

LA--11794-MS

DE90 014818

*Plutonium Deposition and Distribution
from Worldwide Fallout in Northern
New Mexico and Southern Colorado*

*William D. Purtymun
Richard J. Peters
Max N. Maes*



DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

PLUTONIUM DEPOSITION AND DISTRIBUTION FROM WORLDWIDE FALLOUT IN NORTHERN NEW MEXICO AND SOUTHERN COLORADO

by

William D. Purtymun, Richard J. Peters, and Max N. Maes

ABSTRACT

Samples of soil, river, and reservoir sediment were collected and analyzed for two isotopes of plutonium from 1979 through 1987. The plutonium is principally a manmade element, but it has been distributed throughout the world as a result of fallout from nuclear weapons tests and from reentry of satellites that contained a nuclear power source. Fallout is deposited on soil and is subject to erosion and transport as sediment in rivers and deposition in reservoirs. The soil samples collected from northern New Mexico and southern Colorado indicated slight, but significant, differences in the concentrations and ratios of $^{239,240}\text{Pu}$ to ^{238}Pu in soil, river, and reservoir sediments. The differences in plutonium concentrations and ratios found in the environment can be attributed to regional and local weather patterns and to distribution by physical transport into rivers and reservoirs. The variability in plutonium particle size also contributes to the inconstancy of plutonium concentrations and ratios found in soils and sediments.

I. INTRODUCTION

Nuclear weapons tests in the atmosphere and the reentry and burnup of satellites containing plutonium power sources have resulted in the worldwide fallout of plutonium. The plutonium deposited on land surface is adsorbed onto the silt and clay-size fraction of the soil (Nyhan 1976A and 1976B, Tamura 1975, Little 1977). The soil and plutonium are subject to transport and distribution through erosion processes. Thus, plutonium in the natural environment can be found in soil, river, and reservoir sediments in measurable, but very small, concentrations.

The fallout in soils and sediments measured in this study consisted of the isotopes ^{238}Pu and $^{239,240}\text{Pu}$. The amount of $^{239,240}\text{Pu}$ released from weapons tests has been estimated at 320 kCi; about 45 kCi of ^{238}Pu is estimated to be from weapons tests and power sources (Perkins 1980, Harley 1971, Hardy 1973). If the

plutonium were deposited uniformly over the earth, the ratio of $^{239,240}\text{Pu}$ to ^{238}Pu would be about 7.

Of the $^{239,240}\text{Pu}$ that is released, almost 100% is from weapons tests; of the ^{238}Pu released, 60% is from weapons tests and the remaining 40% is from burnup of satellite power sources upon reentry into the atmosphere (Perkins 1980). The fallout is not deposited uniformly over the land surface because weapons tests are located in the northern hemisphere whereas satellite reentry occurs in the southern hemisphere. The distribution of plutonium is further complicated by the variable and inconsistent weather patterns in both hemispheres. Local differences in weather patterns, such as those in northern New Mexico, also result in differences in plutonium concentrations and ratios.

Another variable in the fallout of plutonium concentrations is the particle size of the plutonium. The activity (concentration) of the plutonium particle is

proportional to the third power of its diameter; a ten-fold increase or decrease in the diameter will give a thousandfold increase or decrease in activity (Sills 1971). A single large particle of plutonium could contribute as much activity as a thousand smaller ones with one-tenth the diameter.

Materials from the detonation of nuclear weapons are completely vaporized. As the materials recondense, the particles contain only a very small fraction of plutonium rather than separate, discrete particles of pure plutonium oxide. As a consequence, little, if any, homogeneity can be expected of plutonium concentrations deposited in soils from global fallout (Sills 1971).

The purpose of this report is to examine the variations in plutonium concentrations and ratios resulting from fallout in soils and in river and reservoir sediments in northern New Mexico and southern Colorado. These data can be used to determine if plutonium in the environment is from worldwide fallout from weapons tests and satellite power sources or from plutonium released from nuclear laboratories or weapons facilities.

Regional soils and river sediments are collected near the Los Alamos National Laboratory and are analyzed for radionuclides as part of an ongoing environmental-surveillance program. The analytical results from these samples are used as background (fallout concentrations) to evaluate effects of the Laboratory's operation on the local and regional environment (Purtymun 1987).

The background for plutonium is determined by using 10-g samples with limits of detection of about 2 fCi/g (10^{-15} Ci). Although the methods of analyses used to determine the fallout concentrations of plutonium for the environmental-surveillance program are satisfactory, the authors, in this study, increased the sensitivity of the analysis by using a larger sample. The size of the sample was increased from 10 to 1000 g.

Soil samples and river and reservoir sediment samples (1000 g) were collected from 1979 through 1987. Aliquots of the samples were spiked with a ^{242}Pu tracer. Each sample was then dissolved in acid. The plutonium in the acid was isolated by anion exchange and electroplated onto a stainless steel disk, which was then counted on an alpha spectrometer. The recovery

was determined from the plutonium tracer, and the plutonium concentrations were calculated (ESG 1988, Gautier 1987). It should be noted that, in this report, the numbers in parentheses represent either analytical uncertainties, if used with a single sample, or a standard deviation of a number, if used as a mean.

II. SOIL

Soil samples were collected at five stations within a 15-m (50-mi) radius at Los Alamos in 1981 and 1983 and at nine stations in northern New Mexico and southern Colorado in 1986 (Fig. 1).

At each of these stations a square area 9 m (30 ft) on a side was designated as a sampling area. One sample was collected from each of five locations in the sampling area (the center and four corners), using a soil ring 7.5 cm in diameter driven 5.0 cm into the soil. The five samples were then combined to form one composite sample from each sampling area for analysis.

