CHAPTER VII

SEDIMENT MONITORING

Long-lived contaminants, deposited in aquatic ecosystems by windborne deposition or by fluvial deposition, tend to accumulate in bottom sediments in discrete layers. There is little or no vertical migration of radionuclides such as ¹³⁷Cs and plutonium between undisturbed sediment layers. Thus, the temporal history of atmospheric or liquid releases to a region can be preserved in the sediment of a nearby body of water. Cesium-137 and isotopes of plutonium have been used as indicators of the amount of atmospheric radioactive fallout deposited in sediment over time (Krey et al. 1990; Whicker et al. 1994).

The Rocky Flats watershed includes several creeks and water impoundments (see Chapter VI, Figures <u>VI-1</u> and <u>VI-2</u>). Two significant bodies of waters east of the Rocky Flats site are the Great Western Reservoir and Standley Lake. The Great Western Reservoir received liquidborne contaminants from the site, via Walnut Creek, and it lies in the downwind area that was measurably impacted by deposition of airborne contaminants. Standley Lake may have received some contaminants from Woman Creek, but it was primarily impacted by deposition of airborne contaminants. Mower Reservoir, which is located upstream of Standley Lake, is also of some interest to the dose reconstruction.

The contractor of the Rocky Flats Plant (RFP) conducted some routine monitoring of sediment for radionuclides in the early 1950s and 1970s. Generally, however, that routine monitoring is not useful to the dose reconstruction study. The programs involved few sampling sites, locations and methods were not identified, and results were infrequently reported.

Various special sediment studies have been conducted in the Rocky Flats environs by different organizations. These studies have focused primarily on the Great Western Reservoir, and to a lesser extent on Standley Lake, and are a source of data that may be useful to the dose reconstruction. This chapter presents the results of an evaluation of these data.

OVERVIEW OF MAJOR STUDIES

Various studies have been conducted at and around Rocky Flats. <u>Table VII-1</u> summarizes the sediment studies conducted to date. Most studies have focused on measuring plutonium because this was the primary contaminant of concern released from Rocky Flats operations and it persists in sediment layers. Cesium-137 was also measured often because it is easily analyzed and it provides clues to the temporal history of past depositions. The studies most pertinent to the dose reconstruction are summarized below. Items of particular interest to the dose reconstruction are highlighted.

<u>Poet and Martell</u> (1972) focused primarily on studying contaminated soils surrounding the RFP site following the May 11, 1969, fire in Building 776/777. Only two surface sediment samples were collected at the Great Western Reservoir. The remaining six samples were obtained at a pond located on Walnut Creek and at other surface impoundments. Standley Lake was not included. This study is most useful for the measurements of ^{239,240}Pu concentrations in background lakes (i.e., Calkins Lake, Boulder Reservoir, and Boyd Lake).

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Study date	Organization ^a / Reference	Location(s) ^b	Sampling method	No. of samples	Contaminants	
Aug. 1969– Jul. 1970	NCAR/Poet and Martell (1972)	GWR, CL, Boulder R., Boyd Lake, HP	Not reported	8	Pu, ²⁴¹ Am (GWR and HP)	
Feb. 1970	EPA / <u>EPA</u> (1971)	GWR, SL, MR, CL, WLC, WMC	Hand trowel	16	Pu, ⁹⁰ Sr, U, Gross alpha, total alpha radium	
Sept.	EPA / <u>EPA</u> (1973)	MR, CL, WLC, WMC	Hand trowel	9	Pu	
1970		GWR, SL	Dredge	22 (20 – GWP)	Pu	
		GWR, SL	Core	14 (12 – GWR)	Pu	
Sept. 1973	EPA / <u>Lammering</u> (1975)	GWR, SL, RR, Marston Lake, Cherry Creek Res.	Dredge	46 (20 – GWR, 17 – SL)	²³⁸ Pu, ²³⁹ Pu ^c , ¹³⁷ Cs, ⁸⁹ Sr, ⁹⁰ Sr	
		GWR, SL	Core	24 (15 – GWR)	²³⁸ Pu, ²³⁹ Pu ^c	
Oct. 1973	Dow/ <u>Kunert and</u> <u>Werkema</u> (1974)	GWR, SL	Dredge, core	Dow ~1/3 of 1973 EPA samples	²³⁹ Pu ^c , ¹³⁷ Cs	
Jan. 1971– Apr. 1974	CSU/Johnson et al. (1974)	НР	Polyvial (composites of 5 gm samples collected monthly)	17 to 24 at each of 7 HPs - $\sim \frac{1}{2}$ before and $\sim \frac{1}{2}$ after recon-	²³⁹ Pu ^c	
		WLC HP	Dredge Core	8 18	²³⁹ Pu ^c ²³⁹ Pu ^c	
Apr.– May 1974	PNL/ <u>Thomas and</u> <u>Robertson</u> (1981)	GWR, SL	Core	9	²³⁹ Pu ^c , ¹³⁷ Cs	
Aug. 1976	EML / <u>Hardy et al.</u> (1980)	SL	Core	1	²³⁸ Pu, ²³⁹ Pu, ²⁴⁰ Pu, ¹³⁷ Cs, ²⁴¹ Am	
Mar. 1979	Rockwell / Hurley (1979)	GWR spillway	Hand trowel	14	²³⁹ Pu ^c , ²⁴¹ Am	
Mar. 1980	Rockwell/ Hurley (1980)	GWR spillway	Core Core	14 7	²³⁹ Pu ^c , ²⁴¹ Am ²³⁹ Pu ^c , ²⁴¹ Am	
Aug.	Rockwell/	GWR	Dredge	43	²³⁹ Pu ^c	
1985	Campben (1985)		Core	4	²³⁹ Pu ^c	
Aug. 1984	Rockwell/ Setlock and Paricio (1984)	SL	Dredge	63	²³⁹ Pu ^c	
1990	CO School of Mines/ <u>Cohen et al.</u> (1990)	Wellington Lake, Halligan Reservoir	Core Core	4 2	²³⁹ Pu ^c ²³⁹ Pu ^c , ¹³⁷ Cs, ²¹⁰ Pb	
1991	City of Broomfield/<u>Schmidt</u> (1993)	GWR	Core	30	²³⁹ Pu ^c	

