CHAPTER III

AMBIENT AIR MONITORING

INTRODUCTION

Phase I of the Rocky Flats Dose Reconstruction Project identified inhalation as the most important route of exposure to contaminants released from Rocky Flats (ChemRisk 1994c). Therefore, a particularly careful look at ambient air monitoring data is warranted. *Ambient* air monitoring, which monitors air in the open environment, should be distinguished from *effluent* air monitoring, which monitors effluent air leaving facilities via stacks, exhaust fans, etc.

As indicated in the introduction of this report, there is a break point in the contractor's ambient air monitoring program in the early 1970s, when routine plutonium analyses began. Before that time, measurements of total long-lived alpha activity were used as an indication of plutonium in air. We began our evaluation of the usefulness of the contractor's air monitoring data by examining these plutonium-specific measurements. These data sets were not examined in Phase I of the dose reconstruction. Based on our framework exercise (Chapter I), these relatively recent data could potentially be useful for analyzing spatial or temporal trends and/or for verifying release quantities from routine operations. Unfortunately, plutonium was not measured in air during the time period when the highest releases from Rocky Flats occurred. Therefore, we have examined the contractor's historical measurements of long-lived alpha activity, particularly in the 1960s.

Spatial trend refers to how a material is distributed in the environment, for example, with distance away from the facility. _Temporal trend_ refers to how the concentration of a material changes over time.

This chapter begins with a review of routine monitoring programs for plutonium in air that were run by agencies independent of the operation of the Rocky Flats Plant (RFP). These data establish a history of fallout plutonium concentrations that is necessary for understanding the RFP monitoring data. Also, the independent monitoring programs provide an important cross-check on the contractor's results. Subsequent sections of this chapter address the Rocky Flats contractor air monitoring programs. A few nonroutine air monitoring studies are addressed, and data quality issues are discussed. Maps and lengthier detailed material relating to air monitoring are included in Appendix B.

ROUTINE MONITORING OF PLUTONIUM IN AIR BY INDEPENDENT AGENCIES (1965–1990)

In a public study, independent monitoring by groups not associated with the operation of the site can be important to developing trust in historical monitoring results. There are also important technical reasons to investigate monitoring done at some distance from the site. This section provides an overview of independent monitoring programs. It should be understood that the localized monitoring of these independent monitoring programs is rarely as comprehensive as the site's program, but they can provide a cross-check of results generated by the site at a similar location and time.

In addition to programs that sampled air near the RFP, other programs were identified that were useful in determining historical background levels of plutonium in air. An understanding of

background can be particularly important to a dose reconstruction for U.S. Department of Energy (DOE) sites because often the time periods of highest releases from these sites overlap or coincide with the time periods of highest depositions of fallout from weapons testing.

Other Sources of Plutonium in the Environment

The term *background* usually refers to amounts of materials that occur naturally in the environment, without any input from human activities. However, some materials in our environment are there primarily from human activities. Essentially all plutonium in the environment is due to human activities. For the purposes of this discussion, therefore, background plutonium in the environment around Rocky Flats is defined as that *from sources other than the RFP*.

Before the early 1970s, measurements of total long-lived alpha activity in air were used by the RFP contractor as an indication of plutonium activity. However, there are a number of naturally occurring radionuclides that emit alpha particles. Most of the independent agencies discussed in this chapter did not monitor alpha activity in air. We have estimated the background alpha activity in air in other ways, described later in this chapter. The scope of this section is long-term monitoring of plutonium in air by independent agencies.

Two isotopes, or forms, of plutonium, ^{239}Pu and ^{240}Pu, cannot be distinguished from one another by typical analytical methods. Although often written just as ^{239}Pu, both isotopes are included. An analysis reported as total Pu includes all isotopes of plutonium, such as ^{238}Pu .

The primary source of non-Rocky Flats plutonium in the environment is fallout from global weapons testing. The first nuclear explosion occurred in 1945 in New Mexico. Fallout became recognized as a possible public health problem in the mid-1950s. In 1958, the U.S., United Kingdom (U.K.), and the Soviet Union, who were all conducting aboveground testing, declared a moratorium. However, in 1961, without advance warning, the Soviet Union unilaterally broke the moratorium agreement and exploded about 50 devices. The U.S. responded, and major escalation of testing occurred. In early 1963, a test ban agreement was signed by the U.S., U.K., and the Soviet Union. France and China continued to conduct aboveground tests (particularly in 1968–1970), but not on as large a scale as the other countries had conducted before 1963. A more detailed review of the timing of weapons testing and the resulting plutonium deposition to soils is given in Appendix H of this report. In general, the highest levels of weapons testing fallout in air and deposition samples occurred in the first half of the 1960s.

Plutonium is a component of the weapons fallout that remains after the nuclear detonation. Nuclear fallout has been injected both into the troposphere (lower atmosphere) and stratosphere (upper atmosphere). These two layers of the atmosphere circulate rather independently, with material injected into the stratosphere being distributed globally. The size of the explosion and the height of the detonation are primary factors in whether fallout occurred close to or far away from the test location. The larger megaton bombs injected a large fraction of their fallout into the stratosphere. Some fallout debris stayed in the stratosphere for many months, coming down into the lower atmosphere during seasonal mixing of the atmospheric layers. Essentially all the debris from smaller (kiloton) bombs was deposited within a few months following injection into the atmosphere, because most fallout from these was confined to the troposphere.

A significant source of ^{238}Pu in the environment was the burnup of a satellite in April 1964 (Hardy et al. 1972). This malfunction resulted in an estimated 17 kilocuries of ^{238}Pu being released to the upper atmosphere. Although most of this material ended up falling out in the southern hemisphere, there were detectable amounts in the northern hemisphere that affected the ratio of ^{238}Pu to ^{239,240}Pu in fallout after that time.

Two independent monitoring programs began measuring plutonium in surface air in the U.S. in 1965. No plutonium-specific monitoring of ambient air was occurring as far back as the 1957 fire at Rocky Flats. However, independent monitoring programs were in place before the highest releases from the 903 Area at Rocky Flats occurred (Meyer et al. 1996).

Monitoring of Plutonium in Ambient Air by the Public Health Service and U.S. Environmental Protection Agency (1965–1990)

In 1959, the President of the U.S. directed the Secretary of Health, Education, and Welfare to intensify that Department's activities in the field of radiological health. The Department was assigned, among other things, primary responsibility within the Executive Branch for the collation, analysis, and interpretation of data on environmental radiation levels. Within the Department, this responsibility was delegated to the Division of Radiological Health, Public Health Service (PHS). As a step in the discharge of this responsibility, the PHS initiated the monthly publication *Radiological Health Data* in April 1960.

Initially, the radiological measurements were made on surface water, but they expanded to include air, vegetation, and food as part of a nationwide monitoring network. Gross beta activity of radioactive particulates in air at a number of U.S. cities, including Denver, is tabulated in the PHS monthly reports. Data collected by several agencies are published, including the PHS Radiation Surveillance Network (RSN) and the U.S. Naval Research Laboratory (80th Meridian Network). The RSN was established in 1956 in cooperation with the Atomic Energy Commission to provide a means of promptly determining increases in environmental radioactivity from fallout during nuclear weapons tests. In May 1960, there were 44 stations in urban locations.

Particulates **are small particles suspended in air. Sampling of particulates is conducted by drawing the air through a filter that captures the particles.**

Beginning in November 1965, the RSN started routine analysis for plutonium in air particulates. The monitoring was begun because plutonium had been injected into the atmosphere by nuclear devices, and inhalation was believed to be the principal route of exposure. The cities monitored are shown in Figure III-1.

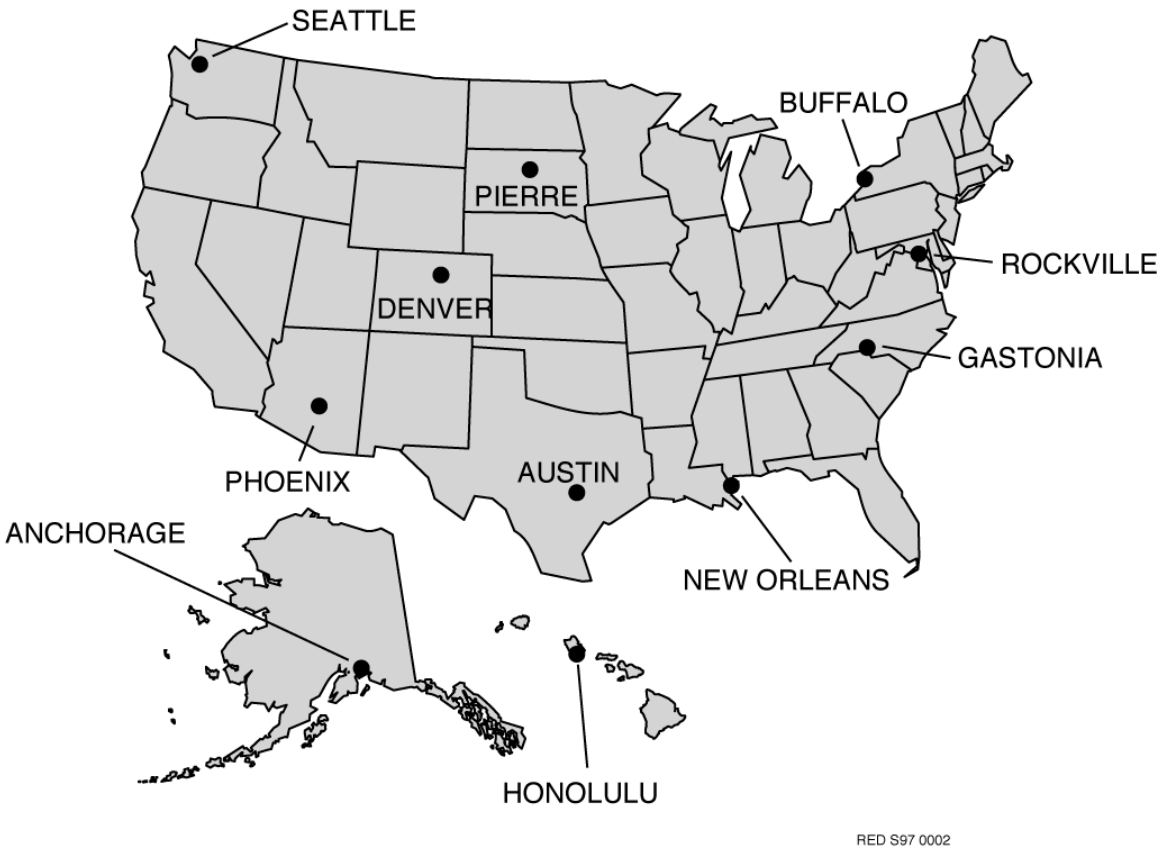

Figure III-1. Public Health Service locations for monitoring of plutonium in airborne particulates in the late 1960s.

A monthly composite of air filters from these stations was analyzed for total plutonium. The sampling and analytical procedures, as described in Russell et al. (1966), are summarized in the following text. The samples were collected on carbon-loaded cellulose filters. Sampler air flow was determined by a gauge that read the pressure drop across the orifice and was calibrated in cubic feet per minute. The air filters were ashed in a platinum crucible, brought completely into solution with a nitric acid-hydrofluoric acid treatment, converted to the chloride ion, and brought to volume in a volumetric flask. The solution was then divided into two aliquots, 70% and 30%, and each aliquot was analyzed and counted separately for total plutonium according to the procedure documented in Levine and Lamanna (1965). The total activity was the sum of the results for both aliquots. The chemical yield of the analytical procedure was approximately 80% as determined by spiked samples. The samples were counted for 1000 minutes in a proportional counter. The counting error at the 95% confidence level was less than 10% for most of the samples, and the accuracy of the air flow measurement was estimated to be approximately 20% at the 1 picocurie (pCi) activity level.

Radiological Assessments Corporation (*RAC)* has compiled the results of the PHS monitoring from the published reports. Figure III-2 illustrates the time trend for Denver, Colorado; Pierre, South Dakota; and Rockville, Maryland. In July 1970, the Rockville station was discontinued and monitoring was picked up in Baltimore, Maryland. The two Maryland cities are plotted as one record (called "Maryland") in Figure III-2. The detailed data that make up this plot are included as Table B-1 of Appendix B. The femtocurie (fCi) is a convenient unit to present concentrations of this magnitude. The fCi is 1×10^{-15} Ci, or 0.001 pCi.

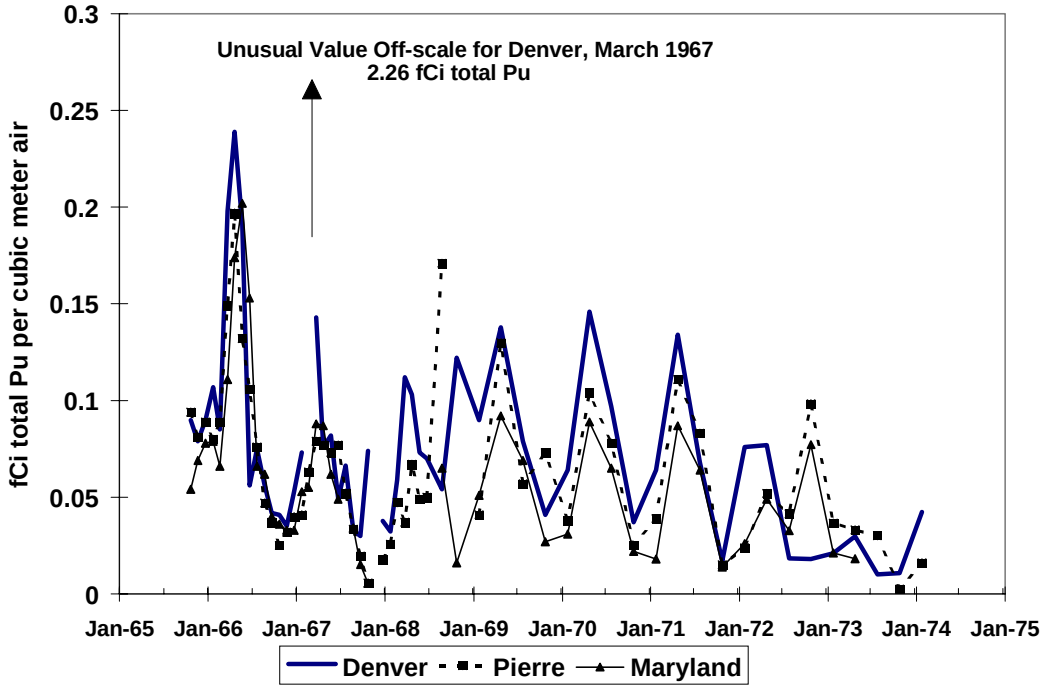

Figure III-2. Total plutonium concentrations in ambient air in Denver, Colorado; Pierre, South Dakota; and Rockville, Maryland, as measured and reported by the Public Health Service. The program was transferred to the U.S. Environmental Protection Agency in 1973. An unusual high value in Denver air in March 1967 is omitted from the plot (shown as off-scale with an arrow).

The seasonal mixing effect (exchange between stratosphere and troposphere) is very apparent in the air monitoring record of the late 1960s. Exchange between the two atmospheric layers is increased in spring, when plutonium in the upper atmosphere is brought into the lower atmosphere and picked up by the samplers. This cycling produces lower concentrations in surface air in the winter and higher concentrations in the spring/summer in the mid-northern latitudes. The same trends in plutonium concentrations in air were observed at these three locations, which are widely spaced across the U.S. This similarity indicates that the stations are responding to global rather than local events and cycles. In general, the concentrations in Denver air are slightly higher than at the other two locations.

Seasonal and longer-term time trends are important to consider when interpreting the historical monitoring record of radionuclides that were present in weapons fallout.

Over the interval of time shown here, the median ratio for Pierre-to-Denver is 0.8 and the median Maryland-to-Denver ratio is 0.7.

***Compositing* refers to a combining of air filters before analysis. A long compositing time is necessary when concentrations are low.**

Beginning with the August 1967 samples, the PHS air filters were analyzed for ^{238}Pu and ^{239,240}Pu rather than total plutonium. The ratio of these isotopes is also presented in the PHS data tables. The analytical procedure for separating the isotopes is described in PHS (1968). The isotopic concentrations are summed to give a total plutonium concentration for the purposes of our trend graph (Figure III-2). Samples were composited monthly until the 4th quarter of 1968, when the program changed to quarterly composites.

The high value in Denver in March 1967 (2.259 pCi/1000 m^3 or 2.259 fCi m^{-3}) was footnoted in the PHS report (PHS 1967) as follows:

> This sample was counted by alpha-particle spectrometry for individual isotopes of plutonium. The approximate isotopic breakdown is 1.85 pCi/1000 m^3 239-240Pu and 0.41 pCi/1000 m^3 238Pu.

There was no indication in this report that the sample result was incorrect in any way. A statement was made in a subsequent RFP contractor memo that "The people at the Northeast Radiological Health Lab think the sample was a 'dog'" (Lee 1970).

An activity ratio of 0.2 (^{238}Pu /^{239,240}Pu), which was measured for this high sample, is not indicative of Rocky Flats plutonium (discussed later in this section). Air monitoring conducted closer to the RFP at this time was only for long-lived alpha activity, and the minimum detectable concentration for this analysis was high (around 5 fCi m^{-3}). Concentrations of alpha activity in onsite air, measured by Dow, were much less in the spring of 1967 than they were in 1968 and 1969 (discussed later in this chapter). The isotopic ratio and the alpha count trend suggest that a release from Rocky Flats was not the source of this relatively high value.

The minimum detectable concentration is the lowest concentration of a material that can be measured accurately by a particular measurement technique.

In 1973, the PHS radiological air monitoring program was integrated into the Environmental Radiation Ambient Monitoring System (ERAMS) of the U.S. Environmental Protection Agency (EPA) Office of Radiation Programs. Monitoring continues to this day. The publication *Environmental Radiation Data* is currently compiled and distributed quarterly by the EPA's Eastern Environmental Radiation Facility in Montgomery, Alabama. The radiochemical procedures currently used are documented in EPA (1984).

RAC has obtained the complete EPA report series to continue the earlier PHS data record. Fallout plutonium concentrations have continued to decline. Beginning in 1971, an uncertainty was reported with each measurement. The EPA has continued to report data for plutonium in air throughout the last 15 years even though recent concentrations are largely not detectable. (When the analytical uncertainty is greater than the estimated concentration, the material can be considered to be not detected.) A long-term trend of plutonium in air reported by the PHS/EPA is plotted in the next section along with data from the Health and Safety Laboratory.

Monitoring of Plutonium in Ambient Air by the Health and Safety Laboratory/ Environmental Measurements Laboratory (1965–1984)

The Health and Safety Laboratory (HASL) of New York City changed its name in 1975 to the Environmental Measurements Laboratory (EML). Operated by DOE and its predecessors (Energy Research and Development and the Atomic Energy Commission), the HASL was not independent of the DOE complex, but it was not involved in operating the RFP. The HASL was a high-quality research laboratory that was instrumental in developing many techniques for radiochemical analyses. As opposed to a DOE production site's compliance-based monitoring program, the objective of the HASL surface air program was to study the spatial and temporal distribution of natural and man-made radionuclides in surface air. Therefore, these results are very relevant to a dose reconstruction study. In addition, the HASL did some relatively long-term monitoring near Rocky Flats in the 1970s. Figure III-3 shows the U.S. locations that were being monitored for plutonium in air in the 1970s.

This surface air monitoring program (for radioactivity) began in 1963 as a continuation of a program initiated by the U.S. Naval Research Laboratory in 1957. Like the PHS, analysis of airborne particulates for plutonium began in 1965. The U.S. stations still active in 1983 were New York City, Miami, and Mauna Loa. In addition, they were then sampling at Barrow, Alaska; Beaverton, Oregon; Rexburg, Idaho; and Chester, New Jersey. New York City has always been one of the cities monitored.

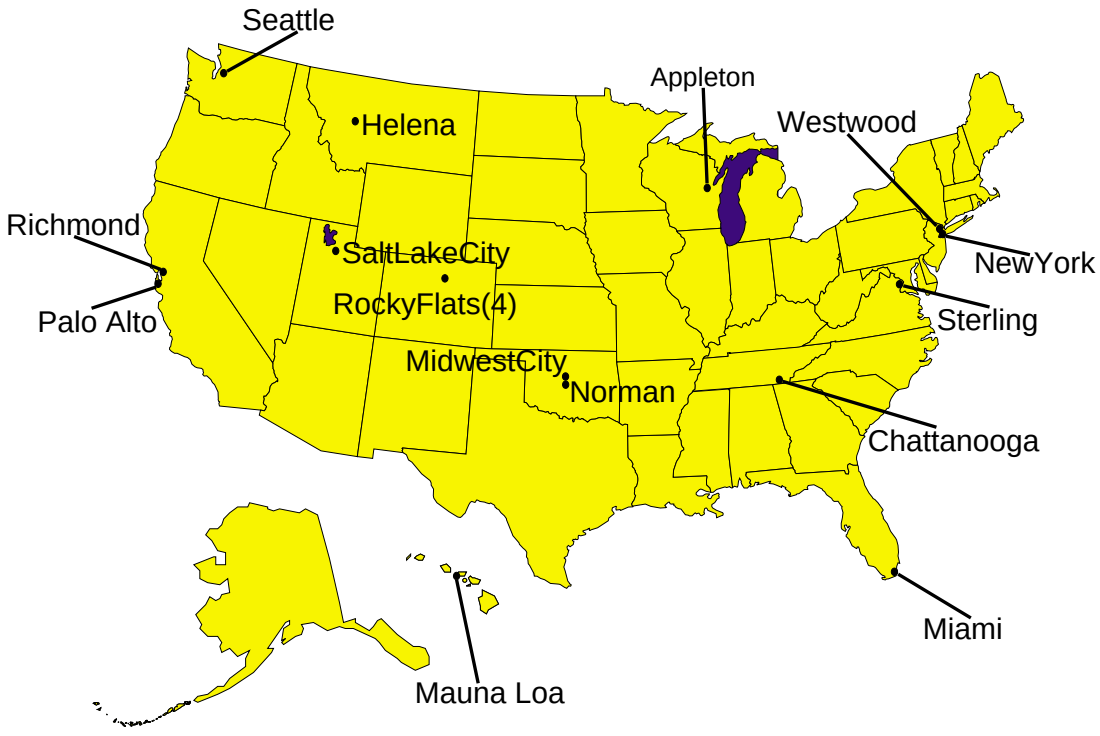

Figure III-3. Health and Safety Laboratory surface air monitoring locations (U.S.) in the 1970s. Other locations were monitored in other countries.

These monitoring data were reported periodically in HASL (and later EML) reports (e.g., Feely et al. 1985). Quality assurance included routine analysis of reference materials (standards),

blanks, and duplicates, which were submitted without identifying the source to the analyst. Data quality results and issues are discussed in a later section of this chapter.

The data reported for air samples from several cities, as well as Rocky Flats, have been compiled by *RAC* into an electronic spreadsheet for plotting and analysis. In the earlier period of this program, the reporting and analysis frequency was monthly. Figure III-4 illustrates the temporal trend of plutonium in air in three cities in the eastern U.S. between 1965 and 1971.

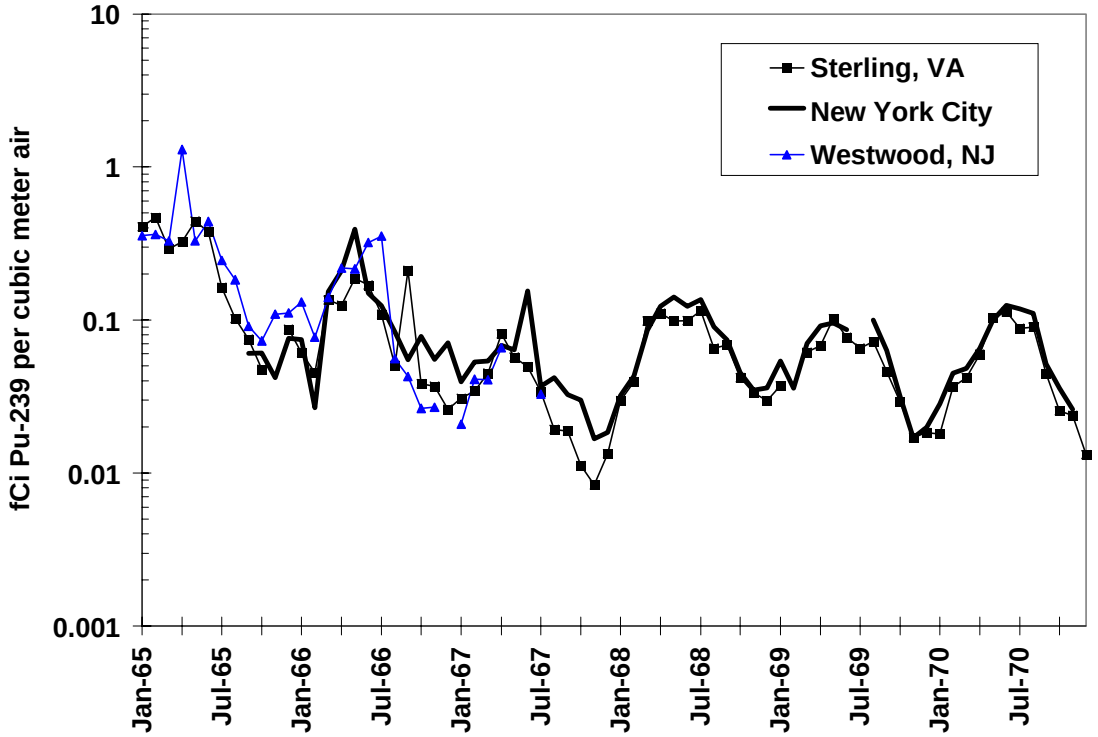

Figure III-4. Concentrations of ^{239,240}Pu in surface air in three cities in the eastern U.S. (Health and Safety Laboratory data). Note the semilogarithmic scale, unlike Figure III-2.

Again, the most striking feature of the time trend is the cycling of concentrations in surface air, which is apparent at all three of these locations. In 1968 and 1970, France conducted a relatively extensive series of nuclear tests in the Pacific. China exploded a large-yield nuclear device in December 1968 and another in September 1969. These tests added to the atmospheric inventory of plutonium that was involved in the seasonal mixing evident in Figure III-4.

Figure III-5 shows data from the same monitoring program, carried into the next decade. Note that this concentration scale extends to 10 times less than Figure III-4. The Salt Lake City monitoring began in 1971. In the 1970s, China was responsible for most of the aboveground testing of nuclear weapons. The last atmospheric nuclear weapons test of a high yield occurred at the Lop Nor test site in China on October 18, 1980. Zirconium-95 (indicative of fresh fallout) was clear in the New York City and Mauna Loa sites, especially in March through May 1981. As late as 1983, a springtime peak in residual fallout was still discernible, but shortly after this, the plutonium levels were frequently at or near detection limits. Therefore, the EML discontinued its

routine analysis of plutonium in air. In May 1986, fallout from the Chernobyl accident arrived in New York City, but by that time EML was no longer reporting plutonium in air.

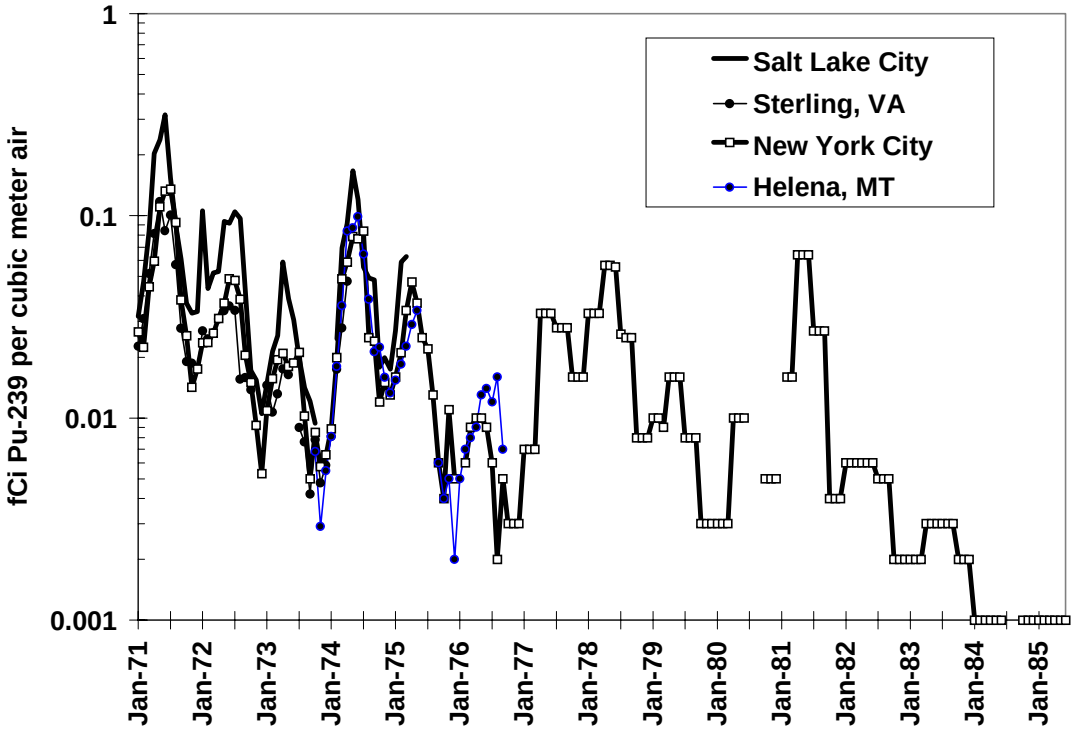

Figure III-5. Plutonium-239,240 in air at four U.S. locations (Health and Safety Laboratory data, 1971–1985). The blocky appearance of the plot in the latter half of this time period is due to quarterly compositing of filters; filters were composited monthly in the earlier period.

Summary of Time Trend of Fallout Plutonium

Figure III-6 presents a long-term view of the time trend in measured fallout plutonium in surface air of the U.S from 1965, when monitoring began, through 1992. The data from the PHS/EPA program and the HASL/EML program track each other well.

The largest amount of global fallout from nuclear weapons testing occurred before monitoring for plutonium in surface air began. For this reason, it is important to estimate concentrations that might have been present during earlier years. This is necessary for interpreting alpha activity measurements made in the peak fallout years. It also provides a benchmark for risk comparison.

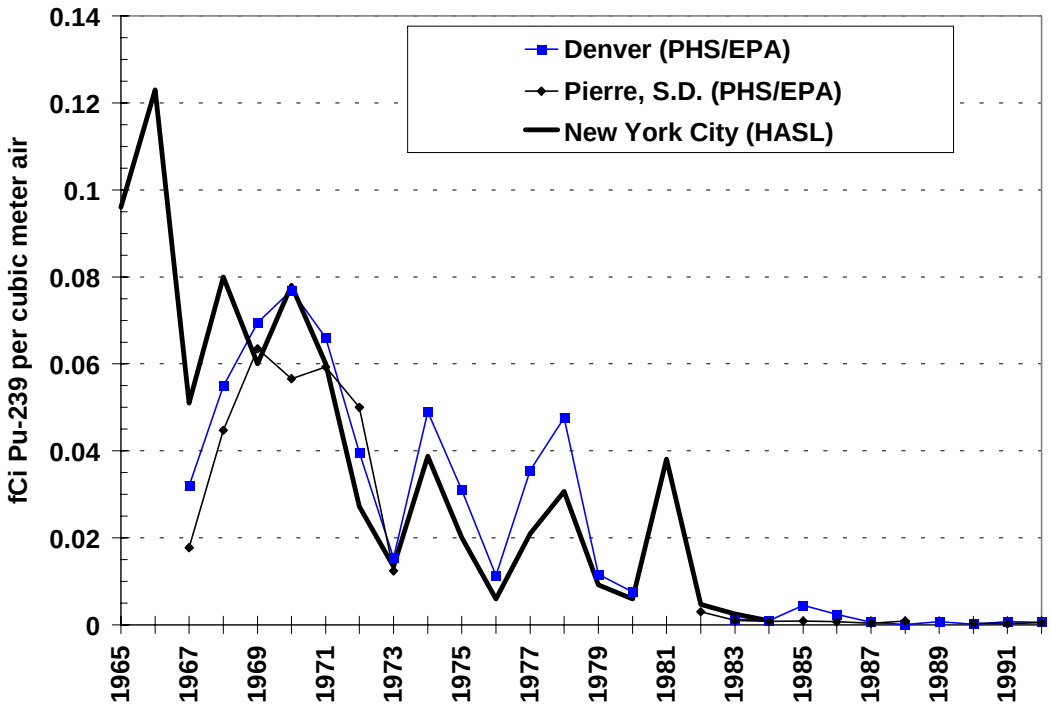

Figure III-6. Long-term view of fallout plutonium in surface air (annual averages).

A back-projection of the concentration of plutonium in Denver air is illustrated in Figure III-7. Before 1965, the concentration of plutonium in air was estimated from the relationship between plutonium concentrations in air and measured depositions of strontium-90 (^{90}Sr), which were monitored before 1965. The HASL was monitoring deposition of fallout ^{90}Sr in Denver during 1959 to 1975 and in New York as early as 1954 (HASL 1977). The ratios of these ^{90}Sr deposition measurements to the measured plutonium concentrations in Denver air after 1965 were used to extrapolate the plutonium concentrations in air to earlier times. A 90% uncertainty range in the back-projection of the plutonium concentration in 1954–1964 (dashed line in upper plot of Figure III-7) ranged from 0.45 to 2.37 times the nominal values, based on the uncertainty in the ratio of strontium deposition to plutonium in air.

During the first 2 years of monitoring plutonium in Denver air, the results were reported as total plutonium, not separately as ^{239,240}Pu and ^{238}Pu. To make those data comparable to the later period, we multiplied the total plutonium results by 0.8, the average proportion of ^{239,240}Pu/Pu for 1967–1971. The range in this proportion was 0.41 to 0.97, with 90% of the values between 0.53 and 0.94. In other words, the 90% uncertainty range on the ^{239,240}Pu concentration for 1965 and 1966 is 0.66 to 1.18 times the nominal values. In 1981 and 1982, there were no data reported for plutonium in air in Denver. Data from New York City were used for those 2 years.

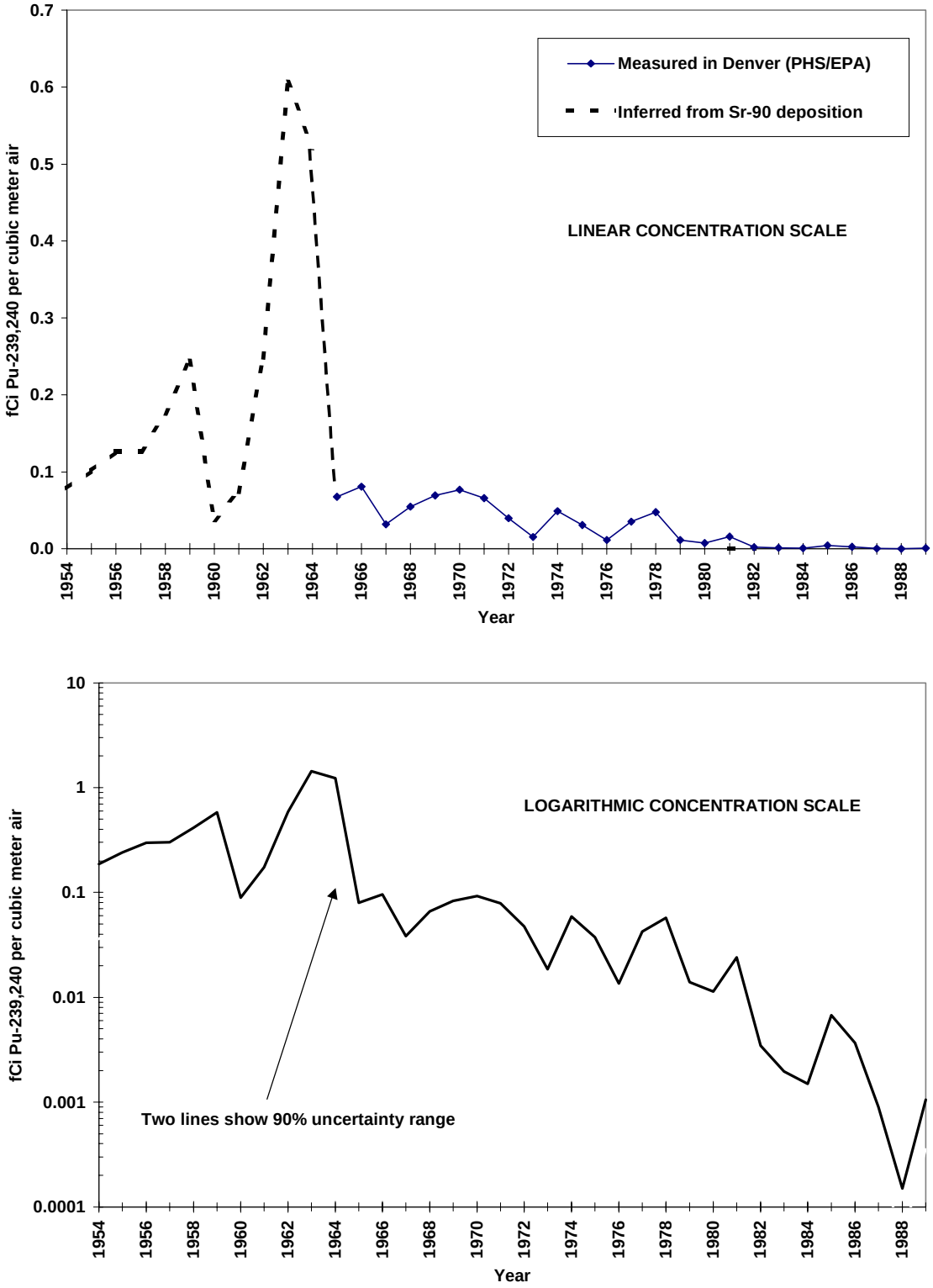

Figure III-7. Time trend in annual average ^{239,240}Pu concentrations in Denver air. See Table B-14 in Appendix B for data and text of this chapter for source of values.

For measured concentrations, the uncertainty range on the annual average was assumed to be ±20% when the concentration was greater than 0.01 fCi m^{-3} and ±50% when it was less. The uncertainty range on the annual average concentrations, both measured and calculated, is illustrated by the two lines in the lower plot of Figure III-7. The nominal values and uncertainty range for plutonium in Denver air between 1954 and 1989 are given in Table B-14 of Appendix B.

In Denver, 95% of the total exposure to fallout plutonium occurred before 1975, with an estimated 37% in 1963 and 1964. The peak annual average concentration of 0.61 fCi m^{-3} in 1963 can be compared to peak estimated concentrations (using a similar plutonium/strontium ratio approach) in Japan of 0.23 fCi m^{-3} in 1964 and 1.7 fCi m^{-3} in New York City in 1963 (Kasai et al. 1984). The integrated air concentration in Denver over the period 1954–1989 is estimated to be 3.0 fCi-y m^{-3}, with a 90% uncertainty range of 1.6 to 6.4 fCi-y m^{-3} (Table B-14). This integrated exposure can be used for risk comparison with estimated exposure from past Rocky Flats releases.

Understanding the differences in the time trends for global fallout and for plutonium releases from Rocky Flats can help to interpret environmental data. Fallout plutonium peaked in 1963–1964. In contrast, the highest releases of airborne plutonium from Rocky Flats are believed to have occurred in 1957, 1968, and 1969.

Monitoring of Plutonium in Air near Rocky Flats by the Health and Safety Laboratory

The HASL began monitoring air near Rocky Flats in June 1970. This was after the 903 Area asphalt pad was in place, but adjacent contaminated soil areas remained (see Chapter VIII). The HASL's first Rocky Flats station was on the east security fence downwind of the 903 Area (Figure III-8). A memo located in the DOE archives indicated that this HASL sampler was near Dow's sampling station S-8. Other sources (e.g., DOE 1980 and Barker 1981) confirm this general location. Feely et al. (1985) describes the location of the Rocky Flats monitoring sites as follows:

- Rocky Flats, Colorado No. 1 was located at the eastern margin of the security fence, about 2300 m west of Indiana Street and 100 m south of the eastern entrance to the plant. The sampler was between the security fence and a dirt road regularly patrolled by plant security personnel.
- Rocky Flats, Colorado No. 2 was located approximately 15 m west of the eastern boundary of the RFP at Indiana Street.
- Rocky Flats, Colorado No. 3 was located approximately 6.5 km west of the RFP.
- Rocky Flats, Colorado No. 4 was located at the eastern margin of the old cattle fence, about 1800 m west of Indiana Street. This station was adjacent to a dirt road regularly patrolled by security personnel.

Station #2 was installed in July 1972, and the upwind station #3 (also called Coal Creek) was installed in October 1972. In May 1974, the HASL discontinued the Coal Creek station and added another, #4, which was east of the security fence at the original perimeter (cattle) fence (Figure III-8). Inspection of stations #1 and #4 in 1981 by EML staff showed that the samplers were subject to gross contamination by dust from the dirt roads (Feely et al. 1985). The samplers were

at ground level[1] and only a meter or so away from the dirt roads. The wooden louvered shelters did not prevent dust raised by passing vehicles from settling on the samplers and on the exposed filters. It seemed likely that a large part of the aerosol being collected was resuspended coarse dust, not necessarily representative of the aerosol that would have been collected if the sampler had been sitting on a metal stand, as were most of the HASL sites in the Surface Air Sampling Program.

Feely (1978) responded to questions from Rockwell (the Rocky Flats contractor) about the air sampling method used for their Rocky Flats sampling. The samplers used by EML at Rocky Flats were Roots rotary lobe blowers, AF size 24. The filters were Microsorban polystyrene, which were effectively 100% efficient for collecting small particles at the air velocities (~40 cm s^{-1}) used (Lockhart et al. 1964). Flow rates were calculated from the observed pressure drop across the filter and ambient temperature. Adjustments were made to air volumes at standard temperature and pressure, which was standard practice for the HASL surface air monitoring program.

Although not part of HASL's permanent monitoring program for as long as most stations, this sampling near Rocky Flats continued for a relatively long period of time. Station #1 was monitored continuously for about 11 years, and the record at station #4 continued for 7 years. The Rocky Flats contractor monitoring program for plutonium in air was just beginning to get underway, whereas the HASL had been monitoring plutonium in air for the previous 5 years. The HASL data provide an important cross-check of the contractor's results at station S-8 and S-32 (at Indiana Street). They are also used to evaluate what proportion of total long-lived alpha activity in air at the eastern security fence was due to plutonium (later in this chapter).

[1] See photo in Figure III-55. The filter was around 2–3 ft (0.6–0.9 m) above ground level.

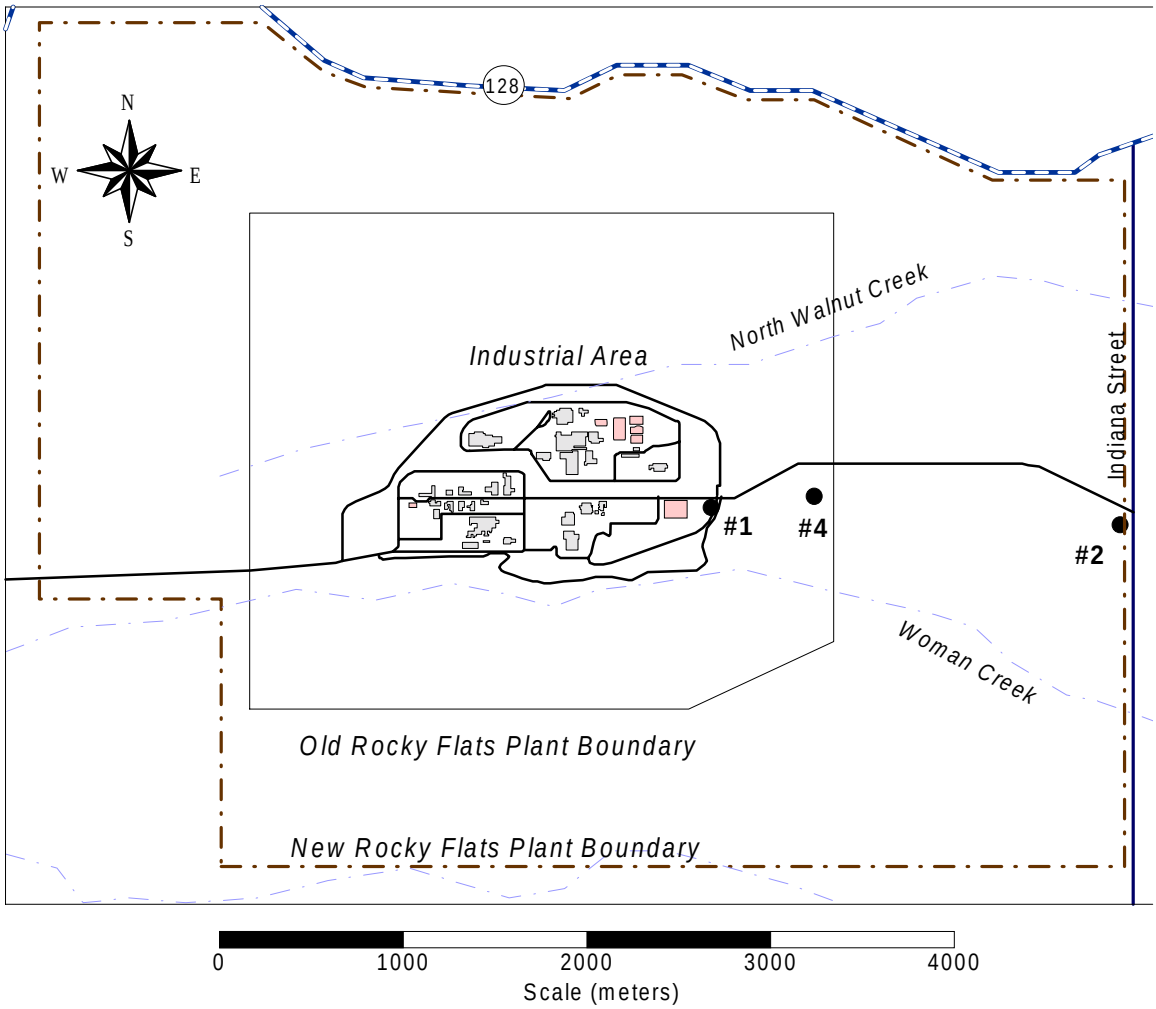

Figure III-8. Health and Safety Laboratory air sampling locations east of Rocky Flats.

