CHAPTER VIII

SOIL MONITORING AND PENETRATING RADIATION SURVEYS

Soil represents a significant sink for plutonium and other long-lived radionuclides and metals. It has been estimated that 99.7% of the plutonium in the grassland ecosystem at the Rocky Flats Plant (RFP) is contained in the soil (Little et al. 1980). There is strong evidence that the majority of plutonium released to the environment of this area came from leaking barrels of plutonium-laden cutting oil, which were stored at the 903 Area.

Background plutonium is defined as plutonium in the environment from sources other than the RFP.

Numerous soil studies have been conducted around the RFP, most of which have focused on the spatial distribution of ^{239,240}Pu, which provides a fingerprint of airborne plutonium released from the Rocky Flats site. Table VIII-1 summaries the major soil research studies conducted to date at or near the RFP that are of interest to the reconstruction of historical public exposures. The studies are listed in the approximate order in which they were conducted. A more detailed account of these studies is given in Appendix H.

This chapter provides an overview of the soil data that are useful to the historical public exposures studies. Information is presented on the definition and determination of background concentrations of plutonium; early soil analyses; the fate of plutonium in soils; baseline suspension; and deposition patterns, inventories, and inventory comparisons. In addition, the results of penetrating radiation surveys are evaluated because they also reflect the extent of surface soil contamination. A discussion of beryllium in soil is also provided.

SOURCES OF BACKGROUND ENVIRONMENTAL PLUTONIUM

The evaluation of background levels of plutonium in the environment around the RFP is important to determine the impacts of historical releases from the plant. Essentially all plutonium in the environment is artificial, arising from activities of humans. We say essentially because there are extremely small amounts of plutonium formed in nature, though such amounts are negligible for our purposes. For our work, we define background plutonium to be plutonium in the environment from sources other than the RFP. The primary source of background plutonium in the Rocky Flats area is global fallout from the atmospheric testing of nuclear weapons. Of the alpha radiation-emitting isotopes of plutonium (the alpha-emitting isotopes are of most concern for this project), the primary constituents of this global fallout are ^{239}Pu and ^{240}Pu. A secondary source of background plutonium is global fallout from the 1964 atmospheric burnup of a satellite, which contributed about 1% of the total background (alpha-emitting) plutonium activity in the northern hemisphere. Plutonium from the satellite was essentially all ^{238}Pu. Regional fallout from surface and subsurface activities at the Nevada Test Site may also have contributed small amounts to the plutonium background. Appendix H provides more detailed information about these different sources of environmental plutonium.

The primary source of background plutonium around Rocky Flats is global fallout from atmospheric testing of nuclear weapons. A secondary source was the burnup of a satellite in 1964 that contributed about 1% to the total. Fallout from the Nevada Test Site area probably contributed minor amounts.

Table VIII-1. Summary of Some Important Soil Sampling Studies Reviewed

Agency (reference)[a]	# of samples and location	Depth (cm)	Analyses	Comments
RFP (Thackeray 1953; Hammond 1957a, 1957b; others)	Onsite and offsite locations	Not stated	Gross alpha	Results are less useful than from other studies because no plutonium results were obtained.
CCEI and **NCAR** (CCEI 1970; Poet and Martell 1972)	38 total, 18 within 10 km (6 mi) of RFP	1	^{239,240}Pu, ^{90}Sr	Presented first public proof of offsite contamination.
HASL (Krey and Hardy 1970)	33, from RFP vicinity up to 64 km (40 mi) downwind	to 20	^{238}Pu, ^{239,240}Pu	Study used most often in past source term estimates. Depth distribution studied at 8 of the 33 locations.
HASL (Krey and Krajewski 1972)	2, about 16 and 32 km (10 and 20 mi) from RFP	10	^{238}Pu, ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu	Used isotopic ratios to separate RFP and global fallout contributions.
HASL (Krey et al. 1976a)	26, east of RFP to Indiana St	10	^{239,240}Pu, ^{241}Am	Soil plutonium concentrations related to air sampler data.
HASL (Krey 1976)	19, downwind to Denver	10	^{239}Pu, ^{240}Pu	Identified source of plutonium (RFP vs. fallout) using isotopic ratios.
HASL (Krey et al. 1976b)	27, to 3 km (1.9 mi) downwind of 903 Area	20	^{239,240}Pu, ^{241}Am	Plutonium and americium studied with depth.
CSU (Little 1976)	10, from two plots near 903 Area	21	^{238}Pu, ^{239,240}Pu	Plutonium concentrations and soil particle size ranges measured in 3-cm (1.2-in.) segments of each deep sample.
CSU (Little and Whicker 1978)	15, transects near plots of Little (1976)	21	Plutonium	Plutonium concentrations and soil particle size ranges measured in 3-cm (1.2-in.) segments of each deep sample.
CDPHE (CDH 1977)	13, offsite up to 161 km (100 mi)	0.32	^{137}Cs, ^{239,240}Pu	Includes comparative evaluation of Dr. Johnson sweep method with CDH methods.
CDPHE (CDH 1990)	13 near and 8 remote	0.48; 0.64	^{239,240}Pu	Summary of 1970 to 1989 results.
CDPHE (Love 1994; Jones and Zhang 1994)	13 within 11 km (7 mi) and 9 remote	0.64	^{239,240}Pu	Summary of results for 1970–1991, with statistical comparison of datasets and contour plots.
CSU (Webb 1992)	10, up to 19 km (12 mi) from 903 Area	21	^{238}Pu, ^{239,240}Pu	Repeated earlier study of Little (1976) to determine changes.

Table VIII-1. Summary of Some Important Soil Sampling Studies Reviewed

Agency (reference)[a]	# of samples and location	Depth (cm)	Analyses	Comments
CSU (Webb 1996; Webb et al. 1997)	>1200, up to 19 km (12 mi) along 4 transects NE, E, SE, SSE from 903 Area	0.3, 3, and to 21	^{238}Pu, ^{239,240}Pu, ^{241}Am	Depth profiles, inventory estimation and 3-dimensional spatial model. Background samples from 10 locations along the front range.
CSU (Ibrahim et al. 1996)	13, within 4.8 km (3 mi.) along 2 transects E and SE of 903 Area	0.3, 3, and to 21	^{239,240}Pu, ^{241}Am	Depth profile samples and inventory estimates.
CSU (Ibrahim et al. 1997)	14, onsite and offsite up to 19 km (12 mi) east from 903 Area	0 to 3	^{239,240}Pu	Used isotopic ratios to distinguish RFP vs. fallout plutonium. Background samples along the front range.
DOE (Litaor 1993; Litaor et al. 1994; Litaor 1995a)	118 and 26 pits, <3 km (1.9 mi) from RFP	0.64, 0 to 96	^{239,240}Pu, ^{241}Am	Geostatistical analysis of surface soil sampling data. Depth profiles.
DOE (Litaor et al. 1995)	240 (CDH method); 167 (RFP method); <3 km (1.9 mi) from RFP	0.64 (CDH) 5 (RFP)	^{239,240}Pu	Geostatistical analysis of data. Also analysis of historical and exhaustive datasets.
DOE (Litaor and Allen 1996)	118 onsite, 60 offsite up to ~16 km (~10 mi)	0.64	^{241}Am	Geostatistical analysis to determine spatial distribution of RFP ^{241}Am.
DOE (Litaor et al. 1996, 1998; Litaor and Ibrahim 1996)	5 pits, ~250 m (~820 ft) east of 903 Area		^{239,240}Pu, ^{241}Am	Investigated vertical distribution and transport processes for actinides. Analysis of plutonium associated with different soil phases.
DOE (Litaor 1999)	61 offsite surface plots and 11 pits	0 to 100	^{239,240}Pu	Spatial and vertical distribution of plutonium in soils and an inventory estimate.
DOE (EG&G 1995)	50 remote, 20 offsite samples	5	Fallout, metals, organics	Extensive study of background soils, including soil classification.
CESC (CESC 1996)	32, offsite, <1.6 to > 64 km) (<1 to >40 mi)	3 and 21	^{238}Pu, ^{239}Pu ^{241}Am, ^{235}U, ^{238}U, ^{137}Cs, ^{90}Sr	Some locations not previously sampled.

[a] CCEI = Colorado Committee for Environmental Information, CDPHE = Colorado Department of Public Health and Environment, CESC = Citizens Environmental Sampling Committee, CSU = Colorado State University, DOE = U.S. Department of Energy, HASL = Health and Safety Laboratory of the U.S. Atomic Energy Commission, NCAR = National Center for Atmospheric Research, RFP = Rocky Flats Plant.

MEASUREMENT OF PLUTONIUM

The most common analytical technique used for plutonium analysis is alpha spectroscopy, a procedure that segregates alpha particle radiations by energy. This allows the differentiation of ^{239,240}Pu from ^{238}Pu. However, because the alpha energies of ^{239}Pu and ^{240}Pu are quite similar, these two isotopes cannot be determined separately with alpha spectroscopy. In essentially all cases, values reported as ^{239}Pu are actually measurements of ^{239,240}Pu, unless it is specifically noted that isotopic measurements or other isotopic information are available. Mass spectrometry can be used to differentiate ^{239}Pu from ^{240}Pu, although it is not as readily applied to field samples as alpha spectroscopy. Alpha spectroscopy results are generally reported in radiological units, while mass spectrometry results are usually reported in mass units or as atom ratios relative to ^{239}Pu.

To prepare a sample for alpha spectroscopy counting, plutonium must first be extracted from the sample media. This is usually performed with concentrated acid, or filter samples may be wet ashed. Chemical separations are then performed, and the plutonium is plated out on a probe (or planchet) for counting in an alpha spectrometer. Plutonium-236 or ^{242}Pu can be added as a tracer at the start of sample preparation to help measure the chemical recovery (Krey and Hardy 1970; Holleman et al. 1987).

Other analytical details that may be important in evaluations of environmental plutonium measurements include physical sample preparation, such as sieving or grinding; fusion of samples versus acid leaching of samples to extract the plutonium; chemical separation techniques used to separate plutonium from other elements in the sample; interferences in the analysis; detection limits of the analytical procedure; and quality control and assurance procedures used in the analyses. Holleman et al. (1987) provides a summary of analytical techniques commonly used for plutonium analyses in environmental samples.