Study date	Organization ^a / reference	Location(s) ^b	Sampling method	No. of samples	Contaminants
1992	CSU / <u>Whicker et al.</u> (1994)	GWR	Core	48	²³⁹ Pu ^c , ¹³⁷ Cs
1992	DOE / <u>DOE</u> (1995)	GWR, SL, MR, WLC, WMC, DC, VD, CD, CC, BDC	Dredge or scoop	128 (most at GWR and SL)	²⁴¹ Am, ¹³⁷ Cs, ²²⁶ Ra, ^{233,234} U, ²³⁵ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu ^c , metals
			Core	12 (5 - GWR, 4 – SL, 3 –	²⁴¹ Am, ¹³⁷ Cs, ²²⁶ Ra, ^{233,234} U, ²³⁵ U, ²³⁸ U, ²³⁸ Pu, ²³⁹ Pu ^c , elements
1992	USGS/ <u>Clow and</u> Johncox (1995)	GWR, SL, MR	Dredge	MR) 34 (15 – GWR, 16 – SL, 3 - MR	²³⁹ Pu ^c

^aCSU = Colorado State University; DOE = U. S. Department of Energy; EML = Environmental Measurements Laboratory, DOE; EPA = U.S. Environmental Protection Agency; NCAR = National Center for Atmospheric Research; PNL = Pacific Northwest Laboratory.

^bBDC = Big Dry Creek; CC = Coal Creek; CD = Church Ditch; CL = Calkins Lake; DC = Dry Creek; GWR = Great Western Reservoir; HP = RFP holding ponds; MR = Mower Reservoir; RR = Ralston Reservoir; SL = Standley Lake; VD = Valley Ditch; WLC = Walnut Creek; WMC = Woman Creek.

^cUnless specified otherwise, ²³⁹Pu is actually ²³⁹Pu plus ²⁴⁰Pu.

In 1970 and 1973, the U.S. Environmental Protection Agency (EPA) performed the first indepth studies of plutonium in surface sediments of the Great Western Reservoir and Standley Lake (EPA 1973; Lammering 1975). These studies are significant because the samples were collected before and after the RFP holding pond reconstruction in 1972, thus, providing data for before and after comparisons.

In 1973, the Dow Chemical Company, contractor for the RFP, conducted a study of the Great Western Reservoir and Standley Lake (Kunert and Werkema 1974). The EPA asked Dow to participate in the analysis of one-third of the core samples collected by EPA. The results were exchanged, and from these data Kunert and Werkema generated ¹³⁷Cs and ^{239,240}Pu concentration contour drawings and depth profiles for the Great Western Reservoir and Standley sediments.

From the spring of 1971 through the summer of 1973, Colorado State University (CSU) investigated changes in ^{239,240}Pu concentrations in biota, water, and surface sediment collected from six holding ponds (A-1, B-1, B-2, B-3, B-4, and C-1) located on the Rocky Flats site and from a small impoundment or pond located downstream of the holding ponds on Indiana Street (see Figure VI-1). The in-depth study chronicled the impacts of pond reconstruction in 1972 and an accidental release of process waste solutions low in ^{239,240}Pu in March 1971. A total ^{239,240}Pu inventory for the holding ponds was also estimated.

Lake bottom sediment cores and surface samples were collected in 1974 from Standley Lake the Great Western Reservoir by a U.S. Department of Energy (DOE) contractor, Battelle Pacific Northwest Laboratory (PNL). The surface grab samples were analyzed for gamma-emitting radionuclides. The cores were analyzed for gamma-emitting radionuclides and, in some cases, for ^{239,240}Pu. The Standley Lake core was not analyzed for ^{239,240}Pu. Using these data, Thomas and Robertson drew radionuclide (^{239,240}Pu, ²⁴¹Am, and ¹³⁷Cs) concentration contours for the Great Western Reservoir (Thomas and Robertson 1981). They also prepared graphs of radionuclide concentrations versus depth of sediment in the Great Western Reservoir and estimated sedimentation rates and radionuclide inventories for the two lakes.

In 1976, the Environmental Measurements Laboratory of DOE collected a core at Standley Lake to determine the history of plutonium releases (<u>Hardy et al.</u> 1980). The history of deposition

Sediment sampling techniques usually fall into two broad categories: (1) bottom grab (dredge) sampling and (2) core sampling. Bottom grab samplers have the advantage of obtaining a larger sample over a broader expanse of bottom; however, they can only sample a shallow depth (up to 15 cm. [6 in.]). Core samplers are used to sample a much greater vertical depth; however, an extremely small area of bottom is studied.

of ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, ²⁴¹Am, and ¹³⁷Cs was elucidated and used to date specific fallout and RFP release events. This study differed from previous studies in that mass spectroscopy was used to determine ²³⁹Pu and ²⁴⁰Pu concentrations. Specific ²³⁹Pu/²⁴⁰Pu atom ratios were used to indicate the sources of the plutonium (i.e., nuclear weapons testing, nuclear-powered satellite failure, or releases from Rocky Flats).