A slightly modified sampling plan was used at the station at Santa Cruz Lake in 1984. Five samples were collected at each of the five locations in the sampling area at this station. The samples from each location were then combined for a composite sample, producing five composite samples from the sampling area. These five composite samples from each of the five locations were analyzed to determine if there was any variation in the concentration of plutonium within the sampling area (Table I). For ^{239}Pu , the concentrations ranged from 0.6 to 2.2 fCi/g; for $^{239,240}\text{Pu}$, from 8.8 to 26.4 fCi/g (Fig. 2). The ratio of $^{239,240}\text{Pu}$ to ^{238}Pu ranged from 5 to 17, indicating that the ratios of total plutonium also vary by a factor of about 3.

For the samples from the five soil stations located within a 15-m (50-mi) radius of Los Alamos that were collected and analyzed for plutonium in 1981 and 1983 (Fig. 1), the total plutonium ranged from 2.8 to 17.8 fCi/g in 1981 and from 2.7 to 15.6 fCi/g in 1983 (Table II). This compares favorably with the plutonium concentrations in the midwest, where the surface layer of soil had a concentration of 15 to 20 fCi/g (Muller 1977, 1978). The soil background for plutonium from soils in northern New Mexico (1974-1986) was similar, with an upper limit of 30 fCi/g (Purtymun 1987).

The concentrations of plutonium in soils sampled in 1981 and 1983 varied from station to station, and the

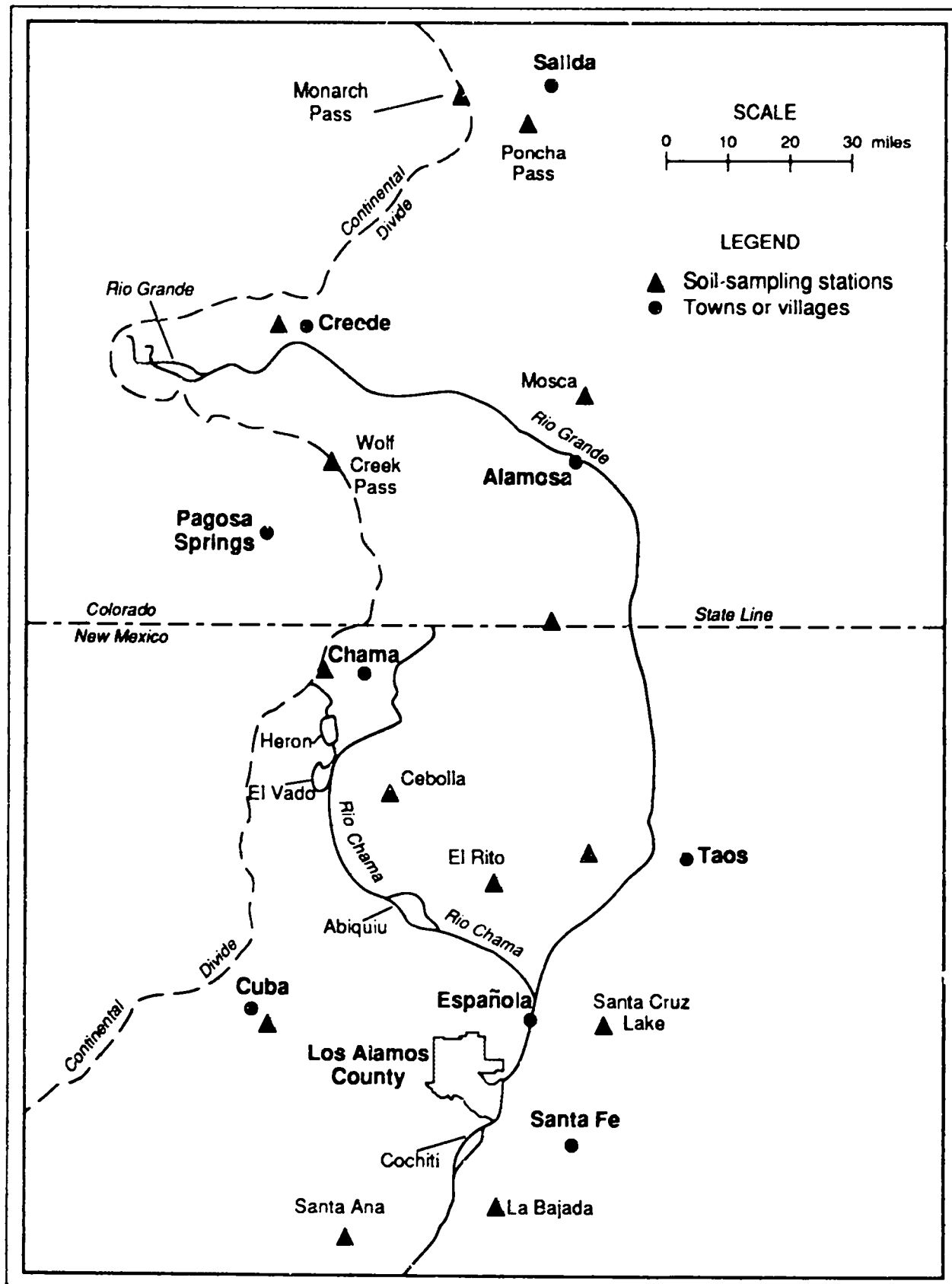


Fig. 1. Locations of soil-sampling stations and reservoirs on the Rio Chama and the Rio Grande.

Table I. Comparison of Plutonium from Five Locations at Soil Station* Near Santa Cruz Lake.

Station	^{238}Pu (fCi/g)	$^{239,240}\text{Pu}$ (fCi/g)	Total Plutonium (fCi/g)	Ratio $^{239,240}\text{Pu}/^{238}\text{Pu}$ (fCi/g)
Location				
Center	2.2 (0.0)	26.4 (0.0)	28.6	12
NW corner	1.0 (0.0)	9.4 (0.0)	10.4	9
SW corner	0.6 (0.0)	10.1 (0.0)	10.7	17
SE corner	0.6 (0.0)	8.8 (0.0)	9.4	15
NE corner	2.2 (0.0)	10.6 (0.0)	12.8	5

*The soil station covered an area 9 m (30 ft) on a side.

Note: The numbers in parentheses represent analytical uncertainties.

correlations by year indicated no particular pattern. The ratio of $^{239,240}\text{Pu}$ to ^{238}Pu ranged from 17 to 31 in 1981 and from 9 to 26 in 1983.