The plutonium concentrations in air near Rocky Flats were reported by HASL/EML on a monthly resolution along with other data from the surface air monitoring program. Filters were changed weekly (Barker 1981) and composited monthly or quarterly. A complete set of Rocky Flats air monitoring data from HASL/EML is compiled in Feely et al. (1985) and is included in Appendix B, Table B-2. Only 3 of the 272 ^{239}Pu measurements made at the four Rocky Flats stations between June 1970 and September 1981 had an analytical error between 20 and 100%; the rest were <20%. However, there are other errors associated with air sampling (see the "Data Quality" section later in this chapter). Figure III-9 plots the data from the four Rocky Flats sites, as well as New York City, which is included for the global fallout perspective.

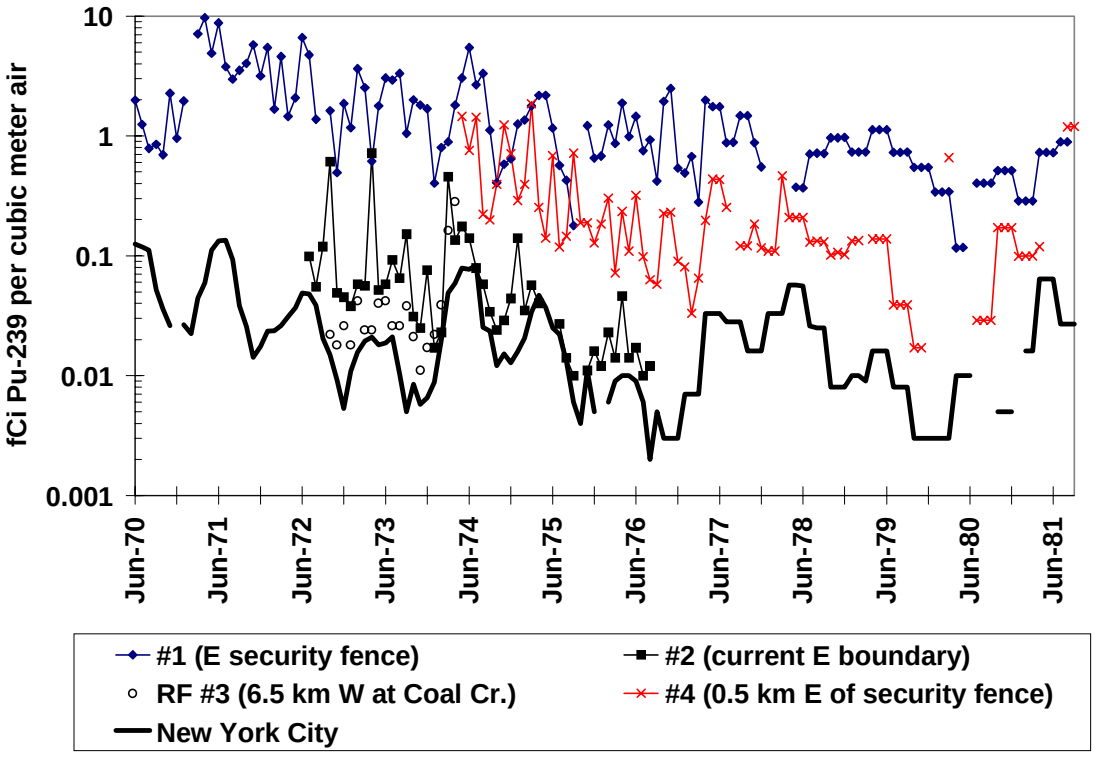

Figure III-9. Plutonium in air near Rocky Flats and in New York City measured by the Health and Safety Laboratory/Environmental Measurements Laboratory. The blocky appearance of the plot in the latter time periods is due to compositing and analyzing samples quarterly but still reporting the data monthly. (See Figure III-10 for smoothing of these data into semiannual averages.)

Figure III-9 presents the most detailed view of these monthly monitoring data. The spatial and temporal trends can be seen more clearly when the data are smoothed into semiannual average concentrations in Figure III-10. It is clear that plutonium near the Rocky Flats site is elevated compared to U.S. background. A downward trend over time at the station closest to the site is apparent. The concentration at this station (#1) in the first half of 1971 (6.5 fCi m^{-3}) is over 10 times that in the first half of 1981 (0.5 fCi m^{-3}), 10 years later. The HASL researchers theorized that contaminated soil material at RFP was becoming less available for resuspension with time (Volchok et al. 1977). In addition, the contaminated soil was being removed and stabilized during this interval.

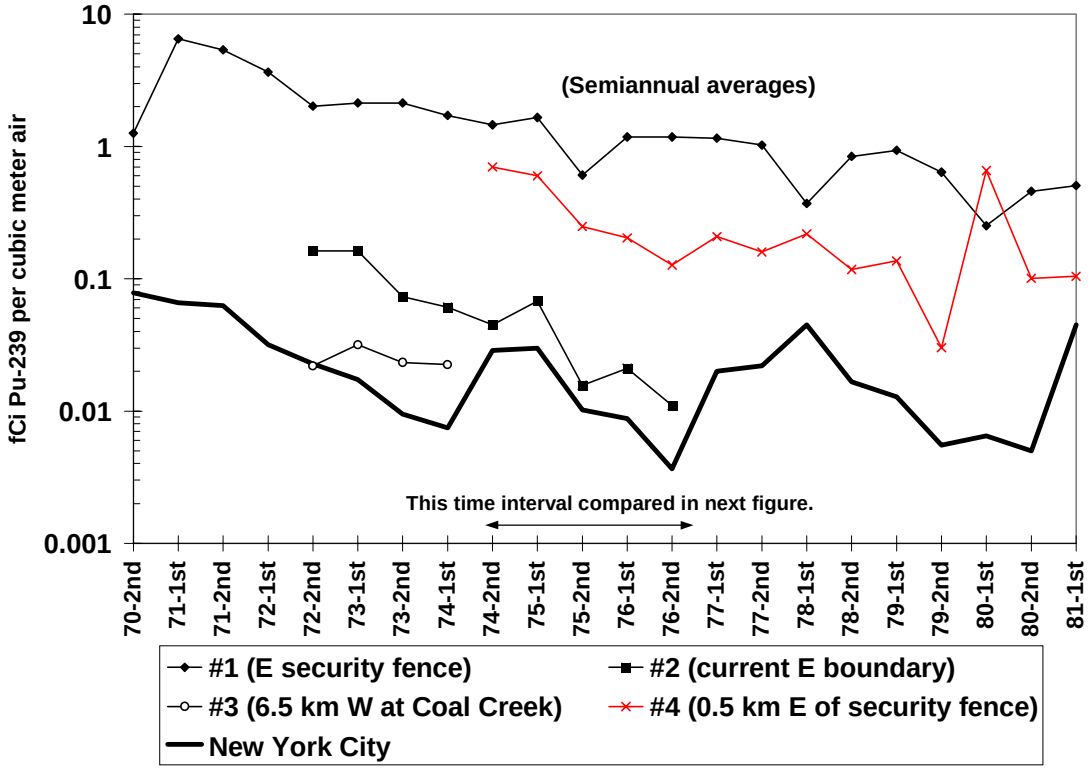

Figure III-10. Plutonium in air near Rocky Flats and in New York City, measured by the Health and Safety Laboratory. Monthly and quarterly measurements have been combined into semiannual averages.

There is a period of about 2 years (June 1974–August 1976) when plutonium in air from the three HASL locations to the east of Rocky Flats can be compared. As stressed earlier in this chapter, it is important to compare concentrations measured at the same time because of seasonal cycles and differences in the levels of background fallout. Figure III-11 shows the data for this period as a column chart, with the three Rocky Flats monitoring stations first, followed by the measurements at Helena, Montana, and New York City over the same time period. The rapid decrease in plutonium in air with distance from the site is clear. There is a decrease of about 3 times in the first 0.5 km east of the security fence and a further 10-fold decrease in the next 1.8 km to Indiana Street. Combined, this makes a 30-fold decrease in the concentration in a 2.3-km distance east of the security fence. The concentrations at Indiana Street in 1974–1976 are roughly twice what the HASL measured in air in New York City and Helena, Montana, during the same time period.

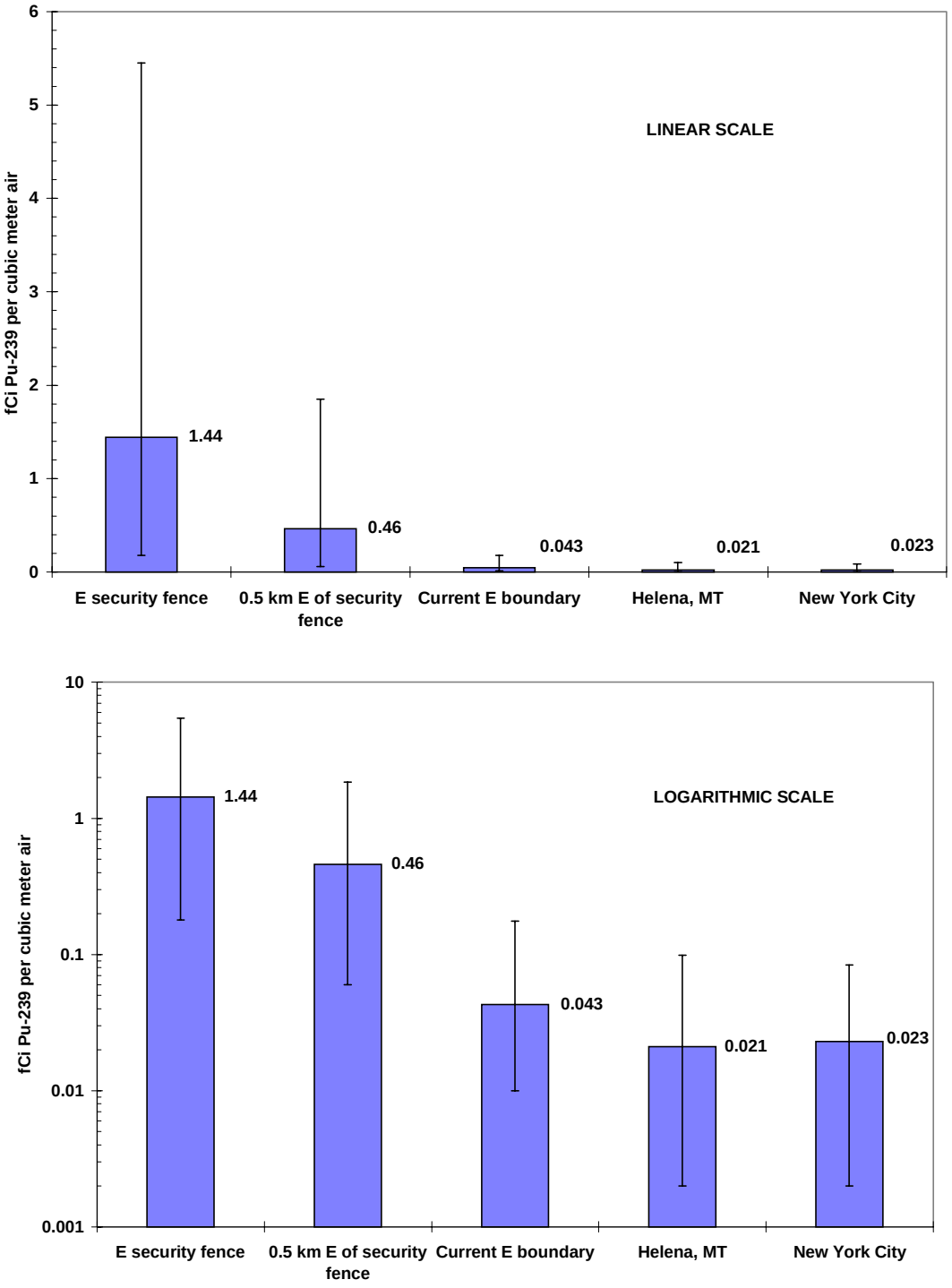

Figure III-11. Comparison of plutonium in air at three locations east of the Rocky Flats Plant; Helena, Montana; and New York City. Data plotted here are the average, minimum, and maximum of monthly measurements by the Health and Safety Laboratory over the time interval May 1974 through August 1976.

The plutonium concentration measured at New York City, the distant HASL location with a continuous record of monitoring, can be used as an estimate of fallout background, to obtain a net concentration for Rocky Flats plutonium. This was done monthly for data from 1970 through 1981, and the results were averaged to obtain a net annual average concentration of plutonium from Rocky Flats at the four HASL locations (Table III-1).

Table III-1. Annual Average Concentrations of Plutonium in Air at Four Locations around Rocky Flats, Measured by the Health and Safety Laboratory

	Net plutonium concentration[a] in air (fCi ^{239,240}Pu m^{-3})			
	Station #1	Station #4	Station #2	Station #3
Year	E security fence	0.5 km E of security fence	Current E boundary	6.5 km W of RFP at Coal Creek
1970	1.23	na [b]	na	na
1971	5.00	na	na	na
1972	2.88	na	0.14	0.01
1973	2.12	na	0.10	0.01
1974	1.72	0.76	0.06	0.09
1975	1.16	0.40	0.02	na
1976	1.22	0.16	0.01	na
1977	1.07	0.17	na	na
1978	0.70	0.14	na	na
1979	0.78	0.07	na	na
1980	0.34	0.29	na	na
1981	0.61	0.51	na	na

[a] Monthly concentration measured in New York City was subtracted from monthly concentration near Rocky Flats to obtain net concentration above fallout background.
[b] na = data not available.

To better understand the trend of concentrations with distance, the concentrations in Table III-1 are normalized (expressed in relative terms) in Table III-2. To normalize, concentrations at each station are divided by the concentration measured at the eastern security fence. This normalization allows us to relate these data to other data obtained from different media or times. For example, these relative concentrations compare favorably with those measured in lichens (slow-growing plants) discussed in Chapter IX. The average concentration of plutonium in lichens near Indiana Street is 0.01 (1%) of the concentration measured in lichens near the security fence.

Table III-2. Net Concentrations of Plutonium in Air Measured by the Health and Safety Laboratory at Three Locations near Rocky Flats, Relative to the Concentrations Measured at the Eastern Security Fence

	Station #4	Station #2	Station #3
Year	0.5 km E of security fence	Current E boundary	6.5 km W of RFP at Coal Creek
1972		0.049	0.004
1973		0.050	0.007
1974	0.44	0.036	0.054
1975	0.35	0.017	
1976	0.13	0.010	
1977	0.16		
1978	0.20		
1979	0.10		
1980	0.84		
1981	0.84		
All-year average	**0.38**	**0.032**	**0.022**

For risk comparison purposes, it is helpful to relate the concentrations of plutonium in air near Rocky Flats to the concentrations from fallout background. The net concentrations of plutonium in Table III-1 are divided by the fallout background concentrations in New York City and the ratio is shown in Table III-3. Concentrations at the eastern security fence range from 16 to 200 times fallout plutonium concentrations between 1970 and 1981 (average, 75 times background). Concentrations at Indiana Street are between 1 and 8 times background between 1972 and 1976 (average, 3.5 times background).

Table III-3. Net Concentrations of Rocky Flats Plutonium Relative to Fallout Background

	Station #1	Station #4	Station #2	Station #3
Year	E security fence	0.5 km E of security fence	Current E boundary	6.5 km W of RFP at Coal Creek
1970	16			
1971	83			
1972	110		5.1	0.4
1973	160		7.8	1.0
1974	44	20	1.6	2.4
1975	58	20	1.0	
1976	200	26	2.0	
1977	51	7.9		
1978	23	4.5		
1979	85	8.0		
1980	57	48		
1981	16	13		
Average	**75**	**19**	**3.5**	**1.3**

The HASL air monitoring data discussed thus far have been for samples collected monthly or quarterly. We located one exception to the monthly or quarterly compositing for HASL

monitoring of plutonium in air near Rocky Flats. In a letter to Rockwell, Volchok (1971) presents both ^{238}Pu and ^{239}Pu concentrations from the #1 sampler, in the second half of 1970, which are from samples generally collected weekly. The data are presented in Appendix B, Table B-3, and the ^{239}Pu concentrations are plotted in Figure III-12. The weekly maximum concentration was 4.7 times the average concentration (1.42 fCi m^{-3}) over the approximately 5-month interval.

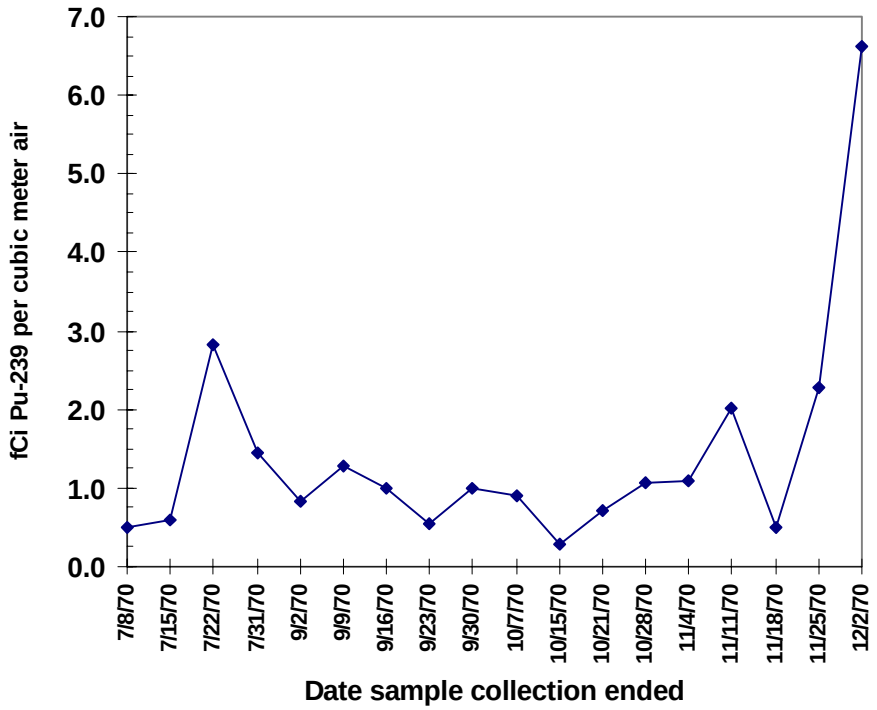

Figure III-12. Weekly concentrations of ^{239}Pu measured by the Health and Safety Laboratory at station #1 at the security fence east of the 903 Area during the second half of 1970.

Monitoring of Plutonium in Air near Rocky Flats by the Colorado Department of Health

The Colorado Department of Health (CDH), currently called the Colorado Department of Public Health and Environment, began monitoring the air around Rocky Flats in 1969. The CDH has been reporting results of plutonium in ambient air around the RFP since July 1970 in monthly reports originally titled *U.S. AEC Rocky Flats Surveillance*. *RAC* obtained a complete set of these reports through 1991. The CDH has maintained onsite continuous air sampling stations as well as a regional air sampling system (15 stations in 1977), which provide additional surveillance in the vicinity of Rocky Flats and in the metropolitan Denver area. The CDH also has monitoring locations in Colorado that are remote from the RFP (such as Durango and Colorado National Monument).

Some of the CDH monitoring locations have samplers for both TSP, or total suspended particulate, and PM-10, or particulate matter less than 10 micrometers (μm). These samplers can help separate the respirable material from the total aerosol, which also contains larger particles.

The first CDH monitoring station for plutonium in air near Rocky Flats was station APC-56 on the east security fence at Central Avenue. Monitoring data from this station are available starting in January 1969. Shortly after, four other stations were added along the eastern security fence, numbered D-1 through D-4. D-1 and D-2 were north and D-3 and D-4 were south of Central Avenue. A physical description or map of the "D" locations on the RFP perimeter is not given in the early monthly CDH reports. The first map of the onsite and offsite locations is in the April 1975 monthly report (CDH 1975). This map is reproduced in Appendix B as Figure B-7. Four of the five sampler locations are also shown on Figure III-13.

Like the HASL monitoring near RFP, the CDH air monitoring record can provide an independent confirmation of contractor onsite monitoring data in the post-1970 period.

The CDH D-x samplers are housed in a General Metal Works aluminum shelter with 8×10-in. glass microfiber filters, flow control, and timers. Quality assurance results are presented in the monthly reports from the CDH. The Air Pollution Control (APC) filters were normally operated for one 24-hour period every 4 days, resulting in a total volume of air sampled of about 15,000 to 20,000 m^3 $month^{-1}$ at each station. The D-x station filters normally operated continuously, with filters being changed every other day. Total volumes sampled at each of the D-x stations were around 60,000 to 80,000 m^3 $month^{-1}$ (CDH 1972a). The minimum detectable concentration for $^{239,240}Pu$ in air in the 1970s, based on monthly compositing of the air filters, was 0.03 and 0.08 fCi m^{-3} for the D-x stations and for the APC stations, respectively (CDH 1971, 1980). This minimum detectable concentration for the D-x stations is about 10 times poorer than that obtained from the HASL air sampling, and is in the range of background fallout plutonium concentrations at this time. Data from the D-5 station were used, along with HASL and Dow contractor monitoring along Indiana Street, to validate airborne releases of plutonium from Rocky Flats in the 1970s (Rood and Grogan 1999c).

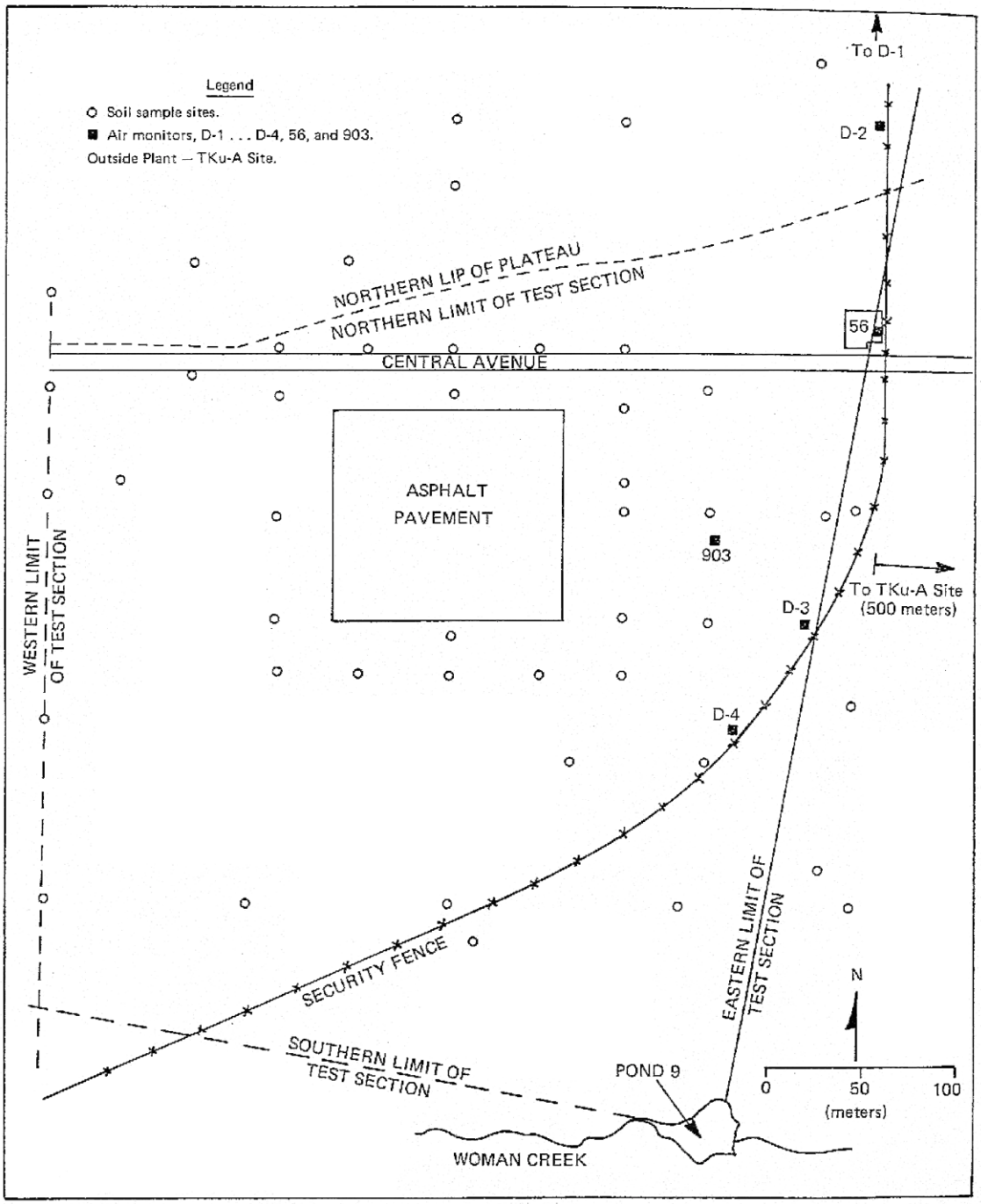

Figure III-13. Map of eastern part of Rocky Flats industrial area, showing locations of Colorado Department of Health air monitoring stations D-1 through D-5. A physical description of all the Colorado Department of Health air sampler locations near Rocky Flats is given in Table III-4. This map was obtained from Michels (1973) and shows features relevant to that study.

Plutonium in air results for station D-5 on the southeastern boundary of the RFP at Indiana Street were reported beginning in June 1970 and for stations D-6 through D-8 in July 1979. These locations are shown in Figure B-8 of Appendix B. Airborne releases of plutonium from Rocky Flats as late as 1979 were insignificant in the context of historical public exposures. Therefore, data from stations D-6 through D-8 were not examined further. Data from stations D-1 through D-5 and APC-56 are presented in this report. The data for 1969 through 1980 are included as Table B-4 in Appendix B. They provide a set of measurements that can validate the impact of contaminated soil remaining in the 903 Area after placement of the asphalt pad. In this respect, they complement the air monitoring conducted by the RFP contractor and by the HASL.

Annual average concentrations of ^{239,240}Pu at five locations at the east security fence and one location on the southeastern boundary (D-5) of the RFP for 1969–1980 are illustrated in Figure III-14. The annual average data for this plot were generated from the tabular summary of monthly results obtained from the Colorado Department of Health (Terry 1992b), which is included as Table B-4 of Appendix B. We double-checked those data against values presented in monthly reports.

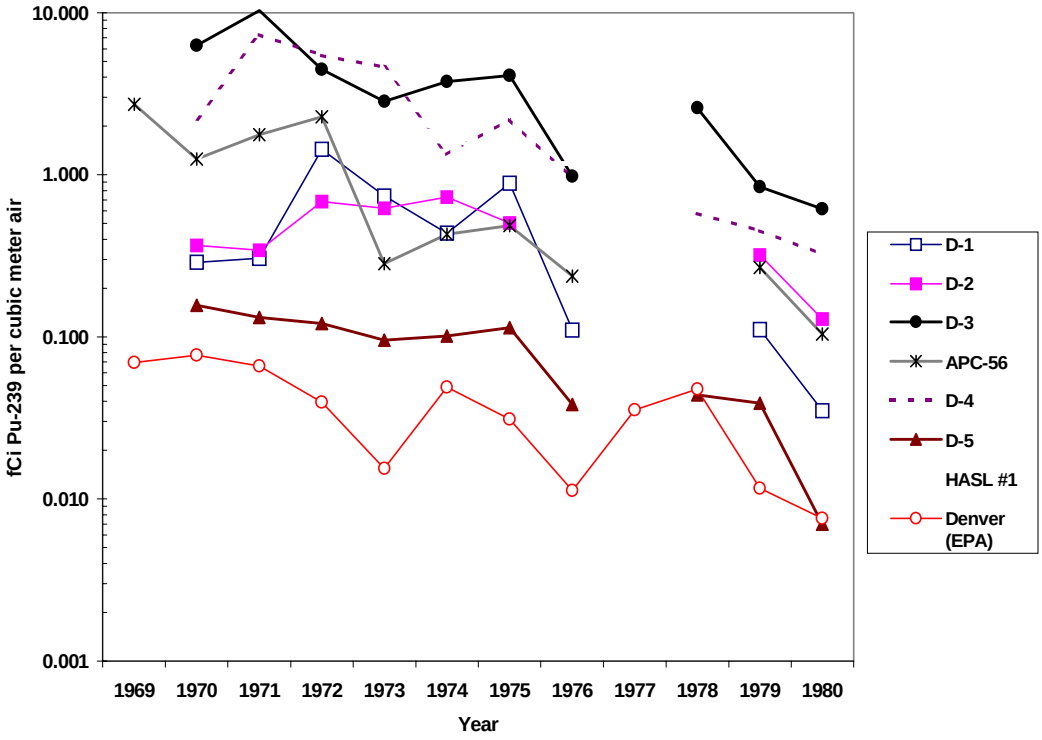

Figure III-14. Annual average concentrations of ^{239,240}Pu at five locations at the east security fence and one location on the southeastern boundary (D-5) of the Rocky Flats Plant, 1969–1980, as measured by the Colorado Department of Health. The Health and Safety Laboratory (HASL) station #1 is included to illustrate that concentrations measured by that agency in a similar location fall in line with the onsite measurements of the Colorado Department of Health. The concentrations in Denver are included as a fallout background comparison.

The amount of plutonium in air was considerably higher at stations D-3 and D-4 than the other locations. These samplers were closest to the contaminated soil areas remaining after the 903 Pad was placed. These contaminated soil areas are discussed in Chapter VIII. Over the same period of time, plutonium concentrations in air at the other CDH samplers were generally about 10% of those measured at D-3. The other three onsite samplers were approximately 200 m to 400 m from the contaminated soil areas. These data illustrate a steep concentration gradient (drop-off) with distance from contaminated areas. The net concentrations (above fallout background) at station D-5 on the current southeastern boundary at Indiana Street averaged 1.6% of that at station D-3 during the 1970–1980. This can be compared with the 3.2% measured at HASL station #2 at Indiana Street during 1972–1976 relative to their #1 station near the 903 Area.

The CDH air sampler D-3 was closest to the HASL sampler #1 (DOE 1980). Monthly average concentrations of plutonium in air from the two programs over a 3-1/4-year period are compared in Figure III-15. Because the HASL reported concentrations of radionuclides in air using standard temperature and pressure air volumes and the CDH did not (until around 1977, Terry 1992b), we corrected the CDH concentrations[2] to standard temperature and pressure conditions for the comparison shown in Figure III-15. Average monthly temperature and pressure data for the Rocky Flats site were obtained from EG&G (1995).

In general, the CDH sampler results varied more widely from month to month than the HASL sampler. There was no HASL measurement in September 1972 when the CDH reported a rather large increase, but other CDH peaks in August 1974 and February 1975 were not observed by the HASL sampler (Figure III-15). The CDH concentrations were higher by about a factor of 1.8 (median ratio). Fifty-nine percent of the measurements made by the HASL and the CDH agree within a factor of 2. This comparison roughly indicates the uncertainty in determining plutonium in ambient air at this place and time.

The part of the RFP industrial zone near the 903 Area contaminated soil is probably one of the more challenging locations for accurate air sampling. As discussed above, the concentration gradient appeared to be a decrease of a factor of 10 within a few hundred meters from contaminated areas. The CDH and HASL samplers could have been separated by as much as 100 m. At these onsite locations, plutonium is probably associated with a wider range of particle sizes than locations further downwind. The sampler housings, flow rates, filters, etc., from these two programs are completely different, and collection efficiency of larger particles by air samplers is strongly affected by sampler housing design, orientation, and other factors.

At the time this project began, the CDH did not have an electronic database of historical air monitoring results, but we were able to obtain a database for stations D-1 through D-8 late in the project (Love 1999). The database extended back to 1971 whereas our hard copy tables started in 1969. Some annual averages generated from this database differed from those generated from the tabulations obtained earlier. It appeared that some database values could have been quarterly averages or that some data had been omitted, because there were fewer results for a given year in the electronic spreadsheet. It was determined to be unimportant for our project to reconcile these differences. Data from the onsite stations were not used quantitatively. Annual average concentrations from boundary station D-5 were compared to predicted concentrations from Rocky Flats airborne releases (Rood and Grogan 1999c). The annual averages used for this

[2] On average, this correction resulted in multiplying CDH values by 1.29. The monthly minimum correction was 1.24 for January, and the maximum was 1.33 for July and August.

purpose were those obtained from the hard copies, which had been checked against the monthly reports. The uncertainty in the predicted concentrations is so large that a small difference in the measured concentration would not affect the qualitative conclusions drawn from this comparison.

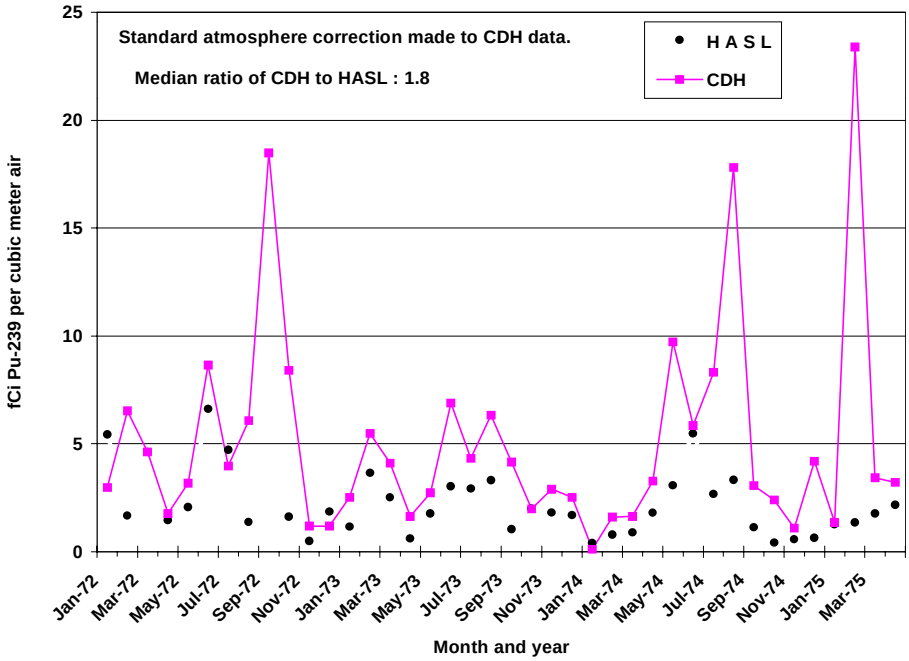

Figure III-15. Comparison of ^{239}Pu measurements made by the Colorado Department of Health at Station D-3 and the Health and Safety Laboratory at Station #1 at the east security fence of the Rocky Flats Plant. These samplers may have been up to 100 m away from each other. Both are on the eastern perimeter fence, generally east to southeast of the 903 Area.

The CDH monitoring of plutonium in air at offsite locations began in 1969 at Golden and Longmont. Monitoring at Arvada and Boulder began in 1970. In 1972, monitoring began at remote locations (Durango, Ft. Collins, Walsenburg, and Rangely). Concentrations of plutonium in air at CDH offsite stations in the early 1970s were very close to the minimum detectable concentrations at that time. In addition, the minimum detectable concentration for the APC stations used offsite was essentially at the concentration expected from global fallout at that time. Table III-4 shows the annual averages for onsite and offsite stations through 1974. Because of the limited sensitivity of the methods used, these data are not useful for quantitative validation of airborne releases from Rocky Flats. They do provide confirmation of the small influence of Rocky Flats releases on plutonium in air at offsite locations. The CDH methods were quite adequate to detect the Rocky Flats plutonium in air samples collected along the eastern security fence of the RFP.

In May 1969, the CDH apparently made arrangements for the Public Health Service to analyze some of their air filters for plutonium, perhaps in connection with the fire that month. The results are presented in the July 1970 monthly report (CDH 1970). The isotopic ratio of ^{238}Pu/^{239,240}Pu for three offsite stations on May 12, 1969 (Arvada, Edgewater, and Golden) was 0.3, whereas for the onsite station APC-56, the ratio averaged 0.03 for 5 days that month. This demonstrated little contribution of Rocky Flats plutonium at those offsite locations (see next section).

Table III-4. Comparison of Plutonium-239,240 Concentrations (fCi m^{-3}) in Onsite and Offsite Air, Measured by the Colorado Department of Health[a]

Location	1969	1970	1971	1972	1973	1974
Onsite						
D-1		0.29	0.31	1.44	0.74	0.44
D-2		0.37	0.34	0.69	0.62	0.73
D-3		6.29	10.29	4.47	2.87	3.77
D-4		2.21	7.34	5.48	4.63	1.33
APC-56	2.73	1.25	1.77	2.28	0.28	0.43
Onsite average[b]	**2.73**	**2.08**	**4.01**	**2.87**	**1.83**	**1.34**
Offsite (metro):						
D-5 (SE boundary)		0.16	0.13	0.12	0.10	0.10
APC-2 Denver (EPA data) [c]	0.070	0.077	0.066	0.040	0.015	0.049
APC-15 Arvada		0.16	<0.08	0.06	<0.06	0.25
APC-16 Golden	0.08	<0.11	0.08	<0.06	<0.06	<0.08
APC-19 Boulder		0.13	<0.07	<0.05	<0.06	<0.06
APC-22 Longmont	0.08	0.12	0.07	<0.02	<0.05	<0.1
Offsite (metro) average[b]	**0.08**	**0.13**	**0.08**	**0.06**	**0.06**	**0.12**
Offsite (remote):						
APC-29 Durango				<0.12	<0.02	<0.05
APC-42 Fort Collins				<0.04	<0.08	0.14
APC-81 Walsenburg				0.07	<0.06	0.38
APC-108 Rangely				0.1	<0.08	0.13
Offsite (remote) average[b]				**0.08**	**0.06**	**0.18**

[a] Source: Terry 1992a, 1992b, 1992c.
[b] Averages in this table are arithmetic averages of the annual averages from each station.
[c] Lower minimum detectable concentration for EPA data (0.001 compared to 0.08 for APC-x stations and 0.03 for D-x stations.

Plutonium Isotope Ratios from Independent Routine Air Monitoring Programs

Of the plutonium in global fallout, the primary constituents are ^{239}Pu and ^{240}Pu, which are indistinguishable by routine radioanalytical techniques. The data trends illustrated in this chapter have all been concentrations of ^{239,240}Pu or total plutonium, which includes ^{238}Pu. A secondary source of ^{238}Pu was the accidental burnup of a satellite in the upper atmosphere in April 1964. Plutonium-238 from the burnup of the SNAP-9A satellite was first detected by the HASL in ground-level air in Ispra, Italy in early 1966. In New York City, the elevated levels were first detected in April of that year. After 1970, HASL stopped reporting ^{238}Pu at many stations, because concentrations were barely detectable and soil sampling indicated that 95% of the SNAP-9A ^{238}Pu had deposited by that time. Because of the low levels of ^{238}Pu in the air after October 1971, the errors associated with the measurements are large (Figure III-16).

The ^{238}Pu/^{239,240}Pu activity ratio can give a rough indication of the source of plutonium. For very precise fingerprinting of plutonium sources, separation of the ^{239}Pu isotope from ^{240}Pu is necessary, which requires sophisticated measurement methods that are not routinely used. The HASL measured ^{238}Pu, as well as ^{239,240}Pu, at the locations around Rocky Flats for a little more than 3 years (Table III-5). At the east security fence, where essentially all the plutonium in air was from Rocky Flats, there was relatively more ^{239,240}Pu than the globally dispersed material

(indicated by New York City), and the range of the ratio was small. This observation was also made by Volchok (1971) for *weekly* air filters collected at the HASL #1 site in 1970. Although the weekly ^{239,240}Pu concentrations varied by over a factor of 20, the ratios of ^{238}Pu/^{239,240}Pu were reasonably consistent, ranging from 0.018 to 0.037 (a factor of 2) and averaging 0.026 (Table B-3).

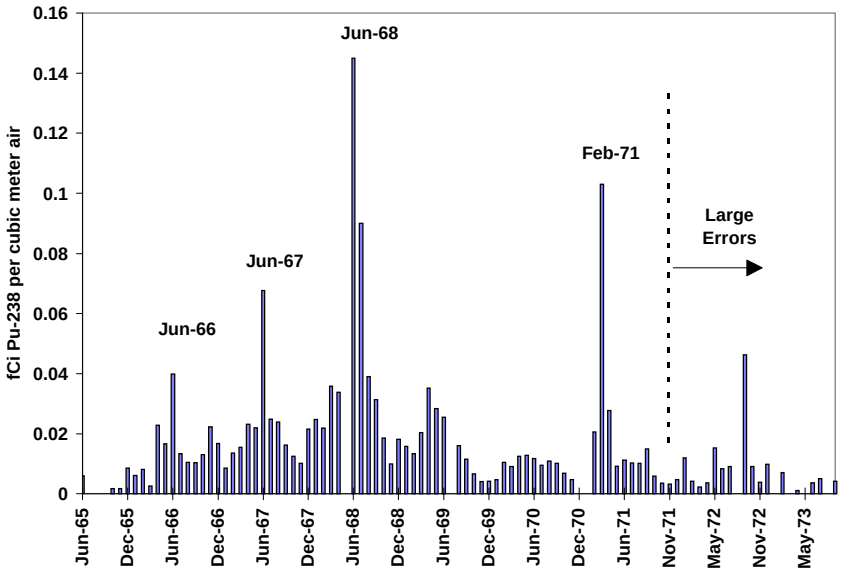

Figure III-16. Plutonium-238 in air in New York City (Health and Safety Laboratory data).

At Indiana Street, the range of ratios of monthly measurements (Table III-5) indicates a mixture of Rocky Flats and fallout plutonium, which is in agreement with the ^{239,240}Pu concentration data presented earlier. The same ratios can be obtained from the PHS/EPA monitoring data. The ratios measured in Denver are similar to the other U.S. locations, indicating that plutonium in Denver is predominantly fallout, according to this method.

Table III-5. Plutonium-238/Plutonium-239,240 Activity Ratios Measured in Air

Location	Agency[a]	Years	Average ratio	Range in ratio
New York City	HASL	1970–1973	0.2	0.06–5
Pierre, S.D.	PHS/EPA	1967–1974	0.15	0.05–0.75
Denver	PHS/EPA	1967–1974	0.14	0.03–1.5
Maryland	PHS/EPA	1967–1974	0.10	0.03–1.1
RFP boundary at Indiana Street	HASL	1970–1973	0.04	0.01–0.1
RF east security fence	HASL	1970–1973	0.02	0.02–0.03

[a] HASL=Health and Safety Laboratory; PHS/EPA=Public Health Service/Environmental Protection Agency.

ROUTINE MONITORING OF ALPHA ACTIVITY IN AIR BY ROCKY FLATS CONTRACTOR (1950S AND 1960S)

Before 1970, air samples collected by the RFP contractor (Dow Chemical Co.) were routinely analyzed for total long-lived alpha (TTLa) activity, not for specific alpha-emitting radionuclides. A gross beta analysis was also performed. Based on our framework exercise for evaluating the usefulness of historic environmental data (Chapter I), the pre-1970 alpha data were believed to have potential for evaluating spatial and temporal trends for RFP-released materials. Beta activities were highly influenced by weapons testing, and consequently were not as useful as alpha counts for evaluating Rocky Flats materials. Meyer et al. (1996) emphasized examining daily onsite air monitoring data, which were located in records storage in Denver, to formulate models for initial suspension and resuspension of alpha contamination from the 903 barrel storage area. This was completed by Weber et al. (1999).

Sources of Information for Routine Contractor Air Monitoring Data

Table III-6 summarizes the document types we obtained that contain routine air monitoring data collected by the RFP contractor. Original daily data sheets have been obtained for the time period of greatest interest for releases from the 903 Area and the 1969 fire (shaded area in Table III-6). Communications with the Kirkland and Ellis law firm, which is representing Dow Chemical Company, turned up another original document type, air sample record cards, for onsite and offsite samplers for 1952 through 1956. These cards are also available for offsite sampling locations only in 1957. Because of our interest in the dispersion of releases from the 1957 fire, we obtained copies of the 1957 cards. There were several types of monthly reports (document type 3) that contained summary air monitoring information.

Onsite Monitoring for Alpha Activity in Air

The RFP contractor began onsite ambient air monitoring at a single station in 1952. By early 1953, 10 onsite stations had been established. The July 1953 monthly site survey report states that calibration of samplers to a flow rate of 2 cfm (cubic feet per minute) was underway. In February 1954, Whatman 41 filter paper[3] was substituted for HV70 paper[4] on all *offsite* samplers, because the HV70 filters ruptured in the week-long sampling duration used offsite. This statement implies that HV70 filters continued to be used for onsite (daily) sampling. In Phase I, ChemRisk also came to the conclusion that ambient air samplers during the 1957 fire probably used HV70 filters or Whatman-41 cellulose filters [both are 47 mm (about 2-in.) in diameter] (Mongan et al. 1996a). Photographs of stations used for offsite air monitoring are included in the data quality section of this chapter. In March 1956, manometers were installed on onsite units. In 1969, two more stations were added to make a total of 12 stations (Figure III-17).

[3] Whatman-41 cellulose filter media. Whatman Paper Ltd., Clifton, NJ 07014.

[4] HV70 cellulose-asbestos filter media. Hollingsworth and Vose Company, East Wadpole, MA 02032.

