CHAPTER II

WHAT CAN ENVIRONMENTAL DATA TELL US ABOUT PAST RELEASES FROM ROCKY FLATS?

The Rocky Flats historical public exposures studies are limited by a lack of quality environmental data during the time period of highest releases. This conclusion underscores the need to approach the reconstruction of important release events, such as the 1957 fire, by methods other than the use of environmental data, specifically by looking at events that happened inside the facility. This "inside-out" approach used in Phase II to reconstruct releases from the 1957 fire (Voillequé 1999a), complements the "outside-in" approach used in Phase I (ChemRisk 1994a).

The environment around the RFP is an important link between contaminants released from operations at the site and the exposure of the surrounding public to these contaminants. The ambient air and water monitoring data collected during the time period of interest (pre-1970) are not adequate to estimate population exposures due to inhalation or ingestion of materials released from the Rocky Flats Plant (RFP). This led us, early in the planning for this task, to develop the general principles and the framework for evaluating the usefulness of historical environmental data at Rocky Flats, which are presented in Chapter I, to guide our efforts. Figure II-1 highlights the types of monitoring data and time periods that were evaluated for this project.

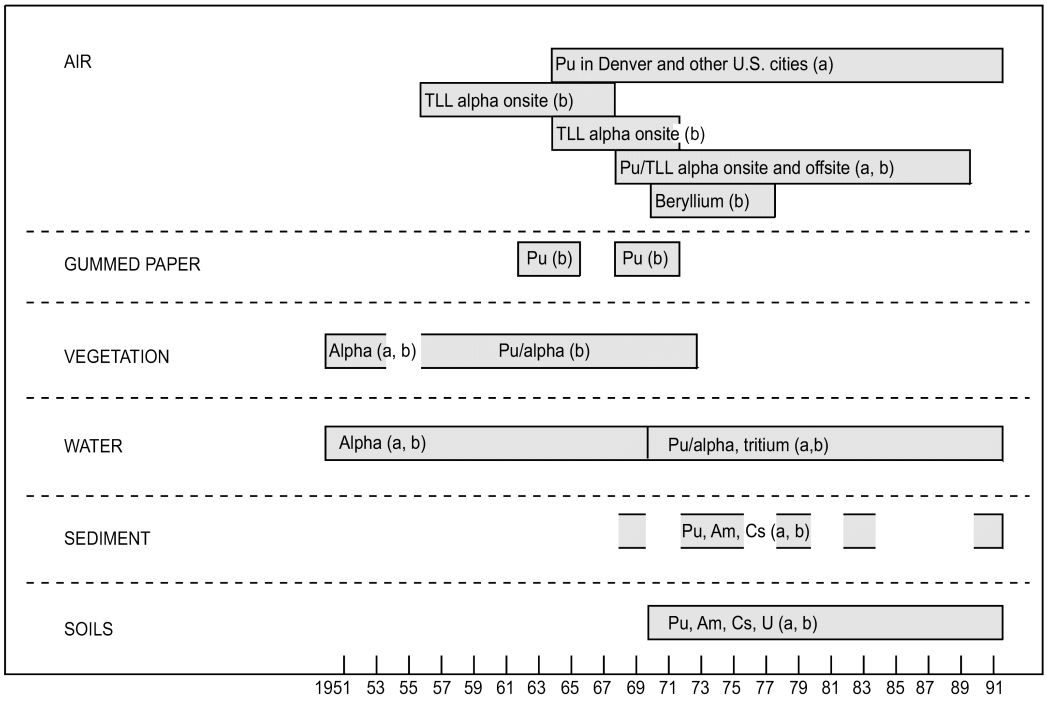

Figure II-1. Timeline of the types of environmental measurements for contaminants of interest and the time interval over which they were evaluated for this project. "TLL alpha" is total long-lived alpha.

The specific uses of the environmental data to support other aspects of the study continued to evolve until the final stages of the project, when all the pieces of available information came together. There was a natural progression from the collection and evaluation of environmental data (Task 4), to application of those data in source term development (Task 2) or risk assessment (Task 3). This process continued throughout the Phase II work, as models were refined and data needs were identified.

Figure II-2 illustrates the primary airborne release points within the production area of the RFP. The Building 771 stack was the source of routine plutonium releases and releases from a major fire in 1957. Roof vents in Buildings 776/777 and 707 were the source of routine plutonium and carbon tetrachloride releases and plutonium releases during a fire in 1969. Building 444 was the source of routine beryllium releases. The 903 Area was a storage area for barrels of waste oil. The barrels stored at the 903 Area leaked and contaminated the soil, which became suspended and transported offsite, mainly in 1968 and 1969. The main storage pad was covered with asphalt in 1969, but adjacent contaminated soil areas continued to be a source of contamination.