Rockwell International conducted additional studies in 1983 and 1984 to improve the existing knowledge of environmental radionuclide concentrations and relevent transport phenomena in Standley Lake and the Great Western Reservoir (Campbell 1985; Setlock and Paricio 1984). In each of these studies, numerous sediment grab samples and cores were collected the Great Western Reservoir and Standley Lake and were analyzed for ^{239,240}Pu by Rockwell, the City of Broomfield, and the Colorado Department of Health. Contour drawings and core profile graphs were constructed from the data.

In 1992, DOE initiated a large Resource Conservation and Recovery Act (RCRA) Facility Investigation/Remedial Investigation (RFI/RI) of Operable Unit 3 (OU3), which is the offsite areas surrounding the RFP. Included in the study was an investigation of the sediment from the surface water reservoirs and drainages in the area. The objective of the sediment study was to determine the contaminants-of-concern (COCs) for the RFI/RI. Not surprisingly, only ^{239,240}Pu was determined to be a COC with respect to human health in the Great Western Reservoir. Other radionuclides and elements in the surface sediments of the Great Western Reservoir, Standley Lake, and Mower Reservoir were determined to be below baseline levels established for the RFI/RI. This study is useful to the dose reconstruction because of the amount of high quality data available, including radioisotope sediment dating of sediment horizons, and because it provides validation of data collected by Rockwell in 1983 and 1984. The sampling design included collecting grab and core samples at sites collocated with randomly selected sites previously sampled by Rockwell. This portion of the study was conducted by the U.S. Geological Survey (USGS) (Clow and Johncox 1995).

Plutonium-239,240 was the only contaminant of concern for cleanup of Great Western Reservoir based on measurements in 1992.

Inventory refers to the total amount of material present at a given place. Plutonium inventory in reservoir sediment is determined by multiplying the concentration (pCi g^{-1}) by the total mass (grams) in a given layer and summing the results over the depth of sediment.

Researchers at CSU also collected numerous sediment

core samples from the Great Western Reservoir in 1992. They either sectioned or homogenized the cores and then analyzed them for ^{239,240}Pu and ¹³⁷Cs. They then described the horizontal and

vertical distributions of ^{239,240}Pu and made inventory estimates. In addition, they calculated the fraction of plutonium in sediments that was contributed from the aquatic pathway.

BACKGROUND CONCENTRATIONS

Few measurements have been made of contaminants in lake and reservoir sediments outside the area of possible impacts from Rocky Flats releases. Table VII-2 presents plutonium levels in the sediments of other Colorado front range lakes and reservoirs. Except for Boyd Lake sampling in 1992, the results in this table represent single measurements. However, they provide a range of background values that reflect global fallout rather than releases from Rocky Flats. Based on these measurements, it could be concluded that 239,240 Pu concentrations in sediments resulting from global radioactive fallout are less than 0.2 pCi g⁻¹, although most results are less than 0.1 pCi g⁻¹.

Table VII-2. Maximum Concentrations of Plutonium in SomeColorado Front Range Lakes and Reservoir Sediments

Location	Date sampled	^{239,240} Pu (pCi g ⁻¹)	References
Calkins Lake ^a	Sept. 1969	0.049	Poet and Martell (1972)
Boulder Reservoir ^a	Sept. 1969	0.007	Poet and Martell (1972)
Boyd Lake ^a	Sept. 1969	0.006	Poet and Martell (1972)
Calkins Lake ^b	Feb. 1970	0.06	<u>EPA</u> (1971)
Autrey Reservoir ^b	Feb. 1970	0.03	<u>EPA</u> (1971)
Cherry Creek Reservoir ^b	April 1974	< 0.05	Lammering (1975)
Marston Lake ^b	April 1974	0.13	Lammering (1975)
Ralston Reservoir ^b	April 1974	0.06	Lammering (1975)
Halligan Reservoir ^c	1988	0.07	<u>Cohen et al.</u> (1990)
Wellington Lake ^d	1988	0.19	<u>Cohen et al.</u> (1990)
Boyd Lake ^e	1992	0.009	Schoep and Whicker (1995)

^a Sampling method not reported.

^b Grab or dredge sample.

^c Sediment core peak concentration at 21–22 cm (8.3–8.7 in.) sediment depth.

^d Sediment core peak concentration at 10–11 cm (4–4.3 in.) sediment depth.

^e Result represents median concentration of seven homogenized cores.

MEASUREMENTS AT GREAT WESTERN RESERVOIR AND UPSTREAM LOCATIONS

The most probable sources of Rocky Flats contaminants, in particular plutonium, in the Great Western Reservoir are RFP disposal practices in the 1950s and 1960s; construction activities at the holding ponds in the early 1970s; and the deposition of airborne plutonium, particularly following the 903 Area disturbance (DOE 1991). Moreover, the plutonium released

into surface waters is efficiently retained by the bottom sediment and is immobile, thus, providing an historical record of release events. These conclusions are supported by sediment studies conducted at the Great Western Reservoir, at the Rocky Flats site holding ponds, and on Walnut Creek.

The RFP began operations in 1952, with the primary mission of producing plutonium triggers and other metal components for nuclear weapons. During operations, liquid waste effluent (in the form of process wastes, sewage treatment plant effluent, and laundry wastewater) was released to Walnut Creek, an intermittent stream draining the northern portion of the RFP site. As discussed in <u>Chapter VI</u>, a series of holding ponds was built on the north and south branches of Walnut Creek between 1952 and 1980 to collect RFP liquid releases and control onsite surface runoff. From June through October 1972, repair and construction of the retention ponds resuspended sediment into the water column. This resulted in the redistribution of the radionuclides in the water and sediment of the holding ponds of Walnut Creek.