Soil samples from nine stations located in northern New Mexico and southern Colorado were collected and

analyzed for plutonium in 1986 (Fig. 1). The total plutonium concentrations varied from 1.3 to 84.9 fCi/g (Table II). The concentrations of total plutonium from the nine stations can be divided into three classes according to location of the sampling stations:

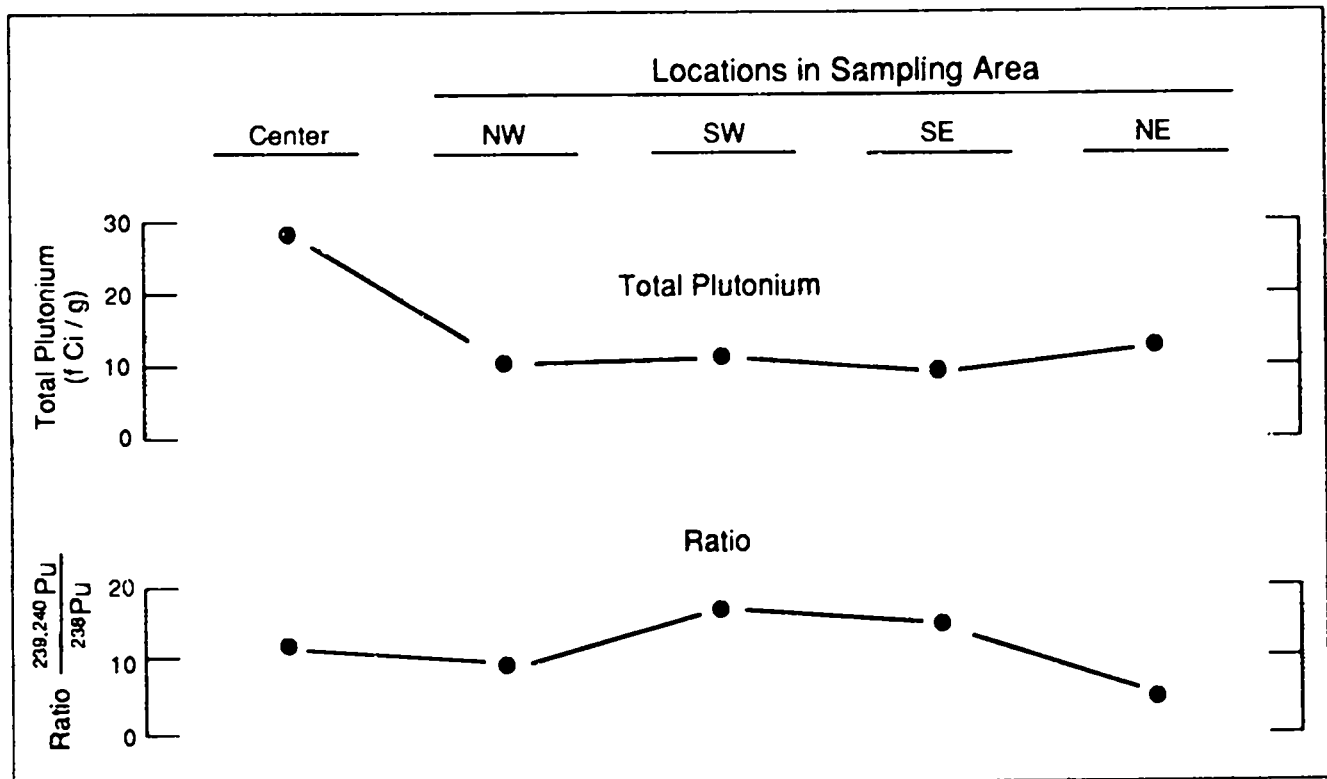


Fig. 2. Total plutonium and the ratio of $^{239,240}\text{Pu}/^{238}\text{Pu}$ from soil-sampling station near Santa Cruz Lake.

Table II. Plutonium in Regional Soils.

Station	^{238}Pu (fCi/g)	$^{239,240}\text{Pu}$ (fCi/g)	Total Plutonium (fCi/g)	Ratio $^{239,240}\text{Pu}/^{238}\text{Pu}$ (fCi/g)
1981				
La Bajada	0.6 (0.2)	10.0 (0.1)	10.6	17
Cuba	0.8 (0.1)	17.0 (0.4)	17.8	21
El Rito	0.3 (0.0)	9.4 (0.3)	9.7	31
Santa Cruz Lake	0.4 (0.1)	10.3 (0.7)	10.7	26
Gallina	0.2 (0.0)	5.4 (0.2)	5.6	27
Santa Ana Pueblo	0.1 (0.0)	2.7 (0.1)	2.8	27
1983				
La Bajada	0.7 (0.1)	13.4 (0.1)	14.1	19
Cuba	0.1 (0.0)	2.6 (0.1)	2.7	26
El Rito	0.2 (0.0)	3.2 (0.1)	3.4	16
Santa Cruz Lake	1.5 (0.8)	14.1 (8.2)	15.6	9
Santa Ana Pueblo	0.3 (0.1)	3.7 (0.2)	4.0	12
Gallina	0.6 (0.1)	13.2 (0.5)	13.8	22
1986				
South Tres Piedras, New Mexico	0.3 (0.1)	7.6 (0.3)	7.9	25
New Mexico–Colorado State Line	0.3 (6.1)	6.2 (0.3)	6.5	21
Mosces, Colorado	0.1 (0.1)	1.2 (0.1)	1.3	12
Poncha Pass, Colorado	0.7 (0.1)	12.2 (0.4)	12.9	17
Monarch Pass, Colorado	3.9 (0.4)	81.0 (8.0)	84.9	21
Creede, Colorado	0.6 (0.1)	15.6 (0.6)	16.2	26
Wolf Creek Pass, Colorado	1.9 (1.0)	43.6 (1.9)	45.5	23
Chama, New Mexico	0.8 (0.2)	19.3 (1.1)	20.1	24
Cebolla, New Mexico	0.4 (0.0)	10.7 (0.4)	11.1	27
$\bar{X}(S)$	0.7 (0.9)	14.4 (17.7)	18.6 (15.1)	21 (6)
Minimum	0.1 (0.0)	1.2 (0.1)	1.3	9
Maximum	3.9 (0.4)	81.0 (8.0)	84.9	31

Note: The numbers in parentheses represent (a) analytical uncertainties if used with a single sample, or (b) a standard deviation of a number if used as a mean.

