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Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley

Los Alamos
NATIONAL LABORATORY

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PREFACE

This study documents the extent of plutonium and uranium releases to the Rio Grande and tributary streams after 50-plus years of operations at the Los Alamos National Laboratory, through 1998. The impacts reflect the intermittent but somewhat steady downstream movement of LANL-derived plutonium and uranium in sediments by relatively small-magnitude flood flows.

During the summer of 2000, the hydrologic conditions in the Los Alamos area were dramatically altered following the Cerro Grande wildfire that burned approximately 43,000 acres, principally in the hillsides above Los Alamos and the Laboratory. With the burning of the grasses, brush, and soils, post-fire runoff magnitudes and frequencies were significantly increased.

What effect these hydrologic changes will have on offsite movement of radionuclides is uncertain. This report describes pre-fire conditions against which we can gauge the impacts of the Cerro Grande fire.

Plutonium and Uranium from Los Alamos National Laboratory in Sediments of the Northern Rio Grande Valley

by

Bruce M. Gallaher and Deward W. Efurd

ABSTRACT

This study was undertaken during 1991–1998 to identify the origin of plutonium and uranium in northern New Mexico Rio Grande and tributary stream sediments. Isotopic fingerprinting techniques help distinguish radioactivity from Los Alamos National Laboratory (LANL) and from global fallout or natural sources. The geographic area covered by the study extended from the headwaters of the Rio Grande in southern Colorado to Elephant Butte Reservoir in southern New Mexico. Over 100 samples of stream channel and reservoir bottom sediments were analyzed for the atom ratios of plutonium and uranium isotopes using thermal ionization mass spectrometry. Comparison of these ratios against those for fallout or natural sources allowed for quantification of the Laboratory impact. We also reviewed several decades of historical monitoring results for evidence of LANL impacts.

Of the seven major drainages crossing LANL, movement of LANL plutonium into the Rio Grande can be traced only via Los Alamos Canyon. The majority of sampled locations within and adjacent to LANL have little or no input of plutonium from the Laboratory. Samples collected upstream and distant to LANL show an average fallout $^{240}Pu/^{239}Pu$ atom ratio of 0.169 ± 0.012, which is consistent with published worldwide global fallout values. These regional background ratios differ significantly from the $^{240}Pu/^{239}Pu$ atom ratio of 0.015 that is representative of LANL-derived plutonium entering the Rio Grande at Los Alamos Canyon. Mixing calculations of these sources indicate that the largest proportion (60% to 90%) of the plutonium in the Rio Grande sediments is from global atmospheric fallout, with an average of 22% ± 19% from the Laboratory. These results compare favorably with an earlier geology-based study that concluded 90% of the plutonium was fallout-derived. The LANL plutonium is identifiable intermittently along the 35-km reach of the Rio Grande to Cochiti Reservoir. The source of the LANL-derived plutonium in the Rio Grande was traced primarily to pre-1960 discharges of liquid effluents into a canyon bottom at a distance approximately 20 km upstream of the river. Thus, only early LANL operations resulted in measurable releases of plutonium to streams offsite. Plutonium levels decline exponentially with distance downstream after mixing with cleaner sediments, yet the LANL isotopic fingerprint remains distinct for at least 55 km from the effluent source.

Levels of $^{239,240}Pu$ in sediments near the contaminant source are more than 200 times above background levels and decline to about 10 times above background levels near the entry with the Rio Grande. Activities of $^{239,240}Pu$ within this sample set ranged from 0.001 to 0.046 pCi/g in the Rio Grande to 3.7 pCi/g near the effluent discharge point. Levels in the Rio Grande are usually more than 1000 times lower than Environmental Protection Agency cleanup levels.

Uranium in stream and reservoir sediments is predominantly within natural concentration ranges and is of natural uranium isotopic composition. None of the sediments from the Rio Grande show identifiable Laboratory uranium, using the isotopic ratios. Historical monitoring records, however, indicate that uranium concentrations in the Rio Grande sediments increase by about one-third along a 10-km reach below the confluence with Los Alamos Canyon. These records suggest a slight LANL impact, although the concentrations remain within background levels for the region. Natural variability caused by differences in texture or mineralization also could be factors.

INTRODUCTION

Los Alamos National Laboratory (LANL or the Laboratory) is a major research and development facility located in northern New Mexico within the Rio Grande drainage system. Established in 1943, the Laboratory was a part of the Manhattan Project, and its original mission was to design, develop, and test nuclear weapons. Currently, the Laboratory's program is multidisciplinary (including initiatives in the areas of health, national infrastructure, energy, education, and the environment) with the central mission of reducing the global nuclear danger (ESP 1999).

During the 50-plus years of research operations, some of the canyons draining the Laboratory received varying amounts of radioactive and nonradioactive waste materials, particularly from untreated liquid effluents during the 1940s (DOE 1981). Owing to these releases, some canyon-bottom sediments and surface waters within the Laboratory contain elevated levels of plutonium ($^{239,240}Pu$) and total uranium (ESP 1999). A relatively small amount of these materials has been carried offsite into the Rio Grande by floods. The Rio Grande is the master stream in the area and flows from its headwaters in the San Juan Mountains in southwestern Colorado, across the center of New Mexico, to El Paso, Texas (Figure 1).

While there is a considerable amount of data on radionuclide concentrations in the Los Alamos area, the source(s) of the radionuclides may not be clear, particularly at locations distant from LANL. Radionuclides from the LANL sources could be mixed with radionuclides derived from fallout or natural sources. Uranium is abundant naturally in soils and waters, and plutonium and other radionuclides have been deposited on the landscape worldwide via global fallout and from satellite re-entry and burnup in the earth's atmosphere (Krey 1967). As an added complication, previous measurements show that background concentrations of soil plutonium from fallout may range as much as 100-fold because of variation in precipitation, soil texture, and geographical location relative to cloud paths from the Nevada Test Site. These factors often make it difficult to establish a baseline to compare values against (Purtymun et al. 1990; Krey et al. 1990; Gallaher et al. 1997).

This report presents the results of a survey initiated to identify the likely source(s) of plutonium and uranium within the northern Rio Grande drainage system. Using analytical "fingerprinting" techniques, the Laboratory contribution of these contaminants was measured in river sediments and canyons in the vicinity of the Laboratory. This approach has been successfully used to evaluate radionuclide sources in the Columbia River (Beasley et al. 1981), the Savannah River (Alberts et al. 1986), the Arctic Ocean (Efurd and Rokop 1997; Cooper et al. 1998), and near nuclear complexes at the Rocky Flats Facility in Colorado (Krey 1976; Krey and Krajewski 1972; Krey and Hardy 1970; Ibrahim et al. 1997; Efurd et al. 1995, 1994, 1993), the former Soviet Union (Beasley et al. 1998), and Los Alamos (Gallaher et al. 1999, 1997).

The specific objectives of this investigation were to

- establish the isotopic signatures of known Laboratory sources for ^{235}U and ^{238}U and ^{239}Pu and ^{240}Pu,
- determine background isotopic signatures for ^{239}Pu and ^{240}Pu derived from global fallout deposited in northern New Mexico,
- conduct sampling of the Laboratory and beyond to track the movement of Laboratory-derived plutonium and uranium, and
- quantify the contribution of Laboratory sources of plutonium and uranium throughout the Rio Grande drainage system in New Mexico.

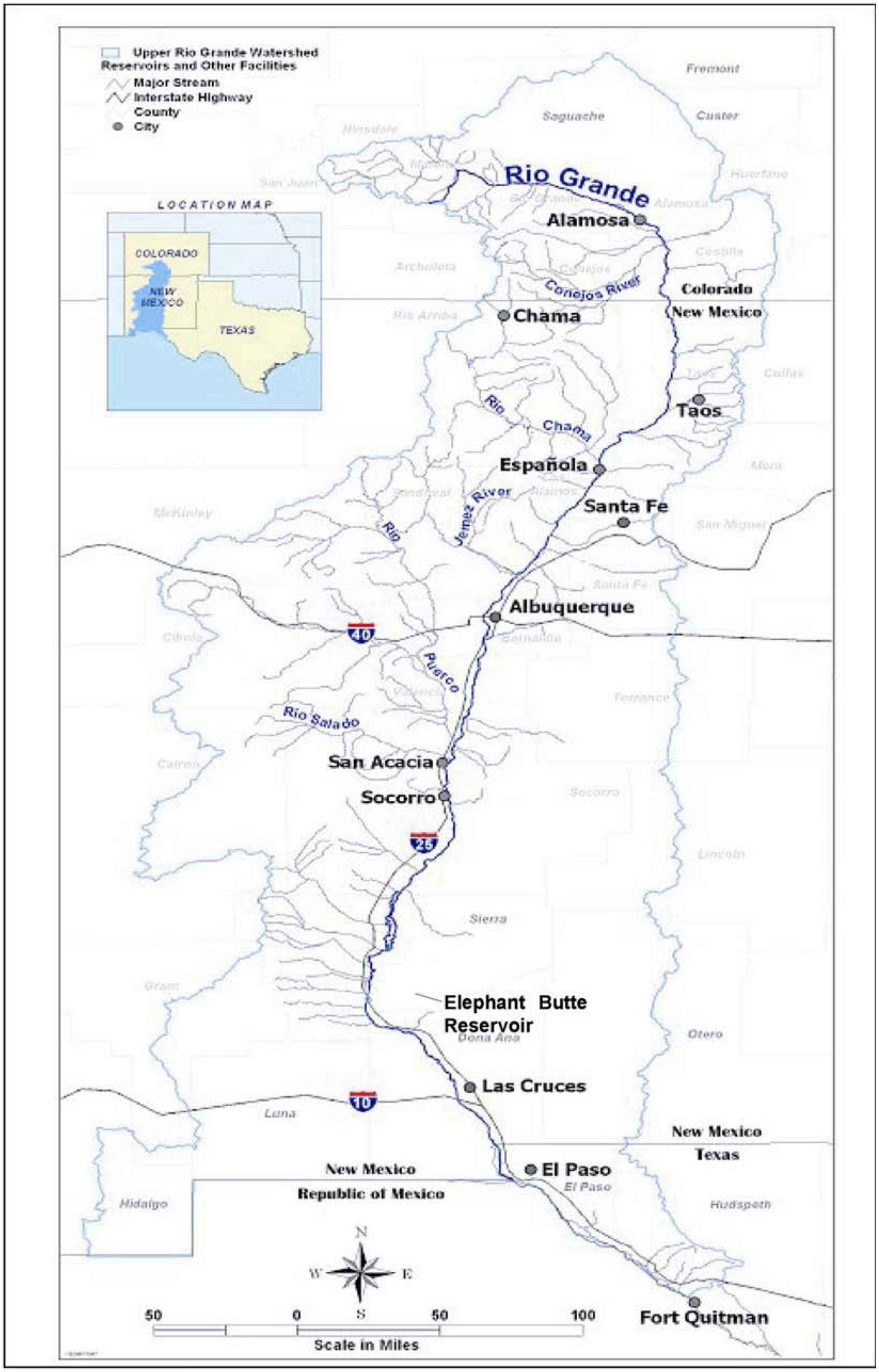


Figure 1. Rio Grande drainage map.

SETTING AND HISTORY

THE PAJARITO PLATEAU

LANL is located in northern New Mexico, approximately 20 miles northwest of Santa Fe (Figure 2). The 43-square-mile (110 km^2) Laboratory is situated on the Pajarito Plateau, a broad, dissected apron of Bandelier Tuff, gradually sloping from the Jemez Mountains caldera eastward toward the Rio Grande and the Rio Chama. Erosion by intermittent streams has cut deep east-to-west-oriented canyons into the relatively smooth surface of the plateau. Weathering and erosion of the Bandelier Tuff produce mostly sand to gravel sized particles that are transported in the region's stream and river systems (Graf 1994; Nyhan et al. 1976).

Most Laboratory and community developments are on the finger-like mesa tops. The surrounding land is largely undeveloped, and large tracts of land north, west, and south of the Laboratory site are held by the Santa Fe National Forest, Bureau of Land Management, General Services Administration, and Los Alamos County. The Pueblo of San Ildefonso borders the Laboratory to the east.

NUCLEAR MATERIALS AT LOS ALAMOS

The Laboratory was established in 1943 as a part of the Manhattan Project, the effort to construct the first atomic weapons. From those early days, much of the research at the Laboratory centered on the fissionable isotopes ^{235}U and ^{239}Pu because they would make up the cores of the first nuclear weapons. Except for small experimental quantities, these materials were shipped to Los Alamos from other industrial plants across the country (DOE 1997).

The Oak Ridge, Tennessee, plants supplied most of the ^{235}U. Uranium-235 occurs naturally within the earth's crust, but only makes up 0.72 percent by weight of natural uranium. The Oak Ridge facilities selectively isolated (that is, enriched) ^{235}U from the other uranium isotopes by diffusion and electromagnetic processes. Oak Ridge also supplied Los Alamos with large quantities of depleted uranium, uranium left over from the enrichment process that has less ^{235}U than natural uranium. Depleted uranium is used in testing at Laboratory explosives firing sites. Before quantities of enriched and depleted uranium were ample to meet the Laboratory's research needs, natural uranium also was used in early Los Alamos research.

The ^{239}Pu used at Los Alamos through the early 1960s was primarily produced at the reactors at Hanford, Washington. In the 1970s the source of ^{239}Pu changed to the reactors at Savannah River, South Carolina. Plutonium-239 is created in a nuclear reactor by adding neutrons to ^{238}U. For the purposes of this study, we will refer to the Hanford plutonium as "pre-1960s plutonium" and the rest as "modern plutonium."

Laboratory-derived plutonium can be distinguished from fallout by variations in its isotopic compositions. Uranium discharged by the Laboratory is identifiable by two separate methods: The enriched uranium and depleted uranium are distinguished by isotopic composition, having artificially different proportions of ^{235}U than natural uranium. Laboratory-derived natural uranium is only distinguishable from "background" or "native" uranium by looking for anomalous concentration patterns.

MAJOR RADIOACTIVE RELEASE AREAS

Effluent Discharges

Historically there have been three principal radioactive liquid effluent discharge areas at Los Alamos. Two of these are located within the Pueblo/Los Alamos Canyon watershed near the northern boundary of the Laboratory. The other discharge area is in Mortandad Canyon within

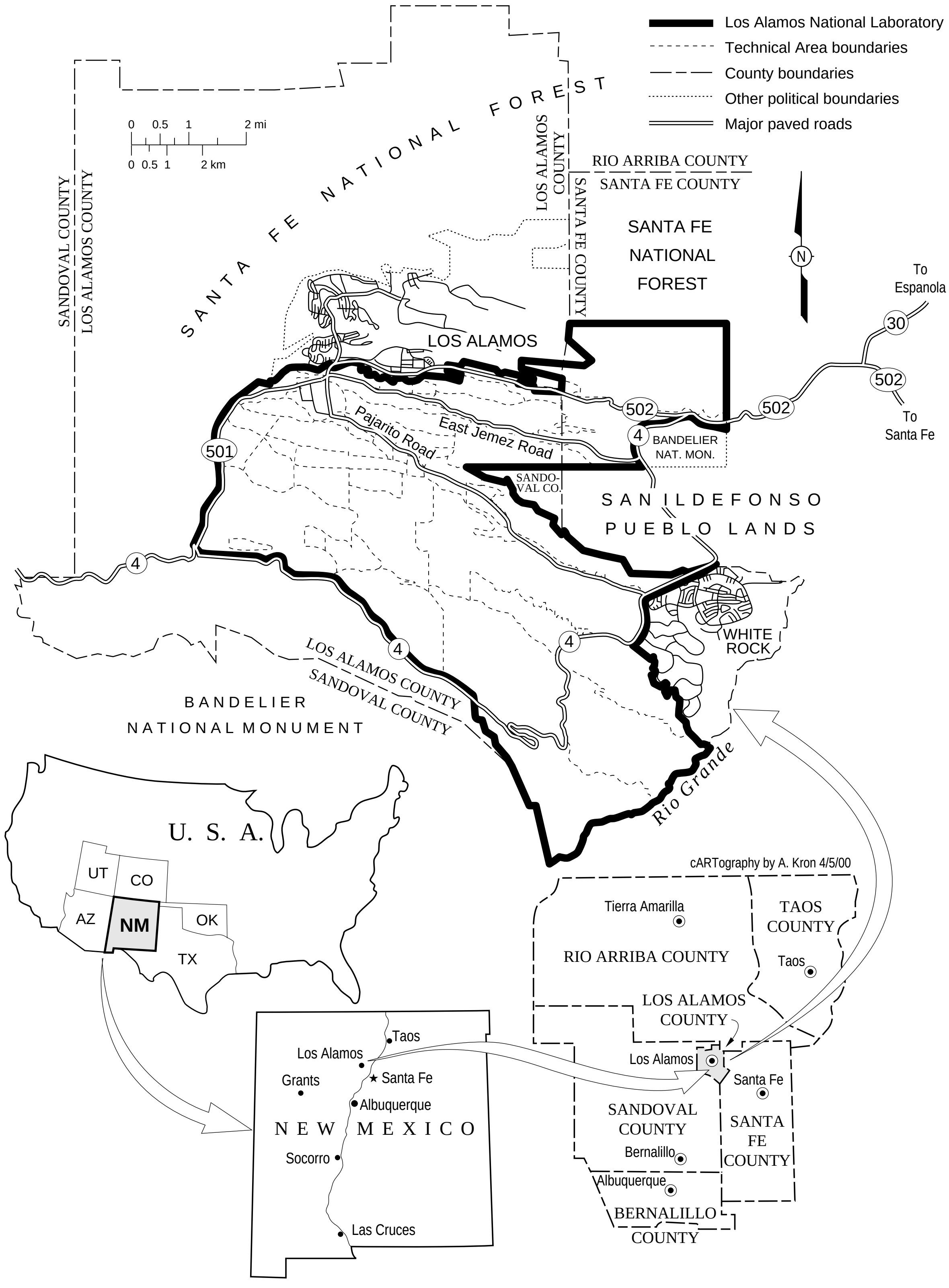


Figure 2. Location of Los Alamos National Laboratory.

the central portion of the Laboratory; it is the only active radioactive liquid effluent discharge (Figure 3).

Technical Area (TA) 45 was the site of the first radioactive liquid waste treatment plant at the Laboratory. Radioactive effluent was discharged into Acid Canyon, a small tributary of Pueblo Canyon, between 1944 and 1964 (DOE 1981). This effluent was untreated before 1951 and the highest concentrations were probably discharged before this time. Discharges from TA-45 directly entered Acid Canyon and flowed into Pueblo Canyon and Los Alamos Canyon, infiltrating into the streambeds. TA-45 was the source for most of the ^{239,240}Pu within the Pueblo/Los Alamos Canyon watershed (DOE 1981; ESP 1999).

The second radioactive discharge originated from TA-21, established in 1945 on DP Mesa and the site of a plutonium processing plant and research laboratories. Treated radioactive effluent was discharged into DP Canyon, a small tributary of Los Alamos Canyon, between 1952 and 1985 (DOE 1979).

The primary use of Mortandad Canyon has been for liquid waste disposal. There are multiple historical sources of plutonium and uranium in this watershed. Mortandad Canyon and its tributaries have received liquid effluents from various Laboratory operations possibly since the Laboratory began operation in 1943. Discharge records for the earlier decades are incomplete. Beginning in 1963, radioactive liquid wastes from most Laboratory operations have been collected and treated at the Radioactive Liquid Waste Treatment Facility (RLWTF) at TA-50. The TA-50 RLWTF has operated continually since and represents the major source of radionuclides released in Mortandad Canyon. The discharge has been regulated as National Pollutant Discharge Elimination System outfall 051 since 1990 and contains relatively low levels of radionuclides and other chemical constituents. The discharge flows a short distance downstream in Mortandad Canyon before infiltrating into the alluvium (LANL 1997).

Many of the radionuclides in the effluents would have tended to adsorb onto sediment or organic colloids. With time, the radionuclide inventory associated with the sediment particles would be remobilized downstream by surface flows.

Explosives Testing

The canyons and mesas of the southern half of the Laboratory contain most of the firing sites where dynamic tests are conducted. The largest operations are within the watersheds of Potrillo, Water, and Ancho Canyons. Other historical sites were located in central (Pajarito Canyon) and northern canyons (Bayo Canyon). Becker (1992) describes the releases that occur from the firing sites:

> "During a dynamic weapons test, depleted uranium is substituted for enriched uranium in a weapons component. The component is then explosively detonated, or is impacted against a target in the open air environment. This results in both the production of a wide range of depleted uranium particles as well as particle scattering over a large distance away from the firing pad. The explosive detonation process of aerial distribution over the watershed distinguishes this contaminant transport problem from others where the source is spatially discrete (e.g., transport away from a waste pile or landfill) . . . . In terms of historical usage of uranium, it has been estimated that on the order of 100 metric tons of depleted and natural uranium have been expended by Los Alamos National Laboratory since the 1940s. Uranium usage was greatest during the early years of Laboratory operation."

Despite the large mass of uranium released to the environment in dynamic testing, detailed field studies near the firing sites show that most of the expended uranium remains close to the firing pads (Becker 1991).

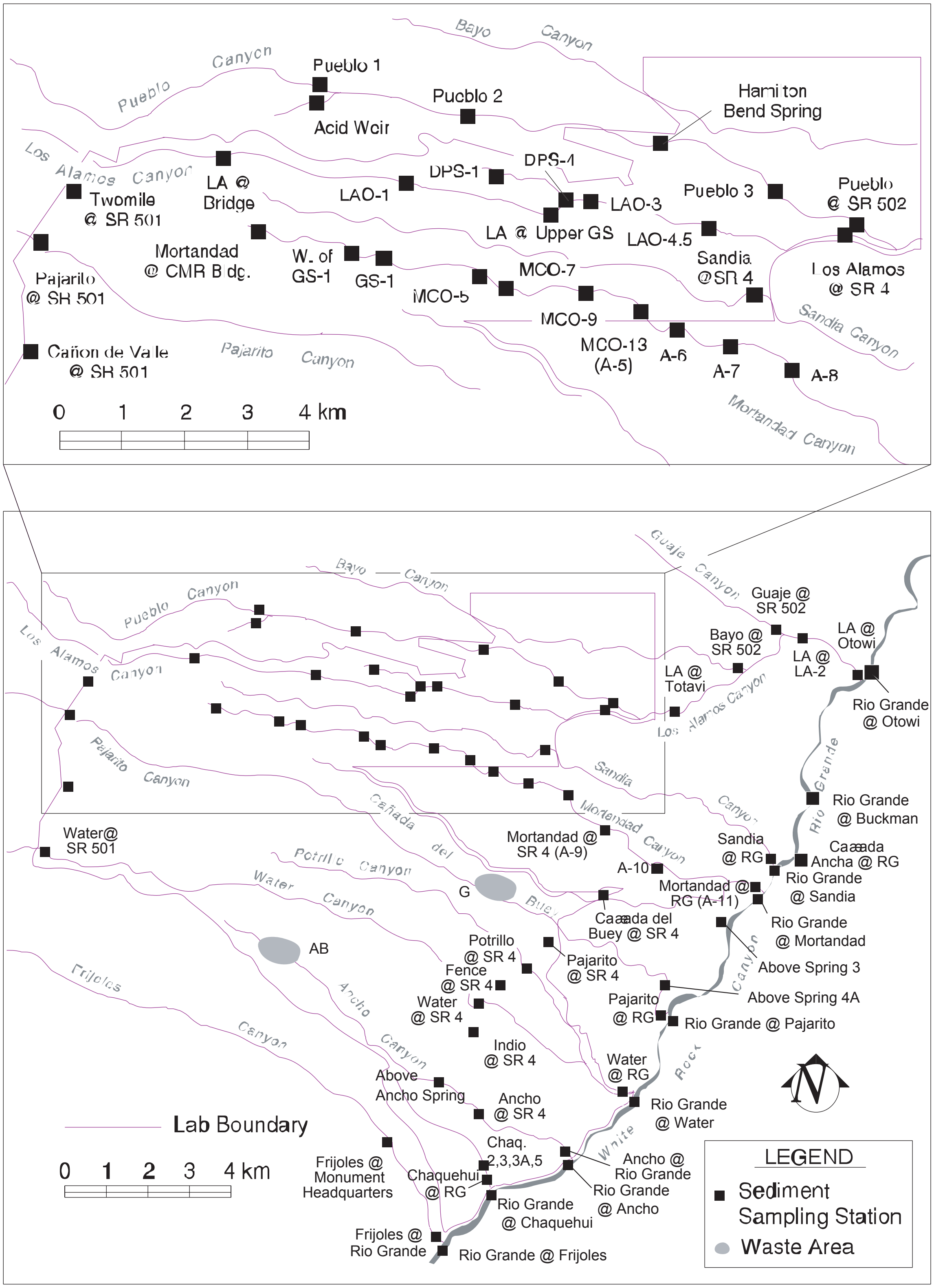


Figure 3. Sediment sampling locations in Los Alamos area.