Some important characteristics related to plutonium in soils should be considered in evaluations of soil sample results around the RFP. As noted above, the measurement technique and, more specifically, the plutonium isotopes actually measured should be determined. If isotopic results, such as the ratio of ^{240}Pu/^{239}Pu, are available, it may be feasible to determine more accurately whether the source of the plutonium is truly background or if it has been influenced by RFP sources (see Appendix H). When comparing samples near the RFP to background samples, the time of sample collection can be important because temporal trends exist in the global fallout of plutonium from nuclear weapons testing (see Appendix H). Finally, depth distributions of plutonium should be considered relative to the goals of a particular sampling program or analysis. Soil samples taken from the surface soils (e.g., to approximately 1 cm [0.4 in.]) are generally not representative of the total deposition of plutonium that exists in the soil column. Quantitative comparisons between results of sampling programs with widely disparate sample depths should be performed when information is available to develop relationships between soil layers of different depths.

DEPTH DISTRIBUTION OF PLUTONIUM IN SOILS

Plutonium that is deposited on ground and vegetation surfaces will gradually weather into the soil. Many of the background soil samples analyzed for plutonium content were taken from surface soils of 0–1-cm (0–0.4-in.) depth or less or from relatively thick samples of 0–20-cm (0–

8-in.) or 0–10-cm (0–4-in.) depth. It is important to consider the depth distribution of plutonium in soils when samples of such different depths are evaluated.

A few historical studies around the RFP have investigated the depth distribution of plutonium in soils. Krey and Hardy (1970) investigated the depth distribution at eight sampling locations around the RFP, from just next to the plant to distances up to about 64 km (40 mi) from the site. Based on these samples, between 9 and 61% of the ^{239,240}Pu from the surface to 20 cm (8 in.) deep was below 5 cm (2 in.) deep. Two other sampling locations, in New York City and Waynesville, Ohio, showed results within this same range. From limited sampling at finer depth resolution, it was concluded that less than 1% of the plutonium occurred below 13 cm (5 in.).

Whicker et al. (1974) performed preliminary investigations of the inventories of ^{239,240}Pu in several compartments of the terrestrial ecosystem near the RFP. Two locations were studied: a contaminated area very close to the former oil barrel storage pad (the 903 Pad) and a relatively less contaminated area (the control) south of the main plant (though not representative of background plutonium). Results from this study indicated that, for both the contaminated study area and the control area, about 40% of the plutonium in soil occurred below 3 cm (1.2 in.) deep.

Little and Whicker (1978) performed more detailed investigations of the depth distributions of plutonium in soil near the RFP (as well as particle size distributions). This work used the same sampling areas used earlier by Whicker et al. (1974). Samples were taken in 3-cm (1-in.) thick slices to a depth of 2 1 cm (8 in.). Four replicate samples from each layer were obtained from 10 locations in the contaminated area and from 5 locations in the control area. We used the average concentrations reported by Little and Whicker, for the less than 2-mm (0.08-in.) size particles, to calculate the fractional contributions of each depth interval to the total plutonium content. For these calculations, we assumed a constant soil density for all layers of soil. Results are shown in Table VIII-2. Because the soil densities for each sample layer were not available, the calculated relative plutonium activities are not exact. (Also, because of rounding, the percentages do not add to exactly 100%.) Qualitatively, it is clear that although much of the plutonium activity occurs in the top 3 cm (1.2 in.) of soil, layers to as deep as 21 cm (8 in.) are significant contributors to the total soil column plutonium. The ^{239,240}Pu inventory in the top 3 cm of soil for Little and Whicker's data is reported as 72% in Webb (1996).

Table VIII-2. Estimated Depth Distributions of ^{239,240}Pu in Soils from Two Areas Near the Rocky Flats Plant (Little and Whicker 1978)

Area	Average relative plutonium activity in each sample layer (percent of total)						
	0–3 cm	3–6 cm	6–9 cm	9–12 cm	12–15 cm	15–18 cm	18–21 cm
Contaminated	58	11	18	4.6	5.8	2.3	0.82
Control	70	6.1	3.4	3.2	4.4	7.1	6.1

Webb et al. (1993) presents the results of a 1989 study of the contaminated area previously documented in Whicker et al. (1974). Webb et al. (1993) also presented additional results from sampling in 1972–1974. The inventory of ^{239,240}Pu was measured for soil layers 0–3 cm (0–1.2 in.) and 3–21 cm (1.2–8 in.). It was concluded that for both periods, about 50% of the soil inventory was in each of these layers. However, the soil concentration in the 0–3-cm (0–1.2-in.) layer was significantly less in 1989 than in 1972–1974. To account for this, Webb et al. made the preliminary speculation that about 2.75 cm (1.08 in.) of the surface soil had been lost from the study area by soil erosion over the 15 years between the two studies.

The studies described here resulted in differing estimates of plutonium depth distributions in the soil column. However, it is clear from the results that significant quantities of plutonium have migrated to depths of at least 10–20 cm (4–8 in.). Whenever possible, these results should be considered in the evaluations of soil plutonium concentration measurements around the RFP. In particular, we note that it is generally not reasonable to use measurements of the surface soil concentrations of plutonium (to depths of 1 cm [0.4 in.] or so) to estimate the total inventory (or total deposition) of plutonium in the soil. In some cases, information may be available to develop relationships between soil layers of different depths, which allow careful, quantitative comparisons (for example, this has been done in Jones and Zhang [1994]). This difficulty is the reason that we do not try to directly compare background surface soil plutonium concentrations (expressed as becquerels per kilogram [Bq kg^{-1}] or similar units) to background soil plutonium inventories (expressed in becquerels per square meter [Bq m^{-2}] or similar units).

BACKGROUND PLUTONIUM CONCENTRATIONS IN SOILS NEAR THE ROCKY FLATS PLANT

Two types of results for plutonium in soils have been reported: (1) concentrations of plutonium per unit mass of soil (mass concentrations), with units such as becquerels per kilogram or picocuries per gram (pCi g^{-1}) and (2) total quantity of plutonium that has been deposited on and remains in the soil, per unit soil surface area (deposition or inventory), with units such as becquerels per square meter or millicuries per square kilometer (mCi km^{-2}). As indicated above, it is generally inadvisable to try to convert results between mass concentrations and values of total deposition without knowledge of the depth distribution of plutonium.

Measurements performed by a variety of organizations including National Center for Atmospheric Research (NCAR); Colorado Department of Public Health and Environment (CDPHE) (formerly Colorado Department of Health [CDH]); Colorado State University (CSU); and EG&G are summarized in Appendix H. These studies indicate that the background mass concentration of ^{239,240}Pu in surface soils (0.16 to 5-cm [0.06 to 2-in.] depths) of eastern Colorado is in the range of about 0.3–4.5 Bq kg^{-1} (0.008–0.1 pCi g^{-1}), although only one value was greater than 3.3 Bq kg^{-1} (0.09 pCi g^{-1}). The wide variability in results may be due to the very shallow surface layers of soil that were sampled and the spatial patterns of fallout deposition across the large area covered by sampling.

Average background concentrations of plutonium in soils around Rocky Flats were 0.038 and 0.058 pCi g^{-1} (2.14 and 1.40 Bq kg^{-1}) in two recent studies.

Deposition measurements and calculations performed by the Health and Safety Laboratory of the U.S. Atomic Energy Commission (HASL) (and the analysis of HASL results in Seed et al. [1971]) and by CSU indicated that the total deposition of ^{239,240}Pu from global fallout, in the general area around the RFP and along the front range, was probably in the range of 40–120 Bq m^{-2} (1.1–3.2 mCi km^{-2}). This range is within that seen for other states in the 37.5–42.5 °N latitude range (10–260 Bq m^{-2} or 0.27–7 mCi km^{-2}), although RFP deposition values tend to be at the higher end of measured concentrations ranges (see Appendix H for further details).

Two studies have been identified that provide a clearer definition of background levels of plutonium around the Rocky Flats environs. The first is a project conducted by CSU from 1992 to 1994 that measured soil plutonium concentrations at 10 locations along the front range of the Rocky Mountains, from north of Fort Collins, south to Colorado Springs (Webb 1996). The

second is a study by EG&G Rocky Flats called the Background Soils Characterization Program (EG&G 1995). The EG&G study sampled at 50 locations along the Colorado front range that were considered unaffected by Rocky Flats releases; these were referred to as remote locations. The EG&G study also sampled at 20 locations in an area just north of the RFP, which has soils similar to those found at Rocky Flats (EG&G 1995). The EG&G study provides extensive data on the geology, soil units, and soil taxonomy of the region and data on fallout radionuclides (including plutonium), naturally occurring radionuclides, metals, and organic compounds.

These studies add considerably to the knowledge about background concentrations of plutonium in soils and, thus, can be used to determine the extent of contamination from deposition of Rocky Flats airborne effluents. Table VIII-3 provides summary information on ^{239,240}Pu concentrations in background soils. The CSU and EG&G results are in good agreement considering the slightly different sampling depths that were used.

Table VIII-3. Plutonium-239,240 Background Concentrations (pCi g^{-1}) [a] in Soils Considered Unaffected by Rocky Flats

Study [b]	Number of samples	Mean	Standard deviation
CSU	10	0.058	0.021
EG&G	20	0.038	0.014

[a] To convert the units to Bq kg^{-1}, multiply the value by 37.

[b] CSU: Webb et al. (1993). Soil samples taken to a depth of 3 cm (1.2 in.). EG&G (1995). Soil samples taken to a depth of 5 cm (2 in.).

OTHER ALPHA ACTIVITY CONCENTRATIONS IN SOIL NEAR ROCKY FLATS

Early environmental measurements of radioactivity in soil around Rocky Flats were reported as total alpha activity. Background concentrations of alpha-emitting radionuclides in soils around Rocky Flats (including uranium, thorium, radium, and daughter products) have been reported (Richmond 1970). Radiochemical results were presented for natural alpha activity in seven soil samples (5-cm [2-in.] depth), including four within 3.2 km (2 mi) of Rocky Flats, two from Denver and one from 72 km (45 mi) east of Rocky Flats. Total uranium concentration was given as 1.9 $\pm$ 0.4 pCi g^{-1} (14.8 Bq kg^{-1}) and ^{238}U was 1.0 $\pm$ 0.2 pCi g^{-1} (37 $\pm$ 7.4 Bq kg^{-1}). Total uranium, thorium, and radium concentration in soil averaged 8.0 pCi g^{-1}(14.8 Bq kg^{-1}). Including daughter products[a] produces a total natural alpha activity content of soils at the sampled areas of about 20 pCi g^{-1} (740 Bq kg^{-1}).