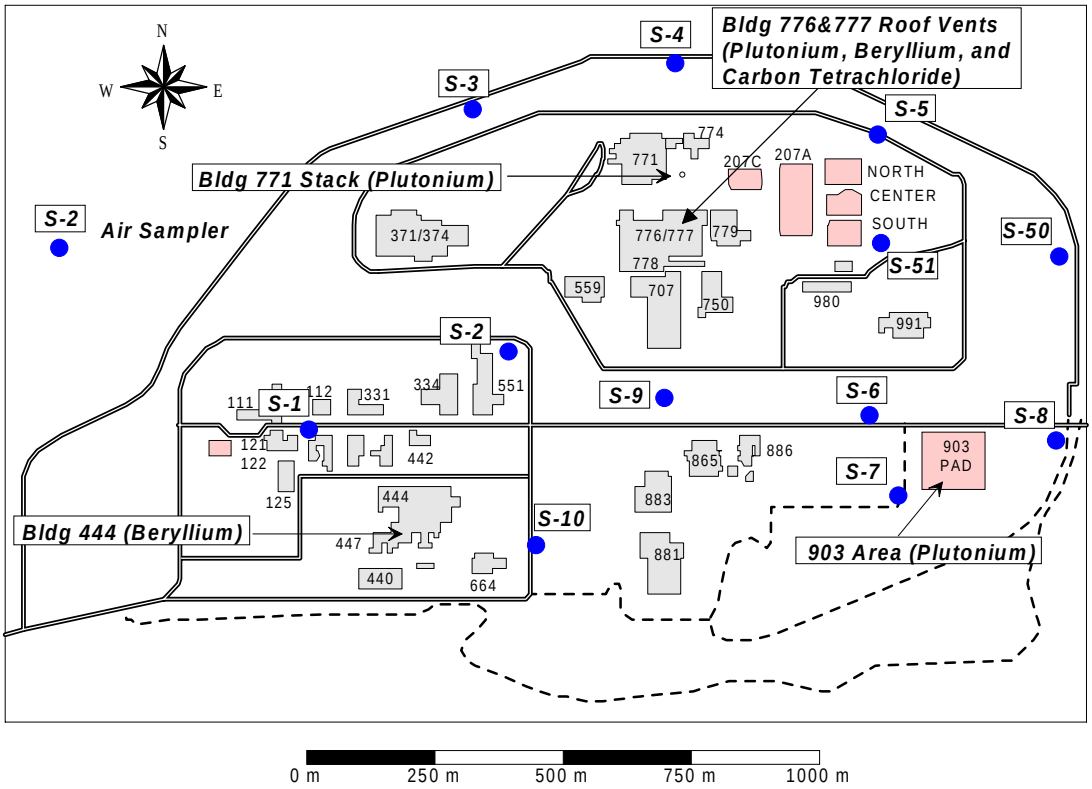

Figure II-2. Main atmospheric release points for plutonium, beryllium, and carbon tetrachloride. Major fires occurred in Building 771 in September 1957 and in Buildings 776/777 in May 1969. The 903 Area was the outside storage location of drums of plutonium-contaminated waste oils**.** This figure illustrates the major building and roads as they existed around 1990. However, placement of air samplers is based on air sampler locations and numbers that existed before 1973. See Chapter III and Appendix B for maps of air sampler locations as they existed with respect to past features.

Chapters III–IX of this report are organized by the different types of environmental media (air, water, vegetation, soil, sediment, etc.) that were historically monitored around Rocky Flats. A general overview of the types of useful data located for each medium is presented in this chapter. This is followed by a series of relevant questions that can be partially answered by evaluation of the historical environmental data.

GENERAL DESCRIPTION OF MONITORING DATA BY MEDIUM

Ambient Air

The first routine monitoring of plutonium in air of the region was conducted by the U.S. Public Health Service (PHS) in Denver, beginning in November 1965. We examined the time history of this monitoring program, which was later transferred to the U.S. Environmental Protection Agency, through 1989. In addition, monitoring of plutonium in air of other U.S. cities by the PHS and the Health and Safety Laboratory (HASL) provided a time history of plutonium from global weapons fallout for 1965–1989.

Before 1970, the RFP contractor routinely monitored ambient air for total long-lived alpha (TLLa) activity, but not for plutonium. Measurement of TLLa activity was a very rough screening method for plutonium, partially because of the contribution of naturally occurring radionuclides that also contributed long-lived alpha activity. More importantly, air sample volumes were small, and counting times were very short, resulting in poor sensitivity, especially in offsite air samples. TLLa activity in air near a waste oil storage site (the 903 Area) within the RFP industrial area clearly showed the impact of disturbance of contaminated soil there in the late 1960s.

After the fire in May 1969, air monitoring programs near Rocky Flats were expanded and refined. The RFP contractor began to routinely analyze air samples for plutonium in addition to TLLa activity. However, there was a transition period in their collection and analytical methods between 1970 and 1975, making trend analysis in that period difficult.

Other agencies also began monitoring after the fire. High quality independent monitoring by the HASL at several locations near Rocky Flats began in 1970 and continued through 1981. The Colorado Department of Health (CDH)[a] began routine monitoring of air for plutonium and TLLa activity near Rocky Flats in 1969 and at other locations in the region shortly thereafter. A number of shorter term special studies were reviewed, which provided information about data quality, particle size, and spatial distribution of plutonium.

Based on the framework outlined in Chapter I, a number of sets of air monitoring data were identified as likely to be useful to the historical public exposures study. These data were evaluated in this report, which served as a reference document for other source term and risk reports produced in Phase II. The air monitoring data that were used in Phase II are summarized in Table II-1.