Table III-6. Document Types in *RAC* Possession that Contain Air Monitoring Data from Routine RFP Contractor Monitoring Programs[a]

	Onsite									Offsite							
1952	b		3							b							
1953	b		3							b		3					
1954	b		3							b		3					
1955	b		3							b							
1956	b		3							b							
1957			3							1							
1958			3														
1959			3														
1960			3									3				7	
1961			3									3			6	7	
1962			3									3			6	7	
1963			3									3			6	7	
1964	1	2	3								2	3			6	7	
1965	1	2	3								2	3			6	7	
1966	1		3							1		3			6	7	
1967	1		3									3			6	7	
1968	1	2	3							1	2	3			6	7	
1969	1	2	3							1	2	3			6	7	
1970	1	2	3	4						1	2	3	4				
1971	1	2	3	4				8		1	2	3	4				8
1972		2	3	4				8			2	3	4				8
1973	1			4	5			8		1			4	5			8
1974				4				8					4				8
1975				4				8					4				8
1976				4				8					4				8
1977				4				8					4				8
1978				4				8					4				8
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1984				4				8					4				8
1985				4				8					4				8
1986				4				8					4				8
1987				4				8					4				8
1988				4				8					4				8
1989				4				8					4				8
1990				4				8					4				8

Table III-6. (continued).

[a] Key to document types in Table III-6:

1. Site Survey Routine Air Sample Results for onsite or offsite samples. Handwritten on standard form. One page per sample collection date. Each sampler's data listed separately. Date on, date off, time and volume, gross, background, and net counts for 4-hour and 1-week decay times. Same type of information on Site Survey Air Sample Record Cards (1957 offsite).
2. Result and Calculation Sheet for onsite or offsite samples. Handwritten on standard form. 4-hour and 1-week count data for each sample. Includes contractor's computation of monthly average concentration.
3. Monthly progress reports. Organizations producing the monthly reports that contain the air monitoring data vary over time, but they include the Site Survey group from the early 1950s through mid-1960s and various Health Physics reports. Data from each sampler location are not given. Sometimes there is no tabular information, only plots. For most of 1955 and for 1956–1960, only 4-hour delayed alpha counts were reported (no long-lived alpha).
4. Monthly Environmental Monitoring Report (titles vary). Data for each sampler are given separately.
5. Computer printouts – summary statistics.
6. Public Health Service monthly summaries of Dow monitoring data. Data from all offsite stations are combined.
7. Environmental Survey Reports. Typically quarterly or semiannual reporting of monthly results. Offsite stations only. Average, maximum, minimum. Data from all stations combined until 1969 reports.
8. Annual Environmental Monitoring Report.

[b] Kirkland and Ellis has air sample record cards for these years.

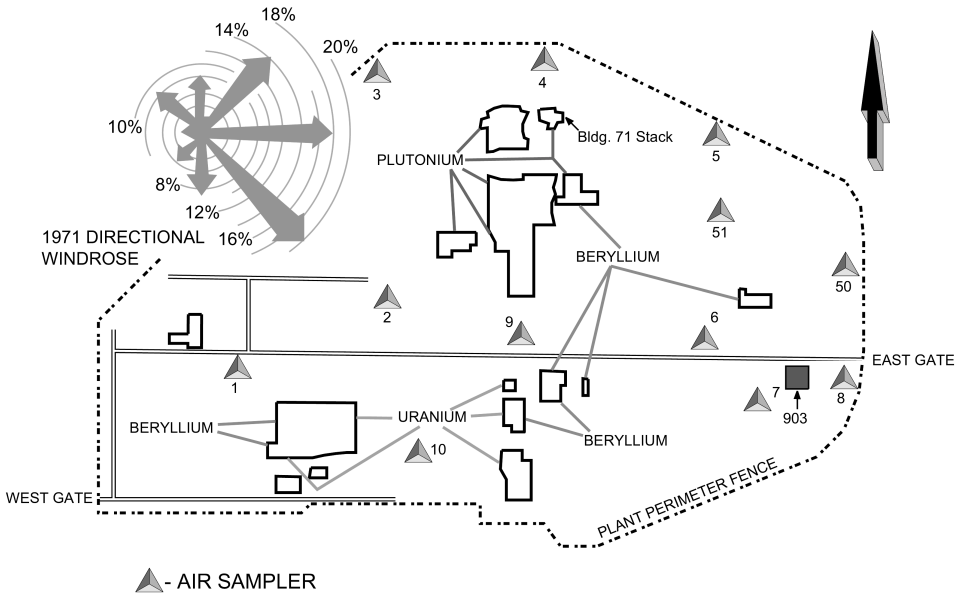


Figure III-17. Location and numbers of early onsite air samplers (adapted from a figure in the 1971 annual environmental report). The geographic coordinates of the samplers are given in Table I-3 of ChemRisk (1994b). Samplers were renumbered in the mid-1970s (Appendix B). The distance from the west to east across the industrial zone of the Rocky Flats Plant is roughly 1 mile. The 903 barrel storage area, between samplers S-8 and S-7, was a primary source of plutonium releases. Building 71 was a major release point for routine releases of plutonium and was the site of a major fire in September 1957. Areas of the industrial zone in which beryllium and uranium were processed are noted.

Early Monitoring Data: Evaluation of Utility of 4-hour Delayed Alpha Counts (1950s)

For most of 1955 and for 1956-1960, only "4-hour alpha counts" were reported in the site survey monthly reports. The 4-hour count, as the name implies, was made about 4 hours after sample collection ended and includes a large contribution from natural alpha-emitting radionuclides like decay products of radon. There were no data given for long-lived alpha activity, which is more indicative of Rocky Flats materials like plutonium, americium, and uranium. Typically the 4-hour count data were presented as a time-trend plot, with no numerical information given in tables. We needed to address the question, "Do the 4-hour alpha count data contain information of any use to the historical public exposures studies?"

To answer this question, the relationship between the 4-hour and 1-week counts was examined. The data were drawn from an existing electronic database of air monitoring results compiled by ChemRisk in Phase I of this study. This database contains the 4-hour and 1-week net alpha counts for several onsite locations in the 1960s. Two time periods were examined: 1968–1969, when the alpha concentrations were high, and 1964–5, when the concentrations were lower. The relationship between the two counts is illustrated in Figures III-18 and III-19, and is explained in the figure captions.

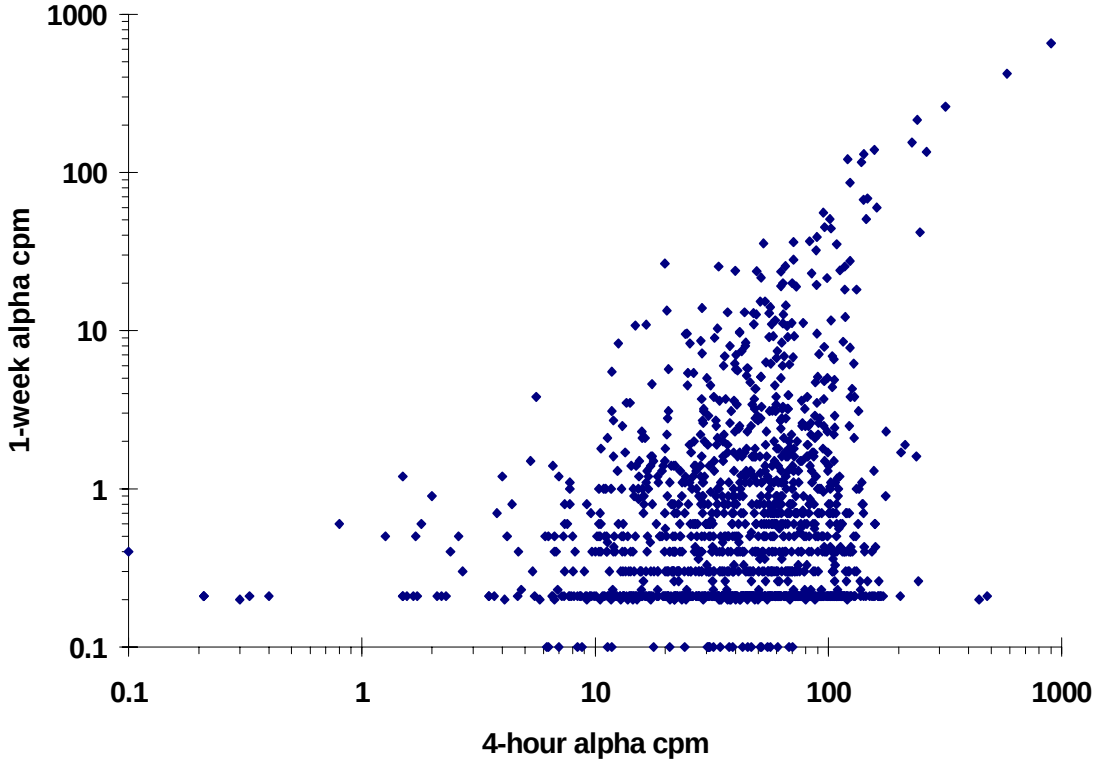


Figure III-18. Relationship between 4-hour alpha count and 1-week alpha count for daily onsite air samples collected in 1968 and 1969. There were a total of 1,515 pairs of data. The 4-hour count ranged from 0.1 to 902 cpm and the 1-week count ranged from 0 to 655 cpm. Only for very high alpha counts, where long-lived alpha emitters comprise the majority of the activity, is there a significant correlation between the two values. For the set of data where 4-hour counts are >100 cpm, the correlation coefficient is 0.79. For the set of data where 4-hour counts are <100 cpm, the correlation coefficient is only 0.18. For the lower count rates, the activity concentrations are dominated by shorter-lived radionuclides such as natural uranium and thorium decay products.

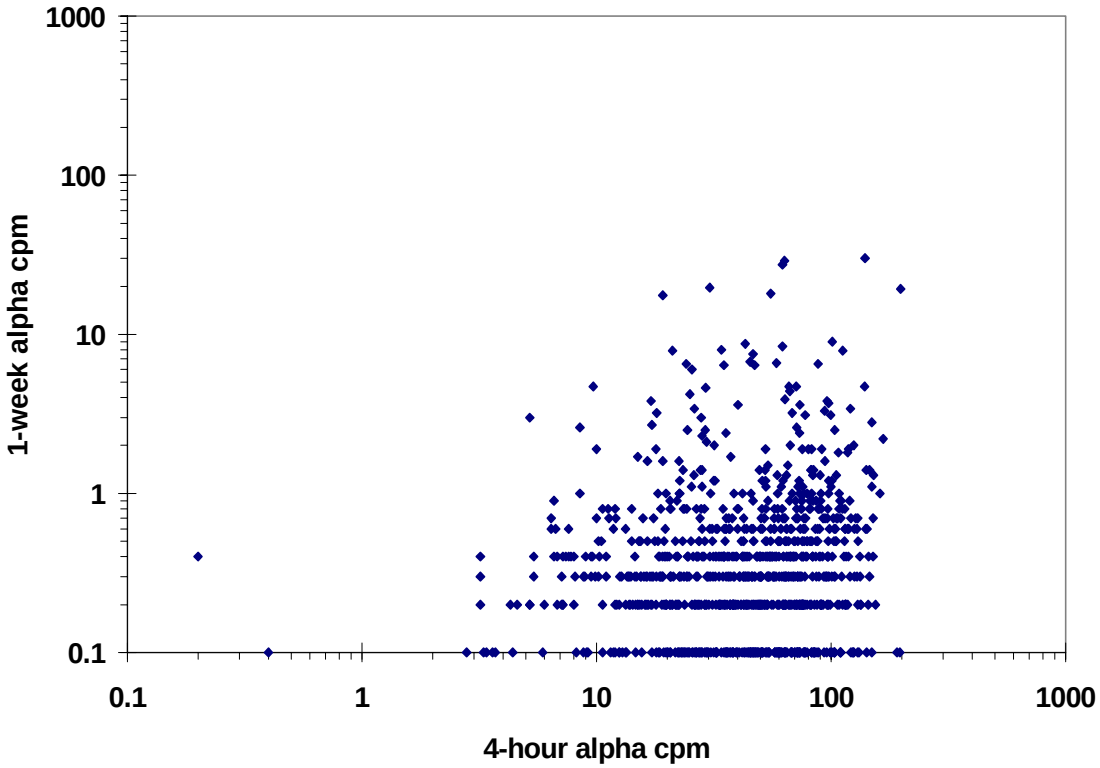


Figure III-19. Relationship between 4-hour alpha count and 1-week alpha count for daily onsite air samples collected in 1964 and 1965. There were a total of 1,492 pairs of data. The 4-hour count ranged from 0.2 to 244 cpm and the 1-week count ranged from 0 to 30 cpm. There is no significant correlation between the two values (r = 0.09). The 4-hour activity concentrations are dominated by shorter-lived radionuclides such as natural uranium and thorium decay products. The long-lived radionuclides are not present in high enough levels to affect the 4-hour counts.

Our conclusion is that the 4-hour count results are of no value in assessing the concentrations of long-lived alpha-emitters released from Rocky Flats unless those counts are very high (greater than several hundred cpm), which was not the case in the 1950s. Therefore, no effort was expended to extract the 4-hour count data from the plots in the monthly site survey reports from the 1950s.

Air Monitoring Associated with the September 11, 1957 Fire

During the 1957 fire, eight onsite samplers (S-1 through S-8) and one offsite sampler located at the Wagner school (≈3.3 mi southeast of the plant) were operating (ChemRisk 1994b). In addition to these routine air samplers, portable samplers were deployed at 13 onsite locations during the fire. Sampling results were reported as the percent of the maximum permissible level (MPL) of 9 disintegrations per minute (dpm). The routine air monitoring equipment were low volume air samplers that operated at a flow rate of 2 cfm. Portable high-volume samplers were maintained for events like the fire and operated at a flow rate of 12 to 40 cfm. Onsite sampler

filters were typically changed daily at 8:15 a.m. However, following the fire on the evening of September 11th, onsite sampler filters were changed at 3:30 a.m. and again at 3:30 p.m. of the following day (September 12th). Filters of offsite samplers were reported to be changed biweekly, but it was unclear whether this meant twice a week or once every two weeks (ChemRisk 1994b). The filter on the Wagner school sampler that would have captured releases from the fire was installed on September 10 at 11:15 a.m. This filter was changed at 2:15 a.m. on September 12th and again sometime on September 13th; however, the exact time of replacement was not reported.

A review of the air monitoring data revealed inconsistencies between activity detected and the timing of the release. For example, the highest air concentrations from the portable sampling network appear to have occurred the morning of September 12th at around 6:00 a.m., well after the fire had been put out and the major release had occurred. The onsite samplers (S-1 through S-8) reported concentrations of 0% of the MPL for the period 8:15 a.m. September 11 to 3:30 a.m. September 12. After that, an average of the onsite samplers operating from 3:30 a.m. to 3:30 p.m. September 12 reported a concentration of 0.28% of the MPL (ChemRisk 1994b). It is postulated that during the main release from fire (10:15 to 10:45 p.m.), the plume was lofted over onsite locations resulting in low ground-level concentrations. It is not clear why measurable activity was detected in the samplers for the later measurement period. Similar discrepancies were noted for the Wagner School air sampler. A concentration of 0% MPL was reported for the time period September 10, 11:15 a.m. to September 12, 2:15 a.m. The second filter that was removed sometime on September 13 reported a concentration of 0.56% MPL.

Air monitoring data collected during the 1957 fire as reported by ChemRisk (1994b) were carefully reviewed by Rood and Grogan (1999a) for applicability towards model validation of predicted air concentrations from the fire. They also considered the data quality issues discussed in this Task 4 report. Based on these data quality issues and discrepancies between the timing of the event and the measurements, they concluded that the air monitoring data were of little use in validating model-predicted air concentrations for the 1957 fire.

Sampling, Counting, and Data Reporting for Daily Onsite Air Samples (1960s)

Beginning in January 1960, a 1-week count and 4-hour counts of alpha and beta activity were presented in tables in the monthly reports. The data were reported as one onsite group, with the maximum and minimum individual count value. Because results from individual samplers cannot be obtained, this monthly summary information is severely limited for use in the dose reconstruction.

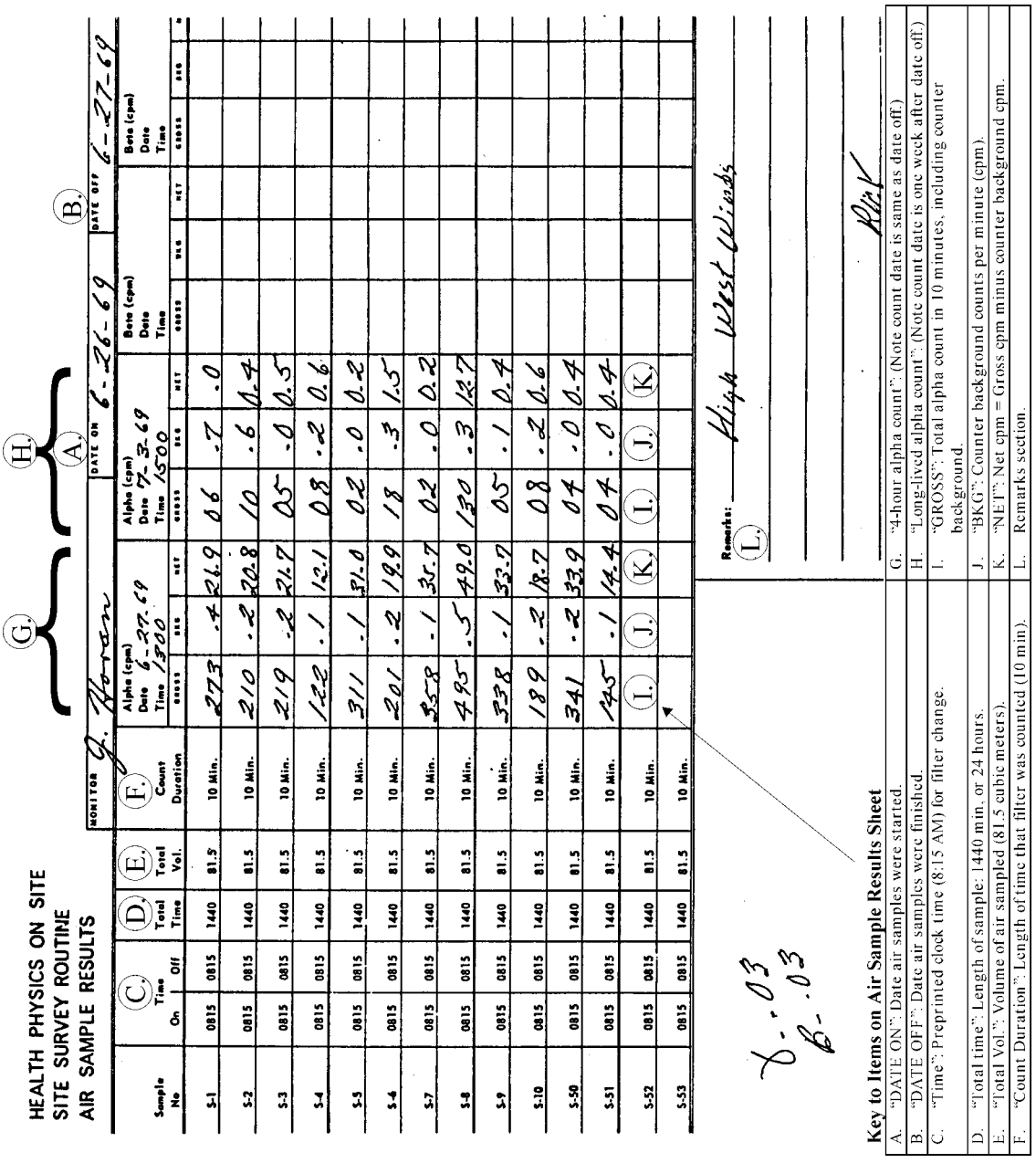
Fortunately, copies of handwritten daily results sheets titled, “Health Physics On Site, Site Survey Routine Air Sample Results,” were retrieved from the Denver Federal Record Center. These data bracket the key period of interest for releases from the 903 Area. The complete set of daily records includes data for air samplers S-1 through S-10, S-50 and S-51 (Figure III-17). The S-1 through S-10 data span the time period October 10, 1964, through December 29, 1971, (with the exception of S-9 data, which is only available for September 18, 1967 to March 30, 1969). The S-50 and S-51 data are limited to October 4, 1968, through December 29, 1971. (Subsets of these data are presented in the ChemRisk Task 6 report (1994b), Table M-1 and Appendix O.)

The daily alpha and beta count information were recorded on a standard form. An example is included here (Figure III-20). Note that the total sampling time (1440 minutes) and volume (81.5 m^3) were preprinted as standard values for all daily samples, that is, specific times and flow rates were not recorded for each individual sample. This is one source of uncertainty in the measurements, which is considered in the data quality section of this chapter. Standard practice in more recent times was to record the start and stop clock times and flow rates for each sampler, which would then be used to obtain the actual volume of air sampled at each station. The flow rate of the onsite samplers at this time was 2 ft^3 min^{-1} (cfm); thus they are low-volume air samplers. High-volume samplers were employed in the mid-1970s.

Original daily air sample count sheets were located for the time period of highest releases from the 903 barrel storage area.

The filter from the air sampler was counted in an alpha scintillation detector. Energy from the alpha particles produced scintillations, or light flashes, which were amplified and detected, resulting in a "count" on the scalar of the detector. The sample filters were each counted for a duration of 10 minutes until a change in procedure on August 25, 1969, when the routine count duration was increased to 30 minutes. The total (gross) count during the 10-minute sample count time was recorded on the result sheets (Figure III-20).

A background count rate (counts per minute, or cpm) was recorded on the data sheet and was subtracted from each gross sample count to obtain the net counts per minute from alpha activity on the sample filter. On the example sample sheet here, for S-8, the net cpm was 130 counts/10 minutes, or 13 cpm – 0.3 cpm = 12.7 cpm (Figure III-20). The background count rate refers to the counter background, not an estimate of background alpha activity in the environment. Putzier (1996) recalled that the background count rate was determined with a blank air filter in the counter. Trace quantities of alpha-emitting radionuclides in the air filter, contamination of the counter from a previous sample, and spurious electronic noise could contribute to the counter background. The length of time that the background was counted was not given on the data sheets. Putzier (1996) recalled that a statistical approach was taken to determining how long the background count rate should be, and that it was perhaps about 1 hour. There were a number of scintillation counters dedicated to site survey samples. Selvidge (1975) states that the background for each counting instrument was measured at the beginning of every counting day. Uncertainties in alpha counts due to the statistics of radioactive decay and counter background are discussed in the data quality section of this chapter.



HEALTH PHYSICS ON SITE
SITE SURVEY ROUTINE
AIR SAMPLE RESULTS

MONITOR G. Horan (G.) DATE ON 6-26-69 (A.) (H.) DATE OFF 6-27-69 (B.)

Sample No	Time On (C.)	Time Off (C.)	Total Time (D.)	Total Vol. (E.)	Count Duration (F.)	Alpha (cpm) Date 6-27-69 Time 1500 GROSS	BKG	NET	Alpha (cpm) Date 7-32-69 Time 1500 GROSS	BKG	NET	Beta (cpm) Date Time GROSS	BKG	NET	Beta (cpm) Date Time GROSS	BKG	NET	*
S-1	0815	0815	1440	81.5	10 Min.	273	.4	21.9	06	.7	.0							
S-2	0815	0815	1440	81.5	10 Min.	210	.2	20.8	10	.6	0.4							
S-3	0815	0815	1440	81.5	10 Min.	219	.2	21.7	05	.0	0.5							
S-4	0815	0815	1440	81.5	10 Min.	122	.1	12.1	08	.2	0.6							
S-5	0815	0815	1440	81.5	10 Min.	311	.1	31.0	02	.0	0.2							
S-6	0815	0815	1440	81.5	10 Min.	201	.2	19.9	18	.3	1.5							
S-7	0815	0815	1440	81.5	10 Min.	358	.1	35.7	02	.0	0.2							
S-8	0815	0815	1440	81.5	10 Min.	495	.5	49.0	130	.3	12.7							
S-9	0815	0815	1440	81.5	10 Min.	338	.1	33.7	05	.1	0.4							
S-10	0815	0815	1440	81.5	10 Min.	189	.2	18.7	08	.2	0.6							
S-50	0815	0815	1440	81.5	10 Min.	341	.2	33.9	04	.0	0.4							
S-51	0815	0815	1440	81.5	10 Min.	145	.1	14.4	04	.0	0.4							
S-52	0815	0815	1440	81.5	10 Min.	(I.)	(J.)	(K.)	(I.)	(J.)	(K.)							
S-53	0815	0815	1440	81.5	10 Min.													

6-.03
6-.03

Remarks: High West Winds (L.)

Mark

Key to Items on Air Sample Results Sheet

A.	"DATE ON": Date air samples were started.	G.	"4-hour alpha count": (Note count date is same as date off.)
B.	"DATE OFF": Date air samples were finished.	H.	"Long-lived alpha count": (Note count date is one week after date off.)
C.	"Time": Preprinted clock time (8:15 AM) for filter change.	I.	"GROSS": Total alpha count in 10 minutes, including counter background.
D.	"Total time": Length of sample: 1440 min. or 24 hours.	J.	"BKG": Counter background counts per minute (cpm).
E.	"Total Vol.": Volume of air sampled (81.5 cubic meters).	K.	"NET": Net cpm = Gross cpm minus counter background cpm.
F.	"Count Duration": Length of time that filter was counted (10 min).	L.	Remarks section.

Figure III-20. Site survey routine air sample results sheet for onsite air samples: June 26, 1969. (Key to items on sheet added.)

There are two columns of alpha count data on the result sheet. The first column represents an initial count taken about 4 hours after sample collection. As discussed earlier in this chapter, this count is dominated by natural, short-lived alpha emitters like decay products of uranium and thorium. The second count was made 1 week later, when essentially all of the obscuring, short-lived alpha emitters had decayed to insignificant levels. Thus, the second column of total long-lived alpha (TLLa) is the value correlated with Rocky Flats releases of plutonium, americium and uranium. The minimum detectable activity (MDA) stated by the RFP for a 10-minute sample count was 0.21 cpm. From our analysis of the data sheets, this MDA is approximately the median of the counter background. That is, half of the background count rates would have been greater than the stated MDA, and half would have been less. A 90% confidence interval on the counter background for a subset of data from 1969 was 0 to 0.7 cpm. Gross counts were in the same range as counter background for many locations and times.

Occasionally the technician would note, in the remarks section of the data sheet, the presence of some operational activity or high winds suspected of contributing to airborne contamination. For example, on the data sheet for April 5, 1971, on which a relatively high alpha concentration at S-8 was recorded, the remarks section contains the note "Cutting ditch North of 903 Pad." On June 26, 1969, the technician noted "High West Winds" (Figure III-20). In the event of a sampling problem, for example, a power outage or pump malfunction, the technician would note this on the data sheet; in those cases the sample usually was not analyzed.

For some of our purposes, the counts per minute results from the data sheets must be converted to units of activity per unit volume of air sampled. The femtocurie is a convenient unit for concentrations of this magnitude; the femtocurie (fCi) is 1×10^{-15} Ci, or 0.001 picocurie (pCi). For daily onsite air samples analyzed for alpha activity, this conversion from counts to activity concentration is

$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / [(0.21 \text{ cpm dpm}^{-1}) \times (2.22 \text{ dpm pCi}^{-1}) (10^{-3} \text{ pCi fCi}^{-1}) (81.5 \text{ m}^3)]$$

or,

$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / 0.038.$$

In the conversion equation above, the total efficiency factor of 0.21 dpm (disintegrations per minute) per cpm, or 21%, is comprised of two subfactors: a 30% detector counting efficiency (determined from counting thinly plated standard alpha-emitting sources) and a 70% correction factor for self-absorption of the alpha particles within the depth of the air filter ($0.7 \times 0.3 = 0.21$). These factors are discussed in more detail in the Phase I Task 5 report, Section 2.2.2.1 (ChemRisk 1994a) and the data quality section of this chapter.

Analysis of Results of Daily Measurements of Total Long-lived Alpha (TLLa) Activity in Onsite Air (1964–1971)

As noted previously in this report, the TLLa data were transcribed from the handwritten results sheets and analyzed by *RAC* using the spreadsheet Microsoft Excel®, V5. For samples collected over periods longer than 1 day (e.g., weekends and holidays), the total net counts per

minute was divided by the number of days of sampling, so that all results in the database are estimates of average daily counts at each sampling station.

Table III-7 presents some summary statistics of this large data set. The information base is quite complete. There were over 27,700 data points over this time period. Of the 12 onsite samplers, 11 of them have ≤2% of the days with missing data. The second part of Table III-7 contains summary statistics of the net count results. The alpha counting method was not very sensitive. With the exception of sampler S-8, ≥ 70% of the measurements were less than the site's reported minimum detectable activity of 0.21 cpm (5.5 fCi m^{-3}), and more than 84% of counts were greater than 0.5 cpm (net). Counting errors are very large for net counts less than 0.5 cpm (see Data Quality section of this chapter). The mode (most frequently observed value) for all onsite sampler data sets was zero. Table III-8 provides some perspective on these levels. The counting method is not adequate to quantitatively measure typical background or fallout concentrations, which were established in the first sections of this chapter. On the other hand, the method is quite adequate to measure the gross contamination that occurred around the 903 Area during high wind events and operational activities.

Table III-7. Summary Descriptive Statistics for Daily Measurements of Total Long-Lived Alpha Activity in Onsite Air, October 1964 through December 1971

Statistic	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-50	S-51
Number of data points	2603	2594	2599	2596	2597	2580	2577	2610	2021	2588	1174	1172
Days of missing data[a]	30	39	34	37	36	53	56	23	612[c]	45	7	7
Percent days missing	1%	2%	1%	1%	1%	2%	2%	1%	30%	2%	1%	1%
Mean[b] (cpm)	0.25	0.18	0.23	0.23	0.23	0.29	0.39	1.97	0.16	0.19	0.20	0.17
Median (cpm)	0.1	0.1	0.1	0.1	0.1	0.1	0.1	0.3	0.1	0.1	0.1	0.1
Standard deviation (cpm)	0.54	0.31	0.45	0.45	1.14	1.02	2.78	17.50	0.25	0.36	0.36	0.33
Minimum (cpm)	0	0	0	0	0	0	0	0	0	0	0	0
Maximum (cpm)	10.6	5.7	9.9	7.6	54.5	39.0	130.3	654.3	2.7	5.9	7.8	8.1
Percent < MDA (0.21 cpm)	70%	77%	71%	73%	74%	70%	70%	45%	80%	77%	75%	78%
Percent > 0.5 cpm	15%	9.9%	13%	12%	12%	16%	16%	37%	9.2%	11%	9.9%	7.4%
Percent > 3.0 cpm	0.6%	0.2%	0.4%	0.6%	0.4%	0.8%	1.6%	7.3%	0%	0.3%	0.2%	0.1%

[a] Monitoring began at S-50 on 10/4/68, and at S-51 on 10/7/68. Days missing are computed based on operating interval.

[b] The mode (most frequently observed value) for all data sets was zero. All counts per minute results in this table are net, after subtracting counter background.

[c] There was no monitoring at S-9 from 12/22/66 through 1/23/67 and from 9/18/67 through 3/30/69.

Table III-8. Perspective on Magnitudes of Long-lived Alpha Activity Concentrations in Air

Measured quantity	Net cpm[a]	fCi m^{-3}
Monthly average fallout plutonium in Denver air in 1965 (see Figure III-2)		<0.3
Long-term average background due to naturally occurring long-lived alpha emitters in air (see later section of this chapter)	<0.1	1.4
Lowest reportable value on daily air sample results sheets	0.1	2.6
Site's quoted minimum detectable activity	0.21	5.5
Maximum daily background due to naturally-occurring long-lived alpha emitters in air (see later section of this chapter)	0.3–0.4	7–10
Long-term average concentrations at onsite stations, except S-7 and S-8	<0.3	<8
Alpha counter background range	0–1	0–26
Annual average concentration at S-7 in 1969	0.76	20
Annual average concentration at S-8 in 1969	7.0	185
Maximum monthly average concentration at S-8 (January 1969)	58	1525
Maximum daily concentration at S-8 on 1/30/69	654	17,200

[a] For daily onsite air samples, fCi m^{-3} equals net cpm divided by 0.038 (see previous section of this report).

The two locations with the highest maximum concentrations (S-8 and S-7) are those which are closest to the 903 barrel storage area. The sampler showing the largest amount of alpha activity is S-8, which was downwind of the 903 Area to the east. Figure III-21 is a plot of the daily TLLa activity concentration in air at S-8. The highest daily value was 654 cpm (17,200 fCi m^{-3}) for the sample collected at S-8 on January 30, 1969.

For a history of operations at the 903 waste storage area, see one of our Task 2 reports, *The Rocky Flats Plant 903 Area Characterization* (Meyer et al. 1996).

Monthly concentrations, computed from the daily data, are tabulated and plotted in a subsequent section of this chapter. The monthly average concentrations also show a strong correlation to 903 Area activities, although the peak concentrations are not as apparent as the daily count data shown in Figure III-21.

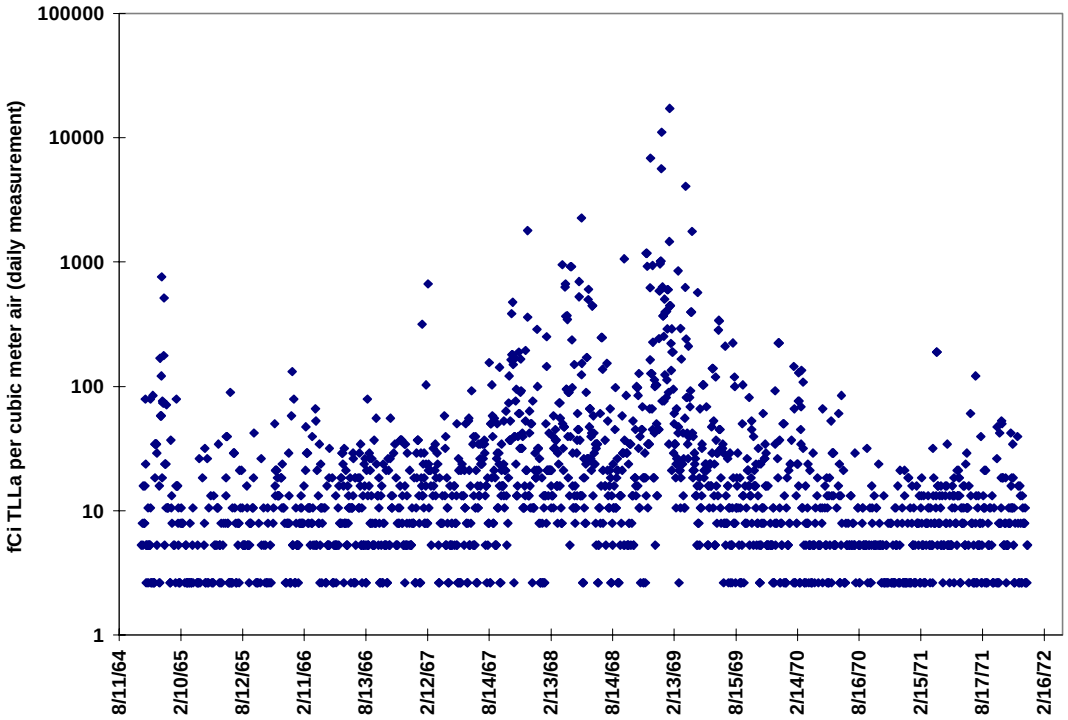


Figure III-21. Daily measurements of total long-lived alpha activity in air at the S-8 sampler near the 903 Area. Zero values were omitted from this semilogarithmic plot, although a net counts per minute of zero was the mode (most frequently observed measurement) in this and all other onsite air datasets. Horizontal-line appearance of plotted points is due to conversion of data reported in 0.1 cpm increments. For example, the lowest reportable positive count was 0.1 cpm, which is 2.8 fCi m^{-3}, or the lowest set of points on this plot.

Association between 903 Area Disturbance Activities and Air Sampling Results

The detailed air sampling data suggested a number of research possibilities for evaluating releases from the 903 Area, which were evaluated as part of Task 2 source term development (Meyer et al. 1996, Weber et al. 1999). Identifying days with particularly high counts was of interest to assess their association with high wind events. To isolate and examine the high airborne contamination days, the entire set of over 27,000 daily counts was sorted, and those with an activity of ≥ 5 cpm were selected. There were 166 days out of this 2,633-day period in which at least one sampler recorded a TLLa count of 5 cpm or greater. A tabulation of data for these high-count days, sorted in three ways (by location, by date, and by activity) is included as Table B-5 of Appendix B. This exercise also pinpointed a few arithmetic errors on the data sheets, which were corrected in the database. In another instance, a high value was due to the fact that the second count of the sample was conducted *2 days* after collection, rather than after 1 week's decay time. Because that count is not comparable to the others, those data were deleted from the database.

A plot of the number of high-count days per month clearly reflects the dramatic fluctuations in onsite airborne alpha activity (Figure III-22). The month with the most high-count days is

January 1969, followed by April and May of 1968. Table III-9 summarizes key operational activities and weather that likely affected the timing of releases from the 903 Area.

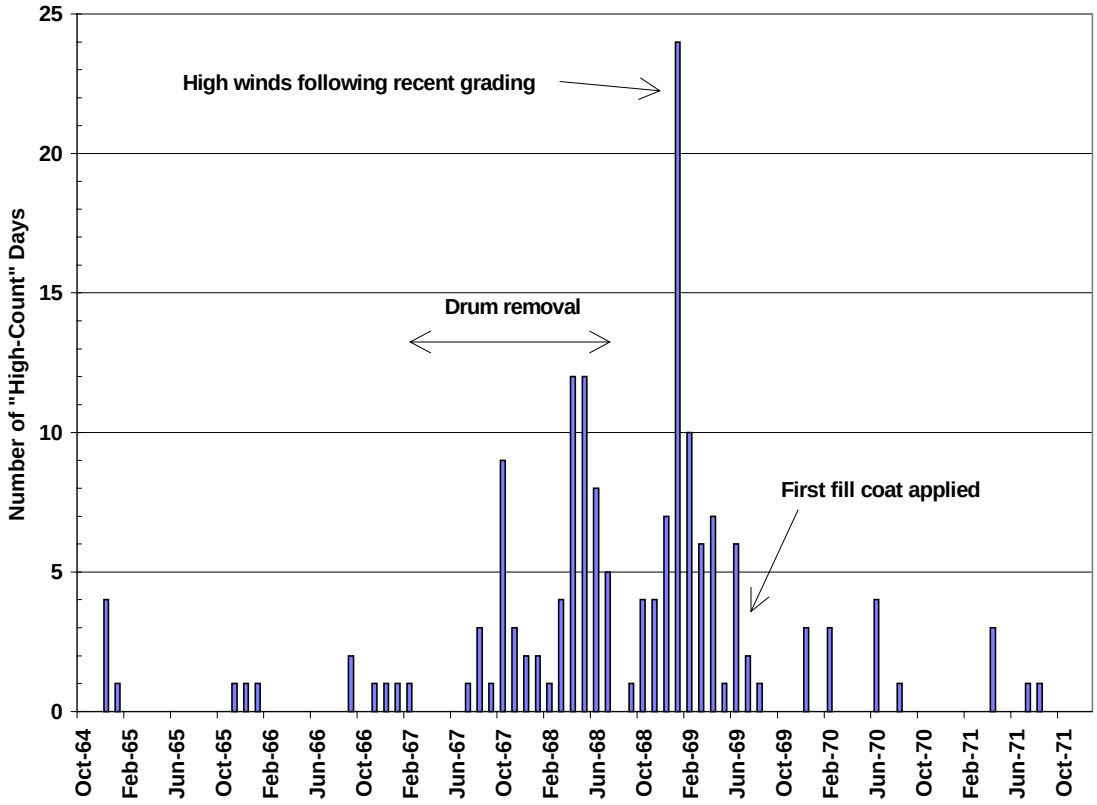


Figure III-22. Number of days per month between October 1964 and December 1971 in which one or more onsite air samplers recorded a daily net count of at least 5 cpm TLLa activity (equivalent to 130 fCi m^{-3} air). There were 166 high-count days out of this 2,633-day period. January 1969 was the most active month, with 24 out of 31 days registering high alpha counts. The shape of the curve substantiates the timing of releases from the 903 Area. Note that the impact of the May 1969 fire is not distinguishable from the larger 903 Area impact at these onsite air sampler locations.

Table III-9. Highlights of Operational Activities and Weather That Likely Affected the Timing of 903 Area Releases Before Covering with Asphalt

Time Interval	Events
Jan 1967–June 1968	Drum removal
October 1968	Weeds burned
November 1968	Grading
December 1968–Jan 1969	High winds
First Quarter 1969	Rock and rubble removal
Mar or Apr 1969	Grading
July 23, 1969	First coat of fill (6-in. road base)
November 1969	Asphalt pad cover work complete

Covering the old drum storage area with asphalt did not completely eliminate airborne contamination, as there were still days with high counts (Figure III-22). Rather extensive areas of soil outside of the asphalt pad were contaminated (see Chapter VIII), and operational disturbances affected airborne contamination levels. For example, four sample wells were dug in November 1969, with concurrent increases in air concentrations. Similarly, a new drainage ditch was completed in the area in April 1971, correlating with high airborne contamination for a few days.

Apportioning Total Long-lived Alpha Count into Specific Radionuclides

At least one approach to assessing releases from the 903 Area before 1970 involves quantitative examination of air monitoring data from station S-8 (Meyer et al. 1996, Weber et al. 1999). In order to apportion those TLLa concentrations into specific radionuclides released by Rocky Flats, the contributions from natural alpha-emitting radionuclides must be addressed. Two approaches were used to estimate the alpha activity in ambient air from naturally occurring alpha emitters (Meyer et al. 1996). The first was examining data presented in a special study documented in Illsley (1982), which was conducted to determine the contributions to the TLLa count made by emitters other than plutonium and americium. A long-term average of 0.1 to 0.3 fCi m^{-3} TLLa activity from natural isotopes of uranium, thorium, and radium was obtained from examining those data. Secondly, a dust loading approach was used, which was based on the natural alpha content of soil and the amount of dust in the air. This method produced a long-term estimate of 0.3 fCi m^{-3} TLLa activity.

Some of the TLLa activity in onsite air samples is due to naturally occurring alpha emitters and fallout plutonium. The remainder can be attributed to plutonium, americium, and uranium from Rocky Flats.

After publication of Meyer et al. (1996), an interested reviewer commented that ^{210}Po, a naturally occurring decay product of radon, is present in the air in greater quantities than would be expected based on its uranium series parent radionuclides in airborne dust. Polonium–210 is an alpha-emitter with a half-life of 138 days; therefore, it would remain on an air filter at the time of the 1-week delayed count. An estimated concentration of ^{210}Po in surface air is 40 μBq m^{-3} (1.1 fCi m^{-3}), according to NCRP (1987). Therefore, to interpret TLLa counts in ambient air, we intend to use a natural background concentration of 1.4 fCi m^{-3} (1.1 fCi m^{-3} from ^{210}Po and 0.3 fCi m^{-3} from thorium, uranium, and radium) for TLLa activity. It is recognized that this value represents a long-term average estimate. An individual monthly background value was not expected to be more than a factor of 3 higher than the long-term average concentration (i.e., about $1.4 \times 3 = 4$ fCi TLLa m^{-3}). An individual daily background concentration might be higher, perhaps up to a factor of 5 or more, or around 7–10 fCi m^{-3}.

CDH monitored TLLa activity in air since 1970, and their data confirm these general levels. Their minimum detectable concentration was 3 fCi m^{-3}. However, it is important to note that quite high TLLa measurements, often attributed to ^{210}Po, did occasionally occur. A plutonium analysis was triggered by any individual TLLa concentration that exceeded a screening level of 40 fCi m^{-3} (CDH 1978). Based on observations in 1969 through 1971, the CDH concluded that the off-site average concentration of TLLa in air was 11 fCi m^{-3} (CDH 1972b). Naturally occurring ^{210}Po was a typical cause of high long-lived alpha concentrations in offsite air; in July 1972 the high individual sample concentration in Longmont, Colorado was 150 fCi m^{-3} (CDH

1972c). A plot of TLLa activity in air particulates during February and March 1974 showed individual results as high as 8–12 fCi m^{-3} in offsite air and somewhat less (up to 9 fCi m^{-3}) in onsite air (CDH 1974a). In April 1974, both onsite and offsite air occasionally contained over 12 fCi m^{-3} TLLa activity (CDH 1974b). CDH reported a long-term (1970–1975) average concentration of TLLa activity in onsite, offsite (metro) and offsite (remote) air of 7, 7, and 6 fCi m^{-3}, respectively (CDH 1977). The RFP action level for triggering plutonium analysis on an ambient air sample in the 1970s was 10 fCi m^{-3}.

After subtracting the natural radionuclide contribution, the remainder of the TLLa activity from an onsite air measurement can be attributed to Rocky Flats materials, namely plutonium, americium, and uranium. Several sources have indicated that the plutonium concentration is only a fraction of the TLLa activity in air, even after considering natural background. Michels (1973) examined the TLLa activity and plutonium content on air filters collected in 1970 and 1971 by the five CDH air samplers (D-1, D-2, D-3, D-4, and 56) near Rocky Flats (see a previous section of this chapter for CDH sampler locations). According to Michels, the CDH detection limit at that time was about 3 fCi m^{-3} and values up to about 5 fCi m^{-3} corresponded to the range found in ordinary air around Denver. Michels wished to use the more abundant TLLa data in his study, so he examined the fraction of the TLLa activity that was due to plutonium, for those filters that were analyzed for both TLLa and plutonium. The CDH data as analyzed by Michels are illustrated in Figure III-23.

The data are quite scattered. However, Michels notes that the data trend falls near the 33 and 50% curves, which suggests that between one-third and one-half of the airborne alpha activity is plutonium. For purposes of his study, Michels attributed 40% of the TLLa activity over 5 fCi m^{-3} to plutonium.