Johnson et al. (1974) and Paine (1980) found that plutonium concentrations in the water and sediment increased in the holding ponds during construction activities. Figure VII-1 illustrates this trend for Pond B-1, which was estimated to have the highest inventory of ^{239,240}Pu among the retention ponds (Table VII-3). Plutonium concentrations also increased in the sediment sampled at Walnut Creek at Indiana Street (a small pond located downstream of the holding ponds and west of the Great Western Reservoir), which indicated that considerable activity escaped the holding pond system during the period of reconstruction (Figure VII-2). The plutonium concentrations in the water column of the small impoundment increased during the period of pond reconstruction and remained relatively high for about 5 months following the increase. This suggests a flushing of resuspended sediment down Walnut Creek toward the Great Western Reservoir.

Studies conducted in 1970 and 1973 by EPA (<u>EPA</u> 1973; <u>Lammering</u> 1975) at Great Western Reservoir also support the conclusion that resuspended sediment was flushed down Walnut Creek toward Great Western Reservoir during the 1972 holding pond reconstruction.

When the spatial distributions of plutonium from 1970 and 1973 were compared, it was evident that the patterns had changed (Appendix F, Figure F-1a). In 1970, the highest concentrations of plutonium were detected in sediments in the Walnut Creek inlet area and the central section of the reservoir (leading to the dam inlet). The lowest concentrations were found in the south arm, the shoreline area between the south arm and the dam, and the western portion of the north arm. By 1973, the highest concentrations were found in the deepest areas of the reservoir rather than in the Walnut Creek inlet area, indicating that the ^{239,240}Pu in the uppermost sediment layer had increased substantially in the 3 years between the studies. These changes are illustrated in Figures <u>VII-3</u>, <u>VII-4</u>, and <u>VII-5</u>. Figures <u>VII-4</u> and <u>VII-5</u> were created using the EPA datasets and the SurferTM Surface Mapping System. Note that the highest concentration in 1970 was 0.85 pCi g⁻¹ in 1970, and the highest concentration in 1973 was 4.1 pCi g⁻¹. The average concentration of 239,240 Pu in surface sediments, collected with a dredge, was 0.3 pCi g⁻¹ (n = 20) in 1970 and 1.6 pCi g⁻¹ (n = 20) in 1973. Comparable results were noted for the top 5 cm (2 in.) of core samples. The average concentration of 239,240 Pu in the top 5 cm (2 in.) of core



Figure VII-1. Mean plutonium concentrations in surface sediments (pCi g^{-1}) and in unfiltered water (pCi L^{-1}) for Pond B-1 from April 1971 through August 1973 (from Paine 1980).



Figure VII-2. Mean plutonium concentrations in surface sediments (pCi g^{-1}) and in unfiltered water (pCi L^{-1}) for an unnamed impoundment downstream of the holding ponds from April 1971 through August 1973 (from <u>Paine</u> 1980). This sampling station is a small pond located where Walnut Creek crosses under Indiana Street.

VII-7

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Table v11-5. Inventories of Pu in Holding Ponds During I			
Pond	^{239,240} Pu (mCi)	^{239,240} Pu (%)	
B-1	84.5	62.4	
B-2	27.0	19.9	
B-3	19.4	14.3	
B-4	4.6	3.4	
Total ^a Source: Paine (1980).	135.5	100.0	

 Table VII-3. Inventories of ^{239,240}Pu in Holding Ponds During 1971^a



Figure VII-3. Concentrations of 239,240 Pu (pCi g⁻¹) in surface sediments of Great Western Reservoir versus water depth in 1970 and 1973 (<u>EPA</u> 1973; <u>Lammering</u> 1975). Samples were collected using a dredge. Note the 5-fold difference in vertical scales of the two plots.



Figure VII-4. Spatial distribution of 239,240 Pu (pCi g⁻¹) in Great Western Reservoir during 1970. Contours were created from data (n = 20) in <u>EPA</u> (1973). Samples were collected using a dredge.



Figure VII-5. Spatial distribution of 239,240 Pu (pCi g⁻¹) in Great Western Reservoir during 1973. Contours created from data (n = 20) in <u>Lammering</u> (1975). Samples were collected using a dredge.

Data collected from the Great Western Reservoir in 1974 by PNL also demonstrated that the highest radionuclide concentrations, including ¹³⁷Cs, ^{239,240} Pu, ²⁴¹Am, could be found in the

A study by PNL in 1974 indicated that the main source of ^{239,240}Pu and ²⁴¹Am in surface sediments of Great Western Reservoir was the RFP and not fallout from nuclear weapons testing. However, ¹³⁷Cs concentrations were indicative of fallout background levels and could be used to date sediment layers. deepest areas of the reservoir (see Appendix F, Figures <u>F-2</u>, <u>F-3</u>, and <u>F-4</u>). The authors also observed that concentrations of ^{239,240}Pu and ²⁴¹Am exceeded expected baseline levels (<u>Thomas and Robertson</u> 1981). On the other hand, ¹³⁷Cs concentrations were typical of fallout background levels observed at other locations in the United States. Using ¹³⁷Cs to age date sediment layers (the highest fallout levels were deposited in 1963), <u>Thomas and Robertson</u> (1981) observed that two large ^{239,240}Pu deposition events were apparent in a core collected in the deep part of the Great Western Reservoir (see <u>Figure VII-6</u>). The maximum ^{239,240}Pu concentration peak occurred at a depth corresponding to sediments deposited around 1968–1969. A smaller peak corresponds to sediments deposited around 1960. The authors attributed these maxima to controlled waterborne releases from the plant, although

they also noted that the later peak could have a fallout component. Current opinion, based on subsequent studies, is that the 1968-1969 peak corresponds to a period of windborne releases from the 903 Area. An increase from 1972–1974, not addressed by <u>Thomas and Robertson</u> (1981) is also evident in <u>Figure VII-6</u>, and it possibly corresponds to the 1972 holding pond reconstruction disturbances.