Litaor (1995b) described soil distribution of uranium at Rocky Flats. Highest values found were in areas affected by Rocky Flats operations. For ^{234}U, this range was 0.70 to 2.51 pCi g^{-1} (25.9 to 92.9 Bq kg^{-1}) and the median was 1.20 pCi g^{-1} (44.4 Bq kg^{-1}). For ^{235}U, the range was 0.0027 to 0.68 pCi g^{-1} (0.1 to 25.2 Bq kg^{-1}) and the median was 0.049 pCi g^{-1} (1.8 Bq kg^{-1}). The range in ^{238}U concentration was 0.83 to 7.72 pCi g^{-1} (30.7 to 286 Bq kg^{-1}) with a median of 1.20 pCi g^{-1} (44.4 Bq

a The activity of ^{232}Th parent was multiplied by six to include the other alpha-emitting daughters in secular equilibrium. The ^{226}Ra activity was multiplied by five to include ^{222}Rn, ^{218}Po, ^{214}Po, and ^{210}Po (Richmond 1970).

kg^{-1}). Litaor referred to an unpublished study that was conducted west of Rocky Flats along Rock Creek in an area thought to be unaffected by Rocky Flats activity. Measured concentrations of ^{238}U were 1.12 ± 0.20 pCi g^{-1} (41.4 ± 7.4 Bq kg^{-1}), ^{235}U were 0.05 ± 0.04 pCi g^{-1} (1.9 ± 1.5 Bq kg^{-1}), and ^{234}U were 1.13 ± 0.17 pCi g^{-1} (41.8 ± 6.3 Bq kg^{-1}).

In summary, the background concentration of ^{238}U in surface soil of the RFP area is around 1 pCi g^{-1} (37 Bq kg^{-1}), with a total uranium concentration about twice that. Total long-lived alpha content is about 8 pCi g^{-1} (296 Bq kg^{-1}), and the total natural alpha content (with short-lived daughter products included) of soils is about 20 pCi g^{-1} (740 Bq kg^{-1}). This natural alpha background is the main reason why early Rocky Flats monitoring of total alpha activity in soils was not a sensitive indication of plutonium releases from Rocky Flats operations.

EARLY SOIL ANALYSES

Few data exist that address radionuclides in soil at and near the RFP during the very early years of operation. What information has been uncovered was reported by the Site Survey Group of Dow Chemical Company and refers to total alpha activity in soils from scattered locations.

The first survey was conducted during August and October 1952 (Thackeray 1953) to demonstrate alpha activity trends so that any subsequent plant-caused contamination could be detected. As part of a designed sampling program (also including water, vegetation, and algae), results of soil samples taken from 18 locations on the immediate plant site (twice from each location) were reported. These soil data, however, were mingled with data from 55 mud samples that were also collected on the plant site at about the same time. The average activity concentration of the total of 91 soil and mud samples was reported as 30,022 dpm kg^{-1}.

Early results of radioactivity in soils were expressed in dpm per kilogram (kg). The abbreviation "dpm" stands for *disintegrations per minute*, a rate of radioactive decay. There are 2.22 dpm per picocurie.

Individual values were not given, but a range of from 10,300 to 64,200 dpm kg^{-1} was given, with one hot mud sample of 109,000 dpm kg^{-1} activity taken from the bank of Woman Creek in the southwest corner of the plant site (a sample taken 23 m [25 yd] from the hot sample indicated 48,000 dpm kg^{-1}). The Site Survey Group concluded that they could roughly establish the statistical distribution of activity concentration, but they did not know laboratory analytical error. The Site Survey Group stated that samples from dry beds of runoff streams (mean of 27 samples = 37,100 dpm kg^{-1}) were statistically higher than those from regular soil sampling locations (mean of 36 samples = 27,000 dpm kg^{-1}). They concluded that the utility of soil samples for the detection of plant-caused airborne contamination was strictly limited. The limits of detection and the large data spread indicated that many soil samples from the same universe would need to increase by about 5000 dpm kg^{-1} before any change above background could be detected.

The Site Survey Group later reported total alpha results of soil analyses in the September and November 1957 monthly progress reports. This work closely followed upon the September 11, 1957, fire in 71 Building. The narrative of the September report (Hammond 1957a) stated that offsite soil and vegetation samples indicated possible low-level contamination from the fire incident. This tentative conclusion may have been reached primarily from observation of vegetation data. For soils, however, there was no 1952 offsite background with which to compare the 1957 postfire data, and the 1952 onsite background values tended to exceed the 1957 postfire values for offsite collections. Statistical summaries were not given, but pulse-height analyses of 8 offsite soil

samples lead to a conclusion of possible plutonium peaks in three samples. The November report (Hammond 1957b) provided no narrative analysis of soils data, but it did list total alpha activity for eight offsite locations apparently close to watercourses or water bodies.

FATE OF PLUTONIUM IN SOILS AROUND ROCKY FLATS

Information on the fate and transport of plutonium in soil is needed to provide input for the environmental transport models used in the historical public exposures studies. Much research has been performed that elucidates the fate of plutonium in soil. Studies of plutonium in soil profiles and in air by the HASL (Krey et al. 1976b; Volchok et al. 1977) and EPA (1973) indicate decreasing availability of plutonium in surface soils for resuspension and, thus, lower airborne concentrations of plutonium with time. This has been attributed by several authors to stabilization of plutonium in the surface soil. Thus, it is possible that much of the released plutonium may still remain in the soil; however, it may have been transported further down in the soil column. Evidence for the latter supposition is mixed.

Based on studies of plutonium particle size distribution and soil profiles of plutonium near the 903 Area (Figure VIII-1), CSU researchers theorized that the following scenario occurred (Little and Whicker 1978): (1) the plutonium contaminant was in the form of an oxide, (2) the plutonium oxide became attached to soil particles, (3) gusty winds, combined with periods of surface disturbance, heterogeneously redistributed the particles to the east and southeast of the 903 Area, and (4) the soil-plutonium particles were eventually broken down by weathering and were dispersed laterally and downward in the soil profile. Webb (1992), on the other hand, suggested that some of the initial plutonium may have been deposited as a more soluble chloride form, allowing it to penetrate the soil rapidly. This chloride form would have bound to the soil as it oxidized. More recent work in Higley (1994), which studied the movement of actinides in soils taken from a natural grassland site at Rocky Flats, suggests that a more soluble form of plutonium is not required to explain the rapid penetration down the soil profile. Physical processes, in particular, cracks formed in the soil as a result of freezing, result in significant migration of clay particles down the soil column. Radionuclide migration was largely attributed to bypass flow through macropores created during soil shrinkage from drying. Work by Litaor et al. (1994, 1996) supported this conclusion and also pointed to earthworm activity. Higley (1994) also attributed less extensive but significant radionuclide migration to volume changes caused by frost heaving and subsequent soil contraction upon thawing. Higley (1994) determined that the highest plutonium activity concentration was associated with the colloidal particle size fraction (<0.45 µm), followed by the clay particle size fraction (0.45–2 µm). However, approximately 50% of the total inventory resided in the coarse silt fraction (10–53 µm), reflecting the abundance of this size fraction in the loam soil. Higley found no evidence that contaminated soil particles of different sizes migrated at different rates in the soil.

Recently, Hulse et al. (1999) demonstrated that ^{137}Cs serves as a good tracer for plutonium in soils. Both cesium and plutonium are very strongly sorbed to small soil and clay particles. The ^{137}Cs profile in soil is also very useful as it provides evidence of disturbances, such as plowing, in the soils.

Another important observation is that soil samples collected within 2 to 4 years of the original deposition from the 903 Area revealed an exponentially decreasing concentration of plutonium with increasing depth in soil (Krey et al. 1976b; Little 1976). Although some plutonium was detected as deep as 20 cm (8 in.) in the soil profile, approximately 50% of the

inventory was found in the top 3 cm (1.2 in.) of soil and greater than 90% was in the top 12 cm (5 in.). A comparison of plutonium concentrations measured in the same plot near the 903 Area in 1989 showed the same, statistically indistinguishable, gradient of plutonium versus depth (Webb 1992). It was concluded that 50% of the total plutonium was in the top 3 cm (1.2 in.) of soil, and concentrations decreased rapidly with depth (Webb et al. 1993). However, the soil concentration in the 0–3-cm (0–1.2-in.) layer was significantly less in 1989 than in 1972–1974. To account for this, Webb et al. speculated that about 2.75 cm (1.08 in.) of the surface soil had been lost from the study area by soil erosion over the 15 years between the two studies.

Figure VIII-1. Aerial view of Rocky Flats, looking to the northwest. The 903 Pad, 903 Lip Area, Pad Field, and East Field are noted.

Based on a study of plutonium particle frequencies and sizes near the 903 Area, McDowell and Whicker (1978) hypothesized that the heterogeneity in the spatial distribution of plutonium particles in soil may partially explain observed plutonium concentrations in the area. They thought that weathering may have broken down particles attached to plutonium with the larger particles remaining comparatively unmoved and smaller particles subject to resuspension.

A study conducted in 1975 (Hayden et al. 1994) on particle sizes of plutonium in surface soil in the RFP buffer zone east of the plant indicated that larger particle sizes are associated with releases from the 903 Area and smaller particle sizes (mean diameter of 0.08 micron) with stack releases. This work was done with in situ autoradiographs (alpha track stars) by using cellulose nitrate film. The larger discrete plutonium particles were found directly east of the plant (and west of Indiana Street), with little spread to the northeast or southeast. It was assumed that the source material was pure PuO_2. Control films were placed at locations in Arvada, Boulder, Dillon, and 1.6 km (1 mi) northwest of the RFP. Star tracks were not observed on the control

films. Hayden did no further investigation of offsite areas and did not provide a quantitative analysis of any concentration trend with distance east from the 903 Area.

The Resource Conservation and Recovery Act Feasibility Investigation/Remedial Investigation (RFI/RI) for Operable Unit 2 (OU2) studied plutonium transport pathways in soil near the 903 Area. In this study, 26 pits were dug and investigated, mostly northeast to southeast from the 903 Area (DOE 1995). The vertical distributions of ^{239,240}Pu and ^{241}Am were evaluated. The study also investigated the mechanisms of vertical and lateral redistribution of plutonium and americium in the soil, as well as physical and chemical parameters that may be important to understanding transport in soil. Results of this study indicate that 90% of plutonium was in the upper 12 cm (5 in.) of the soil column. It was also concluded that biological activities, such as those of earthworms and burrowing animals, were a significant factor in the vertical transport of plutonium and americium in the soil column. Vertical mobility may be followed by increased plutonium transport because of surface flow and wind erosion as hypothesized following a CSU study (Webb et al. 1993). Webb (1996) stated that plutonium appears to be bound tightly to the soil, under normal oxygenated conditions, and is not subject to transport other than with the soil.

