FINAL REPORT

Development of the Rocky Flats Plant 903 Area Plutonium Source Term

Task 2: Independent Analysis of Exposure, Dose, and Health Risk to Offsite Individuals

August 1999

"Setting the standard in environmental health"

RAC

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EXECUTIVE SUMMARY

OBJECTIVES OF THE PROJECT AND THIS REPORT

The Rocky Flats Plant (RFP) is a 6500-acre site located within a few kilometers of the cities of Arvada, Westminster, and Broomfield, Colorado and about 26 km northwest of downtown Denver. The site served as a research, development, and production location for the nuclear weapons complex, with primary production focused on plutonium weapons components. Primary releases from the RFP consisted of plutonium to air through a number of discrete events. *Radiological Assessments Corporation (RAC)*[a] was hired by the Colorado Department of Public Health and Environment as the Phase II contractor for the Historical Public Exposure Studies at Rocky Flats. In Phase II, *RAC* has calculated source term, dose, and risk to the public from releases of plutonium from the RFP identified during Phase I. This report documents the releases of plutonium from a specific source at the site, an outdoor waste storage area known as the 903 Area.

RELEASES OF PLUTONIUM TO THE ENVIRONMENT

The 903 Area served as a waste storage area for barrels of plutonium-contaminated oil during the late 1950s and 1960s. This oil was contaminated with plutonium during its use at the plant to lubricate the plutonium-cutting tools. In addition to the plutonium and oil, the barrels also contained carbon tetrachloride that had been used as an agent to clean the oil from the tools. This combination of contaminants precluded its storage in traditional waste disposal locations at that time. With no other disposal options, the barrels were stored outside on a grassy area to the east of the RFP.

As early as 1962, evidence of barrel corrosion and subsequent leakage onto the soil was detected by site personnel. The area became a primary source of offsite contamination during the period of time after the barrels were removed and sent to the Idaho National Engineering Laboratory for disposal and before the area was covered with asphalt. The potential offsite hazard resulted from suspension of plutonium attached to soil particles from the 903 Area during high wind events.

Methodology

Release estimates were determined by calibrating a wind speed-dependent soil suspension model to air monitoring data taken primarily at the S-8 sampler, which was located about 200 m east from the center of the 903 Area. Deficiencies in air monitoring data, including particle size sampling efficiency and air volume reductions because of filter clogging, were accounted for in the calculation. The soil suspension model was linked to the corrected measured air concentrations at the S-8 sampler using an atmospheric transport model capable of addressing dry deposition and gravitational settling. Meteorological data were obtained from the National Centers for Atmospheric Research from portable anemometer stations used in the Boulder and Rocky Flats areas during the late 1960s. These data were compared to Rocky Flats hourly wind

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[a] *Radiological Assessments Corporation* changed its name to *Risk Assessment Corporation* in 1998.

data, proved to be a good match, and were substituted for missing continuous wind data from the site.

Release quantities were estimated daily for the 24 days that had the highest activity recorded at the S-8 sampler and averaged annually for all remaining days between the years 1964 and 1970. Results were obtained using information available from Rocky Flats and the surrounding areas. This information was incorporated into a source and transport model using downwind concentrations of plutonium as a starting point, using all available wind and weather information, and calculating plutonium releases that would result in the given downwind plutonium concentrations. This methodology ensured the use of all available data for calculating the 903 Area source term.

Release Estimates

The median release estimate of plutonium from the 903 Area for high wind periods during which the source was open and available for particle suspension activity was 3.1 Ci $^{239+240}$Pu, with 5th and 95th percentile values of the distribution of 1.4 and 15 Ci, respectively. The results indicated that 24 dates with high wind events throughout the period from 1968–1969 were responsible for this plutonium release. These releases were specific to certain days and have been summed here merely for ease of presentation. Additionally, continuous baseline suspension of plutonium during the years from 1964–1970 resulted in a source term with a median release estimate of 0.14 Ci and 5th and 95th percentile values of 0.025 and 0.79 Ci, respectively. The continuous releases are much smaller than day-specific releases because of the large influence of wind speed on the release of contamination from the soil area.

After 1970, suspension at the 903 Area was limited by placing an asphalt pad over the contaminated soil. This action inhibited further suspension of contamination from the source areas (fields to the east of the 903 Area that had been contaminated with plutonium during the suspension period), but the fields continued to act as a secondary source for resuspension of plutonium. This secondary source of contamination has been the subject of a number of research studies. This report discusses those studies and other information (i.e., plutonium concentrations in air after 1970) supporting the development of a source term for this area. A source term for resuspension of plutonium is not calculated in this report because the results of the 903 Area east field resuspension studies after 1970 were not comparable and ingestion of this material is expected to be a potential pathway. The cancer incidence risk resulting from this source of plutonium from resuspension after 1970 will be treated as a location-specific source instead of a time-specific source. The total risk from the 903 Area will be reported into the subsequent risk report released as a part of this study.

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ACRONYMS

AEC   U.S. Atomic Energy Commission
AED   aerodynamic diameter

CDPHE Colorado Department of Public Health and Environment
cfm   cubic feet per minute
cpm   counts per minute
CV   coefficient of variance

DOE   U.S. Department of Energy

EPA   U.S. Environmental Protection Agency

FDM   Fugitive Dust Model

GM   geometric mean
GSD   geometric standard deviation

HASL   Health and Safety Laboratory
HPSR   Health Physics Site Report

INEL   Idaho National Engineering Laboratory

NCAR   National Centers for Atmospheric Research
NOAA   National Oceanographic and Atmospheric Administration

P/O   predicted to observed

RAC   *Radiological Assessments Corporation*
RFP   Rocky Flats Plant

STAR   STAbility ARray

TLLa   total long-lived alpha

CHAPTER I

INTRODUCTION

The Rocky Flats Environmental Technology Site is owned by the U.S. Department of Energy (DOE) and is currently operated by Kaiser-Hill Company. For most of its history, the site was called the Rocky Flats Plant (RFP) and was operated by Dow Chemical Company as a nuclear weapons research, development, and production complex (Figure I-1). The RFP is located on approximately 6500 acres of Federal property, about 8–10 km from the cities of Arvada, Westminster, and Broomfield, Colorado, and 26 km northwest of downtown Denver, Colorado. The original 385-acre main production area is surrounded by a 6150-acre buffer zone that now delineates the RFP boundary.

Through a 1989 Agreement in Principle between the DOE and the State of Colorado, DOE provided the State with funding and technical support for health-related studies. The purpose of the Historical Public Exposures Studies on Rocky Flats is to identify potential health effects to residents in nearby communities who may have been exposed to past toxic and radioactive releases. The Colorado Department of Public Health and Environment (CDPHE) first invited a national panel of experts to help design the health studies. Because of intense public concern about Rocky Flats contamination among Denver metropolitan area residents following a Federal Bureau of Investigation raid of Rocky Flats in June 1989, the panel decided to stress public involvement and to separate the research into two major phases conducted by two different independent contractors to enhance credibility.
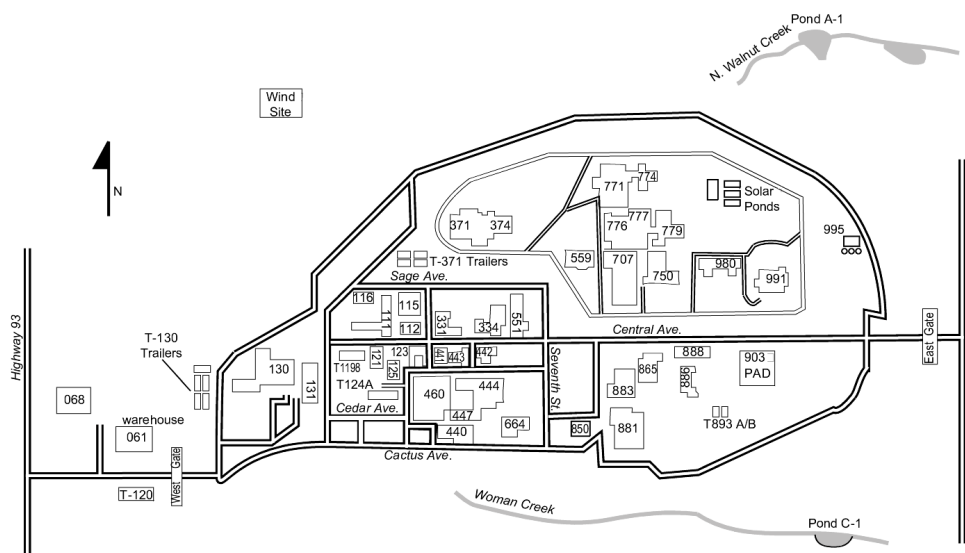
Figure I-1. Main production area of the Rocky Flats Plant. Originally, the buildings were identified with two-digit numbers. Later, a third digit was added. The production area, now sometimes called the industrial area, is surrounded by a security perimeter fence. The area between the perimeter fence and Indiana Street to the east is the buffer zone. The buffer zone was expanded to Indiana Street in the 1970s (map is not to scale).

Phase I of the study was performed by ChemRisk (a division of McLaren/Hart Environmental Engineering). In Phase I, ChemRisk conducted an extensive investigation of past operations and releases from the RFP. The Phase I effort identified the primary materials of concern, release points and events, quantities released, transport pathways, and preliminary estimates of dose and risk to offsite individuals. The conclusions from Phase I were released in a public summary document (ChemRisk 1994), a series of task reports by ChemRisk, and several articles in the journal *Health Physics*.

Radiological Assessments Corporation (*RAC*) was awarded the contract to conduct Phase II of the study, which is an in-depth investigation of the potential doses and cancer risks to the public from historical releases from the RFP. Recommendations for work to be performed in Phase II are outlined in the Phase I summary document (ChemRisk 1994).

This report documents one source of contamination at the RFP that may have led to public exposures—an outside waste storage area, commonly referred to as the 903 Area.

CHAPTER II

BACKGROUND AND HISTORY

The 903 Area, located near the eastern edge of the RFP, was the primary storage area for steel barrels containing ^{239}Pu[b]-contaminated waste oil from the mid-1950s to the late 1960s. This oil was used as a lubricant for plutonium cutting tools in the industrial area and was cleaned from the tools and plutonium parts using a chemical solvent. After long-term storage, chemical reactions of this solvent corroded many barrels, allowing plutonium-contaminated oil to leak onto the ground. In the late 1960s, the drums were removed from the 903 Area, but the contaminated soil was left uncovered and unprotected against wind erosion. High wind events moved contaminated soil particles into the air, creating a source of contamination from the 903 Area. This source of contamination is called the source term, which defines the quantity, chemical and physical form, and the time history of the material released to the environment from the 903 Area. This portion of the Phase II study provides estimates of the amount, form, and timing of the releases that define this source term.

The objectives of this report are to detail the assumptions and calculations that led to the 903 Area source term estimates and to summarize those results for future portions of this study. The calculations leading to estimates of the 903 Area source term were separated into two time periods: from 1964 through 1969 and from 1970 through 1989. These two time periods are defined by two separate mechanisms that controlled the movement of soil. From 1964–1969, wind erosion was the primary mechanism causing soil movement. This period is called the initial suspension period throughout this report; it is the interval during which contaminated soil was first moved off the 903 Area and into the air or onto the field adjoining the area. During this time, no vegetation or other erosion controls were in place to limit soil movement. For the remainder of the study period, from 1970–1989, the contaminated area was covered with an asphalt cap. The only remaining source of air contamination during these later years resulted from resuspension off grassy areas where material had previously been deposited.

The terms used above, suspension and resuspension, have a specific meaning to specialists in particle movement. *Suspension* describes the wind-driven movement of particles from the original soil surface. *Resuspension* describes wind-driven movement of particles from a surface contaminated by suspension. Figure II-1 illustrates the movement of contamination by suspension and resuspension.

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[b] Plutonium-239. Common analytical methods cannot distinguish ^{239}Pu from ^{240}Pu. Unless otherwise indicated, ^{239}Pu implicitly includes both isotopic forms.
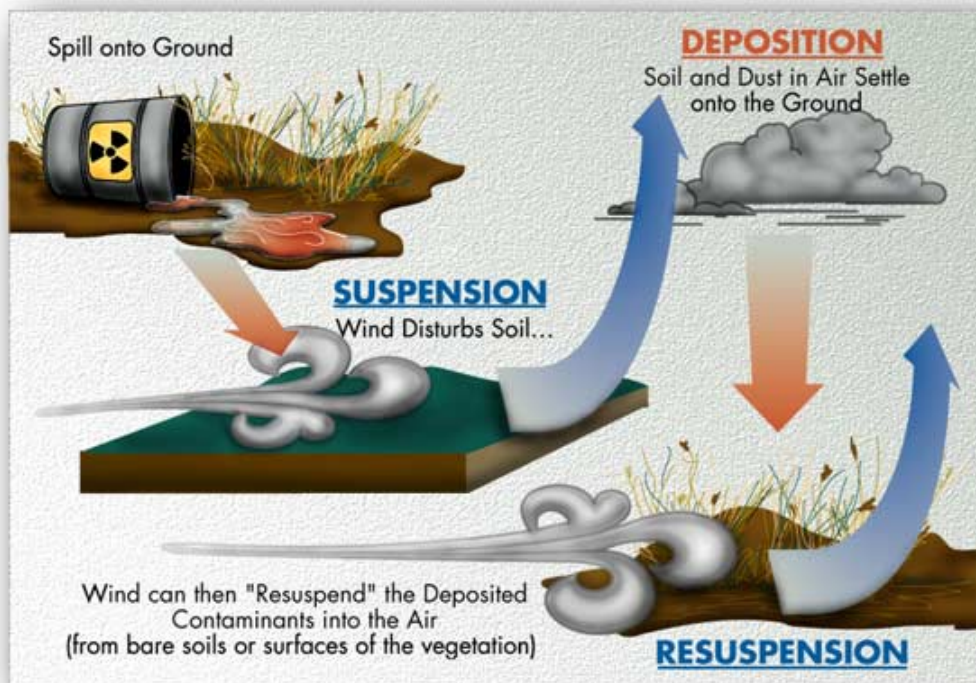
Figure II-1. The processes of suspension, deposition, and resuspension are separate events. This figure illustrates contamination by some nonatmospheric process, such as a spill. Following a spill, winds can suspend the contaminated soil into the air. This atmospherically transported material is then deposited onto the ground and may be resuspended into the air by wind and weather processes.

For the 903 Area, the time period defines the difference between these two types of particle movement. The initial contamination of the 903 Area was caused by barrel leakage, a nonatmospheric process. During the 1964–1969 time period, contaminated soil moved off the area and into the air stream by *initial suspension* events. By 1970, the major source of contamination was covered with an asphalt pad. Then the east field, which had been contaminated by deposition from 903 Area *initial suspension*, became the source of contamination. Winds, rain, and other forces acting on this area caused *resuspension* to occur.

Releases before 1970 were estimated using *suspension* modeling. After 1970, releases resulting from 903 Area contamination of adjacent ground surfaces before the 903 Area was capped were assessed using the results of *resuspension* research conducted at the Rocky Flats 903 Area during the 1970s and 1980s and environmental contamination data collected during this same period.

HISTORY OF THE 903 AREA

Because the time history of the facility is important to a meaningful interpretation of the available information, we provide a brief, chronological history of Rocky Flats Plant operations associated with the 903 Area releases (Meyer et al. 1996). Details and references are provided in the discussion that follows this chronology.

Chronology of 903 Area Events

1954   Approximately 10 barrels, or drums, of oily liquid wastes from RFP were sent to the Arco, Idaho, waste storage facility.

4/54   Further disposal at Arco was refused; the Mound area (near 903 Area, but separate) was created onsite at the RFP for drum burial. Nine hundred drums containing uranium-contaminated wastes were in place in the Mound area by early 1954.

8/55   The first potentially plutonium-contaminated drums were placed in the Mound area.

8/56   Uranium-contaminated oil from 10 drums was burned in RFP Pit 3.

4/57   Uranium-contaminated oil from 169 drums was burned in a pit next to the Mound area. Additional, intermittent burning took place from 6/61 until 5/65. Estimates indicate nearly 1100 drums of uranium-contaminated oil were burned during the entire period.

1957   Building 776 began plutonium operations; quantities of plutonium-contaminated oil and cleaning solvents increased significantly. Most liquids were straight-chain hydrocarbon mineral oils, some hydraulic oils, vacuum pump oil, carbon tetrachloride, trichloroethylene, perchloroethylene, silicone oils, acetone, and still bottoms. Centrifuging was initially used to separate the bulk of plutonium from waste oil. Barrel release limits were 10^{-2} to 10^{-3} g plutonium per liter. Fifty-five gallon barrels contained about 50 gal of centrifuged oil. Thus, a first estimate of the quantity of plutonium contained in each barrel ranges from 0.2 to 2 g.

1958   First reports of stored drum corrosion.

9/58   One thousand four hundred and five drums, including 89 containing perhaps 300 g of plutonium total, were in storage in the Mound area as of this date.

7/59   Ethanolamine was added to drums to neutralize acids and reduce internal corrosion.

7/59   Certain drums from the Mound area were moved across the road to the 903 Area, marking the beginning of 903 Area drum accumulation. (The Mound area was excavated in 1970. No detectable plutonium was reported, although recent remediation programs have identified potential contamination; 1400–1600 barrels were removed.)

1962   Budget was submitted to research early waste solidification; the justification was that 50–60% of the 903 Area drums were corroded and that the contents were spilling onto the ground.

6/62   Date of photograph of a drum turned on its side, showing weeds penetrating the bottom of the barrel where corrosion had occurred. The date of another photo of a drum showed possible distortion from corrosive gases. It was mentioned that there was an odor of carbon tetrachloride.

1964   E. Putzier notes the "first indication" that drums in 903 Area were leaking significantly. Earlier records, noted above, indicated that corrosion was noticed as early as 1958 in the Mound area, and leakage was noted by 1962. Health Physics Department records showed "soil contamination increases" in the 903 Area. A rabbit fence was constructed to prevent the spread of contamination by wildlife.

7/64   "First evidence of large-scale deterioration" of drums; drums were packed tightly in the 903 Area, making inspection of interior drums impossible.

Mid-
1960s   New filtering devices were added to the plutonium machining lines, capturing smaller plutonium particles and "greatly reducing" contamination levels in waste oil.

4/65   Date of photo showing near-maximum number of drums in the 903 Area.

1967   "Highly plutonium contaminated" rabbits were reported on RFP property.

1/67   903 Area clearance operations began. The work lasted until mid-1968 and was hampered by bad weather and other problems. Reports that many barrels were rusted and contained little liquid; filtering of oils in place failed. Sludges were left in the drained drums and shipped.

4/67   Date of photo showing 903 Area still at near-maximum barrel loading.

4/68   Photo showing most barrels removed.

6/68   Last drums were transferred out of 903 Area. Over 5200 drums were reported as removed; one report notes a total of 3572 drums of plutonium-contaminated oil removed. "Difference *(in total number of drums)* attributed to leakage of oil into the soil and some of the drums were not full when first transferred to the field..." About 265,430 gal of oil were transferred to Building 774 for processing; of that volume, 196,460 gal were plutonium-contaminated.

6/68   Health Physics Department report noted that "some slight contamination" was spread from 903 Area by wind and rain. 903 Area oil transfers were completed. It was reported that packaging for shipment of the empty (sludge-containing) drums and pallets "should be completed by June 17."

6/11/68   Leaking solvent drum being transported from 903 Area to Building 774 caused "significant contamination of roadway." A 7.6-m grid was staked out over the contaminated 903 Area (a 168 × 145-m rectangle) and soil radioactivity was checked at the grid intersections. Levels of contamination from less than 100 counts per minute (cpm) to over 1,000,000 cpm were found.

7/68   Memo from Mr. Owen noting that plutonium contamination within the rabbit fence was "extremely hazardous"; and that wind and water had carried contamination outside the area. A Health Physics Department survey of the area was completed and forwarded to Division Services, Manufacturing and Facilities.

10/68   Weeds and vegetation were burned off the 903 Area. Dow requested approval to pave the area to contain contamination.

11/68   Second request to pave area was submitted to management because contamination
              was being spread by wind and rain. Use of a road grader to scrape and cover some
              contamination into a relatively small area was allowed.

12/68   Health Physics Department report noted, "...high winds have blown over as many as
              150 of the 5000 drums east of the nitrate ponds." Details concerning the contents or
              more exact location or identity of these barrels are unknown.

1/69   Health Physics Department reported, "The high winds of January 31 blew all the
              roofing material off Building 889." Further spread of contamination from the 903
              Area was shown by area surveys and perimeter air samples east of the area. Notation
              that, "…29 leaking drums have been detected in the drum storage area east of the
              nitrate ponds. These leakers have resulted in high level contamination of about 200
              square feet of soil." The soil was removed and shipped to Idaho.

1969   Late March to early April, leveling the 903 Area by grading. The mechanically faulty
              grader ran at high speed in third gear; dust was visible as far as 0.8 km past the
              perimeter security fence, the cloud was approximately 3–4.6 m high. Grading
              continued for 2 to 3 days, 5 to 6 hours per day. The soil was dry and not snow-
              covered. There were light breezes. Putzier noted that the road grader was used outside
              the fenced area to move slightly contaminated soil to the more contaminated, fenced
              area.

4/69   "...large rocks and stakes have been removed from the 903 drum storage area
              preparatory to the filling and leveling scheduled to start soon." An interview notes
              that the rock cleanup took a "couple of days." Oily rocks were visible, alpha counts
              were as high as 800,000 cpm.

5/69   Thirty-three drums of contaminated rocks were removed from 903 Area. The main
              contamination concern was the "south edge" of the area. "…filling, leveling, and
              capping of the 903 Area have not yet started." Cleanup of the road grader was
              completed.

5/24/69   Date of photo showing 903 Area cleared and ready for placement of cover.

7/69   First coat of fill was applied to the 903 Area on July 23. Fifteen centimeters of road
              base was applied.

9/69   According to the Health Physics Department report, "...base fill has been applied to
              the 903 Area and the area has been rolled." The asphalt prime coat was reported to be
              in place.

11/69   Asphalt work was completed; sampling wells at four corners of the asphalt pad were
              finished on November 11. The asphalt-covered area was 116×122 m.

2/70   Remaining 903 Area was covered with 10 cm of road base.

5/70   Photo shows 903 Area and gray, road base area.

5/25/71   Photo showing three gray rectangles, evidence of dust suppressant tests, using a
              material called Coherex.

   Following is a more detailed discussion of the above events and other occurrences
contributing to 903 Area releases.

Early Production and Handling of Waste

Buildings 44 and 81, the first production buildings operating at the RFP, were involved in the machining of depleted uranium (i.e., having a reduced fractional abundance of the isotope ^{235}U and corresponding greater abundance of the isotope ^{238}U compared to naturally occurring uranium) and produced oils or oil/water wastes that were defined as radioactive. Even in early 1954, it was difficult to find a disposal site for these radioactively contaminated oily liquids, although approximately 10 barrels were shipped for burial in Idaho. Because of potential leakage problems, Arco later refused to accept such organic liquids for burial (Putzier 1970a).

Creation of Plutonium-contaminated Wastes

During the late 1950s, changes in weapons designs required using metal lathes that used oil as a cutting lubricant and left plutonium-contaminated cutting oil as a by-product. Increasing quantities of carbon tetrachloride were used to clean the plutonium weapons components produced (Barrick 1980). The quantities of contaminated oil and cleaner became significant after Building 776 was opened in 1957. Most of the contaminated liquid was straight-chain hydrocarbon mineral oil (Shell Vitrea), but hydraulic oils, vacuum pump oil, trichloroethylene, perchloroethylene, silicone oils, acetone, still bottoms,[c] and other liquids were also present (Seed et al. 1971). There were no records kept of the specific contents of each barrel. Centrifuging was initially used to separate the bulk of the plutonium turnings from the waste oil, with release limits to barrel storage set at between 0.01 and 0.001 g plutonium per liter (Seed et al. 1971). About 50 gal of centrifuged oil were placed in each 55-gal drum. These figures led to a first estimate of between 0.2 and 2 g of plutonium waste per drum. There were no long-term waste disposal methods available and no good alternatives to storage in barrels.

Burning Uranium-contaminated Oils

Putzier (1970b) stated that uranium-contaminated oil from 10 drums was burned in a pit onsite in August 1956. In March and April 1957, oil from another 169 drums was burned at night, and a portable sampler was reportedly used by health physics workers to monitor air concentrations. Although Health Physics logbooks of air concentration data were said to be maintained, these have not been located. No further burning was reported until June 1961, "after which time oils were burned frequently" (Putzier 1970b). The oil burning continued through May 1965 with over 1000 drums of uranium-contaminated oil burned (Putzier 1970b). An unknown number were 30-gal drums; the remainder were 55-gal barrels.

Creation of Early Soil-Covered RFP Storage–the Mound Area

It is probable that the early loss of access to the Arco burial site led to the creation of the Mound area at Rocky Flats. The Mound area contained solid and liquid uranium wastes in drums buried under several feet of soil (Putzier 1970b). Biles (1970) reported about 900 drums in place

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[c] The term still bottoms refers to residual solids/liquids from a distillation process.

by early 1954. Records from the Waste Disposal Coordination Group indicate that the first drums containing plutonium-contaminated oil were placed in the Mound area in August 1955; however, these may have been hydraulic oils from machinery in plutonium processing areas and not significantly contaminated. Over 1400 55-gal drums were buried in the Mound area through September 1958 (Biles 1970). Biles also stated that twelve, 30-gal drums containing plutonium-contaminated waste were placed in the Mound area. Putzier (1970b) stated that 89 plutonium-contaminated oil drums from Building 776, and 46 from Building 771 were in the Mound area by September 1958. After this time, additional drums were placed at the Mound area but not buried, and they were moved across the road to the 903 Area in July 1959 (Putzier 1970b).

Excavation of the Mound Area

The Mound area was excavated in 1970, and oil-containing barrels were processed in Building 774. According to the records available, no detectable plutonium contamination was found in these barrels (Biles 1970). Of the 1400–1600 barrels placed in the Mound area, up to 1500 contained organic chemicals, with 135 of those containing oil from the plutonium facility (Putzier 1970b). Reports indicated that many barrels were corroding in 1958 (Bronesky 1977).

Development of the 903 Barrel Storage Area

After several interim waste-handling methods were used in the early years, the waste barrels began to accumulate "in the SW corner" of the 903 Area in mid-1959 (Biles 1970; Putzier 1970b). There were suggestions that drum storage at the 903 Area began in the mid-1950's, but photographic evidence from mid-1955 is not conclusive. A series of photographs referenced in the text of this report have been included as Appendix A. Photographs from 1959 show two parallel sets of barrel rows in the 903 Area on July 9, 1959. Most of the drums transferred to the 903 Area were 55-gal capacity; some were 30-gal drums (Seed et al. 1971). Not all the drums were full, and perhaps 75% contained plutonium-contaminated oils (Seed et al. 1971). In July 1959, ethanolamine was first added to the barrel oils to reduce internal corrosion (Seed et al. 1971).

A 1962 waste solidification research budget request noted that 50–60% of these drums was corroded, and that the contents were spilling onto the ground (Bronesky 1977). An aerial photo taken August 11, 1962, showed a large number of waste oil barrels tightly packed in the 903 Area. A June 1962 photo shows a drum, tipped on its side for viewing, with weeds penetrating a heavily corroded area on its bottom. Delays in developing solidification technology also resulted from the production emphasis stressed by upper management: work on waste product treatment was viewed as nonproductive.

Leakage from the drums was noted as early as July 1959, although Putzier (1970b) states that "1964 was the first indication that drums were leaking in the field." Funding to construct a building to store the barrels was requested early on, but it was not approved in the site budget. Efforts to develop solidification alternatives for the contaminated oil were also hampered by budget problems as well as technical difficulties including corrosion of stainless steel components (Dow 1974).