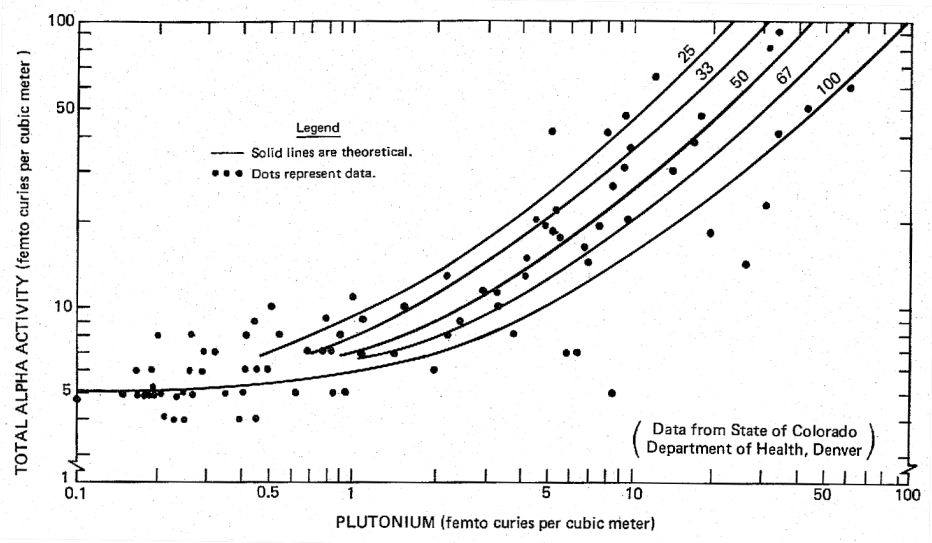


Figure III-23. Percentage of total long-lived alpha activity from plutonium (Michels 1973), based on data collected by the Colorado Department of Health in 1970–1971. Solid lines represent theoretical curves if plutonium comprised 25, 33, 50, 67, or 100% of the TLLa activity over 5 fCi m^{-3}. Dots represent actual measurements. Michels (1973) used a percentage of 40% in his study.

Also in the early 1970s, the HASL was measuring plutonium at the eastern security fence (see a previous section of this chapter). We examined the relationship between the monthly average concentration of plutonium measured by HASL at the eastern security fence and the TLLa activity measured by Dow at sampler S-8. The concentration of plutonium measured each month by HASL in New York City was subtracted as an estimate of global fallout plutonium background. The data are tabulated in Appendix B. The fallout concentration was less than 0.1 fCi m^{-3}, small compared to plutonium concentrations measured at Rocky Flats. As discussed earlier, an alpha background of 1.5 fCi m^{-3} (1.4 fCi m^{-3} from natural sources plus 0.1 fCi m^{-3} from fallout plutonium) was subtracted from the TLLa count to represent the alpha activity from Rocky Flats materials alone. The TLLa concentrations measured by Dow were also converted to standard temperature and pressure conditions (0°C, 760 mm Hg), which was standard HASL practice. This assures that the Dow and HASL concentrations are comparable in terms of standard volumes of air sampled. The data are plotted in Figure III-24. Although the data are scattered, the least-squares best fit to the data results in a best estimate of 36% plutonium, close to the 40% value obtained by Michels.

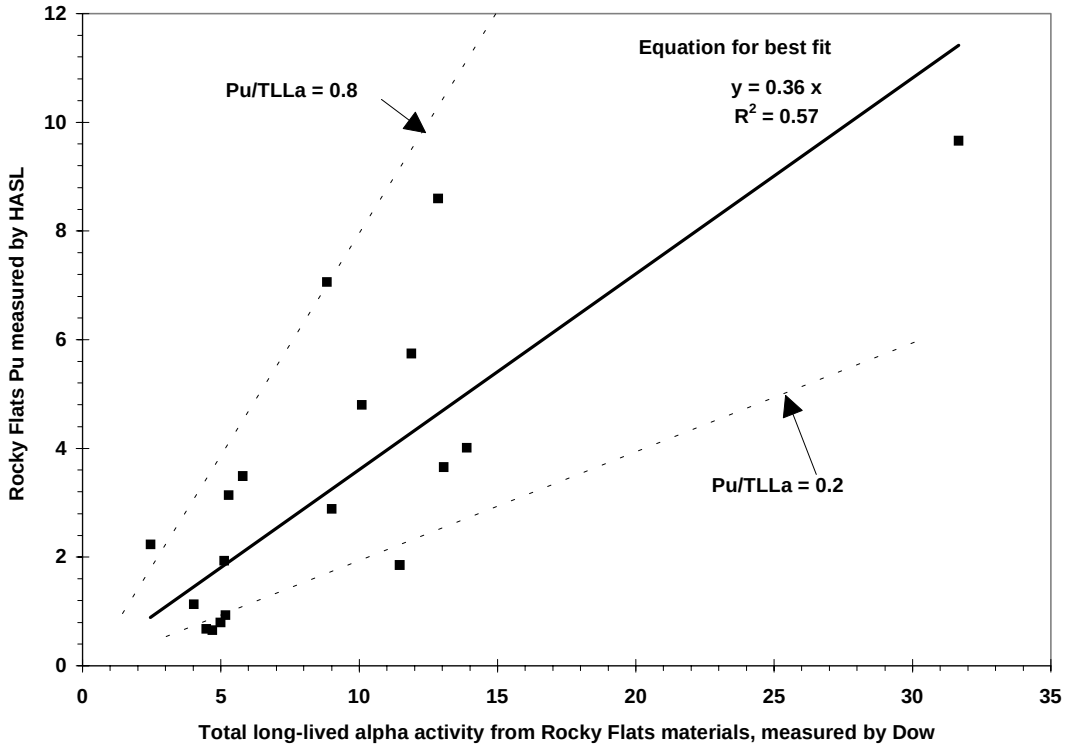


Figure III-24. Relationship between total long-lived alpha activity (fCi m^{-3}) measured by Dow and plutonium measured by the Health and Safety Laboratory at the eastern security fence during an 18-month period in 1970 and 1971. Each point represents the monthly average measurement. Natural and fallout activity have been subtracted so that the measurements represent only Rocky Flats materials. A best fit to the data indicates ~40% plutonium, although a range of 20 to 80% is plausible based on these data.

If 40% of the TLLa activity from Rocky Flats materials is due to plutonium, what constitutes

the remainder? The two most likely contributors are ^{241}Am and isotopes of uranium. According to Phase I estimates of airborne releases, americium comprised 18% of the TLLa activity in effluents from plutonium areas, and ^{239,240}Pu contributed the other 82%. The ratio of ^{241}Am to ^{239,240}Pu activity in airborne effluents from plutonium areas was 0.22, or 22%.

Americium-241 would also be produced by decay of ^{241}Pu, which was a small fraction of the plutonium processed at Rocky Flats. Poet and Martell (1972) states that the maximum concentration of ^{241}Am from ^{241}Pu decay would occur 70 to 80 years after chemical purification of the plutonium mixture. At that time, the ^{241}Am alpha activity would be about 54% of the ^{239,240}Pu activity if the initial ^{241}Pu activity were 1.0% (maximum estimate according to Owen 1968) of the total plutonium alpha activity. This does not take into account the independent emission of ^{241}Am from Rocky Flats. Poet and Martell's analysis of environmental samples in the early 1970s indicated that ^{241}Am was between 3 and 30% of the ^{239}Pu activity, with the more reliable results in the lower half of this range.

In the 1980s, Langer (1986b) measured the activity ratios of Rocky Flats materials in resuspended dust from contaminated soil areas remaining in the 903 Area. The ratio of ^{241}Am to ^{239,240}Pu activity at that time was 0.13, 0.20, and 0.11 at three different sites with different vegetative conditions.

Releases of uranium from the Rocky Flats production area were most important in the 1950s. Before the 1957 fire in Building 771, releases of TLLa activity to air from uranium areas exceeded releases from plutonium areas (ChemRisk 1994a). Hammond (1958) indicated that normal operations of uranium plants at Rocky Flats had contaminated the surrounding area (up to the cattle fence and beyond into the Church property) with detectable amounts of depleted and enriched uranium. Of three air samples taken September 12–13, 1957, and analyzed by pulse-height analysis, two indicated mainly enriched uranium with some plutonium, and the other contained "approximately equal amounts of 3 plant materials."

These three materials that Hammond mentioned were referred to in early site documents by code names: "A" material (depleted uranium), "B" material (enriched uranium), and "C" material (plutonium). The environmental measurements in the 1950s support the importance of uranium releases at that time. Scattered results of pulse-height analyses of onsite air samples are reported in the monthly site survey reports for 1955. "A" and "B" materials (total uranium) comprised between 24 and 100% of the TLLa activity of these onsite air samples, with up to 76% being plutonium.

The October 1955 monthly site survey report states that "some of the air sampling stations near the NO_3 pond consistently showed high counts during the time of definite wind trends." (The location of the nitrate pond, later called solar evaporation ponds, is shown in Figure VI-2 of Chapter VI). A special high-volume air sample collected October 28, 1955 on the east side of the nitrate pond, when winds were 30–35 mph from the west, contained TLLa activity that was all enriched uranium. Deposition of TLLa activity onto gummed paper near the nitrate pond in November of 1954 was mostly uranium and only 5% plutonium alpha activity (see Chapter IV). These few measurements indicate that releases of uranium from Rocky Flats operations to surface water could also contribute to measured amounts of uranium alpha activity in air.

By 1965, however, the cumulative TLLa activity released to air from plutonium areas (mostly plutonium and americium) was about 39 mCi compared to a cumulative release of about 15 mCi from uranium areas. These totals were reconstructed as part of Phase I of this study (ChemRisk 1994a, Table 2-32) and do not include releases from the 903 barrel storage area. Thus, of a total of 54 mCi long-lived alpha activity released to air by 1965 from RFP operations,

about 27% (39 mCi/54 mCi) might have been uranium. It is not known to what extent these percentages can be applied to the isotopic composition of the waste oil on the 903 Area, and consequently the airborne alpha activity resulting from leakage of that oil.

Voillequé (1999) reexamined the historical releases of uranium to air from Rocky Flats and assessed their importance (in terms of public exposures) relative to plutonium. He concluded that plutonium releases were 4.5 times more radiologically significant than enriched uranium and 2.2 times more significant than depleted uranium in routine releases. Routine releases of plutonium were small relative to accidental releases (particularly the 903 Area and the 1957 fire), further diminishing the overall importance of uranium. A conservative assessment of the accidental burning of a pallet of depleted uranium concluded that the radiological significance of uranium released was 60 times less than one high-wind day of releases from the 903 Area. The emphasis on plutonium as the primary radionuclide with respect to historical public exposures was substantiated.

Weber et al. (1999) used data from the onsite air sampler S-8 in the development of the 903 Area source term. For the fraction of the TLLa count attributed to plutonium, they used a triangular distribution with a most likely value of 0.4 (40%) and minimum and maximum values of 0.2 and 0.8.

Average Concentrations: General Trends in Airborne Alpha Activity of Onsite Air

For some purposes, an average concentration of airborne alpha activity over a period of time is more useful than the daily measurements. For example, it is easier to visually compare one location with another or examine trends over time without the extremely large number of data points. (There are over 2,500 data points for each of the onsite samplers S-1 through S-8 for the period October 1964 through December 1971.) It should also be remembered that a cumulative exposure assessment, e.g., determining the intake of a material from breathing contaminated air, will produce the same result when using average concentrations as the daily concentrations.

Our computer file containing the onsite alpha activity measurements is structured so that the date associated with each daily alpha count is the date the air sampler was started. This date is the one during which the majority of air was collected, because change-out of air filters was done in the morning. The date the sample was stopped is the following day. Averages were produced for all dates falling within a given month

Table B-6, Appendix B, contains the monthly average concentrations in femtocuries of long-lived alpha activity per cubic meter of air. The femtocurie unit was chosen so that the numbers could be tabulated and plotted without the use of scientific notation. The average concentrations for all onsite samplers are plotted in the following figures.

These are the first long-term plots for *all* onsite samplers at Rocky Flats that we have seen. A Dow committee evaluation of plutonium levels in soil near Rocky Flats (Seed et al. 1971) presented a similar monthly average plot for sampler S-8, which was downwind of the 903 Area, and which consistently showed the highest readings. This plot has been widely reproduced in various reports to illustrate the timing of historical releases of radioactivity from the 903 Area. After producing our plot of monthly averages for sampler S-8, it was evident that there was a significant discrepancy for the peak month, January 1969 (Figure III-25). There are no numerical data in the Seed report, only the plot. We estimated the concentrations from the Seed plot to illustrate the comparison in Figure III-25. Our monthly average for January 1969 is 1,525 fCi m^{-3}, whereas the Seed report value is about 150 fCi m^{-3}.

We double-checked our computation from the daily data sheets, and cross-checked several other types of historical Dow records. The peak *daily* concentration of 17,200 fCi m^{-3}, which we computed from the counts per minute on the data sheet, is confirmed in the January 1969 site survey report, which gives an onsite maximum value of 1718.04 uc × 10^{-14}/cc. Applying conversion of units, this is equivalent to 17,200 fCi m^{-3}. In addition, our total activity (in cpm) collected at S-8 during January 1969 agrees with a monthly computation sheet, on which Dow summarized the daily records for this month.

ChemRisk informed us that they had also investigated this discrepancy and found the January 1969 TLLa activity concentration at the S-8 sampler reported by Seed et al. (1971) to be erroneous. This issue is addressed in their Phase I Task 6 report (ChemRisk 1994b), Appendix M.

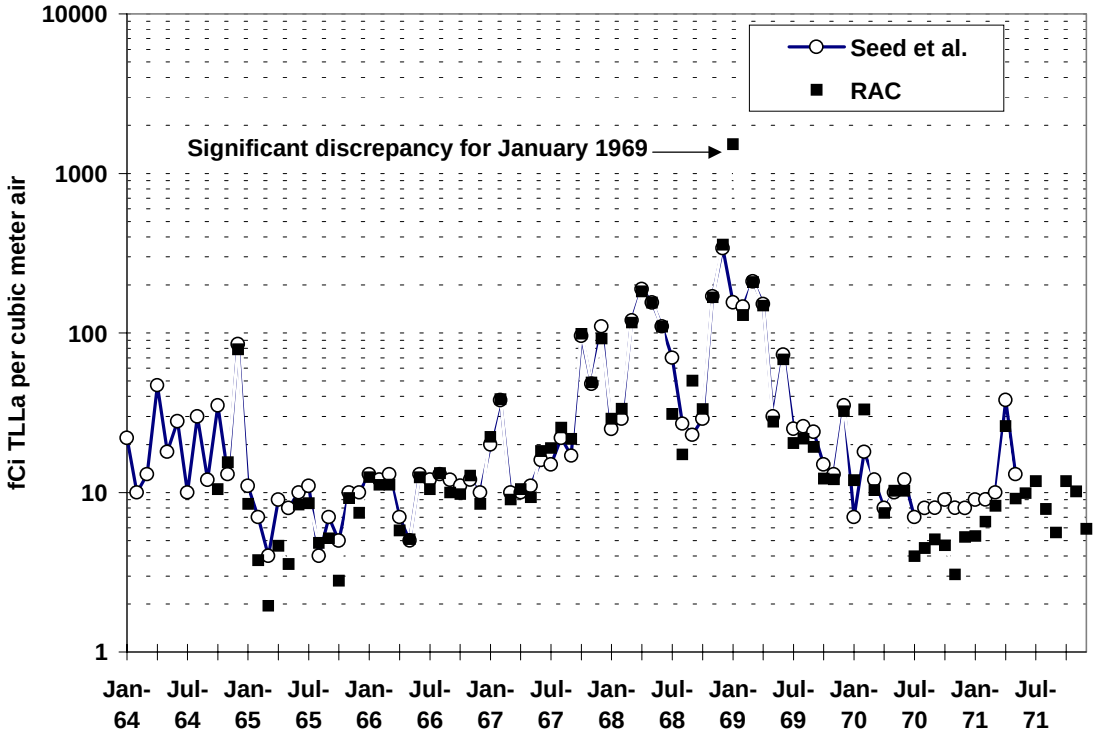


Figure III-25. Comparison of monthly average concentrations of total long-lived alpha activity in air at the S-8 sampler computed in this study by *Radiological Assessments Corporation (RAC)* and those previously plotted by Seed et al. (1971). Note the semilogarithmic scale. The minimum detectable activity was 5.5 fCi m^{-3}.

Our averages for the other months agree reasonably well with the Seed report. There are several known reasons for slight discrepancies between our reconstructed averages and Dow's. For weekend or holiday samples, we divide the total activity collected (in cpm) by the number of days and assign that amount to each day of the interval. Dow assigned all the activity to the sample change-out day. We assigned a measurement to the date the majority of sampling was conducted; Dow often assigned the count to the date of change-out. For example, the maximum recorded long-lived alpha concentration of 654.3 cpm was obtained from a sample that was started on January 30, 1969, and stopped on January 31, 1969. This sample is associated with a

date of January 30 in our computer file and with January 31 in the Dow document titled "Result Sheet for Offsite Air Samples," on which the monthly average concentration is computed. Depending on how dates fell with respect to the end of the month, we might include a count (or part of it) in January, where Dow would have included it in February. This 1-day difference in how the count is recorded is one minor reason why our reconstructed monthly averages may differ from those calculated by the RFP.

Our reconstructed monthly average concentrations of total long-lived alpha activity in air were compared to those produced by Dow Chemical Company.

Our reconstructed averages are frequently less than Dow's, for low contamination months, because of the site practice of using the MDA of 0.21 cpm for any sample that had a net sample count less than the MDA (0.21 cpm translates to 5.5 fCi m^{-3}). In contrast, we used the actual net cpm, which frequently was zero, in computation of averages. Dow's procedure introduces a known positive bias when concentrations are low but no significant bias for time periods when there was a large amount of airborne activity.

There are 2 months, September 1968 and January 1970, in which our reconstructed averages are about 2 times the estimated Seed value. For September 1968, Dow's monthly calculation sheet reported as "missing" the highest daily result for that month. We had no reason to invalidate that datum. We could determine no reason for the January 1970 discrepancy—our values are consistent with Dow's monthly calculation sheet; thus, it appears that the Seed value is plotted too low. However, both concentrations are low for that month, and further investigation did not appear warranted.

The monthly average concentrations at the three samplers closest to the 903 Area are illustrated in Figure III-26. As opposed to Figure III-25, Figure III-26 and subsequent monthly average plots in this section use a linear scale. These data support the timing of releases from the 903 Area. The peak concentrations in 1968 and 1969 coincide with known dust-suspension periods in the 903 Area. January 31, 1969, was the date of high winds that "blew all of the roofing material off Building 889" (Putzier 1969).

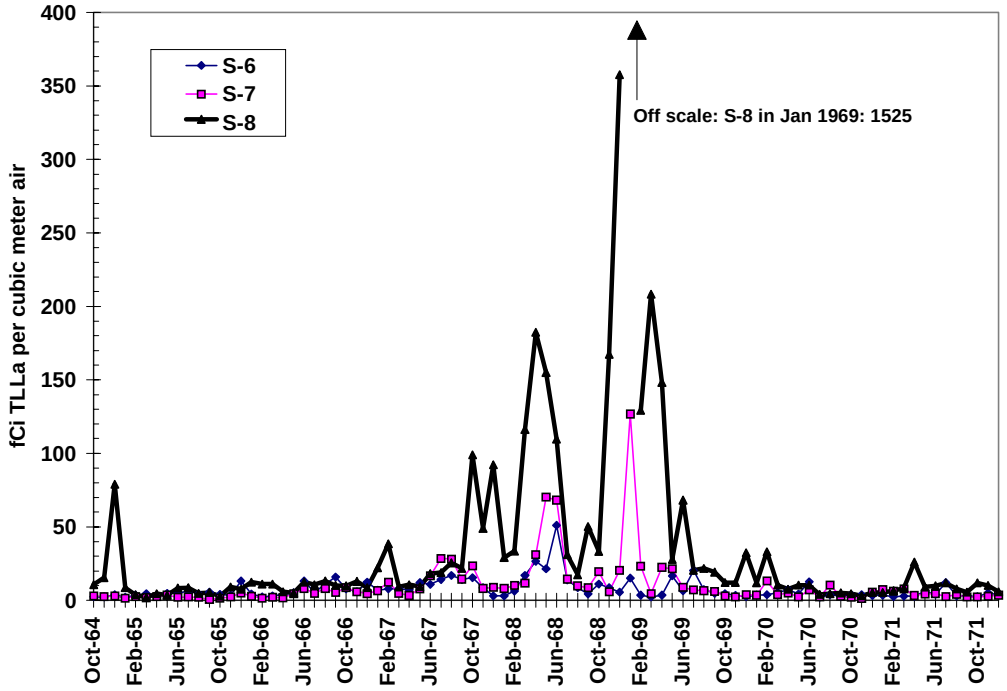


Figure III-26. Time trend in monthly averages of daily measurements of total long-lived alpha activity in air at the three samplers nearest the 903 Area.

Relatively speaking, the other onsite locations do not pick up much contamination from the 903 Area events. Figure III-27 illustrates the annual average concentrations measured at all onsite samplers in 1969, the year of highest airborne activity at S-8. With the exception of S-7, the concentrations at the other samplers are less than 10% of the S-8 level and are close to nondetectable (MDC = 5.5 fCi m^{-3}).

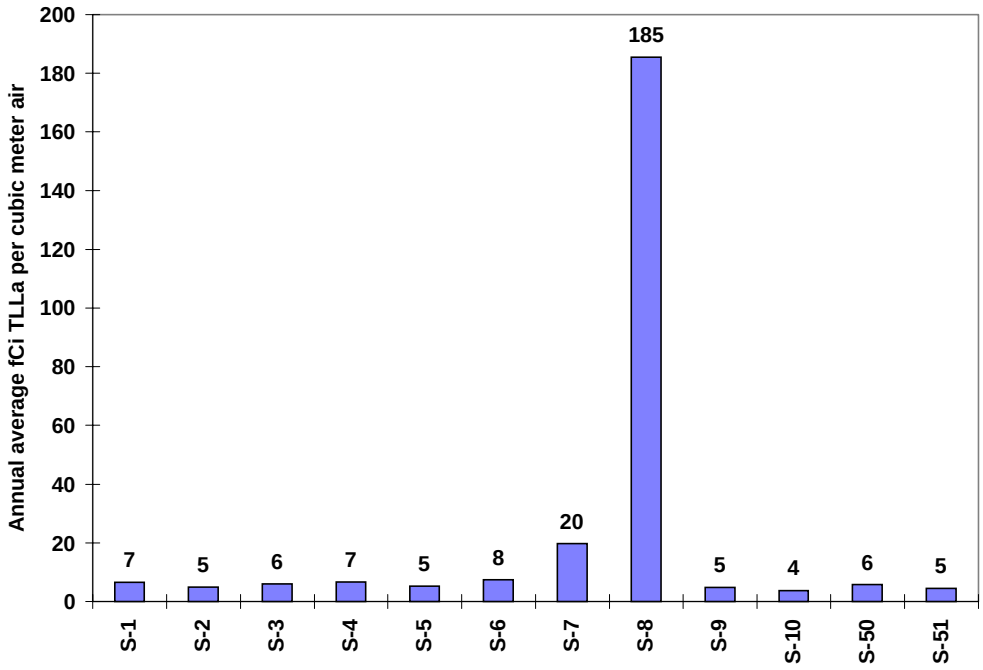


Figure III-27. Annual average concentrations of total long-lived alpha activity at onsite air samplers in 1969. Minimum detectable concentration is 5.5 fCi m^{-3}.

Figures III-28 through III-31 display the time trends in monthly averages for the other onsite samplers that are not near the 903 Pad. The vertical scale of all four figures was purposefully set to the same maximum value, 60 fCi m^{-3}, to permit a comparable visual perspective. It is obvious that the samplers near the 903 Area (Figure III-26) show much higher airborne contamination levels than the others. Many of the averages at the samplers not located near the 903 Area are below the MDA, shown by dotted lines on the plots.

The peaks in the air sampling record were investigated. In addition to the 903 Area, stack releases, spills, operational disturbances, and other sources of fugitive emissions (such as the solar ponds) contributed to sporadic increases in airborne alpha activity. As discussed earlier, the MDA is higher than concentrations that would be expected from naturally occurring or fallout alpha emitters, although daily fluctuations could perhaps account for isolated days with high alpha activity concentrations.

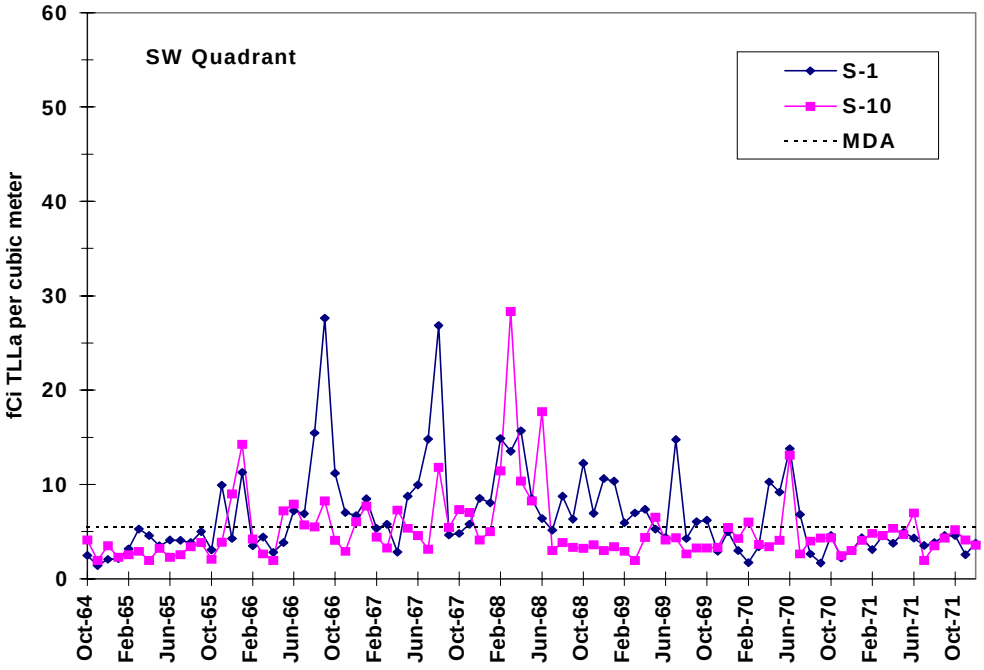


Figure III-28. Monthly average concentrations of total long-lived alpha activity in air at onsite samplers S-1 and S-10, in the southwest quadrant of the RFP.

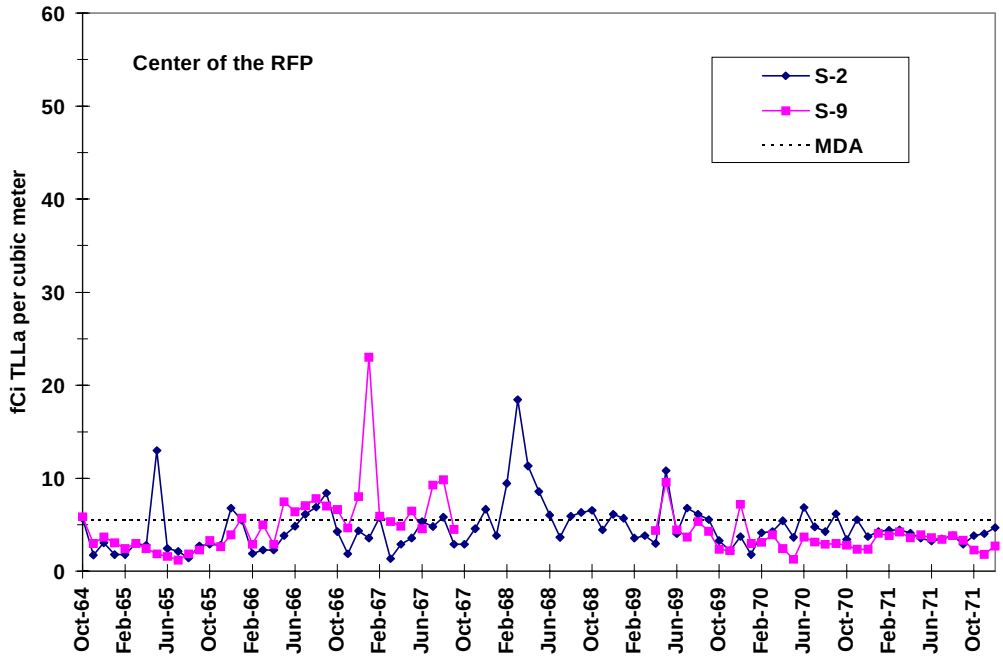


Figure III-29. Monthly average concentrations of total long-lived alpha activity in air at onsite samplers S-2 and S-9, roughly in the center of the RFP.

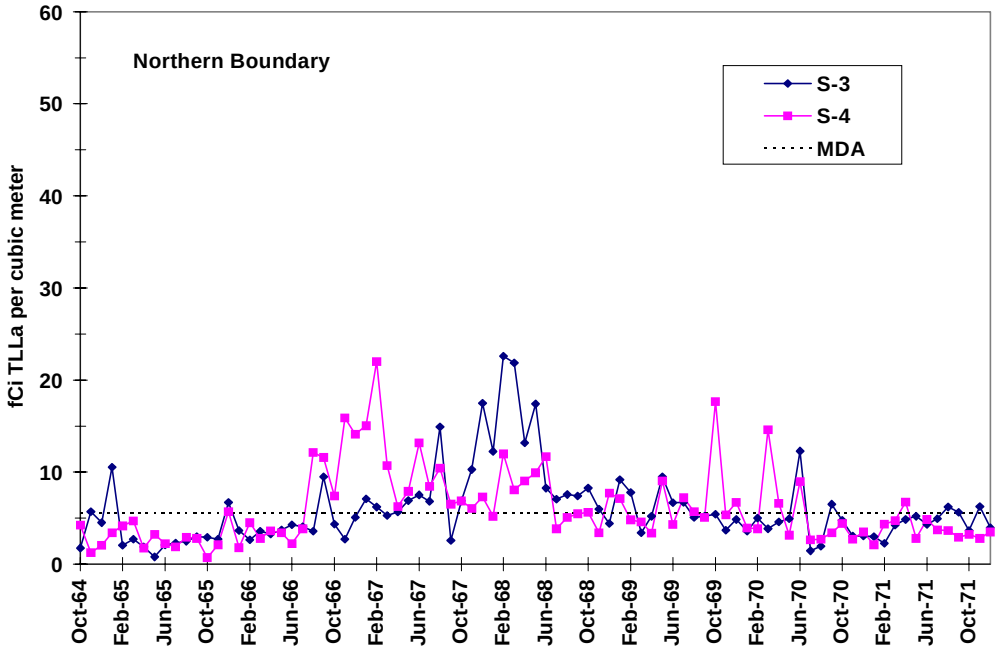


Figure III-30. Monthly average concentrations of total long-lived alpha activity in air at onsite samplers S-3 and S-4, on the northern part of the RFP.

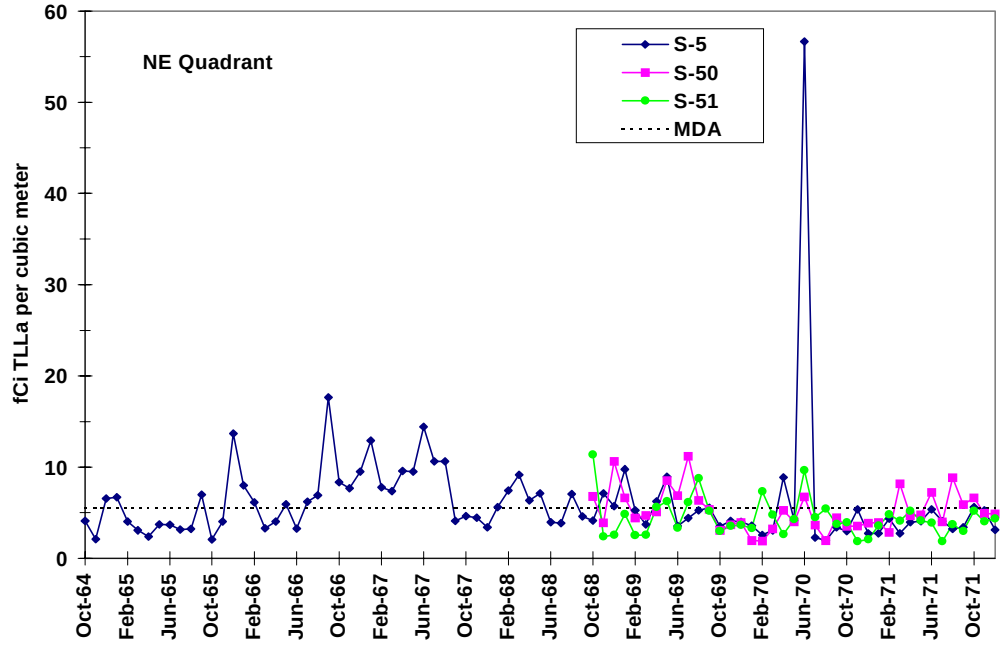


Figure III-31. Monthly average concentrations of total long-lived alpha activity in air at onsite samplers S-51 and S-5 (near solar ponds), and S-50 (near sludge beds), in the northeastern quadrant of the RFP.

Air Monitoring Associated with the May 1969 Fire

Rood and Grogan (1999b) used onsite air sampler data from the daily concentration database described earlier in this report to compare with predicted 24-hour average plutonium concentrations the May 1969 fire. The fire occurred on Sunday, May 11, 1969 in Building 776. The standard practice at that time for sampling over a weekend period was to allow the sampler to run from 8:15 a.m. Friday to 8:15 a.m. the following Monday. The results were then reported for the 3-day sampling period. To arrive at the estimated count rate of TLLα for the day of the fire, we first calculated the average net daily count rate for May, excluding those data that were taken over the weekend the fire occurred (Table III-10). The daily averages were then multiplied by 2 (to represent the 2 pre-fire days in the 3-day sampling period) and subtracted from the net count rate observed on the filter paper for the weekend that included May 9, 10, and 11. The new estimated counts for May 11 were then converted to activity concentrations in air using the conversion factor of 0.038 cpm $(fCi\ m^{-3})^{-1}$ (Table III-10). All excess TLLα activity was assumed to be plutonium that originated from the fire.

The time trend in daily net count rate for the month of May 1969 is shown in Figure III-32. The upper graph in Figure III-32 shows the raw data. Note that for each 3-day weekend period, all three days have the same count rate. That is because, in our database of TLLα in onsite air, we divided the total count equally among the sampled days. Also note that the weekend of May 9, 10, and 11 shows substantially higher count rates than the other days. The lower graph shows the same data but the weekend of May 9, 10, and 11 has been segregated into estimates for the three individual days.

Table III-10. Measured Data at Onsite Air Samplers for the Weekend of May 9–11, 1969 and Estimated Concentrations for May 11, 1969 (Rood and Grogan 1999b)

Sampler	Daily average count rate May 1969 (cpm)	Corrected count rate May 9–11, 1969 (cpm)	Estimated 24-hour average plutonium concentration May 11, 1969 ($fCi\ m^{-3}$)
S-1	0.17	1.5	30.4
S-2	0.13	6.3	159
S-3	0.17	6.0	149
S-4	0.18	5.4	132
S-5	0.15	6.0	150
S-6	0.45	6.6	150
S-7	0.66	6.3	131
S-8	0.74	11.7	269
S-9	0.16	6.3	157
S-10	0.23	1.2	19.4
S-50	0.22	3.9	91.1
S-51	0.088	4.8	122

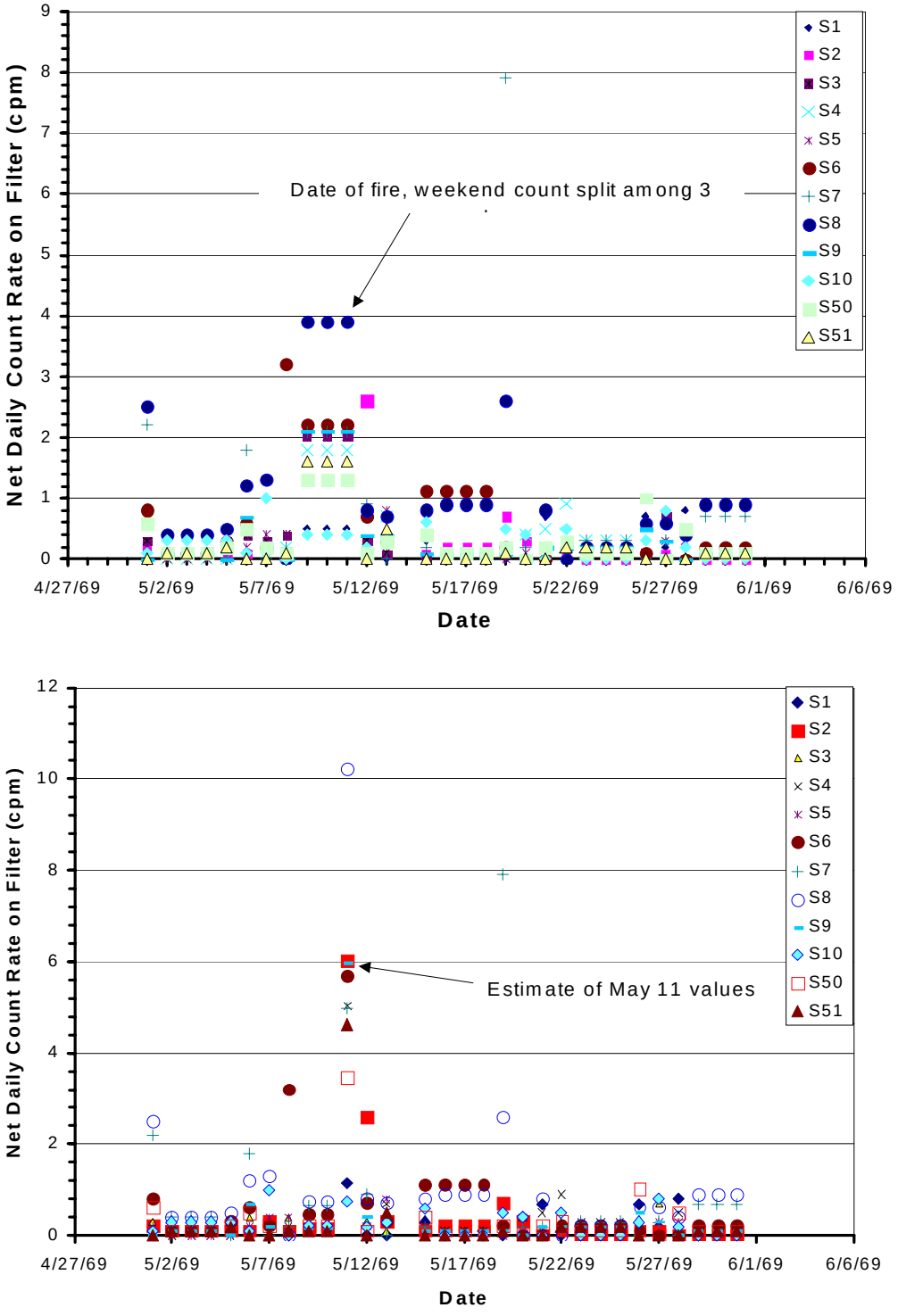


Figure III-32. Net daily count rates for the month of May, 1969. The upper graph shows the average daily count rate for the weekend of May 9, 10, and 11. In the lower graph, the count rate for May 11, 1969 has been segregated from May 9th and 10th. Also note the different vertical scales on the two graphs.

Offsite Monitoring for Alpha Activity in Air

As stated previously, there were no routine plutonium-specific measurements in ambient air during the time period of key interest for this dose reconstruction (pre-1970). Instead, the air samples were analyzed for TLLa concentrations. Based on our framework exercise for evaluating the usefulness of environmental data (Chapter I), we supposed that these data could be useful for spatial comparisons (location A versus location B) or time comparisons (trend analysis). However, based on our evaluation of onsite air data, we suspected that poor sensitivity would be a problem for TLLa measurements in community air samples.

History

Chapman (1952) documents the recommendation to establish offsite air sampling stations to "determine the normal background level of radioactivity in the area, detect air-borne activity in the event of inefficient operation or an accident and to serve as negative evidence in the event of legal action from imagined injury." Fourteen potential offsite locations were identified and described in Chapman (1952) in the communities of Plainview, Eldorado Springs, Marshall, Superior, Lafayette, Arvada, Wheatridge (Denver), Golden, Westminster, Eastlake, Leyden, Wagner School, Boulder, and Broomfield. All samplers were to be mounted on power poles that supplied power to the unit. A weatherproof housing had been designed by the Engineering Group.

Boss (1972) indicates that Dow's initial air sampling program began in 1953 and consisted of one offsite and nine onsite samplers. During the fall of 1953, and continuing through 1956, the number of offsite samplers increased to 15. In 1956, the offsite sampling was reduced to nine locations. The distances to those stations are given by Hammond et al. (1969) as Coal Creek Canyon (4.3 mi.), Marshall (4.7 mi.), Boulder (7.6 mi.), Lafayette (9 mi.), Broomfield (6.2 mi.), Wagner School, southeast of RFP (1.9 mi.), Golden (9 mi.), Denver, roof of Customs House (14.7 mi.), and Westminster (9.4 mi.).

We verified the historical sequence described by Boss by examination of the monthly reports of the site survey group for 1952–1963. These reports also provided some information about the sampling methods. The July 1953 report confirmed that the offsite samplers were low-volume samplers, calibrated to a 2 cfm flow rate. In August 1953, elapsed time meters were installed at all stations. In January 1954, offsite air samples were being collected weekly and analyzed for alpha and beta activity. In February 1954, the monthly report indicates that Whatman 41 paper had been substituted for HV70 paper on all offsite samples beginning February 8th. The HV70 paper had ruptured in week-long samples. A geometry of 50%, or 0.5 cpm/dpm, was used by Dow in the 1950s when converting net alpha counts to activity. The November 1955 report confirms the use of Gast brand pumps at the ambient air monitoring stations. By February 1960, the monthly report states that the Gast pumps were "reaching the point where frequent maintenance is required." In June 1961, the monthly report states that all offsite sample pumps were equipped with new screw type heads to reduce leakage; these new 2-in. samplers would draw 1.5 cfm. Hammond et al. (1969) state that the samplers were placed 6 ft above ground level. However, in a congressional hearings document (Congress 1970), it was stated that all but one of the ambient air samplers (presumably the Custom House roof in Denver) were placed 8–10 ft above ground.

Boss et al. (1972) indicates that Whatman 41 paper was still used, and that the average sampling rate was 2 cfm. However, the sampler type at that time was a Casse Fume Model 0465-V4A-025. The same nine communities were being sampled.

The *Health Physics Guide for Rocky Flats Division* confirms that the *community* air sampling in 1961 was not rigorous or quantitative:

> . . . air sampling stations located in populated areas within 25 miles of the plant site which are maintained in an operating condition in order that they may be put into immediate service if there is a release from the plant. A fortnightly check of these remote stations gives a qualitative confirmation of no discharge of airborne activity.

There is very sketchy information in the monthly site survey reports about the sampling frequency and duration. The April 1960 monthly site survey report stated that the Coal Creek station was changed daily. The March 1961 monthly report states that the S-13 station at Marshall was reactivated and that this station was also changed daily. The third-quarter 1961 environmental survey report indicates that 24-hour air samples were collected at all nine offsite stations. The fourth-quarter 1961 environmental survey refers to the offsite air samples as "continuous." However, the offsite air samplers were actually programmed to sample 5 minutes of each hour in 1969, which was probably the case in earlier time periods as well. This fact was not stated explicitly in the environmental survey reports until 1969. By 1972, most community samplers were still not operating continuously; they were programmed to sample 10 minutes of every hour (Boss et al. 1972). The samples at Coal Creek Canyon and Broomfield were operated continuously beginning July 1972. As a result, the apparent concentration at these two locations was substantially reduced after July, because of the reduced minimum detectable concentration (MDC) used in calculating those averages. The MDC for community samples collected 10 minutes each hour was typically $<0.0045 \times 10^{-12}$ μCi mL^{-1}, and for continuous community sampling, the MDC was $<0.0008 \times 10^{-12}$ μCi mL^{-1} (Boss et al. 1972). These MDC levels correspond to 4.5 and 0.8 fCi m^{-3}, respectively. Boss et al. (1972) stated that, based on the Coal Creek and Broomfield experience, monitors in the surrounding communities should be capable of operating continuously without serious mass filter loading.

In the 1969 environmental survey report, it also states that 8-hour high-volume air samples were periodically collected at one of the sampling stations, and that these samples were analyzed specifically for plutonium (see next major section of this chapter). The first such sample was taken on December 15, 1969, at the east security fence and showed a plutonium concentration of 2.2 fCi m^{-3}.

Early in 1971, the offsite sampling network was complemented by the addition of 12 high-volume air samplers located approximately 2 mi from the plant perimeter. The original low-volume air samplers (2 cfm) were upgraded to high-volume samplers (25 cfm) around 1975.

Reported Monthly Average Concentrations of TLLa in Offsite Air

The reporting of offsite air monitoring data in the contractor's site survey reports in the 1950s was limited to statements like "Negative results were obtained for the presence of significant quantities of alpha emitters." Whereas the beta activity in air was strongly influenced by weapons testing activities, the alpha measurements did not appear to be correlated with weapons testing (Figure III-33). This is consistent with our conclusion that average concentrations of alpha-emitting fallout radionuclides were below the MDC of the Dow alpha counting method. With the exception of a period of several months in the summer of 1955, the long-lived alpha concentrations in onsite and offsite air were generally not detectable in the 1950s. By October 1959, the site survey monthly reports had stopped including the offsite monitoring results.

Beginning in 1959, the offsite monitoring *only* was summarized in environmental survey reports, which were typically quarterly or semiannual. These reports were prepared in compliance with a Presidential executive order of August 14, 1959. The air monitoring data tabulated were the maximum, minimum, and average of *all stations combined* for each month. A continuous public-exposure standard of 4×10^{-14} µCi cm^{-3} (40 fCi m^{-3}) of unidentified alpha activity was used for comparison with TLLa monitoring results.