The average ^{239,240}Pu concentration in the top 5 cm (2 in.) of the eight cores collected by PNL in 1974 (Thomas and Robertson 1981) was 3.6 pCi g⁻¹, compared to a mean of 1.6 pCi g⁻¹ estimated using the 1973 EPA results. The maximum ^{239,240}Pu concentration in 1974 was 6.04 pCi g^{-1} , compared to the maximum concentrations of 4.5 pCi g^{-1} measured in 1973. However, the PNL results are biased high because eight of the nine cores were collected in the deepest sections of the reservoir (Zones A and B in Appendix F, Figure F-4). Thomas and Robertson (1981) divided the reservoir into three zones of plutonium activity and sediment thickness. Zone A represents the deepest layer of sediments [16 to 20 in. (40 to 50 cm)] containing the highest plutonium activity, and it is located in the eastern end of the reservoir. Sediment deposition rates were estimated to be 2.08 to 3.68 cm (0.82 to 1.45 in.) per year in this region (based on 137 Cs concentrations to date core segments). Zone B was designated a region of intermediate thickness (estimated to average about 20 cm [8 in.] deep) and plutonium activity, and it extended to the west from Zone A. Sedimentation rates ranged from 0.25 to 2.08 cm (0.1 to 0.82 in.) per year in Zone B. Zone C included the remaining area, which was characterized by a thin deposit of sediments (about 5 cm [2 in.] deep) of relatively low activity. Sedimentation rates appeared to be less than 0.25 cm (0.1 in.) per year in the shallow, perimeter area. Using average plutonium concentrations and sediment thicknesses estimated for each area, Thomas and Robertson (1981) estimated the total inventory of ^{239,240}Pu to be 244 mCi. Based on a measured ²⁴¹Am/^{239,240}Pu activity ratio of 0.3, the authors calculated an ²⁴¹Am inventory of 73 mCi in the reservoir sediments.



Figure VII-6. Depth distribution of ¹³⁷Cs and ^{239,240}Pu in a sediment core collected from the Great Western Reservoir in 1974 (<u>Thomas and Robertson</u> 1981).

The Rockwell studies conducted in 1983 (<u>Campbell</u> 1985) confirmed earlier observations that plutonium occurred in discrete sediment horizons corresponding with historical releases from the RFP and that these horizons had been buried to varying depths by subsequent sedimentation. Unfortunately, ¹³⁷Cs was not measured, so the cores were not dated using the 1963 fallout peak. However, the peak ^{239,240}Pu concentration in the shallower horizon was thought to correspond with 1969 releases from the 903 Area and the 1969 fire. Sedimentation was estimated to be approximately 2.5 cm (1 in.) per year.

In general, the Rockwell data from 1983 appear to be of acceptable quality because there was good agreement between split samples analyzed by Rockwell, the City of Broomfield, and the Colorado Department of Health. A mean ^{239,240}Pu concentration of 0.2 pCi g⁻¹ was estimated for surface sediments based on measurements reported in <u>Campbell</u> (1985) (<u>Appendix F</u>, Figure F-5). The maximum concentration was 1.0 pCi g⁻¹. It is interesting that this is comparable to pre-1973 measurements, indicating the deposition of less-contaminated materials over the older sediments. In fact, sediment samples co-located with 30 stations used in the 1973 EPA study had lower concentrations than their 1973 counterparts.

A comparison of ^{239,240}Pu surface sediment data grab samples collected by the USGS for the 1992 Resource Conservation and Recovery Act Facility Investigation/Remedial (RFI/RI) Report for Operational Unit 3 (OU3) with surface sediment data collected in 1983 by Rockwell shows a statistically significant lower mean concentration in 1992 (lower by 0.14 pCi g⁻¹) (see Figure VII-7) (Clow and Johncox 1995). This seems to provide additional confirmation that new sediments with relatively low ^{239,240}Pu concentrations have buried older sediments containing higher concentrations of ^{239,240}Pu. In general, trends in ^{239,240}Pu concentrations with depth in 1992



Figure VII-7. Concentrations of ^{239,240} Pu in surface sediment of Standley Lake, Great Western Reservoir, and Mower Reservoir in 1992 (<u>Clow and Johncox</u> 1995). See <u>Figure VI-1</u> for location of Rocky Flats Plant and these water bodies.

were similar to trends in the 1983 study (Figures <u>VII-9</u> and <u>VII-10</u>). Maximum concentrations occurred at depths ranging from 33 to 79 cm (13 to 31 in.) below the sediment-water interface at most sites. Based on the deposition rates determined via age dating of sediments cores (Figure <u>VII-8</u>), core profiles corresponding to deeper areas of the impoundment indicate that an elevated deposition period peaked at around 1970–1971 (Figures <u>VII-8</u>, <u>VII-9</u>, and <u>VII-10</u>). Temporal differences in sediment deposition rates, as well as compaction during core sampling, does not allow for accurate dating of each section of the core using only the ¹³⁷Cs peak. The ^{239,240}Pu peak most likely corresponds with the 903 Pad release in 1969. However, the 1971–1972 holding pond release may be superimposed on the elevated concentrations from 33 to 56 cm (13 to 22 in.).