Ken Freiburg, a Health Physics Department technician, stated that the closely spaced, stored barrels were corroding from the inside out, and that they were bulging and popping in the

sunlight (Bronesky 1977). A June 1962 photo shows such a distorted drum. Freiburg noted the odor of carbon tetrachloride in the area (Bronesky 1977). Even though leakage and contamination were occurring, Freiburg noted that the (then-current) standards for radioactivity in air "were not being violated," so it was difficult for health physics staff to convince RFP management that a significant problem existed. Atomic Energy Commission (AEC) staff were aware of the contamination, but it does not appear that any serious effort to deal with the problem was initiated at the Agency level.

It proved difficult to deal with leaking drums except individually. Putzier reported that once, when a routine site survey showed contamination from leaky drums, uncontaminated soil was brought in to cover that area. One former employee stated that oil from a leaky drum was once used to suppress dust near the 903 Area.

Evidence Concerning Barrel Storage Conditions

Drum corrosion and leakage proceeded rapidly during the early 1960s. By 1964, the Health Physics Department's Site Survey Group was reporting soil contamination increases in the 903 Area in weekly reports. In July 1964, evidence of large-scale deterioration of drums was reported. Photographs from 1963–1965 show the area of stored barrels increasing from May 1963 through March 1964 to April 1965.

Many of the drums were packed so tightly that inspecting barrels located toward the interior of the group was not feasible. A retired health physics manager interviewed by *RAC* stated that, "they tried to keep an eye out for leakers," but that they could not see into the tightly packed pile. An April 1965 photograph views the problem from ground level. A perspective of the Plant taken from the west is provided in a January 6, 1966, photograph. The 903 Area is just visible above the twin-stack building toward the lower right side of the photo. Great Western Reservoir is visible in the distance, near the photo's top right corner.

Wildlife contributed to the spread of contamination. One of the health physics personnel, Joe Ferrerese, reported the spread of contamination by rabbits in the early 1960s. A rabbit fence was constructed during 1964. Measurements of killed rabbits showed "grossly" contaminated fur, confirmed recently during a *RAC* interview with a union representative who worked at the 903 Area during the period. Dale Bokowski noted that he and another Rocky Flats worker had trapped two or three live rabbits from around the barrel storage area in 1967. They analyzed lungs, long bones, and livers, with results that were "screaming" with ^{239}Pu. He believes that the confirming data may have been lost.

Influence of Wind and Rain

Ferrerese recalled that winds occasionally caused the contaminated soil to become airborne. When this occurred, he halted sampling work in the area. He and another monitor discovered contaminated dirt from the 903 Area at the Rocky Flats site perimeter (Bronesky 1977). A worker involved in the eventual barrel removal noted that, "one evening a dust devil came from the east" and caused measurable contamination on a guard car. Litaor et al. (1995) noted that, "A detailed analysis of wind direction, frequency and average velocity for each direction for a period between 1953 and 1970 suggested that >75% of the winds exhibited a westerly component." He stated that, "The spatial plume of ^{239,240}Pu and ^{241}Am clearly followed this wind pattern," and,

"This spatial distribution could only be explained by a wind dispersal mechanism." Litaor further noted, "The isopleth configuration (of plutonium and americium isotopes) was consistent with the hypothesis that the dominant dispersal mechanism of ^{239,240}Pu was wind dispersion from west to east. The actinides (plutonium and americium) activity in the soils decreased extremely (rapidly) in the north and south directions..." (words in parentheses were added for clarity). Recent evidence (Litaor et al. 1996) indicates that significant surface movement of soil plutonium may also have been observed as a result of recent high rainfall events. However, the focus of the current Historic Dose Reconstruction study is on atmospheric transport and potential inhalation dose.

In the mid-1960s, new filters were added to the lathe equipment in Building 776, greatly reducing the contamination levels in the waste oil. Efforts to solidify the oil were also successful at about this time, and in early 1967 clearance operations began on the drums in the 903 Area. However, problems in procuring funding to construct a building to house the filters and pumps needed for this work delayed actual cleanup efforts for some time.

Cleanup: Estimating Plutonium Releases to Soil

Clearance operations in the 903 Area began in January 1967. A mid-April 1966 photograph was taken before cleanup began. The removal of some barrels in the center and lower right corner of the 903 Area can be seen in an April 29, 1967, photograph. An April 10, 1968, photograph shows most drums removed from their previous locations. The overall clearance operation lasted until mid-1968 (Biles 1970); it was hampered by bad weather and difficulties in handling the badly corroded barrels that contained viscous sludge. The clearance process itself has been addressed in numerous reports and in *RAC* and ChemRisk interview summaries.[d] One person who worked on the barrel clearance operation stated that "many" barrels were rusted and contained little liquid. Cleanup initially focused on the most recent drums from Building 776, and delays occurred during attempts to refilter the oil in these drums. The attempt to filter drum contents was soon stopped because it was very slow. The revised method involved transferring the liquids, leaving sludge in the barrels. The last drums, containing uranium, were transferred in June 1968.

Volume Estimates

During processing, 5237 drums containing uranium, plutonium, and trash or tars were removed from the 903 Area (Biles 1970; Putzier 1970a). Putzier noted that the barrel accumulation process, shown in Table II-1, accounted for only 90% of the barrels actually found and removed. These records showed only 4729 drums were transferred to the 903 Area over the years. Putzier also noted that some of the uranium-contaminated oils initially stored in the 903 Area had been burned before the 903 Area cleanup operation. If these barrels were included in the totals shown in Table II-1, the discrepancy between recorded transfers and actual number of barrels recovered would be even larger.

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[d] Contact the Colorado Department of Public Health and Environment, Rocky Flats Health Studies Archives, for copies of the most recent *RAC* and ChemRisk interview summaries.

Table II-1. Drums Transferred to 903 Area[a]

Source building	Number of drums
444	405
881	795
883	165
771	119
77	3245
Total	**4729**

[a] Source: Putzier (1970a).

Oil from the 903 Area drums was transferred into new 55-gal drums during cleanup, and volumes were recorded. Putzier states that 4,826 drums containing repackaged oil were transferred to the plutonium waste processing plant during the cleanup operation. The "difference of 411 drums was attributed to leakage of oil into the soil and some of the drums were not full when first transferred to the field..." (Putzier 1970a). Putzier is apparently comparing the number of new drums (4826) transferred from the 903 Area to the number of drums (5237) he reports were originally delivered to the 903 Area. The difference between these two counts is 411, and this difference is assumed to be due to the fact that a number of barrels were not full (either originally or because of leakage). In summary, 265,430 gal of oil (4826 drums each containing 55 gal) were transferred to Building 774 for processing and 196,460 gal (3572 drums each containing 55 gal) were contaminated with plutonium and the remainder with uranium.

If we assume that the contents of 411 drums leaked to soil, this would amount to approximately 22,605 gal (411 drums each containing 55 gal). However, Don Anderson recalled (Bronesky 1977) that the barrels exhibiting the most leakage were those with uranium-contaminated oils. This leads to difficulty in estimating the quantity of plutonium-contaminated oil leaked to soil based on the (22,605 gal) total leakage (uranium and plutonium-contaminated liquids) inferred from Putzier's estimates.

Health Physics Department Reports

The Health Physics Department surveyed the ground contamination in the vicinity of the 903 Area following barrel removal and recorded their findings in a series of reports. Health Physics Department staff reported in their June 1968 monthly that, "...some slight contamination spread from the contaminated oil drum storage area by the wind and rain is being experienced. The transfer of the contaminated oil from the area has been completed. The packaging for shipment of the empty drums and pallets should be completed by June 17, 1968. On May 20, 1968, a second shift was added to expedite the cleanup of this area" (Dow 1968, June).

On June 11, 1968, "...a leaking solvent drum being transported by forklift truck from the 903 Area to Building 774 caused significant contamination to the roadway. No personnel or vehicle contamination resulted. The contaminated roadway was reopened to traffic the following day after a seal coat had been applied. A 25-foot grid has been staked out over the 903 contaminated area to facilitate contamination surveys. The area is approximately 550 feet by 475

feet and the levels of contamination vary from less than 1000 counts per minute to greater than 1,000,000 counts per minute" (Dow 1968, July).

J. Bruce Owen wrote a memo to J. Seastone in July 1968, noting that plutonium contamination within the rabbit fence around the 903 Area was "extremely hazardous." The report noted that wind and water had carried contamination outside the area. Also in July, "a survey of the plutonium contamination on the surface of the soil in the 903 Area was completed. The results of the survey[e] and the Health Physics Department recommendation for containment of the contamination have been forwarded to Division Services, Manufacturing and Facilities" (Dow 1968, August). After processing to varying extents, all of the contaminated materials removed from the 903 Area were shipped for burial to Arco, Idaho (Putzier 1970a).

In October 1968, "weeds and vegetation were burned off the 903 contaminated oil drum storage area prior to applying an asphalt cap over the area. No contamination problems were encountered" (Dow 1968, November). Also in October, Dow Engineering requested initial approval to cover the area with asphalt, at a cost of $185,000. This proposal was not acted upon, and in November, action was again requested because of the spread of contamination by wind and rain. Dow was allowed to use a road grader to move some of the plutonium-contaminated soil into a smaller area (Dow 1968, December) and to cover it with dirt to reduce the spread of contamination.

The occurrence of strong winds is noted in the Health Physics Department report in December 1968, "...high winds have blown over as many as 150 of the 5000 drums east of the nitrate ponds" (Dow 1969, January).[f] In early 1969, "The high winds of January 31 blew all the roofing material off Building 889. Also, further spread of contamination from the 903 Area is evidenced by area surveys and perimeter air samples east of that area. Approximately 29 leaking drums have been detected in the drum storage area east of the nitrate ponds. These leakers have resulted in high level contamination of about 200 square feet of soil. The contaminated soil is being dug up and shipped as hot waste" (Dow 1969, February). (These latter drums were not in the 903 Area, which had been partially remediated with all drums removed by this time.) Delays in approvals for asphalt coverage of the 903 Area were resolved by April 1969, "...large rocks and stakes have been removed from the 903 drum storage area preparatory to the filling and leveling scheduled to start soon" (Dow 1969, May).

Mel DiLorenzo, interviewed by *RAC* in 1994, worked as a radiation protection technician during the 903 Area cleanup, after barrel removal. He stated that a crew of laborers picked up the larger rocks in the barrel storage area, taking "a couple of days." The Health Physics Department used a Ludlum proportional counter to monitor for alpha contamination. Oily rocks were visible; some exhibited contamination as high as "800,000 counts per minute." Putzier (1970a) noted that during May 1969, 33 drums of contaminated rocks were removed and "discarded as hot waste." He recalled that the main contamination was at the south edge of the area. He did not recall whether soil or smear samples were collected in this area.

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[e] This survey report has not been discovered during the dose reconstruction research.

[f] Included to demonstrate the occurrence of high winds during this key period; details concerning the disturbed barrels are unavailable.

Leveling the 903 Area

After the rock was removed from the area, a road grader was used for leveling. This machine could only be operated "in third gear." Mr. Mel DiLorenzo, interviewed by *RAC* in the summer of 1994, described dust rolling from under the grader's tires and being carried by the breeze to the southeast, visible as far as 0.8 km past the perimeter security fence. The dust cloud may have been "10 to 15 feet high." The grading operation took 2 to 3 days, at 5 to 6 hours per day. He recalls only "slight" breezes during the time, 12 mph or less. Mr. DiLorenzo recalls that the soil was dry and not snow-covered.

A photograph taken in 1995 demonstrates wind suspension of dusts. The photo was taken near Lafayette, Colorado, well north of the RFP. The photograph shows that dust suspension is occurring only in a specific area, recently disturbed by plowing. A 1995 photo of tractor disturbance of soil in the area is also included in Appendix A.

In May 1969, "...the filling, leveling and capping of the 903 Area have not yet started. The road grader used to move contaminated soil and rocks outside of the 903 fenced area was decontaminated and released to surplus" (Dow 1969, June). A May 24, 1969, photograph shows the 903 Area cleared and ready for application of cover materials.

Putzier (1970a) indicated that the road grader was used outside the hot fenced area to move slightly contaminated soil to the fenced area. He noted that the Health Physics Department wanted the area covered as soon as the drums were removed; however, this was not done and the site was exposed to the high winter winds.

The Health Physics Department reported in August 1969, "the first coat of fill was applied to the 903 Area on July 23. Building 903 was moved to a location immediately east of Building 666" (Dow 1969, August).

Placement of the Cover

In September 1969, the Health Physics Department reported, "The base fill has been applied to the 903 Area and the area has been rolled" (Dow 1969, September). "The base course material overlay, soil sterilant and asphalt prime coat for the 903 contamination barrier were completed on 9/24. The asphalt paving, seal coat, drainage ditch and sampling wells should be done soon." (Dow 1969, October). "...sampling wells and seal coat are all that remain to complete the 903 contamination barrier" (Dow 1969, November).

The asphalt work was completed in November 1969 (Loser 1970). The sampling wells at the four corners were finished on November 11 (Dow 1969, December). The asphalt area was 116 x 233 m, with the closest corner located some 88 m west and 12 m south of the east guard station on the Rocky Flats Central Avenue.

Putzier (1970a) provided a chronology of the structure of the asphalt pad and its surroundings. During July 1969, the area was covered with 15 cm of road base. During September, a 7.6 cm asphalt layer was applied. During February 1970, the remaining 903 Area was covered with 10 cm of road base. An April 1970 photo in Appendix A shows both the asphalt pad and the gray, road base area. A May 25, 1971, photograph displays the results of dust suppressant tests.

Basis for Dow Estimates of Leakage to 903 Area Soil

Biles (1970) reported that about 5000 gal of oils had leaked onto the ground during the entire period of barrel storage in the 903 Area, and that about "five or six curies" of plutonium were contained in the leaked oil. This value is apparently from an estimate made by Morrie Maas, a site employee, some time after the cleanup operation. In 1980 private conversations, Maas reported to C.W. Barrick the method by which he calculated an 85 g plutonium release. The maximum volume of oil stored was estimated, and the measured volume of oil recovered was subtracted. The remainder was multiplied by the plutonium concentration measured in the recovered oil, yielding a maximum estimate of ~85 g plutonium. In 1970, Putzier estimated that about 420 drums leaked some of their contents to soil, with about 50 of those drums emptying completely. In 1970, Putzier estimated about 86 g of plutonium leaked to soil. Loser (1970) estimated 85 grams of ^{239}Pu was lost from the drums to soil, based on a private communication with the Health Physics Department site survey technician (probably Maas). It appears that the accuracy of the release estimates noted above is not based on specific measurements. Other information points toward a broader estimated range of plutonium releases to soil. This broader range is used in *RAC* estimates of wind-driven releases from the 903 Area.

After processing in 1968, 594 g of plutonium was recovered from drum liquids, 2471 g stayed with the processed liquids, and 5152 g remained in the emptied drums. These estimates were made using RFP's drum counting system. Rocky Flats staff published several papers describing the waste drum plutonium counting capability (Harlan et al. 1972; Lawless and Chanda 1970).

James Morrison, involved in waste operations onsite for many years, estimated in an interview with ChemRisk that "20–25% of the contents of the (903 Area) drums leaked out" (Morrison 1991). Morrison noted that many of the barrels had become extremely fragile and could not be moved without falling apart. If Morrison's recollection is correct, it would suggest 40,000–50,000 gal of plutonium-contaminated liquid leaked to soil, compared with the site estimate of 5000 gal. Using plutonium concentrations measured during cleanup, this would imply more than 1000 g of plutonium leaked to soil. While other evidence suggests that perhaps 10% of this value is the best estimate, *RAC* uses the Morrison value as an upper limit to the overall range of estimates.

Disposal of 903 Wastes at the Idaho National Engineering Laboratory

The Idaho National Engineering Laboratory (INEL) received the soil, barrels, and related wastes from the 903 Area cleanup at Rocky Flats. A report written by D.H. Card (1977) contains a brief history of each of the pits used for waste burial at what is now called the Radioactive Waste Management Complex (RWMC) at the INEL. Those pits that could have received 903 Area waste are listed in Table II-2. The empty drums disposed in INEL Pits 6 and 9 may have come from the RFP 903 Area.

During the earliest years when Rocky Flats wastes were shipped to the INEL, descriptions of the contents of waste containers were limited; however, the numbers of containers and disposal volume are known. A recent report by EG&G (1994) notes the annual volumes of material disposed (see Table II-2). There was a substantial increase in disposal volume in 1968, about 90% above the mean for 1966–1967 (~4600 m^3). Disposal volumes for 1969 and 1970

were about 36% and 96% greater, respectively, than the 1966–1967 average. Cleanup following the 1969 RFP fire is no doubt responsible for much of the volume in these 2 years.

Luckett et al. (1982) reports the soil volume removed from the lip area[g] near the 903 Area as 458 m^3. This would fill approximately 2200 55-gal drums (the actual number would depend upon packing efficiency and whether the barrels were lined).

Table II-2. Burial of RFP Transuranic Waste at the
Idaho National Engineering Laboratory[a]

Pit #	Disposal period	Contents	Comments
5	2/65–12/66	18,486 drums; 673 boxes; 677 cartons	
6	5/67–9/68	14,396 drums; 3423 boxes	Includes 2451 boxes of contaminated drums
7, 8			Not used for transuranic waste
9	5/68–9/68	3921 drums; 2029 boxes	Includes 1302 boxes of contaminated drums
10	9/68–10/70	26,645 drums; 2849 boxes	Most 1969 fire debris received this period
11	4/70–10/70	13,542 drums; 90 boxes	
12	7/70–9/72	4838 drums; 26 boxes	Transuranic waste burial from July–November 1970 (after which transuranic was no longer buried).

[a] Source: EG&G (1994).

[g] The area to the east of the 903 Area where the terrain drops sharply for a short distance.

CHAPTER III

APPROACH AND BACKGROUND FOR ESTIMATING RELEASES BEFORE 1970

Suspension modeling was used to estimate releases of contaminated soil from the 903 Area before 1970 as described earlier in this report. To effectively apply these models, it is important to first understand the release events.

NATURE OF THE RELEASE EVENTS

There is much evidence to support the *episodic*[h] nature of the release events from the 903 Area using the environmental data collected by the site during the period of interest. Dow Chemical Company had a series of air samplers located outdoors across the RFP. These samplers were low-volume air samplers, operating at a flow rate of about 2 cubic feet per minute (cfm). Particulates were collected on a 2-in diameter filter, and the filters were retrieved every 24 hours. The activity collection period lasted longer over weekends and holidays when filters were not changed out as frequently. The total long-lived alpha (TLLa) activity on daily air filters was used as an indication of the presence of plutonium. The location of these air samplers is shown in Figure III-1.
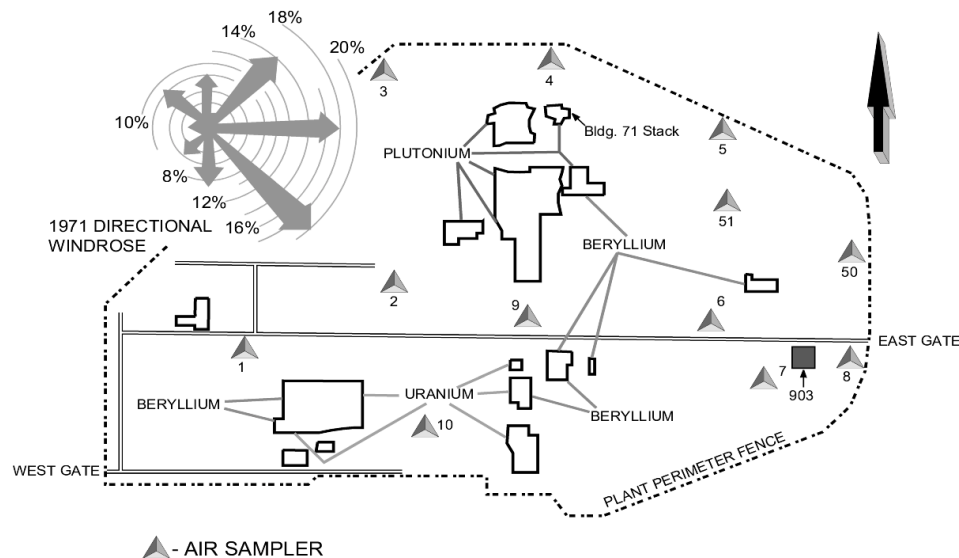
Figure III-1. Location of early onsite air samplers (adapted from a figure in the 1971 RFP annual environmental report). The distance from west to east across the RFP site is roughly 1 mi. The 903 Area is between air samplers S-7 and S-8, in the right of the figure, near the East Gate.

The S-8 sampler was one of the 10 onsite ambient air samplers, located to the east of the 903 Area. Examining historical readings from the S-8 sampler revealed a small number of days

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[h] Episodic events occur over discrete, or separable, periods of time.

during which sampler readings were unusually high. These dates all fell within the 1964–1969 time period, while the 903 Area soil was exposed. Only the S-8 sampler exhibited these very high readings; however, a high reading at S-6 or at S-7 occasionally appeared on one of these same dates.

A review of wind records from nearby meteorological stations showed that the dates of high sampler readings coincided with higher than normal wind readings. Figure III-2 illustrates this phenomenon.
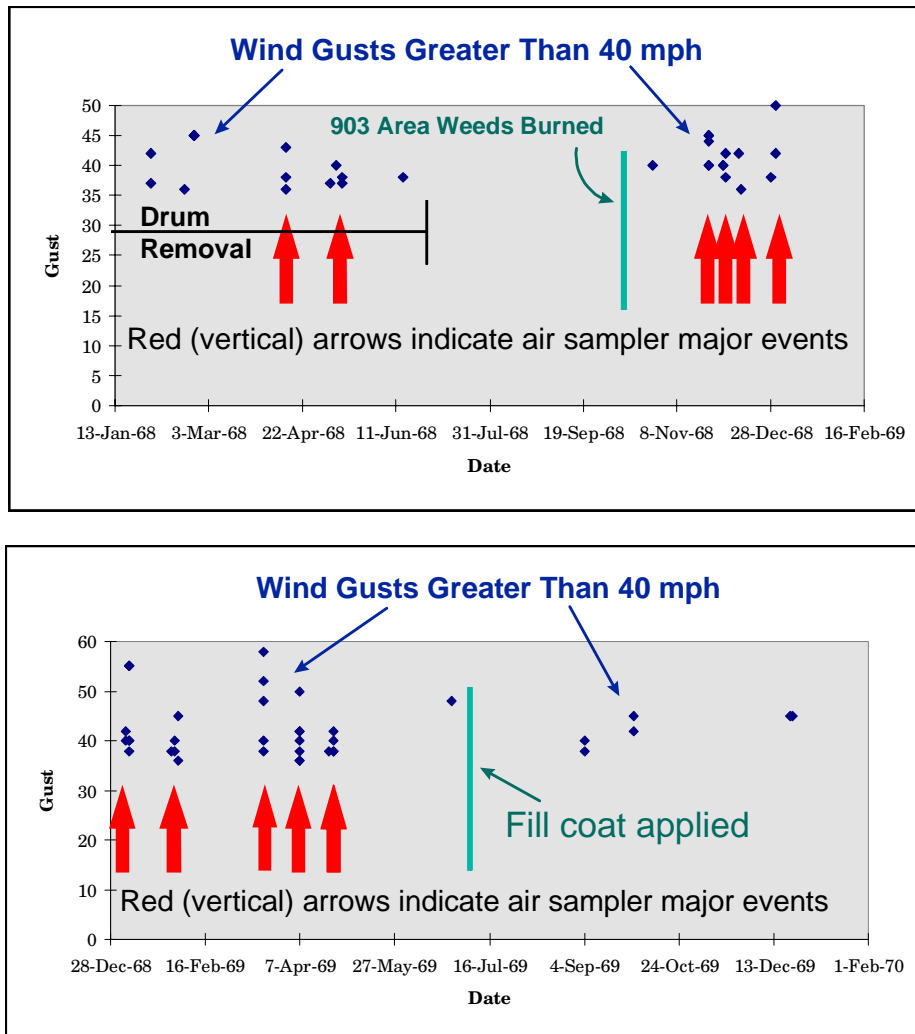
Figure III-2. Correlation over time of high total long-lived alpha measurements at the S-8 air sampler (shown as large vertical arrows) with high winds (defined as gusts greater than 40 mph) and 903 Area disturbance events denoted. Major events at the 903 Area included drum removal, burning weeds from the area, and applying the asphalt pad.

The large arrows in the above figure denote high readings of alpha activity at the S-8 air sampler and are called S-8 events in this report. These S-8 events occurred only during that time

before a cover was applied to the 903 Area. Most of the major S-8 events occurred after the weeds had been burned off the area and only when significant wind gusts also occurred. These pieces of information led Meyer et al. (1996) to conclude that a large fraction of the plutonium released from the 903 Area at Rocky Flats was associated with specific high-wind events.

Erosion of Plutonium–contaminated Soil Particles

The next step in the process of understanding the nature of release events from the 903 Area involved researching erosion because it appeared to be the force moving the majority of the contamination from the 903 Area to other locations. Soil erosion is the process by which surface particles are moved by wind, water, or other mechanisms. The process of erosion has been studied for years. As early as 1941, Bagnold studied the physics of the movement of desert sand and derived an aerodynamic theory to describe the motion of sand grains (Bagnold 1941). Research continued in later years and eventually resulted in formulating general equations and prediction systems for erosion processes over a variety of natural and man-made surfaces based on mass continuity. From this research, two conclusions can be drawn: (1) wind speed is the primary predictor of erosion and is the variable that dominates the outcome of the calculation, and (2) a large number of soil properties that are difficult to specify numerically can either inhibit or promote wind-driven erosion.

If erosion were simply an interaction between the soil and the wind, it should be possible to accurately quantify the amount of soil lost to erosion under given wind conditions. Many factors act to complicate matters, however. Soil moisture can act to inhibit erosion. At Rocky Flats, this factor is particularly important because of the semi-arid nature of the soils. Even a small amount of precipitation or residual soil moisture from snowfall may limit erosion from a surface. Vegetation has a similar effect on erosion. Farmers commonly use vegetation to limit the topsoil erosion from fallow fields. In Figure III-2, the effect of vegetation on the erosion of material from the 903 Area surface seems evident. There appears to be a correlation between the 903 Area weed burn in the fall of 1968 and the increased frequency of S-8 air sampler events after that date. Other factors may also have influenced this increase, but this observation provided another important clue that erosion may be the force at work in the 903 Area.

Promoters of erosion can include mechanical disturbances, human activity, and previous wind disturbances. Many attempts have been made to quantify the effects of a disturbance on erosion, but the outcome depends on disturbance parameters, measures of which are usually unavailable.

Because of the impossibility of defining so many parameters for events that took place over 30 years ago, *RAC* researchers focused on understanding the forces that caused the plutonium releases from the 903 Area, collecting available site-specific data, and building mathematical models that could fully use the available information. Two separate processes were at work during the erosion of the 903 Area: suspension and resuspension. These terms are illustrated in Figure II-1 and are used throughout this report to describe releases from the 903 Area from the erosion that occurred pre-1970 (suspension) and post-1970 (resuspension).

Contribution of Releases from Discrete Events to Total Releases

Figure III-2 showed the correlation between high winds and large plutonium releases from the 903 Area, as indicated by the S-8 air sampler. *RAC* analyzed the historic S-8 air sampler data quantitatively to confirm this correlation. Of the 2269 total days in the pre-1970 time period, 24 exhibited high TLLa readings at the S-8 air sampler, which we assume indicates high airborne plutonium concentrations. Because some of the 24 days occurred over weekends when the S-8 sampler filter paper was not changed, *RAC* was able to reduce the number of wind events to be evaluated to 17. Adding together the gross TLLa concentration values for the filters from the 17 wind events showed that over 51% of the airborne plutonium collected at the S-8 sampler resulted from fewer than 1% of the total days for which air sampler data are available. The S-8 data have limitations (discussed in the following section), but this evaluation gave *RAC* researchers some boundaries for the detailed evaluation of suspension releases from the 903 Area during the pre-1970 time period.

