097	4.97E-07	1.55	2.09E-06	0.64	2.77E-06	0.019	1.02E-08	0.000	0	0.001	0	0.043	1.65E-08	0.980	1.64E-05
162 - 186 cm (hand augered)	**2/12/1999**	1.13	2.81E-07	0.055	2.82E-07	1.33	1.79E-06	0.40	1.73E-06	0.020	1.07E-08	0.006	4.42E-08	-0.002	0	0.066	2.54E-08	0.700	1.17E-05
186 - 213 cm (hand augered)	**4/10/2000**	1.14	2.84E-07	0.065	3.33E-07	1.18	1.59E-06	0.17	7.36E-07	0.001	0	0.004	2.75E-08	0.001	0	0.014	0	0.091	1.52E-06
213 - 229 cm (hand augered)	**4/10/2000**	1.14	2.84E-07	0.065	3.33E-07	1.18	1.59E-06	-0.06	0	0.015	0	-0.001	0	0.001	0	0.034	1.3E-08	0.050	8.38E-07
323 - 335 cm (hand augered)	**4/10/2000**	1.14	2.84E-07	0.065	3.33E-07	1.18	1.59E-06	0.11	4.76E-07	0.000	0	-0.001	0	0.001	0	0.003	0	0.028	4.69E-07

Table 1. Risk calculated for all radionuclide measurements, cumulative risk per horizon in last column (continued)

Cañada Ancha Site (slough area during 1940-1958 or 1967) unit 3B		60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
0 - 30.5 cm (hand augered)	**2/16/1999**	0.13	3.6E-06	0.74	0	0.79	0	0.37	0	0.073	0	0.66	0	1.20	0	0.14	1.62E-06	8.36E-06
30.5 - 61 cm (hand augered)	**2/16/1999**	0.11	3.05E-06	0.84	0	0.86	0	0.79	0	0.120	0	0.95	0	0.84	0	0.13	1.5E-06	9.39E-06
61 - 91.4 cm (hand augered)	**2/16/1999**	0.092	2.55E-06	0.77	0	0.98	0	0.50	0	0.079	0	0.98	0	0.85	0	0.08	8.9E-07	7.63E-06
91.4 - 101 cm (hand augered)	**2/16/1999**	0.14	3.88E-06	0.81	0	0.91	0	0.51	0	0.150	0	0.92	0	1.50	0	0.17	1.97E-06	1.3E-05
101 - 110 cm (hand augered)	**2/16/1999**	0.41	1.14E-05	1.85	0	2.56	0	1.30	0	0.410	0	1.85	0	4.10	0	0.29	3.35E-06	3.02E-05
110 - 125 cm (hand augered)	**2/12/1999**	0.18	4.99E-06	1.33	0	1.21	0	0.95	0	0.460	0	1.21	0	0.98	0	0.15	1.73E-06	1.49E-05
125 - 137 cm (hand augered)	**2/12/1999**	0.088	2.44E-06	1.10	0	1.41	0	1.05	0	0.450	0	1.18	0	1.13	0	0.07	8.32E-07	1.94E-05
137 - 162 cm (hand augered)	**2/12/1999**	0.16	4.43E-06	1.65	0	1.44	0	1.09	0	0.160	0	1.36	0	1.10	0	0.16	1.85E-06	2.84E-05
162 - 186 cm (hand augered)	**2/12/1999**	0.15	4.16E-06	1.42	0	1.24	0	0.93	0	0.600	0	1.32	0	1.50	0	0.08	8.79E-07	2.09E-05
186 - 213 cm (hand augered)	**4/10/2000**	0.041	1.14E-06	0.74	0	1.33	0	1.30	0	0.480	0	1.45	0	1.03	0	0.05	5.2E-07	6.15E-06
213 - 229 cm (hand augered)	**4/10/2000**	0.049	1.36E-06	1.38	0	1.42	0	1.65	0	0.570	0	1.47	0	1.46	0	0.06	6.47E-07	5.06E-06
323 - 335 cm (hand augered)	**4/10/2000**	0.026	7.2E-07	0.74	0	0.65	0	0.54	0	0.202	0	0.64	0	0.63	0	0.03	3.58E-07	4.23E-06

[1]Calculated risk - negative or less than E-8 assigned 0 risk value

[2]Risk calculated for reported values or detection limits for measurements reported below sample specific detection, those values identified in Table 2 and risk recalculated