[a] Current name Colorado Department of Public Health and Environment (CDPHE).

Table II-1. Application of Air Monitoring Data in the Historical Public Exposures Studies on Rocky Flats, Phase II

Data set (agency)[a]	Years	Application
Plutonium (Pu) in Denver air (PHS/EPA)	1954–1964 extrapolated 1965–1989 measured	• risk comparison for integrated exposures closer to the plant (HAP 1999) • subtraction of fallout component for validation, 1970–1989 (Rood and Grogan 1999a)
Sr-90 in deposition, Denver and New York City (HASL)	1954–1964	• back-extrapolation of Pu in Denver air (see above)
Pu in U.S. cities other than Denver (HASL, PHS, EPA)	1965–1989	• fallout time trend and magnitude • illustration that Pu in Denver air is consistent with global fallout in other cities
Pu in air at Indiana Street (RFPcontractor)	1971–1989	• comparison with predicted concentrations from all RFP sources (Rood and Grogan 1999a)
Pu in air at Indiana Street and old RFP boundary (HASL)	1972–1981	• comparison with predicted concentrations from all RFP sources (Rood and Grogan 1999a)
Pu in air at Indiana Street (CDH)	1970–1987	• comparison with predicted concentrations from all RFP sources (Rood and Grogan 1999a)
Pu in air at Broomfield and Leyden (RFP contractor)	1973–1989	• comparison with predicted concentrations from all RFP sources (Rood and Grogan 1999a)
daily total long-lived alpha (TLLa) activity in air at S-8 onsite sampler (RFP contractor)	1964–1971	• calibration of 903 Area source term model for suspension of contaminated soil (Weber et al. 1999) • identification of highest discrete release days (Weber et al. 1999)
TLLa in onsite air at all samplers (RFP contractor)	1964–1971	• illustration of (1) steep concentration gradient from 903 Area and (2) highest releases in 1968–1969 • comparison to predicted concentrations from May 1969 fire (Rood and Grogan 1999b)
Pu and TLLa in onsite air (CDH, HASL, RFP contractor)	1970–1971	• fraction of TLLa in 1960s that was Pu (Weber et al. 1999)
Pu in onsite air (RFP contractor, HASL, CDH)	1969–1985	• illustration of temporal decrease in airborne contamination from suspension and resuspension of contaminated soil and routine releases
various research projects by RFPcontractor and others, scientific text books and articles	varies	• adjustment of TLLa counts for inlet and filter collection efficiency, counting efficiency, volume of air sampled, and counting error; definition of associated uncertainties (Weber et al. 1999)
resuspension experiments (RFP contractor and other agencies)	1970s	• resuspension factor (Rood and Grogan 1999a) • particle-size distributions (Weber et al. 1999) • illustration of steep concentration gradient from 903 Area
Beryllium in onsite and offsite air (RFP contractor)	1973–1976	• comparison to natural background, predicted concentrations and EPA guideline (this report)

[a] PHS = Public Health Service; EPA = Environmental Protection Agency; CDH = Colorado Department of Health and Environment; HASL = Health and Safety Laboratory; TLLa = Total long-lived alpha activity.

In Table II-1, onsite air sampling refers to locations within the industrial area. Note that there are no useful data offsite during the time of greatest interest (before 1970) that could be used to compare with predicted concentrations. Comparison with predictions was possible in the 1970s and 1980s using data from several agencies (RFP contractor, CDH, and HASL). These comparisons were made on an annual average time resolution, with an estimate of fallout

background subtracted from the measured values. The data used in this comparison are tabulated in Appendix B, Table B-15. Some data were not used quantitatively in source term development or risk assessment, but do illustrate fundamental points that influenced our decisions in those areas. Those data are listed in Table II-1 as well.

Deposition (Gummed Paper)

Gummed-paper collectors were first used to monitor deposition of radionuclides from air during 1954–1955. These early collectors were used periodically to measure radioactive fallout at the RFP. Autoradiographs of the paper were used to determine the number of radioactive particles deposited. Particles were seldom observed during these early years, and the autoradiographs provide no data of use to the historical public exposures studies.

In later years, gummed-paper collectors were located on and off the RFP and were routinely analyzed for plutonium. Data are available for the years 1963–1965 for 19 sites located on or near the RFP, primarily at areas downwind of the Building 71 stack. Data are available for the years 1970–1972 for 5 onsite and 12 offsite locations. Three remote locations were added in 1970. All of the measurements were reported as plutonium activity per unit surface area.

Deposition data were quite variable and highly dependent on the collection efficiency of the gummed paper. Collection efficiencies were not reported by RFP scientists. However, based on published empirical relationships between precipitation and collection efficiency, we estimated collection efficiencies, using regional precipitation data for the periods studied. The original data were corrected using the estimated collection efficiencies, but the variability found in the data and small number of results available limit their quantitative usefulness. Data obtained during these years are best used to confirm fallout from atmospheric weapons testing and RFP stack release patterns.