MATHEMATICAL MODELS, METEOROLOGICAL DATA, AND AIR MONITORING DATA

For a detailed quantitative evaluation of the 903 Area suspension, *RAC* scientists had to find usable data and the best available models. The limitations described above and the information gained from the air monitoring data helped direct the *RAC* search for appropriate information. After a brief initial search, we recognized that not enough information about soil moisture and possible disturbances would be available to quantify those parameters. Given the lack of information, the decision was made to focus on the search for detailed meteorological information and use this information in concert with the available models and the air monitoring data to make predictions about 903 Area releases.

S-8 Air Sampling Data: Corrections, Modifications, and Limitations of the Data

Before the S-8 air sampler activity data could be used to make quantitative predictions, many limitations of early air monitoring data had to be recognized and addressed. There have been arguments about the quality of these data, but *RAC* believes that with the appropriate modifications for known inadequacies and recognizing of uncertainty, the data can be used to help quantify the 903 Area releases. Data quality analysis attempts to ensure that the monitoring data are used appropriately and not assumed to reflect more information than they actually represent.

The Task 4 report for this project included a comprehensive section on data quality for ambient air monitoring (see pages III-91–III-123) (Rope et al. 1997). A summary of this information is given here and in Chapter IV of this report.

Data quality for low volume air samplers at Rocky Flats, including the S-8 sampler, is impacted by the following factors:

- Counting error, or the statistically random nature of radioactivity, introduces uncertainty about the central value of the measured count rate based upon the magnitude of that count rate.
- Sampler inlet collection efficiency is a function of particle size and wind speed. It quantifies the effectiveness of the sampler at collecting particles in certain size ranges.
- Filter collection efficiency relates to the efficiency with which the filter collects the particle sizes of interest.
- Filter clogging relates only to the filter collection properties of S-8 data during high wind speeds, such as those during the largest five S-8 events (net TLLa count rate >100 cpm). Filter clogging becomes a factor when particles clog the filter and reduce the flow of air through the filter. The magnitude of this correction is related to some extent to the filter paper type and is approximated by reducing the total daily volume of air sampled used in calculations (Rope et al. 1997).
- Counter efficiency and alpha particle self-absorption are key factors in converting an alpha count to a quantity of alpha particle activity (Rope et al. 1997). Counter efficiency is represented by the fraction of the total alpha particles emitted by the source (filter) that is measured by the detector. Alpha particle self-absorption occurs as a result of the limited range of the alpha particle in matter. The sample (filter) can absorb some fraction of the total activity contained on it. All factors affecting counter efficiency and sample self-absorption are incorporated into a single total efficiency factor for alpha count results.

Applying each of these data quality factors to an estimate of releases is discussed in detail in Chapter IV of this report. The data used in the 903 Area source term analysis are inherently uncertain, but examining data quality issues is a way to quantify the uncertainties and to make appropriate corrections for bias. This series of data quality corrections increases the uncertainty bounds on the air concentrations measured at the S-8 sampler, which if excluded from the analysis, might otherwise make the data unusable.

Meteorological Data

Detailed and site-specific meteorological data were critical to the success of the 903 Area modeling effort. Naturally, RFP data were the most valuable, but other nearby locations where meteorological monitoring took place were also located and used to supplement missing detail from available RFP data.

RFP Data

The first data set of interest to *RAC* was the original set of strip charts[i] from the RFP anemometer.[j] This wind and weather recording station was located near the center of the site and served to record wind speed and direction information for the site.

Early in the project, *RAC* began a concentrated search to find the wind charts from the late 1960s, particularly the period from December 1968 – April 1969. During the search, a number of

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[i] Strip charts refer to long charts (in strips) that were used to record wind data during this period. These charts contain continuous information about winds.

[j] An anemometer is the machine used to measure and record wind speed and direction information.

scientists and former site employees recalled having seen the charts, but they could not locate the charts. The only meteorological data from Rocky Flats that we found were summary hourly wind and weather data for the period of interest. This information gave an average wind speed for each hour, but we required wind data with far greater resolution. Some of this wind information, letters documenting the search for onsite traces, and limited amounts of wind data are located in Appendix B. Without the continuous wind data from Rocky Flats, the search continued for an appropriate set of meteorological data.

Jefferson County Airport Data

The next set of wind data that *RAC* located was the data from the Jefferson County Airport, located southeast of Boulder. Figure III-3 shows the location of the Jefferson County Airport.
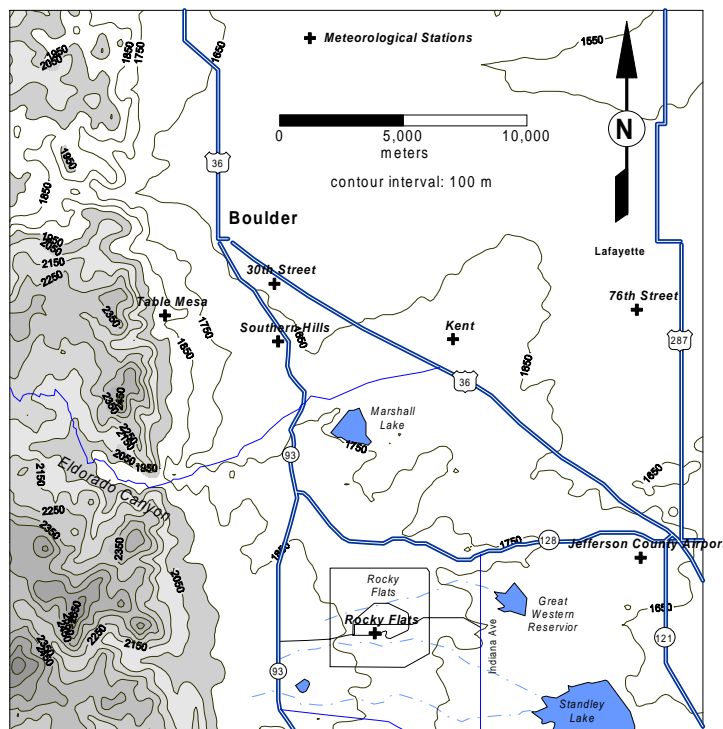
Figure III-3. Area surrounding the RFP. Jefferson County Airport is shown to the northeast of the plant.

Data sets for the critical time period were collected from the airport and assessed by *RAC* scientists. Wind speed, gust speed, and wind direction information was available hourly from 6 a.m. until 10 p.m., the hours of airport operation. We obtained the data from 1967–1970.

Meteorologists who were familiar with wind data collection at airports during that period indicated that these data are less-than-ideal for purposes of modeling releases. The data at an airport are collected not as hourly averages, but rather as hourly point estimates of the wind speed and direction. The person in charge of collecting these data watched the instruments at the top of each hour, collected a 1-minute average for the wind speed and wind direction, and assessed the cloud cover and ceiling height. This type of wind speed and direction information is

not very useful for atmospheric modeling because more detailed information is necessary to accurately predict contaminant movement in the atmosphere.

Another problem with the Jefferson County Airport data was the limited collection time of the data (only during the hours of airport operation). Using these data as the wind speed data would have required finding another data source for the missing hours or producing data from the limited available information. This method was not preferred because many of the strongest wind storms are known to occur during the nighttime and early morning hours. Examples of Jefferson County Airport data are shown in Appendix C.

The cloud cover and ceiling height information from the Jefferson County Airport was helpful, however, for use in our modeling to predict atmospheric conditions that might promote or inhibit dispersion.

National Centers for Atmospheric Research Data

At the suggestion of a member of the Health Advisory Panel overseeing this study (Dr. Niels Schonbeck), *RAC* queried the National Centers for Atmospheric Research (NCAR) as a possible source of data for Front Range winds. A primary NCAR station has existed since the early 1960s atop Table Mesa, above Boulder, Colorado. To facilitate experiments designed to study wind patterns, other stations were temporarily located at various points throughout Boulder and the Rocky Flats area. These stations included Kent, Southern Hills, and 30th Street. See Figure III-3 for approximate locations of NCAR stations.

The data at the NCAR stations were continuously collected by anemometers. Operational errors or power failures were the only conditions that interrupted data collection. These anemometers measured wind direction as well as speed. In some cases, the wind direction was recorded continually, in the same manner as the wind speed. In other cases, including the Kent station, the wind direction was recorded every 5 minutes as a small tick on the wind chart, in one of eight quadrants for wind direction. Wind speed and direction data regularly exist for all periods during which an anemometer operated. Examples of both types of wind charts appear in the appendices to this report. Appendix D contains Table Mesa charts, and Appendix E contains Kent charts.

A search for data from the late 1960s yielded boxes of records retrieved from the local archives with the assistance of the NCAR archivist, Diane Rabson. These boxes contained wind records from a series of NCAR stations located throughout the area for the time period of interest. A listing of the wind records stored at the NCAR archives is given in Appendix F. A map showing approximate station locations (see Figure III-3) suggested that data from the Kent station might prove to be the most applicable to Rocky Flats because it was located at approximately the same latitude as Rocky Flats. A comparison of Kent data to the summary data available from Rocky Flats and other NCAR stations further supported this hypothesis. This comparison is shown in Figure III-4.
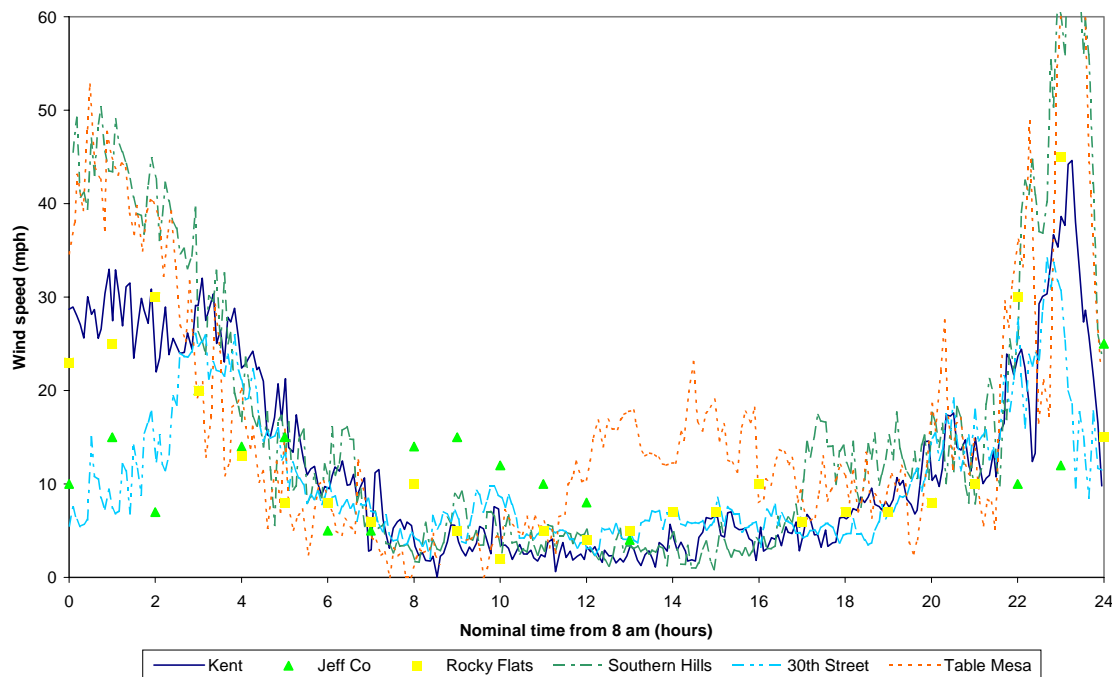
Figure III-4. Rocky Flats summary wind data compared to NCAR stations data for January 30, 1969. Compared with the other NCAR data sets, the Kent data (dark, solid line) appear to most closely represent the Rocky Flats data (light squares).

The Kent data set is the best predictor of Rocky Flats winds of the available NCAR stations. Kent data were compared to Rocky Flats data for five of the largest wind events, and the results of those comparisons are shown in Appendix G. The figures in this appendix indicate that the Kent mean wind speeds are well matched to the Rocky Flats hourly average wind data.

RAC discovered that the former location of the Kent station was at 5691 South Boulder Road in Boulder, Colorado. The details of this search for the station's location, the operation of the station, and some photographs of the area are provided in Appendix H.

The Kent data were collected as part of a temporary experiment conducted by NCAR to examine wind wave patterns as they emanate from the Front Range. Because this was a temporary station, data from this location were only available for a limited portion of the 903 Area study period. Data from the Kent station were obtained for the January 30, March 19, and April 7 events in 1969, dates of three of the six largest wind events that affected the 903 Area.

Because data for the other dates on which suspension events likely took place at the 903 Area did not exist for the Kent station, *RAC* had to find other data to supplement this data set. Located at NCAR headquarters, the Table Mesa station has the most complete data set of all NCAR stations because it was the only nontemporary, continuously operating station at the time. The Table Mesa station, as a result of its higher altitude and location directly in the foothills, generally exhibits higher winds and greater gusts than are seen further out onto the plains. *RAC*

used the best obtainable data, however, to simulate Kent data for dates when Kent data were not available. The process of extracting data from the wind charts and simulating the missing Kent data is described in the following section.

Extracting Data from the NCAR Wind Charts

The NCAR charts exist in several different forms (see Appendices D and E). It was important to develop a consistent method for translating the information in the wind charts to numerical data for use in a model. While the incremental time scale on all of the wind charts was very difficult to read (which prevented the possibility of scanning and digitally integrating the charts), the hourly scale was moderately legible so that each hour could be broken geometrically into 5-minute increments. A ruler was used to create a scale that divided each hour evenly into 12 segments, each representing a 5-minute period. This scale was transferred onto a strip of paper that extended the length of the wind chart to enhance evaluating the wind speed at each increment.

To determine the representative wind speed for each 5-minute segment, the wind speed for that segment had to be analyzed as a whole. Conversations and meetings with Dr. Alfred Bedard, from the National Oceanic and Atmospheric Administration (NOAA) in Boulder, led to an appropriate technique for analysis. Conversations with Dr. Bedard are detailed in Appendix I, which contains a transcript of our first meeting, a letter sent to Dr. Bedard in preparation for our second meeting, and notes from that second conversation.

Understanding the wind charts from this period at the locations for which we had data requires knowledge of wind patterns and wind effects on the Front Range. Dr. Bedard has been working at NOAA and studying wind patterns there since the early 1970s. We took the wind charts from Kent and Table Mesa to his offices in Boulder for advice on characterizing them.

Dr. Bedard suggested that although the wind charts indicate some short-term microgusts during a number of time increments during a day, these bursts are rarely more than eddy effects; within 20 to 30 feet of occurring they will have lost their coherency and broken down. Examples of eddy effects are shown in Appendix D on the Table Mesa charts. Dr. Bedard also noted that the eddy effect is reduced and the coherency of the winds is usually greater at locations farther from the mountains on the west side of Boulder. Thus, the Kent station and Rocky Flats would be expected to exhibit more coherent wind patterns, as shown in the wind charts from the Kent station in Appendix E. Wind records from the Kent station are also characteristic of winds at Rocky Flats because the Kent station was closest to Rocky Flats and was situated about the same distance from the foothills of the Front Range.

To represent the dominant wind speed for the Table Mesa charts that exhibit eddy effects, Dr. Bedard suggested that we draw smooth-fit curves to the "valleys of the peaks," the wind speeds that represent the maximum and most representative speed for each increment of time without being influenced by eddy effects. Figure III-5 shows a representation of this technique.
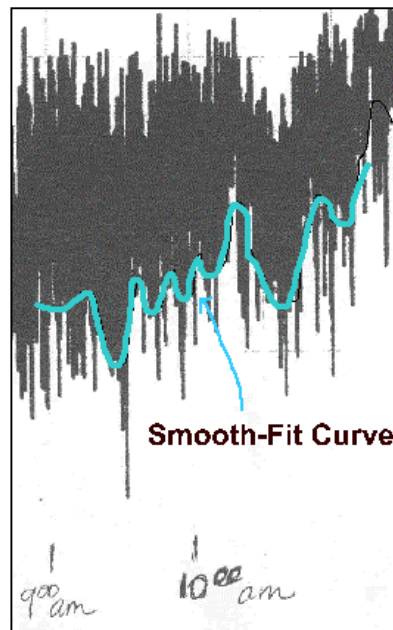
Figure III-5. Example of how representative wind speeds were characterized (Table Mesa data, larger wind speed toward bottom of chart).

Dr. Bedard agreed with our technique of wind speed characterization, and he indicated that he did not know of a better way or a computerized/mechanized way to accomplish what is easily and commonly done by meteorologists with wind charts of this type. Although the line is hand-drawn, the body of scientific knowledge that supports the technique is vast.

Once the best-fit line was drawn on the charts to indicate the characteristic wind speed for each increment of time, a wind speed scale was added. The wind speed during each time increment was then determined by measuring the location of the smooth-fit line within the increment.

This technique allowed a numerical evaluation of the wind speeds from the otherwise difficult to read anemometer charts. With the NCAR wind charts as our only source of continuous data for the time period of interest at appropriate locations, this technique made it possible to use data that we may not have otherwise been able to use.

It is important to recognize that this approach to extracting detailed wind speed data from older wind charts need not be perfectly accurate. For our purposes, the primary goal was that data read from a series of such charts be extracted in a consistent manner so that the results could be compared. From our understanding of the data charts and their history and the assistance provided by Dr. Bedard in their interpretation, we are reasonably confident that we can compare the resulting extracted data.

Using Table Mesa Data to Simulate Kent Data

For the dates when Kent data were not available, we established a method for simulating Kent data. We compared the Table Mesa and Kent data for dates when data existed for both locations to establish a ratio of wind speeds. A cursory comparison indicated that, for the higher

wind speeds of interest, there was a consistent wind speed relationship between the two stations (with the Kent data consistently lower than the Table Mesa data).

The data were digitized and the numerical values compared. The average Kent value was divided by the average Table Mesa value for each increment of time over the entire day. Summary statistics were run on these ratio values, and for March 19 and April 7, a mean value of 0.6 was obtained.

The January 30 wind data yielded a mean ratio close to 1. Because our information on wind patterns at the Front Range indicated that high winds are always stronger nearer the mountains, the January 30 data were examined more carefully.

Plotting these data revealed that at lower speeds on this date, winds at Kent were slightly stronger than at Table Mesa. During periods of strong, gusty winds, however, wind speeds at Table Mesa were consistently higher than at Kent. The mean of the ratios was calculated again, this time using only periods of high winds; the method revealed a ratio of Kent to Table Mesa of 0.6, matching that from the other two dates.

Thus, the ratio of 0.6 was adopted to simulate Kent data from Table Mesa data. For all wind events where Kent data did not exist, Table Mesa data were digitized and multiplied by a factor of 0.6 to simulate Kent data.

Suspension Models

To adequately model the erosion processes that moved soil off the 903 Area and into the air, *RAC* researched the most widely used equations to model the suspension of dust from a bare soil or vegetated surface, those of Dr. Dale Gillette (Gillette 1974).

The model originally developed by Gillette (1974) and studied further by Gillette and a number of other scientists (Gillette 1977; Gillette and Cowherd 1982; Gillette et al. 1998; Cowherd et al. 1985; Porch 1979) describes the wind-driven suspension of fine particulate matter. Fine particulate matter consists of particles with physical diameters less than 20 μm, or particles that are readily inhalable. Larger particles may saltate ("hop" along the ground), impacting and suspending smaller particles, as originally detailed in Bagnold's theory (Bagnold 1941). Saltation may increase the suspension of fine matter by breaking up larger aggregates, increasing the number of particles in the smaller size range. The saltation effect on suspendible material is incorporated into the Gillette suspension model.

The suspension flux equation suggested by the work of Gillette is

$$F_v = X \left(\frac{u}{u_t} \right)^{\gamma}$$

(III-1)

where
F_v = vertical flux of dust from soil surface (g cm^{-2} s^{-1})
X = constant related to soil type (g cm^{-2} s^{-1})
u = wind speed at some reference height z (m s^{-1})

u_t = threshold wind speed for saltation[k] (m s^{-1})
γ = wind speed exponent (dimensionless).

The constant (X) in this equation is calibrated with experimental data for a particular soil surface type. The wind speeds needed for Equation (III-1) were obtained from the NCAR data charts. The 24-hour time period beginning at 8 a.m. on the day of interest was digitized for each chart into approximately 5-minute increments, as described previously. Also extracted from the NCAR charts were median and maximum wind speeds during each 5-minute increment. Examining the charts also led to values for fractions of time during each interval that gust (or maximum) wind speeds were observed. These fractional values were used to adapt Equation (III-1) to predict total vertical particle flux for periods of time during which median wind speed was not the only predictor of suspension. The use of these gust fractions is shown in Equation (III-2).

$$F_v = X\left[\left(\frac{u_g}{u_t}\right)^{\gamma} \cdot f_g + \left(\frac{u_m}{u_t}\right)^{\gamma} \cdot \left(1 - f_g\right)\right] \tag{III-2}$$

where
u_g = gust speed (m s^{-1})
u_m = median wind speed (m s^{-1})
f_g = fraction of the time gust speeds prevail (unitless).

These two equations were used to model 903 Area releases using calibration techniques as described in the following sections.

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[k] The threshold wind speed for saltation is the threshold for particle movement. Saltation activates the mechanism of suspension, linking the two processes. It is likely that suspension thresholds are somewhat higher than saltation thresholds, but they are also more difficult to quantify. Saltation thresholds are used throughout this study.

CHAPTER IV

ESTIMATION OF RELEASE QUANTITIES

As discussed in Chapter III, high alpha activity readings at the S-8 air sampler are correlated with the high wind events at the RFP during the period of interest in 1968 and 1969. Thus, the high activity readings at the S-8 sampler are assumed to be due to the suspension of plutonium-contaminated soil from the 903 Area. In this chapter, we quantify the plutonium activity released from the 903 Area for the 24 highest days measured at the S-8 sampler and those that occurred on a more-or-less continuous basis for the years 1964 through 1969 (referred to as baseline releases). Release estimates are calibrated to measured activity levels in air taken at the S-8 air sampler, accounting for the deficiencies in the air sampler and subsequent sample and data analysis. Release estimates are presented as distributions of possible values that represent the overall precision in the methods used to arrive at the quantities. Release estimates from the 24 highest days are presented first and are followed by the continuous release estimates.

RELEASE ESTIMATES FOR THE 24 HIGHEST S-8 SAMPLER DAYS

To calculate release quantities for the 24 highest days measured at the S-8 sampler we used daily sampler recording sheets (original, handwritten data) and meteorological data from several recording stations. Meteorological stations providing data included the Jefferson County Airport, portable NCAR stations running at Kent and Table Mesa during the period of interest, and digitized data from Rocky Flats provided in electronic format by the Phase I contractor. These data were used in conjunction with an air dispersion model capable of quantifying the effects of gravitational settling and dry deposition to estimate the quantity of plutonium released from the 903 Area. With this approach, we assumed that all activity detected at the S-8 sampler was due to 903 Area sources. Contributions from routine releases and from resuspension of previously deposited material were not considered. The following sections outline the conceptual and mathematical formulation used to derive release quantities and formulate corrections for the deficiencies in the S-8 air sampler data. The remainder of the chapter describes our treatment of uncertainty, presents the results of this portion of the dose reconstruction study, and discusses the sensitivity of these results to input parameters.

Conceptual Model and Mathematical Formulation

The airborne concentration of plutonium attached to soil particles at a sampler located *x, y, z* from an area source for a time increment where the meteorological conditions are assumed to remain constant can be estimated by

$$C = q \, T_f \tag{IV-1}$$

where q = the source strength (Ci s^{-1}) and T_f = the transfer function (s m^{-3}). The transfer function describes the diffusion, dilution, and depletion of the airborne mass as it is transported from the source to the sampler, and it is a function of the meteorological modeling parameters,

stability class, wind speed, and wind direction. The transfer function goes to zero when the wind blows in the opposite direction of the sampler, and it is maximized when the wind blows along the azimuth line connecting the source to receptor. The source strength is a function of wind speed and a variety of other factors, such as soil moisture, vegetation, surface roughness, and distribution of particle sizes (of which we know very little). If we assume that the transport time from the source to the receptor is relatively short compared to the meteorological time increment, then steady-state conditions may be assumed and a model based on the straight-line Gaussian Plume model may be used to determine the transfer function. The source term (q) then becomes the steady state release over the meteorological time increment. For the high release days, wind gusts have a large effect on the total release because of the strong dependence of soil suspension on wind speed. Therefore, it was important to use meteorological data with the finest resolution possible to capture the effect of wind gusts. Longer meteorological averaging times tend to decrease the overall effect of wind gusts and thereby underestimate releases. The meteorological data taken at the portable NCAR stations had the finest resolution; therefore, they were used in the calculations. Wind speed and wind direction were digitized every 5 minutes as previously described.

Measured concentrations of alpha activity in air at the S-8 sampler were reported daily. Therefore, model calibration can consider only total daily releases from the 903 Area. However, within the 24-hour measurement period, the source strength (curies per second) varies as a function of wind speed, which varies as a function of time of day. In addition, sampler efficiency also varies with wind speed and particle size. Large particles (>30 µm aerodynamic equivalent diameter [AED]) go virtually undetected by the samplers; thus, these results only apply to the smaller size fractions. These factors were accounted for by correcting the measured S-8 sampler concentration for deficiencies in sampler efficiency based on the wind speed and an assumed airborne particle size distribution.

The average airborne concentration over the 24-hour sampling period must be computed for comparison with the measured concentrations. The total amount of plutonium attached to particles <30 µm AED that is released for a given day (q_{mod}) is given by

$$q_{mod} = \sum_{i=1}^{n} q_i \Delta t \qquad \text{(IV-2)}$$

where q_i = the quantity of plutonium released in a meteorological sampling period ($\Delta t = 5$ minutes) and n = the number of meteorological sampling periods in a 24-hour measurement period. The release rate (q_i) varies as a function of wind speed raised to a power. The 24-hour average modeled concentration (C_{mod}) can be estimated by

$$C_{mod} = \frac{\sum_{i=1}^{n} q_i T_{f\,i}}{n} \qquad \text{(IV-3)}$$

where T_{fi} = the transfer function for each meteorological sampling period. We are interested in finding the time-integrated release rate that corresponds to a measured concentration at the sampler (i.e., Σq_i). The quantity, q_i cannot be solved directly from Equation (IV-3). Therefore, we derive an additional relationship to arrive at the quantity. The term C_{mod} is comparable to the average airborne concentration measured at one of the samplers. The actual release from the 903 Area for a 24-hour period may be determined by multiplying the modeled release (q_{mod}) as given by Equation (IV-2) by the ratio of the modeled and measured air concentrations. The calibrated release (q_{cal}) that corresponds to measured concentrations at the S-8 sampler is then given by

$$q_{cal} = \frac{C_{meas}\, q_{mod}}{C_{mod}} \tag{IV-4}$$

where
q_{cal} = calibrated release from the 903 Area (Ci)
C_{meas} = measured air concentration (fCi m^{-3})
q_{mod} = modeled total release (Ci)
C_{mod} = modeled air concentration (fCi m^{-3}).

Several limiting assumptions are inherent in this approach:

- The concentration observed at the sampler is entirely due to suspension of plutonium attached to soil particles from the 903 Area. Other sources are ignored, such as resuspension of contaminated soil particles from previously deposited material.
- Distribution of activity among the particle sizes is based on site-specific activity particle size distributions reported in the literature.
- The source is assumed to remain active for the duration of the sampling period. Depletion of suspendable material before the end of the sampling period is not considered.