There was a small, but statistically significant, difference in ^{239,240}Pu concentrations in two co-located sediment cores collected in 1983 and in 1992 (<u>Clow and Johncox</u> 1995). Measured concentrations tended to be higher in 1983 than in 1992, and differences tended to increase with concentration. Spatial variations in sediment and ^{239,240}Pu deposition may contribute to this difference.

The CSU study of 1992 indicates that over onehalf of the ^{239,240}Pu inventory in the Great Western Reservoir is in the bottom sediments of the deepest parts of the basin. The investigation conducted by CSU at the Great Western Reservoir in 1992 included the evaluation of the greatest number of sediment cores (48) ever collected on this reservoir. Only three of the cores were examined as a depth profile. The remaining cores were homogenized in an attempt to determine the total inventory of ^{239,240}Pu in the sediment. Depth-integrated core inventories and homogenized core concentrations of ^{239,240}Pu (Figure VII-11) are highly variable across the reservoir, but they are highest in the deepest



Figure VII-8. Relation of concentrations of ^{239,240}Pu and ¹³⁷Cs to depth at sediment sampling site SED09192 collected from Great Western Reservoir in 1992. The ¹³⁷Cs peak, which corresponds to the peak fallout year, was used to date the core segments (DOE 1995).



Figure VII-9. Relation of concentrations of ^{239,240}Pu to depth at sediment sampling site SED08592 collected from Great Western Reservoir in 1983 and 1992. A sedimentation rate of 2.3 cm (0.9 in.) per year was assumed for the 1983 data (<u>Clow and Johncox</u> 1995).



Figure VII-10. Relation of concentrations of ^{239,240}Pu to depth at sediment sampling site SED09192 collected from Great Western Reservoir in 1983 and 1992. A sedimentation rate of 1.2 in. (3.0 cm) per year was assumed for the 1983 data (<u>Clow and Johncox</u> 1995).



Figure VII-11. Concentrations of ^{239,240}Pu in homogenized cores collected from the Great Western Reservoir in 1992 (<u>Schoep and Whicker</u> 1995) (1Bq kg⁻¹ = 0.027 pCi g⁻¹).

One becquerel per kilogram is equivalent to 0.027 picocuries per gram. portions of the basin. Both integrated and homogenized core inventories and concentrations were significantly correlated to water depth. The increase in average concentration of plutonium [Pu] (becquerels per kilogram) with water depth (D, in meters) was described by

$$[Pu] = 0.648 \times e^{0.326D}$$
(VII-1)

There was also evidence that the primary plutonium-bearing sediments are buried relatively deep in the sediment bed. Core lengths in the study ranged from 9.5 to 29.5 cm (3.7 to 11.6 in.). Homogenized and integrated core inventories and plutonium concentrations were significantly correlated with core length. Plutonium concentration [Pu] (becquerels per kilogram) was related to core length (L, in centimeters) by

$$[Pu] = 0.088 \times e^{0.087L}.$$
 (VII-2)

Based on three core profiles, the observation made in previous studies that plutonium could occur in distinct layers of the sediment was apparent, although peak concentrations and depths varied among cores. The highest concentration (3.2 pCi g⁻¹) was seen in the 60 to 65-cm (24 to 26-in.) segment of a core collected near the dam, which is an area of high sedimentation (Figure VII-12).

The other two cores were collected near the middle of the reservoir where sedimentation rates are lower. All three cores had one primary peak of activity at 20 to 30 cm (7.9 to 12 in.) of



Figure VII-12. Plutonium concentration as a function of sediment depth in a sediment core collected in the deep portion of Great Western Reservoir in 1992. Error bars are 95% confidence intervals (<u>Schoep and Whicker</u> 1995).

sediment. Sedimentation rates were estimated, from core data, to be approximately 1.2 and 3.1 cm (0.5 and 1.2 in.) per year, respectively, in the middle and eastern portions of the reservoir. These estimates are similar to those made in Thomas and Robertson (1981) and Campbell (1985).

<u>Schoep and Whicker</u> (1995) estimated the ^{239,240}Pu inventory in sediments of the Great Western Reservoir using three methods:

- 1. Take the product of the median reservoir-wide activity per unit area (0.08 μ Ci m⁻²) and the area of the reservoir (0.67 km² [0.26 mi²]), which yielded an estimate of 54 mCi.
- 2. Divide the reservoir into 4-m (13-ft) contours and multiply the median plutonium activity per unit area for the depth. These were summed to produce a median plutonium inventory of 62 mCi.
- 3. Try to duplicate the method used by <u>Thomas and Robertson</u> (1981). Using this method, the inventory was estimated to be 92 mCi, compared to 244 mCi calculated by <u>Thomas and Robertson</u> (1981).

Schoep and Whicker attribute the differences in total inventory estimates, using the method of Thomas and Robertson, to several possible factors, including

- 1. Insufficient core lengths in the more recent study to obtain complete plutonium profiles in sediment.
- 2. Inaccuracy in the <u>Thomas and Robertson</u> (1981) estimate, which was based on relatively few samples (eight cores) and used several simplifying assumptions.
- 3. Loss of inventory between 1974 and 1992 because of resuspension and flushing of sediments. However, there is no evidence of significant sediment losses from the reservoir.

The CSU inventory estimates are reasonably close (54, 64, and 92 mCi) and should be used only as minimum estimates of ^{239,240}Pu in the Great Western Reservoir because of the question of whether the core samples were adequate enough to obtain complete plutonium profiles. The sediments near the dam appeared to be deeper than 1 m (3.3 ft), and the longest core tubes available were 0.96 m (3.1 ft) long. In some cases, 51-cm (20-in.) core tubes were used. To eliminate possible bias resulting from different tube lengths, only samples collected using the 96-cm (38-in.) core tubes were used in the summary statistics reported by <u>Schoep and Whicker</u> (1995).