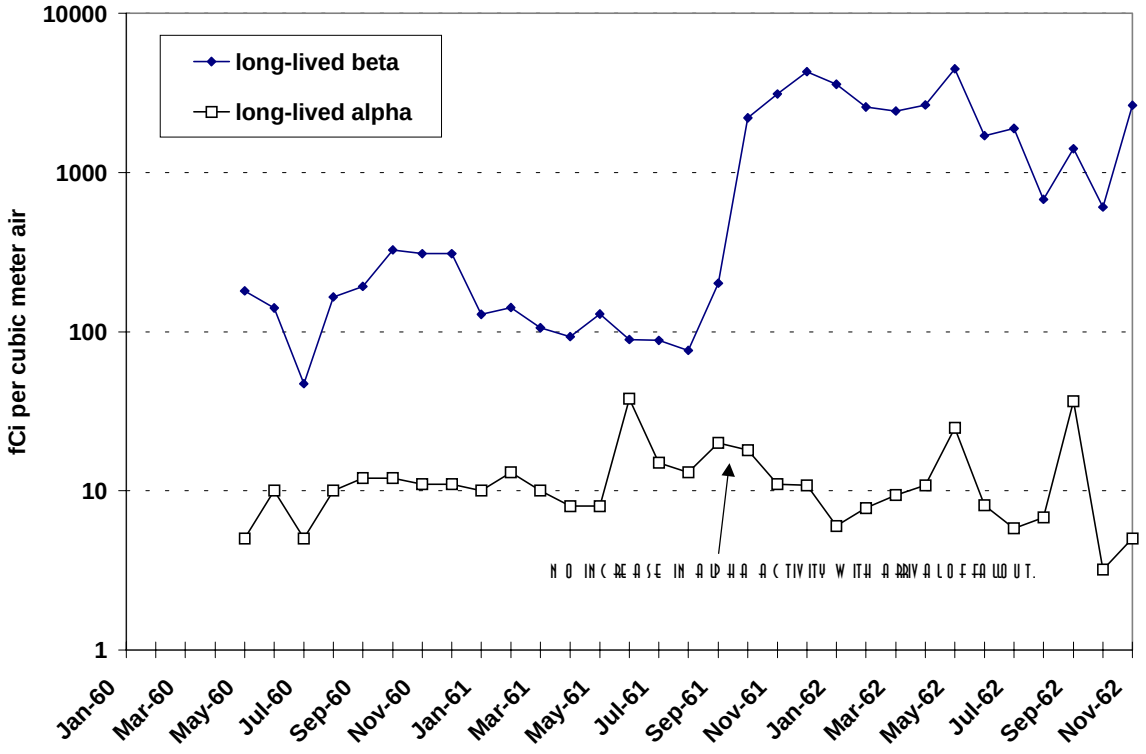


Figure III-33. Monthly average concentrations of long-lived beta and alpha activity in air at the Coal Creek air monitoring station, 1960–1962. The air filters at this station were changed daily. Data were obtained from the contractor's monthly site survey reports. Arrival of fallout in the fall of 1961 is clear from the increase in long-lived beta activity in air. There is no corresponding increase in long-lived alpha activity.

Beginning in January 1960, the onsite and offsite air-monitoring data (alpha and beta, 4-hour and 1-week counts) were presented in table form in the monthly site survey reports. There are basically five location groupings: (1) onsite (all stations combined), (2) Coal Creek, (3) Marshall, (4) an offsite group consisting of S-15, S-16, S-17, and S-18 (Boulder, LaFayette, Broomfield, and Wagner), which we call "offsite group S-1x", and (5) an offsite group consisting of S-20, S-23, and S-25 (Golden, Westminster, and Denver), which we call "offsite group S-2x." Group S-2x is further from the site than group S-1x. The distances to each station were given earlier in this chapter. We examined data for a 19-month period (May 1961 through November 1962) during which there were data for all stations. Those data are plotted in Figure III-34. Annual average concentrations for the five groups in 1960–1962 are shown in Table III-11.

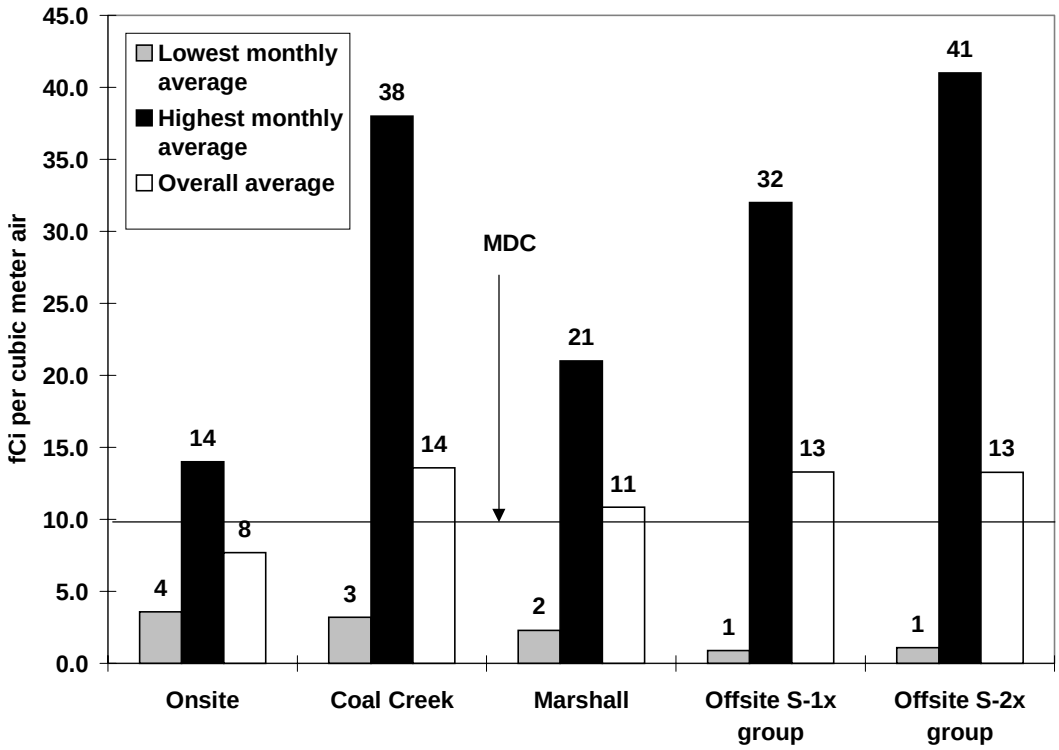


Figure III-34. Monthly average concentrations of total long-lived alpha activity in air, May 1961–November 1962.

Table III-11. Annual Average Concentrations (fCi m^{-3}) of Total Long-Lived Alpha Activity in Air at Five Locations around Rocky Flats in 1960-1962 (Dow monitoring data)[a]

Year	Onsite	Coal Creek	Marshall	Offsite group S-1x	Offsite group S-2x
1960	9.4 ± 3.7	9.5 ± 2.9	na	na	na
1961	10.0 ± 2.7	14.6 ± 8.3	13.7 ± 4.0	18.7 ± 10.3	17.9 ± 15.4
1962	6.1 ± 3.0	11.2 ± 10.1	8.0 ± 4.6	8.8 ± 5.6	9.9 ± 8.0

[a]Values in table are the mean and standard deviation of monthly averages reported in that year.

Most of the data were actually reported as a less-than-detectable concentration, due to the site practice at the time of reporting an average with a "<" designation even when only one or two of the individual measurements making up the average were not detectable. In compiling the statistics for Table III-11 and Figure III-34, we treated all reported concentrations as if they were detectable. Based on examination of more detailed records, discussed in the next section, the minimum detectable concentration was 9 fCi m^{-3}, very close to the measured concentrations. Basically, long-term average concentrations of TLLa activity in air in 1960–1962 were similar all over the region. If anything, concentrations appeared to be higher at offsite locations than onsite.

The Dow monitoring information in the environmental surveys was also summarized and published periodically in the PHS publication, *Radiological Health Data and Reports* (various titles). Annual environmental reports were published by Dow beginning in 1971.

Figure III-35 presents the all-station average for TLLa in offsite air samples reported by Dow from April 1960 through June 1969. These monthly averages were taken directly from the environmental survey reports. Various statements are made in the environmental survey reports to the effect that the offsite monitoring results are believed to be background from natural radioactivity. For example, the typical statement was: "There is no evidence that the alpha activity is greater than that occurring naturally in the area." (Environmental Survey, October–December, 1961).

As discussed earlier in this chapter, we estimated a typical average TLLa concentration from natural radionuclides, using two different approaches, as around 1.4 fCi m^{-3} TLLa activity. It is recognized that this value represents a long-term average estimate. An individual daily background concentration might be higher, perhaps up to a factor of 5 or more, or around 7–10 fCi m^{-3}. An individual monthly background value was not expected to be more than a factor of 3 higher than the long-term average concentration (i.e., about $1.4 \times 3 = 4$ fCi TLLa m^{-3}). Practically speaking, however, measured values of TLLa in air were higher than our estimated natural background level. Hammond et al. (1969) stated that naturally occurring alpha radioactivity had averaged 5 fCi m^{-3}. Michels (1973) indicated that CDH measurements in the Denver area range up to 5 fCi m^{-3}. The highest concentrations in Figure III-35 could represent concentrations higher than expected from naturally occurring alpha emitters or fallout. However, earlier in this chapter, we presented data and conclusions from the CDH monitoring of TLLa activity in air that showed occasionally quite high TLLa measurements attributed to naturally occurring ^{210}Po.

The very limited information in the environmental survey reports provided no opportunity to assess various aspects relating to the quality of these data. Fortunately, some original data sheets were located, which are discussed in the next section.

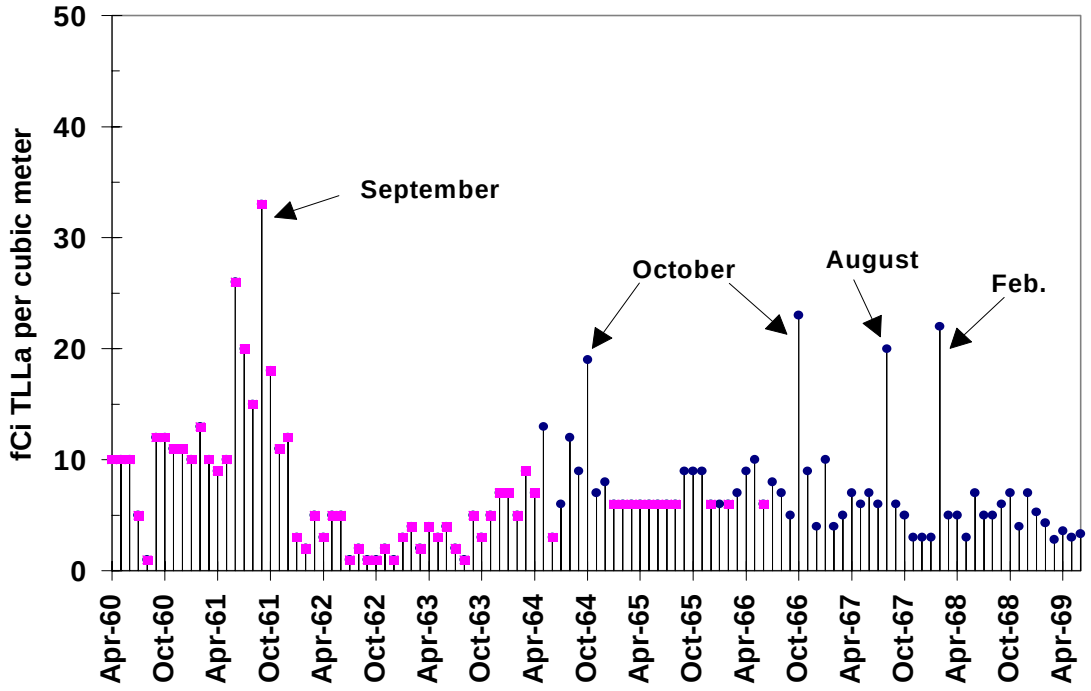


Figure III-35. Monthly average long-lived alpha concentrations in air at offsite monitoring stations (all locations combined), as reported by Dow in the environmental survey reports. Points plotted as squares were reported as "less-than" values; whereas those plotted as circles were not. The minimum detectable concentration for offsite air sampling at this time was around 9 fCi m^{-3}.

Examination of Selected Original Data Sheets for TLLa in Offsite Air from the 1960s

We located some original data sheets in the Federal Records Center for the community air samplers for the years 1966 (first three quarters), 1968, 1969, 1970, 1971, and 1973. These data allowed us to examine the individual station results, as well as other important details involving the data quality, e.g., the duration of sampling, volume of air sampled, and variation in counter background. The time period included the period of peak releases from the 903 Area based on other information sources like the onsite air samplers. The nine offsite sampling locations during this period were Coal Creek (S-11), Marshall (S-13), Boulder (S-15), Lafayette (S-16), Broomfield (S-17), Wagner (S-18), Golden (S-20), Custom House (in Denver) (S-23), and Westminster (S-25).

We chose to examine the data for 1969 first, because this was the peak year for 903 Area releases, based on onsite air data. The 1969 data for two offsite samplers southeast of the RFP, Wagner School (S-18) and Westminster (S-25), were entered into an EXCEL spreadsheet from the original data record sheets. The actual time and volume of air sampled were recorded by the sampling technician on the data sheet for each sample. During 1969, although the sampling duration was most often 7 days, the samplers were purposefully programmed to operate intermittently (5 minutes per hour), presumably to prevent loading of the filters with particulates,

pump failures, and other sampler malfunctions. Thus, the actual operating time was typically around 14 hours per week, or less than 10% of the time. This short time and the low flow rate of these samplers produced a small sample volume, on the order of 40 to 50 m^3 per sampling period (usually one week). This is actually less than the volume sampled in daily measurements made onsite (81.5 m^3).

A small air sample volume resulted in a small amount of particulate material being drawn into the filter. The net sample count rates for filters in 1969 were very low, ranging from 0 to 0.7 cpm after subtraction of counter background. The counter background count rate itself ranged from 0 to 0.9 cpm during this period. In fact, only 10 of the gross counts in 1969 were above a 90% confidence range for counter background.

The monitoring of total long-lived alpha activity in air suffered from two important limitations:

- **It was not specific for alpha-emitting nuclides of interest**
- **It could not measure low levels accurately.**

After examining the variability in counter backgrounds recorded on the original data sheets, we concluded that quantitative use of these data was not warranted because the majority of the gross counts were within the noise range of counter background. The contractor alpha count method at this time suffered not only from nonspecificity (for plutonium) but also poor sensitivity. Figure III-36 illustrates the considerable scatter of the community air sample counts and the high MDC relative to fallout plutonium levels in Denver at this time.

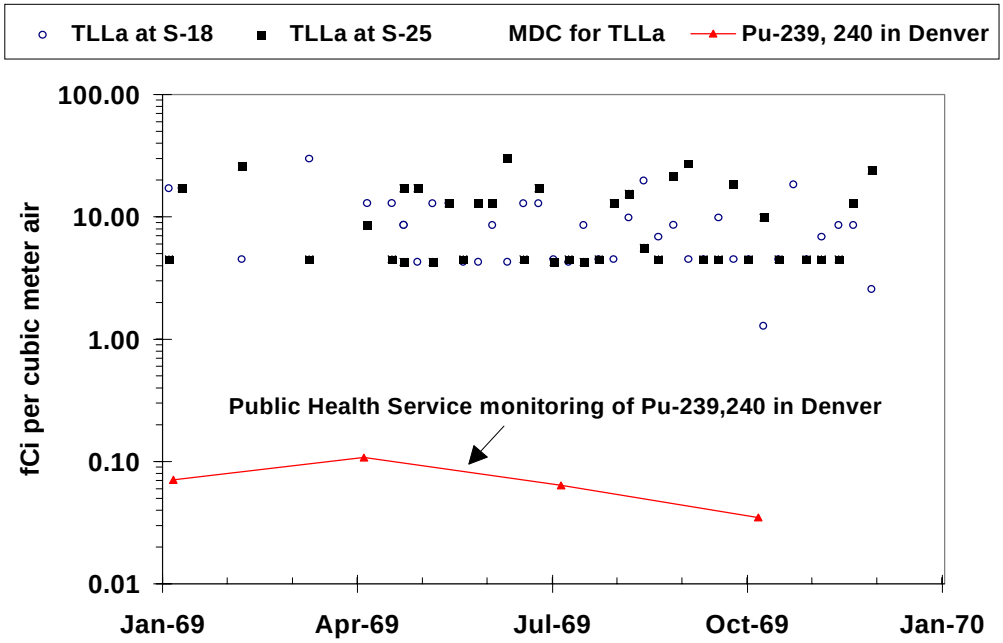


Figure III-36. Results of community air sampling and analysis for long-lived alpha activity in 1969. Sampler S-18 is southeast of Rocky Flats at Wagner School, and Sampler S-25 is further southeast at Westminster. The sensitivity of the analysis was poor, because of small sample volumes, short counting times, and variable counter background.

The MDC, based on a net MDA of 0.21 cpm and a sample volume of 50 m^3, is

$$\text{MDC for offsite samples (fCi m}^{-3}) = (0.21 \text{ cpm})/ [(0.21 \text{ cpm dpm}^{-1}) \times (2.22 \text{ dpm pCi}^{-1}) \times (1 \times 10^{-3} \text{ pCi fCi}^{-1}) (50 \text{ m}^3)],$$

or about 9 fCi m^{-3}. In 1970 and 1971, the community samplers operated about twice as long (10 minutes per hour), producing sample volumes on the order of 100 m^3. This is consistent with an MDC of 4.5 fCi m^{-3}, as reported by Boss et al. (1973) for community air sampling in 1972.

Based on the evaluation of S-18 and S-25 data for 1969, compilation and plotting of community air sampling data from other stations and times did not appear warranted. However, the original data sheets for 1966, 1968, 1970, 1971, which we had located, were scanned for relatively high alpha counts in other years at all samplers. There were over 1,400 offsite air samples collected and analyzed during this period. Only 25 samples, or less than 2%, produced a TLLa activity of 1 cpm or more. These high-count days are included in Appendix B (Table B-7).

Highlights and Conclusions: Monitoring for Total Long-lived Alpha Activity in Community Air (1966–1971)

- Original records were obtained for 1966–1971, when other information sources pointed to high 903 Area releases.

- The sampling and analysis technique for TLLa in community air suffered from poor sensitivity (high detection limit), because of short count times, small volumes of air sampled, and variable counter background. All but two gross sample counts from the Wagner and Westminster stations in 1969 were within the range of counter background.

- Air sampling was not continuous (<10% of time until 1970), and analysis was not specific for contaminants of interest.

- The sampling and analysis technique was not adequate to measure fallout plutonium or naturally occurring alpha emitters under typical dust loading conditions.

- Quantitative use of these data for the historical public exposures studies is not recommended.

ROUTINE MONITORING OF PLUTONIUM IN AIR BY THE RFP CONTRACTOR (AFTER 1970)

In 1970, the Rocky Flats environmental monitoring program began to include routine monitoring for plutonium in air. There was a transition period until about 1975, when routine sampling was conducted in a consistent way at the various locations. Unfortunately for the assessment of historical exposures to the public, this is after most of the releases of plutonium are believed to have occurred. However, information may be gained from examining recent monitoring that can be applied to assessing releases from the earlier period.

As of 1990, there were 51 samplers in the ambient air sampling network. Twenty three were located on the RFP site near the main industrial areas; 14 were perimeter samplers, which bordered the RFP along major highways on the north (Highway 128), east (Indiana Street) south (Highway 72), and west (Highway 93); and 14 were community samplers in adjacent metropolitan areas.

Application of the Framework Exercise to Routine Monitoring of Plutonium in Air

We began our analysis of the utility of the recent air monitoring data by asking the question, "Were concentrations 'onsite' generally greater than concentrations measured 'offsite'?" (See Chapter I, Figure I-3, for logic flow diagram.) This question was easily answered by a quick look at data reported in a relatively recent annual environmental report (Costain et al. 1991). Figure III-37 illustrates the mean plutonium (^{239,240}Pu) concentrations in air at onsite, perimeter, and community stations in 1986 through 1990.

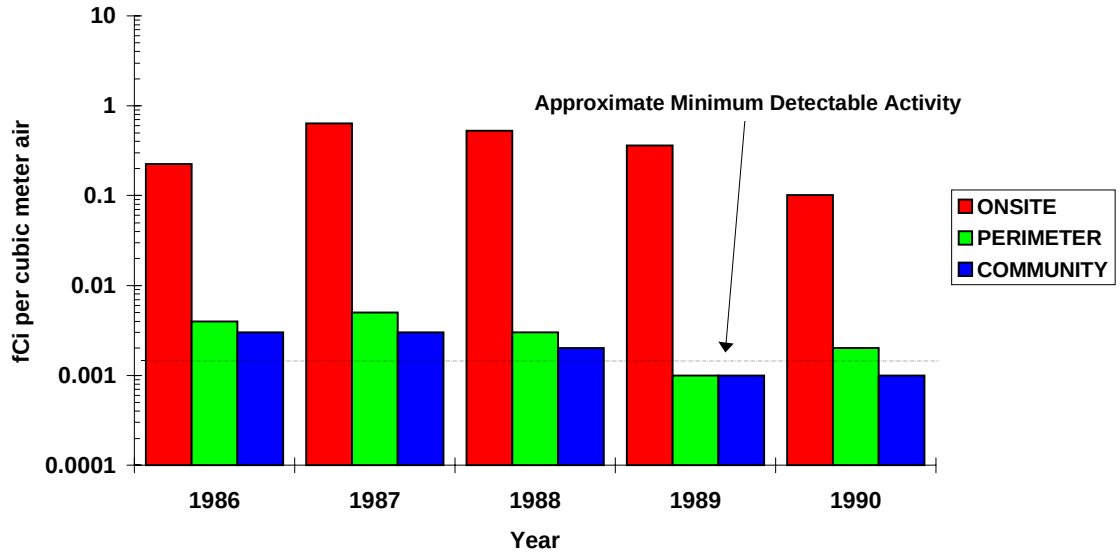


Figure III-37. Mean plutonium concentrations in air at onsite, perimeter, and community stations in 1986 through 1990. One fCi m^{-3} is equal to 1×10^{-15} µCi mL^{-1}, which is the unit used to report concentrations in these annual environmental reports.

The onsite average in Figure III-37 is based on five locations (S-5 through S-9) that have historically shown the highest plutonium concentrations for the sampling network (Setlock and Barr 1987). The location of these onsite samplers is shown in Figure III-38. Onsite samples show significantly higher plutonium concentrations than perimeter and community station samples and are well above detection limits, which were quoted as either 0.002 or 0.003 fCi m^{-3} during this period. Our initial look at recent environmental monitoring data suggested that they may provide useful insights for evaluating the spatial distribution of contaminants around the RFP. Had all the concentrations from the different location groups been similar and/or not detectable, we would not have recommended looking into the recent data any further.

Long-term Trend of Plutonium in Air, Monitored by the RF Contractor, Compared to U.S. Background

After this first look at very recent monitoring, we developed a similar plot based on ambient air monitoring for plutonium back to the early 1970s, which is the earliest that these routine isotope-specific data are available from contractor monitoring. Using the Costain et al. (1991) practice as a precedent, we plotted the average concentrations monitored at locations S-5 through S-9 (onsite), along with the annual average concentrations from perimeter and community stations (Figure III-39). The averages were obtained from the annual environmental reports and are tabulated in Table B-10 of Appendix B. The majority of the annual averages for perimeter and community groups before 1980 were reported as less than the value plotted. We were careful to use the *same five* onsite station locations even though the numbering system for the air monitoring stations changed during expansion of the monitoring program in the early 1970s. For example, the S-7 sampler in 1975 was actually S-8 before that time (see Table B-9 for documentation of the change in sampler numbers and maps of locations in Appendix B).

The plutonium concentrations in perimeter and community samples (Figure III-39) appear to be quite similar to each other from the mid-1970s onward. The difference between onsite and offsite samplers is evident throughout this two-decade period. It is interesting to note the increasing spread between the offsite and onsite concentrations in recent times. Whereas the offsite sampler concentrations decreased by 100 times over this time period, the onsite sampler concentrations changed by less than a factor of 10. The reason for the time trend became clearer to us after examination of fallout trends, which were discussed in the first section of this chapter. The perimeter and community station time trend is basically reflecting the decrease in worldwide fallout, whereas the onsite stations are still being affected by plutonium from Rocky Flats, probably due to localized resuspension of contaminated soil.

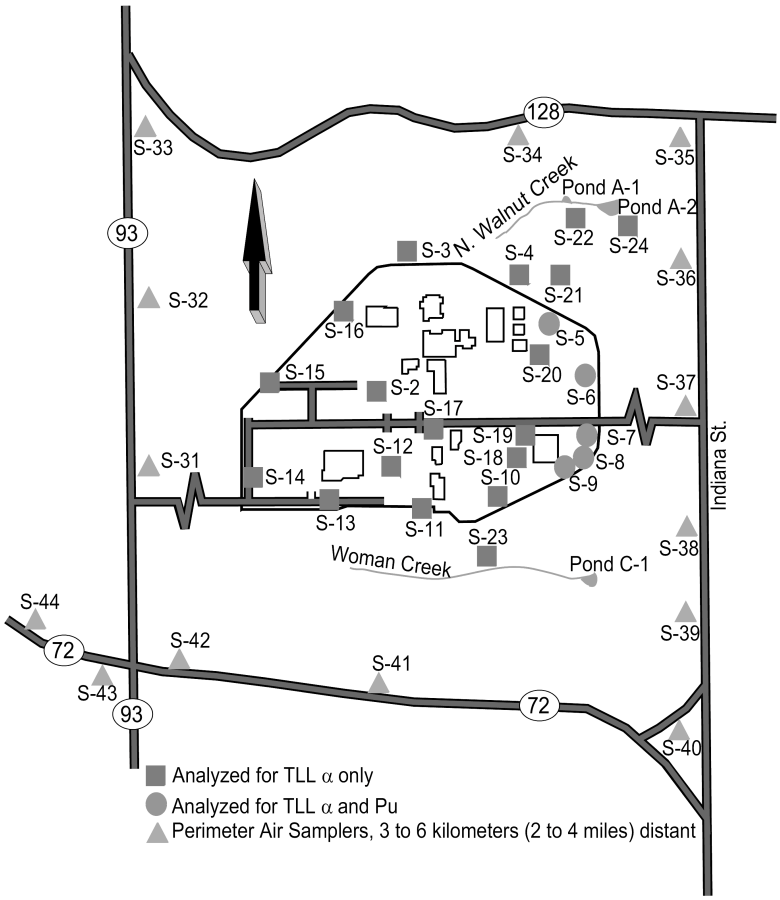


Figure III-38. Location of routine ambient air monitoring stations (onsite and plant perimeter) in 1986. Portions of the map are not to scale. "TLL∝" indicates total long-lived alpha radioactivity. Only five of the onsite air samplers, on the eastern security fence, were routinely analyzed for plutonium at this time. For a period in the mid-1970s, the majority of the onsite samples were analyzed for plutonium.

The community stations in 1973 and 1974 appear to be elevated (Figure III-39). However, this could be due to sampling differences. During the 1970–1975 expansion and transition period for the air monitoring program, the perimeter and community stations were not using the same equipment. Specific information regarding sampler types and location, analytical protocols, and analytical results have been summarized since 1971 in the RFP annual environmental monitoring reports (Dow et al. 1971–1990). *RAC* has compiled the most important information for our purposes into a detailed matrix (Table B-8 of Appendix B). The perimeter stations, which were newly installed in 1971, were high-volume air samplers. The old low-volume samplers at community stations were not upgraded to high-volume samplers until around 1975. Whereas the MDC for the high-volume samplers (perimeter stations) in 1974 was 0.008, the MDC for the low-volume samplers (community samplers) was 0.04 fCi m^{-3}. The samplers were also using different

filters and presumably different weather shelters. These and other issues affecting data quality and interpretation are discussed in the final section of this chapter.

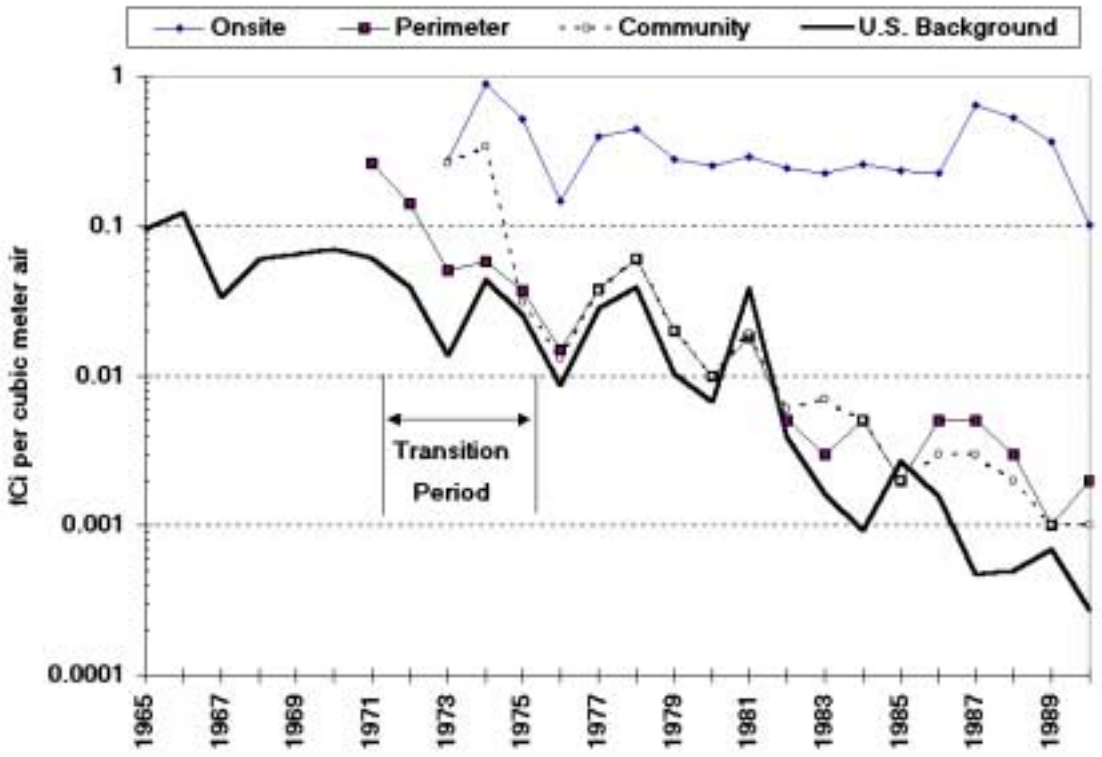


Figure III-39. Annual average concentrations of plutonium in ambient air at community, perimeter, and five onsite locations between 1971 and 1990 (RFP contractor monitoring). Rocky Flats data used to construct this figure are in Table B-10, Appendix B. The monitoring program was in a state of transition until 1975, when consistent sampling and analysis methods were used for perimeter and community stations. Most of the annual averages before 1980 were reported as less-than values. The U.S. background line is the average for Denver Colorado, Pierre South Dakota, and New York City, which were plotted separately in an earlier section of this chapter (Figure III-6). The minimum detectable concentration (MDC) has been between 0.002 and 0.01 for contractor monitoring since 1975. Thus, their method could not measure levels as low as U.S. background concentrations in the last part of this time period.

Figures III-37 and III-39, which illustrate that onsite concentrations of airborne plutonium are generally higher than offsite concentrations, suggested that the ambient air monitoring data from the 1970s and 1980s would be useful for evaluating the spatial distribution of plutonium released from Rocky Flats according to our framework exercise. For example, the data might provide insight into the dispersion of plutonium from the 903 Pad area during earlier time periods. Contaminated soil areas, which were primary contributors to airborne contamination in the 1970s, were within several hundred meters of the asphalt pad that covered the former barrel storage area.

Spatial Distribution of Plutonium in Onsite Air (mid-1970s)

Monthly measurements of plutonium in onsite air are reported in RFP contractor reports (Dow et al. 1970–1990). Individual sample results for each location are given.

We chose a 3-year period, 1975–1977, to do a preliminary examination of the spatial distribution of plutonium concentrations in ambient air for two main reasons. First, the percentage of samples that were below the detection limit was very low during this period, less than 1% in most cases. Second, the plutonium analysis of onsite samples had not yet been reduced to the five samplers on the eastern security fence (S-5 through S-9), so there were more locations on the RFP site in which the plutonium measurements could be examined.

Figure III-40 shows the spatial distribution of plutonium in onsite samples, expressed as the average ratio of the concentration at sampler S-n to that at sampler S-6 (at the east security fence) over the 3-year period. There was a 50-fold variation in average concentrations over this area. Concentrations were highest on the eastern part of the site. Plutonium concentrations in air collected in the western half of the onsite fenced area were generally less than 10% of those measured at the east security fence.

The annual average concentrations of plutonium in onsite air for 1973 through 1977 are given in Table B-11, Appendix B. As discussed previously in this chapter, background plutonium from weapons testing has been widely distributed throughout the world. Most, but not all, of the concentrations of plutonium at onsite locations in 1973 through 1977 were well above fallout levels (Table III-12). Figure III-40 illustrates the spatial distribution of plutonium in onsite air during a time when a number of the onsite location samples were analyzed for plutonium and in which most of the samples were above detection limits.

Table III-12. Number of Onsite Air Sampler Locations Where Annual Average Plutonium Concentration Was Greater than Two Times Background[a]

Year	Number of onsite samplers having plutonium annual average concentration	Number of onsite sampler locations with plutonium concentrations greater than two times background
1973	12	12
1974	15	14
1975	24	13
1976	24	22
1977	8	8

[a] Background was determined based on annual averages for three U.S. cities: Denver, Colorado; Pierre, South Dakota; and New York City (see previous section of this chapter).

Spatial Distribution of Plutonium in Offsite Air (mid-1970s)

In February 1971, a new network of 12 continuous high-volume air samplers were installed at the Rocky Flats perimeter at distances in the range of about 2 to 4 miles (Boss et al. 1972). The sample locations are illustrated in Figure III-41. Airborne particulates were collected daily on Whatman 41 filter paper at an average flow rate of 20-25 cfm. Filters were composited and radiochemically analyzed for plutonium following isolation by ion-exchange. An internal standard of ^{236}Pu tracer was used to determine chemical yield, and alpha pulse-height analysis was used to determine the quantity of plutonium present in the sample.

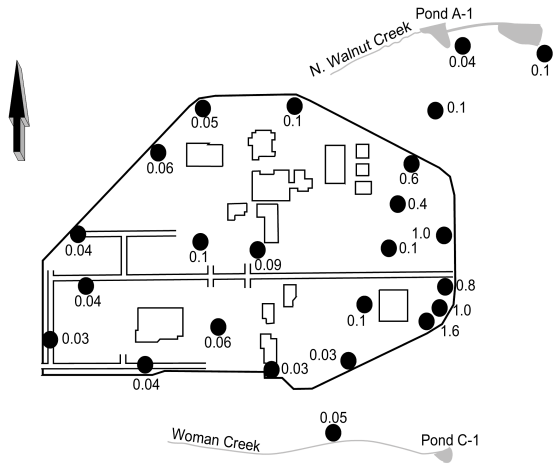


Figure III-40. Spatial distribution of plutonium in air (onsite) in 1975–1977 expressed as the average ratio of the concentration at each sampler to the concentration at sampler S-6. Background (fallout) plutonium levels were not subtracted before determining the ratio. Portions of the map are not to scale.

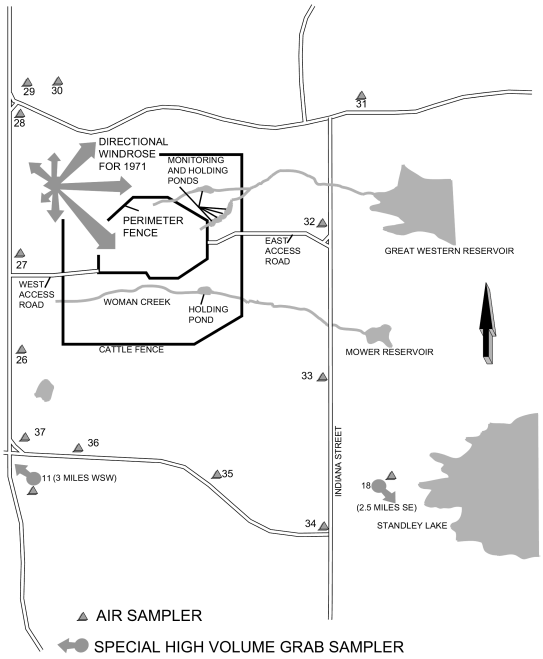


Figure III-41. Location of offsite (perimeter) air samplers in 1971. Note the sample locations and numbers were different than in 1986 (Figure III-38).

Boss et al. (1972) indicated that intended improvements for 1973 included changing from Whatman to Microsorban filter media and electrodeposition of all environmental plutonium air samples. Electrodeposition would more accurately quantify the plutonium through increased spectral resolution of the alpha pulse-height analysis. This would also permit discrimination of ^{239,240}Pu from ^{238}Pu.

This monitoring at the Rocky Flats perimeter represents the first plutonium-specific monitoring of air by the contractor at points of public access. Boss et al. (1973) provided a table of the geometric mean (see glossary) concentrations for each week in 1972. All perimeter sampling locations were combined into this weekly average. The data are plotted in Figure III-42 and compared to PHS monitoring data from Denver and Pierre, South Dakota. Concentrations at the RFP perimeter appear to be elevated above fallout levels in the first quarter of 1972, but no statistical test was applied to the data.

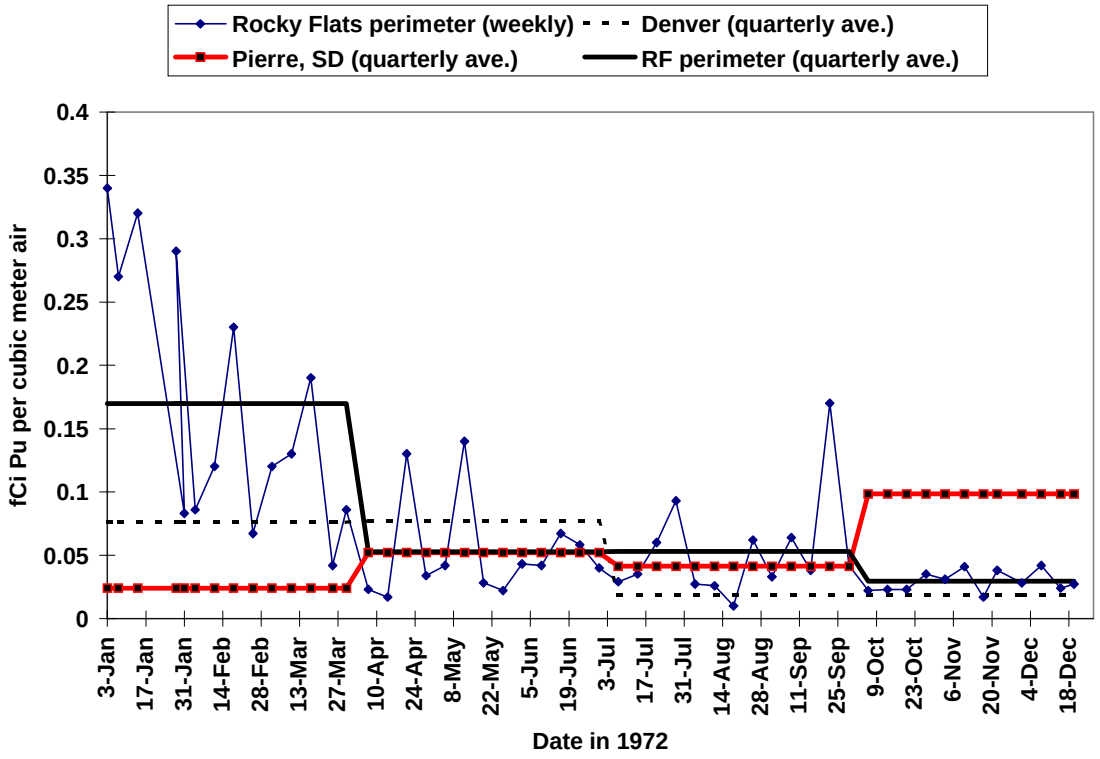


Figure III-42. Comparison of plutonium in air at the Rocky Flats Plant perimeter with concentrations monitored by the Public Health Service in Denver, Colorado and Pierre, South Dakota, in 1972. The Public Health Service data are quarterly averages. The weekly Rocky Flats data are the geometric average of all perimeter stations for that week (computed in Boss et al. 1973). A quarterly average concentration for the Rocky Flats perimeter was estimated (by us), by computing the arithmetic average of the weekly concentrations within that quarter. This permits a more valid comparison to the Public Health Service concentrations. Rocky Flats perimeter concentrations appear to be elevated above fallout levels in the first quarter of 1972 but not for the rest of the year.

The data tabulated in Boss et al. (1973) were from all perimeter stations combined. We compiled data for two perimeter stations (one upwind [S-26] and one downwind [S-32]) and the 903 Area from the start of monitoring at each location through the end of 1972. The data are plotted in Figure III-43, and descriptive statistics are presented in Table III-13. As we have seen in many other environmental data sets, the plutonium concentrations in air near the 903 Area were definitely elevated. However, it would be difficult to distinguish statistically between the concentrations in the upwind and downwind direction at the Rocky Flats perimeter. In a statistical analysis of ambient air monitoring data from 1982 through 1985, Longbotham and Lawton (1986) concluded that the major factors affecting ^{239}Pu concentrations at perimeter and community air sampling stations over that time were similar for all samplers, implying a global as opposed to random or local effect. Only sampler S-37 (previously called S-32), on the perimeter at the east entrance to the plant, had a mean time-adjusted concentration that was significantly larger than all other sampler means.

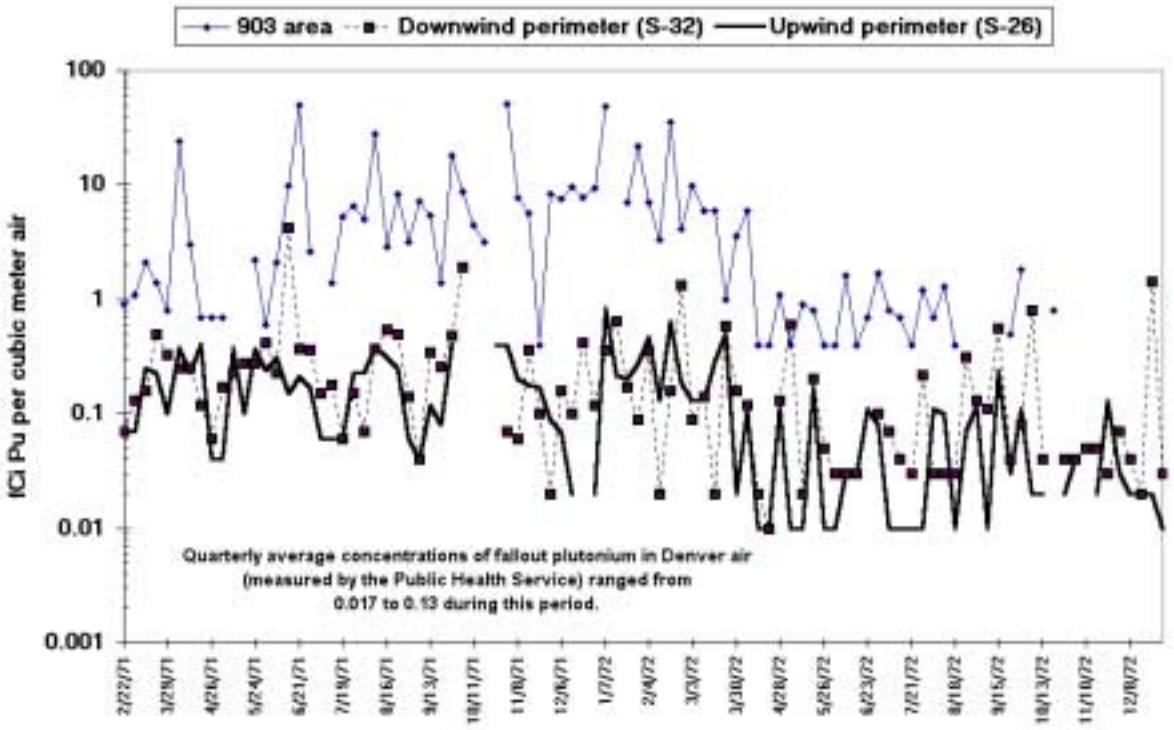


Figure III-43. Weekly measurements of plutonium in air between February 1971 and December 1972 at the 903 Area and at the Rocky Flats perimeter in a downwind and an upwind direction. For a few of these weeks, there was no sample at S-32, and so the value from S-33 (also downwind but more southeast) was used (see Figure III-41). The value plotted for the 903 Area was the maximum of measurements made at five different heights between 5 and 25 feet above ground (see later section of this chapter for description of this monitoring). For 903 Area samples, minimum detectable concentration (MDC) was about 0.4 fCi m^{-3}. For perimeter samples, the MDC was 0.01 fCi m^{-3}. Actual sample results greater than the MDC were often reported as "less-than" values, presumably because of low chemical recoveries.

Table III-13. Descriptive Statistics for Weekly Plutonium Measurements in Air at the Rocky Flats Perimeter between February 1971 and December 1972

	Downwind at Indiana Street (S-32)	Upwind at Highway 93 (S-26)
Arithmetic mean ± standard deviation	0.27 ± 0.52	0.15 ± 0.16
Range	0.01 – 4.3	0.01 – 0.84
Geometric mean ×/÷ geometric deviation	0.13 ×/÷ 3.3	0.08 ×/÷ 3.6

The air monitoring data from the contractor monitoring station directly east of the site at Indiana Street (station S-32, later called S-37) were used, along with data from the CDH and HASL, to compare to predicted concentrations in air from all Rocky Flats releases during the 1970s and 1980s (Rood and Grogan 1999c). Fallout background was subtracted from the annual average measurements before making the comparison with model-predicted concentrations. The data used in this assessment are compiled in Table B-15 of Appendix B.