- i. The low concentrations of plutonium (1.3 to 7.9 fCi/g [south of Tres Piedras, the New Mexico–Colorado state line, and Mosca], with ratios from 12 to 25) are found in the high inter-basin of the San Luis Valley.
2. The intermediate concentrations of plutonium (11.1 to 16.2 fCi/g [Poncha Pass, Creede, and Cebolla], with ratios from 17 to 27) are found on the high mountain slopes west of the continental divide.
3. The high concentrations of plutonium (20.1 to 84.9 fCi/g [Chama, Wolf Creek, and Monarch passes], with ratios of 21 to 24) are located along the high mountain passes along the continental divide.

The concentrations tend to increase northward along the continental divide from Chama to Monarch Pass. Fallout concentrations tend to increase northward from southern New Mexico into northern Colorado because of the prevailing wind pattern (jet stream) eastward from the test areas in the Pacific and in Nevada. The major deposition of fallout was in the corridor in northern Colorado, with a decreasing amount to the south into New Mexico. Local weather patterns, such as upslope winds along the high mountains, tend to deposit fallout on the leeward side. The ratios $^{239,240}\text{Pu}$ to ^{238}Pu in this area are from 27 to 29 (Perkins 1980).

The ratio of $^{239,240}\text{Pu}$ to ^{238}Pu for the soil samples collected in 1981, 1983, and 1986 range from 9 to 31, with a mean of 21(6). The concentrations of the two isotopes of plutonium and their ratios vary considerably (Fig. 3).

III. RIVER SEDIMENTS

River sediments on the Rio Grande were collected at two stations above Otowi and four stations below Otowi in 1979 and 1981 (Fig. 4). The samples were collected from dune build-up in the stream or from sediments deposited along the river channel. The concentrations of ^{238}Pu concentrations ranged from 0.1 to 0.5 fCi/g; the $^{239,240}\text{Pu}$ concentrations ranged from 1.0 to 8.8 fCi/g (Table III). The total plutonium ranged from 1.1 to 9.5 fCi/g, with a mean of 5(3) fCi/g. The background for river sediments for the period 1974-1985 had an upper limit of 28 fCi/g (Purtymun 1987).

The ratio of $^{239,240}\text{Pu}$ to ^{238}Pu of the 1979 and 1981 river sediments ranged from 10 to 71, with a mean of 32(19). The wide range in ratios indicates a variation in