HISTORICAL SEDIMENT MONITORING RESULTS

The sampling of sedimentary material from streams or ponds can provide an indication of the accumulation of undissolved radionuclides in the aquatic environment. For many of the radionuclides, sediment sampling is a more sensitive indicator of many of the waterborne radionuclides than water sampling. Sediment monitoring data provide important information upon which to draw conclusions about the distribution and source(s) of radionuclides in the environment.

For decades, the Laboratory has annually measured the levels of plutonium isotopes and uranium in northern New Mexico sediments. This includes sediment sample stations near LANL, along the Rio Grande, and from Cochiti Reservoir, a 10,690-acre flood and sediment control project located on the river approximately five miles downstream of the Laboratory. The reservoir since 1973 has served to trap most of the Rio Grande sediments from LANL and from northern New Mexico. Statistics of sample results for ^{239,240}Pu and uranium are included in the Appendix.

The analytical results obtained from LANL monitoring activities indicate there are slightly higher concentrations of uranium and ^{239,240}Pu in some river and reservoir sediments downstream of LANL than upstream (ESP 1999; Gallaher et al. 1999). The radiation doses to downstream users of the Rio Grande that are attributable to Laboratory discharges have been calculated to be a fraction (on the order of 1%) of the dose from natural background and worldwide fallout radiation (DOE 1981; Ferenbaugh et al. 1994).

SAMPLE COLLECTION

Samples of stream and reservoir bottom sediments were collected for this survey during 1991–1998. The geographic area covered by the survey extended from the headwaters of the Rio Grande in southern Colorado to Elephant Butte Reservoir in southern New Mexico. Sample locations are shown in Figures 3–5. Figure 3 shows the locations of samples collected on the Pajarito Plateau on or near the Laboratory. The greatest sample density was on Laboratory lands to establish the isotopic signatures of the known contaminant sources. In addition, broad coverage was extended to other less-impacted areas. In total, approximately 100 sediment samples were processed for plutonium and uranium isotopic signatures.

Samples from the least contaminated lower portions of the drainages were collected at different time periods than the samples collected at the most LANL contaminated locations near the sources. All samples were stored and transferred to the Mass Spectrometry Laboratory under full chain-of-custody procedures.

Sediment samples were collected from all the major watercourses crossing the Laboratory and from the major river systems draining northern New Mexico. Samples were collected along Pueblo/Los Alamos and Mortandad Canyons to determine if Laboratory contaminants could be traced into the Rio Grande. Stream and reservoir sediments were collected along the Rio Grande, above and below the Laboratory.

Samples from the smaller watercourses were collected from transects across the active streambed channel at the 0- to 3-cm (0- to 1-inch) depth using pre-wrapped disposable plastic scoops. Emphasis was on collecting the finer-grained materials, avoiding collection of organic matter, cobbles, and pebbles. Rio Grande and Rio Chama river bed sediments were usually collected at the margin of the active channel, at the bank/water interface. At two Rio Grande sites near the Laboratory (Otowi and Frijoles), the bank samples were complemented by width-integrated composite samples of the bed sediment. Floodplain sediments were sampled at three

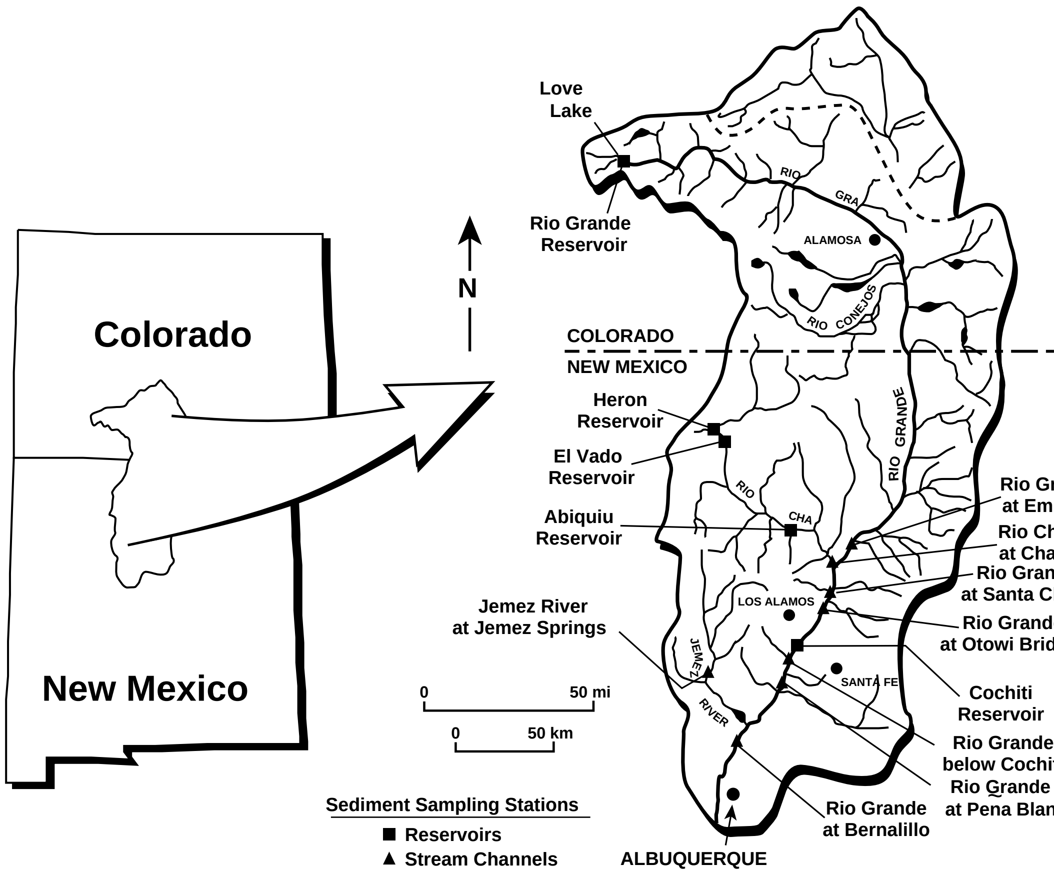


Figure 4. Regional sediment sampling locations within Rio Grande and Rio Chama drainages.

locations identified previously as likely depositional areas for Laboratory contaminants (Graf 1993) that have remained largely undisturbed since deposition.

Reservoir bottom sediments were sampled in two manners: (1) surficial samples at the water/sediment interface and (2) core samples (Figure 6) of the deeper accumulated sediments following the method of Van Metre and Callender (1997). Surficial bottom sediments were collected either with an Eckman dredge or with a box core sampler.

TIMS ANALYTICAL PROCEDURES

Samples collected for this study were submitted to the Los Alamos Clean Chemistry and Mass Spectrometry Laboratory for thermal ionization mass spectrometric (TIMS) analyses. All of the atom ratios reported in this report were derived from the TIMS analyses. The procedures for TIMS analyses of uranium and plutonium were developed by the Mass Spectrometry Laboratory and are described in detail in Efurd et al. (1993). The TIMS procedures are briefly summarized here.

TIMS sample preparation and mass spectrometry are both performed in class-100 clean areas specifically designed for ultra-low-level environmental actinide analyses. During the sample preparation, sediment samples initially are digested with ultra-pure acids. Sediments are then traced with precisely known amounts of reference standards, separated into elements by anion exchange chromatography, and electroplated on mass spectrometry filaments to produce an ionization source for TIMS analysis (Perrin et al. 1985). The filament is then inserted into a

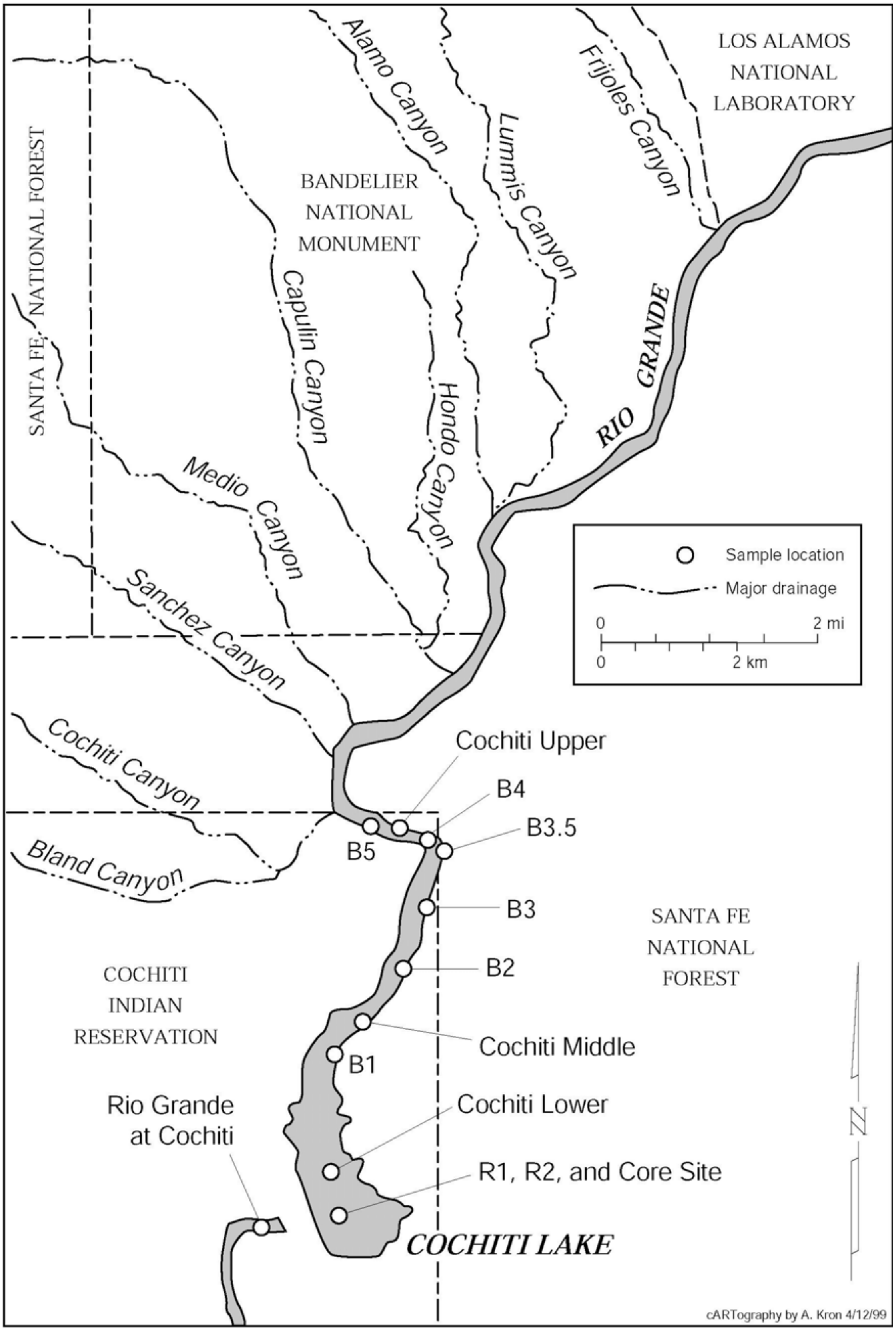


Figure 5. Sediment sampling locations in Cochiti Reservoir.

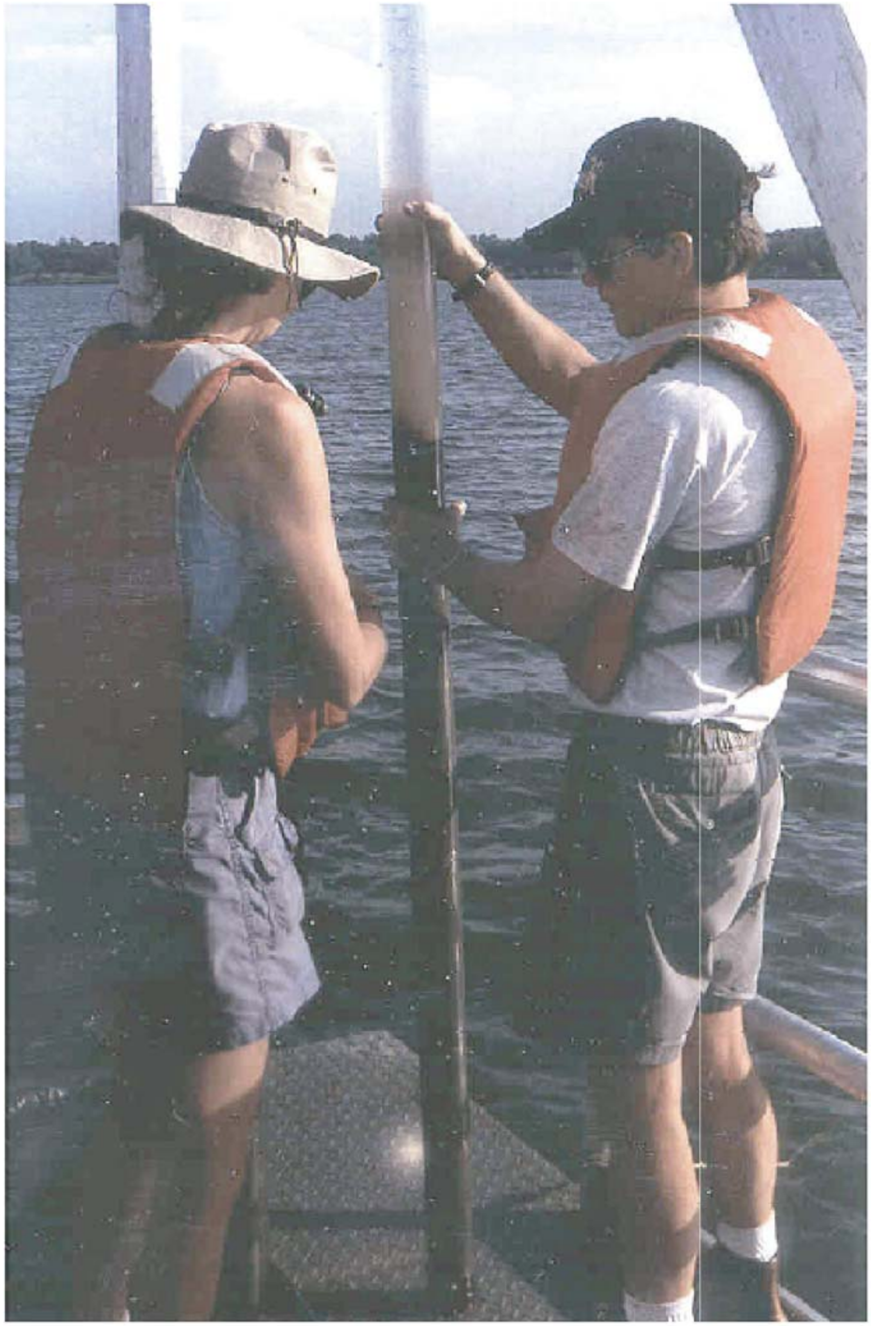


Figure 6. Core samples of Cochiti Reservoir bottom sediments were collected by U.S. Geological Survey scientists and analyzed by the Laboratory for plutonium and uranium isotopes.

thermal ionization mass spectrometer that measures the relative abundance of the isotopes of interest compared with the reference standards.

The TIMS procedure allows for the quantification of the isotopic composition of the plutonium in the sample by measuring the number of atoms of the isotopes ^{240}Pu and ^{239}Pu. Measurement of the $^{240}Pu/^{239}Pu$ atom ratio in samples can be used to distinguish the global fallout component from the Laboratory component(s).

The following uranium isotopes are determined: ^{234}U, ^{235}U, ^{236}U, and ^{238}U. The $^{238}U/^{235}U$ atom ratio can be used to distinguish the naturally occurring uranium from its anthropogenically (human) produced components, in other words, enriched uranium and/or depleted uranium. The enriched and depleted forms of uranium result from the processing of natural uranium to selectively increase (or decrease) the abundance of ^{235}U relative to ^{238}U. Enriched uranium is processed uranium containing more than 0.72 weight percent, the natural mass abundance of ^{235}U; depleted uranium contains less than 0.72 percent ^{235}U. The abundance of ^{235}U in highly enriched uranium may be greater than 90 percent, while the ^{235}U abundance in highly depleted uranium may be on the order of 0.2 percent. The ^{236}U isotope does not exist in nature and its presence indicates an anthropogenic component. The ^{236}U isotope is formed through exposure of ^{235}U to a neutron source, such as a reactor.

IDENTIFICATION OF LANL-DERIVED PLUTONIUM AND URANIUM

PLUTONIUM

Background (Fallout) Plutonium Ratios and Activities

The primary source of plutonium at most locations in New Mexico is global fallout from atmospheric testing of nuclear devices. Areas surrounding the Laboratory may be composed of global fallout or a mixture of LANL plutonium and global fallout. The isotopic composition of global fallout varies from location to location. Global fallout is a complicated mixture with an isotopic composition influenced by the type of nuclear device being tested, the location of the test (Nevada Test Site, Peoples' Republic of China, the former Soviet Union, etc.), and the mechanisms of atmospheric transport and diffusion processes coupled with various fractionation processes (Efurd et al. 1994; Eisenbud and Gesel 1997). Therefore, the isotopic composition of fallout in New Mexico must be determined before the potential contribution of plutonium released from LANL can be assessed.

Three river and 12 lake bottom sediment samples were collected from throughout the Rio Grande drainage system at locations believed to be representative of global fallout in northern New Mexico. The mean atom ratio does not include the result from the Rio Chama at Chamita station because of its large measurement error. Each of the sample sites is located upstream or more than 10 miles distant from the Laboratory. Samples were collected from along the Rio Grande, the Rio Chama, and the Jemez River. Emphasis was given to sampling lake bottom sediments accumulated in reservoirs on the Rio Grande and Rio Chama. The reservoirs capture and integrate suspended and bedload sediments from large upstream contributing drainage areas.

Table 1 summarizes the results obtained on the 15 sediment samples believed to be representative of global fallout in the LANL vicinity. The sample numbers shown in the table are the lab code used to uniquely identify each sample. The sample $^{240}Pu/^{239}Pu$ atom ratios are reported at the 1 sigma (1 standard deviation) uncertainty level. These standard deviations include the errors associated with the TIMS measurements and the uncertainty associated with the concentration of the ^{242}Pu tracer that was used as the isotope dilution tracer. They do not include any errors associated with sample inhomogeneity. The $^{239,240}Pu$ alpha activity levels were determined by multiplying the number of atoms per gram of ^{239}Pu and the number of atoms per gram of ^{240}Pu measured by TIMS by the appropriate specific activity values. The error term associated with each $^{239,240}Pu$ alpha activity is reported at the 1 sigma (1 standard deviation) level.

The $^{239,240}Pu$ activity levels in the 15 samples representative of global fallout in northern New Mexico and southern Colorado ranged from 0.001 to 0.02 pCi/g. The mean $^{239,240}Pu$ activity level was 0.011 pCi/g. The mean $^{240}Pu/^{239}Pu$ atom ratio was 0.169 ± 0.012. The plutonium activity levels and $^{240}Pu/^{239}Pu$ atom ratios are consistent with those obtained by other researchers, as shown in Table 2.

Table 1. Background (Fallout) $^{240}Pu/^{239}Pu$ Atom Ratios in Northern New Mexico and Southern Colorado

Station	Sample Date	Log_ No	240/239_Atom Ratio Ratio	± 1 Std Dev.	$^{239,240}Pu$ Activity (pCi/g)
Colorado Reservoirs					
Rio Grande Reservoir Upper	07/28/1995	12970	0.162	0.005	0.011
Rio Grande Reservoir Upper	06/24/1998	14898	0.160	0.002	0.018
Rio Grande Reservoir Middle	06/24/1998	15352	0.163	0.001	0.020
Rio Grande Reservoir Middle	07/29/1995	13077	0.187	0.016	0.010
Rio Grande Reservoir Lower	07/30/1995	12976	0.181	0.007	0.015
Rio Grande Reservoir Lower	06/24/1998	14899	0.160	0.002	0.020
Rio Grande Reservoir Lower	06/24/1998	15349	0.159	0.001	0.019
Love Lake (CO)	07/28/1995	12973	0.157	0.005	0.023
New Mexico Reservoirs					
Heron Upper	06/23/1998	15350	0.171	0.002	0.006
Heron Middle	06/23/1998	15351	0.170	0.002	0.005
Heron Lower	06/23/1998	15369	0.168	0.002	0.006
Abiquiu Upper	06/22/1998	15348	0.163	0.009	0.001
Abiquiu Middle	06/22/1998	14949	0.170	0.002	0.010
New Mexico Rivers					
Jemez River	11/14/1996	13352	0.169	0.009	0.002
Rio Grande at Embudo	07/25/1994	12114	0.198	0.039	0.001
Rio Chama at Chamita	07/25/1994	12113	0.202	0.110	0.001

Table 2. Comparison of Reported Values for Plutonium Atom Ratios and Activities in Sediments and Soils Attributed to Global Fallout

Site Locations	No. of Sites	Sample Time	$^{240}Pu/^{239}Pu$ Atom Ratio (± 1 S.D.)	$^{239,240}Pu$ Activity (pCi/g)	Reference
Northern New Mexico and Southern Colorado	7	1994–98	0.169 ± 0.012	0.001–0.020	This study
Worldwide (soils)	57	1970–71	0.176 ± 0.014	---	Krey et al. 1976
Colorado (soils)	10	1975	0.169 (avg.)	---	Krey 1976
Colorado (soils)	35	1993–94	0.169 ± 0.005	0.010–0.055 (0.032 avg.)	Efurd et al. 1995
Northern New Mexico and Southern Colorado (soils)	21	1981–86	---	0.001–0.085	Purtymun et al. 1990
Northern New Mexico and Southern Colorado (river and reservoir sediments)	39	1981–86	---	0.0003–0.041	Purtymun et al. 1990
Northern New Mexico (river sediments)	5	1974–97	---	-0.03–0.032 (0.013 statistical 95% upper limit)	McLin and Lyons 2002
Northern New Mexico (reservoir sediments)	4	1982–97	---	0.0002–0.038 (0.022 statistical 95% upper limit)	McLin and Lyons 2002

There are no apparent spatial trends in the atom ratios. However, the plutonium activities in the Colorado reservoir samples often are one order of magnitude greater than in the New Mexico reservoirs. These differences probably reflect a dependence of fallout on precipitation, with greater precipitation occurring at the higher-altitude mountains adjacent to the Colorado reservoirs. Earlier studies by Hardy and Alexander (1962) on the Olympic Peninsula, Washington, showed this effect. The second reason for the differences may be the differences in the distances from the Nevada Test

Site and the more dominant prevailing winds towards Colorado compared to New Mexico. The wind direction is suggested in a map in Krey et al. (1976).

$^{240}Pu/^{239}Pu$ Atom Ratios Used to Identify Laboratory Impacts

We identify Laboratory impacts by comparing $^{240}Pu/^{239}Pu$ atom ratios near Los Alamos with the regional background data set. Evaluation of the 15 background $^{240}Pu/^{239}Pu$ atom ratios indicates that they are consistent with a normal distribution (Shapiro-Wik goodness-of-fit-test, $p<0.01$; Gilbert 1987). Thus, the mean $^{240}Pu/^{239}Pu$ atom ratio of 0.169 ± 0.012 is representative of global fallout in northern New Mexico. Therefore, 99.7% of the background ratios will fall within three standard deviations of the mean: 0.13 to 0.21. Results from Los Alamos area samples are compared against this statistical distribution to evaluate the probability the plutonium is statistically distinguishable from fallout.

No definitive historical information on the $^{240}Pu/^{239}Pu$ atom ratios of Laboratory sources exists, nor do we know how they vary with time and location. However, previous studies from other nuclear research sites indicate the LANL sources likely contained $^{240}Pu/^{239}Pu$ atom ratios considerably lower and statistically distinct from fallout. According to these studies, "pre-1960s plutonium" (Hanford-derived) discharges at LANL probably had $^{240}Pu/^{239}Pu$ atom ratios of approximately 0.01 to 0.03 (Beasley et al. 1981). Waste streams dominated by "modern plutonium" would be expected to have ratios of approximately 0.05 to 0.07 (Efurd et al. 1993; Alberts et al. 1986). Sediment samples with an admixture of LANL-derived plutonium and fallout plutonium will show intermediate $^{240}Pu/^{239}Pu$ atom ratios, ranging between the source and fallout values, depending on mixing proportions.