Transfer Function

The transfer function can be estimated by selecting an air dispersion model capable of handling deposition and plume depletion of particulates. The Fugitive Dust Model (FDM) is an appropriate model to use for this purpose (Winges 1990). The FDM is based on the Gaussian Plume formulation for computing airborne concentrations of dilute substances, but the model has been specifically adapted to incorporate an improved gradient-transfer deposition algorithm originally described by Ermak (1977). The model also incorporates an area source geometry that facilitates simulations for the 903 Area. The FDM simulations incorporated three separate area sources that were within the area later covered by the asphalt pad. Aerial photos and other records indicated the extent of contamination at the 903 Area was limited to an area of 6000 m^2 (total area of the 903 Area is 13,700 m^2). The model was run for a unit release rate (1 Ci s^{-1}), and concentrations at the S-8 sampler location were output for each meteorological period. The transfer function for each meteorological period is then given by

$$T_{f\,i} = \frac{C_i}{Q} \qquad\qquad\qquad\qquad (\text{IV-5})$$

where C_i is the concentration for the ith meteorological period (Ci m^{-3}) and Q = the unit release rate (Ci s^{-1}). Air concentrations predicted by FDM represent the total suspended particulate concentration based on an initial particle size distribution of the source. Particle size distributions are discussed in a later section of this chapter.

Source Release Model

The source release model is based on the premise that release rates are related to the wind speed. In effect, ignoring this premise would assume a constant release rate over the measurement period; therefore, it would not be necessary to develop a source release model. Preliminary calculations showed that ignoring the wind speed dependency of release rate resulted in predicted releases that were lower than when wind speed dependency was included. Soil erosion research has shown the wind speed dependency of suspension. For these reasons, wind speed was included in our calibration model.

The soil suspension model, given in Equation (III-1), is used to estimate the release rate (q_i) in Equations (IV-2) and (IV-3). Wind speed-dependent models typically employ variations of the semi-empirical model proposed by Gillette (1974) to predict suspension of fine particulate matter (<20 μm) from erodible surfaces. These models also implicitly include the effects of saltation on suspension. The source release model uses an equation of the form

$$q = C_s\,AX\left(\frac{u(z)}{u_t(z)}\right)^{\gamma} \qquad\qquad\qquad (\text{IV-6})$$

where

C_s = the concentration of plutonium in soil (Ci g^{-1})
A = area of contamination (m^2)
X = soil suspension calibration constant (g m^{-2} s^{-1})
u = wind speed measured at height z (m s^{-1})
u_t = threshold wind speed at height z (m s^{-1})
γ = wind speed exponent.

The implementation of Equation (IV-6) in Cowherd et al. (1985) includes an additional multiplicative term, $F(x)$, where $x = 0.886\ u_t/u$. We fit the $F(x)$ term presented as a graph in Cowherd et al. (1985) to a polynomial given by

$$F(x) = 1.91207 - 0.0278085x + 0.48113x^2 - 1.09871x^3 + 0.335341x^4 \qquad (\text{IV-7})$$

for $x < 2$ and

$$F(x) = 0.18\left(8x^3 + 12x\right)e^{-x^2}$$ (IV-8)

for $x \geq 2$.

Cowherd's model applies to particles ≤ 10 μm and uses a wind speed exponent of 3. The soil suspension calibration constant used in Cowherd et al. (1985) is 1×10^{-9} g cm^{-1} s^{-1}, which is in the range suggested by Porch (1979). The threshold wind speed is the erosion threshold friction velocity extrapolated to the height of the wind speed measurement. A logarithmic velocity profile is typically assumed and given by

$$u_t(z) = \frac{u_*}{0.4}\ln\left(\frac{z}{z_o}\right)$$ (IV-9)

where
u_* = threshold friction velocity at the surface (m s^{-1})
z = the wind speed measurement height (m)
z_o = the roughness height of the erodible surface (m).

The problem in using any of these models is the lack of soil characterization data for the 903 Area. Of particular interest is the erosion threshold friction velocity, that is, the surface wind speed that corresponds to sustained soil erosion. Cowherd et al. (1985) included methods for determining the threshold friction velocity using an empirical relationship of measured threshold friction velocities to soil aggregate size distribution modes. Threshold friction velocities ranging from 25 to 450 cm s^{-1} are reported for aggregate particle size modes of 0.1 to 100 mm and are described by the equation

$$u_* = 65.5315 p^{0.417672}$$ (IV-10)

where
p = the soil aggregate particle size mode (mm)
u_* = threshold friction velocity (cm s^{-1}).

Suspension models also require estimates of the roughness height of the erosion surface. This parameter is related to the threshold friction velocity. Gillette et al. (1998) give a relationship between the roughness height for a nonsaltating surface and the threshold friction velocity (Figure IV-1). We fit these data to a polynomial given by Equation (IV-11).

$$z_o = 0.022086 - 0.0021654 u_* + 6.29869 \times 10^{-5} u_*^2 - 4.21098 \times 10^{-7} u_*^3 + 9.40549 \times 10^{-10} u_*^4$$ (IV-11)
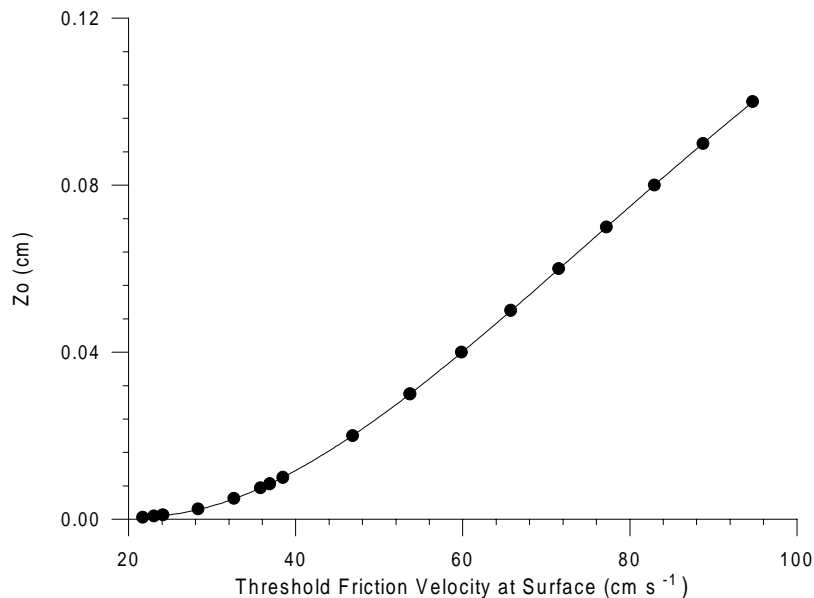
Figure IV-1. Aerodynamic roughness height, Z_o, as a function of the threshold friction velocity for a nonsaltating surface (source Gillette et al. 1998).

During wind erosion events where saltation occurs, the threshold friction velocity is known to increase by some value, Δu_* (Gillette et al. 1998). Experimental data in Gillette et al. (1998) showed that friction velocities on a saltating surface increased by about a factor of 2 for wind speeds of 18 m s^{-1} at a height of 1 m above ground level. During some of the high wind events, saltation is believed to have occurred on the 903 Area. The high plutonium concentrations in the soil within about 100 m of the 903 Area are attributed to movement of activity by saltation and not deposition of suspended particles. For the purposes of calibration, we ignored this effect in our model because detailed characterization of the surface roughness at the time of the events, which is required, is not available.

In general, soil characterization data for the 903 Area before paving are lacking and vehicle traffic, scraping and grading, and other earth moving activities may have altered the particle size distribution and surface roughness over time. To calibrate 903 Area releases with S-8 sampler air concentration measurements, we have treated these parameters stochastically in the source term model given by Equations (IV-6) and (IV-7). Our lack of knowledge about these parameters is reflected in the large distribution assigned to them. A value for the aggregate soil particle size mode was first selected. A surface threshold friction velocity was then determined using Equation (IV-10). Using the surface threshold friction velocity, the roughness height and threshold wind speed were calculated using Equations (IV-11) and (IV-9), respectively.

The overall objective of the calibration process was to calculate the total release of plutonium contaminated soil particles for each suspension event. Equation (IV-4) can be rewritten in terms of the source release model as

$$q_{cal} = \frac{C_{meas} \sum_{i=1}^{n} \left(\frac{u(z)_i}{u_t(z)} \right)^{\gamma}}{\sum_{i=1}^{n} \left(T_{f_i} \left(\frac{u(z)_i}{u_t(z)} \right)^{\gamma} \right)} \tag{IV-12}$$

where n = the number of meteorological time periods in a suspension event. Note that the terms C_S, X, and A cancel out.

Meteorology

Wind speed and direction measurements for the 24 highest release days were derived from NCAR recording stations located at Table Mesa, Kent (Figure IV-2), and other stations located at Jefferson County Airport and near Rocky Flats. NCAR data were recorded on strip charts and later digitized every 5-minutes for the six highest release events and hourly for the other high reading days at the S-8 sampler. Gust data were also obtained from the NCAR data. Details of the NCAR meteorological data are discussed in a previous section of this report. Meteorological data from the Kent station were used if available because this station was closest to Rocky Flats and was situated about the same distance from the foothills. Kent data were not available for all the release events, and in such cases, these data were inferred from Table Mesa data and the other sources. NCAR data were also compared to the Rocky Flats and Jefferson County Airport data for consistency. In cases involving large discrepancies among the data sets (e.g., January 6) a combination of data were used. We selected specific wind speed and wind direction values based on our understanding of meteorological conditions along the Front Range that are favorable for soil suspension. These conditions are typified by strong gusty winds from the west.

Stability classes were calculated hourly using the general classification scheme discussed in Pasquill (1961), Gifford (1961), and Turner (1964), which use wind speed, cloud cover, and ceiling height to estimate seven stability classes (A–G). Stability classes F and G were combined for these simulations because FDM only accepts six stability classes. Cloud cover and ceiling height data were obtained from the Jefferson County Airport. During the period the airport was closed (from 11 p.m. to 6 a.m. the following day), cloud cover was inferred from Denver Stapleton International Airport data when available, or we assumed that cloud cover conditions remained the same as the last hour recorded at Jefferson County Airport. Mixing depth has little bearing on air dispersion calculations when the receptor is close to the source; therefore, we assumed a value of 1000 m for all time periods. A summary of the meteorological data used in the calculations is presented in Appendix J.
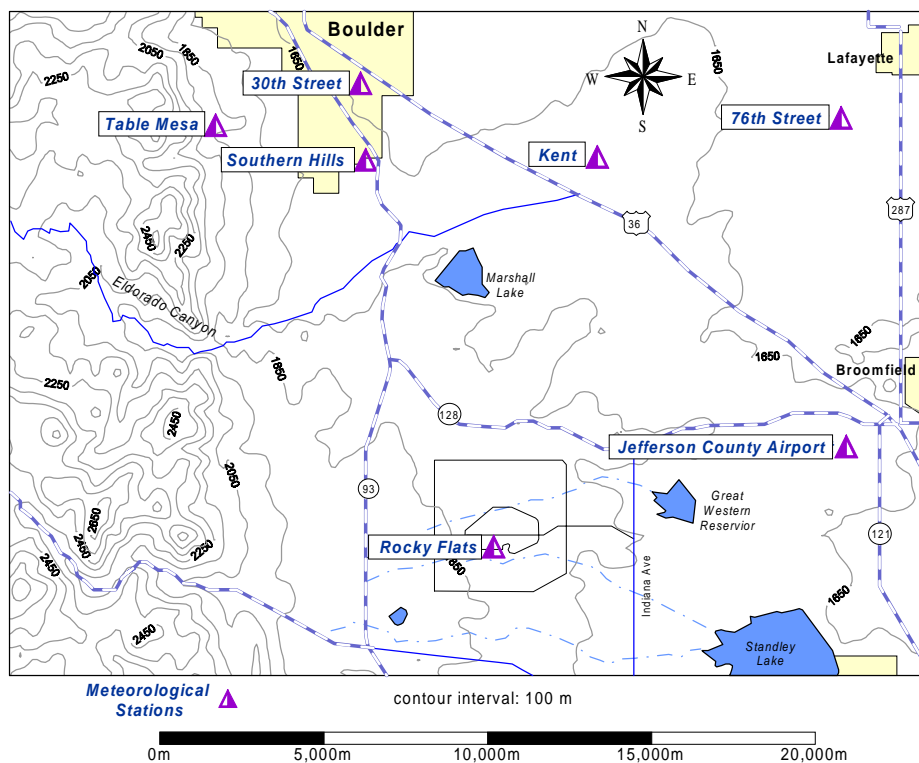
Figure IV-2. Location of the NCAR, Rocky Flats, and Jefferson County Airport meteorological recording stations.

Sampler Data

Measurements of TLLa at the S-8 sampler were used to calibrate the release and transport model using the methodology previously outlined for the 24 highest days recorded at the S-8 sampler. Air sampler results were reported in net counts per minute (cpm) measured on the filter for a given day. Count rates were converted to TLLa airborne activity concentrations by use of a total efficiency factor (which considers detector counting efficiency and alpha particle self-absorption) and the volume of air sampled. These parameters were discussed earlier in this report and were obtained from Rope et al. (1997). Additional corrections were made for inlet and filter collection efficiency, fraction of TLLa attributed to plutonium isotopes, and bias in the sampled air volume; they are discussed later in this section. The S-8 air sampler typically had the highest measured air concentrations of any of the samplers in the vicinity of the 903 Area because of its presence east of the area and the predominance of high winds from the west (Figure IV-3). Other samplers (S-1 through S-10) were considered less desirable for calibration for the following reasons:

- Concentrations at other samplers during the high wind events were at or near the detection limit.
- Other samplers (with the exception of S-50) were located west of the 903 Area requiring easterly winds for material from the 903 Area to be detected at these samplers.

- Measurements at other samplers may have been influenced by releases from nearby facilities other than the 903 Area.
- High readings observed at other samplers showed little correlation to high wind events.
- Other samplers (with the exception of S-6, S-7, S-50, and S-51) were located within an industrial complex containing many structures and obstructions. Dispersion modeling is highly uncertain in such an environment because of the complexity of air flows around building and other structures.
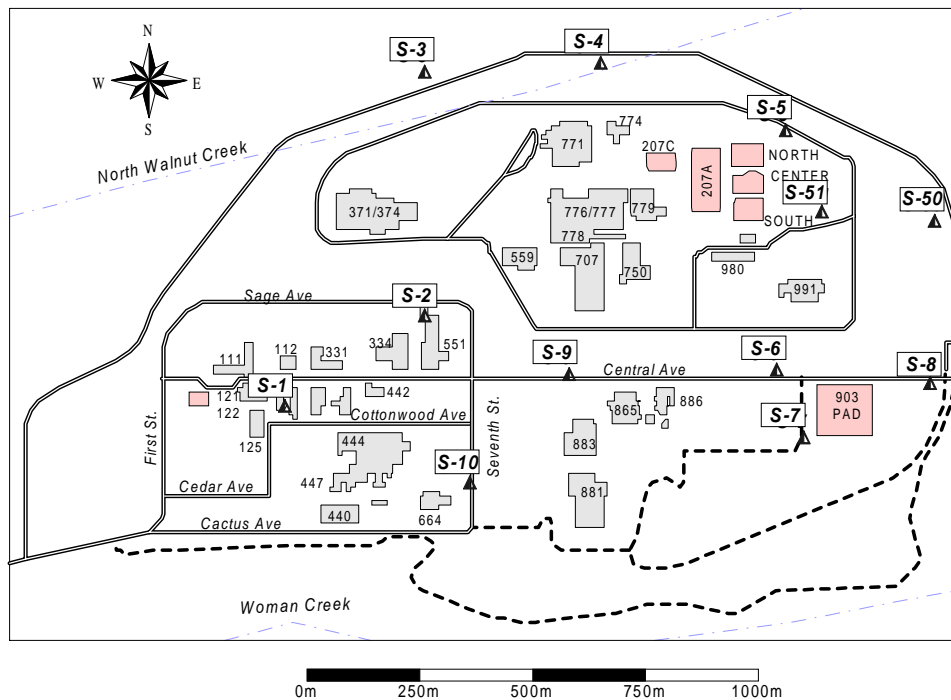
Figure IV-3. Locations of onsite ambient air samplers at the RFP relative to the 903 Area. Alpha activity in air at the S-8 sampler was most affected by the 903 Area. Building and sampler locations shown are those present circa 1971.

For these reasons, calibration of the 24 highest measurement days at the S-8 sampler used the S-8 sampler data only. Data from some of the other samplers were considered for annual average release estimates and are discussed in a later section.

Air filters were changed daily at 8:15 a.m., except for weekend or holiday periods, and were recorded for the day the sampling began. During weekends or holidays, samplers were allowed to collect over the entire period and filter analyses reported the net count rate for the entire weekend or holiday. We computed the average daily count rate by dividing the net count rate by the number of sampling days. If the average daily count rate appeared as one of the high 24 days, then the NCAR meteorological data for that sampling period were examined for high wind events. Three weekend periods appeared in the 24 highest S-8 measurement days: November 22–24 1968, January 3–5 1969, and April 11–14 1968. Review of the NCAR meteorological data

indicated that high wind events occurred on January 3, 1969, and April 12, 1968. A specific high wind event could not be identified for the November 22–24 period. For the January 3–5, 1969, and April 11–14, 1968, events, we assumed most of the activity detected by the sampler over the sampling period was due to single high wind days: January 3, 1969, and April 12, 1969. We assumed the count rate recorded for the entire sampling period was the count rate for an individual day. Calibration was performed using meteorological data for the specific high wind day and the count rate recorded for the entire sampling period. For the November 22–24 period, no specific high wind day could be identified. Therefore, the entire period was modeled using meteorological data that spanned the entire sampling period and the average S-8 sampler count rate measured for that period.

Conversion of Net Count Rate to TLLa Activity Concentration in Air

Conversion of the net count rate (gross count rate minus counter background) to TLLa concentrations in air (fCi m^{-3}) is given by

$$TLLa = \frac{cpm}{(TEF)(CF)(V)}$$ (IV-13)

where
TEF =   the total efficiency factor (counts per disintegration)
CF =   conversion factor from disintegrations per minute to fCi (0.00222 dpm fCi^{-1})
V =   the volume of air sampled (m^3).

Nominal values for TEF and V are 0.21 and 81.5 m^3, respectively (Rope at al. 1997). The TEF consists of two components: detector counting efficiency and alpha particle self absorption. As discussed earlier in this report, filter clogging could reduce the total volume of air sampled under dusty conditions. Corrections for the volume of air sampled are discussed in a separate section of this chapter.

Sampler Inlet Collection Efficiency Correction

As reported earlier, the samplers collected particles <30 μm AED, but the inlet collection efficiency varied as a function of particle size and wind speed. Rope et al. (1997) provides the inlet collection efficiency for particles ranging from 5 to 20 μm AED (Table IV-1). Measured air concentrations were corrected for inlet collection efficiency by computing the average inlet collection efficiency over the measurement period for each particle size.

To correct measured air concentrations for inlet collection efficiency, the airborne activity particle size distribution must be known. The airborne activity particle size distribution is the fraction of the airborne activity that is associated with a given particle size. Particle sizes are reported in AED, which is related to the physical diameter by

$$AED = \sqrt{\frac{\rho_s}{\rho_u}}\ d_p$$ (IV-14)

where
ρ_s = density of the airborne particle (g cm^{-3}),
ρ_u = unit density (1 g cm^{-3}),
d_p = the physical diameter of the particle (µm).

It is important to emphasize a basic premise underlying these calculations, that submicron plutonium particles (Hayden 1974) in the cutting oil were attached to larger soil particles, and it was those soil particles that were suspended and not the individual plutonium particles. Therefore, the properties of the soil particle govern the overall transport of plutonium and not the properties of the individual plutonium particles attached to the soil. For this reason, we used a particle density that represented soil in Equation (IV-14) (2.5 g cm^{-3}).

Table IV-1. Sampler Inlet Collection Efficiency as a Function of Particle Size and Wind Speed[a]

Aerodynamic Equivalent Diameter (µm)	Inlet Collection Efficiency (%)[b]		
	1 m s^{-1}	3 m s^{-1}	7 m s^{-1}
5	95–100	95–100	95–100
10	95–100	80–90	50–55
15	95–100	60–75	10–25
20	95–100	45–55	0

a. From Rope et al. 1997
b. The midpoint in the range was used in the calculations.

Measurements of activity particle size distributions at the RFP (Nathans et al. 1971; Langer 1983, 1986; Sehmel and Lloyd 1976; Volchok et al. 1972) are presented in Table IV-2. These measurements were performed after the 903 Area was paved and they represent resuspension from the contaminated field east of the 903 Area. Airborne activity particle size distributions may have been different from these during suspension events, but we have no measurements to confirm this. We had to assume that these size distributions represent conditions during the suspension events. Measurements indicated that, in general, most of the activity was associated with larger particles (>15 µm AED). Particles <3 µm AED represented about 1.7 to 14% of the total airborne activity collected by the samplers. Nathans et al. (1971) reported difficulties in sampling the smaller particle size fractions and studies documented in Sehmel and Lloyd (1976) only looked at particles up to 7 µm in diameter. Volchok et al. (1972) reported an average respirable activity size fraction of 0.25, but fractions of larger particle sizes were not reported. Langer defines respirable particles as <3 µm AED, but the American Conference of Government Industrial Hygienists reports that particles as large as 10 µm AED may be respirable. However, only 1% of the airborne mass of 10 µm particles actually gets into the deep lung region.

Activity particle size fractions reported in Langer (1986) were measured over a 2 year period and included three measurement heights; 1, 3, and 10 m. Sehmel and Lloyd's work also included multiple sampling heights. We used data at the 1-m level because that was the approximate height of the S-8 sampler inlet. Langer reported average ^{239}Pu concentrations of 3.9, 18, and 91 aCi m^{-3} for the <3 µm, 3–15 µm, and >15 µm particle size classes, respectively. The standard deviations of the averages were close to the mean value, indicating considerable

variation in the activity collected. Also, note that the 3-15 µm size range tended to have the lowest fraction of radioactivity.

Table IV-2. Airborne Activity Particle Size Distribution Measurements from the Field East of the 903 Area at 1 m Above Ground Level

Particle size (µm AED)	Percentage of total activity associated with each particle size				
	Langer (1986)[a]	Langer (1983)	Nathans et al. (1971)[b]	Volchok et al. (1972)[c]	Sehmel and Lloyd (1976)[d]
<3	3.50	1.69	13.00	0.25	43.56
3–10	7.75[a]	0.00	3.20	---	56.44
10–15	7.75[a]	6.67	1.00	---	---
>15	81.00	91.65	82.80	---	---

[a] Particle sizes were reported in only three categories (<3 µm, 3–15 µm, and >15 µm). The 3–15-µm category was split evenly between the 3–10-µm and 10–15-µm categories.

[b] Based on the reported lognormal particle size distribution having a geometric mean of 9.5 µm and geometric standard deviation of 2.05.

[c] Only the respirable fraction was reported (<10 µm AED).

[d] Percentages represent the fraction of activity less that 7 µm that falls within the <3-µm and 3–7-µm category.

Particle size distributions were treated as a stochastic variable in our calculations and are discussed in a later section of this chapter. For deterministic calculations, we used the size fractions derived from Langer (1986) because these measurements were made over 2 years and, therefore, best represent average conditions observed at the RFP. Langer reported particle sizes in three categories (<3 µm, 3–15 µm, and >15 µm). The 3–15-µm category was split evenly between the 3–10-µm and 10–15-µm categories.

The particle size distribution also assumes that the >15-µm size category includes particles up to 30 µm AED. Rocky Flats ambient air samplers that operated during the 1960s and 1970s were very inefficient at collecting particles with diameters greater than >30 µm AED; therefore, releases of particles >30 µm cannot be calibrated using the methodology presented.

The total airborne plutonium concentration (C_T) can be described by the sum of the concentrations of each individual particle size class (C_i) and is given by

$$C_T = \sum_{i=1}^{n} C_i \qquad \text{(IV-15)}$$

where n = the number of individual particle size classes considered. If we know the fraction of activity in each particle size class, then Equation (IV-15) can be rewritten as

$$C_T = C_T f_1 + C_T f_2 + C_T f_3 ... + C_T f_n \qquad \text{(IV-16)}$$

where f_1, f_2, f_3 ... f_n represents the fraction of the total activity in each particle size class. If the inlet collection efficiency for each particle size class is known, then the corrected total concentration (C_{TC}) can be computed by

$$C_{TC} = \frac{C_T f_1}{ICE_1} + \frac{C_T f_2}{ICE_2} + \frac{C_T f_3}{ICE_3} ... + \frac{C_T f_n}{ICE_n} = C_T \sum_{i=1}^{n} \frac{f_i}{ICE_i} \qquad \text{(IV-17)}$$

where ICE_1, ICE_2, ICE_3, ...ICE_n = the inlet collection efficiency over the sampling period for each particle size class. The term, C_{TC} is equivalent to C_{meas} in Equation (IV-12). The inlet collection efficiency must represent the overall inlet collection efficiency for a given particle size over the 24-hour sampling period. We take the average of the inlet collection efficiency computed using the data in Table IV-1 and the measured wind speed for each meteorological period to represent the overall inlet collection efficiency during the sampling period. Thus, ICE is given by

$$ICE_i = \frac{\sum_{j=1}^{n} CE_{i,j}(u)}{n} \qquad \text{(IV-18)}$$

where $CE_{i,j}(u)$ = the inlet collection efficiency for the ith particle size, j^{th} meteorological period, and wind speed, u; and n = the number of meteorological periods during a sampling interval. Inlet collection efficiencies as a function of wind speed and particle size are given in Table IV-1.

Filter Collection Efficiency and Fraction of TLLa Attributed to Plutonium

Rope et al. (1997) estimated the filter collection efficiency to range from 90 to 100%. For these calculations, we have assumed a constant value of 95%. Of the TLLa activity collected, 20 to 80% was attributed to plutonium isotopes, with the remainder being primarily americium and uranium. This fraction was treated stochastically in our calculations and is discussed in a subsequent section on treatment of uncertainty.

Filter Clogging and Air Volume Correction

Filter clogging by dust particles has the potential to reduce the volume of air sampled during a sampling interval by reducing the air flow rate across the filter. An overview of the impact of filter clogging on the volume of air sampled is presented in Section III of the Task 4 report (Rope et al. 1997). At the time of highest releases from the 903 Area, the volume of air sampled was not measured, but it was computed as a fixed value (81.5 m³ d⁻¹) for all samplers from the sample collection time (1440 minutes) and the specified flow rate of the sampler (0.0566 m³ min⁻¹). Rope concluded that under normal operating conditions, filter clogging for daily samples was not expected to be a problem because the samplers were calibrated under such conditions. Normal operating conditions are interpreted to mean days when the dust loading was not abnormally high. High dust loading typically occurs during high wind events and may be enhanced by human

activities. High winds are defined here as events having sustained winds in the gale (15 m s⁻¹) to strong gale (20 m s⁻¹) range or greater (Neiburger et al. 1971).