Soil studies conducted near the 903 Area showed that 50% of the plutonium in a vertical column of soil was in the top 3 cm (1.2 in.) and 90% was in the top 12 cm (5 in.).

A study of the potential mobility of plutonium, americium, and uranium in soils in 11 trenches excavated east of Indiana Street for the RFI/RI for OU3 (offsite) has provided indirect evidence of the fate and transport of these radionuclides in soils (DOE 1996a). Undisturbed soil samples were collected from a vertical profile to a depth of approximately 100 cm (40 in.). The pit studies included measuring soil characteristics and examining pedogenic processes on the vertical distribution of plutonium and americium. Similar to the results of the CSU studies, the highest activities of plutonium and americium were measured in the top 3 cm (1.2 in.) of soil. The detected activities below 10 cm (4 in.) are similar to the upperbound background activities selected for the RFI/RI study (0.04 pCi g^{-1} for plutonium and 0.09 pCi g^{-1} for americium).

Rain simulation tests were conducted during 1993 (Litaor et al. 1996). Actinide transport through the soil to groundwater was studied in five excavated pits located about 250 m (270 yd) to the east of the 903 Area. Controlled rain simulations coupled with measurements of volume flux and actinide activities provided information about fate and transport of plutonium and americium. Actinide activities were not correlated with rainfall intensity characteristics. Approximately 90% of the actinide activity was observed in the upper 12 cm (5 in.) of soil. However, one pit showed appreciable plutonium and americium activities to depths of 36 and 48 cm (14 and 19 in.), respectively. This observation was explained by the influence of preferential flow, large hydraulic conductivity, the location of the pit on a steep slope, and the presence of very coarse textured soil. The authors concluded that this is limited support for theories of continued downward transport of plutonium and americium. A transport mechanism involving discrete plutonium oxide particles, coupled with macropore channeling was proposed by the authors. Litaor believes this phenomenon could be implicated in the movement of plutonium on the soil surface during the wet spring and summer of 1995 (Litaor 1996).

Litaor and Ibrahim (1996) also addressed the geochemical behavior of plutonium that affects the transport of plutonium from the soil to groundwater. In the surface horizons, plutonium was determined to be primarily associated with organic carbon, sesquioxides, and a residual fraction. They suggested that the wet 1995 spring and summer may have led to a partial

dissolution of sesquioxides, followed by a desorption of plutonium and increased plutonium mobility. In a letter to the editor, Marty et al. (1997) disagreed with their conclusion, stating that the results showed the plutonium was not bound to either of these phases (organic carbon or sesquioxides). Marty et al. (1997) considered that most of the plutonium in Rocky Flats soils must be at or near the surface of soil particles or in discrete particles in the soil where it can readily contact solutions. This is consistent with earlier studies that show a portion of the plutonium in Rocky Flats soils occurs as discrete PuO_2 particles (McDowell and Whicker 1978).

Litaor et al. (1998) went on to examine the fractionation of ^{239,240}Pu and ^{241}Am to different particle sizes in soil interstitial water in a series of large-scale rain simulation experiments conducted in five pits located within 250 m (820 ft) of the 903 Area. Most of the radionuclides (83–97%) were associated with particles larger than 0.45 µm, regardless of the level of activity and the depth of sampling. Therefore, Litaor et al. (1998) concluded that plutonium and americium have very limited mobility in soils at RFP because the movement of suspended particles in soil is highly dependent on the spatial arrangement and continuity of macropores in the soil.

Resuspension from Contaminated Soils

The largest wind suspension events that led to plutonium releases from the 903 Area are evaluated in this study as individual pulses or discrete events (Weber et al. 1999). In addition, under routine meteorological conditions, wind-driven suspension of particles occurred on a relatively constant basis but at a lower magnitude than the largest pulses of suspended material (referred to as baseline suspension in Weber et al. 1999). After the asphalt pad was placed on the former oil barrel storage site, remaining contaminated soils continued to contribute to baseline suspension. Some data exist on estimates of plutonium that remained in soils near the 903 Area.

During 1973, a survey using a field instrument for detecting low energy radiation (FIDLER) was conducted in the proximity of the 903 Area and especially on an area just adjacent southeast of the 903 Pad (asphalt) and within the security fence (Krey and Hardy 1973). The FIDLER was adjusted to detect the 60 keV gamma ray from ^{241}Am (the daughter of ^{241}Pu); thus, by assumption, the count rate reading is directly related to the quantity of ^{239}Pu present. Background readings on the FIDLER were about 500. Several qualitative summary statements were made:

1. The 903 Pad and the surrounding plateau to the perimeter road is fill and shows a homogeneous contamination at relatively low levels
2. Highest contamination levels with sharp gradients exist southeast of the 903 Pad on the slope leading to the perimeter road (See area called “903 Lip in Figure VIII-1).
3. Disturbed areas, like the roads and the ground around the gas storage building, show relatively low contamination levels
4. Readings east of the perimeter road range from 400 to 2500 and generally decrease in an easterly direction
5. The highest contamination east of the perimeter road occurs on the plateau just south of the east access road and about 46 m (150 ft) from the perimeter fence.

The work just southeast of the 903 Pad produced an isorad map (Figure VIII-2) (Dow 1974) of ^{241}Am concentrations on an approximately 1-ha (2.5-acre) area. In 1976 and 1978, a soil removal project was conducted in the vicinity of the above-mentioned FIDLER survey, an area by then referred to as the 903 Lip Area (Barker 1982). As a result of the FIDLER survey, it was

believed that 0.56 Ci of plutonium had been deposited at that site. Contaminated soil was first removed in 1976 with hand tools and in 1978 with machines, until the excavated area showed contamination levels below the detection limits of the FIDLER (250 counts per minute). Before removal, the soil was moistened for dust suppression, and work was not conducted if criteria of minimal wind speed, visible dust, and levels of total long-lived alpha activity in portable ambient air samplers were not met. The excavated areas were covered with fresh soil and seeded with grasses. The excavated soil was shipped to the Nevada Test Site. The final estimate provided a value of 0.5 Ci of plutonium removed from the 903 Lip Area, based upon an average concentration of 1200 dpm g^{-1} (540 pCi g^{-1}) of plutonium in removed soil and a soil density of 1 g cm^{-3}.

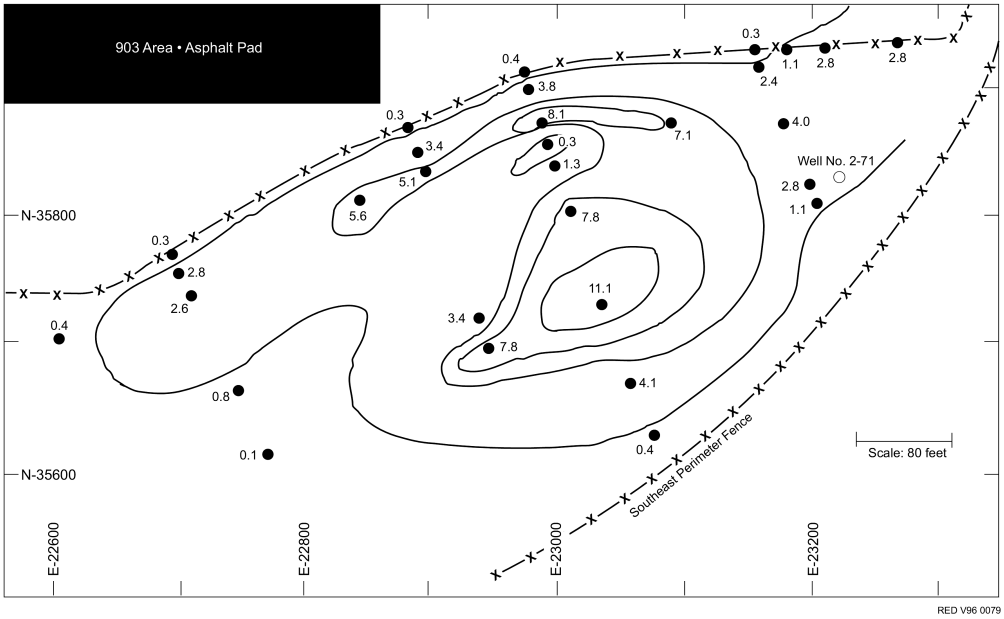

Figure VIII-2. Americium concentrations (in microcuries per square meter) detected by the FIDLER survey in 1973. To obtain approximate plutonium concentrations in microcuries per square meter, multiply values in plot by 10 (Dow 1974).

Langer (1984) described a wind-erosion study of the Pad Field and the East Field (Figure VIII-1). Soil in those areas contained above-background amounts of plutonium and americium. Both of these areas are close to the 903 Area, received windblown contaminated dust during the years of oil barrel leakage, and were themselves susceptible to secondary resuspension in the years following the placement of the 903 Pad. The Pad Field, which during pad installation had been covered with 20 cm (8 in.) of gravely soil from offsite and seeded with tall grasses, nevertheless retains an unspecified amount of surface contamination. A ground gamma survey indicated that approximately 1.2 Ci of plutonium exists in this Pad Field area (Langer 1991). The original soil surface in the East Field area has been left intact, and the same gamma survey indicated 0.67 Ci of plutonium in the East Field. The two fields are separated by the security fence, two dirt roads that run adjacent to the security fence on either side, and a berm between the roads. Hodgin (1984) described a resuspension study begun during fall 1981 and completed in 1982. Hodgin concluded that the berm was responsible for more than one-half of the elevated ^{239}Pu concentrations observed at the S–7, S–

8, and S–9 ambient air samplers. He recommended permanent suppression of wind erosion resuspension from the berm to reduce atmospheric plutonium levels at the site.

From the above, the resultant estimate of plutonium that remained in soil near the oil barrel storage area after placement of the 903 Pad was about 2.43 Ci (89.9 GBq) from 1970 to 1976 and about 1.93 Ci (71.4 GBq) from 1978 on.

DEPOSITION PATTERNS, INVENTORIES, AND INVENTORY COMPARISONS

Most of the plutonium contamination of soils around Rocky Flats probably originated from scraping (road grader work) and wind-driven suspension of contaminated soils in the 903 Area in 1969. Thus, the early soil monitoring studies around the RFP before 1969 probably do not provide any useful information on the primary plutonium release events or contamination presently seen in offsite soils. Moreover, the earlier studies did not include plutonium-specific analyses; rather, gross measurements of alpha-emitting radionuclides (gross alpha) were made. From reviews of these early studies (for example, Thackeray 1953; Hammond 1957a, 1957b), gross alpha concentrations are not greater in onsite soil than in offsite soil. Thus, these results are not useful for reconstructing public exposures, even for establishing ambient conditions.