Based on the background statistical distribution, we developed three general ranges of plutonium isotopic ratios to identify Laboratory impacts (Table 3). These ranges are shown in Figure 7 along with the background distribution and the expected ratios in LANL waste streams. Samples assumed to have a global fallout composition have $^{240}Pu/^{239}Pu$ atom ratios between 0.16 and 0.21. LANL-derived plutonium is assumed for samples with $^{240}Pu/^{239}Pu$ atom ratios between 0 and 0.13. Within this range, the plutonium could be all Laboratory-derived or a mixture of Laboratory-derived and fallout. There is less certainty in the transitional intermediate range (0.13–0.16) of ratios, as the source could be either global fallout or a LANL/global fallout admixture. Approximately one-fourth of the global fallout samples would be expected to have ratios within this range, but there is a larger probability of LANL influences. These ranges are used to initially interpret the data, assuming that the Laboratory is the only source of non-global fallout plutonium.

Plutonium Atom Ratios and Activities near Los Alamos

Approximately 90 sediment samples were collected from locations within and adjacent to the Laboratory. The TIMS results are summarized in Table 4. Results from the background stations also are included in Table 4 to provide comparisons of upstream and downstream conditions.

Combined $^{240+239}Pu$ activity levels from this study ranged from essentially zero to 3.7 pCi/g, compared with the long-term statistical upper limit of background for river sediments of 0.013 pCi/g and for reservoir sediments of 0.020 pCi/g (McLin and Lyons 2002). The highest $^{239,240}Pu$

Table 3. $^{240}Pu/^{239}Pu$ Atom Ratio Ranges Used to Identify Laboratory Impacts

$^{240}Pu/^{239}Pu$ atom ratio	Likely Source of the Plutonium
0.16–0.21	Global fallout
0.13–0.16	Global fallout or mixture of fallout and LANL possible
Less than 0.13	LANL or mixture of LANL and global fallout

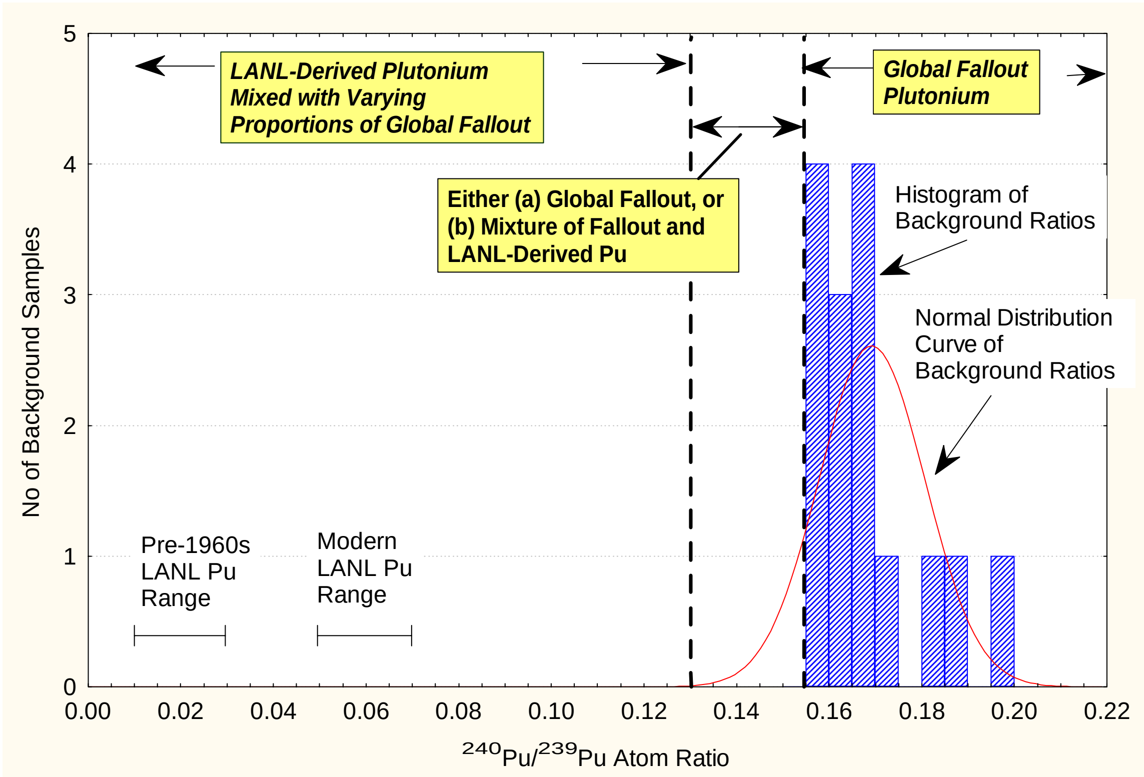


Figure 7. Histogram of background (fallout) $^{240}Pu/^{239}Pu$ atom ratios compared to LANL sources.

activity was found in Pueblo Canyon below the historical TA-45 radioactive effluent discharge (285 times above background). The $^{240}Pu/^{239}Pu$ atom ratio at this location (Acid Weir) was approximately 0.015 and describes the isotopic signature for the TA-45 effluent stream. The activity levels were generally consistent with previous studies. In all cases the plutonium levels were below the current screening action level (SAL) of 44 pCi/g, used as an initial check by the Laboratory's Environmental Restoration Project on whether a contaminant level warrants further study or remediation (ER 2001). The SAL is consistent with Environmental Protection Agency cleanup guidance levels (EPA 1997), which limit industry-caused radiation doses to humans to less than 15 mrem/yr. The total dose from background radiation in the Los Alamos area is about 360 mrem/yr (ESP 1999).

Figure 8 is a graph of the TIMS data and shows the relationship between the atom ratios and plutonium activity levels. For reference, ranges that distinguish fallout-derived plutonium from Laboratory-derived plutonium are displayed. Samples are identified that likely contain Laboratory-derived plutonium, based on the $^{240}Pu/^{239}Pu$ atom ratios. Approximately one-half of the results have a global fallout composition, with $^{240}Pu/^{239}Pu$ atom ratios between 0.16 and 0.21. Plutonium activity levels for this group are within regional background reference levels. All samples measured in this study that contained ≥0.03 pCi/g $^{239,240}Pu$ were consistent with LANL plutonium. At <0.03 pCi/g $^{239,240}Pu$, it was impossible to determine the origin of the plutonium at the sampling locations by $^{239,240}Pu$ activity levels alone.

The most apparent Laboratory impact and perturbation from background are in samples collected from Pueblo/Los Alamos Canyons and in samples of Cochiti Reservoir bottom sediments. Sediments within this group show above-background plutonium activities and very low $^{240}Pu/^{239}Pu$ atom ratios of 0.01 to 0.03. Along the Rio Grande, river sediments often show

Table 4. Plutonium Isotopic Composition of Sediment Samples (TIMS Analyses)

Station Name	Sample Date	Log No.	Sample Size (g)	Atom Ratio $^{240}Pu/^{239}Pu$	Uncert.		Atom Ratio $^{241}Pu/^{239}Pu$	Uncert.	Atoms $^{239}Pu/g$	Uncert.
Regional Stations										
Rio Chama at Chamita	25-Jul-94	12113	48.05	0.202	54.3%	BDL			1.4E+07	14.8%
Rio Grande at Embudo	25-Jul-94	12114	48.53	0.198	19.6%				3.0E+07	6.5%
Rio Grande at Santa Clara Flood Plain 2B (0-0.5 ft.)	17-Aug-95	13036	38.51	0.254	58.2%	BDL			3.0E+06	18.8%
Rio Grande at Otowi (bank)	17-Aug-95	12971A		0.262	10.7%				1.98+07	6.0%
Rio Grande at Otowi (bank)	17-Aug-95	12971B		0.261	9.6%				1.98+07	6.0%
Rio Grande at Otowi (wdth intgrt)	15-Sep-95	13035	50.25	0.124	21.2%				1.4E+07	4.6%
Rio Grande at Buckman Flood Plain 2B (0-0.5 ft.)	1-Jun-91	10411		0.077	1.1%					
Rio Grande at Buckman Flood Plain 2B (1-1.5 ft.)	17-Aug-95	12974A	41.49	0.095	73.7%	BDL			4.2E+06	8.8%
Rio Grande at Buckman Flood Plain 2B (1-1.5 ft.)	17-Aug-95	12974B	41.49	0.098	73.7%	BDL			4.2E+06	10.9%
Rio Grande at Sandia	27-Sep-94	12666	38.15	0.084	19.1%				3.7E+07	3.9%
Rio Grande at Pajarito	28-Sep-94	12629	37.08	0.151	20.9%				3.2E+07	6.5%
Rio Grande at Water	28-Sep-94	12631	36.62	0.146	4.8%				1.1E+08	2.1%
Rio Grande at Ancho	29-Sep-94	12632	42.37	0.114	9.3%				4.0E+07	2.0%
Rio Grande at Chaquehui	29-Sep-94	12635	41.48	0.076	6.1%		0.0071		7.0E+07	1.1%
Rio Grande at Frijoles (bank)	29-Sep-94	12577	40.21	0.168	5.8%				2.4E+07	1.1%
Rio Grande at Frijoles (wdth intgrt)	13-Sep-95	13034	57.41	0.154	26.4%				2.3E+07	7.5%
Rio Grande at Pena Blanca Flood Plain 2B (0-0.5 ft.)	17-Aug-95	12972	32.37	0.173	2.8%				2.5E+08	1.1%
Jemez River	14-Nov-96	13352	52.67	0.169	5.2%				6.0E+07	
Reservoirs on Rio Chama										
Heron Upper	23-Jun-98	15350	51.43	0.171	1.2%				1.4E+08	0.8%
Heron Middle	23-Jun-98	15351	49.99	0.170	1.4%				1.2E+08	0.4%
Heron Lower	23-Jun-98	15369	51.88	0.168	1.1%				1.5E+08	0.4%
Abiquiu Upper	22-Jun-98	15348	50.56	0.163	5.3%				2.5E+07	1.1%
Abiquiu Middle	22-Jun-98	14949	87.50	0.179	1.4%				2.5E+08	0.9%
Reservoirs on Rio Grande										
Rio Grande (S. Colorado):										
Rio Grande Upper	24-Jun-98	14898	100.20	0.160	1.3%				4.7E+08	1.1%
Rio Grande Upper	28-Jul-95	12970	30.53	0.162	3.3%				2.8E+08	2.1%
Rio Grande Middle	29-Jul-95	13077	25.15	0.187	8.7%				2.5E+08	2.2%
Rio Grande Middle	24-Jun-98	15352	51.26	0.163	0.8%				5.1E+08	0.6%

Table 4. cont.

Station Name	Atoms ^{240}Pu/g	Uncert.	Atoms ^{241}Pu/g	Uncert.	Activity ^{239}Pu (pCi/g)	Uncert.	Activity ^{240}Pu (pCi/g)	Uncert.	Activity ^{241}Pu (pCi/g)	Uncert.
Regional Stations										
Rio Chama at Chamita	2.8E+06	52.2%			0.0003	14.8%	0.00026	52.2%		
Rio Grande at Embudo	5.8E+06	18.5%			0.0007	6.5%	0.00053	18.5%		
Rio Grande at Santa Clara Flood Plain 2B (0-0.5 ft.)	7.7E+05	55.1%			0.0001	18.8%	0.00007	55.1%		
Rio Grande at Otowi (bank)	5.18+06	8.9%			0.0005	0.6%	0.00050	0.7%		
Rio Grande at Otowi (bank)	5.17+06	7.5%			0.0005	0.6%	0.00500	0.8%		
Rio Grande at Otowi (wdth intgrt)	1.7E+06	20.7%			0.0003	4.6%	0.00016	20.7%		
Rio Grande at Buckman Flood Plain 2B (0-0.5 ft.)										
Rio Grande at Buckman Flood Plain 2B (1-1.5 ft.)	1.0E+05	72.9%			0.0001	8.8%	0.00004	72.9%		
Rio Grande at Buckman Flood Plain 2B (1-1.5 ft.)	4.2E+05	72.9%			0.0001	10.9%	0.00004	72.9%		
Rio Grande at Sandia	3.1E+06	18.7%			0.0009	3.9%	0.00028	18.7%		
Rio Grande at Pajarito	4.8E+06	19.9%			0.0008	6.5%	0.00043	19.9%		
Rio Grande at Water	1.6E+07	4.4%			0.0026	2.1%	0.00141	4.4%		
Rio Grande at Ancho	4.5E+06	9.1%			0.0010	2.0%	0.00041	9.1%		
Rio Grande at Chaquehui	5.3E+06	6.0%	5.0E+05		0.0017	1.1%	0.00048	6.0%	0.0205	
Rio Grande at Frijoles (bank)	4.1E+06	5.6%			0.0006	1.1%	0.00037	5.6%		
Rio Grande at Frijoles (wdth intgrt)	3.5E+06	25.3%			0.0006	7.5%	0.00032	25.3%		
Rio Grande at Pena Blanca Flood Plain 2B (0-0.5 ft.)	4.3E+07	2.5%			0.0062	1.1%	0.00392	2.5%		
Jemez River	1.0E+07				0.0015		0.00092			
Reservoirs on Rio Chama										
Heron Upper	2.4E+07	0.9%			0.0034	0.8%	0.00216	0.9%		
Heron Middle	2.1E+07	1.4%			0.0030	0.4%	0.00189	1.4%		
Heron Lower	2.6E+07	1.1%			0.0038	0.4%	0.00232	1.1%		
Abiquiu Upper	4.1E+06	5.4%			0.0006	1.1%	0.00037	5.4%		
Abiquiu Middle	4.3E+07	1.1%			0.0063	0.9%	0.00391	1.1%		
Reservoirs on Rio Grande										
Rio Grande (S. Colorado):										
Rio Grande Upper	7.5E+07	0.7%			0.0116	1.1%	0.00684	0.7%		
Rio Grande Upper	4.6E+07	2.5%			0.0070	2.1%	0.00414	2.5%		
Rio Grande Middle	4.6E+07	8.4%			0.0061	2.2%	0.00417	8.4%		
Rio Grande Middle	8.3E+07	0.5%			0.0126	0.6%	0.00753	0.5%		

Table 4. cont.

Station Name	Sample Date	Log No.	Sample Size (g)	Atom Ratio $^{240}Pu/^{239}Pu$	Uncert.		Atom Ratio $^{241}Pu/^{239}Pu$	Uncert.	Atoms $^{239}Pu/g$	Uncert.
Love Lake (S. Colorado):										
Love Lake	28-Jul-95	12973	22.49	0.157	3.1%				5.9E+08	0.7%
Cochiti Reservoir:										
Cochiti Upper	2-Aug-94	12173	54.91	0.135	38.4%	BDL			1.7E+07	9.9%
Cochiti Upper	24-Sep-98	15372	52.38	0.159	0.8%				3.0E+08	0.2%
Cochiti Upper	2-Aug-94	12173	54.91	0.135	38.4%	BDL			1.7E+07	9.9%
Cochiti Upper	24-Sep-98	15372	52.38	0.159	0.8%				3.0E+08	0.2%
Cochiti Middle	24-Sep-98	15373	51.62	0.160	1.0%				3.5E+08	0.8%
Cochiti Lower	24-Sep-98	15371	49.05	0.153	1.2%				3.4E+08	0.4%
Cochiti Resv. Core #3 0-26 cm	17-Sep-96	13384	21.62	0.105	2.5%				4.4E+08	0.5%
Cochiti Resv. Core #3 26-52 cm	17-Sep-96	13385	40.60	0.084	1.5%				5.0E+08	0.4%
Cochiti Resv. Core #3 52-78 cm	17-Sep-96	13386	42.69	0.039	1.3%				1.6E+09	0.2%
Cochiti Resv. Core #3 78-91 cm	17-Sep-96	13350	24.64	0.076	2.4%				1.0E+09	0.7%
Cochiti Resv. Core #3 91-104 cm	17-Sep-96	13388	31.11	0.068	1.7%				8.9E+08	0.4%
Cochiti Resv. Core #3 104-117 cm	17-Sep-96	13351	36.24	0.077	3.1%				8.6E+08	0.8%
Cochiti Resv. Core #3 117-130 cm	17-Sep-96	13346	43.44	0.082	1.8%				8.0E+08	0.6%
Cochiti Resv. Core #3 130-143 cm	17-Sep-96	13389	51.12	0.097	1.2%				5.8E+08	0.4%
Cochiti Resv. Core #3 143-156 cm	17-Sep-96	13345	58.23	0.189	6.2%				5.3E+08	3.8%
Cochiti Resv. Surface R1	17-Sep-96	13387	40.74	0.146	0.8%				5.3E+08	0.4%
Cochiti Resv. Surface B11	17-Sep-96	13349	33.93	0.156	2.1%				6.2E+08	0.6%
Cochiti Resv. Surface B2	17-Sep-96	13341	39.02	0.159	1.0%				5.8E+08	0.5%
Cochiti Resv. Surface B3.5	17-Sep-96	13342	89.34	0.156	1.0%				4.6E+08	0.3%
Cochiti Resv. Surface B4	17-Sep-96	13343	71.19	0.156	1.4%				6.2E+08	0.5%
Cochiti Resv. Surface B5	17-Sep-96	13344	68.49	0.167	1.1%				4.6E+08	0.4%
Elephant Butte Reservoir (S. New Mexico):										
Elephant Butte Core #4 12-15 cm	25-Jul-95	13030A	21.09	0.163	4.6%				2.2E+08	2.0%
Elephant Butte Core #4 12-15 cm	25-Jul-95	13030B	21.09	0.163	4.3%				2.2E+08	2.0%
Elephant Butte Core #4 42-45 cm	25-Jul-95	13037	33.59	0.165	3.1%				3.8E+08	1.1%
Elephant Butte Core #4 87-90 cm	25-Jul-95	13031	29.53	0.174	2.3%				5.3E+08	0.9%
Elephant Butte Core #4 111-114 cm	25-Jul-95	13032A	32.43	0.195	3.4%				5.2E+08	1.4%
Elephant Butte Core #4 111-114 cm	25-Jul-95	13032B	32.43	0.195	3.4%				5.2E+08	1.3%
Elephant Butte Core #4 135-138 cm	25-Jul-95	13033	31.00	0.170	6.5%				3.7E+08	2.4%
Elephant Butte Core #4 159-162 cm	25-Jul-95	13038	37.00	0.173	2.0%				5.7E+08	0.9%
Pajarito Plateau Stations										
Guaje Canyon:										
Guaje at SR-502	28-Jun-94	12008	37.45	0.125	12.3%				2.9E+07	3.3%

Table 4. cont.

Station Name	Atoms ^{240}Pu/g	Uncert.	Atoms ^{241}Pu/g	Uncert.	Activity ^{239}Pu (pCi/g)	Uncert.	Activity ^{240}Pu (pCi/g)	Uncert.	Activity ^{241}Pu (pCi/g)	Uncert.
Love Lake (S. Colorado):										
Love Lake	9.2E+07	3.0%			0.0145	0.7%	0.00837	3.0%		
Cochiti Reservoir:										
Cochiti Upper	2.3E+06	37.1%			0.0004	9.9%	0.00020	37.1%		
Cochiti Upper	4.7E+07	0.8%			0.0074	0.2%	0.00430	0.8%		
Cochiti Upper	2.3E+06	37.1%			0.0004	9.9%	0.00020	37.1%		
Cochiti Upper	4.7E+07	0.8%			0.0074	0.2%	0.00430	0.8%		
Cochiti Middle	5.6E+07	0.7%			0.0086	0.8%	0.00505	0.7%		
Cochiti Lower	5.3E+07	1.2%			0.0085	0.4%	0.00477	1.2%		
Cochiti Resv. Core #3 0-26 cm	4.7E+07	2.4%			0.0110	0.5%	0.00423	2.4%		
Cochiti Resv. Core #3 26-52 cm	4.2E+07	1.5%			0.0123	0.4%	0.00382	1.5%		
Cochiti Resv. Core #3 52-78 cm	6.3E+07	1.3%			0.0402	0.2%	0.00573	1.3%		
Cochiti Resv. Core #3 78-91 cm	7.8E+07	2.3%	1.8E+06		0.0250	0.7%	0.00702	2.3%	0.0747	
Cochiti Resv. Core #3 91-104 cm	6.1E+07	1.6%			0.0219	0.4%	0.00551	1.6%		
Cochiti Resv. Core #3 104-117 cm	6.7E+07	3.0%			0.0211	0.8%	0.00602	3.0%		
Cochiti Resv. Core #3 117-130 cm	6.6E+07	1.7%			0.0197	0.6%	0.00595	1.7%		
Cochiti Resv. Core #3 130-143 cm	5.6E+07	1.1%			0.0142	0.4%	0.00506	1.1%		
Cochiti Resv. Core #3 143-156 cm	1.0E+08	4.9%			0.0132	3.8%	0.00916	4.9%		
Cochiti Resv. Surface R1	7.7E+07	0.7%			0.0130	0.4%	0.00696	0.7%		
Cochiti Resv. Surface B11	9.7E+07	2.0%	1.4E+06		0.0154	0.6%	0.00880	2.0%	0.0579	
Cochiti Resv. Surface B2	9.2E+07	0.8%			0.0143	0.5%	0.00836	0.8%		
Cochiti Resv. Surface B3.5	7.2E+07	1.0%			0.0114	0.3%	0.00654	1.0%		
Cochiti Resv. Surface B4	9.7E+07	1.3%			0.0153	0.5%	0.00874	1.3%		
Cochiti Resv. Surface B5	7.6E+07	1.0%			0.0113	0.4%	0.00692	1.0%		
Elephant Butte Reservoir (S. New Mexico):										
Elephant Butte Core #4 12-15 cm	3.53E+07	4.13%			0.0053	2.0%	0.0032	4.13%		
Elephant Butte Core #4 12-15 cm	3.53E+07	3.80%			0.0053	2.0%	0.0032	3.80%		
Elephant Butte Core #4 42-45 cm	6.2E+07	2.9%			0.0093	1.1%	0.0056	2.9%		
Elephant Butte Core #4 87-90 cm	9.1E+07	2.1%			0.0129	0.9%	0.0083	2.1%		
Elephant Butte Core #4 111-114 cm	1.0E+08	3.1%			0.0127	1.4%	0.0091	3.1%		
Elephant Butte Core #4 111-114 cm	1.0E+08	3.1%			0.0127	1.3%	0.0091	3.1%		
Elephant Butte Core #4 135-138 cm	6.4E+07	6.0%			0.0092	2.4%	0.0058	6.0%		
Elephant Butte Core #4 159-162 cm	9.8E+07	1.8%			0.0140	0.9%	0.0089	1.8%		
Pajarito Plateau Stations										
Guaje Canyon:										
Guaje at SR-502	3.6E+06	11.8%			0.0007	3.3%	0.00033	11.8%		

Table 4. cont.