No direct measurements of dust loading exist for the period of interest. Therefore, we must rely on sampler data and meteorological records to provide some clue as to the magnitude of the dust loading on any given day. Evidence from Langer (1986) suggests there is a relationship between dust loading and airborne plutonium activity concentrations. Figure IV-4 shows good correlation ($r^2 = 0.85$) between dust loading and the airborne plutonium activity concentration in data taken over a 2-year period at the RFP in the early 1980s. This evidence supports our conceptual model for releases from the 903 Area that attributes activity releases from the 903 Area to suspension of contaminated soil particles and not direct suspension of individual plutonium particles. Based on Langer's measurements, we would expect correlation between the net count rate observed at the S-8 sampler and the amount of dust loading on any given day.
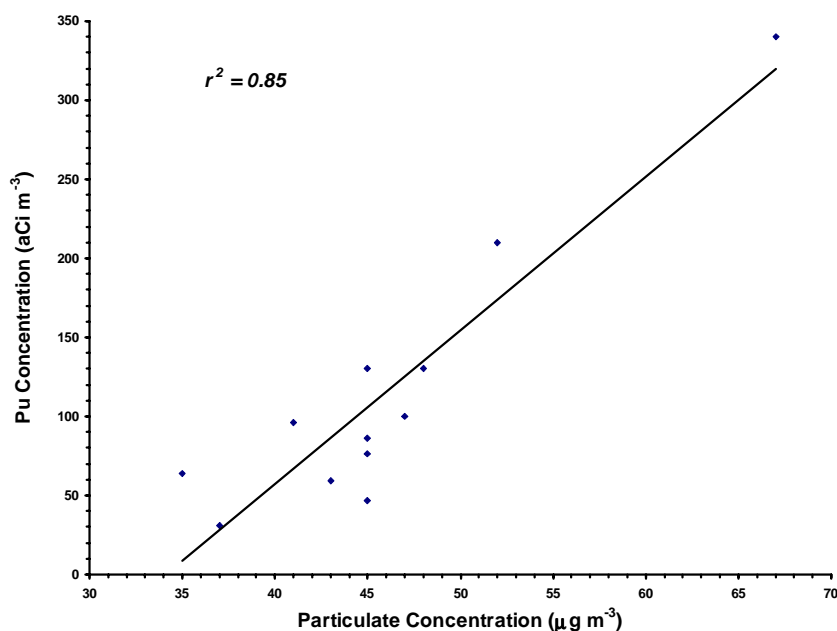
Figure IV-4. Monthly average plutonium concentration as a function of particulate concentration measured in the field east of the 903 Area during 1983 to 1984 (Langer 1986).

Our task is to define a net count rate that would indicate high dust loading conditions. We approached this problem by first observing the distribution of net count rates at the S-8 sampler from 1964 to 1969 and an equally long record of wind speed measurements taken at Rocky Flats. Complete meteorological records were not available for the time frame of interest (1964–1969); therefore, surrogate data taken at Rocky Flats from 1989 to 1993 were used. The U.S. Environmental Protection Agency (EPA) considers 5 years of meteorological data to be representative of long-term conditions. Frequency distributions of these data are presented in Figure IV-5. Note that extremely high values of wind speed and count rate occur rarely. Because both these data sets cover about the same amount of time (≈5 years), we have assumed the frequency of occurrence of extreme values to be related. That is, the frequency of wind speeds in

excess of a given value corresponds to a count rate that exhibits approximately the same frequency of occurrence.

Wind speeds in excess of 15 m s^{-1} occur about 0.6% of the time (or wind speeds less than 15 m s^{-1} occur 99.4% of the time). Wind speeds less than 15 m s^{-1} are assumed to be associated with typical dust loading conditions at Rocky Flats, and no correction to the air volume sampled is necessary. If high winds are related to high dust loading (and, therefore, high count rates observed at the S-8 air sampler), then the frequency of high winds is expected to equate to the same frequency of high count rates associated with high dust loading conditions. Applying the frequency of winds <15 m s^{-1} (99.4% of the time) to the distribution of observed count rates yields a count rate of 40 cpm. In other words, the count rate corresponding to the 99.4 percentile is 40 cpm. We take this value to represent the cutoff where air volume correction is necessary. That is, count rates below this value are not corrected for sampled air volume.
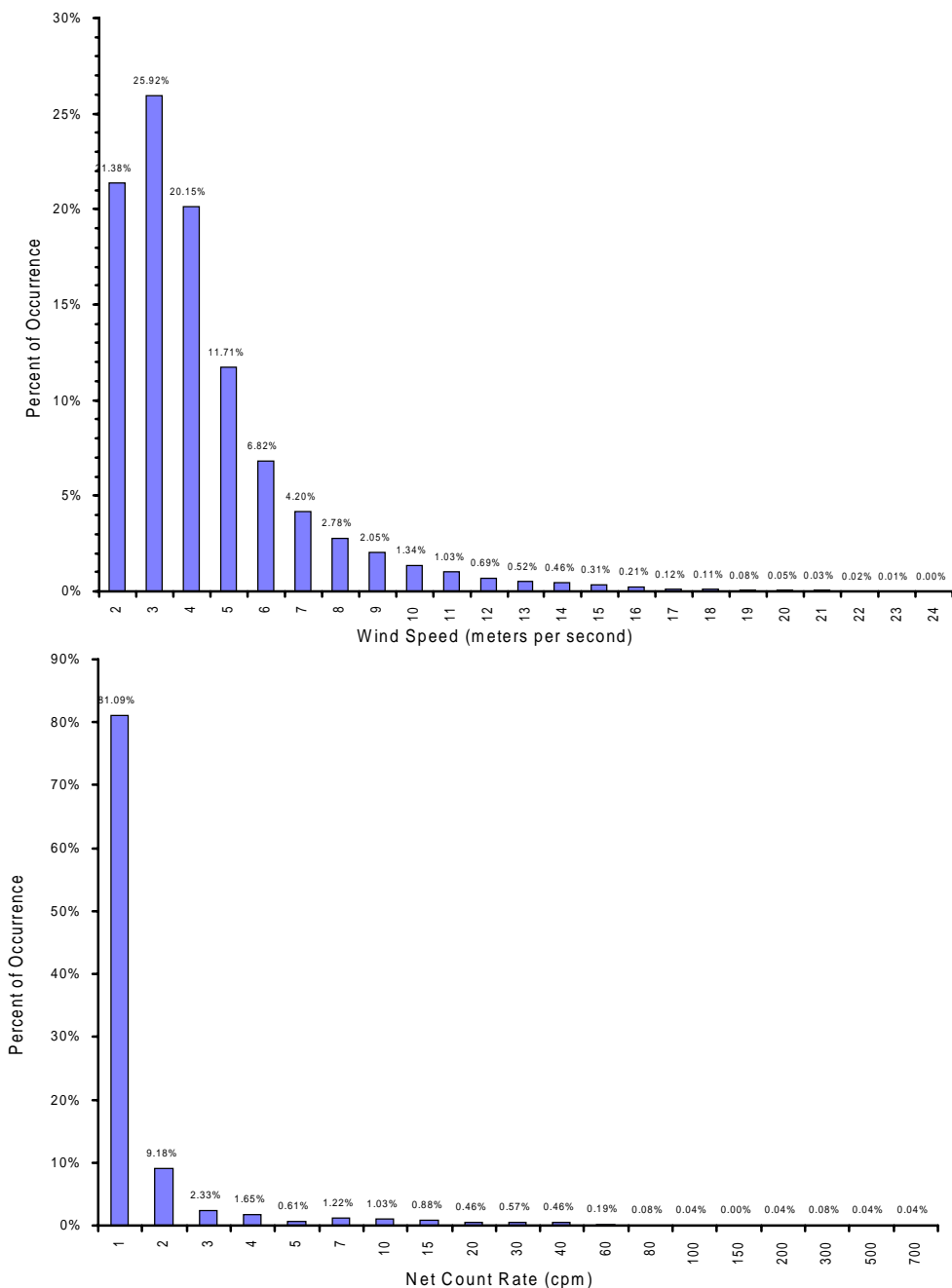
Figure IV-5. Frequency distributions of wind speed (above) and net count rate at the S-8 sampler (below). Wind speed data were 15-minute averages taken at the RFP 61-m tower from 1989 to 1993. S-8 air sampler data includes the years 1964–1969.

All but 5 of the 24 highest S-8 days had count rates in excess of 40 cpm. These five days include March 3, November 25, and December 11, 1968, and January 2 and February 24, 1969. For these 5 days, maximum wind speeds were 16, 15, 12, 24, and 11 m s^{-1}, respectively. It is unknown why the January 2, 1969, day did not result in a higher count rate at the S-8 sampler. If we ignore January 2, the data suggest that lower maximum wind speeds were associated with lower count rates observed at the S-8 sampler.

   We now equate the net count rate to sampled air volume. First, we envision the air sampler to operate as illustrated in Figure IV-6. This figure depicts the volume of air sampled as a function of time under different steady-state dust loading conditions. Under optimum conditions (no significant dust loading), the flow rate is assumed to remain constant over the collection period and the volume of air sampled increases linearly with respect to time (curve labeled C0). Under higher dust loading conditions (curves labeled C1–C5), the flow rate remains constant until sufficient mass has accumulated on the filter so that the flow rate decreases. The curve labeled C5 represents the highest dust loading conditions. Note that higher dust loading conditions affect the sampler flow rate at earlier times during the sampling period (these times are labeled T1–T5 in Figure IV-6). Optimum conditions are interpreted here to represent typical dust loading conditions where any decrease in the sampled flow rate was accounted for in the calibration process. The count rate associated with typical dust loading conditions was estimated to be 40 cpm as determined earlier.
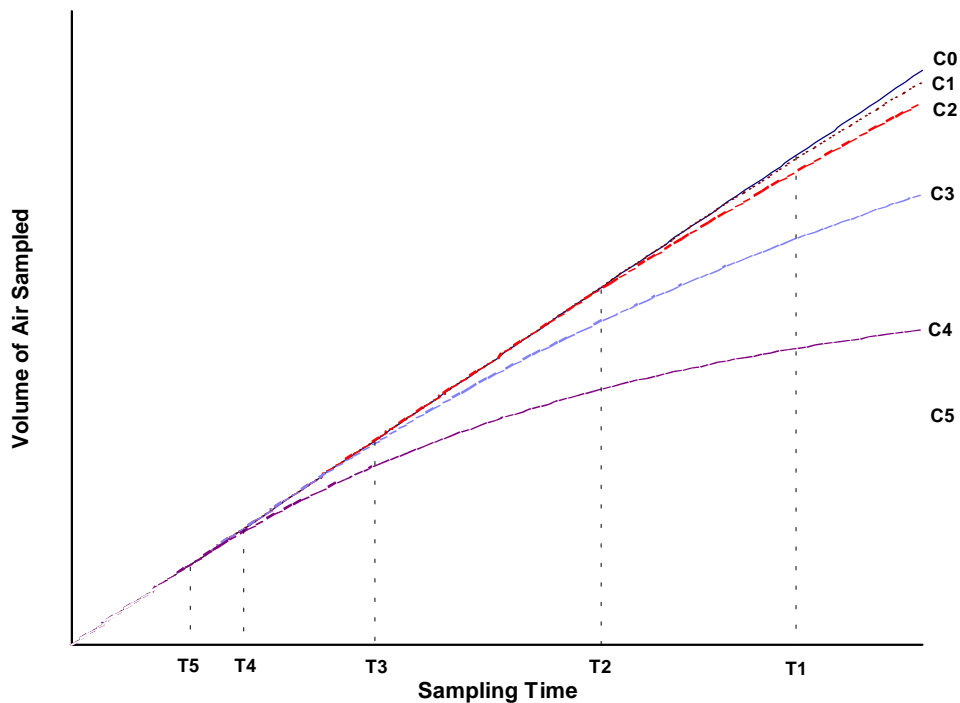
Figure IV-6. Volume of air sampled as a function of sampling time for different steady-state dust loading conditions. The curve labeled C0 represents minimal dust loading and the flow rate remains constant throughout the sampling period. The curves labeled C1–C5 represent increasingly higher dust loading conditions and the sample flow rate falls off with time. The times T1–T5 represent the time when the sampler is affected by dust loading on the filter.

   Rope et al. (1997), citing Lockhart et al. (1964), estimates that under typical dust loading conditions at the RFP, there might have been an 8% decrease in flow for the HV-70 filter paper and 24% for the Whatman 41 paper, both of which were used in the low volume samplers. The

type of filter paper used on the sampler (HV-70 or Whatman 41) was not recorded. If the flow rate is approximated by a first-order process, we can describe its time-rate of change by

$$\frac{dF}{dt} = -kF \tag{IV-19}$$

where F = the flow rate (m^3 min^{-1}) and k = a rate constant describing the fraction decrease in flow rate per unit time (min^{-1}). Note that k depends on the mass loading rate on the filter and is related to the net count rate observed on the filter. Equation (IV-19) applies only after sufficient mass has accumulated on the filter so that the flow rate is affected. Before that, the flow rate is constant. The solution to Equation (IV-19), with initial conditions $F = F_o$ when $t = 0$, is

$$F = F_o\, e^{-k\,t} \tag{IV-20}$$

where F_o is the nominal flow rate (0.0566 m^3 min^{-1}). The term t is given by $t_s - t_o$, where t_s is the sample time and t_o is time when sufficient mass has accumulated on the filter so that the flow rate is affected. The rate constant k is determined from data in Rope et al. (1997) and the value of t_o. For the HV-70 paper, $k = 0.08/t_o$ and for the Whatman 41 paper is $k = 0.24/t_o$.

   The volume of air sampled before time t_o is simply $F_o\,t$, and the volume of air sampled after t_o is the integral of Equation (IV-20). The total volume of air sampled during time t_s is given by

$$V = F_o t_o + \frac{F_o}{k}\left(1 - e^{-k(t_s - t_o)}\right) \tag{IV-21}$$

for $t_S > t_O$ and $F_O\,t_S$ for $t_S \leq t_O$. The term t_O is determined from the net count rate, the sampling time, and the net count rate associated with typical dust loading conditions and is given by

$$t_o = \frac{c_o}{c\,/\,t_s} \tag{IV-22}$$

where c_O = count rate associated with typical dust loading conditions (cpm), c = net count rate of the filter (cpm), and t_S = the sample time (min).

   Figure IV-7 shows the volume of air sampled as a function of time for different net count rates and Whatman 41 filter paper. Note that for a net count rate of 40 cpm, the flow rate is constant throughout the 24-hour sampling period. For higher count rates, the flow rate falls off with time.

   One of the critical assumptions in this approach is that steady-state dust loading conditions persist for the sampling period. The suspension models introduced earlier coupled with the measured wind speed data would suggest dust loading was highly variable during the 24-hour sampling time. We acknowledge this deficiency in the methodology presented here, but we think the methodology represents the overall behavior of the system and incorporating time-dependent dust loading would not significantly affect the results of the calculation. The rationale behind this

decision is based in part on the <u>sensitivity analysis</u> discussed later in this report. In addition, preliminary calculations using the time-dependent dust loading to compute the volume of air sampled showed little difference from values based on a steady-state dust loading. For example, using the January 30, 1969, date and assuming HV-70 paper, the calculated air volume sampled was 49 m^3 assuming steady-state dust loading conditions and 44 m^3 using time-dependent dust loading. Based on these calculations and the sensitivity analysis, we have assumed steady-state dust loading conditions in our calculations.
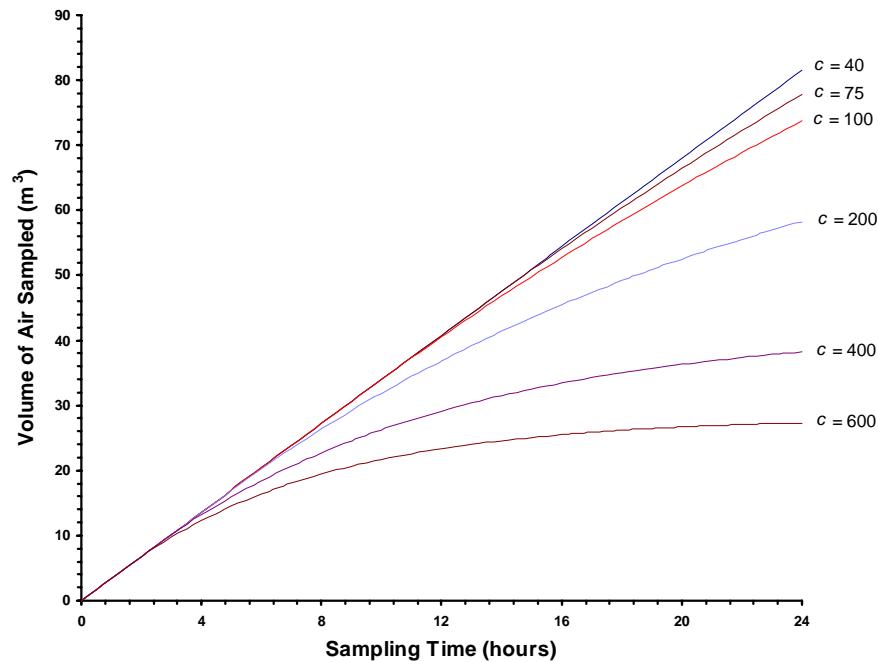
Figure IV-7. Volume of air sampled versus time for Whatman 41 filter paper and several different observed net count rates (c in cpm). Note that for $c = 40$ cpm, the sample flow rate is constant throughout the sampling period.

Treatment of Uncertainty

Numerous sources of uncertainty exist in a calculation of this type, and they include both model uncertainty and parameter uncertainty. The complexity of the suspension process has led to the development of empirical models such as those used in this analysis. Validation of these models under controlled conditions typically results in model predictions that are within about a factor of 2 of the observations. For example, <u>Gillette et al.</u> (1998) showed predicted versus measured roughness heights differed by about a factor of 1.8 for a highly instrumented site in Owens Valley, California. Less than adequate controls and data exist to model releases from the 903 Area using suspension models alone, and for that reason we have chosen to calibrate our release estimates using air monitoring data. By doing so, we hope to reduce the model uncertainty; however, many other sources of uncertainty still exist in the calculation.

In the uncertainty analysis, we have not considered natural variability in a parameter value, but instead we focused on the uncertainty in estimating its true but unknown value under a given

set of conditions. We acknowledge that for some parameters, we have used data that may represent a parameter's natural variability as a means of estimating its uncertainty distribution. In such cases, we have subjectively expanded the distribution to account for our lack of knowledge. In all cases, the distributions presented are intended to represent our confidence in assigning a particular value to a parameter.

For this analysis, the following parameters were treated stochastically:

- Aggregate particle size mode
- Wind speed exponent
- Fraction of TLLa that is attributed to plutonium
- Atmospheric transfer function
- Airborne activity particle size distribution
- Volume of air sampled
- Total efficiency factor.

Counting error (because of the random nature of radioactive decay) was also considered a source of uncertainty. However, for the high release days, sample count rates were relatively high resulting in a low counting error. For example, Rope et al. (1997) shows that for a sample with a net count rate of about 100 cpm, the coefficient of variation due to counting error is about 3%. Count rate uncertainty during high release days was considered negligible compared to the other sources of uncertainty, and for this reason, was not included in the uncertainty analysis.

A distribution was assigned to each of the parameters listed above. Note that the area of contamination, the suspension constant, and plutonium concentration in soil were not considered in the uncertainty analysis because these terms cancel out in the calibration process. Uncertainty was propagated through the calculations using Monte Carlo sampling techniques and a FORTRAN program written specifically for this task. Random sampling routines for various distributions were adapted from Press et al. (1992). Distributions of calibrated releases were generated from 1000 individual realizations. For each realization, the parameters listed above were each sampled once, a calibrated release estimate was calculated, and the process repeated until all 1000 realizations had been completed. The rationale behind assigning distributions to each parameter follows. Each parameter was considered independent of one another.

Aggregate Particle Size Mode

The aggregate particle size mode is used to determine the threshold friction velocity using data presented in Cowherd et al. (1985) and the surface roughness height. The methodology for determining this parameter is based on the work of Chepil (1952), which described a field procedure using a soil sample and sieves to determine the aggregate particle size mode. Soil from the 903 Area was disturbed at different times and would likely exhibit a different particle size distribution than undisturbed, native soils. Unfortunately, this measurement was not performed before paving, so an estimate of the distribution of this parameter must be made. Nathans et al. (1971) measured the soil particle size distribution near the 903 Area at several undisclosed locations. Samples collected on sticky papers gave approximate lognormal distributions with a geometric mean (GM) of 1.9 μm and a geometric standard deviation (GSD) of 2.6. The method used was admittedly not the best, and it is unclear how this relates to the aggregate particle size mode because nonerodible elements may have been excluded from this measurement.

We calculated aggregate particle size modes for various soils and coal mining wastes (Table IV-3) based on threshold friction velocities reported in EPA (1995) and Equation (IV-10). These data were used as the basis for defining a distribution of aggregate particle size modes. It may be argued that it would be easy enough to make a field measurement of the particle size mode. However, a field measurement may not represent the aggregate particle size mode during the high suspension events from the 903 Area. We have chosen instead to treat this parameter with rather large uncertainty.

Table IV-3. Aggregate Particle Size Modes for Soils and Mining Wastes[a]

Material	Threshold friction velocity u_* (m s^{-1})	Aggregate particle size mode p (mm)[b]	Roughness height z_o (cm)
Overburden	1.02	2.9	0.3
Scoria	1.33	5.5	0.3
Ground coal	0.55	0.65	0.01
Uncrusted coal	1.12	3.6	0.3
Scraper	0.62	0.88	0.06
Fine coal dust	0.54	0.63	0.2

[a] Source: EPA (1995).
[b] Calculated using Equation (IV-10) and u_*.

Based on the data in Table IV-3, we assigned a truncated lognormal distribution to this parameter. The GM and GSD represent the GM (1.7 mm) and GSD (2.7) of the five materials. The lower tail of the distribution was truncated at 0.1 mm and the upper tail was truncated at 10 mm. The lower limit represents the minimum particle size mode reported by Cowherd et al. (1985). Cowherd reported threshold friction velocities for particle size modes up to 100 mm. The distribution was truncated at 10 mm because the measured particle size distribution reported by Nathans et al. (1971) suggested that the soil did not contain large particles (on the order of 100–1000 μm). Based on his particle size distribution, the largest particle sizes are on the order of 0.05 mm. Smaller aggregate particle sizes may also result from repeated vehicular traffic and earth moving activities that were know to occur on the 903 Area.

Wind Speed Exponent

Particle flux has been shown to increase as a function of the ratio of the wind speed to the threshold wind speed raised to a power, as shown in Equation (IV-6). The wind speed exponent is an empirical parameter. The higher the value, the higher the susceptibility of the soil surface to erosion. Estimates of the wind speed exponent have varied from 3 to 9. For native soils, a value between 3 and 4 was recommended by Gillette (1977, 1997). Other researchers (Langer 1983; Sehmel and Lloyd 1976) studying resuspension in the field east of the 903 Area confirmed this range and estimated wind speed exponents in the range of 3 to 4. During the periods of highest suspension, the 903 Area was sparsely vegetated and at times disturbed by vehicle traffic and earth moving equipment. For this reason, we looked at a broader range of wind speed exponents

that included higher values. The wind speed exponent was assigned a triangular distribution with a minimum of 3, a maximum of 5, and a most likely value of 4.

Fraction of TLLa Attributed to Plutonium Isotopes

Plutonium isotopes were reported to account for 20 to 80% of the TLLa measured at the samplers (Rope et al. 1997). We assumed a triangular distribution with a minimum of 20%, a maximum of 80%, and a most likely value of 40%.

Atmospheric Transfer Function

The atmospheric transfer function provides the link between the release model and the measured concentration at the air sampler. Uncertainty in the transfer function is directly proportional to atmospheric model prediction uncertainty. Uncertainty in the predicted concentration was represented by a multiplicative correction factor that was applied to modeled air concentrations. The factor represents the combination of the overall expected uncertainty in a model prediction because of both parameter and model uncertainty. The correction factor was based on the inverse of the distribution of predicted-to-observed (P/O) ratios reported in the literature for the Gaussian Plume model, the model upon which FDM is based.

Miller and Hively (1987) report the GSD of P/O ratios for the Gaussian Plume model at highly instrumented, flat-terrain sites for short duration releases and receptors <10 km from the source to range from 1.1 to 1.2. Model evaluation using the 1991 Winter Validation Tracer Study at Rocky Flats (Rood 1997a) indicates that P/O ratios are lognormally distributed. Nine-hour average tracer concentrations measured at 140 samplers located on 8 and 16-km radius circles surrounding the release point were compared with model predictions from five models, including a form of the Gaussian Plume model (Industrial Source Complex Code [EPA 1992]). A paired comparison of predicted and observed concentrations at the 8-km samplers showed the distributions of P/O ratios for the Gaussian Plume model had a GM of 1.1 and a GSD of 6. However, an unpaired comparison of the 24 highest predicted and observed concentrations at the 8-km samplers showed the distribution of P/O ratios to have a GM of 1.2 and a GSD of 2.2. The relatively high GSD for the paired comparisons is due in part to terrain complexities and curvilinear plume trajectories that are not accounted for in the Gaussian Plume model. An unpaired comparison removes plume trajectory from the equation, and model performance improved substantially. Complications related to complex terrain and curvilinear plume trajectories are not likely to affect predicted concentrations for a receptor close to the source, and we would expect the variance in the P/O ratios to be closer to that observed in the unpaired comparison. Other researchers (Robertson and Barry 1989) report GSD values for elevated releases (60 m) and relatively flat terrain to range from 3.4 to 2.1 for averaging times of 12 and 72 hours, respectively, and receptors distances from 1–5 km.

No one study is entirely relevant to this work; however, we expect the distribution of P/O ratios to have a GSD in the range of 2–3 based on the studies cited. For this assessment, we have chosen a lognormal distribution of P/O ratios having a GM of 1.0 and a GSD of 2.5. The GM value of 1.0 assumes no bias in the model. The multiplicative correction factor would also be lognormally distributed with a GM of 1 and GSD of 2.5.

Activity Particle Size Distribution

The activity particle size distribution is a measure of the fraction of the total airborne plutonium activity attached to a given soil particle size. This parameter was treated stochastically for the sampler inlet collection efficiency correction and deterministically for the transfer function calculation. Comparing transfer function values at the S-8 sampler for a large range of different particle size distributions and meteorological conditions showed the maximum difference to be no greater than 3%. The additional complexity needed to treat particle size distributions stochastically in the transport model was not warranted based on these findings.

Measurements of airborne activity particle size distributions taken at the RFP are presented in Table IV-2. These measurements provided the basis for assigning uncertainty to the particle size distribution. Ten particle size distributions were defined (Figure IV-8). The first distribution was weighted toward the smaller size fractions and the tenth distribution was weighted toward the larger size fraction. Particle size distributions between the first and tenth distribution (distributions 2–9) represented a gradation between those at either end. Distributions were represented by the fraction of activity in four particle size classes: <3 μm, 3–10 μm, 10–15 μm, and >15 μm AED.

The maximum size fraction for <3 μm particles reported in Table IV-2 and measured by Nathans et al. (1971) was 0.13. Assuming data from Volchok et al. (1972) represented particles <10 μm, we assigned the 3–10 μm size fraction to be the difference between the <3-μm (defined by Nathans) and <10-μm fraction (defined by Volchok) or 0.25 – 0.13 = 0.12. We assigned the 10–15-μm fraction the minimum measured value reported in Table IV-2 (0.01) and the largest size fraction (>15 μm) made up the remainder (0.74). This distribution was weighted toward the smaller size fractions and is identified as distribution 1 in Figure IV-8.

The other end of the distribution spectrum is labeled as 10 in Figure IV-8. The maximum >15-μm and 10–15-μm size fractions reported in Table IV-2 and measured by Langer (1983) were 0.92 and 0.067, respectively. These size fractions were coupled with the minimum reported size fraction for the <3 μm-size fraction. The minimum <3-μm size fraction was 0.017. This size fraction was reduced to 0.01 and the 3–10-μm size fraction made up the remainder (0.003) of the distribution. This distribution was weighted toward the larger size fractions.