<u>Schoep and Whicker</u> (1995) also estimated the percent of the observed sediment plutonium inventory contributed through aquatic pathways using

$$Pu(a) = \frac{Pu(r) - \left[\frac{Pu(s) \times Cs(r)}{Cs(s)}\right]}{Pu(r)}$$
(VII-3)

where

- Pu(a) = fraction of plutonium from aquatic inputs
- Pu(r) = plutonium in Great Western Reservoir sediments (kBq m⁻²)
- Pu(s) = plutonium in Great Western Reservoir watershed soil (kBq m⁻²)
- Cs(r) = cesium in Great Western Reservoir sediments (kBq m⁻²)
- Cs(s) = cesium in Great Western Reservoir watershed soil (kBq m⁻²).

Using the median reservoir-wide sediment to soil ratios (<u>Table VII-4</u>) and the observed reservoir-wide median activity per unit area of 2.99 kBq m⁻² (96-cm [38-in.] core median), Schoep and Whicker estimated that approximately 87% of the plutonium in the Great Western Reservoir was deposited through aquatic inputs. Walnut Creek was the likely primary source.

The 1992 CSU study indicates that 87% of the plutonium in the Great Western Reservoir originated from aquatic inputs from Walnut Creek. Walnut Creek drains the northern portion of the RFP, where the holding ponds are located.

MEASUREMENTS AT STANDLEY LAKE

Radioactive materials released from the RFP may have been transported to Standley Lake through surface water and airborne particles. Almost all the inflow to Standley Lake is delivered by canals that do not pass through the Rocky Flats site. Woman Creek, which drains the southern side the Rocky Flats site, flows intermittently into Standley Lake. Between 1952 and 1973, the RFP discharged water treatment facility filter backwash into Pond C-1, which discharged into Woman Creek. Standley Lake has been studied less extensively than the Great Western Reservoir. However, it has yielded some important insights on airborne deposition in the region.

In 1970 and 1973, EPA collected lake bottom sediment grab samples to determine ^{239,240}Pu concentrations in the sediment in Standley Lake (Appendix F, Figure F-1). The concentrations were predominantly at baseline levels attributable to global fallout sources of less than or equal to 0.1 pCi g⁻¹. The average concentration of surface grab samples (number of samples = 2) collected in 1970 was 0.13 pCi g⁻¹, with a maximum concentration of 0.21 pCi g⁻¹ (EPA 1971). The average concentration of surface grab samples (number of samples = 17) collected in 1973 was 0.07 pCi g⁻¹, with a maximum concentration of 0.17 pCi g⁻¹ (Lammering 1975). The maximum concentrations seen in cores were 0.37 and 0.11 pCi g⁻¹ in 1970 and 1973, respectively.

Great Western Reservoir and Surrounding Soil				
Pu(sediment):	Pu(soil):	Pu(sediment):	Cs(sediment):	
Cs(sediment)	Cs(soil)	Pu(soil)	Cs(soil)	
1.33	0.18	10.7	1.4	
^a Soil plutonium inventory = 0.26 kBq m ⁻² (<u>Webb</u> 1992) and soil cesium inventory = 1.6 kBq m ⁻²				
(<u>EG&G</u> 1990).				

Table VII-4. Sediment and Soil^a ^{239,240}Pu and ¹³⁷Cs Ratios for Great Western Reservoir and Surrounding Soil In 1974, PNL estimated the average 239,240 Pu surface sediment concentration to be 0.22 pCi g⁻¹ (number of samples = 8), with a maximum concentration of 0.3 pCi g⁻¹ (Thomas and Robertson 1981). A sedimentation rate of 2.5 cm (1 in.) per year was estimated for the eastern third of the lake, which is the deepest part of the lake. Based on the relative size of the lake compared to the Great Western Reservoir and the average 239,240 Pu concentration, a total inventory of 61 mCi of 239,240 Pu and 18 mCi of 241 Am in surface sediments was calculated.

These early studies all indicated average ^{239,240}Pu concentrations in surface sediments within or only slightly above the range observed in background lakes (<u>Table VII-2</u>). However, in 1976, <u>Hardy et al.</u> (1980) collected a long core from Standley Lake to determine the timing of

In a sediment core collected from Standley Lake by the DOE Health and Safety Laboratory in 1976, about 70% of the ^{239,240}Pu occurred in the uppermost 30 cm (12 in.), which was attributed to the release from the 903 Area around 1969. The remaining 30% was attributed to atmospheric fallout during extensive nuclear testing in the 1950s and early 1960s. plutonium releases. Using a combination of actinides, the history of deposition of selected radionuclides was determined (Figure VII-13). Mass spectroscopy was also used to determine ²³⁹Pu/²⁴⁰Pu atom ratios, which were then used to indicate the sources of the plutonium (i.e., nuclear weapons testing, nuclear-powered satellite failure, or releases from Rocky Flats).

A Rockwell study in 1984 represented the most extensive study of Standley Lake conducted to that date. Sixty-three grab samples and four core samples were collected. Some of the samples were collected by the City of Broomfield. The average surface sediment ^{239,240}Pu concentration was determined to be 0.04 pCi g⁻¹. The maximum concentration was 0.21 pCi g⁻¹. Sediment cores

showed fairly consistent concentrations, with a peak at deeper portions of the core. The cores were not dated, but it is presumed that this peak represents the 1969 atmospheric release from the 903 Area.