Station Name	Sample Date	Log No.	Sample Size (g)	Atom Ratio $^{240}Pu/^{239}Pu$	Uncert.		Atom Ratio $^{241}Pu/^{239}Pu$	Uncert.	Atoms $^{239}Pu/g$	Uncert.
Bayo Canyon:										
Bayo at SR-502	3-Jun-94	12012	58.49	2.352	24.9%				1.2E+06	10.0%
Acid/Pueblo Canyons										
Pueblo 1	13-Jul-94	12177	46.67	0.096	23.0%				4.8E+07	3.9%
Acid Weir	9-Aug-94	12665	41.24	0.016	0.7%		0.0001		1.3E+11	0.3%
Pueblo 2	13-Jul-94	12010	39.66	0.013	1.2%		0.0000		2.6E+10	0.1%
Hamilton Bend Spring	13-Jul-94	12009	43.27	0.014	1.3%		0.0001		1.2E+10	0.2%
Pueblo 3	13-Jul-94	12081	52.04	0.233	113.2%	BDL			1.1E+06	18.7%
Pueblo at SR-502	3-Jun-94	12016	51.55	0.013	2.3%				1.9E+10	0.4%
DP/Los Alamos Canyons										
Los Alamos at SR-4	3-Jun-94	12068	49.02	0.032	4.1%				1.4E+09	0.7%
Los Alamos at Totavi	28-Jun-94	12082	53.29	4.517	143.7%	BDL			-9.0E+04	-135.4%
Los Alamos at LA-2	28-Jun-94	12079	57.33	0.014	2.4%		0.0003	3440.0%	3.7E+09	0.5%
Los Alamos at Otowi	14-Jul-94	12080	50.17	0.016	4.6%		0.0011	1830.0%	3.1E+09	0.8%
Los Alamos at Otowi Flood Plain 2B (0-0.5 ft)	14-Aug-95	12975B		0.032	6.7%				6.4E+09	0.6%
Los Alamos at Otowi Flood Plain 2B (0-0.5 ft)	14-Aug-95	12975A		0.033	6.7%				6.4E+09	0.6%
Sandia Canyon										
Sandia at SR-4	3-Jun-94	12015	63.42	0.098	26.9%				1.6E+07	4.1%
SSI-1	27-Jul-94	12104	53.57	0.081	10.1%				3.1E+07	2.7%
SSI-2	27-Jul-94	12116	52.38	0.169	12.9%				5.5E+07	3.9%
SSI-3	27-Jul-94	12105	48.73	0.127	16.9%				1.2E+07	4.0%
Sandia at Rio Grande	27-Sep-94	12667	45.28	0.185	11.1%				1.9E+07	3.8%
Canada Ancha:										
Canada Ancha at Rio Grande	27-Sep-94	12670	53.68	0.097	57.9%	BDL			4.1E+06	6.2%
Mortandad Canyon:										
Mortandad at MCO-13 (A-5)	27-Jul-94	12115	53.43	0.081	47.6%	BDL			1.8E+07	5.3%
Mortandad A-6	27-Jul-94	12103	36.11	0.094	2.7%		0.0030		3.9E+08	0.8%
Mortandad A-7	27-Jul-94	12101	43.78	0.163	4.6%		0.0034		2.7E+08	1.5%
Mortandad A-7	17-Nov-98	15370	52.22	0.144	0.9%				4.0E+08	0.4%
Mortandad A-8	27-Jul-94	12174	46.49	0.139	5.2%				5.1E+07	1.3%
Mortandad at SR-4 (A-9)	3-Jun-94	12013	48.05	0.207	34.8%	BDL			1.5E+07	8.7%
Mortandad A-10	27-Jul-94	12102	45.14	0.203	98.3%	BDL			1.1E+07	23.4%
Mortandad at Rio Grande (A-11)	27-Sep-94	12668	38.86	0.099	8.5%		0.0045		7.2E+07	1.8%
Canada del Buey:										
Canada del Buey at SR-4	3-Jun-94	12066	47.33	0.170	16.6%				2.1E+07	6.5%

Table 4. cont.

Station Name	Atoms ^{240}Pu/g	Uncert.	Atoms ^{241}Pu/g	Uncert.	Activity ^{239}Pu (pCi/g)	Uncert.	Activity ^{240}Pu (pCi/g)	Uncert.	Activity ^{241}Pu (pCi/g)	Uncert.
Bayo Canyon:										
Bayo at SR-502	2.9E+06	22.9%			0.0000	10.0%	0.00026	22.9%		
Acid/Pueblo Canyons										
Pueblo 1	4.6E+06	22.6%			0.0012	3.9%	0.00042	22.6%		
Acid Weir	2.0E+09	0.7%	8.9E+06		3.1858	0.3%	0.18270	0.7%	0.3663	
Pueblo 2	3.3E+08	1.2%	1.1E+06		0.6321	0.1%	0.03001	1.2%	0.0443	
Hamilton Bend Spring	1.6E+08	1.2%	1.2E+06		0.2933	0.2%	0.01473	1.2%	0.0486	
Pueblo 3	2.6E+05	111.6%			0.0000	18.7%	0.00002	111.6%		
Pueblo at SR-502	2.4E+08	2.2%			0.4593	0.4%	0.02171	2.2%		
DP/Los Alamos Canyons										
Los Alamos at SR-4	4.3E+07	4.0%			0.0335	0.7%	0.00390	4.0%		
Los Alamos at Totavi	-4.1E+05	-48.1%			0.0000	-135.4%	-0.00004	-48.1%		
Los Alamos at LA-2	5.4E+07	2.4%	1.3E+06	33.6%	0.0920	0.5%	0.00488	2.4%	0.0524	34.4%
Los Alamos at Otowi	4.8E+07	4.5%	3.6E+06	18.1%	0.0768	0.8%	0.00437	4.5%	0.1471	18.3%
Los Alamos at Otowi Flood Plain 2B (0-0.5 ft)	2.1E+08	6.6%			0.1586	0.6%	0.01890	6.6%		
Los Alamos at Otowi Flood Plain 2B (0-0.5 ft)	2.1E+08	6.6%			0.1586	0.6%	0.01896	6.6%		
Sandia Canyon										
Sandia at SR-4	1.6E+06	26.6%			0.0004	4.1%	0.00014	26.6%		
SSI-1	2.5E+06	9.8%			0.0008	2.7%	0.00022	9.8%		
SSI-2	9.3E+06	12.3%			0.0014	3.9%	0.00084	12.3%		
SSI-3	1.5E+06	16.4%			0.0003	4.0%	0.00013	16.4%		
Sandia at Rio Grande	3.5E+06	10.5%			0.0005	3.8%	0.00031	10.5%		
Canada Ancha:										
Canada Ancha at Rio Grande	4.0E+05	57.6%			0.0001	6.2%	0.00004	57.6%		
Mortandad Canyon:										
Mortandad at MCO-13 (A-5)	1.4E+06	47.3%			0.0004	5.3%	0.00013	47.3%		
Mortandad A-6	3.7E+07	2.6%	1.1E+06		0.0096	0.8%	0.00331	2.6%	0.0474	
Mortandad A-7	4.3E+07	4.3%	8.9E+05		0.0066	1.5%	0.00392	4.3%	0.0368	
Mortandad A-7	5.8E+07	0.8%			0.0100	0.4%	0.00527	0.8%		
Mortandad A-8	7.0E+06	5.1%			0.0012	1.3%	0.00063	5.1%		
Mortandad at SR-4 (A-9)	3.0E+06	33.7%			0.0004	8.7%	0.00027	33.7%		
Mortandad A-10	2.2E+06	95.4%			0.0003	23.4%	0.00020	95.4%		
Mortandad at Rio Grande (A-11)	7.2E+06	8.3%	3.3E+05		0.0018	1.8%	0.00065	8.3%	0.0135	
Canada del Buey:										
Canada del Buey at SR-4	3.6E+06	15.3%			0.0005	6.5%	0.00033	15.3%		

Table 4. cont.

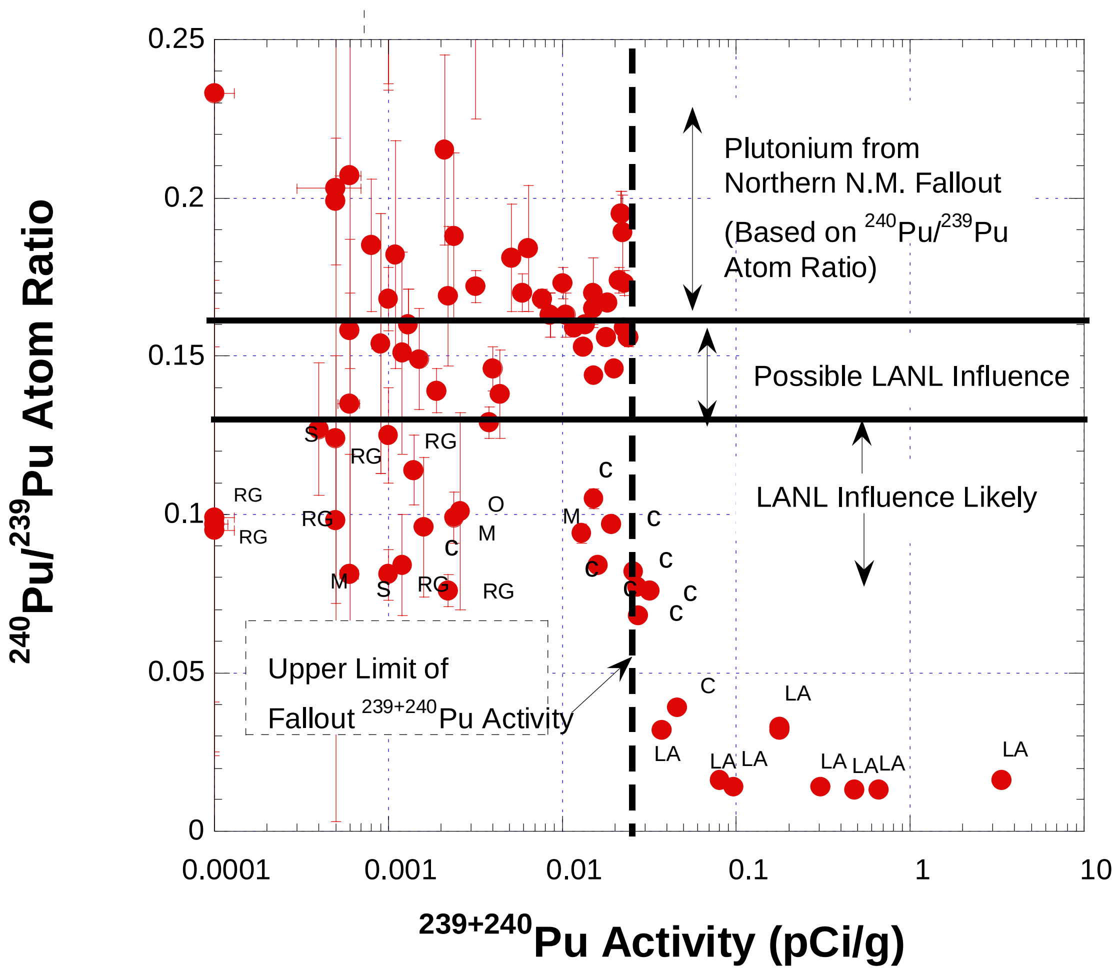
Station Name	Sample Date	Log No.	Sample Size (g)	Atom Ratio $^{240}Pu/^{239}Pu$	Uncert.	Atom Ratio $^{241}Pu/^{239}Pu$	Uncert.	Atoms $^{239}Pu/g$	Uncert.
Pajarito Canyon:									
Pajarito at SR-4	2-Jun-94	12069	46.74	0.181	9.2%			1.2E+08	2.9%
Pajarito at Rio Grande	28-Sep-94	12628	43.86	1.605	11.3%			1.9E+06	4.7%
Above Spring 3	9-Nov-94	12663	37.80	0.138	9.9%			1.2E+08	2.0%
Above Spring 4A	9-Nov-94	12664	37.80	0.170	3.8%	-0.0004		1.5E+08	1.5%
Potrillo Canyon:									
Potrillo at SR-4	9-Aug-94	12014	48.53	0.149	10.6%			4.0E+07	3.7%
Fence Canyon									
Fence at SR-4	2-Jun-94	12669	38.56	0.168	1.6%	0.0024		1.9E+08	0.4%
Fence at SR-4	7-Sep-94	12007	44.32					2.7E+07	64.6%
Water Canyon:									
Water at SR-4	3-Jun-94	12070	50.44	0.215	13.7%			4.7E+07	4.8%
Water at Rio Grande	28-Sep-94	12630	39.99	0.147	3.8%				
Indio Canyon:									
Indio at SR-4	7-Sep-94	12176	47.06	0.188	14.0%			5.9E+07	6.3%
Ancho Canyon:									
Above Ancho Spring	28-Sep-94	12634	52.37	0.158	7.7%	-0.0001	536140.0%	1.5E+07	1.5%
Ancho at Rio Grande	28-Sep-94	12633	38.92	0.129	3.9%	0.0076		1.0E+08	1.4%
Chaquehui Canyon:									
Chaquehui at Rio Grande	29-Sep-94	12575	43.23	0.160	6.7%			3.2E+07	2.3%
Chaquehui Sed Station 2	29-Sep-94	12579	42.00	0.271	16.9%			6.5E+07	7.7%
Chaquehui Sed Station 3	29-Sep-94	12578	42.62	0.172	2.7%	0.0026		7.9E+07	1.1%
Chaquehui Sed Station 3A	29-Sep-94	12581	43.93	0.101	30.5%			7.6E+07	6.4%
Chaquehui Sed Station 5	29-Sep-94	12580	34.83	0.184	11.1%			1.5E+08	3.9%
Frijoles Canyon:									
Frijoles at Monument HQ	28-Jun-94	12067	37.37	0.182	19.7%			2.8E+07	6.8%
Frijoles at Rio Grande	29-Sep-94	12576	46.17	0.199	10.1%			1.1E+07	3.5%

BDL = Below Detection Limits (uncert > 33%)

Table 4. cont.

Station Name	Atoms ^{240}Pu/g	Uncert.	Atoms ^{241}Pu/g	Uncert.	Activity ^{239}Pu (pCi/g)	Uncert.	Activity ^{240}Pu (pCi/g)	Uncert.	Activity ^{241}Pu (pCi/g)	Uncert.
Pajarito Canyon:										
Pajarito at SR-4	2.2E+07	8.8%			0.0031	2.9%	0.00204	8.8%		
Pajarito at Rio Grande	3.0E+06	10.3%			0.0000	4.7%	0.00027	10.3%		
Above Spring 3	1.6E+07	9.7%			0.0030	2.0%	0.00149	9.7%		
Above Spring 4A	2.5E+07	3.5%	-5.8E+04		0.0036	1.5%	0.00226	3.5%	-0.0024	
Potrillo Canyon:										
Potrillo at SR-4	6.0E+06	10.0%			0.0010	3.7%	0.00054	10.0%		
Fence Canyon										
Fence at SR-4	3.2E+07	1.6%	4.6E+05		0.0048	0.4%	0.00294	1.6%	0.0188	
Fence at SR-4					0.0007	64.6%				
Water Canyon:										
Water at SR-4	1.0E+07	12.9%			0.0012	4.8%	0.00092	12.9%		
Water at Rio Grande										
Indio Canyon:										
Indio at SR-4	1.1E+07	12.5%			0.0014	6.3%	0.00100	12.5%		
Ancho Canyon:										
Above Ancho Spring	2.3E+06	7.5%	-1.9E+03	-5389.5%	0.0004	1.5%	0.00021	7.5%	-0.0001	-500.0%
Ancho at Rio Grande	1.3E+07	3.7%	7.9E+05		0.0026	1.4%	0.00121	3.7%	0.0324	
Chaquehui Canyon:										
Chaquehui at Rio Grande	5.1E+06	6.3%			0.0008	2.3%	0.00046	6.3%		
Chaquehui Sed Station 2	1.8E+07	15.0%			0.0016	7.7%	0.00161	15.0%		
Chaquehui Sed Station 3	1.4E+07	2.5%	2.1E+05		0.0020	1.1%	0.00123	2.5%	0.0087	
Chaquehui Sed Station 3A	7.7E+06	29.8%			0.0019	6.4%	0.00070	29.8%		
Chaquehui Sed Station 5	2.8E+07	10.4%			0.0038	3.9%	0.00256	10.4%		
Frijoles Canyon:										
Frijoles at Monument HQ	5.0E+06	18.5%			0.0007	6.8%	0.00045	18.5%		
Frijoles at Rio Grande	2.3E+06	9.5%			0.0003	3.5%	0.00021	9.5%		

BDL = Below Detection Limits (uncert > 33%)



RG = Rio Grande samples, C = Cochiti Reservoir samples, S = Sandia Canyon, M = Mortandad Canyon, LA = Pueblo/Los Alamos Canyon.

Figure 8. Pu ratio vs Pu activity in Pajarito Plateau sediments.

Laboratory-derived plutonium based on the $^{240}Pu/^{239}Pu$ atom ratios, but the combined $^{240,239}Pu$ activities are relatively low and usually within the background activity range.

There is no distinct indication in the sediment samples of typical modern weapons grade materials with a $^{240}Pu/^{239}Pu$ atom ratio between 0.05 and 0.07. Although the Laboratory has used modern plutonium in its research for approximately three decades, there is little indication of its presence in these environmental samples, in contrast to that seen below Acid Canyon with pre-1960s plutonium. In Mortandad Canyon, part of this may be explained by noting that all of the sediment samples were taken below the sediment traps, where much of the contaminant load is settled. These data suggest that most of the modern plutonium is probably retained in Mortandad Canyon along the three-km reach from the effluent release point to the sediment traps. Below the sediment traps, the plutonium in the samples may be a mixture of global fallout and modern plutonium, but the proportion of modern plutonium is relatively low.

Plutonium-239 levels decline logarithmically with distance from the Acid Canyon source (Figure 9) as LANL-derived plutonium progressively is mixed during downstream transport with cleaner sediments. Along the 20-km reach from Acid Canyon to the Rio Grande $^{240,239}Pu$ activity levels decline one order of magnitude. Despite this decline in overall plutonium levels, the $^{240}Pu/^{239}Pu$ atom ratio isotopic signature is maintained in sediment samples taken along the 20-km

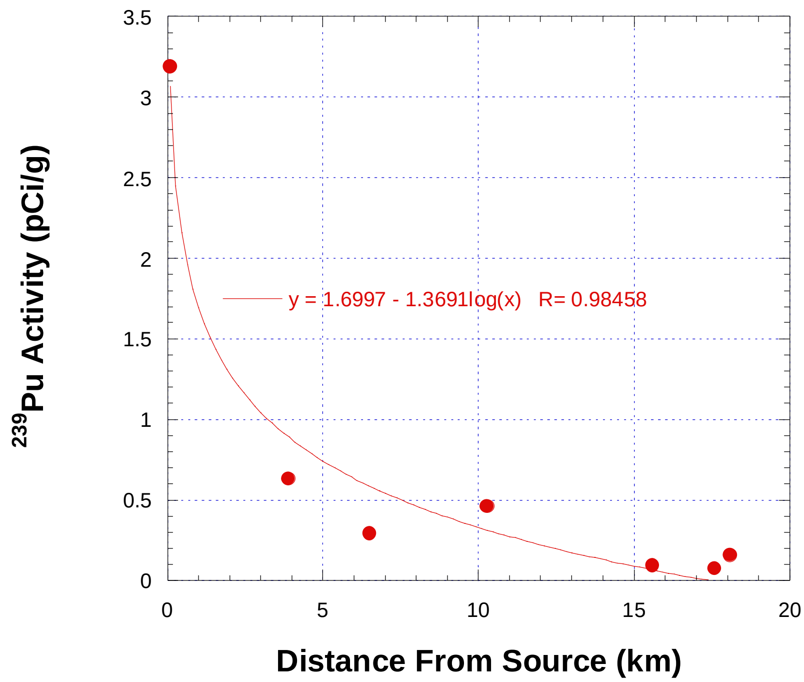


Figure 9. Downstream changes in ^{239}Pu activities in Pueblo/Los Alamos Canyon.

reach below the Acid Canyon source, as shown in Figure 10. We will assume that a ^{240}Pu/^{239}Pu atom ratio of 0.015 is a representative isotopic signature of Laboratory-derived plutonium entering the Rio Grande via the Los Alamos Canyon system.

Figure 11 is a map showing plutonium activities and probable plutonium origins at the Pajarito Plateau sampling sites. Map symbols at the sampling sites are sized proportional to plutonium activities and shaded according to likely plutonium sources, as determined by the three general ranges for ^{240}Pu/^{239}Pu atom ratios described above. Sites impacted most by LANL plutonium are identifiable by the larger-sized symbols and shaded as LANL-influenced.

The TIMS results are generally consistent with historical monitoring data for Pajarito Plateau samples. Median ^{239,240}Pu histories for sediment monitoring stations on the plateau and from the Rio Grande are shown in map perspective in Figure 12. The map is based on data contained in Laboratory annual environmental surveillance reports for years 1973 through 1999. We included all sediment stations having five or more ^{239,240}Pu measurements in this time period. The historic sampling stations provide broader coverage of the plateau than the TIMS data, with denser sampling in Los Alamos Canyon. Plutonium-239, 240 activities greater than regional background levels (the shaded circles in Figure 12) are limited to the Pajarito Plateau. None of the Rio Grande stations had median historic ^{239,240}Pu values greater than the background levels, indicating minimal impact to the Rio Grande.

The data indicate offsite transport of LANL plutonium via Pueblo/Los Alamos, Sandia, and Mortandad Canyons. Movement of LANL plutonium into the Rio Grande, however, can only be traced via the Pueblo/Los Alamos Canyon drainage system. The downstream movement of LANL plutonium in Sandia and Mortandad Canyons is limited, and these canyon systems are not major sources of LANL plutonium to the Rio Grande. We are not aware of a significant LANL source of plutonium in Sandia Canyon, and this finding is not supported by historical data. In Mortandad Canyon, an earlier analysis of TIMS results concluded that offsite movement in the drainage was probably limited to approximately one mile beyond the Laboratory boundary (Gallaher et al. 1997); the Rio Grande lies an additional five miles downstream.

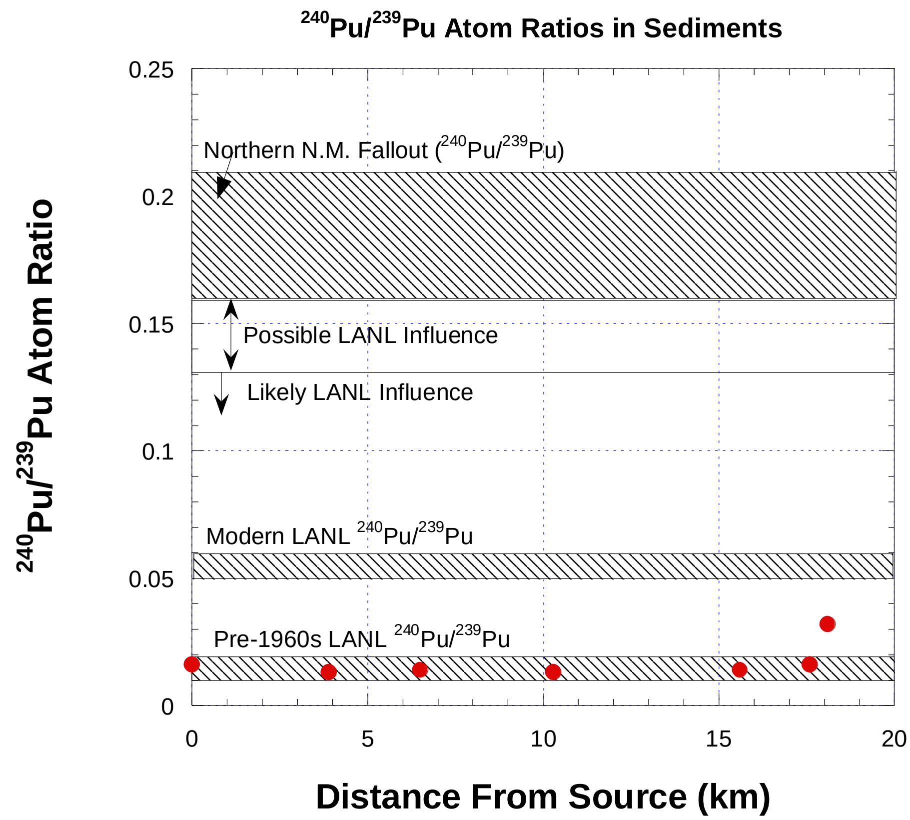


Figure 10. Downstream changes in plutonium atom ratios in Pueblo/Los Alamos Canyon.

Proportion of Laboratory-Derived Plutonium in Sediments

In an attempt to quantify the amount of $^{239,240}Pu$ added to these drainage systems by LANL, we made calculations to separate the plutonium into its LANL component and its global fallout component. Hardy et al. (1972) originally described the method used to separate the plutonium into its two components. A modified form of their equation is presented below:

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

where

$(PuActivity)_L$ = plutonium activity in LANL component,
$(PuActivity)_F$ = plutonium activity in global fallout component,
R_S = $^{240}Pu/^{239}Pu$ atom ratio measured in soil sample,
R_L = 0.015, the $^{240}Pu/^{239}Pu$ atom ratio of plutonium released by LANL,
R_F = 0.169, the $^{240}Pu/^{239}Pu$ atom ratio of global fallout in northern New Mexico, and
3.67 = ratio of half-lives of ^{239}Pu to ^{240}Pu.