The 10 particle size distributions used in the Monte Carlo simulations are illustrated in Figure IV-8. A uniform sampling scheme was applied to the 10 distributions so that there was equal probability that any 1 of the 10 distributions would be used in a given trial. In other words, in any given trial within the overall Monte Carlo simulation, 1 of the 10 distributions would be randomly selected and used in the calculation of inlet collection efficiency.
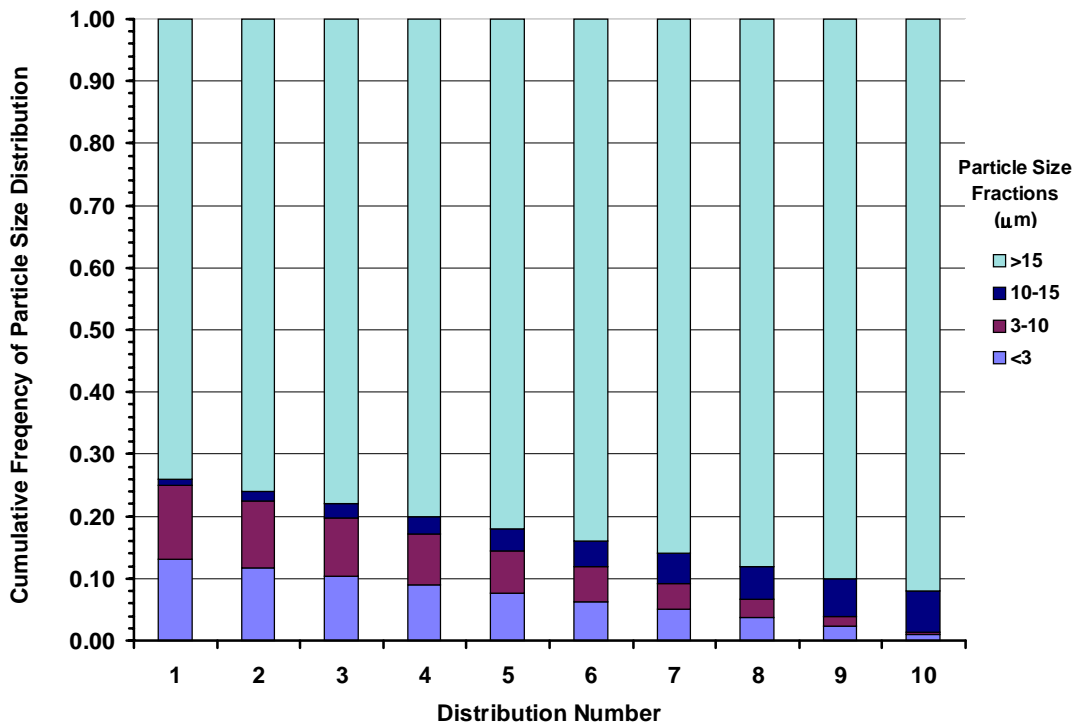
Figure IV-8. Activity particle size distributions used in the uncertainty analysis. A uniform sampling scheme was applied so that there was equal probability that any 1 of the 10 distributions would be used in a simulation.

Volume of Air Sampled

Filter clogging and its effects on the volume of air sampled were discussed in a previous section. Site-specific data indicated a relationship between the airborne particulate mass and the airborne activity concentration. High wind events have the potential to suspend large quantities of particulate matter, and under such conditions, the particulate mass collected on the filter may have impacted the sampled air volume by reducing the sampler flow rate over time. Corrections to the volume of air sampled were made based on the net filter count rate, the count rate associated with normal dust loading conditions, and performance of the filter paper under normal operating conditions. Two types of filters were used in the onsite samplers deployed at the RFP during the 1960s: Whatman 41 and HV-70. Rope et al. (1997), citing Lockhart et al (1964), estimates that under typical dust loading conditions at Rocky Flats there might have been an 8% decrease in flow for the HV-70 filter paper and a 24% decrease for the Whatman 41 paper. The type of filter paper used in any given day of sampling was not recorded; therefore, we have treated this fraction stochastically. A uniform distribution was assigned having minimum and maximum values of 0.08 and 0.24, respectively. These values are used to calculate the rate constant k in Equation (IV-21).

Total Efficiency Factor

The total efficiency factor is discussed in Section IV of the *RAC* Task 4 report (Rope et al. 1997). The total efficiency factor represents the fraction of the total alpha particles emitted by the sample that is measured by the detector. A value of 0.21 was estimated by RFP personnel to convert counts per minute to activity (in disintegrations per minute). Based on recommendations in Rope et al. (1997), a triangular distribution was assigned to this parameter with a minimum, most likely, and maximum value of 0.15, 0.21, and 0.25, respectively.

Summary of Uncertainty

Seven parameters were considered in the uncertainty analysis. These parameters were the aggregate particle size mode, wind speed exponent, fraction of TLLa that is attributed to plutonium isotopes, atmospheric transfer function, airborne activity particle size distribution, volume of air sampled, and total efficiency factor. Uncertainty in the atmospheric transfer function was represented by a lognormally distributed multiplicative correction factor. This factor represented the inverse of the P/O ratio (or the observed-to-predicted ratio) and was based on values reported in the literature and site-specific tracer studies. Each Monte Carlo trial resulted in a single estimate of the total activity released from the 903 Area for a given day. For each trial, values were selected for each stochastic variable, the simulation was run, and the result stored. This process was continued until all trials for a given day were complete. Results obtained in this manner were then post-processed and percentiles were determined from the output distribution. Distributions of release estimates reported here were based on 1000 trials.

Representation of the 903 Area

The 903 Area comprises a 13,700 m^2 area; however, the entire area was not contaminated. Aerial photographs indicated oil spill areas to comprise an area of about 6000 m^2 within the overall 903 storage area. For modeling releases from the 903 Area, the contaminated area was discretized into three sources representing the oil spill areas as seen in the aerial photographs (Figure IV-9). The contaminated area was assumed to be a smooth flat surface of sparse vegetation. The erosion potential was assumed to be unlimited for all releases. That is, it was not limited by the plutonium inventory in the soil or by removal of the erosion surface from earlier erosion events. For calculating releases using the suspension equation alone, additional assumptions about the quantity and depth of plutonium in the soil were also made. Release estimates made with the suspension equation and reported in this report used a bulk density of 1.5 g cm^{-3}, a contamination depth of 1.5 cm, and a 10 Ci plutonium inventory in soil. Estimates of the depth of contamination ranged from 1 to 3 cm and plutonium inventory in soil from 6.1 to 57.4 Ci (Meyer et al. 1996). These calculations were made for comparison with the calibrated release estimates.
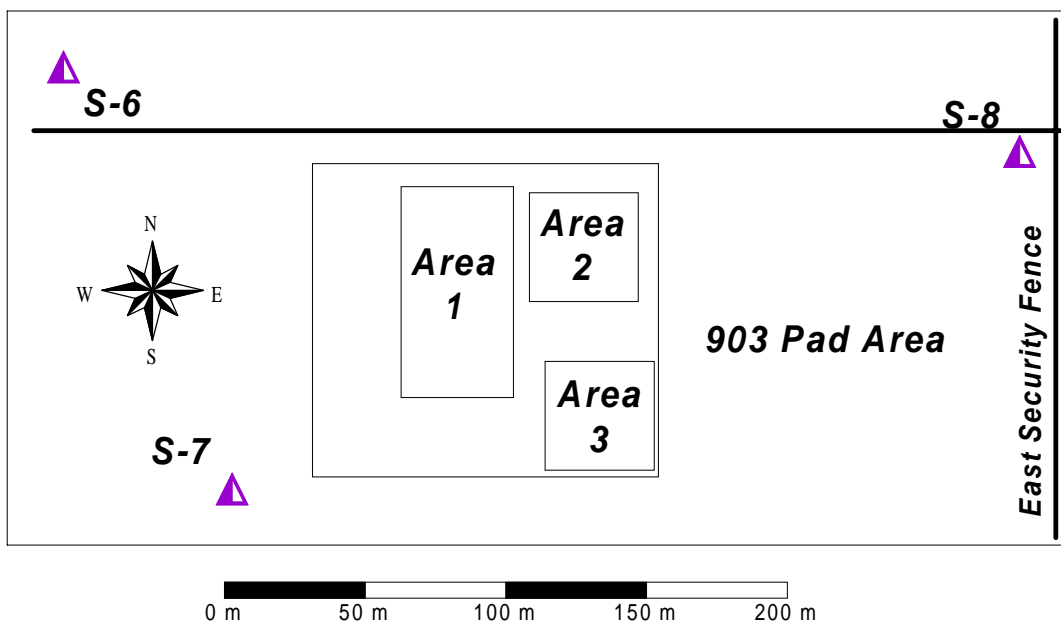
Figure IV-9. Representation of the plutonium-contaminated areas of the 903 Area at the RFP. Contaminated areas were represented by three area sources having a total area of 6000 m2. Ambient air samplers S-6, S-7, and S-8 operated routinely at this time.

Release Estimates and Discussion

Table IV-4 lists the measured TLLa (in counts per minute and femtocuries per cubic meter) and the corrected TLLa converted to activity associated with plutonium isotopes. Corrections for inlet collection efficiency, filter efficiency, sample air volume, and fraction of TLLa associated with plutonium isotopes are factored into the corrected concentration values reported in Table IV-4. Net filter count rates were converted to TLLa in air using the nominal value for the total efficiency factor (0.21) For the most part, corrected plutonium concentrations are greater than the uncorrected TLLa. These differences result from the corrections made for sampler inlet collection efficiency and sampled air volume. For some release days (January 30, February 24, and March 19, 1969), the corrected plutonium concentration is less than the uncorrected TLLa. Remember that about 40% of the TLLa is due to plutonium alone. A compensating factor is the sampler inlet collection efficiency, which is a function of the wind speed. The higher the wind speed, the greater the correction. Days with lower overall wind speeds had higher sampler inlet collection efficiencies; therefore, little correction was needed. Compare the March 19, 1969, and April 7, 1969, release days. The corrected plutonium concentration for April 7 was a factor of 14 greater than the uncorrected TLLa, while for March 19, the corrected plutonium concentration was a factor of 1.2 less than the uncorrected TLLa. This difference is attributed to the overall greater wind speeds for April 7. The median wind speed for April 7 was 11.6 m s^{-1} (mean of 11.5 m s^{-1}), while the median wind speed for March 19 was 1.8 m s^{-1} (mean of 5.3 m s^{-1}).

Calibrated release estimates (Table IV-4) indicate the highest releases occurred on January 7, 1969. The median release for this date was 0.89 Ci with a 5th and 95th percentile values of 0.18 and 4.3 Ci, respectively. A more detail accounting of the release quantity distributions can be found in Appendix K. The sum of the six highest events had a 50th percentile value of 2.8,

with 5th and 95th percentiles of 1.2 and 14.2 Ci, respectively. The sum of all 24 release events had a 50th percentile value of 3.1 Ci with 5th and 95th percentiles of 1.4 and 15 Ci, respectively. Clearly, total releases are dominated by the six highest measurement days at the S-8 sampler. Note that the sum of the releases is not the sum of the 5th, 50th, and 95th percentiles of individual days. Rather, it is calculated by randomly sampling from the distributions comprising each individual day and then summing. Percentiles were then extracted from the output distribution. Overall, release estimates varied by about a factor of 25 within the 5th and 95th percentile interval.

Correlation between median release estimates and the net count rates observed at the S-8 sampler was marginal ($r^2 = 0.57$). Correlation between the net count rate corrected for sampler deficiencies and median release estimates was higher relative to the uncorrected data ($r^2 = 0.70$). However, the regression coefficient indicates the corrected count rate was not directly proportional to the calibrated release estimates. This is because calibrated releases depend not only on the measured count rates but also on the transfer function, which is a function of wind speed, wind direction, and stability class.

Figure IV-10 shows the distribution of calibrated release estimates plotted along with release estimates made by the suspension equation, with and without wind gusts. Suspension equation estimates were made assuming a wind speed exponent of 4, a particle size mode of 0.5 mm, a bulk density of 1.5 g cm^{-3}, a 10-Ci inventory, a 3-cm contamination depth, and a calibration constant of 0.18 g m^{-2} h^{-1}. Suspension equation estimates with gusts were made only for five of the highest S-8 sampler days. In general, calibrated release estimates are higher than those predicted by the suspension equation alone. Inclusion of wind gusts tended to increase suspension release estimates, although the method still underestimated releases relative to the calibrated release estimates. It is evident from Figure IV-10 that calibrated release estimates do not correlate well with release estimates made with the suspension equation alone. This difference brings to light one of the fundamental problems associated with using a soil erosion equation alone to predict releases. That is, soil suspension is dependent on not only the wind speed but also the characteristics of the soil. Mechanical disturbance, soil moisture, and the presence of vegetation have a significant impact on soil erosion susceptibility. We can estimate soil conditions based on precipitation measurements and records of mechanical disturbance, but these estimates are fraught with uncertainty. With no definitive soil characterization data to use in a suspension model, we were left with making the best use of the limited data we did have, which included the air sampling data. Therefore, calibrating releases to measured concentrations, taking into consideration the deficiencies in air sampling, appeared to be the most reasonable approach to obtaining release estimates.

It is instructive to compare the release estimates to the estimated plutonium inventory on the 903 Area. Meyer et al. (1996) estimated the amount of plutonium contained in cutting oil that leaked from barrels on to soil to range from 2.8 to 57 Ci, with a best estimate around 6.1 Ci. Our release estimates are within this range and suggest that about 10–30% of the plutonium was suspended from the 903 Area and transported downwind on soil particles <30 μm. These release estimates do not include contaminated soil that was removed from the 903 Area from saltation effects or larger (>30 μm) suspended particles. We believe the high concentrations of plutonium in soil directly east of the 903 Area are primarily due to saltation and not deposition from airborne plumes. Past researchers (ChemRisk 1994) attributed most of the plutonium in soil 8- to 10-km east of the RFP to 903 Area releases. While we also believe a substantial amount of the

plutonium at that location is from 903 Area sources, a recent investigation into the fire that occurred in 1957 (Voillequé 1998) has indicated this event may have also been a significant contributor.

Table IV-4. Calibrated Release Estimates of Plutonium Attached to <30 μm Soil Particles and Measured and Corrected Concentration for the 24 Highest Release Days

Release date	Measured TLLa concentration (Net cpm)	Measured TLLa concentration (fCi m^{-3})[a]	Corrected plutonium concentration[b] (fCi m^{-3})	Release quantity (Ci) 50%	95%	5%
18-Mar-68	36	955	3,141	1.4×10^{-2}	7.4×10^{-2}	2.9×10^{-3}
11–14-Apr-68[c]	140	3,944	6,831	6.4×10^{-2}	3.4×10^{-1}	1.4×10^{-2}
13-May-68	86	2,319	3,341	3.0×10^{-2}	1.6×10^{-1}	6.6×10^{-3}
17-Sep-68	40	1,059	1,372	6.7×10^{-3}	3.4×10^{-2}	1.2×10^{-3}
22, 23, 24-Nov-68	45	1,178	1,258	6.9×10^{-2}	3.4×10^{-2}	1.3×10^{-3}
25-Nov-68	35	924	1,155	4.9×10^{-3}	2.4×10^{-2}	1.1×10^{-3}
5-Dec-68	262	8,210	12,782	5.1×10^{-2}	2.5×10^{-1}	9.7×10^{-3}
11-Dec-68	36	940	2,593	9.3×10^{-3}	4.6×10^{-2}	2.0×10^{-3}
2-Jan-69	37	966	1,312	2.9×10^{-3}	1.3×10^{-2}	5.7×10^{-4}
3–5-Jan-69[d]	116	3,201	4,146	2.2×10^{-2}	1.0×10^{-1}	4.4×10^{-3}
6 Jan-69	215	6,475	8,621	1.8×10^{-1}	8.6×10^{-1}	3.4×10^{-2}
7-Jan-69	422	15,244	287,988	8.9×10^{-1}	$4.3 \times 10^{+0}$	1.8×10^{-1}
29-Jan-69	56	1,467	1,993	1.3×10^{-2}	6.0×10^{-2}	2.7×10^{-3}
30-Jan-69	654	28,568	26,913	5.3×10^{-1}	$2.4 \times 10^{+0}$	9.6×10^{-2}
24-Feb-69	32	850	651	7.3×10^{-3}	3.7×10^{-2}	1.7×10^{-3}
19-Mar-69	155	4,412	2,949	5.5×10^{-2}	2.7×10^{-1}	1.2×10^{-2}
7-Apr-69	67	1,786	20,045	3.8×10^{-1}	$1.9 \times 10^{+0}$	7.7×10^{-2}
TOTAL[e]				$3.1 \times 10^{+0}$	$1.5 \times 10^{+1}$	$1.4 \times 10^{+0}$

[a] Computed using Equation (IV-13), a total efficiency factor of 0.21, filter efficiency of 0.95, and sample volume given by Equation (IV-21) with c_O = 40 cpm and HV-70 paper.

[b] Based on the fraction of TLLa attributed to plutonium isotopes of 0.4 and an inlet collection efficiency correction based on the particle size distribution reported in Langer (1986).

[c] Releases were based on the meteorological data for April 12, 1968. Count rate is the sum of the 4 days (see sampler data discussion in text).

[d] Releases were based on the meteorological data for January 3, 1969. Count rate is the sum of the 3 days (see sampler data discussion in text).

[e] The distribution of total releases was determined by sampling from the distribution of each individual release event and summing. It is not the sum of the given percentile value.
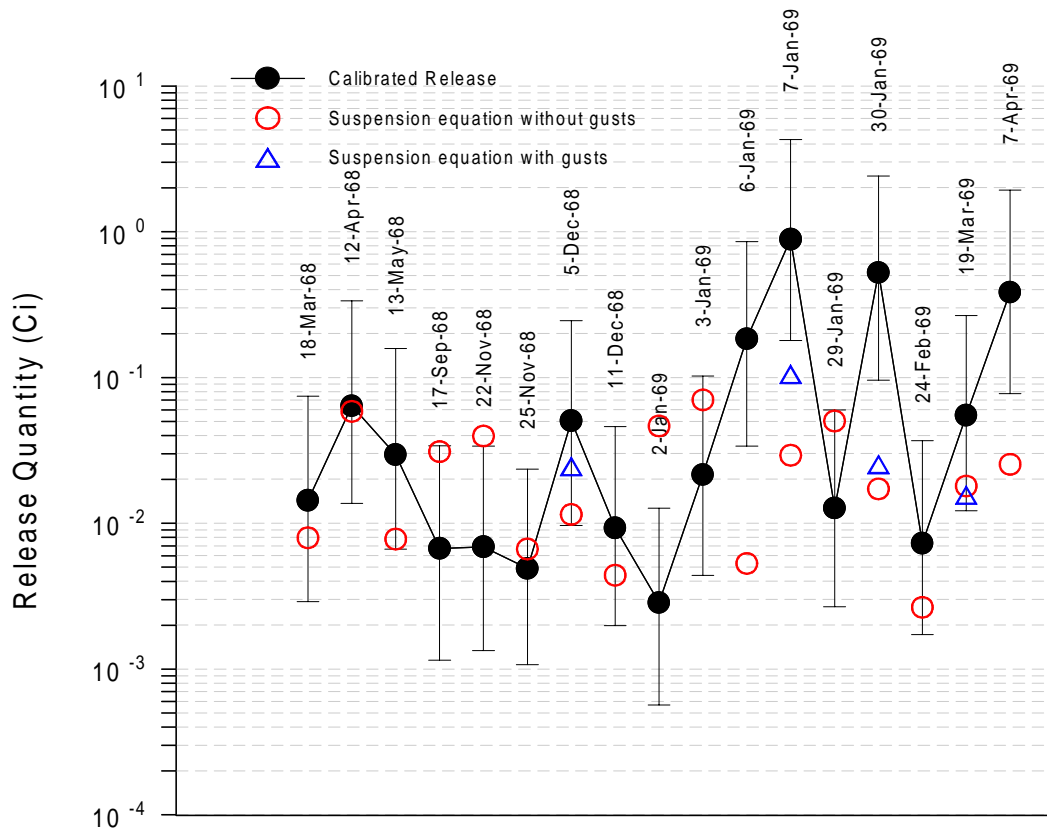
Figure IV-10. Calibrated release estimates for the 24 highest S-8 sampler days and release estimates calculated using the suspension equation alone, with and without gust data. The black dots represent the 50th percentile value with the vertical lines above and below representing the 95th and 5th percentile, respectively. Releases dated November 22, 1968, April 12, 1968, and January 3, 1969, are weekend events and represent 3–4 days of sampling (see discussion on sampler data in this chapter).

Sensitivity of Results to Input Distributions

A sensitivity analysis was performed on the distributions of calibrated release estimates. Parameter sensitivity was evaluated by computing the rank correlation coefficient between input parameter values and corresponding calibrated releases. Rank correlation coefficients range in value from –1 to +1. The higher the absolute value of the rank correlation coefficient, the higher the correlation between the input parameter and calibrated release estimates. A high correlation translates into high sensitivity. A positive correlation coefficient indicates that an increase in an input parameter value results in an *increase* in the calibrated release estimate. A negative correlation coefficient indicates an increase in an input parameter value results in a *decrease* in the calibrated release estimate.

Rank correlation coefficients for each of the stochastic inputs variables using the January 7, 1969, release day (Table IV-5) were greatest for the atmospheric transfer function correction factor, followed by the fraction of TLLa attributed to plutonium isotopes and volume of air sampled. The atmospheric transfer function correction factor, volume of air sampled, detector

counting efficiency, and particle size fraction for particles <10 µm all had negative correlation coefficients, indicating an inverse relationship between the calibrated release estimate and parameter value. Perhaps a simpler way to understand parameter sensitivity is to compute the percent contribution to the total variance. This was *approximated* by squaring the rank correlation coefficients and normalizing them to 100%. Most of the uncertainty in the calibrated release estimates was attributed to the atmospheric transfer function correction factor (86.9%), the fraction of TLLa attributed to plutonium isotopes (8.86%), and volume of air sampled (1.19%).

By contrast, a sensitivity analysis on the suspension equation for the January 7, 1969, release day indicated the particle size mode accounted for 97.7% of the variability in the release estimate, with 1.23% attributed to the wind speed exponent. Release estimates using the suspension equation alone for January 7, 1969, varied by more than 2 orders of magnitude between the 5th and 95th percentile values. This uncertainty analysis did not include uncertainty in the plutonium inventory, depth of contamination, and area of contamination; therefore, the actual distribution is expected to be larger. The calibrated releases for the same day varied by a factor of 35 between the same percentile values. The spread between the 5th and 95th percentiles for the two methods illustrates the precision in the two methods. Greater precision is gained by calibrating release estimates to the S-8 sampler compared to using the suspension equation alone.

Table IV-5. Rank Correlation Coefficient and Percent Contribution to Variance for the Calibrated Release Estimates for 24 Highest S-8 Sampler Days

Parameter	Rank correlation coefficient	% Contribution to total variance
Particle size mode	0.039	0.147
Fraction of TLLa attributed to plutonium isotopes	0.299	8.860
Wind speed exponent	0.054	0.284
Atmospheric transfer correction factor	–0.939	86.946
Volume of air sampled	–0.110	1.186
Total efficiency factor	–0.083	0.673
Fraction of particles <3 um	–0.069	0.476
Fraction of particles 3–10	–0.069	0.476
Fraction of particles 10–15	0.069	0.476
Fraction of particles >15	0.069	0.476

ESTIMATION OF BASELINE RELEASES, 1964–1969

In this section, we consider releases from the 903 Area that occurred on a more-or-less continuous basis during drum storage and removal operations from 1964 to 1969. Evidence of this so-called baseline release is from the generally higher airborne concentrations recorded at samplers near the 903 Area during that period. We approached this problem in a similar fashion to the estimation of the 24 highest S-8 sampler days. That is, we used corrected sampler data and an atmospheric dispersion model to back-calculate release rates from the 903 Area. The approach taken here is somewhat less refined than the approach taken for the 24 highest S-8 sampler days

because releases are estimated annually instead of daily. We justify this approach because episodic releases were already accounted for in the previous analysis.

Conceptual Model and Mathematical Formulation

The objective of these calculations is to provide annual average release estimates of plutonium-contaminated soil from the 903 Area, excluding those days that correspond to the 24 highest measurement days at the S-8 sampler. We assume the release is continuous over the measurement period (1 year) and the annual average concentration at the sampler is proportional to the release rate. With these assumptions and an atmospheric transport model capable of calculating transport from the 903 Area to the samplers, the annual average release rate is given by

$$Q = \frac{C}{T_f} \qquad \text{(IV-23)}$$

where
T_f = transfer function (s m^{-3})
C = concentration at the sampler (Ci m^{-3})
Q = steady state release rate (Ci s^{-1}).

The transfer function in this case is the well known X/Q value, which represents the average concentration at a receptor point divided by the steady-state release rate. The transfer function was calculated using the FDM computer code (Winges 1990), a 5-year meteorological data set, and an activity particle size distribution derived from measured data (Langer 1986 [see Table IV-2]). Dry deposition and gravitational settling are considered in the model but wet deposition is not. To some extent, wet deposition is accounted for in the sampler data because days of precipitation would likely result in little suspension from the 903 Area because moist soil is less susceptible to wind erosion. Evidence of this is shown in Figure IV-11 where the net count rate measured at the S-8 sampler for the first half of 1969 is plotted along with days where measurable precipitation was recorded at the Jefferson County Airport. Note that the low count days are associated with precipitation events. For this reason, we have not considered wet deposition in the calibrated release estimate.
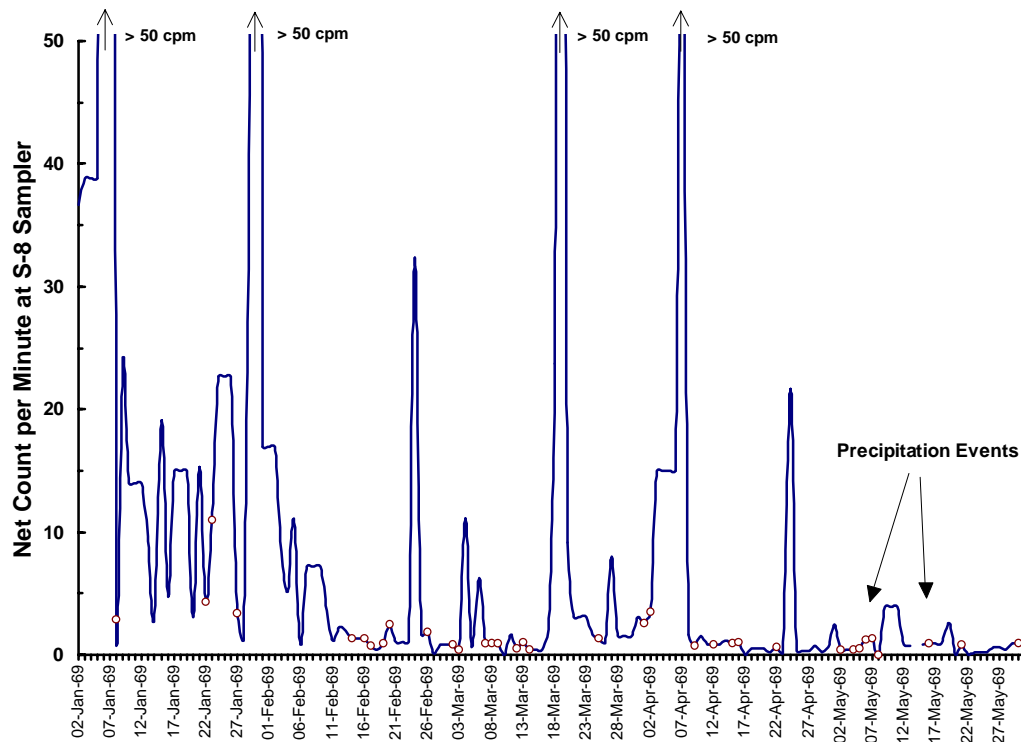
Figure IV-11. Net counts per minute at the S-8 sampler for the first half of 1969. Note that many of the days showing lower count rates are associated with precipitation events (shown as open circles) as recorded at the Jefferson County Airport.