Inventory—the total amount of plutonium—is obtained by multiplying the plutonium concentration in soil by the total amount of contaminated soil.

Soil sampling around the RFP was expanded and intensified following the 1969 fire. Since 1973, the RFP contractor has collected over 50 samples annually from alternate 152-m (500-ft) sections of a grid in the plant's buffer zone. The soil analysis program through 1977 also included the annual collection of samples from 18 degrees of arc on three circles having 1.6-, 3.2-, and 8-km (1-, 2-, and 5-mi) radii concentric with the center of the plant. The data are reported annually in the environmental monitoring reports.

In addition to the RFP-contractor sampling, the CDH has conducted surface soil surveys in the vicinity of the RFP since 1970 (CDH 1990). Composite samples are taken from 13 sectors, within about 11 km (7 mi) around the site. Surveys conducted from 1970 to 1991 are summarized and evaluated in Love (1994).

Soil Studies Resulting in Deposition Patterns and Inventory Estimates

Since 1969, there have been several efforts to produce total inventory estimates and to assess the spatial distribution of plutonium released from the 903 Area at Rocky Flats. The early works of Krey and Hardy (1970), Krey and Krajewski (1972), Krey (1976), Seed et al. (1971), and Poet and Martell (1972) estimated total releases of plutonium to the environment. These were based on relatively few samples as compared to the later efforts of Webb (1996). Webb's data, while more numerous, were collected for a more refined goal.

Krey and Hardy (1970) sampled 33 sites extending up to 64 km (40 mi) east, south, and north of Rocky Flats to establish the first ^{239,240}Pu deposition contour map around the RFP (Figure VIII-3). Of the 33 sites, only 18 locations were within a 16-km (10-mi) radius of the site. The small number of soil samples (52) resulted in significant extrapolation over large unsampled areas and relied heavily on individual data points that may have not been representative of the area. Krey and Hardy (1970) estimated the total plutonium inventory released offsite from RFP

was 251 GBq (6.8 Ci). This was based on 133 GBq (3.6 Ci) total plutonium within the 111 MBq km^{-2} contour, the lowest contour readily discernible as RFP plutonium, and up to 118 GBq (3.2) RFP plutonium outside that area.

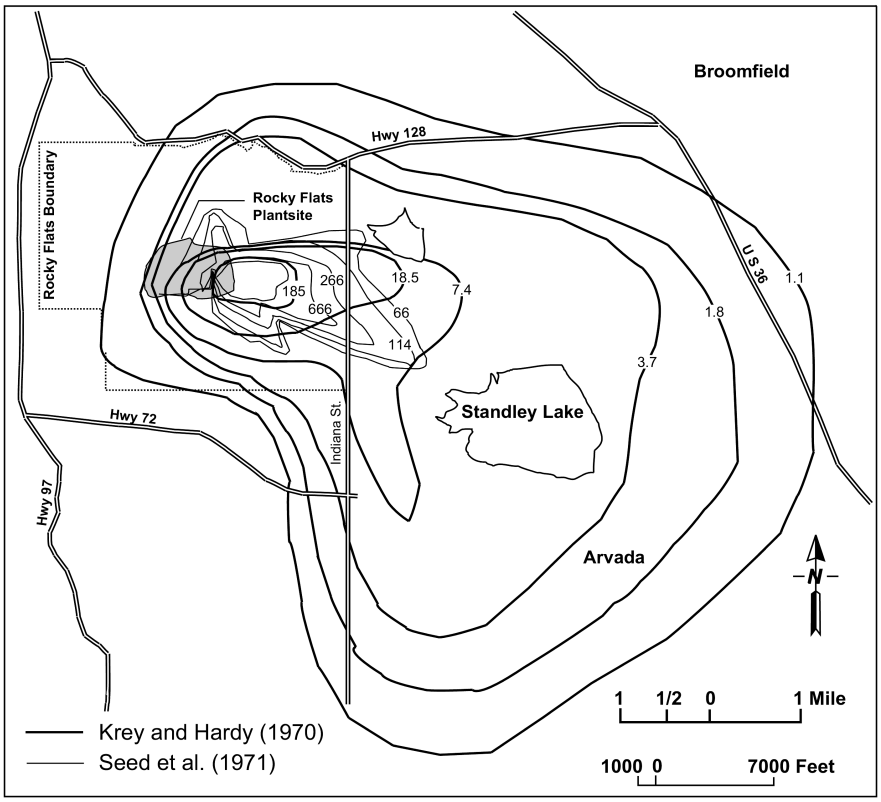

Figure VIII-3. Spatial distribution of $^{239,240}Pu$ ($\times 10^8$ Bq km^{-2}) around the Rocky Flats Plant, as proposed in Krey and Hardy (1970) and Seed et al. (1971) and redrawn from Litaor (1995a). Krey and Hardy sampling depth was 20 cm (8 in.). Seed et al. sampling depth was 3 to 5 cm (1.2 to 2 in.).

Krey and Krajewski (1972) refined the Krey and Hardy (1970) inventory estimate by taking additional soil samples, using plutonium isotopic ratio analyses to refine the predicted weapons fallout levels, and revising the estimation technique. Krey and Krajewski (1972) estimated a total plutonium inventory of 133 GBq (3.6 Ci) with less than a factor of two uncertainty.

Seed et al. (1971) presented a contour map based on 135 soil samples. This map varied significantly from the Krey and Hardy map (Figure VIII-3); however, the sampling depth also differed. The difference in sampling depth is probably the main reason why plutonium areal concentrations measured in Seed et al. (1971) were greater than those reported by Krey and Hardy (1970) in similar areas. Both studies showed dispersion of plutonium to the east of the RFP. Seed et al. (1971) estimated the total release of plutonium to be 32 GBq (0.9 Ci), which did not include the 903 Area itself.

Poet and Martell (1972) conducted an independent sampling program near RFP using about 80 samples from 18 sites to estimate a plutonium inventory of 407 GBq (11 Ci). Poet and Martell (1972) considered their estimate in good agreement with Krey and Hardy (1970), but calculated an

additional 147 GBq (4 Ci) ^{239}Pu out to 68 km (42 mi) in the northwest direction based on the measured ratio of ^{239,240}Pu to ^{90}Sr in those samples. Webb (1996) questioned the use of this ratio because of the different mobility of these radionuclides in soil.

Krey (1976) prepared a final revision to his work of RFP plutonium inventories. Additional samples were collected that allowed the rapidly increasing inventory, particularly at the edge of the 903 Area, to be accounted for. Krey (1976) estimated a total plutonium inventory of 359 GBq (9.7 Ci). This estimate included the edge of the 903 Pad, but not the 902 Pad itself.

Krey (1976) plotted a deposition contour map using 26 samples (Figure VIII-4). Krey also constructed a second isopleth map of plutonium in soils outside the Rocky Flats environs (Figure VIII-5). This map was constructed from 22 samples taken from as far as 60 km (37 mi) north and southeast of Rocky Flats.

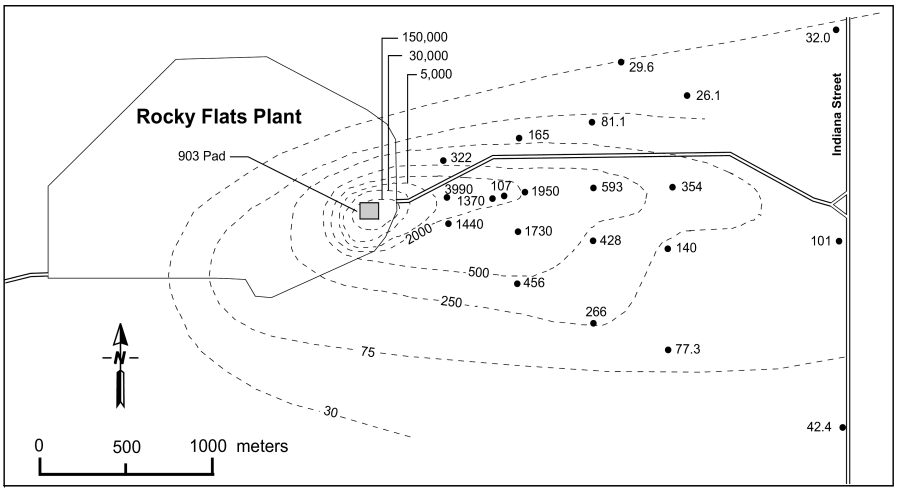

Figure VIII-4. Spatial distribution of ^{239,240}Pu (mCi km^{-2}) in onsite soils east of the Rocky Flats Plant (Krey 1976). The black circles are sampling locations, with measurement results adjacent. For clarity, some of the close-in sampling locations are not shown in this figure. Sampling depth was 20 cm (8 in.).

More recent studies conducted by CSU (Webb 1996) and DOE (Litaor 1995a; DOE 1995, 1996a) attempted to provide the quantity and quality of data necessary to define the extent of plutonium contamination in surface soils. Litaor (1995a) presents an isopleth map (Figure VIII-6) of ^{239,240}Pu concentrations in soils around Rocky Flats that was produced using a statistical interpolation method called kriging (Isaaks and Srivastava 1989). The data for this map were derived from samples collected from a total of 118 plots located near the 903 Area. There were eighty-four 4.0-ha (10-acre) plots and thirty-four 1.0-ha (2.5-acre) plots. Twenty-five subsamples, equally spaced, were collected from each plot using the CDH sampling protocol. Note that the protruding northwest plume shown by Seed et al. (1971) in Figure VIII-3 is not evident. Litaor (1995a) attributed Seed et al.'s northwest plume to one soil sample collected near Walnut Creek, just north of the industrial area of the plant that had a relatively high plutonium activity concentration. Furthermore, the southeastern plume reported in Krey and Hardy (1970) and Seed et al. (1971) and shown in Figure VIII-3 was also not evident (Figure VIII-6).