As a first approximation for these calculations, we chose the ratios of $^{240}Pu/^{239}Pu$ below Acid Canyon (pre-1960s plutonium) to describe Laboratory releases to all drainages except for Mortandad Canyon. A ratio of 0.015 represents the Acid Canyon source term. Other plutonium sources exist within the Los Alamos Canyon drainage system, but they are not significant when compared with Acid Canyon (Graf 1993; Reneau et al.1998a, b). A $^{240}Pu/^{239}Pu$ atom ratio of

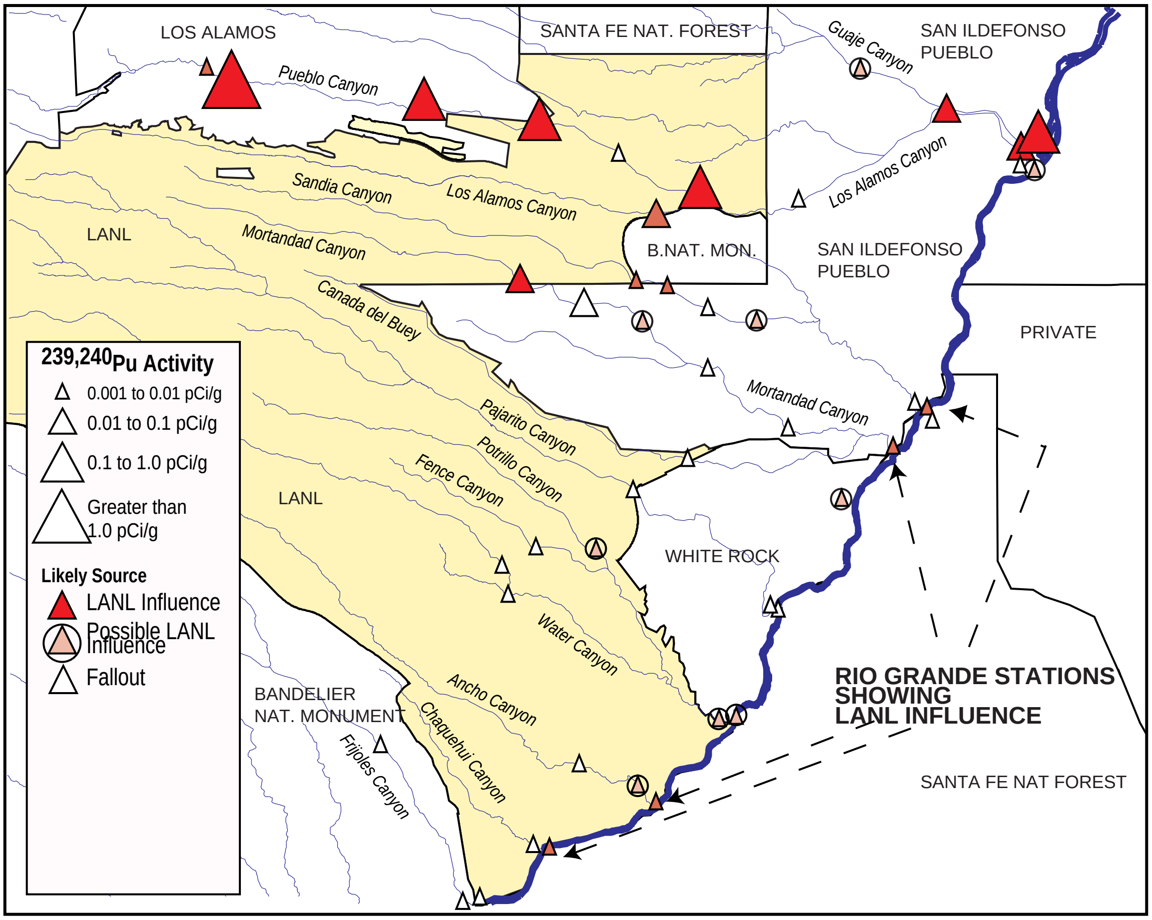


Figure 11. 1994 ^{239,240}Pu activities in sediments from TIMS analyses and likely sources determined by atom ratios.

0.065 describes current Laboratory releases into Mortandad Canyon, as measured just below the RLWTF outfall (Mortandad at GS-1; Gallaher et al. 1997). This ratio is consistent with measurements of modern plutonium at Rocky Flats (Efurd et al. 1993). Because pre-1960s plutonium also was likely released earlier into the upper reaches of Mortandad Canyon, we use a range of results to capture the varied source terms in the canyon. Lastly, we use a fallout ^{240}Pu/^{239}Pu atom ratio of 0.169 for the calculations.

Table 5 presents an estimate of the proportion of Laboratory plutonium that is present at sediment sample stations near LANL. The values assume a simple two-component system. We also assume the average ^{240}Pu/^{239}Pu atom ratio of 0.169 describes fallout, when in reality a range of fallout ratios exists. Some negative values result from using an average value for the end members and are actually zero within the error estimates around the averages.

The calculations show that most samples on the Pajarito Plateau have little or no input of plutonium from the Laboratory, while essentially 100% of the plutonium in the sediments in the Los Alamos Canyon drainage system is from LANL. In Mortandad Canyon up to 81% of the plutonium is LANL-derived in onsite sediments, while plutonium is fallout-derived below the Laboratory boundary at stations A9 and A10.

Along the Rio Grande there is considerable variability in the proportion of LANL plutonium, probably reflecting varied sediment depositional histories at the stations. Laboratory plutonium is

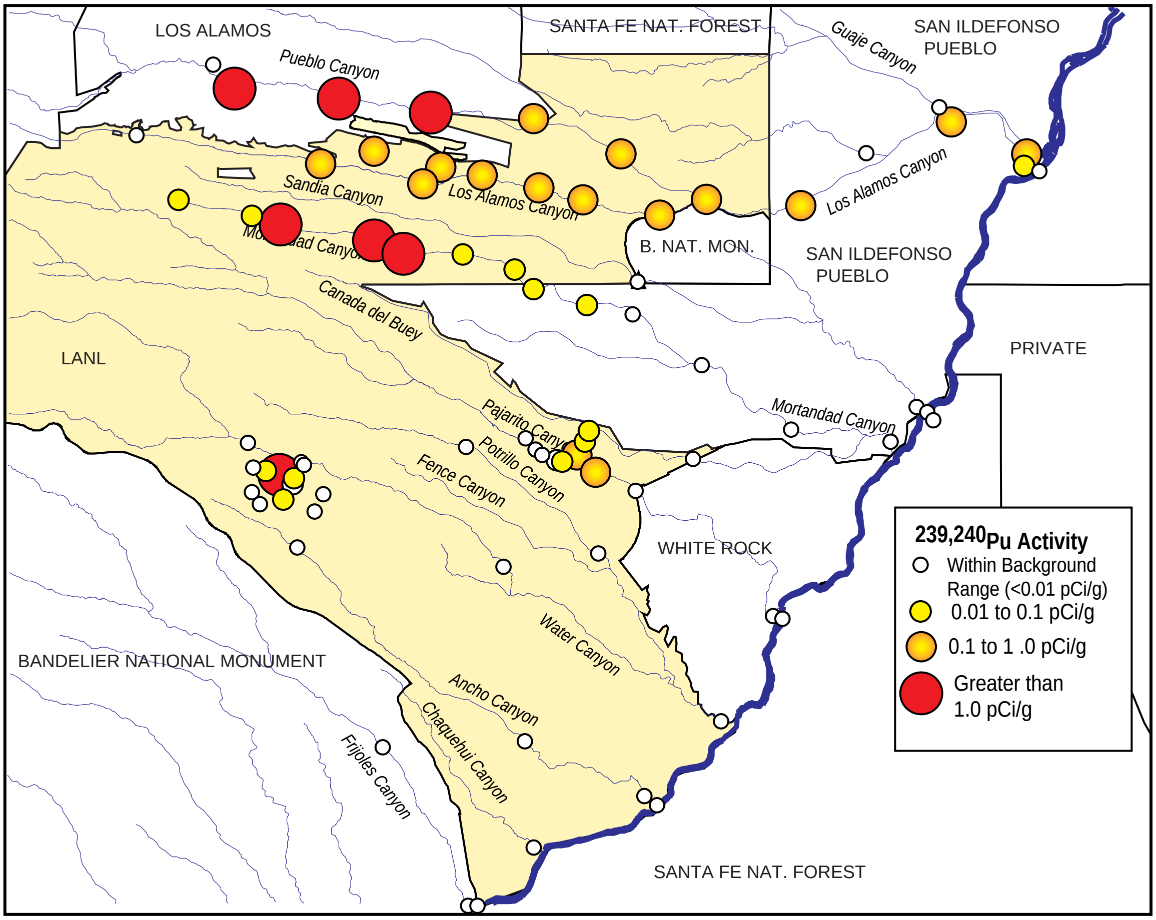


Figure 12. Long-term median ^{239,240}Pu activities in sediments (1970s through 1998).

intermittently indicated along the entire reach between Otowi Bridge and Cochiti Reservoir. At several locations the plutonium is fallout-derived. Where the sediments contain LANL plutonium, between one-third to one-half of the ^{239,240}Pu activity appears to be Laboratory-derived. Averaging the results from the nine Rio Grande sediment locations sampled below the Laboratory shows 78% ± 19% of the plutonium activity is from global fallout, with the remaining 22% ± 19% from LANL. These results compare favorably to an earlier fluvial geomorphology study that concluded global fallout was the source of 90% of the plutonium activity in the northern Rio Grande drainage system and 10% on average was from LANL (Graf 1994). That study calculated the amounts of sediment and plutonium carried by the Rio Grande and tributaries after reviewing historical water and sediment load data.

The shallowest reservoir bottom sediments in Cochiti Reservoir are predominantly fallout derived, with more than 90% of the plutonium activity from fallout, with the remaining an average of 6% from LANL. The deeper sediments collected in a core taken near the dam appear to have a greater proportion of LANL plutonium, however. On a depth-weighted basis, approximately 40% of the plutonium activity in the core segments near the dam is LANL-derived.

Table 5. Percentage of LANL ^{239,240}Pu Activity in Downstream Sediments
Uncertainties shown are 1 Standard Deviation measurement errors.

Station Name	Sample Date	Log No.	% Pu Activity from LANL		
Regional Stations					
Rio Grande at Otowi (bank)	8/17/1995	12971A	0%	+/-	9%
Rio Grande at Otowi (bank)	8/17/1995	12971B	0%	+/-	8%
Rio Grande at Otowi (wdth intgrt)	9/15/1995	13035	21%	+/-	15%
Rio Grande at Sandia	9/27/1994	12666	45%	+/-	11%
Rio Grande at Buckman Flood Plain 2B (0 to 0.5 ft)	1/1/1991	10411	49%	+/-	1%
Rio Grande at Buckman Flood Plain 2B (1 to 1.5 ft)	8/17/1995	12974A	38%	+/-	53%
Rio Grande at Buckman Flood Plain 2B (1 to 1.5 ft)	8/17/1995	12974B	36%	+/-	54%
Rio Grande at Pajarito	9/28/1994	12629	8%	+/-	16%
Rio Grande at Water	9/28/1994	12631	10%	+/-	3%
Rio Grande at Ancho	9/29/1994	12632	27%	+/-	6%
Rio Grande at Chaquehui	9/29/1994	12635	50%	+/-	3%
Rio Grande at Frijoles (bank)	9/29/1994	12577	0%	+/-	4%
Rio Grande at Frijoles (wdth intgrt)	9/13/1995	13034	7%	+/-	20%
Rio Grande at Pena Blanca Flood Plain 2B (0 to 0.5 ft)	8/17/1995	12972	0%	+/-	2%
Reservoirs on Rio Grande					
Cochiti Reservoir:					
Cochiti Upper	9/24/1998	15372	5%	+/-	1%
Cochiti Upper	8/2/1994	12173	16%	+/-	29%
Cochiti Middle	9/24/1998	15373	4%	+/-	1%
Cochiti Lower	9/24/1998	15371	7%	+/-	1%
Core #3 (0 to 26 cm)	9/17/1996	13384	32%	+/-	2%
Core #3 (26 to 52 cm)	9/17/1996	13385	44%	+/-	1%
Core #3 (52 to 78 cm)	9/17/1996	13386	78%	+/-	1%
Core #3 (78 to 91 cm)	9/17/1996	13350	50%	+/-	1%
Core #3 (91 to 104 cm)	9/17/1996	13388	55%	+/-	1%
Core #3 (104 to 117 cm)	9/17/1996	13351	49%	+/-	2%
Core #3 (117 to 130 cm)	9/17/1996	13346	46%	+/-	1%
Core #3 (130 to 143 cm)	9/17/1996	13389	36%	+/-	1%
Core #3 (143 to 156 cm)	9/17/1996	13345	0%	+/-	5%
Surface R1	9/17/1996	13387	10%	+/-	1%

Table 5. cont.

Station Name	Sample Date	Log No.	% Pu Activity from LANL		
Surface B11	9/17/1996	13349	6%	+/-	1%
Surface B2	9/17/1996	13341	4%	+/-	1%
Surface B3.5	9/17/1996	13342	6%	+/-	1%
Surface B4	9/17/1996	13343	6%	+/-	1%
Surface B5	9/17/1996	13344	1%	+/-	1%
Elephant Butte Reservoir:					
Core #4 (12 to 15 cm)	25-Jul-95	13030A	3%	+/-	3%
Core #4 (12 to 15 cm)	25-Jul-95	13030B	3%	+/-	3%
Core #4 (42 to 45 cm)	25-Jul-95	13037	2%	+/-	2%
Core #4 (87 to 90 cm)	25-Jul-95	13031	0%	+/-	2%
Core #4 (111 to 114 cm)	25-Jul-95	13032A	0%	+/-	3%
Core #4 (111 to 114 cm)	25-Jul-95	13032B	0%	+/-	3%
Core #4 (135 to 138 cm)	25-Jul-95	13033	0%	+/-	5%
Core #4 (159 to 162 cm)	25-Jul-95	13038	0%	+/-	1%
Pajarito Plateau Stations					
Guaje Canyon:					
Guaje at SR-502	6/28/1994	12008	21%	+/-	8%
Bayo Canyon:					
Bayo at SR-502	6/3/1994	12012	0%	+/-	9%
Acid/Pueblo Canyons					
Pueblo 1	7/13/1994	12177	37%	+/-	14%
Acid Weir	8/9/1994	12665	99%	+/-	0.1%
Pueblo 2	7/13/1994	12010	100%	+/-	0.2%
Hamilton Bend Spring	7/13/1994	12009	100%	+/-	0.2%
Pueblo 3	7/13/1994	12081	0%	+/-	178%
Pueblo at SR-502	6/3/1994	12016	100%	+/-	0.3%

Table 5. cont.

Station Name	Sample Date	Log No.	% Pu Activity from LANL		
DP/Los Alamos Canyons					
Los Alamos at SR-4	6/3/1994	12068	84%	+/-	1%
Los Alamos at Totavi	6/28/1994	12082	0%	+/-	66%
Los Alamos at LA-2	6/28/1994	12079	100%	+/-	0.3%
Los Alamos at Otowi Floodplain 2B (0 to 0.5 ft)	8/17/1995	12975A	84%	+/-	2%
Los Alamos at Otowi Floodplain 2B (0 to 0.5 ft)	8/17/1995	12975B	83%	+/-	2%
Los Alamos at Otowi	7/14/1994	12080	100%	+/-	1%
Sandia Canyon:					
Sandia at SR-4	6/3/1994	12015	36%	+/-	17%
SSI-1	7/27/1994	12104	46%	+/-	6%
SSI-2	7/27/1994	12116	0%	+/-	10%
SSI-3	7/27/1994	12105	20%	+/-	12%
Sandia at Rio Grande	9/27/1994	12667	0%	+/-	9%
Canada Ancha:					
Canada Ancha at Rio Grande	9/27/1994	12670	37%	+/-	40%
Mortandad Canyon:					
Mortandad at MCO-13 (A-5)	7/27/1994	12115	47% - 81%	+/-	29%
Mortandad A-6	7/27/1994	12103	38% - 66%	+/-	2%
Mortandad A-7	7/27/1994	12101	3% -5%	+/-	3%
Mortandad A-7	11/17/1998	15370	11% -20%	+/-	1%
Mortandad A-8	7/27/1994	12174	14% - 24%	+/-	4%
Mortandad at SR-4 (A-9)	6/3/1994	12013	0%	+/-	30%
Mortandad A-10	7/27/1994	12102	0%	+/-	125%
Mortandad at Rio Grande (A-11)	9/27/1994	12668	35%	+/-	5%
Canada del Buey:					
Canada del Buey at SR-4	6/3/1994	12066	0%	+/-	13%

Table 5. cont.

Station Name	Sample Date	Log No.	% Pu Activity from LANL		
Pajarito Canyon:					
Pajarito at SR-4	6/2/1994	12069	0%	+/-	7%
Pajarito at Rio Grande	9/28/1994	12628	0%	+/-	5%
Above Spring 3	11/9/1994	12663	14%	+/-	7%
Above Spring 4A	11/9/1994	12664	0%	+/-	3%
Potrillo Canyon:					
Potrillo at SR-4	8/9/1994	12014	9%	+/-	8%
Fence Canyon:					
Fence at SR-4	9/7/1994	12007	0%	+/-	1%
Water Canyon:					
Water at SR-4	6/3/1994	12070	0%	+/-	11%
Water at Rio Grande	9/28/1994	12630	10%	+/-	3%
Indio Canyon:					
Indio at SR-4	9/7/1994	12176	0%	+/-	11%
Ancho Canyon:					
Above Ancho Spring	9/28/1994	12634	5%	+/-	6%
Ancho at Rio Grande	9/28/1994	12633	19%	+/-	3%
Chaquehui Canyon:					
Chaquehui at Rio Grande	9/29/1994	12575	4%	+/-	5%
Chaquehui Sed Station 2	9/29/1994	12579	0%	+/-	14%
Chaquehui Sed Station 3	9/29/1994	12578	0%	+/-	2%
Chaquehui Sed Station 3A	9/29/1994	12581	34%	+/-	20%
Chaquehui Sed Station 5	9/29/1994	12580	0%	+/-	9%
Frijoles Canyon:					
Frijoles at Monument HQ	6/28/1994	12067	0%	+/-	16%
Frijoles at Rio Grande	9/29/1994	12576	0%	+/-	8%

URANIUM

Background Uranium Atom Ratios and Concentrations

Natural uranium in its original form contains the following isotopic abundance (Audi and Wapstra 1995):

Isotope	Atom Percent
^{234}U	0.0055
^{235}U	0.720
^{236}U	0.0000
^{238}U	99.2745

The atom ratio of ^{238}U to ^{235}U in naturally occurring uranium is always 137.88. By comparison, enriched uranium may have an ^{238}U to ^{235}U atom ratio below 0.06, while in depleted uranium it may exceed 500.

Due to weathering, the ^{234}U content of uranium may vary slightly in nature. Uranium-234 is a decay product of ^{238}U and, in a closed system, ^{234}U and ^{238}U are in secular equilibrium. Uranium-238 undergoes alpha decay to form ^{234}Th. Thorium-234 undergoes beta decay with a 24.1-day half-life to form ^{234}Pa. Protactinium-234 undergoes beta decay with a 6.7-hour half-life to form ^{234}U. In the natural environment, however, uranium, thorium, and protactinium are chemically different and may be separated by chemical weathering processes. In addition, the alpha decay of ^{238}U may physically damage the mineral containing the uranium. Damage to the mineral's crystal lattice may allow water and air containing carbon dioxide to more readily attack the mineral and increase the availability of the ^{234}U for dissolution. Therefore, naturally occurring uranium may be slightly depleted or enriched in ^{234}U relative to ^{238}U. For example, seawater is 15% enriched in ^{234}U relative to ^{238}U

Uranium-236 is produced by neutron capture on ^{235}U in nuclear reactors. Uranium-236 does not exist in nature, and its presence unambiguously indicates an anthropogenic component of uranium.

For this study, the isotopic composition of the samples is compared against natural uranium, relative to ^{235}U. The ratios $^{236}U/^{235}U$ and $^{238}U/^{235}U$ distinguish natural uranium from anthropogenic components. Because the $^{234}U/^{235}U$ ratio can vary naturally, however, this ratio cannot be used to identify Laboratory-derived uranium. The atom ratios in naturally occurring uranium with ^{234}U and ^{238}U in secular equilibrium are

Isotopes	Natural Uranium Atom Ratio
234/235	0.0079
236/235	0
238/235	137.88

Because ^{236}U does not exist in nature, any sample showing detectable levels of the isotope suggests Laboratory impacts. Laboratory impacts are defined to be present when the $^{236}U/^{235}U$ ratio is more than three standard deviations away from zero.

Samples with $^{238}U/^{235}U$ atom ratios three or more standard deviations from natural indicate Laboratory impacts. Ratios significantly greater than 137.88 typically indicate the admixture of a depleted ^{235}U component with natural uranium, ratios much less than 137.88 include an enriched ^{235}U component.

Background concentrations of total uranium are taken from long-term monitoring of the northern Rio Grande Basin by the Laboratory's Environmental Surveillance Program. A statistical upper limit for regional uranium background concentrations of 4.4 mg/kg was calculated for river sediments (McLin et al. in preparation).

Uranium Atom Ratios and Concentrations near Los Alamos

We collected sediment samples potentially containing a mixture of natural uranium and LANL uranium from locations within and adjacent to the Laboratory. The results obtained by TIMS analyses of these samples are summarized in Figure 13 and Table 6. Table 6 includes the number of standard deviations the measured $^{238}U/^{235}U$ atom ratio is from natural. Positive values typically indicate the admixture of a depleted ^{235}U component with natural uranium, while negative values include an enriched ^{235}U component. Samples greater than three standard deviations from the natural $^{238}U/^{235}U$ atom ratio of 137.88 are considered to contain LANL uranium.

Total uranium concentrations by TIMS analyses in the sediment samples ranged from 0.6 to 10.0 mg/kg. Only two TIMS samples (Bayo at State Route 502, Mortandad at A-7) exceeded regional background ranges.

The preponderance of uranium in sediment samples was of natural uranium isotopic composition (40 of 48 locations, or 83%). Samples showing Laboratory-derived uranium with TIMS were limited to the Pajarito Plateau. None of the samples from the Rio Grande or Cochiti Reservoir showed Laboratory-derived uranium at distinguishable levels.

Depleted uranium was identified along the Laboratory's eastern boundary in several watercourses draining or adjacent to the firing site corridor: Ancho, Chaquehui, Fence, Mortandad, and Water Canyons.

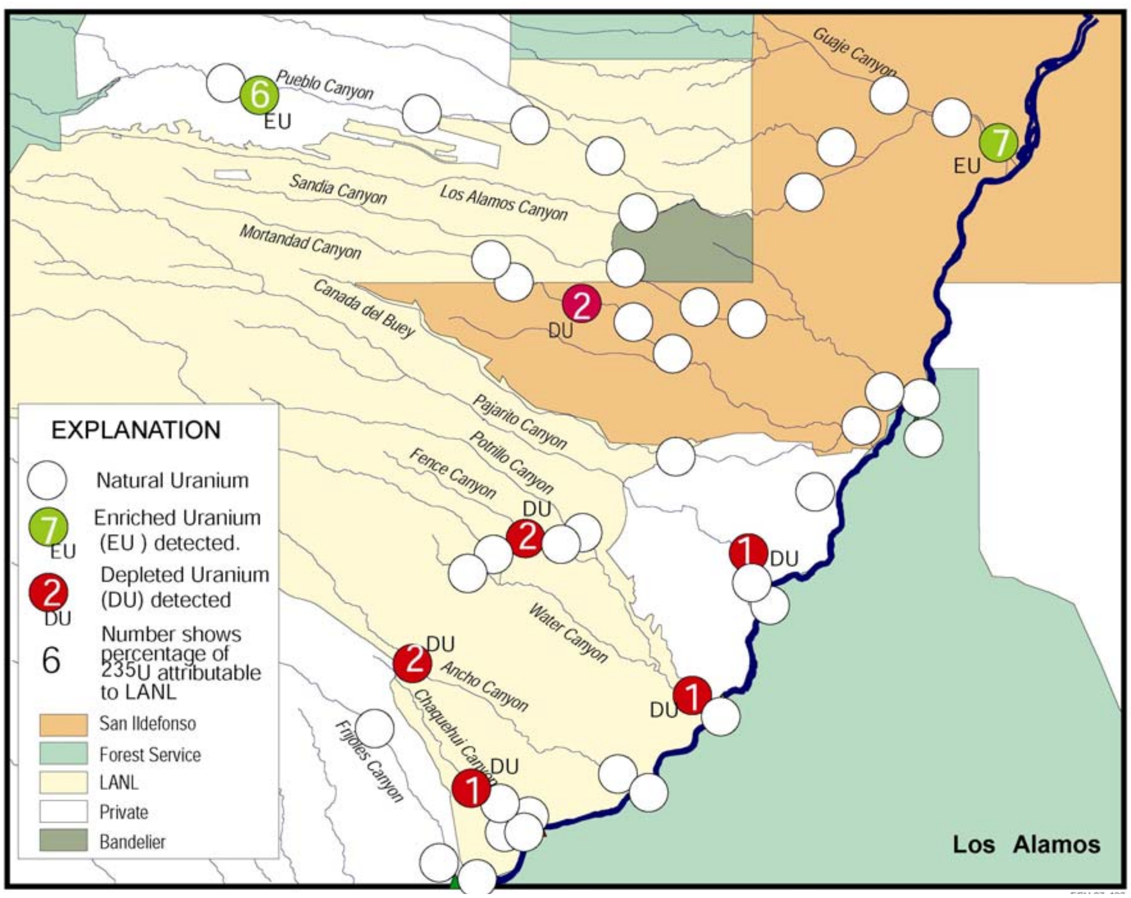


Figure 13. Uranium isotopic composition and source(s) of uranium based on the $^{238}U/^{235}U$ atom ratio.