Vegetation

A preoperational background study in 1951 of alpha activity in vegetation and water began a routine vegetation monitoring program which continued through 1953 (Quimby 1952). In 1951, at the request of the Atomic Energy Commission, the Radiological Science Department of Hanford Works collected 75 vegetation samples from 25 locations within a 6-mi (10-km) radius of the RFP center. Samples consisted of farm crops and vegetation commonly seen during July to October 1951. From April 1952 through late 1953, Dow Chemical Company sampled vegetation bimonthly at 30 onsite locations along a 500-foot grid, at 163 offsite locations along a 2000-foot grid, and at 97 remote offsite locations bounded approximately by Denver, Golden, Boulder, and Brighton, Colorado.

Routine vegetation sampling was discontinued after 1955, but was resumed after the 1957 fire. In response to the fire, samples were collected at all the locations established in the 1952-1953 vegetation monitoring program. The samples collected in conjunction with the fire included 222 vegetation samples in 1957 and 132 vegetation samples in 1958. Over 80 of the 1957 samples were analyzed specifically for plutonium. The data are the earliest plutonium measurements in vegetation around the RFP.

Routine monitoring of vegetation continued from 1963 to 1970, and consisted of gross alpha analyses of samples collected at 55 to 65 locations on the 2000-ft grid and 44 locations off the

grid. Special sampling occurred again in 1969 following the May fire in Building 776-777. Along with the routine samples, 22 samples were collected from 11 offsite locations. Both the routine and nonroutine samples collected in 1969 were analyzed for plutonium as well as TLLa activity. The average TLLa activity in vegetation was 86 pCi kg^{-1}, with a standard deviation of 73 pCi kg^{-1}.

Historic measurements of TLLa activity in vegetation provided some information on the spatial and temporal distribution of alpha emitters in the Rocky Flats area. Plutonium-specific measurements in vegetation, collected after the 1957 fire, from 1969-1972, and after 1979 were evaluated for their usefulness in confirming plutonium release estimates especially during the key release events. However, the lack of specific information on the types of vegetation sampled in the early years and the difficulty of verifying sampling location led to difficulty in data interpretation, thereby limiting the usefulness of the historic vegetation data set. The data do lend support for the deposition patterns measured in soils.

Surface and Drinking Water

Surface water samples were collected and analyzed for gross alpha activity from the Rocky Flats Plant area beginning in 1951, before operations began in 1952. However, the extent of the monitoring program, the spatial distribution of sampling, and the types of materials measured were fairly limited until the early 1970s.

By the early 1970s, all effluent streams leaving the site were sampled. Water was monitored continuously, and samples (including grab samples) were collected daily from the sewage treatment plant influent and effluent, and from Ponds A-3, B-4 and C-1. Continuous sampling was also conducted downstream in Walnut Creek as it crossed Indiana Street. Offsite water sampling was done at the Great Western Reservoir and Standley Lake. In later years, specific isotopic analyses were performed by both the site contractor, the CDPHE and local communities.

The Colorado Department of Health began its own monitoring program for tritium and other radionuclides and nonradioactive constituents following the May 1969 fire. They analyzed water for tritium at Walnut Creek at Indiana Street, Standley Lake and the Great Western Reservoir, and occasionally from Woman Creek at Indiana Street. It was their monitoring that first detected the large tritium release from the RFP in April 1973.

The surface water data were not directly applicable to verifying releases or transport in the environment because airborne, and not waterborne releases, had the greatest impact on offsite radiation dose and risk.

Sediment

Sediment samples were periodically collected, primarily at the Great Western Reservoir, by various government and research organizations from 1969 through 1992. Standley Lake and Mower Reservoir, to a lesser extent, were also sampled. Some background samples were also collected. Colorado State University (CSU) conducted a study of RFP holding ponds upstream of the Great Western Reservoir from 1971 through 1973, which confirmed that liquid releases were an important source of plutonium in the Great Western Reservoir.

Sediment samples were either surface grabs, through the use of a dredge, or cores. Sample analysis focused on plutonium, because this was the primary contaminant released from the RFP

and because it persists in the environment. Other radionuclides, such as cesium-137 (^{137}Cs), were sometimes measured and provided markers for known events, such as global fallout, which were then used to date core segments. Dated core profiles provided a temporal history of plutonium deposition onto lake bottoms.

Results of sediment studies were used to check the magnitude and timing of the model-predicted atmospheric releases of plutonium from the RFP (Rood and Grogan 1999a).

Soil

Extensive monitoring of soil around Rocky Flats for plutonium did not occur until the 1970s. Measurements of TLLa activity in soil from the 1950s and 1960s were not sensitive indicators of plutonium contamination, because of the presence of naturally occurring alpha-emitting radionuclides.

Data collected from soil studies conducted in the Rocky Flats environs were very useful to the historical public exposures studies. In particular, the special studies conducted by CSU, as they provided the most extensive and highest quality data for comparing with the model-predicted spatial distribution of plutonium. The model-predicted soil plutonium inventory estimates were also compared with inventory estimates made by a number of researches based on direct measurements of plutonium concentrations in soil. Background concentrations of plutonium and other contaminants were defined based on a review of many different studies of these materials in environmental media (see Appendix H).

QUESTIONS AND ANSWERS

What Is Plutonium Background?