[3]Cells shaded in yellow, 2 to 6 times greater than 1941 to 1968 Santa Clara globbal fallout reference

Table 1. Risk calculated for all radionuclide measurements (continued)

Pajarito Site (active flood plain and bar surface)1B		234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
0 - 30.5 cm (hand augered)	**9/28/1998**	0.44	1.1E-07	0.020	1.03E-07	0.43	5.81E-07	0.17	7.36E-07	0.006	0	0.002	1.85E-08	-0.003	0	0.003	0	0.059	9.88E-07
30.5 - 45.7 cm (hand augered)	**9/28/1998**	0.73	1.82E-07	0.063	3.23E-07	0.72	9.69E-07	0.16	6.93E-07	0.010	0	0.010	7.69E-08	0.002	0	0.008	0	0.144	2.41E-06
Water Canyon Site (active flood plain and bar surface)1B																			
0 - 30.5 cm (outcrop A)	**9/29/1998**	0.63	1.57E-07	0.042	2.15E-07	0.65	8.75E-07	0.16	6.93E-07	0.003	0	0.001	0	-0.001	0	0.003	0	0.052	8.71E-07
30.5 - 61 cm (outcrop A)	**9/29/1998**	0.60	1.49E-07	0.028	1.44E-07	0.53	7.13E-07	0.16	6.93E-07	0.006	0	-0.003	0	-0.003	0	0.006	0	0.064	1.07E-06
61 - 91.4 cm (outcrop A)	**9/29/1998**	0.73	1.82E-07	0.033	1.69E-07	0.76	1.02E-06	0.17	7.36E-07	0.011	0	0.009	6.92E-08	0.001	0	0.008	0	0.120	2.01E-06
91.4 - 101 cm (outcrop A)	**9/29/1998**	0.82	2.04E-07	0.036	1.85E-07	0.86	1.16E-06	0.15	6.49E-07	0.018	0	-0.001	0	0.003	0	0.008	0	0.132	2.21E-06
101 - 116 cm (outcrop A)	**9/29/1998**	0.72	1.79E-07	0.040	2.05E-07	0.73	9.83E-07	0.16	6.93E-07	0.000	0	-0.002	0	0.002	0	0.002	0	0.064	1.07E-06
91.4 - 101 cm (outcrop B)	**9/29/1998**	0.75	1.87E-07	0.046	2.36E-07	0.85	1.14E-06	0.15	6.49E-07	0.013	0	-0.003	0	0.002	0	0.002	0	0.096	1.61E-06

Table 1. Risk calculated for all radionuclide measurements, cumulative risk per horizon in last column (continued)

Pajarito Site (active flood plain and bar surface)1B		60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
0 - 30.5 cm (hand augered)	**9/28/1998**	0.075	2.08E-06	0.42	0	0.44	0	0.43	0	0.060	0	0.44	0	0.59	0	0.06	6.71E-07	5.29E-06
30.5 - 45.7 cm (hand augered)	**9/28/1998**	0.12	3.32E-06	0.99	0	1.02	0	0.83	0	0.100	0	0.85	0	1.10	0	0.12	1.39E-06	9.37E-06
Water Canyon Site (active flood plain and bar surface)1B																		
0 - 30.5 cm (outcrop A)	**9/29/1998**	0.082	2.27E-06	0.67	0	0.86	0	0.75	0	0.231	0	0.58	0	0.82	0	0.08	9.36E-07	6.02E-06
30.5 - 61 cm (outcrop A)	**9/29/1998**	0.059	1.63E-06	0.57	0	0.49	0	0.24	0	0.222	0	0.67	0	0.73	0	0.07	7.63E-07	5.17E-06
61 - 91.4 cm (outcrop A)	**9/29/1998**	0.097	2.69E-06	0.76	0	0.81	0	0.54	0	0.100	0	0.78	0	0.90	0	0.12	1.39E-06	8.26E-06
91.4 - 101 cm (outcrop A)	**9/29/1998**	0.078	2.16E-06	0.88	0	1.00	0	0.99	0	0.330	0	0.95	0	1.30	0	0.11	1.27E-06	7.84E-06
101 - 116 cm (outcrop A)	**9/29/1998**	0.072	1.99E-06	0.94	0	1.07	0	0.99	0	0.385	0	0.84	0	1.13	0	0.08	9.6E-07	6.09E-06
91.4 - 101 cm (outcrop B)	**9/29/1998**	0.13	3.6E-06	1.02	0	1.18	0	1.08	0	0.091	0	1.06	0	1.40	0	0.12	1.39E-06	8.81E-06