Table 6. Uranium Isotopic Composition of Sediment Samples (TIMS Analyses). (Uncertainties are 1 standard deviation errors.)

Station Name	Sample Date	Log No.	Sample Size (g)	Total Uranium Conc. (mg/kg)	Atom Ratio $^{234}U/^{235}U$	Uncert.	No. Standard Deviations From Natural	Atom Ratio $^{236}U/^{235}U$	Uncert.	No. Standard Deviations From Natural
Regional Stations										
Rio Chama at Chamita	25-Jul-94	11964	0.584	1.760	0.0075	3.30%	-0.6	8.8E-05	91.15%	1.1
Rio Grande at Sandia	27-Sep-94	12531	2.788	2.628	0.0073	1.62%	-3.3	-2.0E-06	2287.85%	0.0
Rio Grande at Pajarito	28-Sep-94	12560	3.165	2.481	0.0076	8.11%	0.0	1.7E-04	240.53%	0.4
Rio Grande at Water	28-Sep-94	12562	3.348	3.528	0.0075	1.97%	-0.8	-9.2E-05	36.51%	-2.7
Rio Grande at Ancho	29-Sep-94	12563	3.484	2.638	0.0077	1.10%	0.9	9.5E-05	92.69%	1.1
Rio Grande at Frijoles (bank)	29-Sep-94	12569	5.220	2.271	0.0077	1.23%	0.7	8.1E-05	41.02%	2.4
Reservoirs on the Rio Grande										
Cochiti Reservoir:										
Cochiti Upper	2-Aug-94	12528	2.894	1.091	0.0070	3.48%	-2.7	-2.2E-04	56.42%	-1.8
Core #3 (104 to 117 cm)	17-Sep-96	13381			0.0077	1.45%	0.5	-8.7E-07	2595.43%	0.0
Core #3 (117 to 130 cm)	17-Sep-96	13367	1.030	3.856	0.0075	1.68%	-1.4	2.8E-05	119.42%	0.8
Core #3 (143 to 156 cm)	17-Sep-96	13382			0.0074	1.45%	-2.2	8.3E-05	30.22%	3.3
Core #3 (78 to 91 cm)	17-Sep-96	13365	0.969	3.765	0.0073	1.52%	-2.7	-1.2E-05	246.40%	-0.4
Surface B11	17-Sep-96	13364	0.980	3.981	0.0073	1.57%	-3.0	-1.3E-05	210.09%	-0.5
Surface B2	17-Sep-96	13378	0.829	4.123	0.0072	1.82%	-3.5	3.7E-05	76.02%	1.3
Surface B3	17-Sep-96	13362	0.862	4.288	0.0073	1.68%	-2.6	-1.8E-05	178.85%	-0.6
Surface B3.5	17-Sep-96	13379	0.923	3.479	0.0074	1.67%	-2.1	-5.0E-05	49.09%	-2.0
Surface B4	17-Sep-96	13380	1.011	3.999	0.0073	1.60%	-2.5	1.6E-04	18.62%	5.4
Surface B5	17-Sep-96	13363			0.0074	1.94%	-1.4	-1.8E-05	178.85%	-0.6
Pajarito Plateau Stations										
Guaje Canyon:										
Guaje at SR-502	28-Jun-94	11881	2.493	2.731	0.0073	1.85%	-2.2	9.4E-05	74.73%	1.3
Bayo Canyon:										
Bayo at SR-502	3-Jun-94	11945	3.941	10.050	0.0102	6.56%	3.8	1.2E-04	331.08%	0.3
Acid/Pueblo Canyons:										
Pueblo 1	9-Aug-94	12524	2.897	1.499	0.0078	1.99%	0.8	3.6E-05	171.63%	0.6
Acid Weir	9-Aug-94	12525	3.297	2.028	0.0075	2.97%	-0.5	2.1E-04	53.26%	1.9
Pueblo 2	13-Jul-94	11950	3.325	2.565	0.0076	1.67%	-0.4	-9.8E-06	435.91%	-0.2
Hamilton Bend Spring	13-Jul-94	11951	3.690	1.990	0.0076	1.88%	-0.3	1.5E-05	338.38%	0.3
Pueblo 3	13-Jul-94	11952	4.395	1.964	0.0075	2.53%	-0.6	5.6E-05	103.02%	1.0

Table 6. cont.

Station Name	Atom Ratio $^{238}U/^{235}U$	Uncert.	No. Standard Deviations From Natural	Atoms $^{234}U/g$	Uncert.	Atoms $^{235}U/g$	Uncert.	Atoms $^{236}U/g$	Uncert.	Atoms $^{238}U/g$	Uncert.
Regional Stations											
Rio Chama at Chamita	137.00	0.35%	-1.8	2.4E+11	3.29%	3.2E+13	0.25%	2.8E+09	91.15%	4.4E+15	0.25%
Rio Grande at Sandia	138.96	0.35%	2.2	3.4E+11	1.60%	4.8E+13	0.25%	-9.7E+07	-2287.85%	6.6E+15	0.25%
Rio Grande at Pajarito	138.37	0.35%	1.0	3.4E+11	8.10%	4.5E+13	0.25%	7.5E+09	240.53%	6.2E+15	0.25%
Rio Grande at Water	138.66	0.35%	1.6	4.8E+11	1.95%	6.4E+13	0.25%	-5.9E+09	-36.51%	8.9E+15	0.25%
Rio Grande at Ancho	138.67	0.35%	1.6	3.7E+11	1.07%	4.8E+13	0.25%	4.5E+09	92.69%	6.6E+15	0.25%
Rio Grande at Frijoles (bank)	137.52	0.35%	-0.7	3.2E+11	1.21%	4.1E+13	0.25%	3.4E+09	41.02%	5.7E+15	0.25%
Reservoirs on the Rio Grande											
Cochiti Reservoir:											
Cochiti Upper	136.58	0.35%	-2.7	1.4E+11	3.47%	2.0E+13	0.25%	-4.5E+09	-56.42%	2.7E+15	0.25%
Core #3 (104 to 117 cm)	137.60	0.35%	-0.7	5.2E+11	1.81%	6.8E+13	0.25%	-5.9E+07	-2595.43%	9.3E+15	0.25%
Core #3 (117 to 130 cm)	137.73	0.53%	-0.2	5.2E+11	1.64%	7.0E+13	0.39%	2.0E+09	119.42%	1.0E+16	0.25%
Core #3 (143 to 156 cm)	137.60	35.00%	-0.6	4.9E+11	1.43%	6.6E+13	0.25%	5.5E+09	30.22%	9.1E+15	0.25%
Core #3 (78 to 91 cm)	138.10	0.35%	0.4	5.0E+11	1.50%	6.8E+13	0.25%	-8.3E+08	-246.40%	1.0E+16	0.25%
Surface B11	138.58	0.35%	1.4	5.3E+11	1.55%	7.2E+13	0.25%	-9.5E+08	-210.09%	1.1E+16	0.25%
Surface B2	137.23	0.35%	-1.3	5.4E+11	1.81%	7.5E+13	0.25%	2.8E+09	76.02%	8.7E+15	0.25%
Surface B3	137.94	0.35%	0.1	5.7E+11	1.66%	7.8E+13	0.25%	-1.4E+09	-178.85%	1.0E+16	0.25%
Surface B3.5	137.50	0.35%	-0.8	4.7E+11	1.65%	6.4E+13	0.25%	-3.2E+09	-49.08%	9.5E+15	0.25%
Surface B4	138.19	0.35%	0.6	5.3E+11	1.58%	7.3E+13	0.25%	1.1E+10	18.61%	9.7E+15	0.36%
Surface B5	138.80	35.00%	1.8	5.1E+11	1.93%	6.9E+13	0.25%	2.0E+08	985.96%	9.5E+15	0.25%
Pajarito Plateau Stations											
Guaje Canyon:											
Guaje at SR-502	137.31	0.36%	-1.1	3.7E+11	1.83%	5.0E+13	0.26%	4.7E+09	74.73%	6.9E+15	0.25%
Bayo Canyon:											
Bayo at SR-502	134.12	2.03%	-1.4	1.9E+12	6.38%	1.9E+14	1.52%	2.2E+10	331.08%	2.5E+16	1.35%
Acid/Pueblo Canyons:											
Pueblo 1	136.57	0.35%	-2.7	2.1E+11	1.97%	2.8E+13	0.25%	1.0E+09	171.63%	3.8E+15	0.25%
Acid Weir	129.28	0.38%	-17.7	3.0E+11	2.96%	3.9E+13	0.28%	8.1E+09	53.26%	5.1E+15	0.25%
Pueblo 2	138.10	0.35%	0.4	3.5E+11	1.65%	4.7E+13	0.25%	-4.6E+08	-435.91%	6.4E+15	0.25%
Hamilton Bend Spring	138.28	0.35%	0.8	2.7E+11	1.87%	3.6E+13	0.25%	5.3E+08	338.38%	5.0E+15	0.25%
Pueblo 3	136.91	0.35%	-2.0	2.7E+11	2.52%	3.6E+13	0.25%	2.0E+09	103.02%	4.9E+15	0.25%

Table 6. cont.

Station Name	Activity ^{234}U (pCi/g)	Uncert.	Activity ^{235}U (pCi/g)	Uncert.	Activity ^{236}U (pCi/g)	Uncert.	Activity ^{238}U (pCi/g)	Uncert.	Max. % Enriched ^{235}U	Max. % Depleted ^{235}U
Regional Stations										
Rio Chama at Chamita	0.589	3.29%	0.027	0.25%	0.0001	91.15%	0.588	0.25%	0.6%	
Rio Grande at Sandia	0.838	1.60%	0.040	0.25%	0.0000	-2287.85%	0.877	0.25%		0.3%
Rio Grande at Pajarito	0.837	8.10%	0.038	0.25%	0.0002	240.53%	0.828	0.25%		0.1%
Rio Grande at Water	1.171	1.95%	0.054	0.25%	-0.0001	-36.51%	1.178	0.25%		0.2%
Rio Grande at Ancho	0.897	1.07%	0.040	0.25%	0.0001	92.69%	0.881	0.25%		0.2%
Rio Grande at Frijoles (bank)	0.778	1.21%	0.035	0.25%	0.0001	41.02%	0.758	0.25%	0.3%	
Reservoirs on the Rio Grande										
Cochiti Reservoir:										
Cochiti Upper	0.341	3.47%	0.017	0.25%	-0.0001	-56.42%	0.364	0.25%	0.9%	
Core #3 (104 to 117 cm)	1.257	1.42%	0.057	0.25%	0.0000	-2595.43%	1.237	0.25%		
Core #3 (117 to 130 cm)	1.271	1.55%	0.061	0.25%	0.0000	-210.09%	1.328	0.25%	0.1%	
Core #3 (143 to 156 cm)	1.183	1.43%	0.056	0.25%	0.0001	30.22%	1.210	0.25%		
Core #3 (78 to 91 cm)	1.309	1.81%	0.064	0.25%	0.0001	76.02%	1.375	0.25%		0.6%
Surface B11	1.380	1.66%	0.066	0.25%	0.0000	-178.85%	1.431	0.25%		0.2%
Surface B2	1.132	1.65%	0.054	0.25%	-0.0001	-49.08%	1.161	0.25%	0.5%	
Surface B3	1.288	1.58%	0.061	0.25%	0.0003	18.61%	1.334	0.25%		0.2%
Surface B3.5	1.233	1.93%	0.058	0.25%	0.0000	985.96%	1.268	0.25%	0.3%	
Surface B4	1.266	1.64%	0.059	0.39%	0.0000	119.42%	1.287	0.36%		0.9%
Surface B5	1.213	1.50%	0.058	0.25%	0.0000	-246.40%	1.256	0.25%		
Pajarito Plateau Stations										
Guaje Canyon:										
Guaje at SR-502	0.891	1.83%	0.042	0.26%	0.0001	74.73%	0.911	0.25%	0.4%	
Bayo Canyon:										
Bayo at SR-502	4.655	6.38%	0.159	1.52%	0.0006	331.08%	3.353	1.34%	2.7%	
Acid/Pueblo Canyons:										
Pueblo 1	0.520	1.97%	0.023	0.25%	0.0000	171.63%	0.500	0.25%	0.9%	
Acid Weir	0.721	2.96%	0.033	0.28%	0.0002	53.26%	0.677	0.25%	6.2%	
Pueblo 2	0.862	1.65%	0.039	0.25%	0.0000	-435.91%	0.856	0.25%		0.1%
Hamilton Bend Spring	0.668	1.87%	0.030	0.25%	0.0000	338.38%	0.664	0.25%		0.1%
Pueblo 3	0.660	2.52%	0.030	0.25%	0.0001	103.02%	0.655	0.25%	0.7%	

Table 6. cont.

Station Name	Sample Date	Log No.	Sample Size (g)	Total Uranium Conc. (mg/kg)	Atom Ratio $^{234}U/^{235}U$	Uncert.	No. Standard Deviations From Natural	Atom Ratio $^{236}U/^{235}U$	Uncert.	No. Standard Deviations From Natural
DP/Los Alamos Canyons:										
Los Alamos at SR-4	3-Jun-94	11878	3.432	1.618	0.0079	1.94%	1.4	4.7E-07	7563.09%	0.0
Los Alamos at Totavi	28-Jun-94	11873	2.841	1.267	0.0072	1.96%	-2.9	-8.6E-05	62.17%	-1.6
Los Alamos at LA-2	28-Jun-94	11879	4.470	1.610	0.0078	3.96%	0.5	1.0E-04	45.82%	2.2
Los Alamos at Otowi	14-Jul-94	11948	2.821	1.851	0.0077	1.38%	0.6	2.7E-05	132.37%	0.8
Sandia Canyon:										
Sandia at SR-4	3-Jun-94	11872	1.622	0.863	0.0075	2.21%	-0.9	7.3E-05	145.34%	0.7
SSI-2	27-Jul-94	11962	2.841	0.680	0.0077	1.57%	0.5	3.8E-05	115.74%	0.9
Sandia at Rio Grande	27-Sep-94	12532	3.351	1.848	0.0076	1.17%	-0.8	7.8E-05	26.22%	3.8
Canada Ancha:										
Canada Ancha at Rio Grande	27-Sep-94	12530	3.229	1.627	0.0072	1.01%	-5.7	1.2E-04	10.29%	9.7
Mortandad Canyon:										
Mortandad at MCO-13 (A-5)	27-Jul-95	11953	2.130	0.809	0.0077	1.69%	0.2	7.8E-05	60.08%	1.7
Mortandad A-6	27-Jul-94	11960	1.774	1.343	0.0079	3.81%	0.9	3.7E-04	20.47%	4.9
Mortandad A-7	27-Jul-94	12536	2.051	2.351	0.0077	1.47%	0.5	2.8E-04	13.04%	7.7
Mortandad A-7	17-Nov-98	15501		5.680						
Mortandad A-8	27-Jul-94	12537	3.152	2.565	0.0074	1.99%	-1.4	-4.3E-05	148.49%	-0.7
Mortandad at SR-4 (A-9)	3-Jun-94	11880	1.992	1.085	0.0076	2.59%	-0.4	7.3E-06	825.62%	0.1
Mortandad A-10	27-Jul-95	11961	1.344	1.592	0.0075	2.39%	-0.6	-1.2E-04	69.29%	-1.4
Mortandad at Rio Grande (A-11)	27-Sep-94	12533	3.501	2.611	0.0074	1.89%	-1.4	8.8E-05	71.58%	1.4
Canada del Buey:										
Canada del Buey at SR-4	3-Jun-94	11959	3.945	0.900	0.0075	2.30%	-0.9	-4.3E-05	100.82%	-1.0
Pajarito Canyon:										
Pajarito at Rio Grande	28-Sep-94	12559	4.078	1.055	0.0087	3.64%	3.4	-2.1E-04	72.08%	-1.4
Above Spring 3	9-Nov-94	12522	3.055	2.654	0.0098	1.81%	12.2	2.6E-05	168.87%	0.6
Above Spring 4A	9-Nov-94	12523	1.894	1.880	0.0077	1.12%	0.8	1.0E-04	24.99%	4.0
Potrillo Canyon:										
Potrillo at SR-4	2-Jun-94	11882	5.634	0.627	0.0076	1.83%	-0.3	7.6E-06	551.57%	0.2
Fence Canyon:										
Fence at SR-4	7-Sep-94	12527	3.275	3.687	0.0078	2.26%	1.0	6.8E-04	16.22%	6.2
Fence at SR-4	2-Jun-94	11874	2.837	1.029	0.0078	2.96%	0.8	4.4E-05	259.88%	0.4

Table 6. cont.

Station Name	Atom Ratio $^{238}U/^{235}U$	Uncert.	No. Standard Deviations From Natural	Atoms $^{234}U/g$	Uncert	Atoms $^{235}U/g$	Uncert.	Atoms $^{236}U/g$	Uncert.	Atoms $^{238}U/g$	Uncert.
DP/Los Alamos Canyons:											
Los Alamos at SR-4	136.89	0.44%	-1.6	2.3E+11	1.92%	3.0E+13	0.27%	1.4E+07	7563.09%	4.1E+15	0.35%
Los Alamos at Totavi	137.80	0.35%	-0.2	1.7E+11	1.94%	2.3E+13	0.25%	-2.0E+09	-62.17%	3.2E+15	0.25%
Los Alamos at LA-2	138.19	0.35%	0.6	2.3E+11	3.95%	2.9E+13	0.25%	3.0E+09	45.82%	4.0E+15	0.25%
Los Alamos at Otowi	128.18	0.35%	-21.4	2.8E+11	1.36%	3.6E+13	0.25%	9.6E+08	132.37%	4.6E+15	0.25%
Sandia Canyon:											
Sandia at SR-4	138.13	0.35%	0.5	1.2E+11	2.20%	1.6E+13	0.25%	1.1E+09	145.33%	2.2E+15	0.25%
SSI-2	137.87	2.11%	0.0	9.5E+10	1.55%	1.2E+13	0.25%	4.7E+08	115.74%	1.7E+15	2.09%
Sandia at Rio Grande	138.56	0.35%	1.4	2.5E+11	1.14%	3.3E+13	0.25%	2.6E+09	26.22%	4.6E+15	0.25%
Canada Ancha:											
Canada Ancha at Rio Grande	138.81	0.67%	1.0	2.1E+11	0.85%	2.9E+13	0.55%	3.6E+09	10.28%	4.1E+15	0.39%
Mortandad Canyon:											
Mortandad at MCO-13 (A-5)	137.46	0.35%	-0.9	1.1E+11	1.67%	1.5E+13	0.25%	1.2E+09	60.08%	2.0E+15	0.25%
Mortandad A-6	138.46	0.39%	1.1	1.9E+11	3.80%	2.4E+13	0.30%	8.9E+09	20.46%	3.4E+15	0.25%
Mortandad A-7	144.53	0.35%	13.0	3.1E+11	1.44%	4.1E+13	0.25%	1.1E+10	13.03%	5.9E+15	0.25%
Mortandad A-7	139.87	0.15%	9.7	4.4E+11	1.60%	5.9E+13	0.20%	5.9E+09	20.52%	8.2E+15	18.00%
Mortandad A-8	138.52	0.35%	1.3	3.5E+11	1.97%	4.7E+13	0.25%	-2.0E+09	-148.49%	6.4E+15	0.25%
Mortandad at SR-4 (A-9)	138.08	0.35%	0.4	1.5E+11	2.58%	2.0E+13	0.25%	1.4E+08	825.62%	2.7E+15	0.25%
Mortandad A-10	136.71	0.35%	-2.4	2.2E+11	2.37%	2.9E+13	0.25%	-3.5E+09	-69.29%	4.0E+15	0.25%
Mortandad at Rio Grande (A-11)	138.81	0.35%	1.9	3.5E+11	1.87%	4.7E+13	0.25%	4.2E+09	71.58%	6.6E+15	0.25%
Canada del Buey:											
Canada del Buey at SR-4	137.74	0.35%	-0.3	1.2E+11	2.28%	1.6E+13	0.25%	-7.0E+08	-100.82%	2.3E+15	0.25%
Pajarito Canyon:											
Pajarito at Rio Grande	137.96	0.35%	0.2	1.7E+11	3.63%	1.9E+13	0.25%	-4.0E+09	-72.08%	2.6E+15	0.25%
Above Spring 3	137.46	0.36%	-0.8	4.8E+11	1.79%	4.8E+13	0.25%	1.3E+09	168.87%	6.7E+15	0.26%
Above Spring 4A	139.55	0.35%	3.4	2.6E+11	1.09%	3.4E+13	0.25%	3.5E+09	24.99%	4.7E+15	0.25%
Potrillo Canyon:											
Potrillo at SR-4	138.79	0.35%	1.8	8.6E+10	1.81%	1.1E+13	0.25%	8.7E+07	551.57%	1.6E+15	0.25%
Fence Canyon:											
Fence at SR-4	143.56	0.37%	10.8	5.0E+11	2.24%	6.5E+13	0.27%	4.4E+10	16.22%	9.3E+15	0.25%
Fence at SR-4	138.44	0.45%	0.9	1.5E+11	2.94%	1.9E+13	0.37%	8.2E+08	259.88%	2.6E+15	0.25%

Table 6. cont.

Station Name	Activity ^{234}U (pCi/g)	Uncert.	Activity ^{235}U (pCi/g)	Uncert.	Activity ^{236}U (pCi/g)	Uncert.	Activity ^{238}U (pCi/g)	Uncert.	Max. % Enriched ^{235}U	Max. % Depleted ^{235}U
DP/Los Alamos Canyons:										
Los Alamos at SR-4	0.567	1.92%	0.025	0.27%	0.0000	7563.09%	0.540	0.35%	0.7%	
Los Alamos at Totavi	0.407	1.94%	0.019	0.25%	-0.0001	-62.17%	0.423	0.25%	0.1%	
Los Alamos at LA-2	0.555	3.95%	0.025	0.25%	0.0001	45.82%	0.537	0.25%		0.1%
Los Alamos at Otowi	0.680	1.36%	0.031	0.25%	0.0000	132.37%	0.618	0.25%	7.0%	
Sandia Canyon:										
Sandia at SR-4	0.286	2.20%	0.013	0.25%	0.0000	145.33%	0.288	0.25%		0.1%
SSI-2	0.232	1.55%	0.010	0.25%	0.0000	115.74%	0.227	2.08%	0.0%	
Sandia at Rio Grande	0.617	1.14%	0.028	0.25%	0.0001	26.22%	0.617	0.25%		0.2%
Canada Ancha:										
Canada Ancha at Rio Grande	0.517	0.85%	0.025	0.55%	0.0001	10.28%	0.543	0.39%		0.3%
Mortandad Canyon:										
Mortandad at MCO-13 (A-5)	0.276	1.67%	0.012	0.25%	0.0000	60.08%	0.270	0.25%	0.3%	
Mortandad A-6	0.469	3.80%	0.021	0.30%	0.0002	20.46%	0.448	0.25%		0.2%
Mortandad A-7	0.765	1.44%	0.034	0.25%	0.0003	13.03%	0.785	0.25%		1.8%
Mortandad A-7										0.6%
Mortandad A-8	0.841	1.97%	0.039	0.25%	-0.0001	-148.49%	0.856	0.25%		0.2%
Mortandad at SR-4 (A-9)	0.363	2.58%	0.017	0.25%	0.0000	825.62%	0.362	0.25%		0.1%
Mortandad A-10	0.536	2.37%	0.025	0.25%	-0.0001	-69.29%	0.531	0.25%	0.8%	
Mortandad at Rio Grande (A-11)	0.856	1.87%	0.040	0.25%	0.0001	71.58%	0.872	0.25%		0.3%
Canada del Buey:										
Canada del Buey at SR-4	0.299	2.28%	0.014	0.25%	0.0000	-100.82%	0.300	0.25%	0.1%	0.0%
Pajarito Canyon:										
Pajarito at Rio Grande	0.407	3.63%	0.016	0.25%	-0.0001	-72.08%	0.352	0.25%		0.0%
Above Spring 3	1.157	1.79%	0.041	0.25%	0.0000	168.87%	0.886	0.26%	0.3%	
Above Spring 4A	0.635	1.09%	0.029	0.25%	0.0001	24.99%	0.627	0.25%		0.5%
Potrillo Canyon:										
Potrillo at SR-4	0.210	1.81%	0.010	0.25%	0.0000	551.57%	0.209	0.25%		0.3%
Fence Canyon:										
Fence at SR-4	1.226	2.24%	0.054	0.27%	0.0011	16.22%	1.231	0.25%		1.6%
Fence at SR-4	0.356	2.94%	0.016	0.37%	0.0000	259.88%	0.344	0.25%		0.2%

Table 6. cont.