SPECIAL (NONROUTINE) ENVIRONMENTAL MONITORING OR STUDIES

Special monitoring studies were carried out over a shorter term than routine monitoring, usually less than several years. Those studies that seemed likely to be particularly important to evaluating historical public exposures are reviewed here. Because they are so closely linked with the resuspension process, most studies addressing the particle size of resuspended plutonium from contaminated soil areas, have been evaluated in the 903 Area source term report (Weber et al. 1999) or the corresponding risk assessment report (Rood and Grogan 1999d). Studies that related to the quality of the air monitoring data are included in the "Data Quality Issues for Ambient Air Monitoring" section later in this chapter.

Vertical Distribution (Profile) of Plutonium in Air

Studies to investigate the vertical distribution of plutonium-contaminated soil particles in air were conducted by the Rocky Flats contractor and others in an attempt to quantify erosion processes and local plutonium movement. The vertical profile would help in defining how far plutonium-carrying dust could be airborne, since it must rise a minimum height above the ground before being caught by the wind and carried further distances (Langer 1986b).

The sources of the plutonium or the plutonium-contaminated soil particles during these vertical profile studies were areas around the original oil storage area, which was covered with asphalt in 1969 (903 Pad). These remaining contaminated soil areas are discussed in Chapter VIII of this report. The particles of plutonium suspended in the waste oil stored on the 903 Pad were relatively small, because oil was filtered prior to storage to remove particles greater than 2 to 3 microns (μm) in physical size (Little and Whicker 1978; Langer 1986b). This physical size is equivalent to aerodynamic diameters ("AD") of 7 to 10 μm, using a plutonium density of 11.5 for PuO_2. McDowell and Whicker (1978) showed that the equivalent sizes of plutonium particles in contaminated soil, collected approximately 200 m southeast of the 903 Pad, were between 0.2 and 0.3 μm. However, if small particles of plutonium became attached to larger ambient soil or dust particles, the soil particle density (around 2.4 g cm^{-3}) would have dominated the particle's aerodynamic properties. Langer (1991) indicated that plutonium had attached itself to host soil particles ranging in size from a few microns to millimeters in diameter.

Three studies are reviewed in this section:

- Measurements were made by the RFP contractor (Dow) from February 1970 to April 1972 and were reported monthly in from May 1970 to June 1972 (Dow 1970–72). A handwritten logbook containing some of these data was also found (Anonymous 1970–1971), which corroborated the data published in the monthly reports and provided additional data for the February 27, 1970, through May 20, 1970 time period.

- A field experiment was conducted by researchers from Hanford, Washington during July 1973. Objectives were to "determine vertical airborne concentrations of plutonium and plutonium attached to the mass of airborne soil, and particle size distributions of plutonium and associated airborne soil at three sampling sites, and to relate measurements to meteorological variables and surface contamination" (Sehmel and Lloyd 1974). Although additional analysis was done on the samples collected during this experiment, many questions remained unanswered.

- From November 1982 to August 1985, a third study was carried out by the RFP contractor (Rockwell) to "understand and quantify the physical processes that lead to the resuspension of soil particles contaminated with plutonium," and to continue the development of a resuspension model (Langer 1987). This work looked more closely at the particle size distribution and the limitations of the sampling equipment.

Dow Monitoring of Vertical Profile of Plutonium in Air

From 1970–1972, special air samples were taken about 250 ft east of the 903 Pad for determination of ^{239}Pu concentration in air at different heights above ground (Dow 1970–1972, Anonymous 1970–1971). High-volume samplers were suspended by ropes and pulleys at 5-foot intervals on a pole to a height of 25 ft. The pole was set in line with the north-south center of the pad. Four-inch Whatman 41 filters were used. Concentrations reported as not detectable ranged from 0.1 to as high as 3 fCi m^{-1}.

A complete listing of the data can be found in Table B-12 in Appendix B, and the additional data (from February 27, 1970, until May 20, 1970) have been indicated. The logbook (Anonymous 1970–1971) noted that during the initial 2 months of sampling there were a number of unusually high values, when compared to the usual level reported during the following months. Although these have been included with the complete set of data, they may be artifacts of the beginning of an experimental program.

Except for around 20 samples collected at 25 ft, 80 to 100 samples were collected at each height (Table III-14). Roughly 20–30% of the reported values were below the lower limit of detection (LLD).

For the summary statistics in Table III-15, the LLD was treated as the value of the sample. This is likely an overestimate of the activity measured, but it allows the use of the total sample set without generating a complete bias toward the higher value samples. The concentrations in air measured through time at each height are presented in Figures III-44 (a-e). No trends are obvious. The highest concentrations of plutonium were seen at 5 ft above the ground, but an expected systematic decrease with height was not clearly shown. The scatter of the data is illustrated by the large standard deviations and large maximum/minimum range (Table III-15).

Table III-14. Detectability of Vertical Profile Air Samples Collected for Plutonium Analysis by Dow East of the 903 Asphalt Pad (1970–1972)

Height (ft)	Total number of samples	Number of samples less than detectable	Fraction of samples less than detectable
5	88	22	0.25
10	96	26	0.27
15	103	26	0.25
20	97	19	0.19
25	21	4	0.19

Table III-15. Summary Statistics of the ^{239}Pu Data Collected East of the 903 Pad from July 1970 to April 1972 (fCi m^{-3})

Concentration (fCi m^{-3})	5 feet	10 feet	15 feet	20 feet	25 feet
Average	24	3.9	10	5.5	14
Number of samples	88	96	103	97	21
Standard deviation	116	8.5	55	29	42
Geometric mean	1.6	1.2	1.4	1.2	1.7
Median	1.1	1.0	1.1	1.0	2.8
Maximum	847	46	532	288	191
Minimum	0.1	0.1	0.1	0.1	0.1

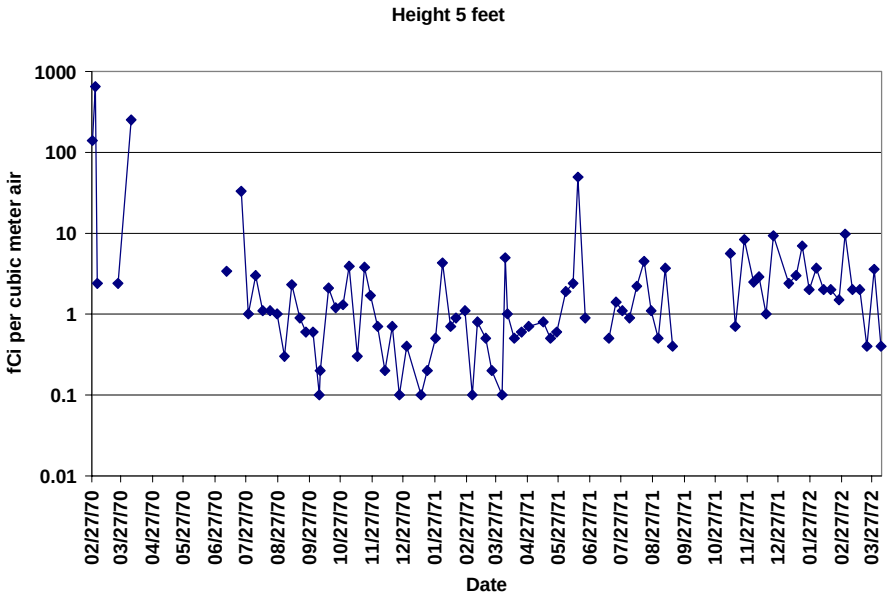
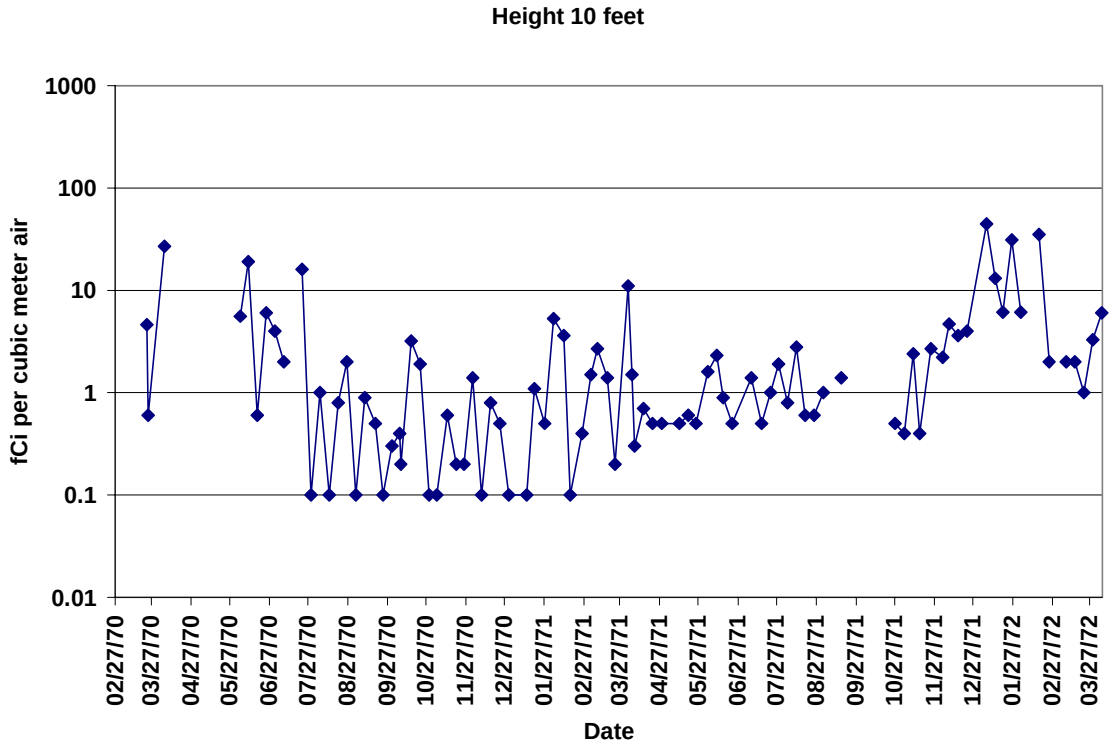


Figure III-44a. Concentrations of plutonium in air at 5 ft above the ground surface and about 250 ft east of the 903 Pad.



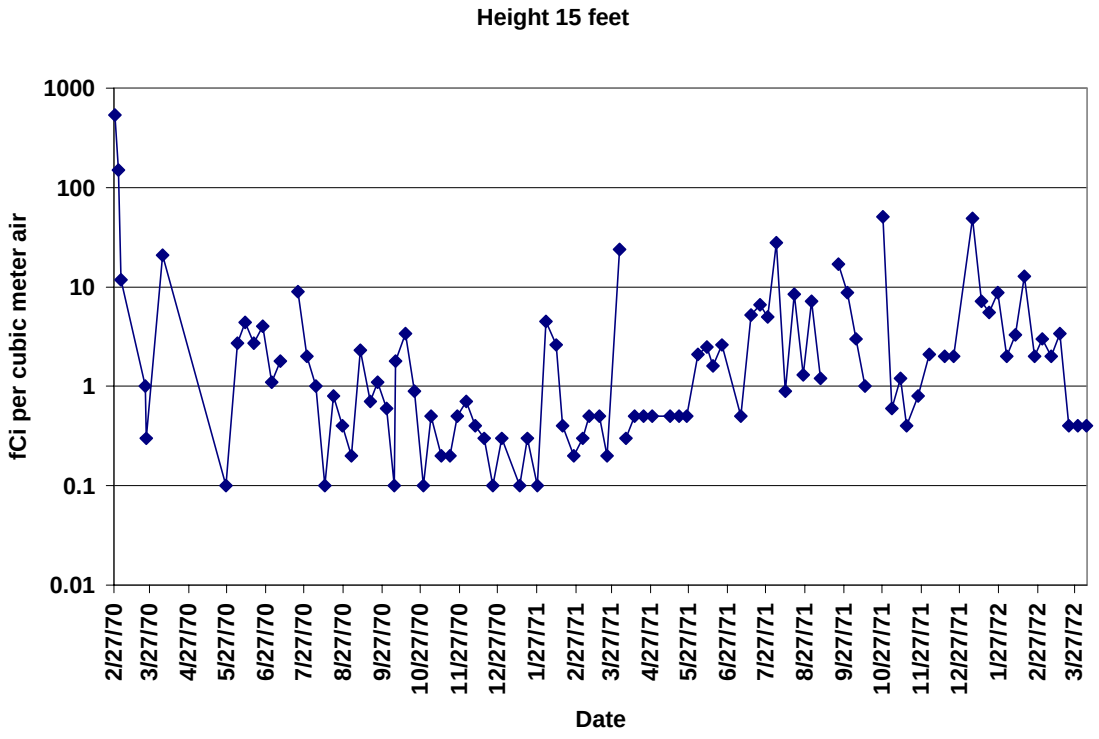
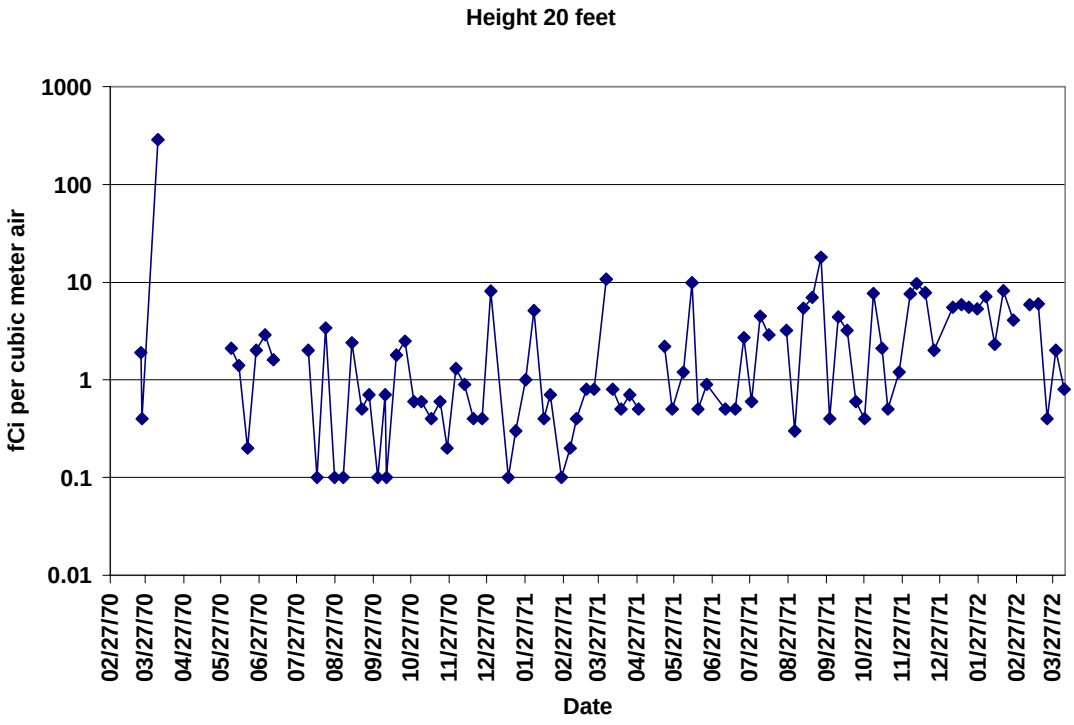


Figure III-44b and c. Concentrations of plutonium in air at 10 and 15 ft above the ground surface and about 250 ft east of the 903 Pad.



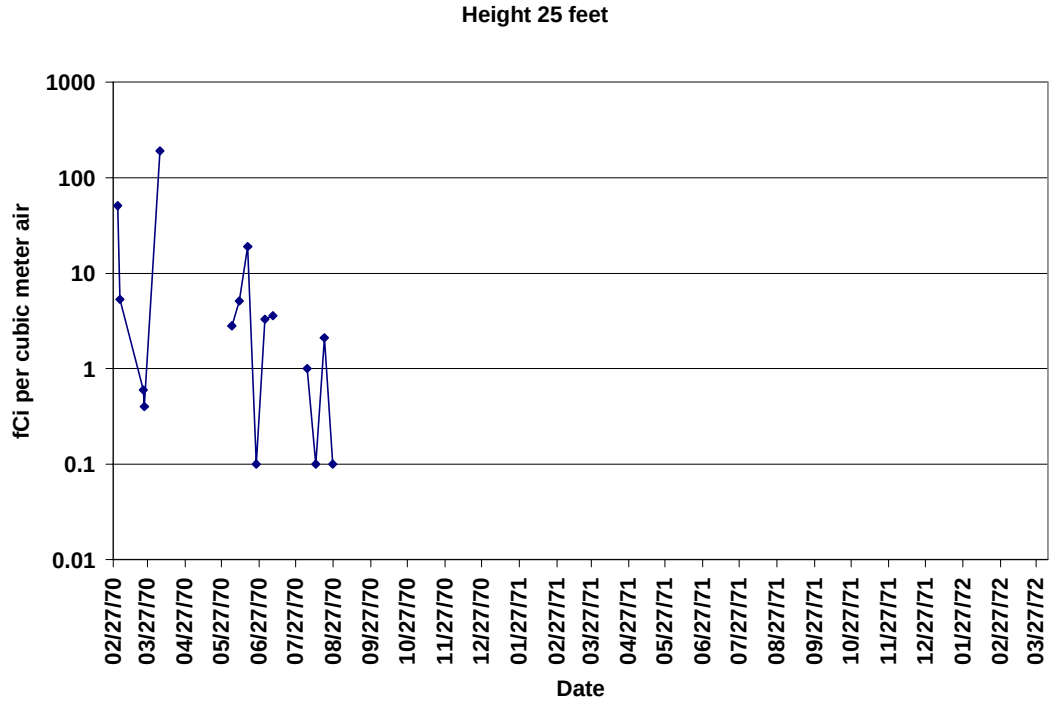


Figure III-44d and e. Concentrations of plutonium in air at 20 and 25 ft above the ground surface and about 250 ft east of the 903 Pad.

We investigated the possibility of consistent fractions of the total resuspended activity being captured at a given height by calculating the fraction of the total plutonium activity detected at each height. These data are shown in Table B-13 in Appendix B.

These same data were used by Michels (1973) to try to ascertain the height of the plume of resuspended plutonium. The sampling data for each height were averaged for each month using a total of 47 profiles (36 of which were complete). Even grouped together, no long term trends were exhibited; however in many cases the lowest and highest heights did collect the higher alpha activities. Due to the plutonium levels measured at 20 ft, it is difficult to determine the uppermost height of the plutonium resuspended into the air.

Resuspension Field Experiment by Battelle Northwest Laboratory at RFP

Experimental measurements of plutonium resuspension were made by Battelle-Northwest at three sampling sites east of the 903 Area in July 1973 (Sehmel and Lloyd 1974; Sehmel 1975; Sehmel 1976; Sehmel 1978; Sehmel 1980). One site was along the eastern security fence (A), another at the old RFP boundary along the east cattle fence (B), and the third in-between (AB), at a distance of 392 m from A and 277 m from B. High-volume cascade impactors were assembled with a rotating cowl that was sensitive to wind direction. In addition, samples could be taken at a rate of 0.57 m^3 min^{-1} for respirable particles, simultaneously with larger nonrespirable particles that would settle on the floor of the cowl. The respirable particles were separated into nominal aerodynamic diameter ranges of 1.1, 2.0, 3.3 and 7 µm. The nonrespirable samples were not usually collected in most sampling systems (Sehmel 1976), but this large size fraction was considered potentially important for a better understanding of resuspension processes. Submicron particles were collected on the impactor backup filter.

These samplers were bolted to sampling towers at heights ranging from 0.3 to 30 m. Sixteen continuous high-volume air samplers, operating continuously at 1.4 m^3 min^{-1} were also included in the experimental setup. Of the 21 cascade impactors at the three locations, five were operated for wind speeds of 4.1 to 6.3 m s^{-1}, four were operated during wind speeds of greater than 6.3 m s^{-1}, and the rest during all wind speeds. A background sampler was operated 13 km to the west.

Sehmel and Lloyd (1974), indicates that the ^{239}Pu results for the respirable particle samples had significant uncertainties from counting error (see Data Quality section) and the loss of about 20% of the plutonium during the chemical separation stage. The maximum airborne ^{239}Pu concentrations detected were 3.7×10^{-15} µCi cm^{-3} of air, 5×10^{-5} µCi g^{-1} total airborne soil, and 7×10^{-3} µCi g^{-1} for the respirable fraction of soil collected in the 2-µm impactor stage. A general trend of decreasing concentration with increasing distance eastward from the 903 Area and increasing height was expected based on simple modeling concepts. This was not always the case, as shown on Figure III-45. The relatively high concentrations seen at the 1-m height at AB, the 10-m height at B, and the 30-m height at AB seemed unusual. At 1 m, the ^{239}Pu concentration at AB was 1 to 2 orders of magnitude higher than at the other heights for this site. The higher concentration found at site B was not expected, because it was the site most remote from the source of the plutonium (903 Area).

The ^{239}Pu concentrations at site AB for all respirable size classes included some unexpected high ^{239}Pu concentrations at this distance and height (Figure III-46). These higher concentrations are likely due to the attachment of the plutonium to larger resuspended particles, clusters of particles, or individual “hot” particles (Sehmel 1980).

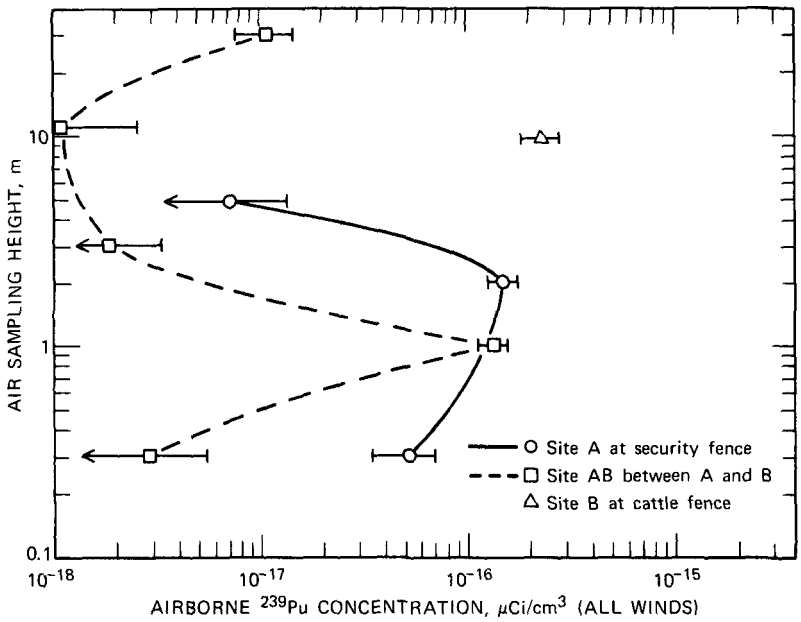


Figure III-45. Airborne ^{239}Pu concentrations from impactor 2.0 µm stage collections (for all wind speeds) (from Sehmel 1980).

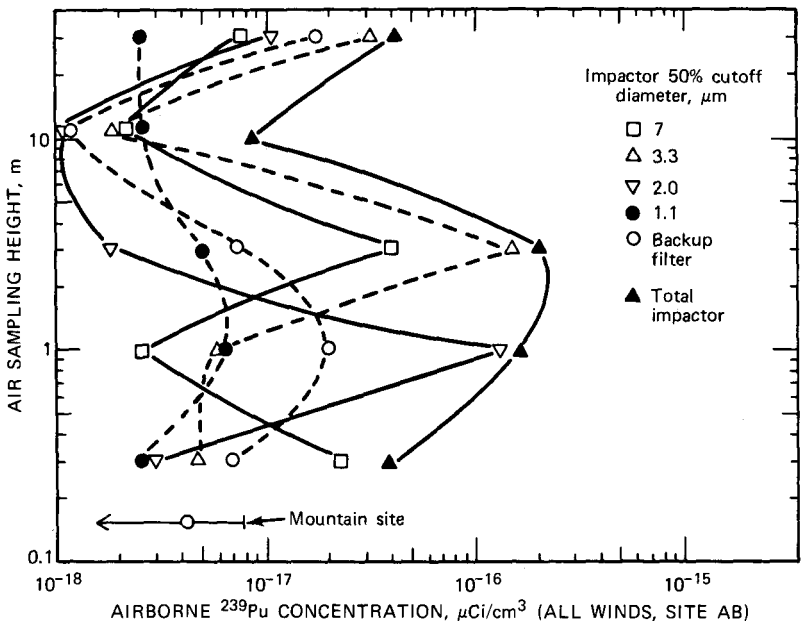


Figure III-46. Airborne ^{239}Pu concentrations, as a function of diameter, at Site AB impactor collection site and a background mountain site (from Sehmel 1980).

In the initial publications describing these experiments, the authors hypothesized that the observed results might be caused by the possibility of a secondary elevated source of plutonium. To address this, further analysis of isotopic content (^{239}Pu, ^{240}Pu, and ^{241}Pu) was carried out for seven samples obtained at elevations from 0.3 to 30 m (Sehmel 1975). Within the variation of individual samples, it was not possible to come to a definite conclusion of the likely source of the plutonium. In later publications (Sehmel 1978, 1980), outliers are attributed to "hot" particles that may have biased the results.

The nonrespirable particles collected at each height were combined to obtain the total average airborne ^{238}Pu and ^{239}Pu moving through a defined area per unit time ("flux") at each height, reported in microcuries per square meter per day ($\mu Ci\ m^{-2}\ day^{-1}$) (Sehmel 1980). At 0.3 m, the samples were sized into 12 increments ranging from 10 to 210 µm. Particles less than 10 µm were retained as the 13th size fraction. The other samples were not sized because of insufficient sample weight. In calculating the airborne flux of plutonium and soil particles, it was assumed that the nonrespirable particles approaching the cowl inlet continued in a straight trajectory and, therefore, were collected in each cowl.

The airborne flux of plutonium decreased almost 100-fold over a distance of 400 meters east of the 903 Area.

The maximum airborne ^{239}Pu flux was 6×10^{-4} µCi m^{-2} day^{-1} at site A, closest to the 903 Area. The flux decreased with distance and height as expected. Airborne fluxes of ^{239}Pu decreased by almost 100 times between A and AB. At A, the flux decreased by more than 10 times as the height increased from 0.3 m to 2 m above the ground (Figure III-47). The same trend was seen at site AB with a decrease of less than 10 times. Closer analysis showed that at both sites the flux did not significantly decrease between 1 and 10 m (the greatest height samples for these larger particles). The nonrespirable particle plume height above 10 m is unknown (Sehmel 1980).

The same general trends were seen for the airborne ^{238}Pu fluxes; however, a decrease with height was seen from 1 to 10 m. The maximum flux at 0.3 m at site A was 1.2×10^{-5} µCi m^{-2} day^{-1}. An unusual outcome was that the ^{238}Pu content in the samples at the farther site B was statistically higher than at the closer site AB.

Measuring Resuspension of Plutonium and Soil Particles at Rocky Flats

A 10-m high sampling scaffold was set up about 100 m from the eastern edge of the 903 field from November 1982 to August 1985 (Langer 1986a, 1986b, 1987, 1991). We believe this is the 903 sampler located on the map in Figure III-13. Three high-volume air samplers collected dust samples at 1, 3, and 10 m above the ground for <3 µm, 3–15 µm and >15 µm (respirable, inhalable and coarse, respectively). Thirty-four months of biweekly samples were collected. The 2-week sampling time was needed to collect sufficient plutonium for analysis.

The amount of respirable dust collected did not change with height, as might be expected for the small size particles, but the decrease in material for the larger sizes was statistically significant (Table III-16). The concentration of ^{239}Pu in the dust was analyzed, but because of the low concentrations, the counting uncertainties were quite large. No statistical decrease with increasing height was seen for the two smaller size fractions, but the coarse fraction did decrease by a factor of 3 between 1 and 10 m height. It was concluded that about 70% of the plutonium activity was associated with the coarse particles, which represent 60% of the mass of dust

resuspended. These data could not be correlated with other factors such as wind speed or direction (Langer 1991).

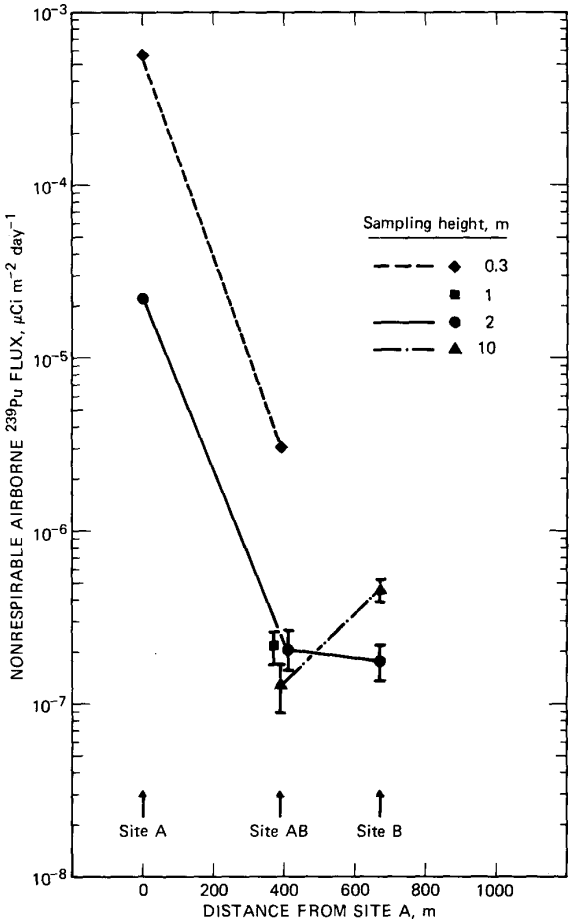


Figure III-47. Variation of total nonrespirable ^{239}Pu particle flux at Rocky Flats (from Sehmel 1980). Flux describes the amount of plutonium moving through a defined area per unit time.

The average concentration of the respirable plutonium (particles <3 µm) at this location in the 903 Pad field was about 3 times greater than the background level due to worldwide fallout. Isotopic analysis (mass ratio of ^{240}Pu to ^{239}Pu) of the soil at the 903 Pad and the respirable airborne dust was conducted by mass spectrometry at Richland, Washington. The mass isotopic ratio for Rocky Flats plutonium was 0.051 and for global fallout in soils was 0.18 (see Appendix H, this report). The soil collected at the 903 Pad had a ratio of 0.054, which was indicative of Rocky Flats plutonium. The isotopic ratio in the respirable airborne dust samples was somewhat higher (0.068), indicating relatively more fallout plutonium in air than in the soil. This slight difference could be due to the particle sizes that were associated with the Rocky Flats plutonium as opposed to fallout plutonium.

Table III-16. Summary of Dust and ^{239}Pu Concentration Data from Vertical Dust Flux Tower, November 1982-August 1985[a]

Sample height (m)	Average dust concentration (μg m^{-3})			
	Respirable[b]	Inhalable	Coarse	Total
1	8.3 ± 1.9 (23)[c]	11 ± 2.4 (22)	29 ± 8.3 (29)	48 ±8.3 (17)
3[d]	8.0 ± 2.4 (30)	11 ± 1.5 (14)	26 ± 11 (42)	44 ± 12 (27)
10	7.8 ± 1.7 (22)	8.2 ± 2.5 (30)	20 ± 7.5 (38)	35 ± 8.9 (25)
average	8.0	10	25	42
Probability that change with height is not significant	0.48	0.0004	0.0066	0.0004

Sample Height (m)	Average ^{239}Pu concentration (× 10^{-3} fCi m^{-3})[e]			
	Respirable	Inhalable	Coarse	Total
1	5.3 ± 6.4 (120)	24 ± 27 (110)	100 ± 100 (100)	130 ±100 (77)
3	15 ± 21 (140)	9.1 ± 5.9 (65)	68 ± 86 (130)	93 ± 96 (100)
10	6.0 ± 8.4 (140)	41 ± 150 (370)	32 ± 35 (110)	78 ± 120 (230)
average	8.8	125	67	100
Probability that change with height is not significant	0.71	0.50	0.014	0.31

[a]Langer 1986a, 1986b, 1987, 1991.
[b]Size ranges: respirable <3 mm; inhalable 3–35 mm; coarse >15 mm.
[c]Coefficient of variation (%).
[d]Covers period from January 1984 through August 1985, only.
[e]Local background is (2.6 ± 1.5) × 10^{-3} fCi m^{-3} at 1 m height.

Respirable Fraction of Plutonium-239 in Air near Rocky Flats

Several experiments were carried out during October 12–14, 1971, close to the RFP to determine the respirable fraction of the resuspended ^{239}Pu (Volchok et al. 1972). The closest site was near Dow Chemical's sampler S-8, at the security fence east of the 903 Area. All of the samplers were operated about 3 ft above the ground. The results indicated that in this area, where airborne concentrations were in excess of 1 fCi m^{-3}, the respirable fraction of the airborne material was about 0.2 to 0.4. This is not in good agreement with earlier studies, which showed a lower fraction (0.05 to 0.07). The experiments also indicated that the main source of resuspended plutonium was probably located somewhat east of the plant site. Also, very steep vertical gradients of plutonium concentrations were probably present in the air in this vicinity, at least in the range of 3 to 8 ft above the ground.

Dynamics of Resuspension from a Contaminated Soil Surface

This short-term special study was conducted by scientists from the Health and Safety Laboratory of New York City and the Oak Ridge National Laboratory during a 3-week period in the summer of 1973 (Krey et al. 1974). Primary conclusions of the study were

- The "hot particle" problem and sampling bias render short term air measurements at Rocky Flats uncertain and possibly not representative of average conditions
- Resuspended total mass and plutonium concentrations are inversely related to precipitation and soil moisture
- The respirable fraction of both mass and activity concentrations in surface air averages about 40%
- Only 2.5% of the respirable mass is derived from local soil surfaces
- At the time of measurement, the resuspension factor was 3×10^{-9} m^{-1}.

The resuspension factor is the ratio of the concentration of a contaminant in air (pCi m^{-3}) to the areal concentration on the surface soil (pCi m^{-2}).

Two years later, Krey et al. (1976) discussed plutonium and americium contamination in soils around Rocky Flats in a HASL report. They present data from the HASL Surface Air Program, which collected continuous air filter samples at the Rocky Flats plant perimeter fence. (Routinely reported data from this program were discussed in a previous section of this report.) Air filter samples were analyzed by the Knolls Atomic Power Laboratory to determine the $^{241}Pu/^{239}Pu$ ratios. The ratios agreed with the ratios in soil, which dated the source of the plutonium to about 1958 to 1961. The authors concluded that little or none of the observed airborne plutonium was being released from current operations. Also, $^{241}Am/^{239,240}Pu$ ratios in surface air at the RFP perimeter fence were consistent with a 1958 source with a 0.4% atom percent of ^{241}Pu.

Pollen and Spores as Radionuclide Transport Vectors

These studies, conducted in the mid-1970s, are reviewed in Chapter IX of this report, "Biological and Miscellaneous Monitoring Studies."

Special Air Sampling Associated with Litigation

In 1975, a lawsuit was filed against the U.S. and other defendants by owners of land around the RFP. A settlement agreement finalized in July, 1985 required the RFP to undertake remedial actions on those portions of the land containing plutonium at concentrations exceeding a CDH special construction requirements plutonium in soil standard (0.9 pCi g^{-1}) adopted by the court (DOE 1991).

In 1976, nine ambient air monitoring stations were installed by the RFP near the lawsuit acreage. These stations were operated from November 1976 through July 1978 specifically to collect monthly airborne plutonium concentration data for the lawsuit. The data are tabulated in Hume (1978) by month and location. The locations of the nine lawsuit samplers (S-45, S-46, S-47, and S-63 through S-68) are shown on a map in the referenced document. Two of these

stations were analyzed by an independent laboratory. Data are included for 18 other stations that were part of the routine monitoring program.

Hume concludes that:

> The data indicate no significant differences between the twenty-seven stations as a group. Likewise, there is no significant difference between the nine litigation stations as compared to the remaining eighteen. The two litigation stations analyzed by the independent laboratory do not differ from the other stations.
>
> It is concluded that the nine sampler stations installed for litigation do not yield information not provided by the routine Rocky Flats monitoring stations and hence should be discontinued. Moreover, the need for all of the stations included in the routine monitoring program should be investigated."

Hume gives no indication of the statistical method(s) used to produce his conclusions.

Our interest in these data for the historical public exposures studies was that they might be useful for assessing the releases from the 903 Area, because

1. Mechanical disturbance of the soil during plowing of the contaminated land is similar to the mechanical disturbance caused by grading of the 903 Pad, and
2. At the time of the litigation property plowing, the air samples would have been analyzed for plutonium, not just TLLa.

In a disclosure to the City of Broomfield in 1985, Rockwell (1985) presents a table showing the relationship between plutonium concentrations in air from samplers located on the different properties (including the Church and Good properties) and the concentrations of plutonium in soils from those properties. A resuspension factor was derived for six areas, and ranged from 1.0×10^{-9} to 3.0×10^{-8} m^{-1}.

Contribution of Plutonium and Americium to Total Long-lived Alpha Activity in Air

Results of these studies (Illsley 1982; Michels 1973) are incorporated into the analysis in the section titled "Apportioning Total Long-lived Alpha Count into Specific Radionuclides," earlier in this chapter.

Analysis of Resuspension Source Area Impacts at Rocky-Flats Surveillance Air Samplers S-7 and S-8

The study documented in Hammer (1984) appears to be a continuation of the previous work of Hodgin (1983a, 1983b, 1984). Air monitoring for this study was done by the Environmental and Occupational Health Branch of Rockwell in 1983. The three samplers S-7, S-8, and S-9 had routinely exhibited ^{239}Pu levels 15 to 25 times above concentrations observed at other onsite samplers, which showed concentrations typical of background for the Front Range area. These experiments in 1983 were designed to measure the horizontal flux (activity per square meter per second) of resuspended material. Ten high-volume air samplers were placed in upwind/downwind positions along an east-west line (approximate) which included samplers S-7 and S-8. The sources of contamination that contributed to the higher levels of plutonium in air at samplers S-7 and S-8 were within 150 m of the samplers. Ten additional high-volume air samplers were placed

to evaluate potential resuspension source regions. A controller switched samplers on or off according to the wind direction. At this time, most of the contamination was coming from the east. Three local source regions to the east of the perimeter fence were named (a) the east berm, (b) the range field, and (c) the east field (also see Chapter VIII of this report).

The sources of contamination that contributed to the higher levels of plutonium in air at samplers S-7 and S-8 in 1983 were within 150 m of the samplers.

Summary of Resuspension Studies after 1970 (Langer 1991)

A large number of short-term resuspension studies that have been conducted by researchers employed by the Rocky Flats contractor. All of these are not reviewed in detail in this report. Periodic updates on many studies have been published in semiannual progress reports of the Environmental Sciences Branch. Langer (1991) is a good overview of special air monitoring conducted as part of research on the resuspension of soil particles from soil contaminated with plutonium areas called the 903 Pad field and east field (see Figure VIII-1).

The results of 2 years of air sampling in these fields (Langer 1983a, 1983b, 1984a) showed that monthly plutonium concentrations in air varied by a factor of 10 to 100 at a given sampler location over a 2-year period. No correlation between wind speed and plutonium concentration was found. Neither was there any correlation between the plutonium concentrations found at four sampler locations when plotted against time. The same was true for 9 years of data from the routine surveillance samplers along the eastern security fence. The concentration data between those routine samplers did not correlate over time. That is, when one sampler was high, the others were not necessarily high. By contrast, the dust concentrations in air showed identical trends with time at the different samplers. So did the concentrations of ^{7}Be, which is a natural radioactive isotope generated in the upper atmosphere that becomes attached to dust particles in the lower atmosphere.

Citing other studies, Langer (1991) concludes that the plutonium particles measured in air in the late 1970s and 1980s were a few discrete, relatively large particles in a huge volume of air. For example, for a relatively high plutonium concentration of 1.4 fCi m^{-3} in the 903 field, 125 plutonium particles 1 micron in size would account for the activity found in 40,000 cubic meters of air sampled during a typical run. Or, a single 5-μm particle could account for all the activity. Such poor sampling statistics make it impossible to establish short-term concentration trends. This conclusion is in agreement with that reached in Krey et al. (1974). At best the averages over a year or longer could be examined for meaningful trends. For the community samplers, it is impossible to establish plutonium trends that relate to RFP activities. Finally, Langer (1991) concludes that the influence of the 903 field could not be discerned beyond about 1.5 km away, but suggests further studies to confirm this observation.

MONITORING OF BERYLLIUM IN AIR

Beryllium is one of the nonradioactive materials of concern identified in Phase I of this historical public exposures study (ChemRisk 1994a). Beryllium (chemical symbol "Be") is a solid metallic element extracted from the mineral beryl, although at least 30 different beryllium-containing minerals are known (Hurlbut 1974). Beryllium is used in nuclear weapons and was processed at Rocky Flats in large scale beginning in 1958 (Barrick 1983). The main production operations took place in Building 444 and included casting (foundry), cutting, heat-treating, rolling, and machining. Beryllium foundry operations ceased in 1975, but other uses of beryllium continued beyond that time.

Estimates of the amounts of beryllium released to air from Rocky Flats were based on historical effluent monitoring of building stacks. The highest beryllium releases were believed to have occurred in 1968, with an annual release estimate of 38 grams. However, the time trend in release estimates is complicated by the fact that analytical detection levels improved over time, and many of the release quantities are reported as "less-than" values (McGavran and Rood 1999).

As with radioactive materials, the environmental monitoring data for beryllium were not examined extensively in Phase I of this study, but have been in Phase II. Environmental monitoring of beryllium in soils of the Rocky Flats area is summarized in Chapter VIII of this report. In general, enhanced concentrations of beryllium in soil due to Rocky Flats airborne releases are not evident.

Background Concentrations of Beryllium in Air

Beryllium is present in small quantities in air mainly due to suspension from naturally occurring beryllium in soils and combustion of coal. Uncontaminated air in North America contains around 1×10^{-4} μg m^{-3} or less (EPA 1987; Drury 1978). Average concentrations in urban air, depending on the amount of coal burning, can be two to forty times higher, or 2×10^{-4} μg m^{-3} to 4×10^{-3} μg m^{-3} (EPA 1987; Merian 1984).

We developed an approximate range for the background beryllium concentration in air using a soil concentration of 1 μg g^{-1} (range 0.3 to 6 μg g^{-1}) (McGavran and Rood 1999) and a dust concentration in the air of 35 μg m^{-3} (range 10 to 100 μg m^{-3}). An uncertainty analysis was performed assuming triangular distributions of these two parameters. The resulting 90% confidence range for background beryllium in air is 3×10^{-5} μg m^{-3} to 3×10^{-4} μg m^{-3}, with a median of 1×10^{-4} μg m^{-3}. Our median estimate is in good agreement with the literature values for uncontaminated air. This background range will be used for perspective for measured concentrations in the environment and for predicted concentrations from Rocky Flats releases.

The following sections of this chapter review the available historical monitoring data for beryllium in ambient air around Rocky Flats. In addition, shorter-term measurements were located, including a few in the 1950s, a special study in 1978, and special monitoring during a fire in February 1978.

Early Monitoring of Beryllium in Ambient Air (1950s–1960s)

A few data on beryllium measurements in ambient air in the 1950s were located in the monthly site survey reports of Dow Chemical Company. The first mention is the March 1954 site survey report (Kittinger 1954a), which states:

> Three air samples from the Wagner School air sampling station were given Be analysis by emission spectroscopy with all reported at about 2 ppm. of Be.

The April 1954 monthly report (Kittinger 1954b), states:

> Only two air samples from the Wagner School Station were submitted for Be analysis. (Others are being held for analysis of fallout decay.) These were reported as 2, and 5–7 ppm of Be *of the ashed sample weight*. (Italics added for emphasis.)

Without the ashed sample weights, it is impossible to convert these reported results to concentrations of beryllium in air. They only serve as evidence that early monitoring of beryllium in ambient air had begun.

Dow Chemical Company also took a few air samples near the Beryl Ore Co., a privately owned operation located on W 100th Avenue, about 1.2 miles east of Indiana Street and about 0.9 miles south of the RFP access road's entrance upon Indiana. The April 1954 monthly site survey report (Kittinger 1954b), states:

> Survey near Beryl Ore Co. (off-site). It was decided to install air sampling equipment on this property to determine airborne Be, Li, and U present in the atmosphere. An air sampling station was installed about 125 feet southeast of the Grinding building on April 28.

Samples were collected twice weekly. In the May 1954 the results were reported as "positive but less than allowable amounts." Quantitative results were given in June and July of 1954: Average concentrations (of biweekly samples) were reported to be 3.7×10^{-3} µg m^{-3} and 3.4×10^{-3} µg m^{-3} for June and July, respectively. Dow dismantled this sampling station on July 30, 1954.

Average concentrations for June–July 1954 near Beryl Ore Co. were about 35% of the air quality "standard," which was (and still is) 1×10^{-2} µg m^{-3}. This standard was developed in 1949, thus preceding the EPA and its ambient air quality standards by about 25 years. This value is now listed in the code of federal regulations (40CFR61.32) as an EPA guideline for an average (30-day) concentration in ambient air. For the remainder of this chapter, we will refer to this value as the "EPA ambient air concentration guideline."

Beginning in December 1957, beryllium concentrations in ambient air are again reported in the monthly site survey reports. The detection level was reported as 0.002 µg beryllium per sample. However, we do not know to what extent the air filters were composited before analysis, and therefore what the corresponding minimum detectable concentration (per unit volume) would have been. At any rate, the site survey reports for December 1957 through February 1958 state that all onsite and offsite samples were at or below detection levels. In March 1958, the site

survey report states that the monitoring program for beryllium was being transferred to the Safety Department, and results would no longer be given in the site survey reports.