[1]Calculated risk - negative or less than E-8 assigned 0 risk value

[2]Risk calculated for reported values or detection limits for measurements reported below sample specific detection, those values identified in Table 2 and risk recalculated

[3]Cells shaded in yellow, 2 to 6 times greater than 1941 to 1968 Santa Clara globbal fallout reference

Table 1. Risk calculated for all radionuclide measurements (continued)

Frijoles Site (pre-1950 flood-plain deposits and reservoir sedimentation)		234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
0 - 30.5 cm (hand augered)	**9/30/1998**	1.22	3.03E-07	0.075	3.85E-07	1.12	1.51E-06	0.15	6.49E-07	0.018	0	-0.001	0	-0.001	0	0.022	0	0.128	2.14E-06
30.5 - 61 cm (hand augered)	**9/30/1998**	1.31	3.26E-07	0.091	4.67E-07	1.28	1.72E-06	0.23	9.96E-07	0.022	1.18E-08	0.007	5.38E-08	0.003	0	0.018	0	0.301	5.04E-06
61 - 91.4 cm (hand augered)	**9/30/1998**	0.63	1.57E-07	0.057	2.92E-07	0.71	9.56E-07	0.22	9.52E-07	0.006	0	-0.001	0	0.001	0	0.009	0	0.098	1.64E-06
91.4 - 122 cm (hand augered)	**9/30/1998**	0.81	2.01E-07	0.027	1.38E-07	0.68	9.15E-07	0.21	9.09E-07	0.004	0	0.003	2.15E-08	0.002	0	0.005	0	0.101	1.69E-06
122 - 152 cm (hand augered)	**9/30/1998**	1.25	3.11E-07	0.083	4.26E-07	1.26	1.7E-06	0.20	8.66E-07	0.009	0	0.000	0	0.003	0	0.016	0	0.172	2.88E-06
152 - 183 cm (hand augered)	**9/30/1998**	1.12	2.79E-07	0.040	2.05E-07	1.07	1.44E-06	0.23	9.96E-07	0.011	0	-0.004	0	0.001	0	0.008	0	0.220	3.69E-06

Table 1. Risk calculated for all radionuclide measurements, cumulative risk per horizon in last column (continued)

Frijoles Site (pre-1950 flood-plain deposits and reservoir sedimentation)		60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
0 - 30.5 cm (hand augered)	**9/30/1998**	0.1	2.77E-06	1.29	0	1.16	0	1.22	0	0.440	0	1.11	0	0.84	0	0.13	1.5E-06	9.26E-06
30.5 - 61 cm (hand augered)	**9/30/1998**	0.085	2.35E-06	1.27	0	0.99	0	1.12	0	0.380	0	1.01	0	1.44	0	0.09	9.94E-07	1.20E-05
61 - 91.4 cm (hand augered)	**9/30/1998**	0.063	1.75E-06	0.98	0	0.89	0	0.85	0	0.259	0	0.77	0	1.09	0	0.10	1.16E-06	6.90E-06
91.4 - 122 cm (hand augered)	**9/30/1998**	0.091	2.52E-06	0.86	0	0.90	0	0.33	0	0.078	0	0.64	0	0.93	0	0.13	1.5E-06	7.90E-06
122 - 152 cm (hand augered)	**9/30/1998**	0.076	2.11E-06	1.23	0	1.15	0	0.87	0	0.310	0	0.95	0	1.18	0	0.07	8.32E-07	9.12E-06
152 - 183 cm (hand augered)	**9/30/1998**	0.16	4.43E-06	1.09	0	1.00	0	0.99	0	0.410	0	1.11	0	1.40	0	0.14	1.62E-06	1.27E-05

[1]Calculated risk - negative or less than E-8 assigned 0 risk value

[2]Risk calculated for reported values or detection limits for measurements reported below sample specific detection, those values identified in Table 2 and risk recalculated

[3]Cells shaded in yellow, 2 to 6 times greater than 1941 to 1968 Santa Clara globbal fallout reference