Plutonium does not occur naturally in the environment. Plutonium background refers to the concentration that is present from sources other than the RFP. Background sources of plutonium measured in air and soil are reviewed in Chapters III and VIII and Appendices B and H. Background plutonium in surface and drinking water are reviewed in Chapter VI. It is important to understand the trends in plutonium background so that the contribution from the RFP can be assessed. The main other source of plutonium in the environment is widely dispersed fallout from nuclear weapons testing.

The time trend of plutonium background in air is reviewed in Chapter III of this report. Highest concentrations were measured beginning in the mid-1960s, and even higher concentrations were extrapolated for the early 1960s by examining data for other fallout radionuclides. In Denver, we estimated that 95% of the total exposure to fallout plutonium occurred before 1975, with 37% in 1963 and 1964. Annual average concentrations of plutonium were less than 1 fCi m^{-3} (femtocuries per cubic meter air) even in the highest years. An integrated concentration of fallout plutonium in Denver air was developed for 1954–1989; this provided perspective to plutonium concentrations predicted from Rocky Flats releases (HAP 1999).

Deposition rates of radionuclides were historically measured by gummed-paper collectors. There are no data to establish background plutonium deposition rates in the vicinity of the RFP during the periods of interest. Regional background measurements were not performed during the

years 1963–1965. Worldwide fallout of plutonium from nuclear weapons tests dominated global measurements during these years and were quite variable geographically (see Holleman et al. 1987 and Appendix H of this report). Three background locations (Alamosa, Berthoud, and Castle Rock) were used in the RFP gummed-paper monitoring program during the years 1970–1972; however, for reasons discussed in Chapter IV, the results are unusable.

The average level of plutonium measured in vegetation after the 1957 and 1969 fires at distances greater than 5 miles from the RFP was approximately 25 pCi kg^{-1}, ranging from 10–50 pCi kg^{-1}. This value is similar to that reported for vegetation taken near the Rocky Flats Plant in 1974 (Little 1980).

Many studies of background concentrations of plutonium in soil have been reviewed in Appendix H to this report. 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. 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.

Some measurements have been made of plutonium concentrations in lake and reservoir sediments outside the area of possible impacts from Rocky Flats releases (see Table VII-2). The concentrations of $^{239,240}Pu$ in the sediments of Colorado Front Range lakes and reservoirs ranged from 0.006 pCi g^{-1} (measured in Boyd Lake by the National Center for Atmospheric Research in 1969) to 0.19 pCi g^{-1} (measured in Wellington Lake by the Colorado School of Mines in 1988). The average concentration of $^{239,240}Pu$ in these lakes, based on 11 measurements made from 1969 through 1992, is 0.06 pCi g^{-1}. The major source of $^{239,240}Pu$ in these sediments is global fallout from nuclear weapons testing.

There are some data on concentrations of plutonium in waters in the general region of the RFP. A small number of measurements were made in 1969 (after the May 1969 fire) by Poet and Martell (1972) in lakes near the RFP that did not receive liquid effluents from the RFP. These values ranged from about 0.0015 to 0.013 pCi L^{-1} (1.5 to 13 fCi L^{-1}). Plutonium concentrations in Colorado drinking water from Denver and Platteville for 1981–1992 ranged from 0.001 to 0.01 pCi L^{-1} (1 to 10 fCi L^{-1}). Similar plutonium levels were measured in water discharged from the holding ponds to Walnut and Woman Creeks after about 1990.

What Is Alpha Background?

Alpha background refers to the concentrations of alpha radioactivity in the environment that are not from Rocky Flats materials. Naturally occurring radionuclides of uranium, thorium, and radium, and their decay products like radon, contribute to the natural alpha activity background. Specifically we are interested in total long-lived alpha activity (TLLa), and not the short-lived decay products of radon, because TLLa activity was used by the RFP contractor as an indication of the presence of plutonium.

Measured concentrations of uranium, thorium, radium and decay products in soils around the RFP and up to 45 miles east have been reported (Richmond 1970). Total uranium concentration (all isotopes) was given as 1.9 ± 0.4 pCi g^{-1} and ^{238}U alone was 1.0 ± 0.2 pCi g^{-1}. Total uranium, thorium, and radium concentrations in soil averaged 8.0 pCi g^{-1}. Inclusion of decay products produced a total natural alpha activity content of soils at the sampled areas of about 20 pCi g^{-1}. Litaor (1995) described soil distribution of uranium at the RFP. Total uranium background concentration in a control area west of the RFP was 2.3 ± 0.4, and ^{238}U was 1.1 ± 0.2 pCi g^{-1}.

The natural alpha background is the main reason why early RFP monitoring of TLLa activity in soils and vegetation was not a sensitive indication of plutonium releases from the plant. In a background survey conducted in 1951 before site operations began, the TLLa activity in vegetation averaged 86 pCi kg^{-1} with a maximum concentration of 310 pCi kg^{-1}. In 1952, concentrations in both onsite and offsite samples remained within this range.