A comparison between 1984 Rockwell and 1992 RFI/RI USGS data indicated no statistical difference between the studies' estimates of mean surface sediment concentrations of ^{239,240}Pu and ²⁴¹Am. The mean ^{239,240}Pu and ²⁴¹Am concentrations were 0.05 and 0.02 pCi g⁻¹, respectively. Maximum concentrations were 0.19 and 0.10 pCi g⁻¹, respectively. The plutonium concentrations were well within background levels measured in sediments of other front range lakes and reservoirs (Table VII-2). Concentrations of ^{239,240}Pu in sediment cores collected in 1984 and 1992 are shown in Figures <u>VII-14</u> and <u>VII-15</u>. In general, the trends are similar, with single peaks obvious below 38 cm (15 in.). However, there were slight statistically significant differences in the matched pairs (Figures <u>VII-14</u> and <u>VII-15</u>). Clow and Johncox (1995) attributes this to spatial variation in sediment and ^{239,240}Pu deposition. The cores confirm earlier conclusions that ^{239,240}Pu concentrations in Standley Lake sediments primarily reflect airborne deposition of contaminants released from the 903 Area in 1969.



Figure VII-13. Depth distribution of ¹³⁷Cs, ^{239,240}Pu, ²³⁸Pu, and ²⁴¹Am in Standley Lake sediment core collected in 1976 (redrawn from <u>Hardy et al.</u> 1980).



Figure VII-14. Relation of concentrations of ^{239,240}Pu to depth at sediment sampling site SED08392 collected from Standley Lake in 1984 and 1992. A sedimentation rate of 3.0 cm (1.2 in.) per year was assumed for the 1983 data (from <u>Clow and Johncox</u> 1995).



Figure VII-15. Relation of concentrations of ^{239,240} Pu to depth at sediment sampling site SED08192 collected from Standley Lake in 1984 and 1992. A sedimentation rate of 3.8 cm (1.5 in.) per year was assumed for the 1983 data (from <u>Clow and Johncox</u> 1995).

MEASUREMENTS AT MOWER RESERVOIR

With the exception of a few samples collected by EPA in 1970, Mower Reservoir was not examined until 1992. Two grab samples collected in February 1970 were analyzed for ^{239,240}Pu, and they yielded results of 0.10 pCi g⁻¹ (on the west end) and 0.09 pCi g⁻¹ (on the east end of the lake). These results are very close to the baseline level of 0.1 pCi g⁻¹ accepted by EPA (EPA 1971). In September 1970, two more surface sediment samples were collected at the same locations. This time, both results were 0.18 pCi g⁻¹. No explanation for the increase was given.

In 1992, <u>Clow and Johncox</u> (1995) reported a mean ^{239,240}Pu concentration of 0.351 pCi g⁻¹ in Mower Reservoir. These concentrations are considerably higher than background levels shown in <u>Table VII-2</u>, as well as the concentrations of plutonium in sediments for Standley (see <u>Figure VII-7</u>). However, they noted that trends examined among Mower Reservoir, Standley Lake, and Great Western Reservoir are in agreement with the patterns of ^{239,240}Pu concentration reported for soil surrounding Rocky Flats. They further state that these findings are consistent with the fact that Mower Reservoir is closest to the drum storage site (903 Pad) at Rocky Flats.

SUMMARY

Data collected from sediment studies conducted from 1969 through 1992 appear to be useful to the historical dose reconstruction. The data associated with the most rigorous quality assurance programs are those collected most recently. However, the majority of earlier studies were documented well enough to determine sampling and analytical methods. In addition, most studies had at least some minimum amount of quality assurance data (such as the use of split samples, replicates, and spiked samples) to provide some confidence in the data. Moreover, the trends observed in early studies were confirmed in the later, more rigorous studies.

Data collected from the Great Western Reservoir and the holding ponds could be used to confirm a liquid effluent source term. Several estimates of total inventory have been made (Table VII-5). According to the CSU studies, the majority of the plutonium inventory in the reservoir is from liquid releases from Rocky Flats. <u>Schoep and Whicker</u> (1995) estimate that 87% of the plutonium in the Great Western Reservoir originated from aquatic inputs from Walnut Creek.

In addition, the historical studies provide evidence that known releases (such as the 1969 903 Area disturbance and the 1971–1973 holding pond reconstruction) are the predominant sources of plutonium in the sediments of the Great Western Reservoir. Current core profiles from the deepest regions of the impoundment still reflect the impact of these early releases (see Figure VII-16). The temporal trends seen in the surface sediments over the time period studied

^{239,240} Pu inventory	
(mCi)	Reference
244	Thomas and Robertson (1981)
54	Schoep and Whicker (1995)
62	(Note: Three different calculation methods were used. See the
92	text for descriptions)

Table VII-5. Estimates of ^{239,240}Pu in Great Western Reservoir



Figure VII-16. Core profiles from various locations across the Great Western Reservoir (from <u>DOE</u> 1995).

(1970–1992) also suggest that the holding pond reconstruction during 1972 may have had the biggest impact on plutonium concentrations in the Great Western Reservoir sediments after 1970 (Figure VII-17).

Past studies provide evidence that Standley Lake, and probably Mower Reservoir, were primarily impacted by airborne deposition of plutonium. Temporal changes in surface concentrations and core profiles help to confirm that airborne deposition from Rocky Flats, particularly from the 903 Area releases, have had the greatest impact on these impoundments. <u>Thomas and Robertson</u> (1981) estimated a total inventory of 61 mCi of ^{239,240}Pu in Standley Lake. However, this estimate was based on only eight core samples.

Finally, background lake data and historical evidence of fallout from nuclear weapons testing in sediment cores can be used to corroborate the timing of fallout ambient plutonium and cesium levels in the Rocky Flats environs.



Figure VII-17. Mean ^{239,240}Pu concentrations in surface sediments of Great Western Reservoir from 1970 through 1992.

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