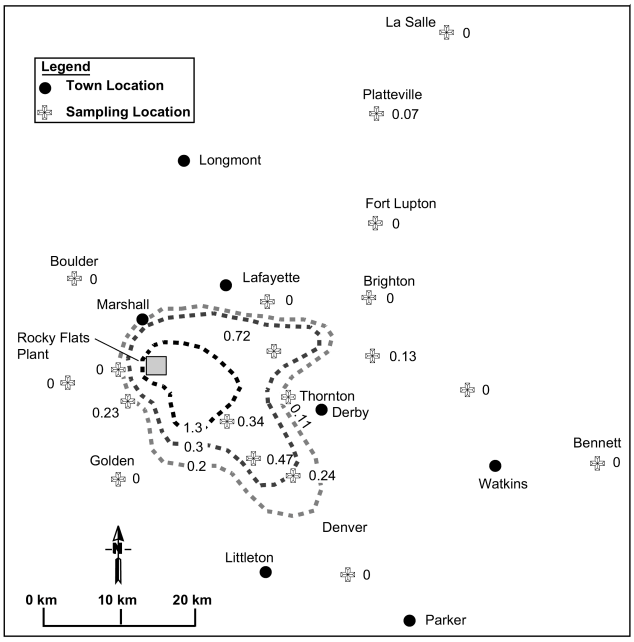

Figure VIII-5. Spatial distribution of ^{239,240}Pu (mCi km^{-2}) east of the Rocky Flats Plant, as proposed by Krey (1976). Figure shows only plutonium thought to be due to releases from the Rocky Flats Plant. Sampling depth to 20 cm (8 in.).

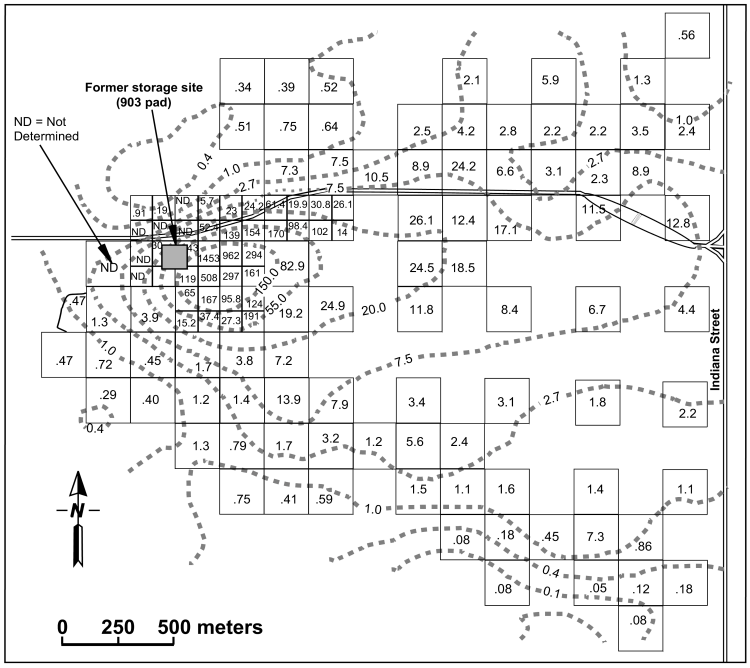

Figure VIII-6. Isopleth map of ^{239,240}Pu (pCi g^{-1}) in soils in the buffer zone east of the Rocky Flats production area using ordinary kriging (Litaor 1995a). The observed values are shown in the center of each 1.01- and 4.05-ha plots. Sample depth to 0.64 cm (0.25 in.). (1 Bq kg^{-1} = 0.027 pCi g^{-1}).

Litaor (1995a) notes that while a few samples in the southeast direction at different distances from the 903 Area had somewhat higher values than the surrounding plots, the ordinary kriging technique failed to produce a southeast plume. Ordinary kriging did not produce the contortions that result from trying to force a polynomial to fit the data, as did the least-squares method employed by Seed et al. (1971). Litaor et al. (1995) went on to revise the kriging technique used (see below and Figure VIII-7) to avoid the possibility of underestimating plutonium concentrations in highly contaminated areas and overestimating concentrations in areas of low contamination.

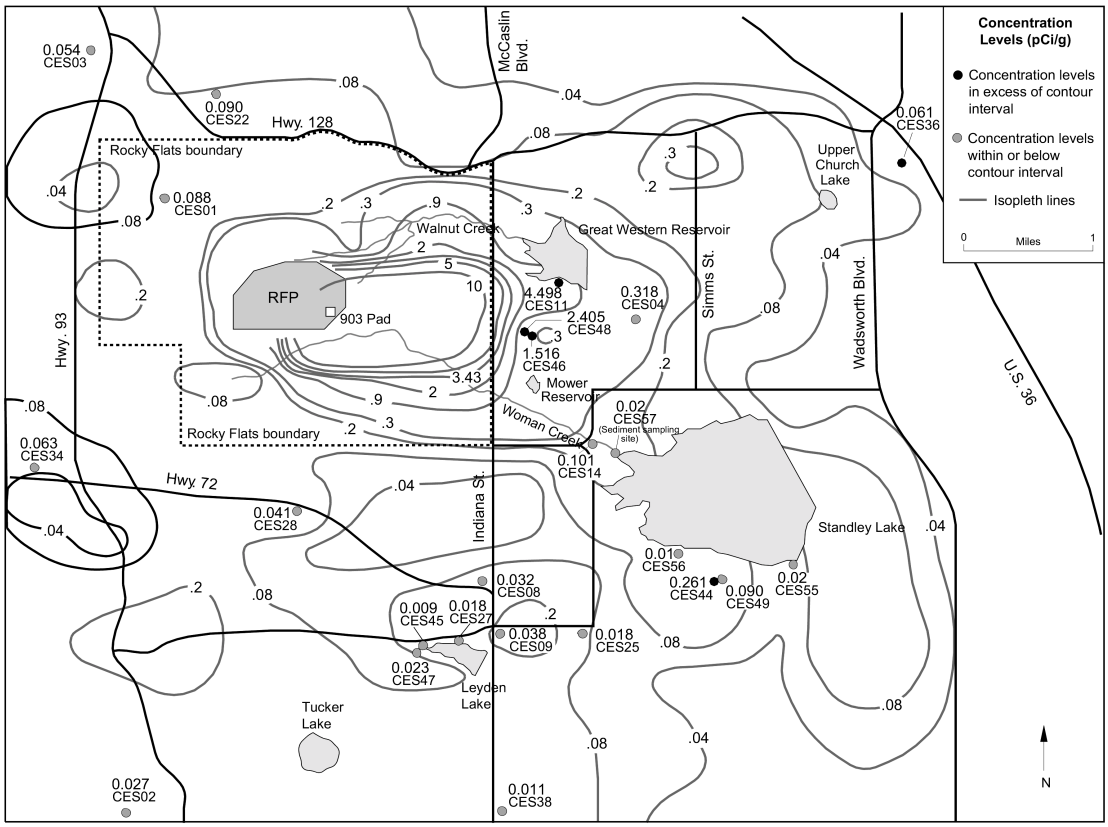

Figure VIII-7. Plutonium isopleth map of ^{239,240}Pu (pCi g^{-1}) in soils around Rocky Flats based on indicator kriging using an exhaustive dataset by Litaor et al. (1995), combined with soil sampling documented in CESC (1996). Sample depth varies. Figure redrawn from CESC (1996). (1 Bq kg^{-1} = 0.027 pCi g^{-1}).

The CDPHE evaluated the spatial and temporal trends of Rocky Flats soil data collected by the CDPHE from 1971 through 1991 (Jones and Zhang 1994). Contour maps were produced using kriging techniques, and when data were controlled for the depth of the sample, there was no evidence of a time trend, either increasing or decreasing. This result implied that data from historical and current studies could be combined, as long as one is comfortable with the different sampling techniques used and the quality of historical datasets.

Litaor et al. (1995) combined datasets from the RFI/RI OU2 study (DOE 1995) and the RFI/RI OU3 study (DOE 1996a) with datasets from CDPHE routine monitoring (Jones and

Zhang 1994), annual RFP monitoring, CSU studies (Webb et al. 1993), Western Technology measurements of soil collected west of the site (Western Technologies 1991), and historical studies (Krey and Hardy 1970; Seed et al. 1971; Krey 1976; Illsley and Hume 1979). The compilation of this "exhaustive" dataset disregarded field sampling technique, time of collection, particle size of the sample, and the analytical method of plutonium analysis. Any redundancy in data (i.e., samples collected by CDPHE and RFP sampling in the same plot) was eliminated by including only the highest activity measured per plot. Indicator kriging (Isaaks and Srivastava 1989) was used for this study rather than ordinary kriging because it does not assume any particular distribution of the data and is resistant to highly skewed distributions and outliers. The resulting isopleth map for the exhaustive dataset is shown in Figure VIII-7. Figure VIII-7 is a complicated map that estimates distribution, but it does not provide plutonium inventories in soil. The values are given as concentration per unit mass and not area. The map supports the existence of a clear east-west trend, with rapid decline in concentration east from the 903 Area. Plutonium activity also decreased rapidly with distance to the north and south. It was indicated that areas west of Rocky Flats have not been significantly impacted by plutonium in soil. The authors considered this plutonium distribution map was significantly different from those produced previously (Figures VIII-3 through VIII-5). Later, Litaor (1999) calculated a total plutonium inventory of 111.2 ±50.8 GBq (3 ±1.4 Ci) for the open space and residential areas (total area 117.4 km^2 [24.3 mi^2]) predominantly east of RFP. The large uncertainty was due to the high variability in plutonium concentration measurements over small distances.

The existence of a southeastern plume remains open to discussion. The southeastern component of the plume reported by Krey and Hardy (1970), Seed et al. (1971), and Krey (1976) is not confirmed in the exhaustive dataset map. Of the four isopleth maps produced (Litaor et al. 1995), the one based on the historical dataset showed evidence of a southeast plume. The two isopleth maps based on the CDPHE and RFP soil sampling techniques did not produce a southeast plume. Furthermore, work on ^{241}Am (Litaor and Allen 1996) showed no plume in the southeast direction. This is noteworthy because there appears to be a strong correlation between plutonium and americium concentrations in soil at Rocky Flats (Ibrahim et al. 1996).

More recently the Citizens Environmental Sampling Committee (CESC) collected surface soil (0 to 3 cm [0 to 1.2 in.]) and composite grab samples (0 to 21 cm [0 to 8 in.]) from 29 locations during a 1994 campaign (CESC 1996). Sample locations ranged from less than 1.6 km (1 mi) to greater than 64 km (40 mi) from the RFP. The highest plutonium concentration was measured in a sample taken about 0.75 mi. (1.2 km) east of the buffer zone boundary. Results were generally in line with previous sampling results, but new information was obtained for locations not previously sampled (Figure VIII-7).