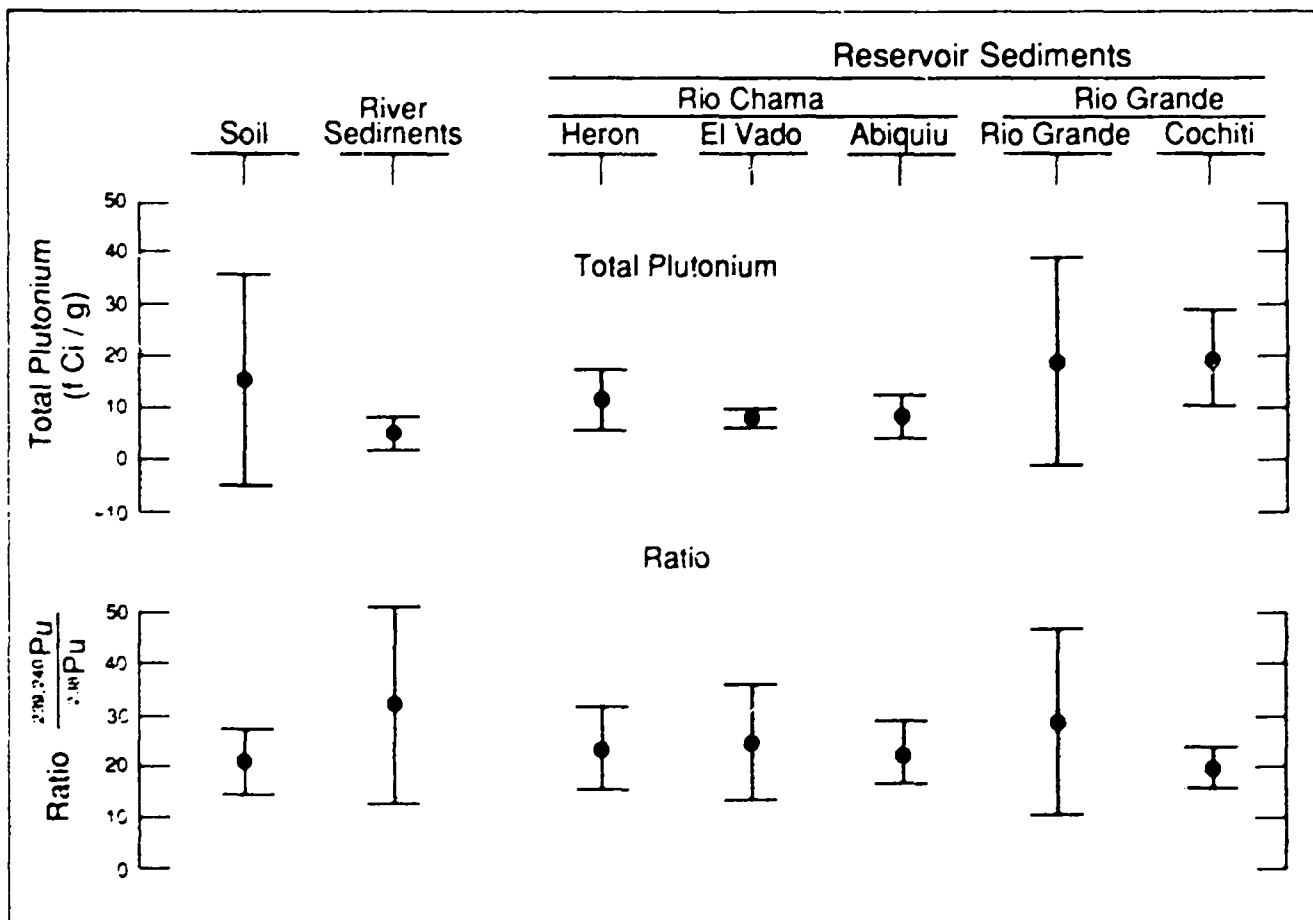


Fig. 3. Mean total plutonium and ratios of $^{239,240}\text{Pu}/^{238}\text{Pu}$, with standard deviation of soil, river, and reservoir sediments (standard deviation is shown as the bar)

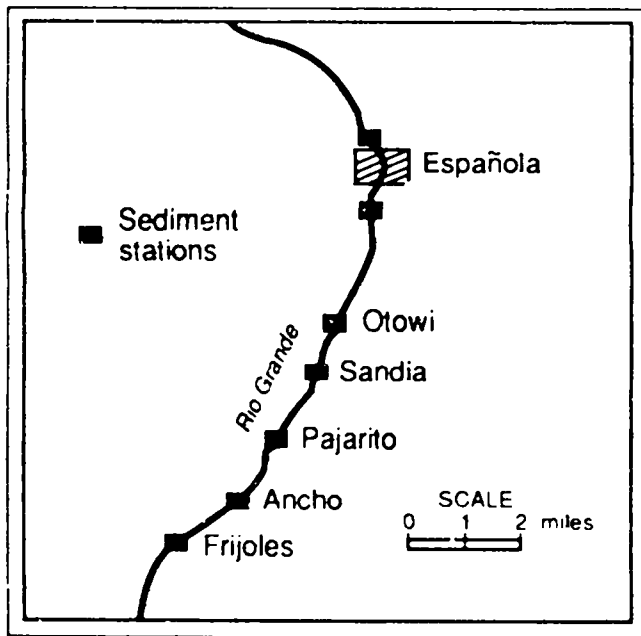


Fig. 4. River sediment stations on the Rio Grande

the concentrations of plutonium. The difference of the ratios is unknown, but it may be due to the smaller particle size of the ^{238}Pu particles, or it may be that the

silts and clay-size particles that adsorb ^{238}Pu more readily are winnowed out of the river sediments and deposited in the reservoirs.

The concentrations of plutonium in the river sediments are lower than those found in soil. The low concentrations are due to the fact that the river sediments contained coarser material than that from the soils and that the larger concentrations of plutonium are found in the silt and clay-size material.

Table III. Plutonium in Sediment in the Río Grande.

Station	^{238}Pu (fCi/g)	$^{239,240}\text{Pu}$ (fCi/g)	Total Plutonium (fCi/g)	Ratio $^{239,240}\text{Pu}/^{238}\text{Pu}$ (fCi/g)
1979				
Otowi	0.2 (0.0)	7.3 (0.2)	7.5	36
Sandia	0.1 (0.0)	4.3 (0.2)	4.4	43
Pajarito	0.1 (0.1)	1.0 (0.2)	1.1	10
Ancho	0.5 (0.0)	8.8 (0.3)	9.3	18
Frijoles	0.1 (0.0)	2.3 (0.2)	2.3	23
1981				
Española (above)	0.1 (0.4)	2.3 (0.2)	2.4	23
Española	0.1 (0.0)	7.1 (0.3)	7.2	71
Otowi	0.1 (0.6)	2.8 (0.4)	2.9	28
$\bar{X}(S)$	0.2 (0.1)	4.5 (2.9)	5 (3)	32 (19)
Minimum	0.1 (0.0)	1.0 (0.2)	1.1	10
Maximum	0.5 (0.0)	8.8 (0.3)	9.3	71

Note: The numbers in parentheses represent (a) analytical uncertainties if used with a single sample, or (b) a standard deviation of a number if used as a mean.

IV. RESERVOIR SEDIMENTS ON THE RIO CHAMA

Sediments were collected from three reservoirs on the Rio Chama (Fig. 1). The samples were collected from a boat, using an Eckman dredge that collects a sample of sediments from an area about 15 cm (6 in.) on a side and to a depth of about 5 cm (2 in.).

Heron Reservoir was completed in the Chama drainage in 1970. The drainage area is about 500 m²

(193 mi²) along the eastern edge of the continental divide. The reservoir is used for storage of trans-mountain water from the San Juan River basin. Three sets of samples were collected—in 1982, 1984, and 1985 (Table IV). Analysis of the results showed no particular pattern of the concentrations from one year to the next. The total plutonium ranged from 3.7 to 20.8 fCi/g, with a mean of 12(6) fCi/g (Table IV). The mean ratio of ^{239,240}Pu to ²³⁸Pu was 24(8), slightly above the mean ratio for soils of 21(6) (Fig. 3).

Table IV. Plutonium in Reservoir Sediments on the Rio Chama.