The August 1962 monthly report from site survey and industrial hygiene (Hill 1962) indicates that "Beryllium air sample results continue low except for isolated cases. Results of beryllium monitoring have been sent to those concerned."

Routine Monitoring of Beryllium in Ambient Air (1970–1976)

Routine monitoring results for beryllium in ambient air are reported beginning in 1970 in the monthly environmental reports issued by Dow Chemical Company. Although after the period of highest releases of beryllium, releases during the 1970s are higher than they were in the 1980s. Concentrations are reported as the average of all offsite or all onsite stations combined. There are no individual results for different air monitoring stations. The longest record is available for the onsite samplers (January 1970 through October of 1976). These data are illustrated in Figure III-48. Sometime in mid-1973, the analytical method or sample collection procedures changed such that the detection level improved from 1×10^{-3} µg m^{-3} to 1×10^{-5} µg m^{-3}. Although this change was not noted until the February 1975 monthly report, we suspect it occurred after the two months of missing data in April and May 1973 (Figure III-48), because after that time much lower concentrations are often reported. In addition, the 1974 annual environmental report gives 1×10^{-5} µg m^{-3} as the detection level for beryllium in ambient air. Monthly average concentrations for beryllium in ambient air before mid-1973 are too near the detection level to be used quantitatively (Figure III-48).

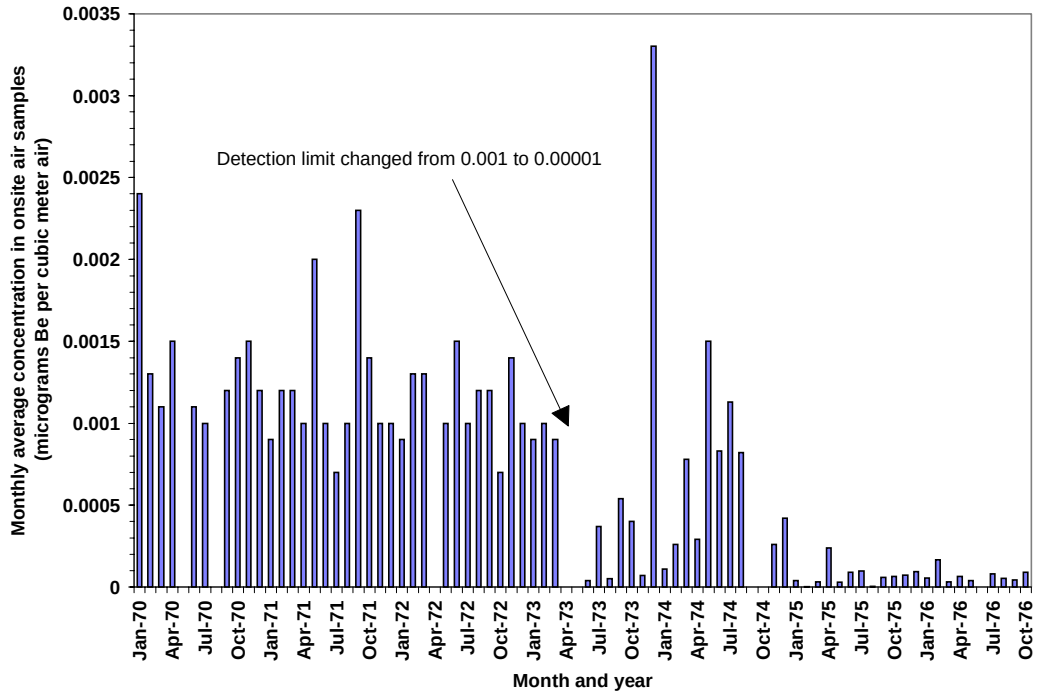


Figure III-48. Time trend in monthly average concentration of beryllium in onsite air (all stations combined).

In January of 1972, the monthly environmental reports began to include a *maximum* beryllium concentration in addition to the all-station average. It is not known exactly what this maximum beryllium concentration represents. For example, it could be the highest value on any daily air filter or on a composited set from one location over some time period. Only rarely was the location of that maximum concentration given with the value.

There is a 14-month period (January 1972 through February of 1973) in which average and maximum concentrations are reported for both onsite and offsite (Figure III-49). Average concentrations onsite are very similar to offsite and are close to the detection level. However, there is a strong suggestion that the maximum concentrations in onsite air reflect Rocky Flats operations.

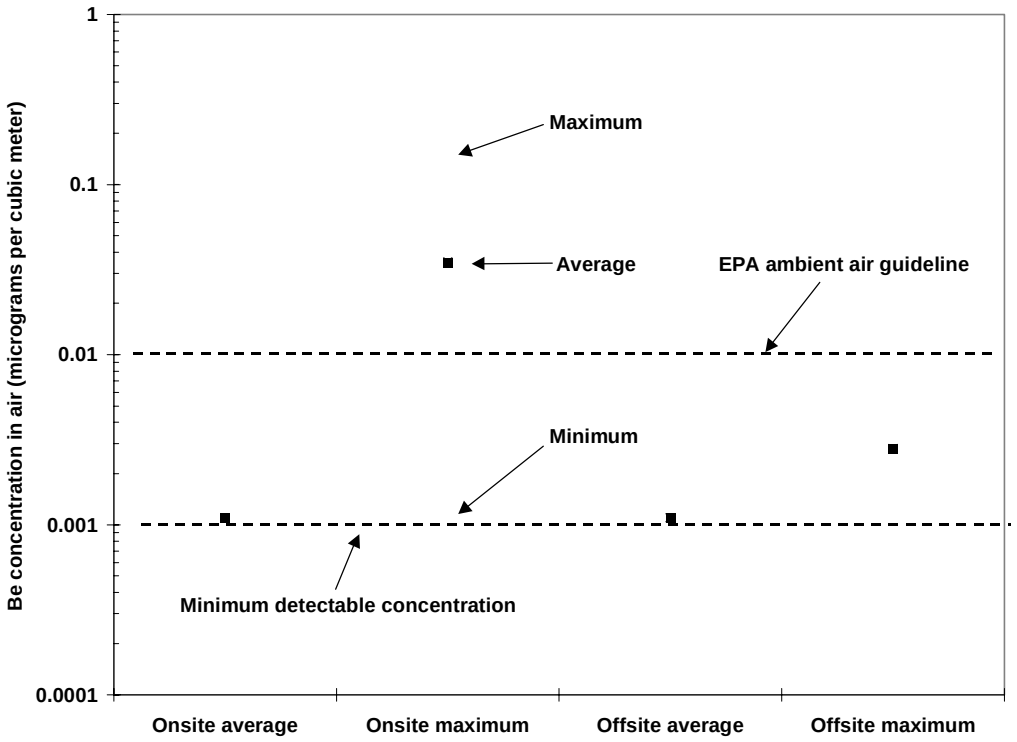


Figure III-49. Monthly average concentrations of beryllium in onsite and offsite air, January 1972 through February 1973. A "maximum" concentration was reported along with the average for each month. It is not known exactly what the maximum represents (see text). The range plotted here represents the highest and lowest concentrations reported during this 14-month period.

The maximum and average concentrations of beryllium in onsite air for June 1973 (after the detection level improvement) through October 1976 are plotted along with the reported monthly stack releases in Figure III-50. There is no correlation between the monthly release quantities and the maximum concentration in ambient air (correlation coefficient, r = 0.03) or the average concentration in ambient air (r = 0.2). The long-term average concentration for the measurements plotted in Figure III-50 is 3×10^{-4} µg m^{-3}, or about 3 times higher than our estimated median background concentration.

The EPA's discharge limitation for beryllium was 10 grams per stationary source in a 24-hour period (EPA 1973). The entire Rocky Flats Plant is considered a single stationary source.

The release amounts are the primary indication of environmental compliance for beryllium at the RFP. This is probably why results of ambient air monitoring for beryllium were never reported in the annual environmental reports, although the data were collected and reported in the contractor's monthly reports in the 1970s.

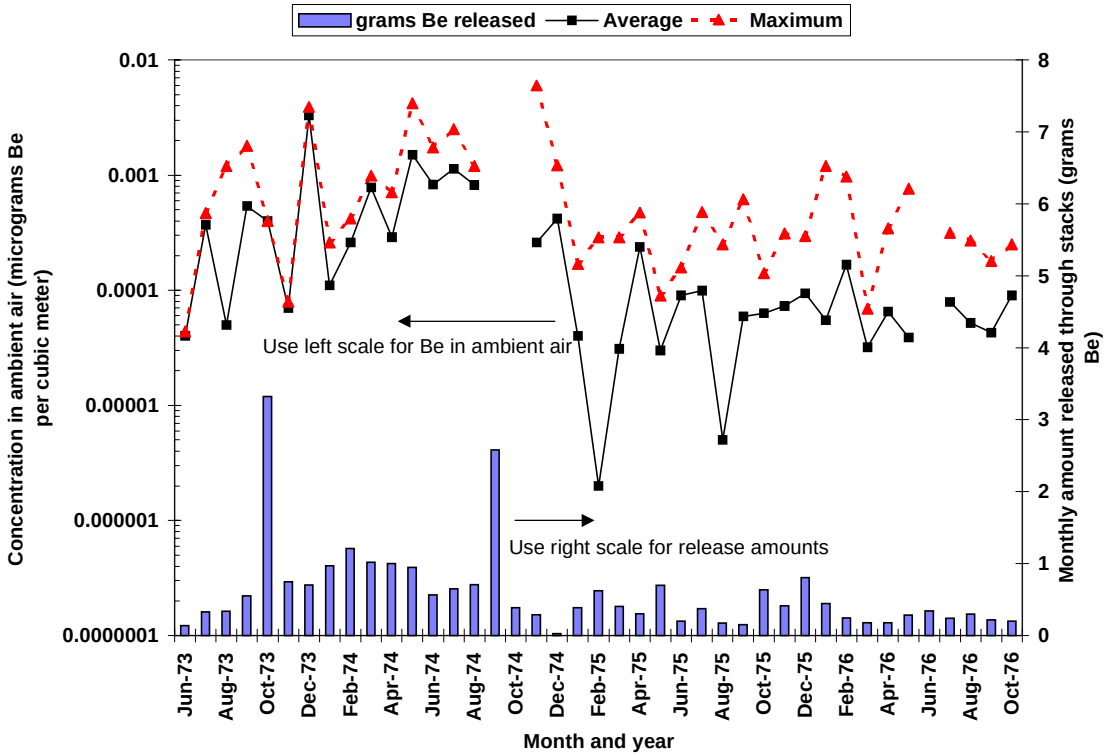


Figure III-50. Time trend in monthly average and maximum concentrations of beryllium in onsite air, along with estimates of amounts of beryllium released per month. There is no correlation between amounts released and concentrations in ambient air.

Short-term Monitoring of Beryllium in Air (1978)

Routine monitoring of beryllium in ambient air by the RF contractor apparently stopped in late 1975. Only short-term environmental monitoring data were located after that time.

A special study examined the concentration of beryllium in ambient air near the beryllium shop dock on the south side of Building 444 (Barker 1978a). Soil in this area had been contaminated with beryllium due to a previous practice of pouring chlorinated hydrocarbon solvents, which had been used to rinse beryllium parts, onto the ground at that location. The time period during which air samples were collected was June 20, 1977 through September 20, 1977. An ambient air sampler (the Rocky Flats designed high-volume sampler in routine use for radiological monitoring at that time) was placed about 3 feet above ground level on a pole 8 ft south of the Door 10 to Building 444. The 8 × 10 inch Microsorban filters were collected weekly and analyzed for beryllium by flameless atomic absorption. The approximate detection level was very low, 7.5×10^{-9} µg m^{-3}.

Beryllium concentrations measured in air at this location were well above detection levels. Thirteen weekly samples ranged from 3.0×10^{-4} to 2.3×10^{-3} μg m^{-3}, averaging 9.0×10^{-4} μg m^{-3}. Because the average concentration in air at the contaminated soil location in 1978 was 9% of the EPA ambient air concentration guideline (1×10^{-2} μg m^{-3}), Barker (1978a) indicated that removal of the contaminated soil was not required at that time.

This contaminated area of soil is one of the few places on the Rocky Flats Plant site where beryllium concentrations in soil are well above background. In general, the concentrations of beryllium in soil appear to be in the range known to occur naturally and show no relationship to distance or direction from the industrial area (McGavran and Rood 1999).

Short-term monitoring of beryllium in air was conducted in response to a fire that occurred in the filter plenum of building 444 on February 23, 1978. The wind direction during the releases was from the east to the west. The highest concentrations observed onsite within the security fence during plume passage (about 30 minutes) were 0.02 μg m^{-3} (Werkema 1978). This concentration was computed based on the assumption that all of the beryllium on the air filters was deposited during 30 minutes, that is, the measured amount of beryllium on each filter was divided by the volume of air sampled in 30 minutes. In reality, the samplers had been operating for several days prior to the fire. Similarly, measured amounts of beryllium on air filters from five samplers on the western and southwestern boundary of the RFP were converted to estimated concentrations which ranged from 0.006 to 0.015 μg m^{-3}, based on the volume of air sampled during an estimated plume passage time of 1 hour. When the entire volume of air sampled was used to compute the beryllium concentrations, air from the onsite stations ranged from 6×10^{-5} to 3.4×10^{-4} μg m^{-3}. Concentrations at boundary stations ranged from 4×10^{-5} to 9×10^{-5} μg m^{-3} when averaged over the entire volume of air sampled in roughly a weekly period (Barker 1978b).

Analytical Considerations for Evaluation of Historical Beryllium Monitoring Data

In addition to the detection level differences discussed earlier, there are other data quality issues that arise when examining the historical beryllium monitoring data. The annual environmental reports in the early 1970s only state that the beryllium analyses were conducted by atomic absorption spectrophotometry and cite the method reference (Bokowski 1968). Beginning in 1974, quality control results are reported for beryllium on air filters. Both error and bias were often large (Table III-17). Hurlbut (1975) investigated possible causes for low yields in beryllium air filter analysis. He indicated that results of beryllium analysis on air filters submitted to the laboratory had been routinely 30% low.

Jameson and Bokowski (1983) address a possible solution for a known negative bias for beryllium analyses on effluent air filters. The reason for the negative bias was loss of a portion of the beryllium in each sample in the fume and mist evolved during digestion of the filter samples. They state that losses ranged from 12 to 17% with an average of approximately 13%. (Note: this does not agree with the large negative bias observed in 1979 quality control samples, Table III-17.) A solution to the problem was to place glass beads in a funnel above the digestion beaker during sample oxidation. The beads allowed gases to escape while acting as impingers to collect beryllium mist. The relative standard deviation after this modification was less than 5% and accuracy was within 2%.

Table III-17. Intralaboratory Quality Control Results for Beryllium on Air Filters Reported in Rocky Flats Annual Environmental Reports

Year and medium	Relative error (%)[a]	Bias (%)[b]	Total number of control analyses
1974 Effluent filters	127.3	–1.2	not given
1975 Effluent filters (Whatman)	17.3	–10.9	240
1975 Effluent filters (Gelman)	40.8	0.1	240
1976 Effluent filters (Gelman)	55.1	41.6	240
1976 Room air filters (Whatman)	9.4	–10	240
1977 Effluent filters	31	35.2	240
1979 Whatman filter	44.4	–67.7	120
1980 Whatman filter	116	not given	120

[a] Also called coefficient of variation (Hornbacher et al. 1981).
[b] Bias is the observed value minus the standard value, divided by the standard value and expressed as a percent. A negative bias means the measured value was low compared to the standard value.

Summary and Perspectives on Beryllium Concentrations in Air

Figure III-51 provides a perspective for the various levels of beryllium concentrations that have been discussed here. The highest concentration, represented by the dashed horizontal line, is the EPA ambient air concentration guideline of 1×10^{-2} μg m^{-3}. The lowest concentration, also shown by a dashed horizontal line, is the detection level for the flameless atomic absorption analytical method, which was used in the special study conducted by Barker in 1978 and for effluent air monitoring in the 1980s. After mid-1973, the detection level is low enough that measured concentrations of beryllium in ambient air are quantitative. The long-term average concentration in onsite air is about 3 times our median estimate of the natural background concentration, but many months are within a background range.

Both maximum and average concentrations in onsite air are unrelated to the amounts released via Rocky Flats stacks. Based on the results of the Barker (1978a) study, it is likely that resuspension of contaminated soil onsite influenced the maximum concentrations observed in onsite air. There was only a small amount of onsite soil identified as contaminated with beryllium; a much smaller amount than was contaminated with plutonium. Based on environmental monitoring data for plutonium, the concentration of beryllium in offsite air is expected to be at least 1000 times less at the site boundary than onsite. Thus, the concentration of beryllium in offsite air from resuspension of contaminated soil onsite would be much less than background.

McGavran and Rood (1999) presents the methodology used in Phase II of this study for predicting concentrations of beryllium in offsite air from stack releases of beryllium from Rocky Flats. For the highest year (1968, 38 grams beryllium released), the corresponding concentrations in offsite areas (median estimate) ranged from 7×10^{-7} µg m^{-3} east of the plant at Indiana street to 4×10^{-9} µg m^{-3} at the southeastern border of the assessment domain, near Aurora (Figure III-51).

The ambient air concentration guideline of 1×10^{-2} µg m^{-3} was used by the Rocky Flats Plant as a level of acceptability for some operational decisions, like removal of contaminated soil. In the light of recent knowledge, that guideline is probably inadequate protection for those individuals who are sensitive to chronic beryllium disease (McGavran and Rood 1999); however, no revised standard has been promulgated. The comparison to natural background concentrations carries less connotation of acceptability. It is clear that most measured concentrations of beryllium in onsite air are comparable to naturally occurring concentrations and that the concentrations in offsite air due to Rocky Flats releases were many times lower than naturally occurring concentrations. In this sense, the incremental risk due to past Rocky Flats beryllium releases was probably small.

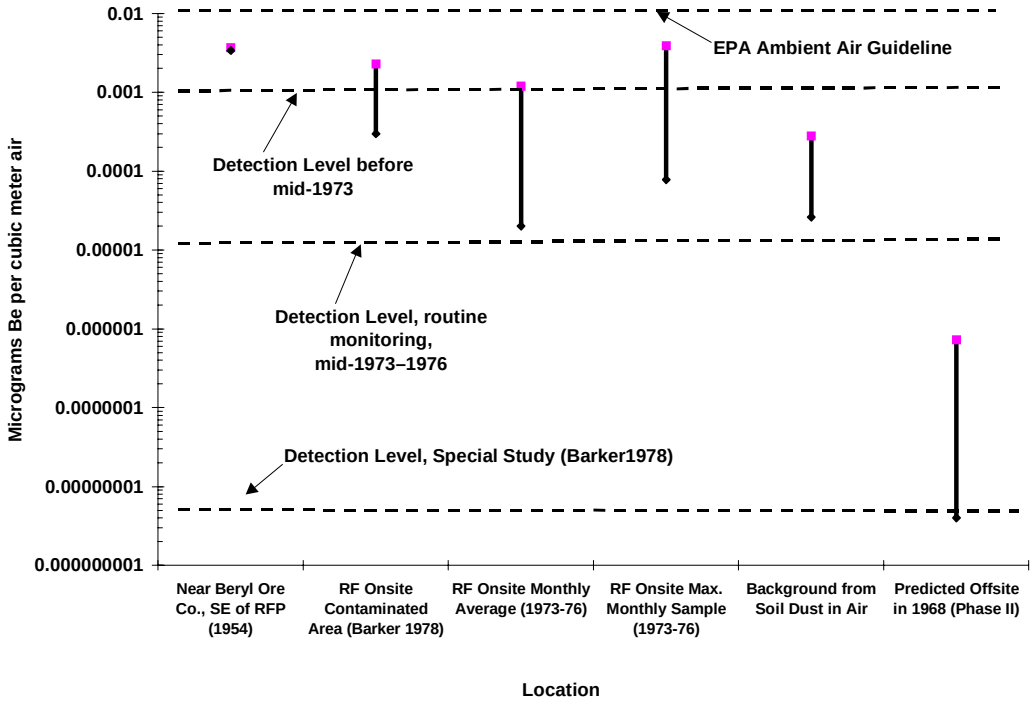


Figure III-51. Perspectives on concentrations of beryllium in ambient air. The 1973–1976 onsite ranges are the 5th to 95th percentiles of all monthly results. The predicted concentrations of beryllium in 1968 (highest release year) are annual average concentrations from Rocky Flats releases (McGavran and Rood 1999) and do not include natural background. The range shown for the predicted concentrations in 1968 is the range of median values within the model domain. The areas representing the highest and lowest predicted concentrations are east of the RFP at Indiana Street and Aurora.

DATA QUALITY ISSUES FOR HISTORICAL AIR MONITORING

Because there are a limited number of historical data sets available, quality is usually addressed by quantifying bias and uncertainty in the measurements, not by disqualifying the data.

As mentioned earlier in this report, data quality issues are being addressed as environmental data sets are identified as being potentially useful to the historical public exposures studies. It was clear early in our work that the daily measurements of alpha activity in air from the RFP contractor's onsite sampler S-8 would be key to at least one approach to evaluating primary releases from that area (1965–1969). That assessment is documented in Weber et al. (1999). In addition, the HASL air monitoring data in the 1970s, as well as the contractor and CDH monitoring in the 1970s and 1980s, would be used to evaluate the spatial trends in secondary resuspension after the 903 asphalt pad was installed.

Although a number of documents relating to historical air monitoring were collected and reviewed, evaluating data quality is a complex task that requires a thorough knowledge of sampling collection, analysis, and data reporting procedures throughout the years. The routine monitoring data or contractor reports, especially older ones, did not provide this information. Nonroutine research projects that were conducted at the RFP and elsewhere provided some help. General discussion of the most important data quality issues is presented here. However, final decisions are described in other reports where the data are applied (Rood and Grogan 1999a, b, c, d; Weber et al. 1999).

Bias and Uncertainty in Air Monitoring Data

In evaluating the data from historical air monitoring programs, sources of uncertainty and bias must be examined. Uncertainty is a general term used to describe the level of confidence in a given measurement. Bias is a systematic distortion of measurements that makes the results inaccurate. Accuracy is a measure of how close a value is to the true number, or a measure of the correctness of a measurement. Precision refers to the ability of an analytical method to reproduce the same result upon repeated trials.

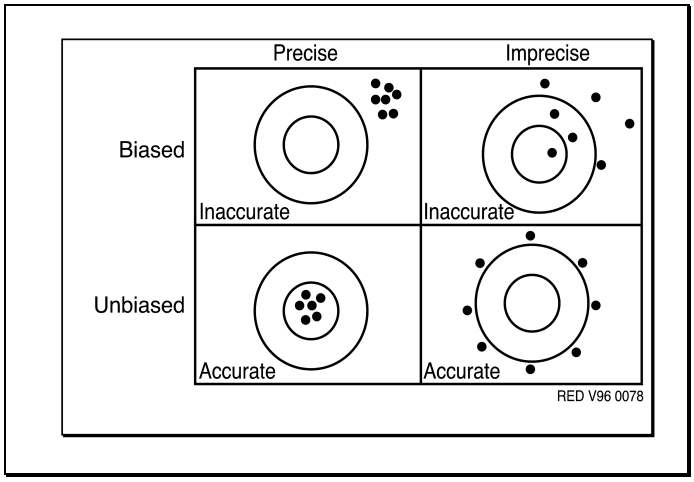


Table III-18 lists some sources of uncertainty in measurements of airborne alpha activity and the expected direction of bias. Information was not located to evaluate all sources of uncertainty for all times. However, a number of these issues were examined. Statistical counting error will be

considered first, because this is a limiting factor that excluded some data (particularly some TLLa counts) from further quantitative analysis.

Table III-18. Direction of Bias for Potential Sources of Uncertainty in Measurements of Airborne Alpha or Plutonium Activity

Source of uncertainty	Direction of bias[a]
Noncontinuous sampling (for early offsite samples)	0
Statistical counting error	0
Variation in sampler flow rate	0
Flow rate decrease because of particulate loading of filter	–
Inlet collection efficiency	–
Filter collection efficiency	–
Loss of particles from filter between collection and counting	–
Deliberate data falsification[b]	–
Contamination of sample because of poor contamination control	+
Self-absorption of alpha particles by filter	[c]
Alpha counter efficiency	[c]
Analytical error in determining plutonium activity	[c]

[a] – Indicates measurement would tend to underestimate true concentration.
+ Indicates measurement would tend to overestimate true concentration.
0 Indicates no systematic bias.

[b] No evidence of data falsification has been uncovered. However this is listed as a possible source of bias because it has been raised by concerned members of the public. It is also presumed that if data falsification occurred, it would be to decrease the observed measurements. We have found some data transcription or arithmetic errors. See, e.g., the discussion about the January 1969 monthly average on pages III-46 and III-47. Misleading or incorrect summary statements have been found, particularly in materials for public release in the early 1970s.

[c] Direction of possible bias depends on whether correction factors used are good estimates.

Uncertainty in Total Long-Lived Alpha Count Rates Because of Statistical Variations in Radioactive Decay (Counting Error)

Radioactive decay is a random process. The number of nuclei in a sample of radioactive material that decay in any time period is not fixed but fluctuates about a mean value. This fact can be demonstrated by making repeated measurements of the activity of a long-lived radionuclide, each for the same time duration. A distribution of counts is represented by the binomial distribution. The normal distribution is an excellent approximation to the binomial when the sample half-life is long compared to the counting time, and when the number of nuclei present is large. The basic statistical principles and equations used to describe the uncertainty in the measurement of radioactivity are well described in many textbooks (e.g., Wang et al. 1975, Chapter 12; Watson 1980, Chapter 6).

For most environmental radiation measurements, the net count rate of a sample (r_s) is determined by subtracting a background count rate (r_b) from a gross (sample plus background) count rate (r_{s+b}). The uncertainty in the net count rate, sigma (σ), (often called *counting error*) is given by

$$\sigma = (r_{s+b}/t_s + r_b/t_b)^{1/2} \quad . \qquad \text{(III-1)}$$

where t_s and t_b are the counting time for the sample and background, respectively.

Stated in words, the counting error of a net count depends on the background count rate, the gross sample count rate, and the length of time the sample and the background are counted. A 95% confidence interval on a net sample count is given by the count rate ± 1.96 [. Longer counting times reduce counting error.

The counting error is important to consider for the TLLa measurements at Rocky Flats in the 1960s, because the count times were short, and gross count rates were often similar to the counter background count rates. As discussed earlier in this chapter, the counter background is the count rate obtained with a blank, unused air filter in the instrument. As an example, the computation of counting error for data reported on the sample data sheet illustrated earlier in this chapter (Figure III-20) is shown in Table III-19. Note that only half of the 12 samples counted on that day can be considered as containing detectable activity based on counting error alone.

Table III-19. Counting Error Example for Data Recorded in Figure III-20[a]

Gross sample count rate (r_{s+b})	Background sample count rate (r_b)	Net sample count rate (r_s)	Counting error for r_s (1σ)	Counting error for r_s (1.96 σ)	Positive activity detected?[b]	Coefficient of variation ([/ r_s)
0.6	0.7	-0.1	0.27	0.52	no	
1.0	0.6	0.4	0.33	0.65	no	
0.5	0	0.5	0.22	0.44	yes	45%
0.8	0.2	0.6	0.29	0.57	yes	48%
0.2	0	0.2	0.14	0.28	no	
1.8	0.3	1.5	0.43	0.84	yes	29%
0.2	0	0.2	0.14	0.28	no	
1.3	0.3	1.0	0.37	0.72	yes	37%
0.5	0.1	0.4	0.23	0.45	no	
0.8	0.2	0.6	0.29	0.57	yes	
0.4	0	0.4	0.20	0.39	yes	50%
0.4	0	0.4	0.20	0.39	yes	50%

[a] Count time of sample was 10 minutes, from Figure III-20. Count time of background assumed to be 60 minutes (Putzier 1996).

[b] If the net sample count rate is less than 1.96[, then there is a 95% probability that no additional alpha activity was present in the sample as compared to the counter background.

Our database of TLLa activity counts in daily onsite air samples contains only the net sample count rate (see previous section of this chapter). We anticipated that the daily net alpha counts will be used in at least one approach to evaluating releases from the 903 Area, and this was the case (Weber et al. 1999). To use the air monitoring data quantitatively, the uncertainty in the measurements must be assessed. Counting error is only one component of uncertainty in these data, but it is an important one. With over 27,700 individual daily measurements in the database over the time interval between October 1964 and December 1971, it is impractical to determine a counting error and significance judgment about the presence of detectable activity for each sample. Rather, some general observations are needed about the levels at which net TLLa counts can be used in a quantitative way, and the uncertainty associated with various levels.

Alpha counter background is an important component of counting error when gross sample counts are low (Equation III-1). The counter background varied from 0 to 0.9 cpm for a set of over 200 counts that we examined from 1969 (Figure III-52). Ninety percent of the counter

backgrounds in this data set were between 0 and 0.7 cpm. The median, 0.25 cpm, is close to the MDA of 0.21 cpm that was used by the RFP contractor.

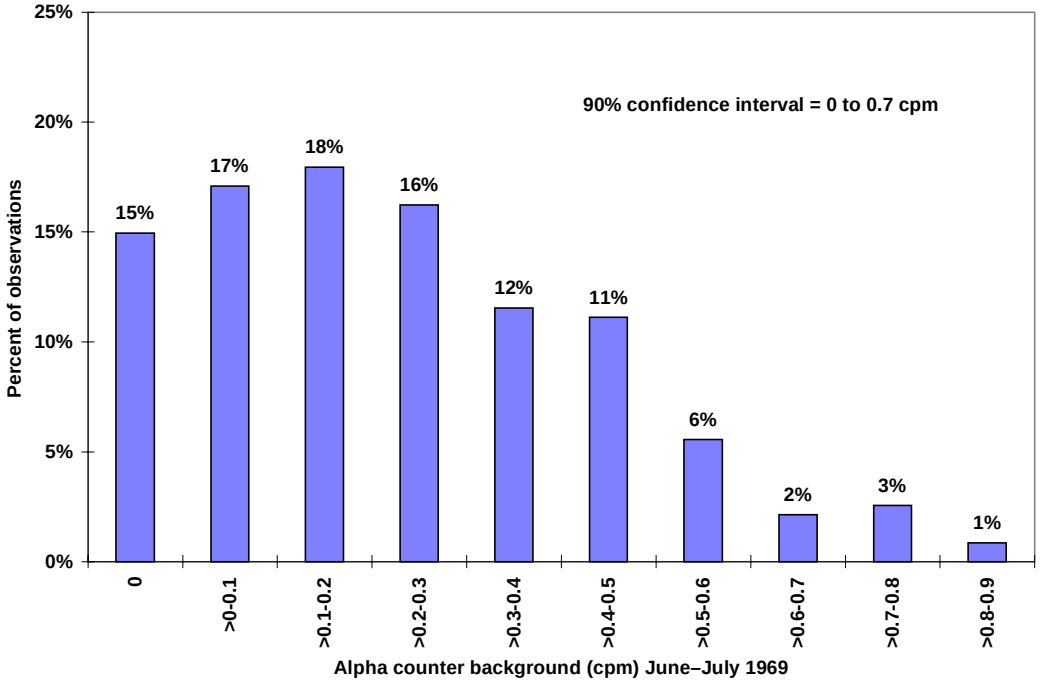


Figure III-52. Histogram of alpha counter background count rates for 234 counts taken between June and July 1969. Data were transcribed from daily alpha count sheets. The median was 0.25 cpm.

Counts of TLLa activity in air from onsite daily samples ranged from 0 to over 600 cpm. The most frequently observed net count rate was 0 and the median was 0.1 cpm. Table III-20 illustrates the counting error as a function of background count rate and gross sample count rate (using Equation III-1). It is clear that counting error (when expressed as a coefficient of variation) for samples with large amounts of activity is small. For net count rates over 100 cpm, 1-[counting error is 3% or less, probably smaller than or comparable to other sources of uncertainty, such as alpha particle self-absorption in the filter. Two cells in Table III-20 where the coefficient of variation is 50% are shown in bold boxes. These correspond to points where 2-[counting error is 100% of (equal to) the net count rate itself. At net count rates $\geq$0.8 cpm, positive alpha activity is quite likely to be present, but the counting error can still be substantial. At 0.4 cpm and below, the count method cannot positively detect alpha activity at a 95% confidence level. Therefore, for the 903 Area risk assessment, individual alpha count rates above 0.5 cpm could be used quantitatively, with counting error considered as one source of uncertainty in the measurements. Counts of 0.4 cpm or less should not be used quantitatively. The coefficient of variation using a background cpm of 0.25 is considered the best estimate of the counting error for a single count, with an uncertainty range based on background count rates of 0 and 0.7 cpm (Table III-21).

Table III-20. Counting Error for Long-lived Alpha Samples[a]

Net sample (cpm)	Counting error (coefficient of variation, %, in the net sample cpm)								
	Background cpm								
	0	0.1	0.2	0.25	0.3	0.4	0.5	0.6	0.7
600	1%	1%	1%	1%	1%	1%	1%	1%	1%
300	2%	2%	2%	2%	2%	2%	2%	2%	2%
100	3%	3%	3%	3%	3%	3%	3%	3%	3%
10	10%	10%	10%	10%	10%	10%	10%	10%	10%
5	14%	14%	14%	15%	15%	15%	15%	15%	15%
4	16%	16%	16%	16%	16%	17%	17%	17%	17%
3	18%	19%	19%	19%	19%	20%	20%	20%	21%
2	22%	23%	24%	24%	24%	25%	25%	26%	27%
1	32%	33%	35%	36%	37%	38%	40%	41%	43%
0.9	33%	35%	37%	38%	39%	41%	43%	44%	46%
0.8	35%	38%	40%	41%	42%	44%	46%	48%	**50%**
0.7	38%	41%	44%	45%	46%	49%	51%	53%	56%
0.6	41%	45%	48%	50%	51%	54%	57%	60%	63%
0.5	45%	50%	54%	56%	58%	62%	66%	69%	73%
0.4	**50%**	57%	63%	66%	68%	74%	78%	83%	87%
0.3	58%	68%	77%	81%	85%	92%	99%	105%	111%
0.2	71%	89%	104%	111%	117%	129%	140%	150%	159%
0.1	100%	147%	183%	198%	212%	238%	261%	283%	303%

[a] Based on count times of 10 minutes for sample and 60 minutes for background.

Table III-21. Counting Errors to be Used in 903 Area Risk Assessment to Evaluate Uncertainty in Daily Counts of Total Long-Lived Alpha Activity

Net count rate (cpm)	Coefficient of variation from counting error	
	Best estimate (%)	Uncertainty range (%)
100–700	2	1–3
3.0–99.9	15	10–20
1.0–2.9	35	22–43
0.8–0.9	40	33–50
0.5–0.7	50	38–73
0.4 and less	Not used quantitatively	

Table III-22 presents the percentage of daily TLLa counts from our database of onsite samplers (October 1964 through December 1971) that fall into each of the ranges of net count rate defined for this counting error analysis. With the exception of sampler S-8, over 80% of the daily counts of TLLa activity should not be used quantitatively based on counting error alone. The percentage of daily counts in the quantitative region ranges from a low of 7.4% for sampler S-51 to a high of 37.1% for S-8.

Table III-22. Percentage of Daily Total Long-Lived Alpha Counts (Onsite Samplers) That Fall into Ranges Defined for Counting Error Analysis

Net cpm range[a]	Onsite sampler											
	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-50	S-51
100–700	0.0	0.0	0.0	0.0	0.0	0.0	0.1	0.2	0.0	0.0	0.0	0.0
3.0–99.9	0.6	0.2	0.4	0.6	0.4	0.8	1.5	7.1	0.0	0.3	0.2	0.1
1.0–2.9	3.7	1.5	2.4	2.7	2.7	3.7	4.4	12.6	1.2	1.9	1.9	0.8
0.8–0.9	2.6	1.2	1.9	2.4	2.0	2.3	2.2	5.1	1.9	1.6	0.9	0.6
0.5–0.7	8.6	7.0	8.0	6.7	6.4	9.1	7.3	12.1	6.1	7.0	6.9	5.9
0.4 and less	84.5	90.1	87.3	87.6	88.5	84.1	84.5	62.9	90.8	89.2	90.1	92.6
0.5 and higher	15.5	9.9	12.7	12.4	11.5	15.9	15.5	37.1	9.2	10.8	9.9	7.4

[a] See previous table for counting error applicable to each count rate range.

The sample count time for TLLa analysis was increased to 30 minutes on August 25, 1969. The effect of this longer count time is a reduction in the net sample counting error to about 60% of what it had been previously for a 10-minute sample count time. Because the largest releases occurred before this date, the uncertainties given in Table III-21 are recommended for use for the entire data set.

It should be emphasized that actual net counts, including zero values, are being used when computing averages for long-term trend charts or other analyses. However, an average that is comprised of daily measurements mostly <0.4 cpm will have little or no quantitative meaning.

Collection Efficiency of Ambient Air Samplers

The collection efficiency of the ambient samplers is one significant source of negative bias (measurement will be lower than true value). Simply speaking, the collection efficiency of an air sampler is a measure of how accurately the sampler captures the true airborne concentration of the contaminant of interest. For example, if the true airborne concentration is 100 units, and the air monitoring procedure produces an estimate of 80 units, then the collection efficiency would be 80%. (Analytical bias is not considered here.)

The total collection efficiency of an air sampler can be viewed as having two components:

1. The *inlet collection efficiency* of the sampling device (how accurately the device draws the ambient aerosol into the filter)
2. The *filter collection efficiency* (the amount of the material drawn into the filter that is retained by the filter, i.e., does not pass through it).

Properties of the air sampler that affect total collection efficiency include

- Inlet face velocity of the incoming air (related to flow rate)
- Placement height and orientation
- Type of weather shelter or housing
- Filter characteristics.

Properties of the environment, such as wind speed and orientation with respect to the sampler inlet, also affect the inlet collection efficiency (Figure III-53). Older ambient air samplers were often constructed from equipment also used for in-plant air monitoring. The pump and filter collection unit was simply installed in a weather shelter, basically a box with louvered sides to allow air to enter. The photographs in Figures III-54 show the community air sampler (low-volume) used by the Rocky Flats contractor in the 1950s. Information about the type of housing used for onsite samplers in the 1950s and 1960s was not located. Figure III-55 is a photograph of the air sampler used by the HASL in their routine air monitoring program, and Figure III-56 shows the samplers used by the RFP contractor and the CDH in the late 1970s.

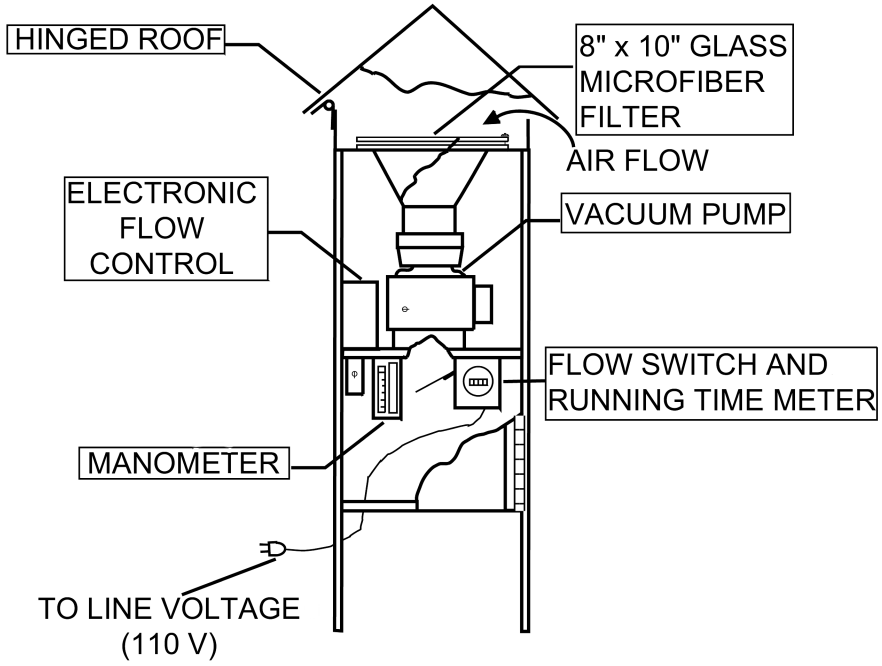


Figure III-53. Cutaway schematic of typical high-volume air sampler used in the 1970s and later. Air entering the sampler must undergo a nonlinear trajectory in order to be drawn into the air filter. Large particles do not follow the air flow as well as smaller ones, resulting in lower inlet collection efficiency for larger particles.

Figure III-54. Low-volume community air sampler maintained by the Rocky Flats contractor in the 1950s. In the upper photo, the sampler housing is closed, illustrating louvered door through which air enters. In the lower photo, the sampler housing is open, illustrating air filter and pump assembly. EG&G Rocky Flats photo numbers 01619-00 dated May 9, 1956, and 02196-00, dated March 20, 1957.

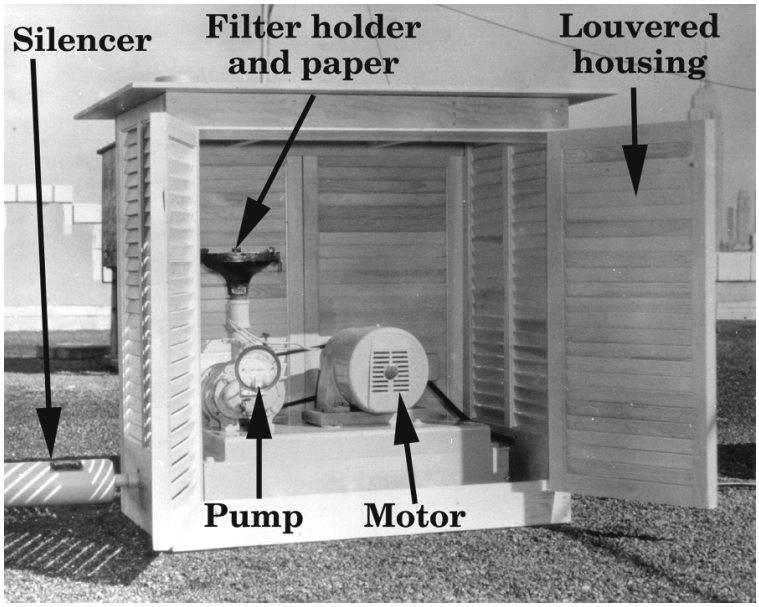


Figure III-55. Air sampler and louvered housing used by the Health and Safety Laboratory (HASL) in the 1970s–1980s, including the sites around Rocky Flats (Volchok and de Planque 1984; Rosen 1996). Different types of samplers were used by HASL near Rocky Flats during intensive experiments performed over shorter periods. The routine samplers used by HASL at Rocky Flats were Roots rotary lobe blowers, AF size 24 (Feely 1978). Filters were Microsorban polystyrene (see next section). Field personnel at the RFP provided readings of temperature and pressure drop across the filter made during the sampling interval. The flow rates were calculated from the observed pressure drops using a calibration curve and are corrected for temperature. Results were adjusted for altitude and temperature to standard air volumes. Variations in the calibration of the samplers during a 5-year period were expected to be less than 10%.

Figure III-56. Air samplers used by the Rocky Flats Plant (left) and the Colorado Department of Health (right) in the mid-1970s and beyond. EG&G Rocky Flats photo number 28761-6 dated ~1980. A schematic of the insides of these samplers is included later in this chapter.

The most important characteristic of the aerosol that affects the inlet collection efficiency of an air sampler is the particle-size distribution. The aerodynamic diameter (AD) of a particle is more relevant to evaluating collection efficiency than its physical size. The AD is the diameter of a sphere of unit density (1 g cm^{-3}) that has the same gravitational settling velocity as the particle. Particles of dense materials behave as if they were larger than their physical size. The AD of a particle is its physical size multiplied by the square root of the particle density. Weber et al. (1999) discusses the particle-size distribution of plutonium associated with contaminated soil in the 903 Area, and how that influenced the source term development.

It is useful—in conducting a general discussion of air sampler collection efficiency—to think in terms of *coarse* and *fine*[5] aerosols. Small particles making up fine aerosols tend to follow the air stream into the sampler; the large particles in coarse aerosols have sufficient inertia that they tend to move in straight lines, not following curved air trajectories. For coarse aerosols, aerodynamic effects outside the sampler are strongly dependent on wind speed, turbulence, orientation effects, etc.; for fine aerosols, such effects are much reduced.

Deposition collectors, such as sticky paper and dust deposit gauges, sometimes are used to collect coarse aerosols. A limited amount of this type of sampling was conducted at Rocky Flats (Chapter IV). Air samplers are used to collect the finer aerosols. They are generally inefficient collectors of coarse aerosols, because of poor inlet collection efficiency and reproducibility (Vincent 1989). For the Rocky Flats samplers, the onsite stations and those downwind of the 903 Area were probably faced with the largest proportion of coarse aerosols containing plutonium.

Garland and Nicholson (1991) summarize some important common features of studies of air sampler performance:

> . . .all the published tests show some common features: the sampling efficiency declines with particle size and also with ambient wind speed. . . . Orientation may be important for non-symmetrical inlets. The dependence of efficiency with so many parameters makes it improbable that any correction can be successfully applied to filter samplers operating in field conditions. Few of the filter samplers investigated have had a satisfactory sampling efficiency for particles larger than 30 μm, and it is unlikely that any can sample particles larger than 100 μm.

Several wind tunnel and field inter-comparison experiments on the Rocky Flats-designed Surveillance Air Sampler used in the late 1970s and 1980s have evaluated the spectrum of particle sizes collected. Site-specific documentation on the efficiency of samplers used before that time has not been located. Therefore, to interpret earlier air monitoring data, we must rely on studies of similar samplers and an assessment of basic physical principles.