A significant effort to estimate the inventory of plutonium released from the 903 Area has been made in Webb (1996). The study is valuable because it includes an analysis of data from earlier studies to determine inventory estimates that can be compared with his own study. The study area ranged from 200 m to 19 km (220 yd to 12 mi) from the 903 Area (the primary source of plutonium contamination) and swept a 60° arc from 60°T (true bearing) to 120°T. Furthermore, the study area covered a large portion of the populated areas near Rocky Flats. Based on more than 1200 measurements, equations were developed to mathematically describe the decrease in plutonium concentrations with depth into soil, distance from the 903 Area, and direction away from 90°T (all three components carried estimates of statistical uncertainty). These equations were used to quantify the total plutonium inventory in soil in the area of the primary plume. The results were adjusted to account for fallout plutonium by subtracting the

mean background concentration measured along the front range (Table VIII-2). Over 80% of the total plutonium in offsite soils within the model region was determined to be from global fallout, yet over 95% of the total plutonium onsite (within the model region) was determined to be from Rocky Flats. A best estimate of total plutonium (^{239,240}Pu) in the study area attributable to releases from Rocky Flats was 126 GBq (3.4 Ci), somewhat less than previously estimated by earlier researchers. Webb (1996) reported a revised best inventory estimate of 53 GBq (1.4 Ci) when soil volume displaced by rock is considered (Figure VIII-8).

Figure VIII-8. Photograph of a typical rocky soil, northwest sector, Rocky Flats Plant.

The two inventory estimates of Webb (1996) are compared with inventory estimates from four other studies (Figure VIII-9). When calculating the plutonium inventory of earlier researchers that fell within his 60º arc, Webb did not redraw their isopleth lines or make any new conclusions regarding the validity of the numbers. Differences in Webb's inventory estimates, in comparison to those made by earlier researchers (Figure VIII-9, Table VIII-4), were generally attributed to such characteristics as the physical area considered, equations for inventory calculation, and estimates of fallout contribution. Figure VIII-9 also provides a summary of the total inventory estimates for plutonium in soil for the various studies.

As an example of how an estimate of inventory within the 60º arc was made using an earlier study, Webb (1996) superimposed the arc over the Krey and Hardy (1970) contour map (Figure VIII-3, evidently originally plotted by manual interpolation). It was possible to estimate the percent of area within each Krey and Hardy contour interval that fell within the arc, and an inventory estimate and an error estimate of Krey's data were generated by Webb (1996).

The Webb (1996) inventory estimate based upon Seed's work was accomplished in the same manner that Seed calculated his total estimate (Seed constructed contours using curve-fitting and least squares analysis). That portion of Seed's estimate that fell within the 60º arc was then calculated. Given these techniques, Webb (1996) provided separate inventory estimates and

associated error estimates, gleaned from several studies, of the total Rocky Flats originating plutonium inventory within the 60º arc and out to 19 km (12 mi).

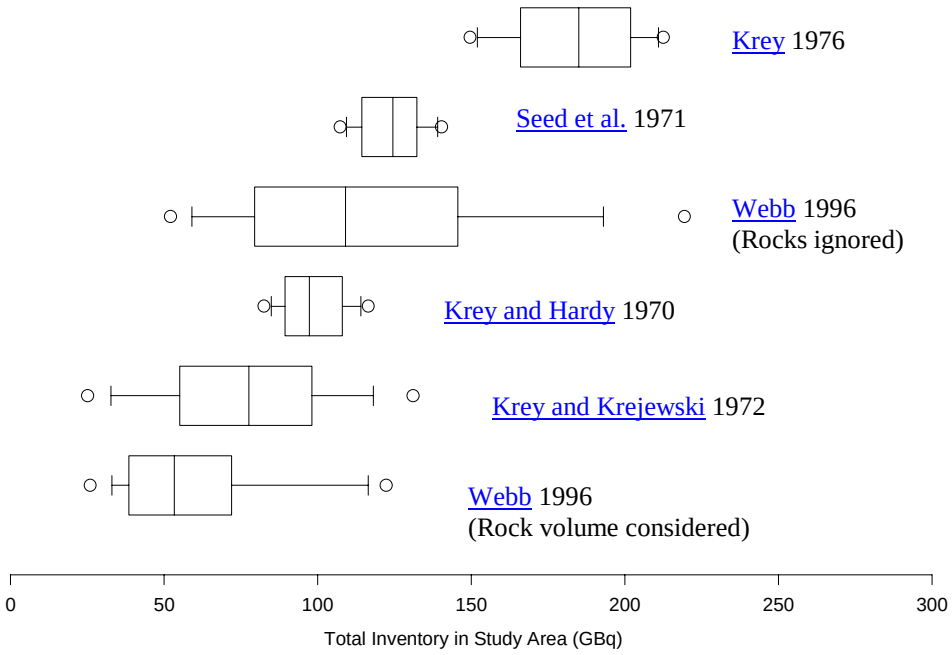

Figure VIII-9. Comparison of plutonium inventory estimates from historical studies with that of Webb (1996). The vertical bar in the center of each box is the geometric mean estimate. The limits of the box represent the 25th to 75th percentiles, end bars on the line represent 5th to 95th percentiles, and the dots at extreme ends are minimum and maximum values.

The Webb (1996) data are not significantly different from several historical estimates if rock volume is not accounted for. When Webb (1996) accounted for the presence of large volumes of rock in nearly all of their soil samples, a substantially lower plutonium inventory estimate was calculated based on his own data. Nevertheless, the upper value of the estimate is not statistically different from any historical value except that of Krey (1976). When rock volume was considered, the best estimate of the plutonium inventory within the study region was (as given above) a geometric mean estimate of 53 GBq (1.4 Ci), with a 95% confidence interval of 21 to 136 GBq (0.57 to 3.68 Ci). The range of historical estimates was 72 to 179 GBq (1.9 to 4.84 Ci). Webb (1996) stated that because it was possible there was no statistical difference between the true mean values within the studies (for the 60º arc), they were tempted to adjust their inventory upward to estimate the total plutonium release surrounding Rocky Flats in all directions. However, they did not do this for two reasons: (1) the current inventory value was based solely on data from within the study area, and (2) another of the transects along 150°T revealed soil plutonium concentrations less than their measured background values.

Table VIII-4. Summary of Plutonium Inventory Estimates in Soils around Rocky Flats (adapted from Webb 1996)

Reference	Total ^{239}Pu inventory	Webb study area[a] inventory	Inventory error estimate	No. of locations/ samples	Weapons testing ^{239}Pu fallout (Bq km^{-2})
Krey and Hardy (1970)	251 GBq	99.4 GBq	±20%	33/52	55.5
Seed et al. (1971)	32.5 GBq	124 GBq	±15%	135/135	86.0
Poet and Martell (1972)	400 GBq	—[b]		18/~80	55.5
Krey and Krajewski (1972)	133 GBq	72 GBq	<factor 2	35/54	55.5
Krey (1976)	359 GBq	179 GBq	±20%	54/~80	62.9
Webb (1996)	—[c]	53.3[d] GBq	GSD[e] 1.6	96/>1200	62.9
Litaor (1999)	111.2 GBq ±50.8[f]	Not applicable	See total inventory	42/~110	[g]

[a] 209 km^2: a sector from 60° to 120° T and 200 m (220 yd) to 20 km (12 mi) from the 903 Area.

[b] Not determined because of incompatible estimation techniques.

[c] Not calculated.

[d] Geometric mean.

[e] GSD = Geometric standard deviation.

[f] ~120 km^2 (24.5 mi^2) of open space and residential areas predominantly east of RFP.

[g] Evaluated based on probability of exceeding an isotopic ratio of 0.155.

It should be noted that Krey and Hardy (1970) state that they employed an alternate sampling method at sites with a preponderance of rocks and included the rocks with the sample. However, 10 sites were described as very rocky and the larger rocks were removed from the sample. Krey and Hardy (1970) determined that approximately 0.2% of the total plutonium was associated with these rocks.

In a related study, Ibrahim et al. (1997) reported total ^{239,240}Pu and the ^{240}Pu: ^{239}Pu ratio around Rocky Flats in an effort to identify the amounts of plutonium contributed by Rocky Flats and global fallout. This work was done in the same 60º sector out to 19 km (12 mi) from the 903 Area. The data from one transect indicated that the total inventory estimate based on total plutonium was not different from that based on the isotopic ratio method. An important point was that, even at certain distant locations where the plutonium in soil was at background concentrations, the atom ratio indicated the presence of a Rocky Flats contribution. Although the Rocky Flats contribution to total plutonium at offsite locations ranged from 24-90%, the overall inventory was relatively small (Ibrahim et al. 1997).

The work reported above suggests that the effort to estimate and compare several inventories of RFP contribution to plutonium in soil in one 60º sector can be worthwhile if differences in research efforts are recognized. Also, estimates of error are of value but may be

quite broad. If there has been no significant redistribution of plutonium in soil over decades, inventory comparisons are more valid. Studies conducted since the 903 Area was paved imply very little, if any, time trend in distribution has been discerned. In the case of plutonium concentration with depth in soil, there is evidence that within a small number of years (<5) of the initial deposition, there has been no significant change with time (see earlier discussion). As for plutonium in surface soil, there has probably been little redistribution over the years in the broad-scale sense, but evidence has not ruled out that some local redistribution may have occurred. Numerous studies have demonstrated that plutonium concentrations and total plutonium deposition (from Rocky Flats) decrease rapidly with distance from the 903 Area and with the deviation in direction from due east. For a discussion of plutonium sediment inventories in water bodies around the RFP, see Chapter VII of this report.

PENETRATING RADIATION AND AERIAL SURVEYS

Penetrating radiation surveys can be used to characterize the concentrations of x- and gamma-emitting radionuclides in soils. The primary radionuclides of concern in the Rocky Flats environs have been isotopes of plutonium. The primary radiations emitted by plutonium are alpha and beta radiations, which are difficult to measure except with laboratory analyses of soil samples. Thus, direct field measurements of plutonium in soil, using radiation survey instruments, are not feasible. However, ^{241}Pu is present in the Rocky Flats plutonium, and it decays to form ^{241}Am. When this nuclide decays, it emits a low-energy x-ray (about 60 keV), which can be detected by some gamma radiation surveys. The results can be used to calculate concentrations of ^{239,240}Pu based on isotopic ratios of ^{239}Pu/^{241}Pu. If the isotopic ratio of ^{241}Am/^{239}Pu can be established, ^{239}Pu concentrations can be indirectly measured (Ibrahim et al. 1995). Radiation surveys can also be used to determine concentrations of other radionuclides, such as ^{137}Cs. Results of aerial gamma radiation surveys around the RFP are presented in Appendix G.