One approach to assessing releases from the 903 Area before 1970 involved quantitative examination of air monitoring data from air monitoring station S-8, east of the 903 Area on the production area perimeter (Meyer et al. 1996). In order to apportion those TLLa concentrations into specific radionuclides released by Rocky Flats, the background contributions from natural alpha-emitting radionuclides had to be addressed. To interpret TLLa counts in ambient air, we estimated a long-term average natural background concentration of 1.4 fCi m^{-3} (1.1 fCi m^{-3} from polonium-210 and 0.3 fCi m^{-3} from isotopes of thorium, uranium, and radium). It is recognized that this value represents a long-term average estimate. Our judgement was that an individual daily background concentration might be higher, perhaps up to a factor of 5 or more, or around 7–10 fCi m^{-3}. CDH monitoring of TLLa in air in the 1970s resulted in even higher measurements of TLLa in air in offsite locations; the contributing radionuclide was naturally occurring ^{210}Po. This variable and sometimes high natural background of TLLa activity in air seriously limited the use of TLLa activity measurements in offsite air.

Alpha activity measured in water samples collected for the background environmental survey in 1951 and at offsite locations not affected by Rocky Flats discharges in the 1950s and 1960s fell between 1 pCi L^{-1} and 3 pCi L^{-1}, with concentrations up to 6 pCi L^{-1} measured occasionally. During this time period, water from the holding ponds that was discharged to Walnut Creek had alpha activity levels of 10–30 pCi L^{-1}.

How Much of a Total Long-Lived Alpha Measurement is Plutonium?

The answer to this question depends on the environmental medium being analyzed and the time and place the measurement was made. This is an important question for interpreting the TLLa activity concentrations in air near the 903 Area in the late 1960s. At that time and place, virtually all of the TLLa activity in air can be attributed to Rocky Flats materials. Several sources indicated that the plutonium concentration in air is only a fraction of the TLLa activity, even after considering natural background. The apportioning of TLLa activity in daily onsite air samples into specific radionuclides is discussed in Chapter III. Two distinct data sets from the early 1970s suggest that an average of 40% of the TLLa activity in air near the 903 Area in the late 1960s is plutonium. The remainder can be attributed to uranium and americium isotopes.

Comparing the measurements of alpha activity in vegetation samples from the background survey in 1951 with the plutonium in vegetation measurements made in 1957 at distant locations,

the ratio of plutonium to alpha activity is roughly 0.2–0.3 (20–30%). This ratio would vary somewhat over the years. In the background survey, much of the activity was attributed to uranium. Rocky Flats airborne releases of uranium were important in the early 1950s, but plutonium releases dominated after 1956 (Chapter III, this report; Voillequé 1999b).

In general, naturally occurring alpha emitters in soil are present in concentrations greater than plutonium. However, soil samples in highly contaminated near the Rocky Flats industrial area contain plutonium and americium at much higher concentrations than natural levels. In those samples, almost all alpha activity is due to RFP materials such as plutonium and americium.

Comparing the background measurements from 1951 and the offsite samples, the ratio of plutonium to alpha activity in water is quite small, roughly 0.001 to 0.01 (0.1 to 1%). For water samples from onsite locations, the ratio of plutonium to alpha activity is much higher, about 0.35 (35%). This latter value was obtained by correlating alpha and plutonium-specific measurements that were done concurrently from 1970 through 1973 on water samples from Pond B-4 to South Walnut Creek. The actual ratio varied considerably, especially in late 1972 and early 1973 when the ratio was close to 1 due to the pond rebuilding. This ratio of almost 1 indicated that the activity measured in water released from the ponds during reconstruction was predominantly plutonium activity.

How Are Rocky Flats Materials Distributed?

Vegetation

The measurements of plutonium in vegetation made after the 1957 fire are difficult to interpret because they are highly variable and depended on localized conditions that were not included in the environmental transport model. They do provide some general indications of spatial distribution onsite, with highest levels measured just south of Building 771 and decreasing to the west and east of that location. Concentrations of plutonium also show some indication of higher levels at onsite locations to the east than to the west of the site center. This pattern follows the general deposition patterns seen in soils, and reflects the pattern expected from the general meteorology of the area, i.e., winds predominate from the west toward the east and southeast. At the time the fire started, the winds were blowing from the east, however, shortly afterwards, the wind direction at Rocky Flats shifted so that it blew out of the northwest and continued to blow from that direction until the early morning.

The plutonium levels measured offsite are at least ten times lower than the peak values measured onsite. Although these data are useful for discerning spatial trends, the actual number of samples showing increased levels of plutonium was small, thus limiting their usefulness for a quantitative validation of releases from the 1957 or 1969 fires.

Looking at the changes in plutonium activity over time is more difficult because of the limited plutonium analysis in vegetation carried out only after the 1957 and 1969 fires. In general, however, the concentration of plutonium measured in vegetation was markedly higher at several locations onsite in 1969 compared to 1957. The plutonium analysis in 1969 alerted site personnel to the very high levels east of the 903 Area, adding support to that location being the source of the highest airborne releases of plutonium.

For alpha activity in vegetation, the levels of alpha activity remained at nearly background levels at locations sampled just to the west of the RFP during the 1950s and 1960s. Alpha activity in vegetation drops off fairly rapidly with distance from the security fence to the east.

Air

Review of the ambient air monitoring data strongly suggests that the 903 Area was the primary source of plutonium contamination near the RFP. Concentrations of plutonium or TLLa activity in air decrease rapidly with distance from that area. After the 903 Pad was covered with asphalt in 1969, contaminated soil areas within several hundred meters were the primary sources of airborne activity. Soil from some of those areas was removed in the mid-1970s; however, some contamination remained.