Station Name	Sample Date	Log No.	Sample Size (g)	Total Uranium Conc. (mg/kg)	Atom Ratio $^{234}U/^{235}U$	Uncert.	No. Standard Deviations From Natural	Atom Ratio $^{236}U/^{235}U$	Uncert.	No. Standard Deviations From Natural
Water Canyon:										
Water at SR-4	3-Jun-94	11871	2.766	2.009						
Water at Rio Grande	28-Sep-94	12561	3.166	2.165	0.0073	1.48%	-2.8	2.3E-05	160.97%	0.6
Indio Canyon:										
Indio at SR-4	27-Sep-94	12526	3.157	1.250	0.0077	5.38%	0.1	8.6E-04	29.94%	3.3
Ancho Canyon:										
Ancho at SR-4	2-Jun-94	11875	4.150	0.819	0.0076	1.73%	-0.3	2.6E-04	21.72%	4.6
Ancho at Rio Grande	28-Sep-94	12564	3.030	4.168	0.0077	0.82%	1.0	2.5E-04	7.81%	12.8
Chaquehui Canyon:										
Chaquehui at Rio Grande	29-Sep-94	12567	5.460	1.481	0.0077	1.47%	0.7	1.1E-04	34.51%	2.9
Chaquehui Sed Station 2	29-Sep-94	12571	4.384	2.826	0.0078	1.23%	1.2	4.6E-05	70.02%	1.4
Chaquehui Sed Station 3	29-Sep-94	12570	4.466	1.979	0.0077	2.42%	0.4	7.4E-05	99.72%	1.0
Chaquehui Sed Station 3A	29-Sep-94	12573	3.780	2.078	0.0077	2.94%	0.1	2.9E-05	165.78%	0.6
Chaquehui Sed Station 5	29-Sep-94	12572	4.062	3.935	0.0076	1.94%	-0.3	2.8E-05	127.07%	0.8
Frijoles Canyon:										
Frijoles at Monument HQ	28-Jun-94	11957	2.952	3.437	0.0077	1.60%	0.5	9.4E-05	55.76%	1.8
Frijoles at Rio Grande	29-Sep-94	12568	5.093	1.157	0.0079	1.63%	1.7	-6.7E-05	77.51%	-1.3

Table 6. cont.

Station Name	Atom Ratio $^{238}U/^{235}U$	Uncert.	No. Standard Deviations From Natural	Atoms $^{234}U/g$	Uncert	Atoms $^{235}U/g$	Uncert.	Atoms $^{236}U/g$	Uncert.	Atoms $^{238}U/g$	Uncert.
Water Canyon:											
Water at SR-4	143.09	2.00%	1.8	0.0E+00	0.00%	3.5E+13	1.94%	0.0E+00	0.00%	5.0E+15	0.48%
Water at Rio Grande	140.16	0.35%	4.6	2.8E+11	1.46%	3.9E+13	0.25%	8.8E+08	160.97%	5.4E+15	0.25%
Indio Canyon:											
Indio at SR-4	136.51	0.46%	-2.2	1.8E+11	5.37%	2.3E+13	0.39%	2.0E+10	29.93%	3.1E+15	0.25%
Ancho Canyon:											
Ancho at SR-4	143.57	0.35%	11.2	1.1E+11	1.71%	1.4E+13	0.25%	3.7E+09	21.72%	2.1E+15	0.25%
Ancho at Rio Grande	139.33	0.37%	2.8	5.8E+11	0.78%	7.5E+13	0.25%	1.9E+10	7.81%	1.0E+16	0.27%
Chaquehui Canyon:											
Chaquehui at Rio Grande	138.41	0.35%	1.1	2.1E+11	1.45%	2.7E+13	0.25%	3.0E+09	34.51%	3.7E+15	0.25%
Chaquehui Sed Station 2	139.87	0.35%	4.0	3.9E+11	1.21%	5.1E+13	0.25%	2.3E+09	70.02%	7.1E+15	0.25%
Chaquehui Sed Station 3	138.15	0.35%	0.5	2.8E+11	2.40%	3.6E+13	0.25%	2.7E+09	99.72%	5.0E+15	0.25%
Chaquehui Sed Station 3A	138.78	0.35%	1.8	2.9E+11	2.93%	3.8E+13	0.25%	1.1E+09	165.78%	5.2E+15	0.25%
Chaquehui Sed Station 5	139.31	0.38%	2.7	5.4E+11	1.92%	7.1E+13	0.28%	2.0E+09	127.07%	9.9E+15	0.25%
Frijoles Canyon:											
Frijoles at Monument HQ	137.41	0.35%	-1.0	4.8E+11	1.58%	6.3E+13	0.25%	5.9E+09	55.76%	8.6E+15	0.25%
Frijoles at Rio Grande	137.48	0.35%	-0.8	1.7E+11	1.61%	2.1E+13	0.25%	-1.4E+09	-77.51%	2.9E+15	0.25%

Table 6. cont.

Station Name	Activity ^{234}U (pCi/g)	Uncert.	Activity ^{235}U (pCi/g)	Uncert.	Activity ^{236}U (pCi/g)	Uncert.	Activity ^{238}U (pCi/g)	Uncert.	Max. % Enriched ^{235}U	Max. % Depleted ^{235}U
Water Canyon:										
Water at SR-4	0.000	0.00%	0.030	1.94%	0.0000	0.00%	0.671	0.48%		1.4%
Water at Rio Grande	0.693	1.46%	0.033	0.25%	0.0000	160.97%	0.723	0.25%		0.6%
Indio Canyon:										
Indio at SR-4	0.430	5.37%	0.019	0.39%	0.0005	29.93%	0.417	0.25%	1.0%	
Ancho Canyon:										
Ancho at SR-4	0.265	1.71%	0.012	0.25%	0.0001	21.72%	0.273	0.25%		1.6%
Ancho at Rio Grande	1.408	0.78%	0.063	0.25%	0.0005	7.81%	1.391	0.27%		0.4%
Chaquehui Canyon:										
Chaquehui at Rio Grande	0.504	1.45%	0.023	0.25%	0.0001	34.51%	0.494	0.25%		0.1%
Chaquehui Sed Station 2	0.958	1.21%	0.043	0.25%	0.0001	70.02%	0.943	0.25%		0.5%
Chaquehui Sed Station 3	0.676	2.40%	0.030	0.25%	0.0001	99.72%	0.660	0.25%		0.1%
Chaquehui Sed Station 3A	0.702	2.93%	0.032	0.25%	0.0000	165.78%	0.694	0.25%		0.2%
Chaquehui Sed Station 5	1.311	1.92%	0.060	0.28%	0.0000	127.07%	1.313	0.25%		0.4%
Frijoles Canyon:										
Frijoles at Monument HQ	1.177	1.58%	0.053	0.25%	0.0002	55.76%	1.147	0.25%	0.3%	
Frijoles at Rio Grande	0.404	1.61%	0.018	0.25%	0.0000	-77.51%	0.386	0.25%	0.3%	

There is evidence that some of the depleted uranium may originate from airborne deposition, probably from dynamic experimentation in the firing site corridor, rather than from streamwater transport. TIMS analyses conducted for this and another related study (McNaughton et al. 1999) show measurable amounts of depleted uranium at three widely spaced locations that do not appear to be related to water-borne sources:

- depleted uranium is indicated in two samples from Mortandad Canyon collected just below the Laboratory boundary (both from station A7), but not indicated in other upstream Mortandad sediment stations (Gallaher et al. 1997). The $^{238}U/^{235}U$ atom ratios were 144 ± 0.7 and 140 ± 0.2, which differ from natural uranium by 13 and 10 standard deviations, respectively.
- depleted uranium is indicated in a soil sample collected from the escarpment of White Rock Canyon, at a location above Spring 4A (Figure 4) and removed from noticeable surface drainage. The $^{238}U/^{235}U$ atom ratio was 140 ± 0.5, which differs from natural uranium by 3 standard deviations.
- A preliminary measurement of depleted uranium was made on a sample of pine needles collected from the central portion of the Laboratory (McNaughton et al. 1999). The $^{238}U/^{235}U$ atom ratio was 165, with a standard deviation of ± 4, which differs from natural uranium by 7 standard deviations.

Enriched uranium was measured in the Pueblo/Los Alamos Canyon drainage system sediments at two locations: Acid Weir, near the historical TA-45 discharge point in Acid Canyon, and in Los Alamos Canyon near the confluence with the Rio Grande at Otowi (Figure 3). The $^{238}U/^{235}U$ atom ratios at these locations were 129 ± 0.6 and 128 ± 0.5, which differ from natural uranium by 18 and 21 standard deviations. The enriched uranium is from water transport, as it is not used in firing site tests. These results are consistent with the detection of enriched uranium in shallow groundwater samples from Pueblo and Los Alamos Canyons (Gallaher and Efurd, in preparation).

The TIMS results for Pajarito Plateau samples are generally consistent with past LANL sediment monitoring results. Uranium concentrations measured at sediment monitoring stations within the eastern portion of the plateau are shown in Figure 14. The graph is based on data contained in Laboratory annual environmental surveillance reports for years 1973 through 1999. Most of the stations included in the graph are located near the Laboratory's eastern boundary, from the Rio Grande upstream to State Route 4 and State Route 502. These stations best describe the levels of uranium entering the Rio Grande from the Laboratory. To put the LANL canyon data in context, we also include uranium levels for Guaje and Rendija Canyons, located north of LANL, and Frijoles Canyon, located south of LANL.

Figure 14 shows that the vast majority of the historical total uranium concentrations are within background levels reported for Rio Grande sediments (McLin et al. 2002). However, samples collected in the 1970s from Pueblo Canyon significantly exceed background levels and suggest an early LANL source in that drainage, and likely also in lower Los Alamos Canyon. This finding is consistent with the TIMS analyses showing enriched uranium near Acid Canyon and near the mouth of Los Alamos Canyon at the Rio Grande.

In Frijoles Canyon, several historical uranium measurements above Rio Grande levels are indicated in Figure 14, but the source of elevated uranium levels is not definitive. The TIMS analyses of Frijoles Canyon stream sediments show natural uranium composition. However, depleted uranium was detected in a surface water sample taken from Frijoles Creek at the Rio Grande (Gallaher and Efurd, in preparation). Given the proximity of Frijoles Canyon to the firing site corridor, it is possible that small quantities of depleted uranium have been carried there from LANL by wind.

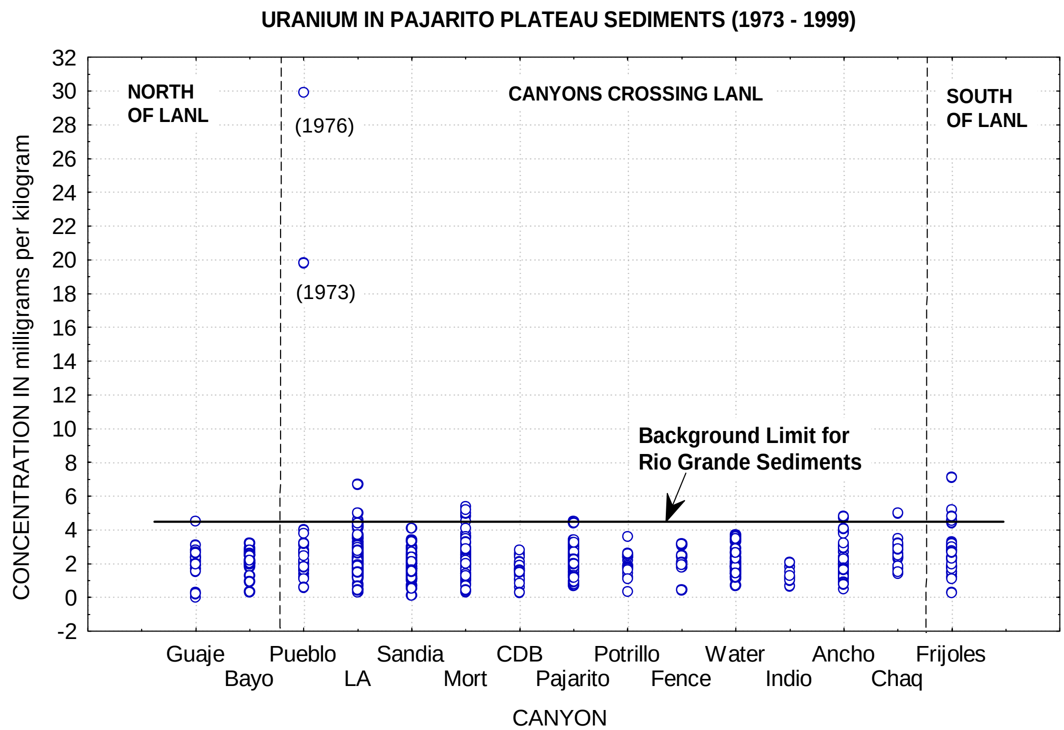


Figure 14. Total uranium concentrations in Pajarito Plateau sediments.
The circles show individual measurements.

Unlike the results for plutonium, none of the TIMS uranium sediment results from the Rio Grande or Cochiti Reservoir show identifiable Laboratory impacts, based on the $^{238}U/^{235}U$ atom ratios. These data indicate that the mass of Laboratory-derived uranium entering the Rio Grande is small relative to the natural levels in stream sediments and soils. The Rio Grande sediments contain abundant natural uranium that obscures the anthropogenic signatures.

Uranium histories for river and reservoir sediments along the Rio Chama and Rio Grande (Figures 15 and 16) are generally consistent the TIMS results. Most of the historical total uranium concentrations are within regional background ranges calculated by McLin et al. (2002). A slight LANL impact is suggested in the Rio Grande sediments along possibly about a 10-km reach below the confluence with Los Alamos Canyon. Median uranium concentrations along this reach increase approximately 30% from upstream stations (2.9 mg/kg at Sandia and Pajarito stations vs 2.2 mg/kg at Otowi station). The increases in uranium concentrations from the Otowi station are statistically significant at Sandia (ANOVA, $p = 0.009$) and at Pajarito (ANOVA, $p = 0.03$). Natural differences in soil texture or mineralization also could potentially cause the uranium concentration increases. Regardless of the source(s) of the increases, the concentrations generally are within natural levels for the region. Farther downstream, median uranium concentrations along the next 40-plus km to Bernalillo are comparable to upstream stations.

Proportion of Laboratory-derived Uranium in Sediments

Some sediment samples from the southern canyons on the Pajarito Plateau were depleted in ^{235}U and some samples from the more northern Los Alamos Canyon drainage were enriched in ^{235}U. Here we estimate the proportion of ^{235}U that is attributable to LANL using the following equation, after Efurd et al. (1993):

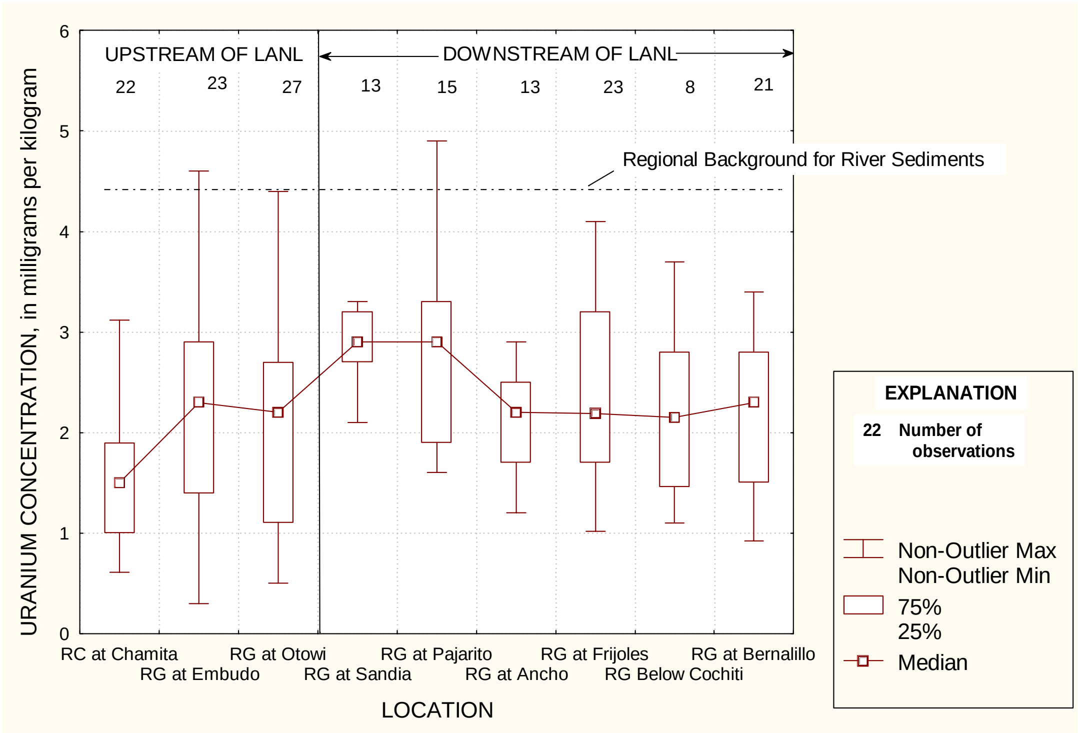


Figure 15. Total uranium concentrations in Rio Grande and Rio Chama river sediments.

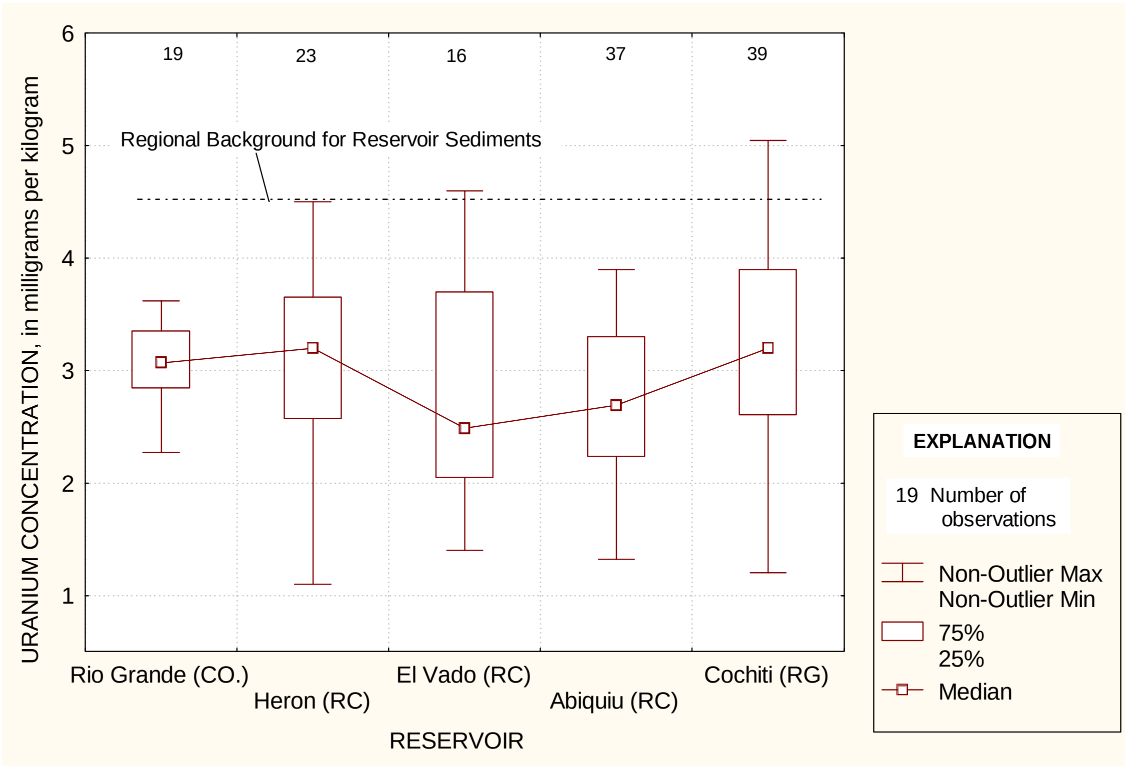


Figure 16. Total uranium concentrations in Rio Grande and Rio Chama reservoir sediments.

$$\left(N238/N235\right)_{Obs} = \left\{\left(N238/N235\right)_{DepOrEnr}\right\} \times \{F\} + \left\{\left(N238/N235\right)_{Nat}\right\} \times \{1-F\} \quad ,$$

where

$\left(N238/N235\right)_{Obs}$ is the $^{238}U/^{235}U$ atom ratio measured in the sample,

$\left\{\left(N238/N235\right)_{DepOrEnr}\right\}$ is the $^{238}U/^{235}U$ atom ratio in the depleted or enriched uranium,

$\left\{\left(N238/N235\right)_{Nat}\right\}$ is the $^{238}U/^{235}U$ atom ratio in natural uranium,

$\{F\}$ is the fraction of depleted or enriched uranium in the sample, and

$\{1-F\}$ is the fraction of the sample that is naturally occurring.

This equation can be solved to provide an estimate of the proportion of depleted or enriched uranium present in the samples.

Table 7 and Figure 13 illustrate the maximum amount of depleted or enriched uranium that is present in each sediment sample. The values assume a simple two component system. Samples with a $^{238}U/^{235}U$ atom ratio larger than natural uranium are assumed to be an admixture of natural uranium and depleted uranium, while samples with low ratios are assumed to contain natural uranium and enriched uranium. The data reported in Table 7 assume that the depleted uranium released by LANL contains 0.2% ^{235}U ($^{238}U/^{235}U$ atom ratio of ~500) and the enriched uranium released by LANL contains 95% ^{235}U ($^{238}U/^{235}U$ atom ratio of ~0.06).

Table 7 and Figure 13 show that the fraction of uranium in sediments that is attributable to release of uranium at the Laboratory is small. In most cases, depleted or enriched uranium comprises less than 1% of the uranium in sediments. The maximum proportion identified of enriched uranium was 7% in Los Alamos Canyon (at Otowi), while the maximum depleted uranium was less than 2% (Mortandad A-7).

CONCLUSIONS

The stream sediments on LANL property and the stream and reservoir sediments of the Rio Grande below the Laboratory have measurable concentrations of the plutonium isotopes ^{239}Pu and ^{240}Pu derived from Laboratory operations. Plutonium isotopes in Rio Grande sediments are not at levels known to adversely affect public health. Offsite movement of plutonium is most apparent in Pueblo/Los Alamos Canyons, with limited offsite movement in Sandia and Mortandad Canyons. Movement of LANL plutonium into the Rio Grande can be traced only via the Pueblo/Los Alamos Canyon drainage system. This means that significant quantities of LANL plutonium enter the Rio Grande only as a result of Manhattan Project and early Cold War operations. The Laboratory plutonium in the Rio Grande is from liquid effluent discharges that occurred during the early days of the Laboratory in the 1940s and 1950s.

Examination of the atom ratio $^{240}Pu/^{239}Pu$ reveals that most sampled locations on the Pajarito Plateau have little or no input of plutonium from the Laboratory, while in Pueblo and Los Alamos Canyons essentially 100% of the plutonium activity in sediments is derived from Laboratory operations.