Sampler Data

Concentrations at each of the onsite samplers for the period 1964–1969 were compiled on a monthly average and reported in the Task 4 report (Rope et al. 1997). Monthly average airborne TLLa at the samplers surrounding the 903 Area (samplers S-6, S-7, and S-8) are plotted with the monthly average airborne TLLa of the other onsite samplers in Figure IV-12. Note that the S-6, S-7, and S-8 sampler averages are generally higher than those of other samplers. High readings are dominated by the S-8 sampler because of its location east of the 903 Area and the predominance of westerly winds. The S-6, S-7, and S-8 sampler TLLa concentrations show poor correlation ($r^2 = 0.028$) to concentrations measured at the other onsite samplers. Concentrations of TLLa at all samplers are above the background concentration of TLLa in air, which was estimated to be about 1 to 5 fCi m^{-3} (Rope et al. 1997, page III-51). Ideally, we would like to have had a network of samplers encircling the 903 Area that were free from obstructions and other alpha activity sources that may have influenced the measurements. The locations of onsite samplers (see Figure IV-3) show this is not the case, and only the S-6, S-7, and S-8 samplers were relatively free of obstructions. The S-50 and S-51 samplers did not begin operation until October 1968 and, therefore, were not used in the calibration. It is not clear how much influence routine releases from other RFP facilities had on the samplers. Releases from the 44-m Building 771 stack under most circumstances were not expected to greatly influence the onsite samplers because the effluent plume would tend to pass over the onsite samplers. However, activity

released from building exhaust ducts has the potential to influence samplers in the vicinity of the building. The sampling efficiency for high-efficiency particulate air-filtered effluent will also tend to be greater than for suspended plutonium-contaminated soil because the particle sizes are considerably smaller.
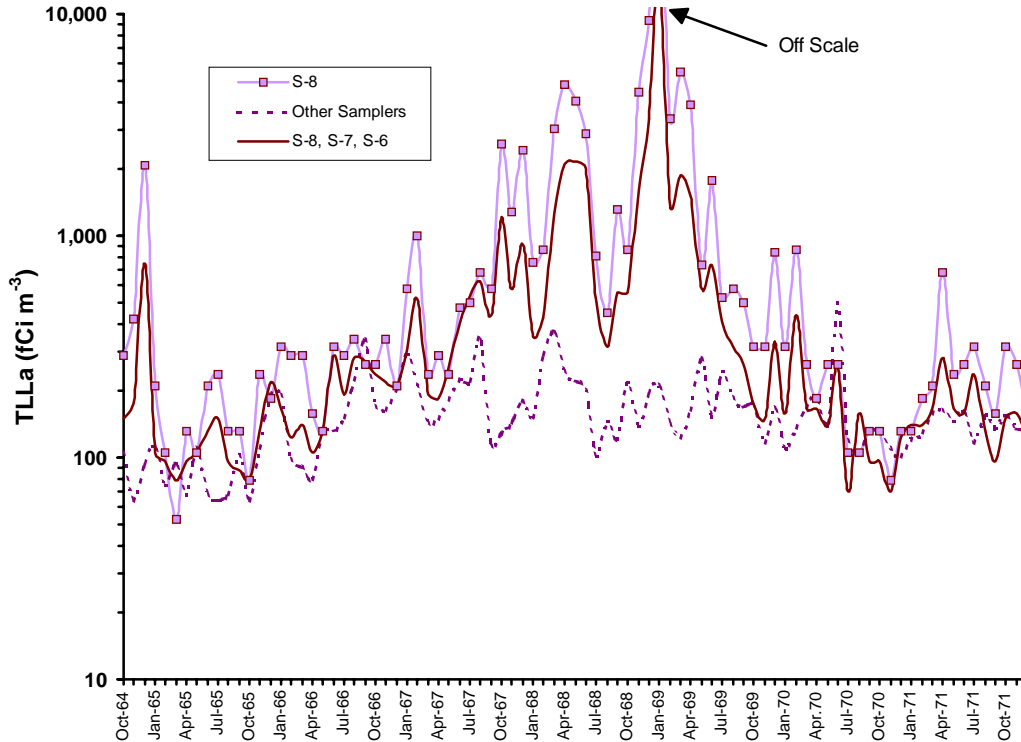
Figure IV-12. Monthly average TLLa in air measured at the onsite samplers from October 1964 to December 1971. The average of the readings at the S-6, S-7, and S-8 samplers was typically higher than the average of the reading at the other samplers.

Modeling air concentrations in an industrial complex such as RFP is feasible, but it would take considerable effort. Such an exercise would have to consider other sources of activity in close proximity to the samplers that may impact measurements. We believe the simplest, most accurate way to obtain calibrated releases is to restrict calibration to only those samplers that are most likely to reflect 903 Area releases. Therefore, calibration was performed using primarily the S-8 sampler, and calibrated release estimates using the S-6 and S-7 samplers were made for comparison.

Measurements of TLLa at the S-8 sampler were corrected for inlet collection efficiency and fraction of TLLa attributed to plutonium isotopes using the procedure outlined earlier. Rope et al. (1997) recommend that count rates less than 0.4 cpm not be used quantitatively because this value is less than the minimum detectable concentration for the alpha detector instrumentation. Count rates <0.4 cpm were, therefore, assumed to be zero. Inlet collection efficiency calculations used hourly wind speeds that were derived from a surrogate meteorological data set (see discussion on meteorological data) because meteorological data for the entire assessment period were lacking. A particle size distribution equivalent to that measured by Langer in 1986 (see

Table IV-2) was used in the calculation. Sensitivity analysis showed the calibrated release estimates were not particularly sensitive to the particle size distribution and, therefore, it was fixed and not considered stochastically. Based on the particle size distribution and meteorological data previously mentioned, an inlet collection efficiency factor (the term $\sum f_i/ICE_i$ in Equation [IV-17]) was calculated to be 2.12.

Measurements that appeared in the 24 highest release days were subtracted from the annual average along with alpha background from naturally occurring sources and plutonium weapons fallout. Natural long-lived alpha background was estimated to be 1.4 fCi m^{-3} (long-term average) based on the evaluation in Rope et al. (1997). Concentrations of ^{239}Pu in air in the Denver area from weapons testing fallout were obtained year-by-year from Rope et al. (1997). These concentrations were inferred from ^{90}Sr deposition before 1964 and measured by the Public Health Service and EPA after 1964.

Meteorological Data

A complete meteorological record of this period (1964–1969) is lacking from any of the nearby recording stations including Jefferson County Airport, NCAR portable stations, and RFP. For this reason, we employed a technique often used in prospective analyses and in retrospective analyses when historical meteorological records are lacking. This technique uses compilations of recently acquired meteorological data as a surrogate for past or future conditions and typically only applies to assessments of long-term (>1 year) dispersion conditions. This technique was previously used to estimate annual average concentrations in the model domain from routine releases (Rood 1997b).

For annual average 903 Area release estimates, we used meteorological data spanning a 5-year period (1989–1993) taken at the RFP. The EPA considers a 5-year database adequate for predicting annual average air quality impacts at a site. Meteorological data were taken at the 10-m level from the 61-m tower located on the south side of the plant complex at universal transverse mercator (UTM) coordinates 482064 E 4414963 N. Data recorded at this station included wind speed, wind direction, temperature, and other parameters (such as heat flux and standard deviation of wind direction) that were not used in these simulations. Stability classes were calculated using the general classification scheme discussed in Pasquill (1961), Gifford (1961), and Turner (1964), which use wind speed, cloud cover and ceiling height to estimate seven stability classes (A–G). Stability class F and G were combined because FDM allows only six stability classes. Cloud cover and ceiling height data were obtained from Denver Stapleton International Airport meteorological station located 24 km east and 14 km south of the RFP. Data of this type were not recorded at the RFP.

RFP data were obtained in electronic format from the Rocky Flats meteorologist. Each record represented the average over a 15-minute recording period. Hourly average wind speed and direction also were calculated from the raw RFP meteorological data using the protocol described in EPA (1987). An arithmetic average of the wind direction was computed first, and then it was segregated into 1 of 16, 22.5-degree sectors as required by FDM. The average wind speed for the hour was computed by taking the average of the four, 15-minute data segments. The wind speed was then segregated into six classes: 1.5, 2.5, 4.3, 6.8, 9.5, and 12.5 m s^{-1}. Meteorological data were then tabulated into a joint frequency distribution represented by a STAbility ARray (STAR) table. A STAR table lists the fraction of time the wind blew in a given

wind speed class, sector, and stability class. Appendix L contains the STAR table used in the
FDM calculations.

Treatment of Uncertainty

The sensitivity analysis conducted for the 24 highest S-8 sampler days indicated that the
atmospheric transfer correction factor, the fraction of TLLa attributed to plutonium isotopes, and
the total efficiency factor accounted for about 97% of the observed variability in the release
estimates. Based on this information, we did not consider uncertainty in the other parameters for
the baseline release estimates. However, because the sample count rates for baseline releases are
lower and near background compared to those for the 24 highest S-8 sampler days, the
uncertainty from counting error was included in the uncertainty analysis for baseline release
estimates. In addition, we used a surrogate meteorological data set in our calculations, which
introduces additional uncertainty into the calculations. Therefore, five parameters were included
in the uncertainty analysis: atmospheric transfer correction factor, fraction of TLLa attributed to
plutonium isotopes, counting error, total efficiency factor, and meteorological uncertainty. Each
parameter is discussed separately below.

Atmospheric Transfer Correction Factor

Uncertainty in the atmospheric transfer function was treated in a manner similar to the 24
highest S-8 sampler days. We used a multiplicative correction factor representing the inverse of
P/O ratios reported in the literature. Models tend to perform better under longer averaging times
thus, we would expect the correction factor for baseline releases to exhibit lower variability than
that for the 24 highest S-8 sampler days.

In the uncertainty analysis for routine releases from Rocky Flats, Rood (1997b) uses a
lognormally distributed correction factor having a GSD of 2.2 for receptors <8 km from the
source. This correction factor was applied to predictions made by the RATCHET model
(Ramsdell et al. 1994) over distances out to about 30 km.

Miller and Hively (1987) reported the GSD of the distribution of P/O ratios for annual-
average Gaussian Plume model predictions to be 1.5. These ratios were reported for a flat terrain
environment, a ground-level release, and receptors out to 10 km. Another study (Simpson et al.
1990) reported the GSD of the distribution of annual average P/O ratios to be 1.5. Conditions for
the Simpson et al. study included a flat terrain environment, elevated release, and receptors out to
50 km.

No one study is entirely relevant to the situation we are researching. However, we would
expect the distribution of P/O ratios to be somewhere in the range reported in the various studies
cited. Lacking a completely relevant study from which to base the distribution of P/O ratios, we
have chosen to error on the side of conservatism and select a larger GSD value for our
distribution. Therefore, the multiplicative correction factor applied to the atmospheric transfer
function was lognormally distributed having a GM of 1.0 and a GSD of 2.2. As was done for the
24 highest S-8 sampler days, we have assumed no bias in the model.

Fraction of TLLa Attributed to Plutonium

The distribution of the fraction of TLLa attributed to plutonium isotopes was identical to that described for the 24 highest S-8 sampler days.

Counting Error

Sampler data were converted from counts per minute to TLLa concentrations by first computing the annual average count rate (subtracting out the 24 highest S-8 sampler days) and applying the total efficiency factor (0.21) and volume of air sampled (81.5 m^3). Counting uncertainty was calculated by assuming each count day represents an independent measurement. Therefore, the variance in the annual average count rate can be computed using

$$\sigma_T^2 = \frac{1}{n}\sum_{i=1}^{n}\sigma_i^2 \qquad \text{(IV-24)}$$

where
σ^2_T = the variance of the mean count rate
σ^2_i = the variance of the ith sampling day
n = the number of sampling days in a year.

The variance in the count rate for each sampling day is dependent on the background counting rate. Background count rates were reported to vary from 0 to 0.9 cpm. Ninety-five percent of the filters analyzed had filter background count rates ≤0.6 cpm. Rope et al. (1997) calculated the coefficient of variation (*CV*) for different net and background count rates. For example, the *CV* for a net sample count rate of 1.0 cpm was 0.33 for a 0.1 cpm background count rate and 0.41 for a background count rate of 0.6 cpm. Note that as the background count rate increases, so does the *CV* value for a given net sample count rate. For count rates exceeding 100 cpm, the *CV* is relatively small (3%) for all background count rates; for that reason, counting uncertainty was not considered for the 24 highest S-8 sampler days. Rope et al. (1997) suggests treating the background count rate stochastically for evaluating uncertainty in the net count rates. To do so would require sampling from a distribution that is itself an uncertain parameter. We have chosen instead to simplify the calculation and assume a constant background count rate. The background count rate chosen was 0.6 cpm because 95% of the background count rates were less than or equal to this value. By choosing this value, uncertainty in the net sample count rates may tend to be overestimated. However, the net count rate itself is not biased (positive or negative) in any way.

The *CV* is then a function of the net sample count rate (Table IV-6). The *CV* is the standard deviation of the count rate divided by the mean count rate and represents the relative error in the measurement. By assuming a mean of 1.0, the *CV* can be converted to variance by squaring the *CV* value. We represented counting uncertainty year-by-year using a normally distributed

multiplicative correction factor having a mean of 1.0 and a variance given by Equation IV-24. To avoid negative values, the distribution was truncated at a value of 0.01.

Table IV-6. Coefficient of Variation in the Net Count Rate
Used to Evaluate Uncertainty in Annual Average
Counts of TLLa Activity[a]

Net count rate (cpm)	Coefficient of variation (%)[b]
100	3
10	10
5	15
4	17
3	20
2	26
1	41
0.9	44
0.8	48
0.7	53
0.6	60
0.5	69
0.4	83
0.3	105
0.2	150
0.1	283

[a] From Rope et al. 1997

[b] Based on a background count rate of 0.6 cpm

Meteorology Uncertainty

Meteorology uncertainty arises because we are using 5 years of meteorological data spanning a recent time period (1989–1993) to define an annual average dispersion that will be applied to the assessment years (1964–1969). We must determine how well this 5-year period represents the past. Comparisons of annual average *X/Q* (air concentration divided by source term) values computed with a 5-year data set to the annual average *X/Q* values computed using the meteorological data for each specific year were recently performed for the Fernald Dosimetry Reconstruction Project (Killough et al. 1996). Meteorological data from the Cincinnati Airport from 1987 to 1991 composed the 5-year composite meteorological data set. Annual average *X/Q* values computed with these data were then compared with the annual average *X/Q* values computed for each specific year using the meteorological data for that specific year. The years spanned from 1951 to 1991. Concentrations were calculated at 160 receptors ranging in distance from 1000 to 10,000 m from the release point. A straight line Gaussian plume model for a 10-m release height was used to generate the *X/Q* values. The 5-year composite *X/Q* divided by the *X/Q* for the specific year (P/O ratio) forms the basis of the upper graph in Figure IV-13 (note the logarithmic scale). A similar procedure was applied to the *X/Q* values generated for routine release evaluations in this study (Rood 1997b) and is depicted in the lower graph in Figure IV-13. However, only the composite period is shown because meteorological data from previous years were not obtained. The lower graph in Figure IV-13 was generated using the RATCHET

model ([Ramsdell et al.](#) 1994) and *X/Q* values from a release point at Building 776 for 2300 receptors in the model domain. [Figure IV-13](#) depicts the 5th, 50th and 95th percentile of the cumulative frequency distribution for all points in the model domain. Note that for the composite period, the spread of the data is similar for both data sets.

As expected, the spread is much larger for those years that do not include the 5-year composite data. The long-term trend of these data may not depend strongly on location. If this procedure is applied to the RFP environs using Denver Stapleton International Airport data, for instance, the locus of the 50th percentiles is likely to look somewhat different, although the amplitudes may be similar. Obtaining meteorological data from past years (1953–1989) for Denver Stapleton International Airport (Rocky Flats data are nonexistent) and performing the uncertainty calculations is not a trivial task, and the overall impact on the results may be similar to what is observed at Cincinnati based on a similar spread of these data for the composite period at both locations. For this reason, we have chosen instead to adapt the Fernald data to our analysis.

The Fernald data were represented by a multiplicative correction factor having a GM of 1.0 and GSD of 1.7. This distribution was developed using the following sampling scheme:

1. Noting from [Figure IV-13](#) that the maximum range in the GMs is a factor of 2, a GM was randomly selected from a log-uniform distribution with a minimum $2^{-1/2}$ and maximum $2^{1/2}$.
2. Using the GM from step (1) and GSD = 1.61 (the maximum GSD calculated from the ratio of the 5-year composite *X/Q* to specific year *X/Q* for the 40 years of data), a sample is drawn from a lognormal distribution with these parameters.
3. Values are stored from step (2) and the process is repeated.

This somewhat conservative procedure accounts for the year-to-year variability in the GM of the 5-year composite *X/Q* to specific year *X/Q* ratio, as well as the uncertainty associated with distance and direction from the source. For a sample size of 1000, a lognormal distribution was fitted with a GM = 1.0 and GSD = 1.7.
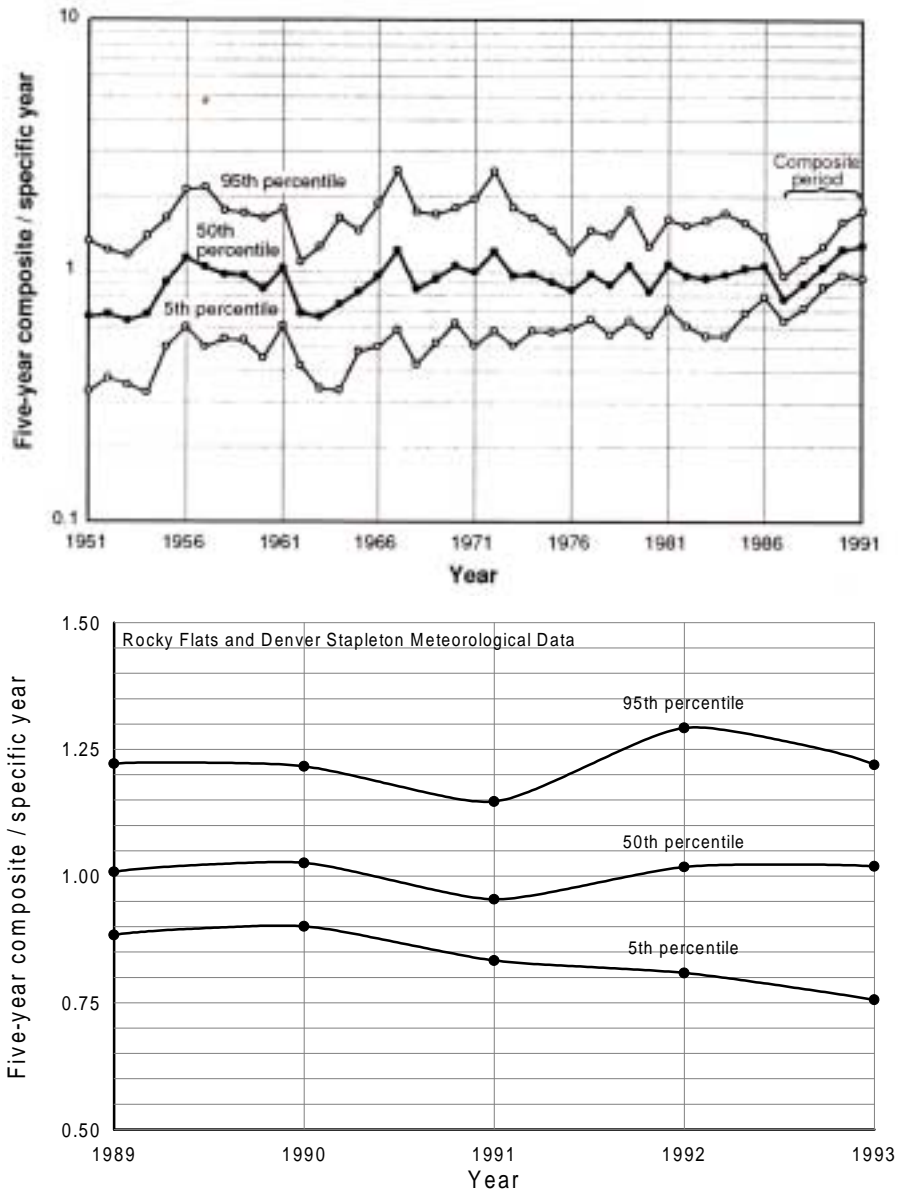
Figure IV-13 Distributions of P/O ratios for *X/Q* calculated with the Cincinnati Airport meteorological data (upper graph) and RFP–Denver Stapleton International Airport meteorological data (lower graph). Predicted (P) corresponds to *X/Q* values for a 5-year composite; observed (O) corresponds to the *X/Q* values for a specific year (from Killough et al. 1996).

Summary

Five sources of uncertainty were accounted for in the baseline release estimates: (1) atmospheric transfer uncertainty, (2) counting uncertainty, (3) total efficiency factor uncertainty, (4) fraction of TLLa associated with plutonium isotopes, and (5) meteorological uncertainty. Except for the fraction of TLLa attributed to plutonium isotopes and the total efficiency factor, uncertainty was represented by multiplicative correction factors. Combining all uncertainty and conversion factors, Equation IV-25 describes the baseline release rate estimate.

$$Q = \frac{\dfrac{CF_1\,ICEF}{(TEF)(V)(FE)}}{DF_2\,F_3} \qquad \text{(IV-25)}$$

where
C	=	net annual average count rate (counts per minute)
F_1	=	counting error uncertainty factor (stochastic)
$ICEF$	=	inlet collection efficiency factor (unitless)
TEF	=	total efficiency factor (stochastic)
FE	=	filter collection efficiency (0.95)
V	=	volume of air sampled (81.5 m³)
D	=	atmospheric transfer function or X/Q (3.85×10^{-5} s m^{-3} for S-8 sampler)
F_2	=	atmospheric transfer function uncertainty correction factor (stochastic)
F_3	=	meteorological uncertainty correction factor (stochastic).

Monte Carlo simulations were performed using the Crystal Ball© software. Release estimate distributions were developed from 1000 individual trials. For each trial, parameter values were sampled once from the input distributions, and a single release estimate was calculated. The process was then repeated for all 1000 trials. Correlation among the parameters was not considered.

Results

Median value baseline release estimates for 1964–1969 (Table IV-7) ranged from 0.0083 Ci in 1966 to 0.052 Ci in 1969. A more detailed accounting of the release quantity distributions can be found in Appendix K. The totals for all 6 years had a 50th percentile value of 0.142 Ci, with 5th and 95th percentile values of 0.025 Ci and 0.79 Ci, respectively. Compared to estimates for the 24 highest S-8 sampler days, the baseline releases represent about 5% of the total estimated releases from the 903 Area. Distributions of releases for each year are illustrated in Figure IV-14. Also shown in Figure IV-14 are release estimates made using the S-6 and S-7 sampler data. For these deterministic calculations, a detector efficiency of 0.21 was used, and the fraction of TLLa attributed to plutonium isotopes was set at 0.5. Except for 1969, estimates made with the S-6 sampler were generally higher than estimates made with the S-7 sampler. The S-7 sampler release estimate for 1964 was outside the 5th and 95th percentiles of the distribution of release

estimates made with the S-8 sampler. Total releases appear to be dominated by releases during the years 1967–1969.

Overall, the release estimates vary by about a factor of 30 within the 5th and 95th percentile interval. This range is similar to the range for the 24 highest release days identified by the S-8 sampler.

Table IV-7 Results of Baseline Release Estimates of Plutonium Contaminated Soil
<30 μm using the S-8 Sampler Data

Year	Average count rate (cpm)	Uncorrected TLLa[a] (fCi m^{-3})	^{239,240}Pu fallout (fCi m^{-3})	Corrected ^{239,240}Pu[b] (fCi m^{-3})	Release quantities (Ci) 50th	95th	5th
1964	1.49	39.3	0.58	33	0.007	0.038	0.0008
1965	0.21	5.65	0.25	5.7	0.013	0.10	0.0009
1966	0.39	10.2	0.11	10	0.014	0.11	0.0013
1967	1.32	34.7	0.054	35	0.026	0.16	0.0030
1968	2.65	69.8	0.042	70	0.055	0.30	0.011
1969	3.60	79.1	0.056	79	0.064	0.34	0.013
TOTAL[c]					0.19	0.98	0.039

a. Based on the nominal values for total efficiency factor (0.21) and volume of air sampled (81.5 m^3)

b. Based on an inlet collection efficiency factor of 2.12, and the fraction of TLLa attributed to plutonium of 0.4

c. The distribution of total releases was determined by sampling from the distribution of each individual year and summing. It is not the sum of the given percentile value.
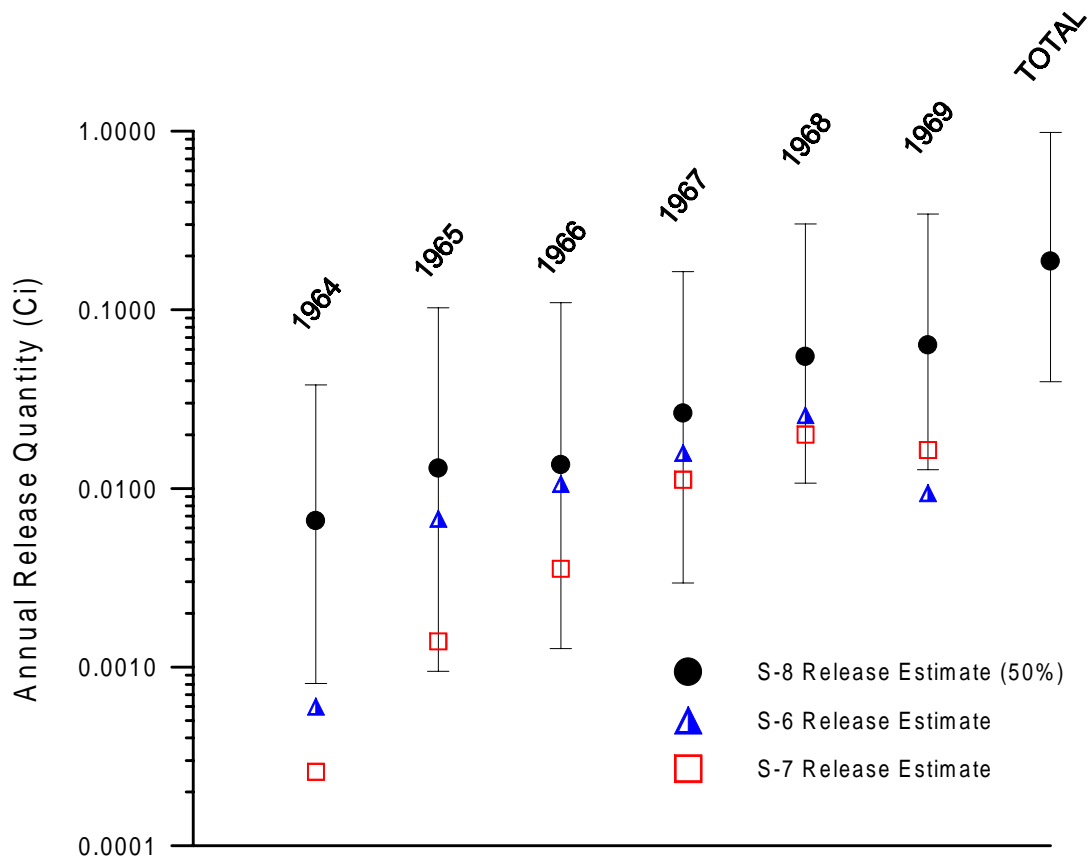
Figure IV-14. Distribution of baseline release estimate quantities by year. The black dots represent the 50th percentile value with the vertical lines above and below representing the 95th and 5th percentile respectively. Release estimates determined with the S-6 and S-7 samplers are deterministic estimates.

Sensitivity of Results to Input Distributions

Using the same procedure previously outlined for the 24 highest S-8 sampler days, we performed a sensitivity analysis for two of the years for which baseline release estimates were made (Table IV-8). The results indicate that the atmospheric transfer correction factor was again the most sensitive parameter. Counting uncertainty was identified as the second most sensitive parameter for the 1964 results, but it was fourth for the 1969 results. This difference highlights the importance of counting error when the count rates are low. The annual average count rate was 1.49 cpm for 1964 and 3.6 cpm for 1969.