A rapid decrease in plutonium in air with distance from the RFP was clear from monitoring of plutonium in air by the HASL in the 1970s. There was a decrease of about 3 times in the first 0.5 km east of the security fence, and a further 10-fold decrease in the next 1.8 km to Indiana Street. Combined, this made a 30-fold decrease in the concentration in a 2.3-km distance east of the production area security fence. The concentrations at Indiana Street in 1974–1976 were roughly twice the fallout plutonium concentrations measured by HASL in air in New York City and Helena, Montana, during the same time period.

The net (above fallout background) concentration of plutonium in air at Indiana Street averaged 3% of that measured at the RFP security fence in the 1970s. The net concentration of plutonium in lichens (slow-growing plants which trap small atmospheric particles) near Indiana Street averaged 1% of the concentration measured in lichens near the security fence. The lichens were present during the time of highest releases from the RFP.

Deposition (gummed paper)

The measurements of plutonium deposited from air during the years 1963–1965 show a definite spatial pattern on and near the RFP, with highest quantities measured near the Building 71 stack and in a downwind (east-southeast) direction from the stack (see Figure IV-12). A maximum deposition of 1600 pCi m^{-2} during 1963 and 1964 was estimated at 0.4 km east of the stack. This spatial pattern clearly implicated the Building 71 stack as the major source of plutonium deposition at the RFP during this time period.

There are no discernible spatial patterns in the deposition measurements made during 1970–1972. This is primarily due to the fact that there were only five onsite stations. Offsite measurements were not statistically different from onsite locations, with two exceptions. Data obtained from collectors located near the solar evaporation ponds and the 903 Area were higher than offsite. Local resuspension of contaminated soil is implicated in both cases. More detailed analysis of the offsite data suggested that the RFP had no measurable impact during 1970–1972.

Surface Water

Both the alpha activity in water measured in the early years and the plutonium concentrations measured after 1970 showed higher levels in samples from onsite locations compared to offsite. Tritium and plutonium measurements in water from Walnut Creek at Indiana

Street confirmed that significant releases of materials occurred from the holding ponds during pond reconstruction in the early 1970s.

Routine surface water monitoring data from Walnut Creek at Indiana Street alerted the site to an accidental tritium release in 1973. The weekly tritium concentrations measured in the Great Western Reservoir from that time period were used to estimate doses to the public from ingestion of drinking water from the Broomfield water supply.

Sediment

Rocky Flats plutonium is distributed in definite patterns in sediment, both horizontally and vertically. Surface measurements of ^{239,240}Pu in the Great Western Reservoir sediment demonstrate that most of the contamination currently is located in the deeper eastern portion of the reservoir. A study by CSU in 1992 indicated that over half of the ^{239,240}Pu inventories in the Great Western Reservoir was in the bottom sediments of the deepest parts of the basin. Inventory estimates ranged from 54 mCi (by CSU, based on 48 samples) to 244 mCi (by Pacific Northwest Laboratories [PNL], based on nine samples) for the Great Western Reservoir (see Table VII-5). The PNL estimate is very conservative and should be considered the upper bound. In 1992, CSU estimated that 87% of the plutonium in the Great Western Reservoir originated from Walnut Creek, which drains the northern portion of the RFP, where the holding ponds are located.

Clear vertical patterns of concentrations of ^{239,240}Pu were observed in many cores collected from the Great Western Reservoir and Standley Lake. These core profiles reflect a time history of deposition rate and are discussed in Chapter VII.

Soil

At and around Rocky Flats, plutonium distribution in soil is a general arc in an easterly direction from the 903 Area and falls off quickly in concentration out to at least 19 km. There are nuances to this pattern, however, and there is dispute over the existence of a plume tending southeast from the 903 Area. There is rapid falloff to the north, south and west of the 903 Area. Migration of plutonium into the soil has been demonstrated to at least 21 cm, but most is contained in the upper few cm. Approximately 60% to 70% of the total plutonium inventory is contained in the top 5 cm of soil.

How Did Rocky Flats Releases Change with Time?

Air

Review of the ambient air monitoring data strongly suggested that the 903 Area was the primary source of localized TLLa and Pu contamination. The time trend in TLLa activity in air near the 903 Area showed definite high concentrations in late 1968 and 1969 when mechanical disturbances combined with high winds suspended contaminated soil. Impact of the May 1969 fire was not noticeable above the greater effect of suspension of contamination from the 903 Area.

In the 1970s, weathering of plutonium in remaining contaminated soil areas as well as removal of some contaminated soil produced a gradual decline in plutonium concentrations in air. The concentration at the HASL station at the RFP production area fence in the first half of 1971 (6.5 fCi m^{-3}) was over 10 times that in the first half of 1981 (0.5 fCi m^{-3}), 10 years later.