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level

Station Description	Date Collected	234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
Santa Clara Sites																			
DOEOB 3 (abandoned flood plain, active 1941-1968)2B																			
5.18 - 30.5 cm (hand augered)	4/24/2001	0.90	2.24E-07	0.039	2.00E-07	0.68	9.15E-07	-0.01	RV BDL	0.003	RV BDL	0.001	RV BDL	0.0006	RV BDL	0.0051	0.00E+00	0.102	1.71E-06
DOEOB 4 (abandoned flood plain, active prior to 1941)3B																			
33.5 - 39.6 cm (hand augered)	4/24/2001	0.90	2.24E-07	0.044	2.26E-07	0.91	1.22E-06	0.12	RV BDL	0.003	RV BDL	0.007	RV BDL	-0.0016	RV BDL	0.0008	RV BDL	0.036	RV BDL
73.2 - 88.4 cm (hand augered)	4/24/2001	0.38	9.45E-08	0.017	8.72E-08	0.39	5.22E-07	0.22	RV BDL	0.010	RV BDL	0.006	RV BDL	0.0010	RV BDL	-0.0006	RV BDL	0.027	RV BDL
DOEOB 5 (abandoned flood plain, active prior to 1941)3B																			
24.4 - 39.6 cm (hand augered)	4/25/2001	1.14	2.84E-07	0.060	3.08E-07	0.98	1.32E-06	0.07	RV BDL	0.008	RV BDL	-0.001	RV BDL	0.0007	RV BDL	-0.0002	RV BDL	0.036	RV BDL
57.9 - 73.2 cm (hand augered)	4/25/2001	0.86	2.14E-07	0.059	3.03E-07	0.86	1.16E-06	0.07	RV BDL	0.006	RV BDL	0.001	RV BDL	0.0000	RV BDL	0.0006	RV BDL	0.033	RV BDL
116 - 158 cm (hand augered)	4/25/2001	1.15	2.86E-07	0.053	2.72E-07	1.09	1.47E-06	0.01	RV BDL	-0.003	RV BDL	0.005	RV BDL	0.0000	RV BDL	0.0000	RV BDL	0.041	RV BDL

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level and cumulative risk in last column

Station Description	Date Collected	60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Total Risk
Santa Clara Sites																		
DOEOB 3 (abandoned flood plain, active 1941-1968)2B																		
5.18 - 30.5 cm (hand augered)	4/24/2001	0.05	RV BDL	0.86	0.00E+00	0.83	0.00E+00	0.64	0.00E+00	0.26	0.00E+00	0.77	0.00E+00	0.79	0.00E+00	0.048	RV BDL	3.05E-06
DOEOB 4 (abandoned flood plain, active prior to 1941)3B																		
33.5 - 39.6 cm (hand augered)	4/24/2001	0.04	RV BDL	1.01	0.00E+00	1.01	0.00E+00	0.86	0.00E+00	0.32	0.00E+00	0.91	0.00E+00	1.10	0.00E+00	0.043	RV BDL	1.67E-06
73.2 - 88.4 cm (hand augered)	4/24/2001	0.04	RV BDL	0.47	0.00E+00	0.43	0.00E+00	0.40	0.00E+00	0.15	0.00E+00	0.41	0.00E+00	0.44	0.00E+00	0.042	RV BDL	7.04E-07
DOEOB 5 (abandoned flood plain, active prior to 1941)3B																		
24.4 - 39.6 cm (hand augered)	4/25/2001	0.04	RV BDL	0.81	0.00E+00	0.78	0.00E+00	0.72	0.00E+00	0.29	0.00E+00	0.76	0.00E+00	0.76	0.00E+00	0.050	RV BDL	1.91E-06
57.9 - 73.2 cm (hand augered)	4/25/2001	0.04	RV BDL	0.92	0.00E+00	0.92	0.00E+00	0.81	0.00E+00	0.26	0.00E+00	0.78	0.00E+00	0.97	0.00E+00	0.039	RV BDL	1.67E-06
116 - 158 cm (hand augered)	4/25/2001	0.04	RV BDL	1.10	0.00E+00	1.08	0.00E+00	0.83	0.00E+00	0.35	0.00E+00	0.88	0.00E+00	1.02	0.00E+00	0.050	RV BDL	2.02E-06

RV BDL = Reported value below its sample specific detection limit

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level (continued)