Reservoirs	²³⁸ Pu (fCi/g)	^{239,240} Pu (fCi/g)	Total Plutonium (fCi/g)	Ratio ^{239,240} Pu/ ²³⁸ Pu (fCi/g)
Heron Reservoir				
1982				
Upper	0.2 (0.1)	7.7 (0.0)	7.9	38
Middle	0.7 (0.1)	13.5 (0.0)	14.2	19
Dam	0.9 (0.0)	19.9 (0.1)	20.8	22
1984				
Upper	0.3 (0.1)	3.4 (0.2)	3.7	11
Middle	0.3 (0.1)	6.5 (0.4)	6.8	22
Dam	0.8 (0.1)	18.1 (0.9)	18.9	23
1985				
Upper	0.7 (0.2)	17.4 (1.2)	18.1	25
Middle	0.5 (0.2)	11.4 (1.6)	11.9	23
Dam	0.2 (0.2)	7.4 (0.4)	7.6	37
$\bar{X}(S)$	0.5 (0.3)	11.7 (5.9)	12 (6)	24 (8)
Minimum	0.2 (0.2)	3.4 (0.2)	3.7	11
Maximum	0.9 (0.0)	19.9 (0.1)	20.8	38
El Vado Reservoir				
1982				
Upper	0.1 (0.0)	10.6 (0.0)	10.7	—
Middle	0.1 (0.0)	5.2 (0.1)	5.3	52
Dam	0.6 (0.0)	12.6 (0.1)	13.2	21
1984				
Upper	0.4 (0.1)	7.7 (0.0)	8.1	19
Middle	0.3 (0.1)	6.8 (0.8)	7.1	23
Dam	0.4 (0.1)	6.8 (0.4)	7.2	17

Table IV (Continued)

Reservoirs	^{238}Pu (fCi/g)	$^{239,240}\text{Pu}$ (fCi/g)	Total Plutonium (fCi/g)	Ratio $^{239,240}\text{Pu}/^{238}\text{Pu}$ (fCi/g)
<i>El Vado Reservoir (Continued)</i>				
1985				
Upper	0.4 (0.2)	7.4 (1.2)	7.8	18
Middle	0.3 (0.2)	7.7 (0.6)	8.0	26
Dam	0.3 (0.2)	7.8 (0.5)	8.1	26
$\bar{X}(S)$	0.3 (0.2)	8.1 (2.2)	8 (2)	25 (11)
Minimum	0.1 (0.0)	5.2 (0.1)	5.3	17
Maximum	0.6 (0.0)	12.6 (0.1)	13.2	52
<i>Abiquiu Reservoir</i>				
1982				
Upper	0.4 (0.3)	8.0 (0.6)	8.4	20
Middle	—	—	—	—
Dam	0.6 (0.1)	11.4 (0.6)	12.0	19
1984				
Upper	0.7 (0.3)	16.3 (1.2)	17.0	23
Middle	0.5 (0.2)	11.0 (1.2)	11.5	22
Dam	0.9 (0.2)	10.7 (0.8)	11.6	12
1985				
Upper	0.3 (0.2)	7.8 (0.8)	8.1	26
Middle	1.2 (0.2)	9.4 (0.8)	10.6	—
Dam	0.5 (0.2)	9.1 (0.8)	9.6	18
1986				
Upper	0.2 (0.1)	6.7 (0.3)	6.9	34
Middle	0.3 (0.1)	6.3 (0.2)	6.6	21
Dam	0.4 (0.1)	9.5 (0.3)	9.9	24
1987				
Upper	0.1 (0.0)	0.2 (0.1)	0.3	—
Middle	0.2 (0.0)	5.0 (0.3)	5.2	25
Dam	0.2 (0.0)	6.0 (0.3)	6.2	30
$\bar{X}(S)$	0.5 (0.3)	8.4 (3.7)	9 (4)	23 (6)
Minimum	0.1 (0.0)	0.2 (0.1)	0.3	12
Maximum	1.2 (0.2)	16.3 (1.2)	17.0	34

Note: The numbers in parentheses represent (a) analytical uncertainties if used with a single sample, or (b) a standard deviation of a number if used as a mean.

El Vado Reservoir, which was completed in 1935, is located downstream from Hecion. The drainage area is about 2260 m² (873 mi²). Three sets of samples were collected—in 1982, 1984, and 1985 (Table IV). Again, there was no particular trend to the concentration of plutonium, either in a single set or from year to year. The total plutonium concentrations ranged from 5.3 to 13.2 fCi/g, with a mean of 8(2). The mean ^{239,240}Pu to ²³⁸Pu ratio was 25(11), slightly above the mean ratio of soil of 21(6) (Fig. 3).

Abiquiu Reservoir was completed in 1961 and is located downstream from El Vado. The drainage area above the reservoir is about 4140 m² (1600 mi²). Five sets of samples were collected—in 1982, 1984, 1985, 1986, and 1987. As before, no pattern was found for plutonium concentrations in a single year or from one set to another (Table IV). The total plutonium concentrations ranged from 0.3 to 17 fCi/g, with a mean of 9(4). The mean ^{239,240}Pu to ²³⁸Pu ratio was 23(6), within the range of the mean soil ratio of 21(6) (Fig. 3).

V. RESERVOIR SEDIMENTS ON THE RIO GRANDE

Sediments were collected from two reservoirs on the Rio Grande (Fig. 1). The sediments from Rio Grande Reservoir were collected just below the water level because the reservoir had been drained for repair. The reservoir is located below the continental divide in southern Colorado. The sediments from Cochiti Reservoir were collected from a boat, using an Eckman dredge in water depths ranging from 4 to 15 m (12 to 50 ft).

Rio Grande Reservoir was completed in 1912 and is one of the oldest reservoirs on the Rio Grande. The drainage area, west of the continental divide and above the reservoir, is about 420 m² (163 mi²).

The reservoir sediments from Rio Grande Reservoir were collected from the upper end and middle of the reservoir and from an area near the dam. The total plutonium concentrations ranged from 5.0 to 41 fCi/g (Table V). The low concentration of 5.0 fCi/g was

Table V. Plutonium in Reservoir Sediments on the Rio Grande.