Soil

Most of the excess plutonium in soils in the vicinity of the RFP is likely to have originated from the 903 Area, as a result of releases between 1965 and 1970. This is despite the fact that the largest offsite releases were probably from the 1957 fire. Particle sizes for the 1957 releases were smaller than 903 Area releases, resulting in lower gravitational settling velocities and deposition. Also, winds were relatively light during the 1957 fire compared to the 903 Area releases during high wind events, and resulted in lower deposition velocities. Finally, the 1957 fire releases were from an elevated plume compared to ground-level releases for the 903 Area. The 1969 fire and routine releases made lesser contributions.

Following the paving of the 903 Pad, there were contaminated soil areas remaining near the pad that were sources for suspension of smaller amounts of Rocky Flats materials.

Vegetation

The vegetation data clearly record the deposition patterns that resulted from key release events onsite, specifically the higher releases of plutonium that were occurring with the increased activity at the 903 Area. The highest TLLa levels measured in vegetation from the RFP area were those measured in 1969 just east of the 903 Area (up to 30,000 pCi kg^{-1}), and coincide with the re-drumming and plowing activities at the 903 Area.

This change in activity over time is also seen at locations near the solar evaporation ponds. The highest TLLa levels just east of the solar evaporation ponds, (1200–1500 pCi kg^{-1}) were measured in 1962 and in 1970, and correspond to times of repairs and modifications being made to the ponds.

Deposition (gummed paper)

There are not enough data to clearly assess temporal patterns of deposition of Rocky Flats releases. However, a comparison of deposition rates measured at a common onsite area during the 1963–1965 and 1970–1972 periods demonstrates that the Building 71 stack releases were relatively greater during the earlier period. This conclusion is supported by the observation that deposition during 1970–1972 appears to be influenced more by localized resuspension of contaminated soil than by stack releases.

Sediment

Core profiles of ^{239,240}Pu in the Great Western Reservoir clearly show a peak concentration at a time corresponding to the 903 Pad area release (1969–1970). There is also some evidence that this peak extends into the early 1970s and may include the aquatic releases from the RFP. The maximum concentrations of ^{239,240}Pu in core samples collected by various researchers range

from 4 to 8 pCi g^{-1}. Concentrations in other sections of the core samples are generally greater than background levels (i.e., >0.2 pCi g^{-1}) before and after the peak concentration until more recent deposits corresponding to the early 1980s. This indicates that liquid, as well as airborne, releases contributed to the contamination in Great Western Reservoir sediments.

Core profiles of ^{239,240}Pu in Standley Lake also show a peak concentration at the time corresponding to the 903 Pad area release (1969–1970). The maximum concentrations (0.2-0.6 pCi g^{-1}) were far less than those measured in the Great Western Reservoir sediments. In addition, concentrations prior to and after the peak were generally within those measured at background locations. These core profiles were one of the environmental data sets used by Rood and Grogan (1999a) to compare to predicted concentrations in sediment resulting from plutonium releases from Rocky Flats.

Water

The gross alpha measurements in water before 1970 correspond fairly well to release events onsite, and the data are useful for supporting the relative magnitude of routine releases to surface water. For example, alpha activity in water samples taken from North and South Walnut Creeks dropped when the holding ponds came on-line. The historical monitoring data clearly show the effectiveness of the holding ponds in decreasing the levels of activity released to offsite streams.

Measurements of plutonium in water from the final holding ponds onsite reveal that the concentrations of plutonium discharged from the site decreased dramatically, from 15,000 fCi L^{-1} (15 pCi L^{-1}) in 1972 to 5 fCi L^{-1} (0.005 pCi L^{-1}) in 1989.

We used a method to estimate plutonium releases for years when only gross alpha activity was measured, by calculating a ratio of monthly plutonium to alpha activity and applying it to measurements of alpha activity from 1952 to 1970 (when plutonium was not measured).

What Do Environmental Data Tell Us About Releases of Nonradioactive Contaminants of Concern?

The principal contaminants of concern from past RFP operations identified in Phase I were isotopes of the radioactive element plutonium and the nonradioactive chemical carbon tetrachloride. Possible exposures and risks of another contaminant of concern, beryllium, were also evaluated further in Phase II (McGavran and Rood 1999).

Historically, there was very little environmental monitoring for nonradioactive contaminants of concern at the RFP. Some special studies of beryllium distribution in soils were located. Also, beryllium was routinely monitored in ambient air by the RFP contractor between 1970 and 1976.

We developed an approximate range for the natural background concentration of beryllium in air in the Rocky Flats area, 3×10^{-5} μg m^{-3} to 3×10^{-4} μg m^{-3}, with a median of 1×10^{-4} μg m^{-3}. This background range was used for perspective when examining the historical environmental monitoring data for beryllium in air. Concentrations of beryllium predicted in offsite air from historical Rocky Flats releases are well below natural background concentrations.

In general, enhanced concentrations of beryllium in soil due to Rocky Flats airborne releases are not evident. One elevated area of beryllium in soil was located within 30 m of the stack of an RFP building that processed beryllium. Average concentrations of beryllium in onsite air are

very similar to offsite and are close to the detection level. However, there is a strong suggestion that the maximum concentrations in onsite air reflect Rocky Flats operations. Both maximum and average concentrations in onsite air were unrelated to the amounts released via Rocky Flats stacks. It is likely that resuspension of contaminated soil onsite influenced the maximum concentrations observed in onsite air.

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