Station Description	Date Collected	234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
Cañada Ancha Site																			
(slough area during 1940-1958 or 1967) unit 3B																			
0 - 30.5 cm (hand augered)	2/16/1999	0.68	1.69E-07	0.036	1.85E-07	0.69	9.29E-07	0.07	RV BDL	0.003	RV BDL	-0.003	RV BDL	0.0029	RV BDL	0.0022	RV BDL	0.093	RV BDL
30.5 - 61 cm (hand augered)	2/16/1999	0.69	1.72E-07	0.044	2.26E-07	0.75	1.01E-06	0.29	1.26E-06	-0.005	RV BDL	-0.003	RV BDL	-0.0013	RV BDL	0.0000	RV BDL	0.130	RV BDL
61 - 91.4 cm (hand augered)	2/16/1999	0.76	1.89E-07	0.033	1.69E-07	0.80	1.08E-06	0.36	1.56E-06	0.014	RV BDL	0.004	RV BDL	0.0007	RV BDL	0.0017	0.00E+00	0.070	RV BDL
91.4 - 101 cm (hand augered)	2/16/1999	1.13	2.81E-07	0.053	2.72E-07	1.06	1.43E-06	0.27	1.17E-06	0.014	RV BDL	0.001	RV BDL	-0.0007	RV BDL	0.0033	RV BDL	0.240	4.02E-06
101 - 110 cm (hand augered)	2/16/1999	1.96	4.88E-07	0.126	6.46E-07	1.99	2.68E-06	0.36	1.56E-06	0.026	1.39E-08	0.009	RV BDL	0.0056	0.00E+00	0.0670	2.58E-08	0.600	1.01E-05
110 - 125 cm (hand augered)	2/12/1999	1.25	3.11E-07	0.070	3.59E-07	1.20	1.62E-06	0.31	1.34E-06	0.004	RV BDL	-0.005	RV BDL	0.0013	RV BDL	0.0121	0.00E+00	0.270	4.52E-06
125 - 137 cm (hand augered)	2/12/1999	1.28	3.18E-07	0.071	3.64E-07	1.26	1.70E-06	0.40	1.73E-06	0.016	0.00E+00	0.014	RV BDL	0.0018	0.00E+00	0.0262	1.01E-08	0.710	1.19E-05
137 - 162 cm (hand augered)	2/12/1999	1.41	3.51E-07	0.097	4.97E-07	1.55	2.09E-06	0.64	2.77E-06	0.019	1.02E-08	0.000	RV BDL	0.0007	RV BDL	0.0430	1.65E-08	0.980	1.64E-05
162 - 186 cm (hand augered)	2/12/1999	1.13	2.81E-07	0.055	2.82E-07	1.33	1.79E-06	0.40	1.73E-06	0.020	1.07E-08	0.006	RV BDL	-0.0020	RV BDL	0.0660	2.54E-08	0.700	1.17E-05
186 - 213 cm (hand augered)	4/10/2000	1.14	2.84E-07	0.065	3.33E-07	1.18	1.59E-06	0.17	RV BDL	0.001	RV BDL	0.004	RV BDL	0.0008	0.00E+00	0.0144	0.00E+00	0.091	1.52E-06
213 - 229 cm (hand augered)	4/10/2000	1.14	2.84E-07	0.065	3.33E-07	1.18	1.59E-06	-0.06	RV BDL	0.015	0.00E+00	-0.001	RV BDL	0.0012	0.00E+00	0.0337	1.30E-08	0.050	0.00E+00
323 - 335 cm (hand augered)	4/10/2000	1.14	2.84E-07	0.065	3.33E-07	1.18	1.59E-06	0.11	RV BDL	0.000	RV BDL	-0.001	RV BDL	0.0013	0.00E+00	0.0029	0.00E+00	0.028	0.00E+00

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level and cumulative risk in last column