Soil contamination by ^{241}Am extends, as a plume, east from the 903 Area to beyond the perimeter fence at Indiana Street. This area of contamination is similar in character and in locations of maximum concentrations to that shown by DOE soil sampling. After three surveys, there is no evidence that the spatial distribution of ^{241}Am in soils has changed over time. Levels of ^{137}Cs showed no pattern indicative of Rocky Flats release. Aerial surveys generally are not as accurate as soil sample analysis for determining radionuclide concentrations. For this reason, we conclude that radiochemical analyses are more useful than aerial surveys for quantitative evaluations of plutonium and americium contamination around Rocky Flats. However, aerial survey results provide useful corroboration.

Stone and Whicker (1995) presented an extensive study of background radiation variation in populated areas along the front range of Colorado. Measurements were made using a pressurized ionization chamber that produces exposure rate readings in micro-Roentgen per hour (μR hr^{-1}). While the study provided large amounts of data that were correlated with environmental factors, such as elevation and geologic formation, it is only useful to the historical dose reconstruction for relative risk comparisons. Results are not related to releases from the RFP.

BERYLLIUM IN SOIL

Beryllium is one of the nonradioactive materials of concern that were identified in Phase I of this historical public exposures study. Beryllium concentrations in soil are of interest because resuspension of beryllium in soil is a potential exposure pathway and a pattern of beryllium contamination in soil could reveal information about discharges from the plant. Beryllium sources that might affect concentrations in soils at the RFP include

- Operations at the plant
- A beryllium ore industry located 2 km (1.2 mi) east of the plant
- A beryllium ceramics industry located 15 km (9.3 mi) south of the plant
- Coal burning and other combustion sources near the plant
- Beryllium in gravels brought into the site
- Naturally occurring beryllium (Barrick 1982; Kray 1992).

A study to characterize sources of beryllium and to quantify environmental beryllium contamination in soil near the RFP was conducted in 1981 (Barrick 1982). That study reported an estimate of 196 g for the total amount of beryllium exhausted from all buildings that processed beryllium during the 24-year period 1958 to 1982. The estimate included releases from two reported filter fire accidents in February 1978, which released 14.5 g (0.51 oz) of beryllium from the main beryllium production building, Building 444. In that study, 241 soil and rock samples from the site and from nearby areas were obtained. Deeper samples were taken at 5 to 10 cm (2 to 4 in.) to establish geological background levels of beryllium. The study concluded that RFP-originated beryllium could not be distinguished from geological, naturally occurring beryllium taken on lands outside plant property. The survey found that natural gravels and an estimated 36 million kg (79 million lb) of gravels brought in and added to RFP surfaces have the highest and most variable beryllium concentrations. The mean concentration in these gravels is 1.1 $\pm$1.4 μg g^{-1} of soil (parts per million or milligrams per kilogram of soil). The background beryllium concentrations in soil (Rocky Flats alluvium) averaged 0.64 $\pm$0.07 μg g^{-1}.

In what appears to be an earlier draft of these results, Barrick (1982) suggested that atmospheric transport of beryllium to soils surrounding the plant had not occurred because no surficial soils near the plant were found to have elevated beryllium concentrations. The mean beryllium concentration in soils in the plant area was reported to be 0.6 μg g^{-1} and ranged from 0.2 to 1.1 μg g^{-1}. Higher concentrations found near roads and buildings were attributed to surficial gravel aggregates, which had the highest background or natural beryllium levels.

One accumulation of beryllium in soil that likely originated from RFP operations was found 30 m (100 ft) from the stack of a plant that processed beryllium. The soil samples at this location had beryllium concentrations 44 to 69 μg g^{-1}, which were well above background. Subsequent to this study, more samples were taken at various depths to try to determine when the accumulation in soil had occurred. A high beryllium soil concentration was found in a 10 m × 10 m (33 ft × 33 ft) plot adjacent to door 10 of Building 444. The contamination was found within the top 5 cm (2 in.) of soil, and the beryllium concentration ranged from 1 to 114 μg g^{-1}. The pattern of contamination suggested the source of the beryllium release was door 10 and not the nearby stack for Building 444 or the filter plenum room. The study's authors recommended removal of 1 m^3 (35 ft^3) of contaminated soil (Barrick 1982). Before 1970, chlorinated hydrocarbon solvent that had been used to rinse beryllium parts was disposed of by pouring it on soil outside door 10 on the south side of Building 444. A special study to sample beryllium in air near this solvent

disposal site was done in the summer of 1977. Filters were collected weekly from an air monitor mounted 0.9 m (3 ft) above ground level. The detection limit for beryllium in air was reported to be approximately 7.5×10^{-9} µg m^{-3}. The maximum beryllium concentration in air reported at this location was 2.3×10^{-3} µg m^{-3} and the average was 9×10^{-4} µg m^{-3}. Because the air concentrations averaged about 9% of the ambient air standard of 0.01 µg m^{-3}, soil removal was not recommended at that time (Barker 1978).

The Colorado Department of Public Health and Environment (CDPHE) conducted studies on beryllium in soil in 1971 and 1989. In 1992, at the request of Bob Quillan, (a Health Advisory Panel member representing the CDPHE), a discrepancy in the 1971 and 1989 beryllium soil sampling results was evaluated by CDPHE personnel (Quillan 1992). The study done in 1989 reported 21 results, all less than the analytical detection limit for beryllium of 2.7 µg g^{-1}. The 1971 data consisted of 13 results, ranging from 2.0 to 60 µg g^{-1}, with no analytical detection limit reported. The pattern of positive values seen in 1971 was not consistent with what would be expected if the beryllium in the soils had been deposited because of atmospheric dispersion from the RFP. Spatial variations did not indicate a plume of beryllium from the plant operations. The 1989 dataset was judged to be more credible because of better documentation of analytical procedures, more rigorous quality assurance, and improved analytical methods and equipment (Kray 1992).

To investigate potential contamination of surface soils from windborne dispersal in Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA) OU3 (offsite), the distribution of metals in OU2 (onsite areas) was evaluated. If contamination of soils onsite was due to activities at the plant, then the soil sampling results were expected to show a distinct spatial distribution trend of decreasing concentrations with increasing distances from areas where beryllium was used. The CERCLA program personnel reasoned that if metal contamination of soil in onsite areas (OU2) was at background concentrations or appeared to be a result of localized incidents of contamination, and if no spatial trends could be identified, then contamination in offsite (OU3) soils was unlikely. Therefore, sampling of OU3 soils (at distances further out) would not be warranted (DOE 1994). Samples were compared with results from two studies of background concentrations: the Rock Creek and the Background Soils Characterization Program. The study found a mean beryllium concentration in OU2 soils of 0.68 µg g^{-1}, with a standard deviation of 0.21 µg g^{-1}. One extreme value of 1.50 µg g^{-1} was determined to be an outlier. The beryllium concentrations in OU2 soils were similar to those for Rock Creek soil samples, which had a mean concentration of 0.68 µg g^{-1} and a maximum concentration of 0.96 µg g^{-1}. The Background Soils Characterization Project study showed a similar mean of 0.66 µg g^{-1}. The U.S. Geological Survey geometric mean for beryllium concentrations in the front range soil was 1.20 µg g^{-1}. Beryllium concentrations in the OU2 soil did not appear to be above background. No spatial trends in the data or recognizable plume were apparent and no preferred direction of beryllium in soils was observed. This study did not provide evidence of airborne contamination (EG&G 1995).

The results of soil monitoring conducted as a part of investigations of CERCLA OU5 (Woman Creek Drainage) and OU6 (Walnut Creek Drainage) do not indicate a windborne deposition pattern (DOE 1996b, 1996c).

Samples of cottonwood leaves, collected from trees growing on soils with 0.1–1.0 µg g^{-1} beryllium, contained beryllium concentrations that were only slightly correlated (r = 0.25) with the concentration in the soil. This observation led researchers to decide that leaf surveys would

not be useful as indicators of soil contamination (Barrick 1982), although analysis of cottonwood trees has been used to locate beryllium ore deposits in the former Soviet Union.

Taken together, the soil data suggest that beryllium deposited on soil from RFP releases is not distinguishable from beryllium in the soil from natural and other sources.

SUMMARY

Data collected from soil studies conducted in the Rocky Flats environs are quite useful to the historical public exposures studies. Numerous background concentration data are available. A recent study documented in DOE (1996a) provides exhaustive data on background soil characteristics and contaminants (fallout radionuclides on offsite areas, metals, and organics). The data are sufficient for defining background contaminant concentrations.

Special studies conducted by CSU, DOE, and Litaor in the 1990s are of particular use to these studies because they provide the most extensive and highest quality data yet seen. The studies conducted by CSU over several years in a permanent plot located east of the 903 Area are relevant in evaluating releases from that site. Exceedingly valuable new information has been produced in several areas of interest. This includes estimates of statistical uncertainty for many variables in regard to plutonium contamination in onsite and offsite soils. Also, knowledge of the extent of contamination in relation to distance and lateral spread (away from due east) from the 903 Area, more information on depth distribution and inventory in soils, and evidence as to mobility in soil have been enhanced.

For the most part, data collected from Rocky Flats soil studies indicate that there is very little, if any, time trend in plutonium distribution and concentrations in soil. Statistical analyses of historical and current CDPHE data combined with RFP data for the years 1984 to 1992 demonstrated no evidence of a time trend for depth distribution in shallow soil (Jones and Zhang 1994). This suggests that plutonium is relatively immobile in surface soils and agrees with studies by CSU on plutonium transport in soils. It also suggests that historical data can be used with current data. Litaor et al. (1995) produced a plutonium isopleth map using “exhaustive” data from previous and current soil studies. This isopleth map could be used as a reference soil plutonium concentration map to compare with reconstructed depositions from historical releases. The isopleth pattern is consistent with the hypothesis that the dominant dispersal mechanism of plutonium was wind dispersion from west to east.

Penetrating radiation field surveys in the early 1970s helped establish the extent of plutonium and americium remaining in soils after the 903 Area was covered with asphalt. Aerial radiation surveys, reviewed in Appendix G, corroborate the spatial distribution of plutonium in soils seen in other studies.

Data on beryllium in soil indicate that deposits from the RFP cannot be distinguished from that of other or natural origins.

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