Reservoirs	²³⁸ Pu (fCi/g)	^{239,240} Pu (fCi/g)	Total Plutonium (fCi/g)	Ratio ^{239,240} Pu/ ²³⁸ Pu (fCi/g)
Rio Grande Reservoir^a				
1986				
Upper	0.1 (0.1)	4.9 (0.4)	5.0	49
Middle	0.5 (0.1)	9.5 (0.3)	10.0	19
Dam	2.2 (0.2)	38.8 (2.7)	41.0	18
$\bar{X}(S)$	0.9 (1.1)	17.7 (18.4)	19 (20)	29 (18)
Minimum	0.1 (0.1)	4.9 (0.4)	5.0	18
Maximum	2.2 (0.2)	38.8 (2.7)	41.0	49
Cochiti Reservoir^b				
1979				
Station 1	0.2 (0.0)	3.9 (0.1)	4.1	20
2	0.1 (0.0)	2.6 (0.1)	2.7	26
3	0.0 (0.0)	0.4 (0.0)	0.4	—
4	1.4 (0.2)	30.3 (0.7)	31.7	22
5	1.3 (0.2)	24.6 (0.7)	25.9	19
6	1.3 (0.4)	26.6 (1.0)	27.9	20
7	0.7 (0.2)	20.7 (1.0)	21.4	30
8	1.4 (0.4)	24.4 (1.1)	25.8	17

Table V (Continued)

Reservoirs	^{238}Pu (fCi/g)	$^{239,240}\text{Pu}$ (fCi/g)	Total Plutonium (fCi/g)	Ratio $^{239,240}\text{Pu}/^{238}\text{Pu}$ (fCi/g)
Cochiti Reservoir (Continued)				
1982				
Station 1	0.7 (0.1)	15.1 (0.8)	15.8	22
2	1.2 (0.1)	25.7 (1.2)	26.9	21
3	0.8 (0.1)	15.6 (0.8)	16.4	20
4	0.8 (0.1)	15.6 (1.0)	16.4	20
5	1.0 (0.1)	17.4 (0.9)	18.4	17
6	1.2 (0.1)	17.9 (1.0)	19.1	15
7	0.8 (0.1)	17.1 (0.8)	17.9	21
1984				
Upper	0.1 (0.1)	14.7 (0.7)	14.8	—
Middle	1.0 (0.1)	16.7 (0.8)	17.7	17
Dam	0.9 (0.1)	27.7 (1.3)	28.6	31
1985				
Upper	2.0 (0.2)	29.2 (2.4)	31.2	15
Middle	1.2 (0.2)	18.9 (1.4)	20.1	16
1986				
Frijoles Canyon	1.1 (0.2)	20.0 (1.2)	21.1	18
Upper	1.0 (0.1)	16.5 (0.6)	17.5	16
Middle	1.9 (0.2)	30.1 (1.6)	32.0	16
Dam	0.9 (0.1)	18.2 (0.7)	19.1	20
1987				
Upper	0.0 (0.0)	2.6 (0.1)	2.6	—
Middle	1.1 (0.1)	29.7 (1.1)	30.8	27
Dam	1.4 (0.1)	20.2 (0.9)	21.6	14
$\bar{X}(S)$	0.9 (0.5)	18.6 (8.5)	20 (9)	20 (5)
Minimum	0.0 (0.0)	0.4 (0.0)	0.4	14
Maximum	1.9 (0.2)	30.3 (0.7)	32	31

^aLocated in Colorado near the Continental Divide.

^bLocated south of Los Alamos.

Note: The numbers in parentheses represent (a) analytical uncertainties if used with a single sample, or (b) a standard deviation of a number if used as a mean.

found at the upper end of the reservoir where current sediments were being deposited. The plutonium increased to 10.0 fCi/g at the middle of the reservoir,

with a further increase to 41 fCi/g near the dam. The high concentration at the dam may reflect the earlier deposition of sediments when fallout was high during

the 1950s. The ratio of the $^{239,240}\text{Pu}$ to ^{238}Pu in the upper part of the reservoir is high, 49; the ratios in the middle of the reservoir at 19 and near the dam at 18 are within the mean ratios of soil, 21(6) (Fig. 3).

Cochiti Reservoir, southeast of Los Alamos, is one of the newer reservoirs on the Rio Grande (Fig. 1). The reservoir was completed in 1972. The drainage area is about $38\,600\text{ m}^2$ ($14\,900\text{ mi}^2$) in southern Colorado and northern New Mexico. It includes water from the Rio Grande and the Rio Chama.

The reservoir sediments were sampled in 1979, 1982, 1984, 1985, 1986, and 1987 (Table V). Eight stations were sampled in 1979, seven in 1982 and two to four in the remaining years (Fig. 5).

The concentrations of plutonium in each sample set are random, and there is no pattern according to location or years of collection. The total plutonium concentrations range from 0.4 to 32 fCi/g, with a mean ratio of about 20(9) fCi/g. The concentrations of plutonium from Cochiti Reservoir are higher than those found in the reservoirs on the Rio Chama but are comparable with those found in the Rio Grande Reservoir. The sediments in Cochiti and Rio Grande reservoirs contain a higher fraction of finer sediment particles and organic materials than occurs in the reservoirs on the Rio Chama. These features enhance the capacity of the sediments to adsorb plutonium, resulting in higher concentrations of plutonium.

The ratios of $^{239,240}\text{Pu}$ to ^{238}Pu in Cochiti Reservoir ranged from 14 to 31, with an average of 20(5). This is not significantly different from the mean ratios of soil, 21(6), which are near the mean ratios in sediments from Rio Grande Reservoir and near those from the three reservoirs on the Rio Chama.

VI. CONCLUSIONS

There was significant difference between the highest and lowest total plutonium concentrations and the ratios of $^{239,240}\text{Pu}$ to ^{238}Pu of five samples collected in a 9-m^2 (30-ft^2) area. The difference was a factor of about 3, indicating that even in a small area plutonium concentrations and ratios from fallout are not homogeneous.