Station Description	Date Collected	60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
Cañada Ancha Site																		
(slough area during 1940-1958 or 1967) unit 3B																		
0 - 30.5 cm (hand augered)	2/16/1999	0.13	RV BDL	0.74	0.00E+00	0.79	0.00E+00	0.37	RV BDL	0.07	RV BDL	0.66	0.00E+00	1.20	0.00E+00	0.140	RV BDL	1.28E-06
30.5 - 61 cm (hand augered)	2/16/1999	0.11	RV BDL	0.84	0.00E+00	0.86	0.00E+00	0.79	0.00E+00	0.12	RV BDL	0.95	0.00E+00	0.84	0.00E+00	0.130	RV BDL	2.66E-06
61 - 91.4 cm (hand augered)	2/16/1999	0.09	RV BDL	0.77	0.00E+00	0.98	0.00E+00	0.50	0.00E+00	0.08	RV BDL	0.98	0.00E+00	0.85	0.00E+00	0.077	RV BDL	2.99E-06
91.4 - 101 cm (hand augered)	2/16/1999	0.14	RV BDL	0.81	0.00E+00	0.91	0.00E+00	0.51	RV BDL	0.15	RV BDL	0.92	0.00E+00	1.50	RV BDL	0.170	RV BDL	7.17E-06
101 - 110 cm (hand augered)	2/16/1999	0.41	RV BDL	1.85	0.00E+00	2.56	0.00E+00	1.30	RV BDL	0.41	0.00E+00	1.85	0.00E+00	4.10	RV BDL	0.290	RV BDL	1.55E-05
110 - 125 cm (hand augered)	2/12/1999	0.18	RV BDL	1.33	0.00E+00	1.21	0.00E+00	0.95	0.00E+00	0.46	0.00E+00	1.21	0.00E+00	0.98	RV BDL	0.150	RV BDL	8.15E-06
125 - 137 cm (hand augered)	2/12/1999	0.09	RV BDL	1.10	0.00E+00	1.41	0.00E+00	1.05	0.00E+00	0.45	0.00E+00	1.18	0.00E+00	1.13	0.00E+00	0.072	RV BDL	1.60E-05
137 - 162 cm (hand augered)	2/12/1999	0.16	RV BDL	1.65	0.00E+00	1.44	0.00E+00	1.09	0.00E+00	0.16	RV BDL	1.36	0.00E+00	1.10	0.00E+00	0.160	RV BDL	2.21E-05
162 - 186 cm (hand augered)	2/12/1999	0.15	RV BDL	1.42	0.00E+00	1.24	0.00E+00	0.93	0.00E+00	0.60	0.00E+00	1.32	0.00E+00	1.50	0.00E+00	0.076	RV BDL	1.58E-05
186 - 213 cm (hand augered)	4/10/2000	0.04	RV BDL	0.74	0.00E+00	1.33	0.00E+00	1.30	0.00E+00	0.48	0.00E+00	1.45	0.00E+00	1.03	0.00E+00	0.045	RV BDL	3.73E-06
213 - 229 cm (hand augered)	4/10/2000	0.05	RV BDL	1.38	0.00E+00	1.42	0.00E+00	1.65	0.00E+00	0.57	0.00E+00	1.47	0.00E+00	1.46	0.00E+00	0.056	RV BDL	2.22E-06
323 - 335 cm (hand augered)	4/10/2000	0.03	RV BDL	0.74	0.00E+00	0.65	0.00E+00	0.54	0.00E+00	0.20	0.00E+00	0.64	0.00E+00	0.63	0.00E+00	0.031	RV BDL	2.21E-06

RV BDL = Reported value below its sample specific detection limit

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level (continued)