In the Rio Grande below the Laboratory, the largest proportion of the plutonium in the stream and reservoir sediments is from global atmospheric fallout, rather than from Laboratory sources. Laboratory-derived plutonium in the Rio Grande is identifiable intermittently along the 35-km reach from Otowi Bridge to Cochiti Reservoir. Where the sediments have been impacted by Laboratory operations, between one-third to one-half of the plutonium activity is attributable to the Laboratory. Averaging the results from the nine Rio Grande river sediment locations

Table 7. Maximum Percentage of Uranium in Sediments that is Attributable to Release of Enriched or Depleted Uranium at LANL

Station Name	Date	Log No.	% 235 Enriched U	% 235 Depleted U
Mortandad A-7	7/27/1994	12536		2%
Ancho at SR-4	6/2/1994	11875		2%
Fence at SR-4	9/7/1994	12527		2%
Water at SR-4	6/3/1994	11871		1%
Water at Rio Grande	9/28/1994	12561		0.6%
Mortandad A-7	11/17/1998	15501		0.6%
Chaquehui Sed Station 2	9/29/1994	12571		0.5%
Spring 4A	11/9/1994	12523		0.5%
Ancho at Rio Grande	9/28/1994	12564		0.4%
Chaquehui Sed Station 5	9/29/1994	12572		0.4%
Rio Grande at Sandia	9/27/1994	12531		0.3%
Canada Ancha at Rio Grande	9/27/1994	12530		0.3%
Mortandad at Rio Grande (A-11)	9/27/1994	12533		0.3%
Potrillo at SR-4	6/2/1994	11882		0.3%
Chaquehui Sed Station 3A	9/29/1994	12573		0.2%
Rio Grande at Ancho	9/29/1994	12563		0.2%
Rio Grande at Water	9/28/1994	12562		0.2%
Cochiti Resv. Surface B11	9/17/1996	13364		0.2%
Sandia at Rio Grande	9/27/1994	12532		0.2%
Mortandad A-8	7/27/1994	12537		0.2%
Mortandad A-6	7/27/1994	11960		0.2%
Fence at SR-4	6/2/1994	11874		0.2%
Chaquehui at Rio Grande	9/29/1994	12567		0.1%
Chaquehui at Rio Grande	9/29/1994	12567		0.1%
Rio Grande at Pajarito	9/28/1994	12560		0.1%
Hamilton Bend Spring	7/13/1994	11951		0.1%
Los Alamos at LA-2	6/28/1994	11879		0.1%
Cochiti Resv. Surface B4	9/17/1996	13380		0.1%
Chaquehui Sed Station 3	9/29/1994	12570		0.1%
Sandia at SR-4	6/3/1994	11872		0.1%
Cochiti Resv. Core #3 (78 to 91 cm)	9/17/1996	13365		0.1%
Pueblo 2	7/13/1994	11950		0.1%
Mortandad at SR-4 (A-9)	6/3/1994	11880		0.1%
Pajarito at Rio Grande	9/28/1994	12559		0.02%
Cochiti Resv. Surface B3	9/17/1996	13362		0.02%
SSI-2	7/27/1994	11962	0.01%	
Los Alamos at Totavi	6/28/1994	11873	0.1%	
Canada del Buey at SR-4	6/3/1994	11959	0.1%	
Cochiti Resv. Core #3 (117 to 130 cm)	9/17/1996	13367	0.1%	
Rio Grande at Frijoles (bank)	9/29/1994	12569	0.3%	
Cochiti Resv. Surface B3.5	9/17/1996	13379	0.3%	
Frijoles at Rio Grande	9/29/1994	12568	0.3%	
Spring 3	11/9/1994	12522	0.3%	
Mortandad at MCO-13 (A-5)	7/27/1995	11953	0.3%	
Frijoles at Monument HQ	6/28/1994	11957	0.3%	
Guaje at SR-502	6/28/1994	11881	0.4%	
Cochiti Resv. Surface B2	9/17/1996	13378	0.5%	
Rio Chama at Chamita	7/25/1994	11964	0.6%	
Pueblo 3	7/13/1994	11952	0.7%	
Los Alamos at SR-4	6/3/1994	11878	0.7%	
Mortandad A-10	7/27/1995	11961	0.8%	
Cochiti Upper	8/2/1994	12528	0.9%	
Pueblo 1	8/9/1994	12524	0.9%	
Indio at SR-4	9/27/1994	12526	1%	
Bayo at SR-502	6/3/1994	11945	3%	
Acid Weir	9/8/1994	12525	6%	
Los Alamos at Otowi	7/14/1994	11948	7%	

sampled below the Laboratory shows 78% ± 19% of the plutonium activity is from global fallout, with the remaining 22% ± 19% from LANL. These results compare favorably to an earlier geology-based study that concluded global fallout was the source of 90% of the plutonium activity in the northern Rio Grande drainage system (Graf 1994). Reservoir sediments collected in the bottom of Cochiti Reservoir appear to contain approximately similar proportions of LANL plutonium. The shallowest 6 inches of the sediments in the reservoir contained an average of only 6% LANL plutonium, while a core sample of the entire accumulated thickness taken near the dam contained 40% LANL plutonium, on a depth weighted basis.

The vast majority of the stream and reservoir sediment samples analyzed are of natural uranium isotopic composition and within natural concentration ranges. Anthropogenic uranium is only identifiable in several watercourses on the Pajarito Plateau, but its abundance is low. Enriched uranium in Los Alamos, Pueblo, and Mortandad Canyons appears to be from effluent discharges. Depleted uranium in Ancho, Fence, Water, and Chaquehui Canyons appears to be derived from water and airborne transport from detonations in the firing site corridor. In most cases, uranium that can be attributed to the Laboratory comprises less than 1% of the total uranium in sediments; the maximum LANL proportion identified is 7%. Unlike the results for plutonium, none of the sediments from the Rio Grande or Cochiti Reservoir show identifiable Laboratory uranium, based on uranium isotope ratios.

This study shows that after 50-plus years of operation, the concentrations of plutonium and uranium in the Rio Grande outside the boundaries of LANL are relatively low and in many cases near concentrations arising from fallout plutonium or from naturally-occurring uranium.

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Appendix

Table A1. Summary Statistics for ^{239,240}Pu and Uranium Levels in Northern New Mexico and Southern Colorado Sediments, 1973–1999

Summary Table of Means
N=2072 (No missing data in dep. var. list)
Plutonium-239,240 (picocuries per gram)

Station Name	Location	N	Means	Std.Dev.	Q25	Median	Q75	Background	Ratio: Mean /Background	Ratio: Median /Background	Estimated % LANL Pu Activity)[a]
AB-1	Pajarito Plateau	16	0.0196	0.0259	0.0055	0.0150	0.0206	0.013	1.51	1.15	13%
AB-10	Pajarito Plateau	14	0.0101	0.0127	0.0030	0.0060	0.0110	0.013	0.78	0.46	0%
AB-11	Pajarito Plateau	14	0.0503	0.1431	0.0040	0.0063	0.0140	0.013	3.87	0.48	0%
AB-2	Pajarito Plateau	14	0.0366	0.0293	0.0090	0.0310	0.0520	0.013	2.81	2.38	58%
AB-3	Pajarito Plateau	16	1.5861	2.5250	0.3575	1.0819	1.6975	0.013	122.01	83.23	99%
AB-4	Pajarito Plateau	14	0.0276	0.0550	0.0070	0.0135	0.0210	0.013	2.13	1.03	3%
AB-4A	Pajarito Plateau	13	0.0137	0.0075	0.0090	0.0140	0.0172	0.013	1.05	1.08	7%
AB-5	Pajarito Plateau	14	0.0205	0.0155	0.0060	0.0150	0.0325	0.013	1.58	1.15	13%
AB-6	Pajarito Plateau	14	0.0162	0.0171	0.0060	0.0113	0.0130	0.013	1.25	0.87	0%
AB-7	Pajarito Plateau	14	0.0106	0.0070	0.0040	0.0101	0.0160	0.013	0.81	0.78	0%
AB-8	Pajarito Plateau	14	0.0050	0.0048	0.0020	0.0035	0.0060	0.013	0.39	0.27	0%
AB-9	Pajarito Plateau	16	0.0110	0.0125	0.0030	0.0079	0.0150	0.013	0.85	0.60	0%
AC-3	Pajarito Plateau	8	347.4300	279.8030	104.3400	297.4800	519.4800	0.013	26725.38	22883.08	100%
Acid Weir	Pajarito Plateau	30	27.5274	62.8189	6.4600	7.5550	11.8000	0.013	2117.49	581.15	100%
Ancho at Rio Grande	Pajarito Plateau	20	0.0047	0.0045	0.0020	0.0030	0.0065	0.013	0.36	0.23	0%
Ancho at SR-4	Pajarito Plateau	23	0.0037	0.0033	0.0020	0.0030	0.0060	0.013	0.29	0.23	0%
Bayo at SR-502	Pajarito Plateau	23	0.0029	0.0034	0.0010	0.0020	0.0030	0.013	0.22	0.15	0%
Canada Ancha at Rio Grande	Pajarito Plateau	14	0.0016	0.0029	0.0010	0.0010	0.0020	0.013	0.12	0.08	0%
Canada del Buey at SR-4	Pajarito Plateau	24	0.0053	0.0063	0.0020	0.0030	0.0063	0.013	0.41	0.23	0%
Chaquehui at Rio Grande	Pajarito Plateau	16	0.0154	0.0170	0.0040	0.0070	0.0266	0.013	1.18	0.54	0%
DPS-1	Pajarito Plateau	33	1.6100	3.1313	0.0246	0.1550	1.1460	0.013	123.84	11.92	92%
DPS-4	Pajarito Plateau	34	0.3226	0.2129	0.1490	0.3480	0.4240	0.013	24.82	26.77	96%
Fence at SR-4	Pajarito Plateau	8	0.0112	0.0082	0.0047	0.0105	0.0150	0.013	0.86	0.81	0%
Frijoles at Monument HQ	Pajarito Plateau	23	0.0038	0.0024	0.0020	0.0030	0.0050	0.013	0.29	0.23	0%
Frijoles at Rio Grande	Pajarito Plateau	20	0.0054	0.0061	0.0020	0.0040	0.0065	0.013	0.41	0.31	0%
G-1	Pajarito Plateau	19	0.0082	0.0080	0.0030	0.0050	0.0110	0.013	0.63	0.38	0%
G-2	Pajarito Plateau	18	0.0077	0.0076	0.0030	0.0045	0.0120	0.013	0.59	0.35	0%
G-3	Pajarito Plateau	20	0.0121	0.0069	0.0075	0.0120	0.0150	0.013	0.93	0.92	0%
G-4	Pajarito Plateau	17	0.0346	0.0395	0.0150	0.0190	0.0370	0.013	2.66	1.46	32%
G-5	Pajarito Plateau	19	0.0694	0.1476	0.0090	0.0390	0.0570	0.013	5.34	3.00	67%
G-6	Pajarito Plateau	17	0.1618	0.1469	0.0400	0.1500	0.2260	0.013	12.45	11.54	91%
G-7	Pajarito Plateau	19	0.2497	0.5429	0.0380	0.1050	0.2140	0.013	19.21	8.08	88%
G-8	Pajarito Plateau	18	0.0627	0.0726	0.0130	0.0315	0.0630	0.013	4.82	2.42	59%
G-9	Pajarito Plateau	18	0.0601	0.1137	0.0130	0.0260	0.0400	0.013	4.62	2.00	50%

Table A1. cont.

Station Name	Location	N	Means	Std.Dev.	Q25	Median	Q75	Background	Ratio: Mean /Background	Ratio: Median /Background	Estimated % LANL Pu Activity)[a]
Guaje at SR-502	Pajarito Plateau	26	0.0098	0.0364	0.0018	0.0020	0.0030	0.013	0.75	0.15	0%
Guaje near well G-4	Pajarito Plateau	5	0.0192	0.0435	-0.0010	0.0000	0.0020	0.013	1.48	0.00	0%
Hamilton Bend Spring	Pajarito Plateau	25	0.4756	0.3447	0.2100	0.4280	0.5660	0.013	36.59	32.92	97%
Indio at SR-4	Pajarito Plateau	7	0.0040	0.0019	0.0025	0.0040	0.0050	0.013	0.31	0.31	0%
Los Alamos at Bridge	Pajarito Plateau	24	0.0039	0.0072	0.0015	0.0026	0.0035	0.013	0.30	0.20	0%
Los Alamos at LA-2	Pajarito Plateau	28	0.2111	0.2754	0.0615	0.1110	0.2490	0.013	16.24	8.54	88%
Los Alamos at LAO-1	Pajarito Plateau	27	0.6129	0.8374	0.1384	0.3600	0.9100	0.013	47.15	27.69	96%
Los Alamos at LAO-3	Pajarito Plateau	23	0.2354	0.1623	0.1240	0.2140	0.3185	0.013	18.11	16.46	94%
Los Alamos at LAO-4	Pajarito Plateau	9	0.3771	0.1538	0.3050	0.3540	0.3950	0.013	29.01	27.23	96%
Los Alamos at LAO-4.5	Pajarito Plateau	23	0.3286	0.3527	0.1220	0.1680	0.4370	0.013	25.28	12.92	92%
Los Alamos at Otowi	Pajarito Plateau	24	0.1412	0.1202	0.0553	0.1310	0.1920	0.013	10.86	10.08	90%
Los Alamos at Rio Grande	Pajarito Plateau	7	0.0754	0.1121	0.0030	0.0540	0.0760	0.013	5.80	4.15	76%
Los Alamos at SR-4	Pajarito Plateau	33	0.1931	0.1703	0.0760	0.1340	0.2720	0.013	14.86	10.31	90%
Los Alamos at Totavi	Pajarito Plateau	32	0.1677	0.2042	0.0180	0.1045	0.2350	0.013	12.90	8.04	88%
Los Alamos at TW-3	Pajarito Plateau	9	0.4550	0.3045	0.2160	0.4880	0.5800	0.013	35.00	37.54	97%
Los Alamos at Upper GS	Pajarito Plateau	25	0.3480	0.2715	0.1940	0.2690	0.5070	0.013	26.77	20.69	95%
Mortand A-6	Pajarito Plateau	13	0.0299	0.0249	0.0150	0.0230	0.0360	0.013	2.30	1.77	43%
Mortandad A-10	Pajarito Plateau	9	0.0021	0.0008	0.0020	0.0020	0.0030	0.013	0.16	0.15	0%
Mortandad A-7	Pajarito Plateau	14	0.0208	0.0265	0.0090	0.0115	0.0190	0.013	1.60	0.88	0%
Mortandad A-8	Pajarito Plateau	9	0.0081	0.0068	0.0040	0.0040	0.0120	0.013	0.62	0.31	0%
Mortandad at GS-1	Pajarito Plateau	28	38.5249	79.1927	3.4060	7.6000	25.1500	0.013	2963.45	584.62	100%
Mortandad at MCO-13 (A-5)	Pajarito Plateau	30	0.0280	0.0211	0.0190	0.0235	0.0310	0.013	2.15	1.81	45%
Mortandad at MCO-5	Pajarito Plateau	32	16.5609	20.6862	5.6350	7.9460	19.2000	0.013	1273.91	611.23	100%
Mortandad at MCO-7	Pajarito Plateau	28	4.0164	4.5430	0.7200	2.0515	5.7600	0.013	308.96	157.81	99%
Mortandad at MCO-9	Pajarito Plateau	25	0.3539	1.6327	0.0130	0.0210	0.0300	0.013	27.22	1.62	38%
Mortandad at Rio Grande (A-11)	Pajarito Plateau	20	0.0019	0.0017	0.0010	0.0010	0.0020	0.013	0.15	0.08	0%
Mortandad at SR-4 (A-9)	Pajarito Plateau	30	0.0033	0.0021	0.0020	0.0030	0.0040	0.013	0.25	0.23	0%
Mortandad near CMR Building	Pajarito Plateau	26	0.0717	0.1336	0.0060	0.0110	0.0630	0.013	5.52	0.85	0%
Mortandad west of GS-1	Pajarito Plateau	22	11.2060	39.3281	0.0070	0.0265	0.0800	0.013	862.00	2.04	51%
Pajarito at Rio Grande	Pajarito Plateau	15	0.0039	0.0042	0.0020	0.0030	0.0030	0.013	0.30	0.23	0%
Pajarito at SR-4	Pajarito Plateau	24	0.0140	0.0198	0.0020	0.0090	0.0154	0.013	1.07	0.69	0%
Pajarito at SR-501	Pajarito Plateau	4	0.0043	0.0013	0.0035	0.0040	0.0050	0.013	0.33	0.31	0%
Pajarito Canyon	Pajarito Plateau	4	0.0300	0.0516	0.0005	0.0065	0.0595	0.013	2.31	0.50	0%
Potrillo at SR-4	Pajarito Plateau	22	0.0058	0.0071	0.0020	0.0040	0.0070	0.013	0.44	0.31	0%
Potrillo at TA-36	Pajarito Plateau	8	0.0281	0.0715	0.0015	0.0035	0.0055	0.013	2.16	0.27	0%

Table A1. cont.

Station Name	Location	N	Means	Std.Dev.	Q25	Median	Q75	Background	Ratio: Mean /Background	Ratio: Median /Background	Estimated % LANL Pu Activity)[a]
Pueblo 1	Pajarito Plateau	26	0.8087	1.8341	0.0050	0.0090	0.2700	0.013	62.21	0.69	0%
Pueblo 2	Pajarito Plateau	24	2.3346	3.2457	0.5325	0.9936	3.2435	0.013	179.58	76.43	99%
Pueblo 3	Pajarito Plateau	26	1.1364	3.2068	0.0050	0.1903	0.5060	0.013	87.42	14.64	93%
Pueblo at PC-1	Pajarito Plateau	6	0.6337	0.9387	0.0230	0.0435	1.8100	0.013	48.74	3.35	70%
Pueblo at SR-502	Pajarito Plateau	32	0.5350	0.3112	0.3760	0.4900	0.7345	0.013	41.15	37.69	97%
Pueblo at TW-2	Pajarito Plateau	6	1.1897	0.7349	0.4540	1.2450	1.7500	0.013	91.51	95.77	99%
Sandia at Rio Grande	Pajarito Plateau	14	0.0039	0.0085	0.0010	0.0015	0.0030	0.013	0.30	0.12	0%
Sandia at SR-4	Pajarito Plateau	23	0.0051	0.0141	0.0010	0.0020	0.0040	0.013	0.39	0.15	0%
Two Mile	Pajarito Plateau	4	0.0053	0.0024	0.0035	0.0060	0.0070	0.013	0.40	0.46	0%
Water at Rio Grande	Pajarito Plateau	15	0.0049	0.0074	0.0010	0.0020	0.0080	0.013	0.37	0.15	0%
Water at SR-4	Pajarito Plateau	27	0.0071	0.0102	0.0020	0.0040	0.0080	0.013	0.54	0.31	0%
Water at SR-501	Pajarito Plateau	5	0.0031	0.0027	0.0010	0.0040	0.0050	0.013	0.24	0.31	0%
Water Canyon at Beta	Pajarito Plateau	8	0.0166	0.0247	0.0025	0.0030	0.0285	0.013	1.28	0.23	0%
Abiquiu Lower	RC Reservoir	15	0.0065	0.0038	0.0033	0.0076	0.0095	0.022	0.29	0.35	0%
Abiquiu Middle	RC Reservoir	16	0.0061	0.0035	0.0038	0.0055	0.0096	0.022	0.28	0.25	0%
Abiquiu Upper	RC Reservoir	16	0.0044	0.0042	0.0011	0.0036	0.0072	0.022	0.20	0.16	0%
Cochiti Lower	RG Reservoir	15	0.0117	0.0079	0.0020	0.0123	0.0179	0.022	0.53	0.56	0%
Cochiti Middle	RG Reservoir	17	0.0249	0.0292	0.0130	0.0167	0.0238	0.022	1.13	0.76	0%
Cochiti Upper	RG Reservoir	17	0.0158	0.0192	0.0072	0.0124	0.0165	0.022	0.72	0.56	0%
Heron Lower	RC Reservoir	10	0.0092	0.0049	0.0049	0.0079	0.0120	0.022	0.42	0.36	0%
Heron Middle	RC Reservoir	10	0.0065	0.0036	0.0038	0.0055	0.0090	0.022	0.30	0.25	0%
Heron Upper	RC Reservoir	9	0.0095	0.0052	0.0055	0.0077	0.0120	0.022	0.43	0.35	0%
Rio Grande Lower	RG Reservoir	6	0.0188	0.0124	0.0164	0.0182	0.0209	0.022	0.85	0.83	0%
Rio Grande Middle	RG Reservoir	4	0.0168	0.0062	0.0119	0.0170	0.0217	0.022	0.76	0.77	0%
Rio Grande Upper	RG Reservoir	4	0.0153	0.0071	0.0107	0.0180	0.0199	0.022	0.70	0.82	0%
Jemez River	RG River	27	0.0040	0.0033	0.0020	0.0030	0.0060	0.013	0.31	0.23	0%
Rio Chama at Chamita	RC River	27	0.0024	0.0033	0.0000	0.0020	0.0030	0.013	0.18	0.15	0%
Rio Grande at Ancho	RG River	16	0.0059	0.0077	0.0015	0.0040	0.0070	0.013	0.45	0.31	0%
Rio Grande at Bernalillo	RG River	26	0.0043	0.0034	0.0020	0.0040	0.0050	0.013	0.33	0.31	0%
Rio Grande at Chaquehui	RG River	4	0.0062	0.0044	0.0025	0.0060	0.0100	0.013	0.48	0.46	0%
Rio Grande at Cochiti	RG River	14	0.0272	0.0730	0.0020	0.0063	0.0240	0.013	2.09	0.48	0%
Rio Grande at Embudo	RG River	28	0.0025	0.0023	0.0020	0.0020	0.0040	0.013	0.19	0.15	0%
Rio Grande at Frijoles (bank)	RG River	21	0.0074	0.0198	0.0020	0.0030	0.0048	0.013	0.57	0.23	0%
Rio Grande at Frijoles (wdth intgrt)	RG River	4	0.0051	0.0050	0.0020	0.0030	0.0082	0.013	0.39	0.23	0%

Table A1. cont.

Station Name	Location	N	Means	Std.Dev.	Q25	Median	Q75	Background	Ratio: Mean /Background	Ratio: Median /Background	Estimated % LANL Pu Activity)[a]
Rio Grande at Otowi (bank)	RG River	34	0.0062	0.0128	0.0010	0.0030	0.0040	0.013	0.47	0.23	0%
Rio Grande at Pajarito	RG River	17	0.0156	0.0502	0.0010	0.0030	0.0060	0.013	1.20	0.23	0%
Rio Grande at Sandia	RG River	14	0.0032	0.0028	0.0020	0.0030	0.0040	0.013	0.25	0.23	0%
Rio Grande at Water Canyon	RG River	4	0.0057	0.0038	0.0025	0.0060	0.0090	0.013	0.44	0.46	0%
El Vado Lower	RC Reservoir	6	0.0069	0.0039	0.0060	0.0070	0.0083	0.022	0.31	0.32	0%
El Vado Middle	RC Reservoir	6	0.0055	0.0020	0.0050	0.0058	0.0068	0.022	0.25	0.26	0%
El Vado Upper	RC Reservoir	8	0.0065	0.0020	0.0052	0.0063	0.0071	0.022	0.29	0.28	0%

[a] This calculation uses a simple additive mixing model to estimate the Laboratory contribution. An increase in the ^{239,240}Pu activity above the background value is assumed to be solely due to the addition of Laboratory ^{239,240}Pu. For example, a sample with a median ^{239,240}Pu activity ratio 5 times that of the background value is assumed to contain a mixture of 4 parts of Laboratory plutonium and 1 part of global fallout plutonium. The algorithm is as follows:

[(PuComponent)L]/[(PuComponent)F] = [(PuActivity)S - (PuActivity)F]/[(PuActivity)F],

where

(PuComponent)L = plutonium activity in LANL component,

(PuComponent)F = plutonium activity in global fallout component,

(PuActivity)S = the median ^{239,240}Pu activity measured in soil samples at a location, and

(PuActivity)F = 0.013, the statistical upper limit for ^{239,240}Pu activity from global fallout in northern New Mexico river sediments (S.G. McLin, LANL Water Quality and Hydrology Group, personal communication).

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