**Table IV-8. Rank Correlation Coefficient and Percent Contribution to
Variance for Calibrated Baseline Release Estimates**

Parameter	1964		1969	
	RCC[a]	% Variance[b]	RCC[a]	% Variance[b]
Fraction of TLLa attributed to plutonium isotopes	0.21	4.48	0.26	7.69
Atmospheric transfer correction factor	–0.66	44.24	–0.74	62.3
Meteorology correction factor	–0.48	23.4	–0.48	26.2
Total efficiency factor	–0.12	1.46	–0.09	0.92
Count rate	0.51	26.0	0.16	2.9

[a.] Rank correlation coefficient
[b.] Percent contribution to total variance.

CHAPTER V

APPROACH TO ESTIMATING RELEASES AFTER 1970

Because of the unique events that took place at the 903 Area, a different technique is required to estimate releases after 1970. The initial suspension events described in this report took place because plutonium-contaminated oil was deposited on the soil. The larger soil particles with greater settling velocities were suspended during these events, but then they settled out rapidly on a grassy field east of the 903 Area. When the original contaminated area was covered with asphalt in late 1969, the field east of the 903 Area (referred to as the east field) became the primary source of airborne contamination.

It is far more difficult to model resuspension[1] than suspension. The variables that affect resuspension are numerous and difficult to quantify. Research has been done in this area for a number of years, but little has taken place since the 1980s. Recent literature on resuspension is very difficult to locate because researchers, it appears, recognized the variable and site-specific nature of resuspension and were hesitant to generalize the parameters that predict it. Resuspension is far better measured in situ than predicted based on other studies.

This fact makes the job of predicting resuspension activity and airborne contamination from the 903 Area at the RFP for periods after 1970 very difficult. Instead of predicting resuspension based on studies done at other areas, we base our approach for estimating releases on research conducted and airborne contamination data collected at the RFP in the 1970s and 1980s. The resuspension studies available are difficult to compare because the studies were conducted by different researchers using different experimental techniques.

Instead of establishing a source term for the resuspension from the east field, we present the available information here for completeness and use the incomparability of data to support the spatial nature of releases from this source.

It is important to remember that suspension and resuspension are different processes and are dominated by different effects. This distinction is important because releases after 1970 were dominated by resuspension from the east field contaminated with previously suspended material.

RESUSPENSION EXPERIMENTS

Gerhard Langer produced a report in 1991 that summarized years of research on resuspension from the east field (Langer 1991). He noted that plutonium from the RFP located in the east field is difficult to mobilize and that the field was stabilized by adding topsoil and by introducing vegetation.

Initial resuspension studies at the east field were directed at saltation processes (Langer 1991). Most of the experimental evidence supported a threshold for resuspension without the influence of saltation, which immediately contrasts this process with suspension from the 903 Area soils. Resuspension was hypothesized to result from the mobilization of plutonium from grass blades and grass litter.

The grass resuspension pathway was then studied further to determine the mechanism for passing the plutonium attached to soil to the grass blade. Both rain splash and soil suspension of

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[1] See Chapter II and Figure II-1 of this report for a definition of resuspension.

material subsequently captured by the fibers on the grass blades were suspected to act as mechanisms for contaminating the grass with resuspendible quantities of plutonium.

Mechanical disturbances were also found to have a rather dramatic impact on releases of plutonium. George Sehmel studied this mechanism more thoroughly than others (Sehmel 1980). These short-term events are difficult to quantify in terms of release rate of plutonium, but they have been shown to influence downwind plutonium concentrations.

The numerous processes suspected to be responsible for resuspension of plutonium from the 903 Area east field are difficult, if not impossible, to combine into a quantitative and predictive model. Because modeling was not feasible, different researchers set up sampling towers in the vicinity of the east field. These towers held impactor particle collectors that preferentially collected particles of a certain size. Each experiment differed in sampling distance, sampling heights, and particle size fractions collected.

One of Langer's experiments showed that most of the activity from the area was associated with nonrespirable particles (Langer 1991). Samplers located on the perimeter of the field showed that the concentration of respirable particles at an undisclosed height was approximately 0.02 fCi m^{-3} ^{239}Pu. The average nonrespirable particle concentration was approximately 0.71 fCi m^{-3} ^{239}Pu. This fraction carried 97% of the total airborne radioactivity.

Another Langer experiment placed samplers at a 100-m distance from the eastern edge of the east field at heights of 1, 3, and 10 m for particle sizes of <3, 3–15, and >15 µm. This research program lasted from November 1982 through August 1985, collecting samples bimonthly. Respirable (<3 µm) and inhalable (3–15 µm) plutonium concentrations did not change with height, with average results of 0.0088 and 0.025 fCi m^{-3}, respectively. Nonrespirable concentrations decreased with increases in height above ground, but they averaged 0.067 fCi m^{-3} ^{239}Pu. Even though there were enough data points to produce significant plutonium collection, the concentrations of plutonium were so low as to be statistically erratic (Langer 1991).

Because Langer's studies were limited to the area near the east field, he indicated that his estimates of total plutonium emissions were also limited to this area. Langer provided an estimate of source release from the east field of 200 µCi y^{-1}. For this prediction, Langer had to estimate average airflow over the field and the resulting fetch for resuspended particles. He did not present either of these parameter estimates in his paper (Langer 1991).

Sehmel also conducted studies on resuspension at the 903 Area east field (Sehmel and Lloyd 1976). Sehmel's studies lasted for the month of July 1973; looked at three different locations east of the east field; heights of 0.3, 1, 2, 3, 4, 10, 11, and 30 m; and particle sizes of 1.1, 2.0, 3.3, and >7 µm. These samplers also operated for different wind speed increments. The results of this experiment are difficult to compare to Langer's results because of the differing parameters.

From comparisons of the maps provided in each study, it appears that Sehmel's site A is the most readily comparable to Langer's perimeter samplers, and Sehmel's site AB might be the closest to Langer's sampler located 100 m from the eastern edge of the field.

At a height of 1 m, site A concentrations for total particle collection (respirable + nonrespirable) range from 0.4 fCi m^{-3} to 1.8 fCi m^{-3} ^{239}Pu and site AB concentrations for total particle collection range from 0.025 fCi m^{-3} to 0.18 fCi m^{-3} ^{239}Pu. This 1-month sampling experiment is even less statistically meaningful than Langer's study, and the results must be viewed in that light.

To facilitate comparison, the values cited thus far are presented in Table V-1. The comparable sites between Langer and Sehmel are listed together.

Table V-1. Comparisons of ^{239}Pu Concentrations at Locations East of the 903 Area

Study and location	Particle sizes measured	^{239}Pu concentration (fCi m^{-3})
Langer, perimeter sampler	Respirable	0.02
	Nonrespirable	0.71
Sehmel, site A	All particle sizes	0.4–1.8
Langer, 100 m east sampler	Respirable	0.0088–0.025
	Nonrespirable	0.067
Sehmel, site AB	All particle sizes	0.025–0.18

When the nonrespirable and respirable fractions are summed for a Langer site, they match reasonably well with the total particle collection for the similar Sehmel site. These comparisons are very tenuous because different sampling and analysis techniques were used and particle sizes collected with each sampler varied. This illustrative comparison tends to support somewhat similar resuspension results from the two experiments.

Sehmel did not estimate total annual plutonium release from the field as Langer did. In a later paper discussing the same experiment, Sehmel did estimate total nonrespirable horizontal airborne plutonium flux. This estimate was made using the data from Sehmel's study, with maximum flux at site A of 6 x 10^{-4} μCi m^{-2} d^{-1}. The flux decreased with distance, with a maximum of 2 x 10^{-6} μCi m^{-2} d^{-1} at site AB (Sehmel 1976).

To translate this horizontal flux to total annual release, we need to know the cross sectional contaminated area that is represented by the sampler. Langer presented the results of the soil sampling study done by the Atomic Energy Commission's Health and Safety Laboratory (HASL) (Langer 1991). The contours in this soil contamination plot represented areas of equal concentration of plutonium in soil. We used these contours to help us estimate cross sectional contaminated area.

At the location of Sehmel's A and AB samplers, we drew a north-south line that spanned the distance across one of the contour areas on the HASL plot. We selected the contour that represents contamination levels of 75 mCi km^{-2} because this contour spanned a comprehensive north-south distance to the east of the east field, and because it is the smallest contamination contour that still appears to represent plutonium contamination from the 903 Area and not other sources.

The north-south distance at the location of sampler A is 920 m and at sampler AB is 1200 m. We assume that the height of the plume at location A (1 m) is lower than at the more distant location AB (2 m) and use this height to calculate cross sectional area as a rectangle. The cross sectional area represented by sampler A is 920 m^2 and by sampler AB is 2400 m^2.

To calculate annual plutonium released, we multiply Sehmel's flux at each location by the appropriate cross sectional area and by the number of days in a year. At location A, we calculated the plutonium released to be 201 μCi y^{-1}. At location AB, the release rate was 1.8 μCi y^{-1}.

Although the release rate at location A matches Langer's value quite well, we must emphasize that this comparison is somewhat coincidental. The contour selected by the authors,

while chosen with care, was still somewhat random and may or may not represent well the area seen by the sampler. It must also be stressed that the Sehmel experiment lasted only for one month, while the Langer work pulls together nearly two years of research. Additionally, the month Sehmel examined, July 1973, may have been a month with less dramatic wind speeds than sometimes seen in this area. Evidence from the wind records used in the first part of this study suggested that greater velocity winds were likely during the winter rather than during the summer. For these reasons, we do not hold much value in the comparison of the Sehmel estimate to the Langer estimate. This exercise does demonstrate the difficulty met in comparing the research of different scientists done at the RFP. Each scientist chose not only a different analysis technique, but also a different endpoint, making it necessary for us to estimate other unknown parameters in order to compare the values.

What this comparison does do is give some range to the releases from the 903 Area east field during periods after 1970. Although 200 μCi y^{-1} seems to represent a reasonable estimate for a source term from this area, the release measured at a minimally more distant location (300 m to the east of the original 903 Area) is much smaller, indicating that airborne quantities probably dropped off dramatically with distance from the site. At any rate, quantities of plutonium released from the remaining source are small when compared to estimates for 1968–1969.

AIR SAMPLER DATA COLLECTED NEAR THE 903 AREA

Another source of information considered to help provide estimates of plutonium releases after 1970 from the 903 Area east field was the data collected by several air samplers located east of the former 903 Area. Air samplers were operated by both the RFP contractor and the Colorado Department of Health.

A difficulty with using these data to establish a model for resuspension from east field releases comes from subtracting fallout background because the fallout data were collected quarterly and the contractor data at the RFP perimeters stations were collected weekly. As with the Langer and Sehmel data, we struggled with the problem of different sampling and analytical techniques. Figure V-1 shows yearly comparisons of annual average plutonium collected at the contractor's perimeter station at Access Road and Indiana Street.
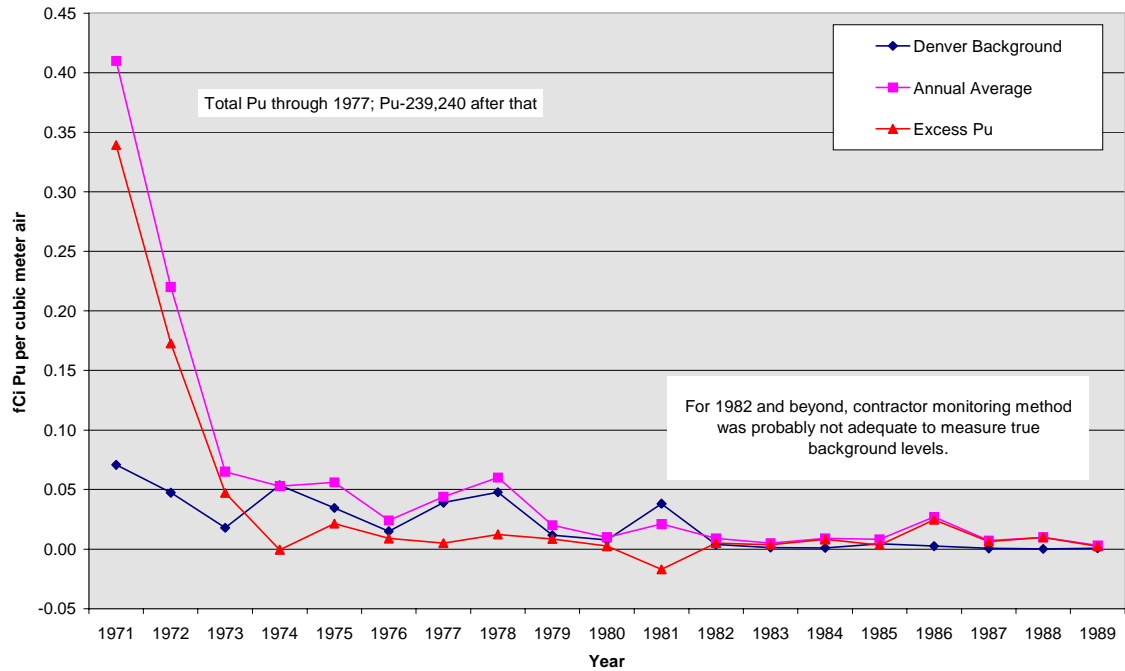
Figure V-1. Plutonium collected at the RFP perimeter station located directly east of the plant (Access Road at Indiana Street). Plutonium air sampling concentrations at this station, shown by squares, are very difficult to distinguish from background collected in Denver, shown by diamonds. Excess plutonium concentrations are shown by the triangles, and, in most cases, they are lower than or the same as fallout concentrations at the background measurement station in Denver.

Data at perimeter stations in all directions around the RFP show similar average and maximum weekly values regardless of the direction from the site. This observation makes it very difficult to employ a modeling technique for resuspension similar to that used to establish suspension releases from the 903 Area.

RESUSPENSION AFTER 1970

Because of the number of difficulties cited here in merging the available information to produce source term estimates from resuspension, we do not feel we have the necessary information to perform a source term and transport calculation. The risk of excess cancer due to resuspension from the 903 Area is closely tied with the spatial deposition of plutonium. As a result, we will use a resuspension factor approach to estimate air concentration from deposited material after 1970.

The resuspension factor is given by

$$C_{a_i} = C_{s_i} \cdot RF \tag{V-1}$$

where

C_{ai} = air concentration at location i (Ci m^{-3})

C_{si} = soil concentration at location i (Ci m^{-2})

RF = resuspension factor (m^{-1}).

Using this equation allows us to determine air concentration from soil concentration, assuming that we know something about the resuspension factor (RF).

The resuspension factor has been estimated by researchers during their resuspension work at the 903 Area east field. Langer estimated a resuspension factor to range from 10^{-13} to 10^{-10} m^{-1} (Langer 1991). Sehmel reported maximum RF values for bare soil to range from 10^{-9} to 10^{-5} m^{-1} (Sehmel and Orgill 1973). We can use these site-specific resuspension factor values in combination with our calculated deposition patterns to determine air concentration and risk from plutonium released from the east field after 1970.

These estimates will be burdened by uncertainty, not only in our deposition estimates but also in the large range of resuspension factors provided in the literature. We also recognize that estimating air concentrations this way means that we will not be accounting for the transport of material once it is deposited. Because of the small quantities, however, we are confident that this will not be a major factor.

It would be virtually impossible to perform source term and transport calculations on a source of this nature. Once the primary source of contamination (the 903 Area) was covered, the remaining source in the east field was not static. This source continually changed, and it would be a major undertaking to model this source, particularly since releases from it were minimal when compared to 1968–1969 releases. The available resuspension experiment literature supports our conclusion that the plutonium releases from the east field were minimal and can be adequately modeled using the resuspension factor technique.

CHAPTER VI

REFERENCES

Bagnold, R.A. 1941. The Physics of Blown Sands and Desert Dunes. Methuen, London.

Barrick, C.W. 1980. *A Review of the Rocky Flats Plant Waste Oil Drum Leak Incident (1958– 1970)*. Environmental Sciences Department. October 17.

Biles, M.B. 1970. *Summary of Contaminated Waste Storage Burial at the Rocky Flats Plant Site*. Division of Operational Safety. 1500656. December.

Bronesky, J.J. 1977. The Factual Development of the Barrel Incident. Draft Memo. Rockwell International proprietary document. December.

Card, D.H. 1977. *History of Buried Transuranic Waste at INEL*. WMP 77-3. March.

ChemRisk. 1994. *Exposure Pathway Identification and Transport Modeling*. Health Studies on Rocky Flats; Phase I: Historical Public Exposures. Colorado Department of Public Health and Environment, Project Task 6. May.

Chepil, W.S. 1952. "Improved Rotary Sieve for Measuring State and Stability of Dry Soil Structure." *Soil Science Society of America Proceedings* 16: 113–117.

Cowherd, C. Jr., G.E. Muleski, P.J. Englehart, and D.A. Gillette. 1985. *Rapid Assessment of Exposure to Particulate Emissions from Surface Contamination Sites*. EPA/600/8-85/002. U.S. Environmental Protection Agency, Office of Health and Environmental Assessment. Washington, D.C. February.

Dow (Dow Chemical Corporation). 1968. *Health Physics Status Reports for Buildings. Site Survey, Equipment, Decontamination, and Construction*. Monthly reports, February– December.

Dow. 1969. *Health Physics Status Reports for Buildings. Site Survey, Equipment, Decontamination, and Construction*. Monthly reports, January–December.

Dow. 1974. A Historical Summation of Environmental Incidents Affecting Soils at or Near the USAEC Rocky Flats Plant, Draft. Repository Document RE-318. January 24.

EG&G (EG&G Idaho, Inc.). 1994. *A Comprehensive Inventory of Radiological and Nonradiological Contaminants in Waste Buried in the Subsurface Disposal Area of the INEL RWMC During the Years 1952–1983*. EGG-WM-10903. June.

EPA (U.S. Environmental Protection Agency). 1987. *On-Site Meteorological Program Guidance for Regulatory Modeling Applications*. EPA-450/4−87−013. Research Triangle Park, North Carolina.

EPA. 1992. *User's Guide for the Industrial Source Complex (ISC) Dispersion Models Vol. 1, User's Instructions*. EPA-450/4−92−008a. Research Triangle Park, North Carolina.

EPA. 1995. *Compilation of Air Pollution Emission Factors, Fifth Edition*. Office of Air Quality Planning and Standards. Washington, DC.

Ermak, D.L., 1977. "An Analytical Model for Air Pollutant Transport and Deposition from a Fugitive Area Source." *Atmospheric Environment* 11: 231–237.

Gifford, F.A. 1961. "Use of Routine Meteorological Observations for Estimating Atmospheric Dispersion." *Nuclear Safety* 2 (4): 47–51.

Gillette, D.A. 1974. "On the Production of Soil Wind Erosion of Soil: Effect of Wind and Soil Texture." *Atmosphere-Surface Exchange of Particulate and Gaseous Pollutants*. ERDA Symposium Series 38, Oak Ridge, Tennessee. 591–609.

Gillette, D.A. 1977. "Fine Particulate Emissions Due to Wind Erosion." *Transactions of the ASAE*. 890–897.

Gillette, D.A. 1997. Personal Communication with Jill M. Weber, *Radiological Assessments Corporation*.

Gillette, D.A. and C. Cowherd. 1982. "The Concept of Resuspension Rates Applied to Problems of Fugitive Dust Emissions and Wind Erosion." In *Precipitation Scavenging, Dry Deposition, and Resuspension.* Edited by Pruppacher et al. Elsevier Science Publishing Co., Inc. 1059–1072.

Gillette, D.A., B. Marticorena, and G. Bergametti. 1998. "Change in the Aerodynamic Roughness Height by Saltating Grains: Experimental Assessment, Test of Theory, and Operational Parameterization." *Journal of Geophysical Research* 103 (D6): 6203–6209.

Harlan, R.A., J.L. Lawless, R.N. Chanda, and R.A. Deal. 1972. "Simplifications Used for Rocky Flats Drum and Small Package Counters. *Trans. Am. Nucl. Soc.* 15: 83.

Hayden, J.A. 1974. Dow Chemical Company Letter to Phil Krey. Subject: Particle Size of Plutonium in Rocky Flats Soil. September 24.

Killough, G.G., M.J. Case, K.R. Meyer, S.K. Rope, D.W. Schmidt, B. Shleien, W.K. Sinclair, P.G. Voillequé, and J.E. Till. 1996. *Task 6: Radiation Doses and Risk to Residents from FMPC Operations from 1951–1988. Volume II, Appendices*. 4-CDC-Fernald-1996-DRAFT (Vol. II). *Radiological Assessments Corporation,* Neeses, South Carolina.

Langer, G. 1983. "Activity, Size, and Flux of Resuspended Particles from Rocky Flats Soil." In *Precipitation Scavenging, Dry Deposition, and Resuspension.* Edited by Pruppacher et al. Elsevier Science Publishing Co., Inc. 1161–1172.

Langer, G. 1986. *Dust Transport–Wind Blown and Mechanical Resuspension, July 1983 to December 1984*. RFP-3914 (DOE/TIC-4500). Rocky Flats Environmental Technology Site, Golden, Colorado.

Langer, G. 1991. *Resuspension of Soil Particles from Rocky Flats Containing Plutonium Particulates.* GHS-0070-91. EG&G Rocky Flats, Inc. Golden, Colorado.

Lawless, J.L. and R.N. Chanda. 1970. "A Plutonium Waste Counter." *Trans. Am. Nucl. Soc.* 13: 748.

Litaor, M.I., D. Ellerbroek, L. Allen, and E. Dovala. 1995. "Comprehensive Appraisal of $^{239+240}$Pu in Soils Around Rocky Flats, Colorado." *Health Physics* 69(6): 923–935.

Litaor, M.I., G.R. Barth, and E.M. Zika. 1996. "Fate and Transport of $^{239+240}$Pu and ^{241}Am in the Soil of Rocky Flats, Colorado." *Journal of Environmental Quality* 25(4): 671-683.

Litaor, M.I and L. Allen. 1996. "A Comprehensive Appraisal of ^{241}Am in Soils Around Rocky Flats, Colorado." *Health Physics* 71(3): 347–357.

Lockhart, L.B., R.L. Patterson, Jr., and W.L. Anderson. 1964. *Characteristics of Air Filter Media Used for Monitoring Airborne Radioactivity.* Report NRL-6054. U.S. Naval Research Laboratory. National Technical Information Service, Springfield, Virginia.

Loser, J.R. 1970. *The general dispersion of plutonium in soil surrounding the Rocky Flats Plant.* An initial study prepared for J.R. Seed. November 16.

Luckett, L.L., A.A. Dickman, C.R. Wells, and D.J. Vickery. 1982. *History of Rocky Flats Waste Streams.* March 10.

Meyer, H.R., S.K. Rope, T.F. Winsor, P.G. Voillequé, K.R. Meyer, L.A. Stetar, J.E. Till, and J.M. Weber. 1996. *The Rocky Flats Plant 903 Area Characterization.* RAC Report #2-CDPHE-RFP-1996. *Radiological Assessments Corporation*, Neeses, South Carolina.

Miller, C.W., and L.M. Hively. 1987. "A Review of Validation Studies for the Gaussian Plume Atmospheric Dispersion Model." *Nuclear Safety* 28 (4): 522–531.

Morrison. 1991. Personal Communication with ChemRisk Scientists.

Nathans, M.W., W.D. Holland, and H.C. Shaw. 1971. *The Size Distribution and Plutonium Concentration of Particles from the Rocky Flats Area.* TLW-6111. LFE Corporation, Richmond, California.

Neiburger, M., J.G. Edinger, and W.D. Bonner. 1971 *Understanding Our Atmospheric Environment.* San Francisco, California: W.H. Freeman and Company.

Pasquill, F. 1961. "The Estimation of the Dispersion of Windborne Material." *The Meteorological Magazine* 90: 33–49.

Porch, W.M. 1979. *Resuspension of Toxic Aerosol using MATHEW-ADPIC Wind Field Transport and Diffusion Codes.* UCID-18204. Lawrence Livermore Laboratory, Livermore, California.

Press, W.H., S.A. Teukolsky, W.T. Vetterling, and B.P. Flannery. 1992. *Numerical Recipes: The Art of Scientific Computing*. New York, New York: Cambridge University Press.

Putzier, E.A. 1970a. 903 Oil Drum Storage Area—A Brief History (includes detailed table of individual barrels to 903 area). April 14.

Putzier, E.A. 1970b. Memo. Status report—waste disposal coordination August 1970 (includes shipments to Idaho Falls). September 30.

Ramsdell, J.V., Jr., C.A. Simonen, and K.W. Burk. 1994. *Regional Atmospheric Transport Code for Hanford Emission Tracking* (RATCHET). PNWD-2224 HEDR. Battelle Pacific Northwest Laboratories, Richland, Washington.

Rood, A.S. 1997a. *Performance Evaluation of Atmospheric Transport Models*. RAC Report 3-CDPHE-RFP-1996. *Radiological Assessments Corporation*, Neeses, South Carolina, December.

Rood, A.S. 1997b. *Estimated Exposure and Lifetime Cancer Incidence Risk from Routine Plutonium Releases*. RAC Report 08-CDPHE-RFP-1997. *Radiological Assessments Corporation*, Neeses, South Carolina. December.

Robertson, E. and P.J. Barry. 1989. "The Validity of a Gaussian Plume Model When Applied to Elevated Releases at a Site on the Canadian Shield." *Atmospheric Environment* 23 (2): 351–362.

Rope, S.K., K.R. Meyer, M.J. Case, D.W. Schmidt, M. Dreicer, T. Winsor, L. Stetar, and J.E. Till. 1997. *Rocky Flats Dose Reconstruction Project Phase II: Toxicity Assessment and Risk Characterization, Task 4: Evaluation of Historical Environmental Data*. RAC Report 01-CDPHE-RFP-1997. *Radiological Assessments Corporation*, Neeses, South Carolina. March.

Seed, J.R. 1970. *Status Report of Committee on Plutonium in Soil*. Document Code L00047. Repository Document IN 560.

Seed, J.R., K.W. Calkins, C.T. Illsley, F.J. Miner, and J.B. Owen. 1971. *Committee Evaluation of Plutonium Levels in Soil Within and Surrounding USAEC Installation at Rocky Flats, Colorado*. Dow Chemical, Rocky Flats Division Report RFP INV 10. Repository Document AC 726.

Sehmel, G.A. 1976. *Airborne ^{238}Pu and ^{239}Pu Associated with the Larger than Respirable Resuspended Particles at Rocky Flats during July 1973*. BNWL-2119, UC-11. Battelle Pacific Northwest Laboratories, Richland, Washington.

Sehmel, G.A. 1980. "Particle Resuspension: A Review." *Environment International* 4: 107–127.

Sehmel, G.A. and F.D. Lloyd. 1976. "Resuspension of Plutonium at Rocky Flats." In *Atmospheric Surface Exchange of Particulate and Gaseous Pollutants*. ERDA Symposium Series, Oak Ridge, Tennessee. 757–779.

Sehmel, G.A. and M.M. Orgill. 1973. "Resuspension by Wind at Rocky Flats." In *Pacific Northwest Laboratory Annual Report for 1972*. BNWL-1751-PT-1. Battelle Pacific Northwest Laboratories, Richland, Washington.

Simpson, D., D.A. Perrin, J.E. Varey, and M.L. Williams. 1990. "Dispersion Modeling of Nitrogen Oxides in the United Kingdom." *Atmospheric Environment* 24 (7): 1713–1733.

Turner, D.B. 1964. "A Diffusion Model for an Urban Area." *J. Applied Meteorology* 3: 83–91.

Voillequé, P.G. 1998. *Estimated Airborne Releases of Plutonium during the 1957 Fire in Building 71*. RAC Report No. 10-CDPHE-RFP-1998. *Radiological Assessments Corporation*, Neeses, South Carolina. March

Volchok, H.L., R.H. Knuth, and M.T. Kleinman. 1972. "The Respirable Fraction of Plutonium at Rocky Flats." *Health Physics* 23: 395–396.

Winges, K.D. 1990. *User's Guide for the Fugitive Dust Model*. EPA-910/9-88-202R. U.S. Environmental Protection Agency, Region 10, Seattle, Washington.