Station Description	Date Collected	234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
Pajarito Site																			
(active flood plain and bar surface)1B																			
0 - 30.5 cm (hand augered)	9/28/1998	0.44	1.10E-07	0.020	RV BDL	0.43	5.81E-07	0.17	7.36E-07	0.006	0.00E+00	0.002	RV BDL	-0.0025	RV BDL	0.0029	RV BDL	0.059	RV BDL
30.5 - 45.7 cm (hand augered)	9/28/1998	0.73	1.82E-07	0.063	3.23E-07	0.72	9.69E-07	0.16	6.93E-07	0.010	0.00E+00	0.010	RV BDL	0.0017	RV BDL	0.0076	0.00E+00	0.144	2.41E-06
Water Canyon Site																			
(active flood plain and bar surface)1B																			
0 - 30.5 cm (outcrop A)	9/29/1998	0.63	1.57E-07	0.042	2.15E-07	0.65	8.75E-07	0.16	RV BDL	0.003	RV BDL	0.001	RV BDL	-0.0013	RV BDL	0.0032	RV BDL	0.052	8.71E-07
30.5 - 61 cm (outcrop A)	9/29/1998	0.60	1.49E-07	0.028	1.44E-07	0.53	7.13E-07	0.16	RV BDL	0.006	0.00E+00	-0.003	RV BDL	-0.0032	RV BDL	0.0061	RV BDL	0.064	RV BDL
61 - 91.4 cm (outcrop A)	9/29/1998	0.73	1.82E-07	0.033	1.69E-07	0.76	1.02E-06	0.17	RV BDL	0.011	RV BDL	0.009	RV BDL	0.0006	RV BDL	0.0082	RV BDL	0.120	RV BDL
91.4 - 101 cm (outcrop A)	9/29/1998	0.82	2.04E-07	0.036	1.85E-07	0.86	1.16E-06	0.15	RV BDL	0.018	0.00E+00	-0.001	RV BDL	0.0025	RV BDL	0.0080	0.00E+00	0.132	2.21E-06
101 - 116 cm (outcrop A)	9/29/1998	0.72	1.79E-07	0.040	2.05E-07	0.73	9.83E-07	0.16	RV BDL	0.000	RV BDL	-0.002	RV BDL	0.0015	RV BDL	0.0021	RV BDL	0.064	RV BDL
91.4 - 101 cm (outcrop B)	9/29/1998	0.75	1.87E-07	0.046	2.36E-07	0.85	1.14E-06	0.15	RV BDL	0.013	0.00E+00	-0.003	RV BDL	0.0015	RV BDL	0.0017	RV BDL	0.096	RV BDL

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level and cumulative risk in last column

Station Description	Date Collected	60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
Pajarito Site																		
(active flood plain and bar surface)1B																		
0 - 30.5 cm (hand augered)	9/28/1998	0.08	RV BDL	0.42	0.00E+00	0.44	0.00E+00	0.43	0.00E+00	0.06	RV BDL	0.44	0.00E+00	0.59	RV BDL	0.058	RV BDL	1.43E-06
30.5 - 45.7 cm (hand augered)	9/28/1998	0.12	RV BDL	0.99	0.00E+00	1.02	0.00E+00	0.83	0.00E+00	0.10	RV BDL	0.85	0.00E+00	1.10	0.00E+00	0.120	RV BDL	4.58E-06
Water Canyon Site																		
(active flood plain and bar surface)1B																		
0 - 30.5 cm (outcrop A)	9/29/1998	0.08	RV BDL	0.67	0.00E+00	0.86	0.00E+00	0.75	0.00E+00	0.23	0.00E+00	0.58	0.00E+00	0.82	0.00E+00	0.081	RV BDL	2.12E-06
30.5 - 61 cm (outcrop A)	9/29/1998	0.06	RV BDL	0.57	0.00E+00	0.49	0.00E+00	0.24	RV BDL	0.22	0.00E+00	0.67	0.00E+00	0.73	RV BDL	0.066	RV BDL	1.01E-06
61 - 91.4 cm (outcrop A)	9/29/1998	0.10	RV BDL	0.76	0.00E+00	0.81	0.00E+00	0.54	RV BDL	0.10	RV BDL	0.78	0.00E+00	0.90	0.00E+00	0.120	RV BDL	1.37E-06
91.4 - 101 cm (outcrop A)	9/29/1998	0.08	RV BDL	0.88	0.00E+00	1.00	0.00E+00	0.99	0.00E+00	0.33	0.00E+00	0.95	0.00E+00	1.30	0.00E+00	0.110	RV BDL	3.76E-06
101 - 116 cm (outcrop A)	9/29/1998	0.07	RV BDL	0.94	0.00E+00	1.07	0.00E+00	0.99	0.00E+00	0.39	0.00E+00	0.84	0.00E+00	1.13	0.00E+00	0.083	RV BDL	1.37E-06
91.4 - 101 cm (outcrop B)	9/29/1998	0.13	RV BDL	1.02	0.00E+00	1.18	0.00E+00	1.08	0.00E+00	0.09	RV BDL	1.06	0.00E+00	1.40	0.00E+00	0.120	RV BDL	1.57E-06

RV BDL = Reported value below its sample specific detection limit

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level (continued)