The concentrations of plutonium in soil samples collected from five stations in a 15-m (50-mi) radius of

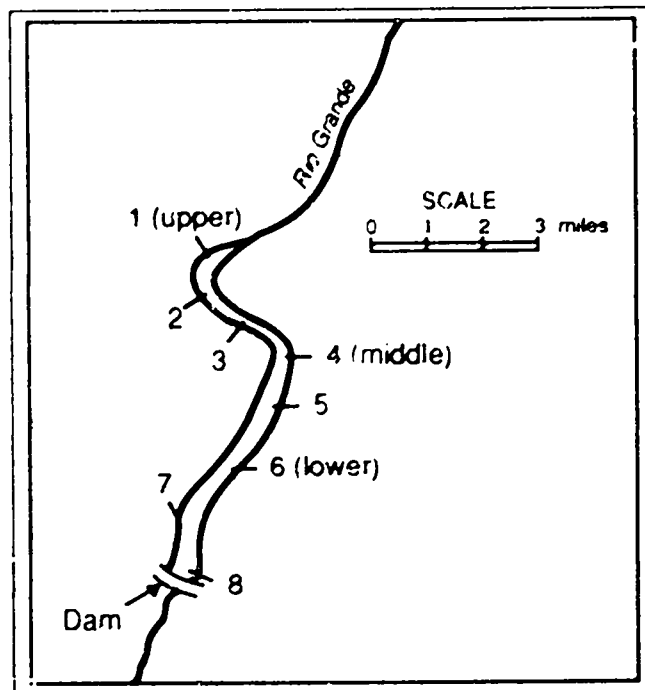


Fig. 5. Sediment stations at Cochiti Reservoir.

Los Alamos in 1981 and 1982 varied from station to station and from year to year. The concentrations varied by a factor of 4 in 1981 and by a factor of 5 in 1982.

Nine soil samples collected in northern New Mexico and southern Colorado in 1986 were divided into three groups according to concentrations. Low concentrations of plutonium (1.3 to 7.9 fCi/g) were found in the high interbasin of the San Luis Valley, intermediate concentrations (11.1 to 16.2 fCi/g) were found on the high mountain slope east of the continental divide, and high concentrations (20.1 to 84.9 fCi/g) were found in the high passes along the continental divide. The concentrations in the high mountain passes increased from south to north. The mean ratio of $^{239,240}\text{Pu}$ to ^{238}Pu in the soil was 21(6).

River sediments were collected from six stations on the Rio Grande. The concentrations of the plutonium in the river sediments were lower than those found in the soils, ranging from 1.1 to 9.3 fCi/g, with a mean of 5.0(3). The ratio of $^{239,240}\text{Pu}$ to ^{238}Pu ranged from 10 to 71, indicating that physical transport by the river has caused a further distribution of plutonium, with the fine-grained sediments being winnowed out of the bed sediments.

Concentrations of plutonium in reservoir sediments from the Rio Chama are much lower than those found in the reservoir sediments on the Rio Grande. The mean total plutonium concentrations from the three reservoirs on the Rio Chama were 12, 8, and 9 fCi/g; the mean total plutonium concentrations for the two reservoirs on the Rio Grande were 19 and 20 fCi/g. The mean ratio of $^{239,240}\text{Pu}$ to ^{238}Pu from the three reservoirs on the Rio Chama ranged from 23 to 25. The same ratio for the upper reservoir on the Rio Grande was 29; that of the reservoir below Los Alamos was 20, about the same as that found in the soils.

The variations in plutonium concentrations and in the ratios of $^{239,240}\text{Pu}$ to ^{238}Pu in soils, river sediments, and reservoir sediments indicate that the deposition of plutonium fallout is not homogeneous, but is due to differences in regional and local weather patterns that cause selective areas for fallout, differences in particle sizes of the plutonium that result in variations of concentration, and differences in distribution of the plutonium because of varying erosion and transport conditions.

REFERENCES

ESG 1988: Environmental Surveillance Group, "Environmental Surveillance at Los Alamos During 1987," Los Alamos National Laboratory report LA-11306-ENV (1988).

Gautier, M. A., E. S. Gladney, W. D. Moss, M. B. Phillips, and B. T. O'Malley, "Quality Assurance for Health and Environmental Chemistry: 1986," Los Alamos National Laboratory report LA-11114-MS (1987).

Hardy, E. P., P. W. Krey, and H. L. Volchok, "Global Inventory and the Distribution of Fallout Plutonium," *Nature* 241(5390), 444-445 (1973).

Harley, J. H., "Worldwide Plutonium Fallout from Weapons Tests," in the "Proceedings of the Environmental Plutonium Symposium," Los Alamos Scientific Laboratory report LA-4756 (1971).

Little, C. A., "Plutonium in a Grassland Ecosystem," U.S. Energy Resource Development Administration (ERDA) report COO-1156-83 (1977).

Muller 1977: R. N. Muller and D. G. Sprugel, "Distribution of Local and Stratospheric Plutonium in Ohio Soil," *Soil Science* 33, 405-408 (1977).

Muller 1978: R. N. Muller, D. G. Sprugel, and B. Kohn, "Erosion Transport and Deposition of Plutonium and Cesium in Two Small Midwestern Watersheds," *Journal of Environmental Quality* 7, 171-174 (1978).

Nyhan 1976A: J. W. Nyhan, F. R. Miera, Jr., and R. E. Neher, "Distribution of Plutonium in Trinity Soils after 28 Years," *Journal of Environmental Quality* 5, 431-437 (May 1976).

Nyhan 1976B: J. W. Nyhan, F. R. Miera, Jr., and R. J. Peters, "Distribution of Plutonium in Soil Particle Size Fraction in Liquid Receiving Areas at Los Alamos," *Journal of Environmental Quality* 5, 50-56 (May 1976).

Perkins, R. W., and C. W. Thomas, "Worldwide Fallout," in "Transuranic Elements in the Environment," W. C. Hanson, Ed. (U.S. Department of Energy, Washington, DC, 1980).

Purtymun, W. D., R. J. Peters, and M. N. Maes, "Background Concentrations of Radionuclides in Soils and River Sediments in Northern New Mexico, 1974-1986," Los Alamos National Laboratory report LA-11134-MS (1987).

Sills, C. W., "The Particle-Size Problem as Related to Sample Inhomogeneity," in the "Proceedings of the Environmental Plutonium Symposium," Los Alamos Scientific Laboratory report LA-4756 (1971).

Tamura, T., "Distribution and Chemical Characterization of Plutonium in Soils from the Nevada Test Site," *Journal of Environmental Quality* 4, 350-354 (April 1975).