Station Description	Date Collected	234U (pCi/g)	RISK	235U (pCi/g)	RISK	238U (pCi/g)	RISK	90Sr (pCi/g)	RISK	241Am (pCi/g)	RISK	237Np (pCi/g)	RISK	^{238}Pu (pCi/g)	RISK	$^{239/240}$Pu (pCi/g)	RISK	137Cs (pCi/g)	RISK
Frijoles Site																			
(pre-1950 flood-plain deposits and reservoir sedimentation)																			
0 - 30.5 cm (hand augered)	9/30/1998	1.22	3.03E-07	0.075	3.85E-07	1.12	1.51E-06	0.15	RV BDL	0.018	0.00E+00	-0.001	RV BDL	-0.0012	RV BDL	0.0217	0.00E+00	0.128	2.14E-06
30.5 - 61 cm (hand augered)	9/30/1998	1.31	3.26E-07	0.091	4.67E-07	1.28	1.72E-06	0.23	RV BDL	0.022	1.18E-08	0.007	RV BDL	0.0026	RV BDL	0.0179	0.00E+00	0.301	5.04E-06
61 - 91.4 cm (hand augered)	9/30/1998	0.63	1.57E-07	0.057	2.92E-07	0.71	9.56E-07	0.22	RV BDL	0.006	RV BDL	-0.001	RV BDL	0.0005	RV BDL	0.0091	0.00E+00	0.098	1.64E-06
91.4 - 122 cm (hand augered)	9/30/1998	0.81	2.01E-07	0.027	RV BDL	0.68	9.15E-07	0.21	RV BDL	0.004	RV BDL	0.003	RV BDL	0.0018	RV BDL	0.0053	0.00E+00	0.101	1.69E-06
122 - 152 cm (hand augered)	9/30/1998	1.25	3.11E-07	0.083	4.26E-07	1.26	1.70E-06	0.20	RV BDL	0.009	RV BDL	0.000	RV BDL	0.0033	RV BDL	0.0162	0.00E+00	0.172	2.88E-06
152 - 183 cm (hand augered)	9/30/1998	1.12	2.79E-07	0.040	2.05E-07	1.07	1.44E-06	0.23	RV BDL	0.011	0.00E+00	-0.004	RV BDL	0.0005	RV BDL	0.0082	0.00E+00	0.220	3.69E-06

Table 2. Calculated Risk for radionuclides measured above their sample specific detection level and cumulative risk in last column

Station Description	Date Collected	60Co (pCi/g)	RISK	212Pb (pCi/g)	RISK	214Pb (pCi/g)	RISK	228Ac (pCi/g)	RISK	208Tl (pCi/g)	RISK	214Bi (pCi/g)	RISK	212Bi (pCi/g)	RISK	22Na (pCi/g)	RISK	Cumulative Risk
Frijoles Site																		
(pre-1950 flood-plain deposits and reservoir sedimentation)																		
0 - 30.5 cm (hand augered)	9/30/1998	0.1	RV BDL	1.29	0.00E+00	1.16	0.00E+00	1.22	0.00E+00	0.440	0.00E+00	1.110	0.00E+00	0.8400	RV BDL	0.1300	RV BDL	4.34E-06
30.5 - 61 cm (hand augered)	9/30/1998	0.085	RV BDL	1.27	0.00E+00	0.99	0.00E+00	1.12	0.00E+00	0.380	0.00E+00	1.010	0.00E+00	1.4400	0.00E+00	0.0860	RV BDL	7.57E-06
61 - 91.4 cm (hand augered)	9/30/1998	0.063	RV BDL	0.98	0.00E+00	0.89	0.00E+00	0.85	0.00E+00	0.259	0.00E+00	0.770	0.00E+00	1.0900	0.00E+00	0.1000	RV BDL	3.05E-06
91.4 - 122 cm (hand augered)	9/30/1998	0.091	RV BDL	0.86	0.00E+00	0.9	0.00E+00	0.33	RV BDL	0.078	RV BDL	0.640	0.00E+00	0.9300	RV BDL	0.1300	RV BDL	2.81E-06
122 - 152 cm (hand augered)	9/30/1998	0.076	RV BDL	1.23	0.00E+00	1.15	0.00E+00	0.87	0.00E+00	0.310	0.00E+00	0.950	0.00E+00	1.1800	0.00E+00	0.0720	RV BDL	5.31E-06
152 - 183 cm (hand augered)	9/30/1998	0.16	RV BDL	1.09	0.00E+00	1	0.00E+00	0.99	0.00E+00	0.410	0.00E+00	1.110	0.00E+00	1.4000	0.00E+00	0.1400	RV BDL	5.61E-06

RV BDL = Reported value below its sample specific detection limit