In Phase I of this project, ChemRisk used a collection efficiency of 100% for particles less than 15 μm, 50% for particles between 15 and 30 μm and 0% for particles greater than 30 μm, for the assessment of releases from the 903 Area (Mongan et al. 1996b). Key references that were cited for these values were Langer (1980) and Hurley (1980). We note, however, that these studies were conducted on the relatively recent (mid-1970s) high-volume sampler used at Rocky Flats.

[5] Fine aerosols are made up of particulates with a diameter of less than 1 to 2 microns, or about 1/50 of the width of human hair. Human activities have increased the amount of fine aerosols in the atmosphere, which can be transported hundreds of kilometers. Natural dust is in the range of 2–10 microns and tends to settle more readily. In urban areas, more than 90% of the number of particles in the air may be "fine" (Perera and Ahmed 1979).

Faced with a similar concern in the Fernald Dosimetry Reconstruction Project (Killough et al. 1996), *RAC* focused its assessment of the bias of the ambient air samplers on the collection efficiency of as a function of particle size. A number of literature sources on aerosol sampling of ambient air were reviewed (May et al. 1976; Pattenden and Wiffen 1977; Wedding et al. 1977; Agarwal and Liu 1980; Liu and Pui 1981; Vrins and Hofschreuder 1983; Vincent 1989; Garland and Nicholson 1991). The work of Wedding et al. proved most applicable to the Fernald study, providing quantitative measurements of collection efficiency for the standard American High-Volume air sampler for particle sizes up to 50 μm aerodynamic diameter (Table III-23). This sampler has been the principal one used to measure total suspended particulate in air and operates at a flow rate of 50 cubic feet per minute (cfm).

Table III-23. Sampling Effectiveness (% of Total Aerosol Collected) of High-Volume Air Sampler at a Wind Speed of 4.6 m s^{-1} (Wedding et al. 1977)

Particle aerodynamic diameter (μm)							
5		15		30		50	
Sampler orientation[a]							
0°	45°	0°	45°	0°	45°	0°	45°
97%	100%	35%	55%	18%	41%	7%	34%

[a] Sampler orientation of 0° is defined as the situation with the high-volume sampler roof ridge parallel to the air flow. The high-volume sampler is extremely sensitive to the angle of approaching wind as is more efficient at 45° than 0° (Wedding et al. 1977). (See schematic, next page.)

In June 1978, Dr. Wedding and Mr. T.C. Carney of the Civil Engineering Department, Colorado State University, completed a study of the inlet collection efficiency of the Rocky Flats high-volume sampler and the efficiency of the filter paper used in that sampler (Wedding and Carney 1978, Hurley 1980). Results of the study were compared with a previous test on the standard high-volume sampler (Wedding et al. 1977) for a variety of simulated field conditions. A schematic of both samplers is shown in Figure III-57. The schematic also illustrates the meaning of sampler orientation with respect to flow of air.

Test results were obtained from measurements conducted in the Environmental Wind Tunnel Facility at Colorado State University using aerosolized oleic acid droplets (density 0.89) tagged with uranine dye. The inlet collection efficiency was determined as a function of particle size (1–34-μm diameter), wind speed 3.4–27 mph (1.52–12.19 m s^{-1}), and sampler orientation to the mean flow of air (0°, 45°, 180°).

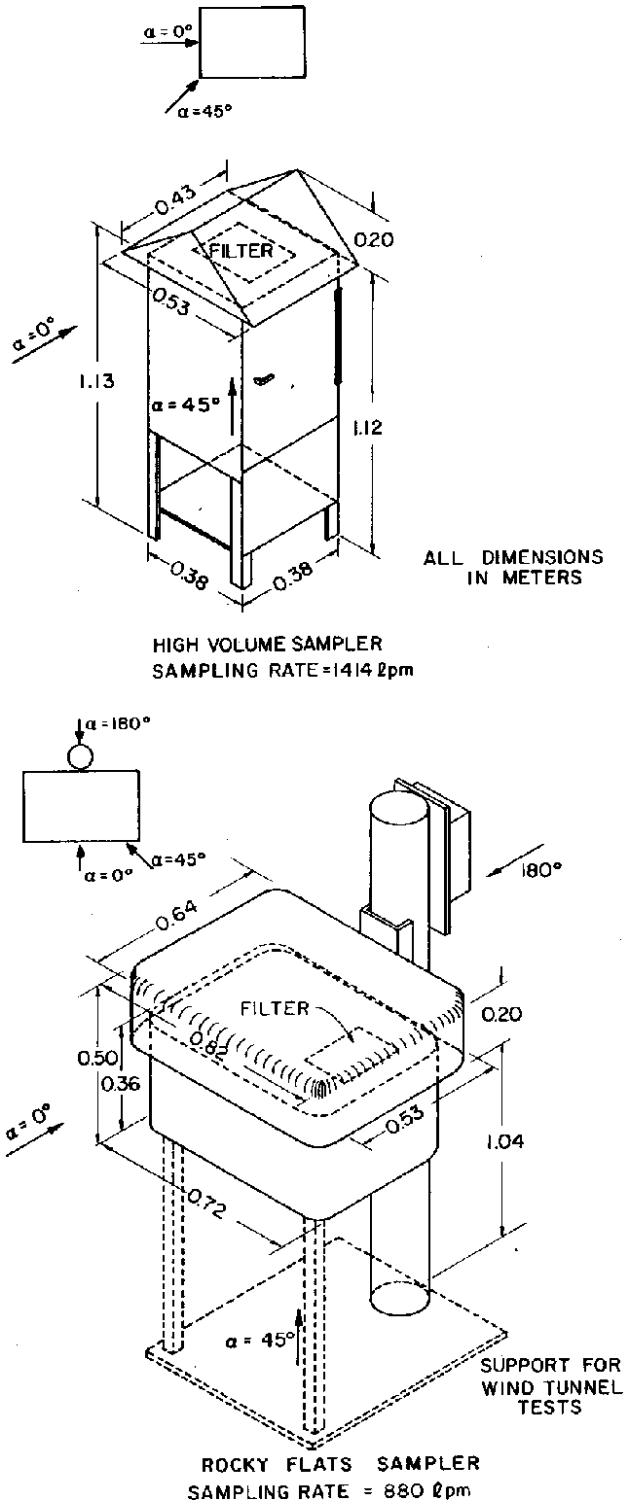


Figure III-57. Schematic of Rocky Flats ambient air sampler and high-volume air sampler tested by Wedding and Carney (1978). A photograph of these samplers is shown in Figure III-56.

The results showed that the Rocky Flats surveillance air sampler, operating at an inlet flow rate of 880 L min^{-1} (31 cfm) had an inlet collection efficiency that is a slight function of orientation angle for 1–10 µm particles, with a larger effect for 20–34 µm particles. A strong effect of wind speed was seen up to 5 m s^{-1} (11 mph) where a further increase showed only a slight decrease in collection efficiency. At the 0° orientation and 6–12 m s^{-1} wind speeds, the effectiveness ranged from 75% for 10-µm particles to 15% for 34-µm particles. Results compare favorably with previous work on the standard EPA high-volume sampler. The largest particle size (50 µm) used for previous testing of the EPA sampler was not used in the Rocky Flats tests, but the trendline for the Rocky Flats sampler suggests that the large particle collection characteristics of the Rocky Flats sampler would be similar (Figure III-58). Based on this study, Langer (1983a) stated that the Rocky Flats high-volume sampler had a 15-µm aerodynamic diameter cut-point, that is, 50% of the 15-µm particles were rejected by the sampler inlet.

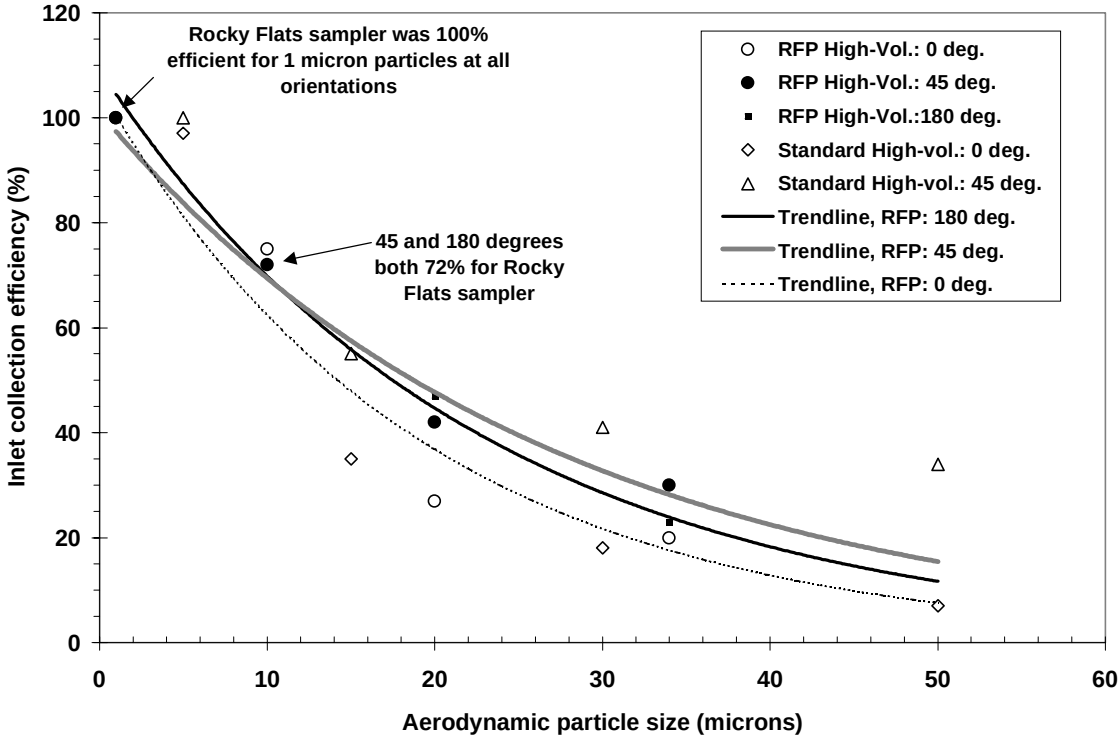


Figure III-58. Inlet collection efficiency of Rocky Flats high-volume sampler and standard EPA high-volume sampler for wind velocities of 4–6 m s^{-1} (9–13 mph) and sampler orientations from 0° to 180° (data plotted by *RAC* from Wedding and Carney 1978). Although the trendlines drawn here suggest that efficiencies for particles around 5 µm are 85–90%, experimentation with the standard high-volume sampler indicated that efficiencies are 97–100% for this size.

Figure III-58 illustrates the collection efficiency results for an intermediate wind speed. At the higher wind speed (12.19 m s^{-1}), the collection efficiency was similar to that shown here, and at the lower wind speed (1.52 m s^{-1}), the inlet collection efficiency was higher,

particularly for the 45° orientation. Plots of those data are included in Wedding and Carney (1978).

High-volume air samplers were routinely employed at Rocky Flats starting in the 1970s. Before that time, a low-volume sampler had been used, and much less is known about them. The sample flow rates for the RFP low-volume samplers were about 15 times lower than the high-volume samplers, and smaller (~2-in. diameter) filters were used. The collection efficiency studies done on the Rocky Flats high-volume sampler are not applicable to the much different samplers used in the 1950s and 1960s. In particular, the inlet collection efficiency of low-volume air samplers is more dependent on wind speed than are high-volume samplers. This distinction is important, because the air sampling data from sampler S-8 during high wind events was used to assess releases from the 903 Area (Weber et al. 1999).

The inlet collection efficiency for six low-volume air samplers is given in Vincent (1989, Chapter 15). Figure 15.16 of that reference presents the inlet collection efficiency as a function of wind speed (up to 9 m s^{-1}, or 20 mph) and aerodynamic diameter (AD) up to 25 µm. Qualitatively, the samplers show very consistent patterns. For all six, collection efficiency varies strongly with both AD and wind speed, decreasing with increasing AD and with the rate of fall being more rapid at higher wind speeds. Table III-24 presents the data for two of the six samplers that are closest (both higher and lower) to the flow rate of the Rocky Flats low-volume sampler. It is clear that at a relatively high wind speed (>7 m s^{-1} or 16 mph), only very small particles (less than 5 µm) are collected efficiently. At low wind speeds, a wider range of particle sizes are efficiently collected. The average wind speed at the RFP is around 9 mph (4 m s^{-1}).

Table III-24. Effect of Particle Size and Wind Speed on Inlet Collection Efficiency of Low-Volume Air Sampler

Aerodynamic particle diameter (µm)	Collection efficiency (%)[a]		
	1 m s^{-1}	3 m s^{-1}	7 m s^{-1} [b]
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] Range of two low-volume samplers bracketing flow rate of Rocky Flats sampler (from Vincent 1989, Chapter 15).

[b] Collection efficiency of two other low-volume samplers at a wind speed of 9 m s^{-1} was 0% for particles greater than 10 µm.

The collection efficiency of the low-volume sampler is an important consideration in this study because the air monitoring data from S-8 were used to calibrate the suspension model for primary releases from the 903 Area (Weber et al. 1999). However, it should be emphasized that the particle size range that is important for internal dose assessment should have been efficiently collected by the samplers. This is particularly true for the high-volume samplers that operated beyond the mid-1970s. In addition, the total mass concentration of radioactive particles is a rather poor indication of the inhalation hazard of an aerosol. It is now a widespread view that if just one aerosol fraction is to be collected relevant to health for a wide range of types of aerosol in the ambient atmosphere, then that fraction should be relevant to the deposition of particles in the lung (Vincent 1989). Inhalability and deposition of particles in the lungs is a function of particle size. Modern-day samplers are often designed to cut off the largest particle sizes and collect only the

respirable fractions, sometimes subdivided into certain size categories. Depending on the particle sizes of the contaminant, the total mass concentration and the respirable mass concentration may be quite different. In recent years, the RFP contractor and the CDPHE have routinely monitored plutonium in specific particle size categories.

The particle size distributions of plutonium from the various sources at Rocky Flats were examined as part of the reconstruction of those source terms and are presented in other Phase II source term or risk assessment reports. In those reports, the useful air monitoring data have been incorporated. Correction of air sampling data was most critical for the 903 Area source term development (Weber et al. 1999). An approach generally illustrated in Figure III-59 was used to correct the data for collection efficiency, as well as other sources of bias and uncertainty. For other Rocky Flats sources, the uncorrected air sampling data are compared to predicted concentrations of respirable particles only. The uncertainties in the predicted concentrations were so large that minor adjustments in the air monitoring data were unnecessary.

Similarly for monitoring of global fallout by independent agencies (discussed earlier in this chapter), inlet collection efficiency is not as important to consider as it is for the 903 Area releases. The particle sizes that are subject to large-scale transport are small enough that inlet collection efficiency is expected to be high.

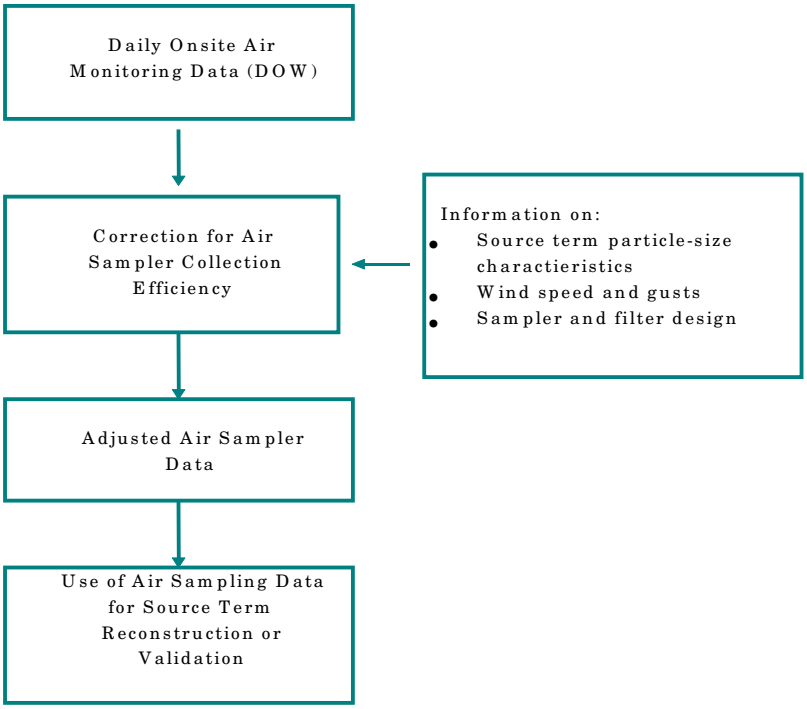
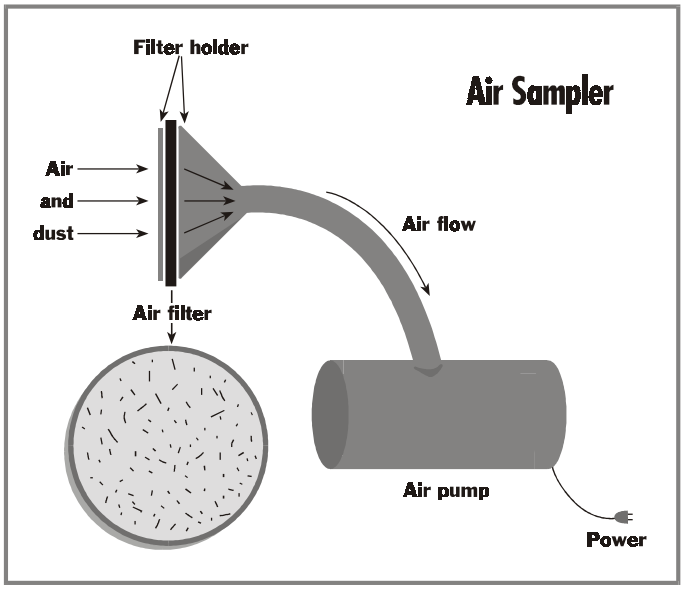


Figure III-59. Correction of daily onsite air monitoring data for collection efficiency.

Filter Collection Efficiency and Clogging

From our review of the technical literature, collection efficiency of the filter is a much less important source of bias in ambient air sampling than the inlet collection efficiency. A similar conclusion is reached by Eisenbud (1973), which states:



"The uncertainties in estimating the human hazard from inhaling radioactive dust are so great that small differences of the order of 10 to 20% owing to imperfection in filter performance are relatively unimportant and would not affect one's evaluation of a given set of data. All the commercial filter media, when used properly, have efficiencies that are more than adequate to serve the purpose."

There is a common misconception that very small particle sizes are not efficiently captured by air filters (e.g. Biggs 1997). In fact, air filters are least efficient for particle sizes of about 0.3 microns and collect smaller and larger particles more efficiently. This minimum efficiency results from the competing physical collection mechanisms of inertia and diffusion (Lindeken et al. 1963). As discussed in the previous section, the inlet collection efficiency of small particles is also high.

The efficiency and other characteristics of air filters used by different monitoring groups included in this chapter are discussed below.

Health and Safety Laboratory/Environmental Measurements Laboratory

Feely (1978) asserted that the air filters used by the HASL (Microsorban polystyrene filters) in their monitoring near the RFP in the 1970s were "effectively 100% efficient for the collection of small particles at the air velocities used" (about 40 cm s^{-1}, or 80 fpm). Feely cites Lockhart et al. (1964) for this claim, which we verified by examining that reference. Larsen (1995) prepared a chronology of the filter media, and their associated aerosol collection efficiencies, which were used in the HASL/EML Surface Air Sampling Program since the mid-1960s. Microsorban filter material was used from the 1960s until 1988 when HASL's stock was depleted following termination of the filter's manufacture. Microsorban consists of a layer of cellulose gauze onto which microfibers of polystyrene are deposited in "random disorder." This in turn is protected with another layer of cellulose tear-resistant gauze. The filter has relatively low air resistance and is very resistant to clogging. Two filter geometries were mentioned by Larsen (1995), a ~200-mm (~8-inch) diameter filter using a Roots Blower-24AF, resulting in a filter face velocity from ~45 to 90 cm s^{-1}; and a ~200 mm by ~250 mm filter using a Fuji Ring Compressor Type 302P,

resulting in a filter face velocity from ~30 to 45 cm s^{-1}. From Lockhart et al. (1964), the expected filtration efficiencies are >99%.

Public Health Service

The Radiation Surveillance Network of the PHS used 4-in cellulose, carbon-loaded filters (MSA BM-2133) with an effective area of 9.6 in^2 (62 cm^2) in its samplers with a flow rate of 40-46 cfm (Lockhart and Patterson 1962). This resulted in a filter face velocity of about 300 cm s^{-1}. Lockhart et al. (1964) gives a filter collection efficiency of 98.1–99.8% for these conditions. Additional data quality considerations for PHS monitoring was presented in an earlier section of this chapter, along with a description of the monitoring and analytical methods.

Colorado Department of Public Health and Environment (1970s–1980s)

The CDH high-volume air samplers for total suspended particulates use 8 × 10-in. glass microfiber filters, Whatman EPM–2000 (Harrison 1995). Lodge (1989) gives a collection efficiency of 99.0–>99.99 for Whatman EPM–1000 glass fiber filters.

Rocky Flats High-Volume Samplers (1970s–1980s)

Wedding and Carney (1978) documents tests of the Microsorban-98 filters used in Rocky Flats high-volume air samplers over the size range of particles from 0.01 to 1 μm and with three different face velocities using sampler flow rates of 600, 800, and 1000 L min^{-1} (21–35 cfm), corresponding to pressure drops of 20–24 inches of water (3.74–4.49 cm Hg). The filter was found to be greater than 99.9% efficient over the range of particle sizes and pressure drops tested.

In 1982, the RFP was notified that production of the Delbag Microsorban filter media was being discontinued. A variety of media were tested that year, and a decision was made to switch to Schleicher and Schuell glass microfiber filters (S&S-29) at selected onsite and all perimeter and community sampler locations (Anonymous 1983). Manufacturer's test specifications rate this filter media to be over 99% efficient for relevant particle sizes under conditions typically encountered in routine ambient air sampling (Schleicher and Schuell 1982).

Rocky Flats Low-Volume Samplers (1950s-1960s)

Until the early 1970s, low-volume samplers (2 cfm) were used by Rocky Flats for routine ambient air monitoring. These samplers used a different type and size of filter than the later high-volume samplers. The February 1954 monthly site survey report states that Whatman 41 filter paper[6] was substituted for HV70 paper[7] on all *offsite* samplers, because the HV70 filters often ruptured in the week-long sampling duration used offsite. This statement could imply that HV70 filters continued to be used for onsite (daily) sampling, but we are not certain. In Phase I, ChemRisk also came to the conclusion that ambient air samplers during the 1957 fire probably used HV70 filters or Whatman-41 filters (both are 47 mm [about 2 in.] in diameter) (Mongan et al. 1996a). Assuming an effective diameter of 47 mm (surface area, 17 cm^2), the filter face velocity would have been 54 cm sec^{-1} or 107 fpm.

[b] Whatman-41 cellulose filter media. Whatman Paper Ltd., Clifton, NJ 07014.

[c] HV–70 cellulose-asbestos filter media. Hollingsworth and Vose Company, East Wadpole, MA 02032.

Lindeken et al. (1963) presents the collection efficiency of Whatman 41 filter paper for submicron solid particles at different collection velocities. The authors state that the collection efficiency is minimal in the velocity range of 20–30 fpm (10–15 cm s^{-1}), which is explained by the effect of two opposing mechanisms (diffusion and impaction), each of which influence retention of particles by the filter. The lowest collection efficiency was 74%. At a collection velocity of about 100 fpm (closest to the Rocky Flats sampling condition), the collection efficiency would have been between 90 and 95% for submicron particles.

Lockhart et al. (1964) presents the efficiencies of various filters for three different aerosol types. For a face velocity of 50–60 cm sec^{-1}, the efficiency of Whatman 41 paper was 84% for mono-disperse 0.3 μm particles of dioctyl phthalate (DOP), 76% for natural radioactive aerosols in the atmosphere, and 99% for airborne fission products. The HV70 efficiencies were 99.2% for DOP, 98.6% for natural radioactive aerosols in the atmosphere, and 100% for airborne fission products. For the 903 Area source term development (Weber et al. 1999), *RAC* used a filter collection efficiency of 90–100%. Hammond et al. (1969) states that the filters used in the ambient air sampling program were 99.7% efficient, but no details are given.

Filter Clogging

An RFP investigator suggested that measured alpha activity in air from the low-volume samplers might be biased low because of clogging of the filter with particulates (Chapman 1960), thereby reducing collection efficiency (Mongan et al. 1996b). Particulate loading of an air sampler filter actually does not decrease the filter's efficiency. In fact, dust particle loading has been shown to *increase* the filter efficiency of Whatman 41 cellulose filters (Lindeken et al. 1963). However, the effect of particulate loading on the air filter may be to decrease the actual air flow rate through the filter. In turn, this would result in an overestimate of the volume of air sampled. Recall that in the early years, the volume sampled was a fixed average value, pre-printed on the data sheet (Figure III-20), and not computed for each actual sample. Sampling of air may even stop with severe dust loading if the motor overheats.

This topic was addressed by Lockhart et al. (1964) for a number of different filter types. Compacted cellulose filters are the most quickly affected by clogging. The percent decrease in flow is given by Lockhart et al. for a unit volume of air drawn through a unit filter surface area (m^3 air cm^{-2} filter). Whatman 41 paper was found to have a 5.0% decrease in flow and HV70 a 1.7% decrease in flow per m^3 cm^{-2}. The Rocky Flats onsite samplers collected 81.5 m^3 day^{-1} across an effective area of 17 cm^2, or 4.8 m^3 cm^{-2}. Therefore, under typical dust loading conditions with the Rocky Flats sampler, flow might have decreased 8% ($4.8 \times 1.7\%$) over a day using the HV70 paper and 24% ($4.8 \times 5.0\%$) if Whatman 41 paper were used.

Flow-rate changes under typical sampling conditions should have been incorporated into the original calibration of the samplers. This calibration would have produced the estimated volume of air sampled in a day, which was pre-printed on the data sheets. A low-volume air sampler and a daily filter change-out should not produce filter clogging problems under most sampling conditions. However, filter clogging would have been more likely to occur on windy days with high dust loading in the air. For this reason, ChemRisk used *ratios* of alpha concentrations from different onsite air samplers to select appropriate models in Phase I.

The effect of filter clogging/flow rate decrease on the measurements of alpha activity during high wind events was examined further in the source term development for the 903 Area (Weber

et al. 1999). For wind speeds higher than 15 m s^{-1}, a correction was made to the volume of air sampled.

Selvidge (1976), based on experience at Rocky Flats, gives a standard deviation for the total daily volume of air sampled (81.5 ± 2.04 m^3), or a relative standard deviation of 2.5%. However, this is an assumed value for the purposes of illustrating the factors involved in assessing overall precision of an air sampling result. An example of the correction of an air sampler count due to filter clogging and variation in sample volume is illustrated later in this chapter.

Determination of Amount of Radioactive Material on Air Filter

After airborne particulate material from the environment has been collected on the air sampler's filter, the filter is taken to the laboratory where the amount of radioactive material present is determined. That amount, divided by the volume of air sampled, produces the estimated concentration of the material during the sampling interval. This section of the report includes the available information to assess the bias and uncertainty in the historical measurements. Determination of beryllium on air filters was addressed in a previous section.

Counter Efficiency and Alpha Particle Self-absorption

The two key variables in determining the bias for measurements of alpha activity on the air filter are the counter efficiency (also sometimes called counter geometry) and the self-absorption of alpha particles within the air filter. Counter efficiency represents the percentage of total alpha particles emitted that are measured by the detector; this was determined by counting thinly plated standard sources containing a known amount of alpha activity. The counter efficiency used by the RFP was 30%. In addition, a 70% correction factor was applied for self-absorption when counting air filters instead of very thin standard sources. This means that 70% of the alpha particles would escape the filter matrix, and 30% would be absorbed. These two subfactors were combined to produce a total efficiency factor of 0.21 cpm per dpm ($0.7 \times 0.3 = 0.21$), which was used by RFP personnel to convert alpha count rates to activity concentrations. The basis for these two subfactors in alpha counting efficiency was investigated in Phase I of this project and discussed in section 2.2.2.1 of their Task 5 report (ChemRisk 1994a). With regards to the counting geometry, the report states

> Plant personnel verified counting efficiencies by performing daily measurements with electroplated standard alpha radiation sources that were traceable to the National Bureau of Standards. The values remained reasonably consistent over the years, due in large part to plant procedures which required technicians to adjust each instrument's amplifier gain in order to maintain 30 percent counting efficiency."

Selvidge (1976), based on experience at Rocky Flats, gives an standard deviation for the 30% counter geometry as 0.5%, or a relative standard deviation of 1.7%. However, this is an assumed value for the purposes of illustrating the factors involved in assessing overall precision of an air sampling result.

Documents that describe the technical basis for the self-absorption correction factor were not located during Phase I, although interviewees described a study that derived or confirmed that value (ChemRisk 1994a). Our review of the literature supports the Phase I conclusions that 30%

loss due to absorption by the filter is reasonable. An early study (Alercio and Harley 1952) was located that examined three uranium compounds (alpha-emitters) collected on Whatman 41 paper. The alpha particle energy of ^{238}U (~4.2 MeV) is less than ^{239}Pu (~5.2 MeV), so the Alercio and Harley results should be conservative when applied to plutonium. (Plutonium alpha particles have greater energy and would be somewhat more likely to escape the filter and be detected). The filters containing various amounts of uranium compounds were analyzed by alpha counting (to ± 1% precision), and those results were compared to the mass of uranium present (determined chemically) after filter dissolution. Counter geometry was considered separately, so the difference between the alpha-count result and the chemical result reflected the amount of alpha-particle absorption. As a result of the study, Alercio and Harley recommended a 30% absorption correction factor for routine alpha counting of air particulates, which is in agreement with the value used by the RFP.

Some other details of interest can be derived from the Alercio and Harley results. Three uranium compounds were tested: two insoluble compounds and one gas, uranium hexafluoride, which hydrolyzes rapidly with moisture in air to form a finely divided mist. The two solid compounds are most applicable to our situation at Rocky Flats. When large amounts of uranium were collected on the filter, the absorption of alpha particles *decreased*. The authors do not speculate on the reasons for this, but it could be from collected dust not going as deeply into the filter. The lowest amount of absorption was 15% for samples containing more than 1.1 mg of UF_4. The authors do state that there was no evidence for self-absorption of alpha particles by the uranium itself or inert dusts collected; that is, the absorption occurred in the filter. The amount of dust required for complete absorption was estimated to be 35 mg per paper, which was never approached in practice. In a very dusty field situation, when total dust loading could exceed 100 µg per m^3 (Langer 1984a, 1984b), the total mass collected by a daily air sample (81.5 m^3) might be over 8 mg if the flow of the sampler was maintained (see previous section). These results suggest that counts on air samples collected during very windy conditions, in which a large amount of mass is collected, will not be underestimated because of increased self-absorption; in fact the reverse may be true.

Another similar study was located from the Fernald, Ohio, uranium facility (Boback 1963), in which the absorption of uranium alpha particles by Whatman 41 paper was examined. Before this study, the Fernald site had been using an absorption loss of 30%, based on the Alercio and Harley work. A total of 250 samples containing less than 50 µg of sample particles were analyzed. Results are presented for five different uranium compounds. The average absorption was 42%, ranging from 29% for U_3O_8 to 50% for UF_4. For 23 samples containing larger amounts (50 to 690 µg), an absorption loss of 34% was obtained.

At the Oak Ridge National Laboratory, Struxness (1954) reported the absorption of alpha particles from uranium dioxide on six filter media. This was determined by the ratio of the mass obtained by a direct counting method to the mass obtained by a chemical method. The count-to-chemical mass ratio was 0.879 ± 0.17 (absorption of 12%) for HV70, 18 mils filter media, 0.861 ± 0.12 (absorption of 14%) for HV70, 9 mils filter media, and 0.489 ± 0.07 (51% absorption) for Whatman 41 filter media.

From our review of these three studies, we agree with the Phase I researchers that the 70% correction factor (30% absorption) for loss of alpha particles in the filter is reasonable. However, we also believe that an uncertainty on that value should be incorporated along with other sources of uncertainty. Documentation of the uncertainty of this factor was not located in Phase I.

The uncertainty in the amount of absorption of alpha particles in the air filter has been researched and incorporated into the total efficiency factor for alpha count results.

Selvidge (1976), based on experience at Rocky Flats, gives a standard deviation on the 70% correction factor as 8%, or a relative standard deviation of 11%. However, this is an assumed value for the purposes of illustrating the factors involved in assessing overall precision of an air sampling result. Alercio and Harley (1952), from analysis of over 100 filters, presents a relative standard deviation for the self-absorption correction factor of 25%. This corresponds to a 2-sigma confidence interval for the absorption factor from 15 to 45%. To include the higher (~50% absorption) observed by Boback (1963) and Struxness (1954), we propose a triangular uncertainty distribution with a best estimate of 30% absorption, an upper bound of 50%, and a lower bound of 15%. Incorporating this distribution into the total efficiency factor produces a best estimate of 0.21 cpm per dpm, with lower and upper bounds of 0.15 and 0.25 cpm per dpm, respectively. This distribution was used by Weber et al. (1999) to describe the uncertainty in the total efficiency factor in the development of the 903 Area source term.

In 1978 and 1979, the results of inter-laboratory comparison programs are reported in the annual environmental reports; total alpha activity on air filter media was one the comparisons made with the EPA Environmental Monitoring Systems Laboratory. For both these years, the ratio of the Rocky Flats results to the standard result was 1.0, indicating excellent agreement. In 1980, the relative percent error was 16% for analysis of alpha activity on only one air filter.

Analytical Error in Determining Plutonium Activity on Air Filters at Rocky Flats

As discussed earlier in this chapter, the routine analysis of plutonium on ambient air filters began in the early 1970s, with the first half of that decade being a time of transition both in sampling equipment and in analytical techniques. The analytical procedure used by Rocky Flats for ambient air filters in 1979 is documented in Bokowski (1979).

Sill (1975) discussed some problems in measuring plutonium in the environment. The major problems identified were in obtaining sufficient samples to be statistically representative of the populations involved and in obtaining homogeneous samples for analysis. Compared to the problems and uncertainties involved in sampling, Sill viewed analytical problems as relatively minor. In large part, this is due to the availability of isotopic tracers. As long as complete exchange is achieved between the tracer and the sample isotopes, and contamination and errors in standardizing the tracer are avoided, virtually all other deficiencies in the analytical procedure will be accounted for, and accurate results will be obtained. Sample decomposition then becomes the most critical part of the determination.

Plutonium, uranium, and americium were determined in air filters at Rocky Flats by chemical separation from the sampling matrix, isolation from other radionuclides by ion exchange techniques, electrodeposition onto stainless steel disks, and analysis by alpha pulse-height spectrometry. A tracer was added to each sample to determine chemical yield and counting efficiency. The tracers used for plutonium, uranium, and americium were ^{236}Pu, ^{236}U, and either

^{244}Cm or ^{243}Am, respectively. In 1980, environmental samples were counted for a minimum of 16 hours (Hornbacher et al. 1981) and up to 168 hours for samples needing a lower detection limit.

Quality control results began to be reported in the annual monitoring reports beginning in 1974 (Thompson and Hornbacher 1975). By the end of 1974, approximately 15% of all samples analyzed by the internal Rocky Flats Environmental Analysis Laboratory (EAL) were quality control samples. The preparation of standard samples and numerical analysis of sample results were conducted by the Rocky Flats Chemical Standards Laboratory. A summary of the quality control results of the EAL is provided in Table III-25.

Table III-25. Intralaboratory Quality Control Results for Plutonium on Air Filters Reported in Rocky Flats Annual Environmental Reports

Year and medium	Relative error (%)[a]	Bias (%)[b]	Total number of control analyses
1974 Effluent filters	33.6	3.6	not given
1975 Effluent filters (Gelman AE)	16.6	7.0	240
1976 Ambient air filters (Microsorban)	8.6	9.2	48
1977 Effluent filters	9.1	6.7	240
1977 Ambient air filters	9.1	6.7	48
1978 Gelman E filters	16.3	3.9	75
1978 Whatman filters	8.7	5.0	24
1979 Whatman filters	-12.1	-23.2	120
1979 Microsorban	-14.2	-29.2	48
1980 Microsorban	5.0	not given	48
1980 Whatman filter	35	not given	120

[a] Also called coefficient of variation (Hornbacher et al. 1981).
[b] Bias is the observed value minus the standard value, divided by the standard value and expressed as a percent. A negative bias means the measured value was low compared to the standard value.

In addition, the EAL participated in inter-laboratory comparison programs sponsored by the EPA's Quality Assurance Branch in Las Vegas, Nevada, as early as 1974 (Thompson and Hornbacher 1975). Only qualitative statements about the results of this program are given in the Rocky Flats environmental reports. Thompson and Hornbacher (1976) states, "Computer reports and laboratory performance charts periodically furnished by the EPA indicate that the Rocky Flats Environmental Laboratory performance is satisfactory." In 1978, Hornbacher et al. (1979) state that the (renamed) Rocky Flats Health and Environmental Laboratory participated in the EML's Quality Assessment Program. Rocky Flats results of analyses of EML samples were submitted to EML, who published both a quarterly report and an annual summary listing the results reported by all participants.

Longbotham and Lawton (1986) states that the laboratory procedure for measuring the amount of ^{239}Pu in high-volume air sampler filters was changed substantially at the beginning of

1980, increasing the accuracy of the results. Hence, it states that the concentrations before and after January 1980 are not comparable.

Analytical Error in Determination of Plutonium Activity on Air Filters (HASL/EML)

Krey et al. (1976), referencing Krey et al. (1975), states that the HASL had maintained a good inter-laboratory calibration with the Environmental Analysis Laboratories for plutonium in filter paper samples for many years, and that the accuracy of those analyses had averaged better than 95%.

The quality of analyses for the HASL surface air sampling program in 1974 is discussed in Toonkel et al. (1976). LFE-Environmental Analysis Laboratories of Richmond, California, performed the radiochemical analysis of ^{239}Pu, as well as some other contaminants, under contract. The percent deviation averaged 4% for ^{239}Pu analyses of all standards and blanks in 1974. Duplicate samples, prepared by splitting an air filter in half, averaged 9.1% deviation from the mean for ^{239}Pu analyses. Mean blank values were ≤0.06 dpm.

Feely et al. (1985) presents 48 pairs of results from reference samples and blanks analyzed radiochemically for ^{239}Pu between February 1980 and November 1983. Reference samples, which contained known quantities of plutonium that had been added to the filter, were submitted to the laboratory along with routine samples. Blank samples were clean filters with no plutonium added. At this time, the plutonium analyses were being conducted by Environmental Analysis Laboratories (EAL) of Richmond, California. Those data are plotted in Figure III-60.

At the low levels encountered in the environment, the uncertainty in determining the amount of plutonium on the air filters can be largely due to counting error. This source of error was discussed earlier in this section with respect to determination of gross alpha activity on air filters. It was standard reporting practice for the HASL/EML to code their data as follows:

A – one standard deviation of the counting error is between 20% and 100%

B – one standard deviation of the counting error exceeds 100%. (The actual result is omitted from the tables when the error is this great.)

If there is no code letter next to the datum, the counting error was less than 20%. For the monitoring performed by HASL/EML near Rocky Flats between June 1970 and September 1981 (data tabulated in Table B-2), only three of the 272 ^{239}Pu measurements had an analytical error between 20 and 100%; the rest were <20%.

In a special study conducted by HASL in the summer of 1973, the average accuracy of plutonium analyses on air filters was within 2% (Krey et al. 1974). However, the precision of field replicates (side-by-side samplers) was very poor–the average percent deviation between duplicates for the plutonium activity concentration in air, averaged over a 4-day interval, reached as high as a factor of 5. The authors attributed this to discrete, infrequent particles in the tens of micron size range that were not evenly mixed in the surface atmosphere. These hot particles rendered short-term measurements uncertain and possibly not representative of average conditions. However, the authors noted that weather shelters used for most long-term ambient air monitors would discriminate against these larger particles and minimize the hot particle problem.

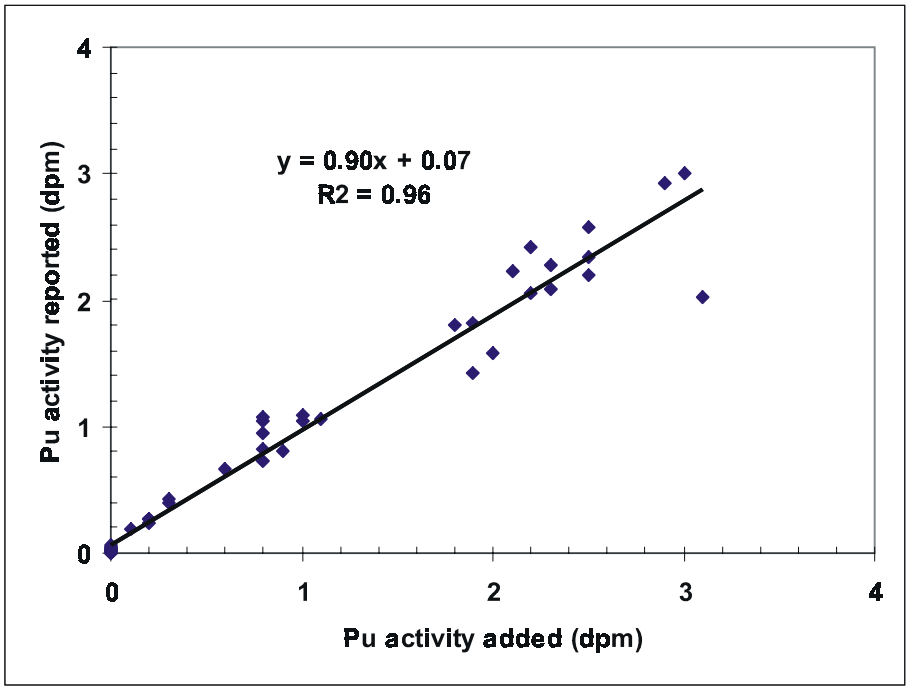


Figure III-60. Results of radiochemical analyses of ^{239}Pu in reference samples and blanks by the EML contractor laboratory (EAL, Inc.) between February 1980 and November 1983 (data from Feely et al. 1985). One outlier was omitted, a reference sample in which 1.1 dpm was added but the laboratory reported 4.15 dpm.

Data Reduction Procedures

An example of a data reduction procedure is how average concentrations are generated from individual measurements. This procedure has varied over the history of the Rocky Flats environmental monitoring program.

Paricio (1985) compiled the history of reporting negative values in the ambient air monitoring program. "Blank" values of radioactivity were subtracted from the total counted activity of a dissolved air filter to correct for the background activity of the solvent and the counting apparatus. Negative values could result if the blank value was greater than the activity of the sample. The following notes from Paricio (1985) are relevant to interpreting air monitoring data for the historical public exposures studies:

1976 – The current network of ambient air samplers was established. The results were not corrected with blank values.

1980 – HS&E Labs started correcting sample data with blank values. However, any negative values accrued during this year were reported as less than the Minimum Detectable Activity (MDA) and are not traceable.

1981 – The practice of reporting negative values was instituted in the Monthly Environmental Monitoring Report. However, no negative values were reported for 1981.

In the February 1989 Monthly Environmental Information Summary, the footnote to Table I states:

> The plutonium, uranium, americium, and beryllium measured concentrations in this report include values that are less than the corresponding calculated minimum detectable concentrations (MDCs). In some cases, the values are less than zero. This method of reporting began in January 1981. These negative values result when the measured value for the laboratory reagent blank is subtracted from an analytical result which was measured as a smaller value than the reagent blank. This may happen when measuring concentrations which are very close to zero.

Examples: Correction of Total Long-Lived Alpha Counts for Uncertainty and Bias

The previous sections of this chapter have discussed a number of sources of bias and uncertainty for air monitoring data and have recommended approaches for correcting the data. This section presents three examples that illustrate the correction of a daily alpha count. Each example begins with the net count from the daily air filter and the computation of an uncorrected concentration of alpha activity in air. Then, a corrected air concentration is produced, in the form of an uncertainty distribution. The correction incorporates bias and uncertainty in the following parameters:

- Volume of air sampled (sampler calibration and effect of filter clogging)
- Inlet collection efficiency for three wind speeds and four particle sizes
- Filter collection efficiency
- Alpha-particle absorption by filter (incorporated into total efficiency factor distribution, see previous section)
- Counting error.

The first example assumes a moderately high count rate that is associated with moderate winds and equal apportioning of the alpha activity among four particle size categories. The second example describes a high-wind condition in which more of the airborne activity is associated with larger particles. In addition to the normal uncertainty in the volume of air sampled, a bias in the volume of air sampled is shown to illustrate the possible effect of filter clogging. The third example assumes a low but positive count rate associated with low wind speed and relatively more of the airborne activity associated with small particles.

An uncertainty distribution was generated for the corrected air concentration for each example. These are shown as charts in Figures III-61 through III-63. The ratio of the uncorrected air concentration, which would have been reported by the RFP for that net count rate, to the 50th percentile of the corrected concentration distribution is computed for each example. This is a measure of average bias. The amount of uncertainty in the corrected air concentrations is described by the ratios of the 50th percentile to the 5th and 95th percentiles. A sensitivity chart is included to show the input parameters that have the greatest contribution to the uncertainty in the corrected air concentration (Figures III-61 through III-63).

The three most important contributors to uncertainty are the volume of air sampled, the total efficiency factor (which incorporates the uncertainty due to alpha-particle absorption in the air filter), and counting error. Uncertainty from counting error dominates uncertainty for lower count

samples (example #3), but it is small relative to the other two contributors for high-count samples (example #2). Although not explicitly reflected in these examples, another critical input assumption is the distribution of activity among various airborne particle sizes. The bias is small for the low-wind/low-activity example (example #3), but the uncertainty in the concentration is large.

The bias is largest for example #2. This most closely represents the high daily counts that were examined for assessing primary releases from the 903 Area. The average bias in this example is 12%; that is, the uncorrected concentration is only 12% of the activity that would have been associated with airborne particle sizes of 20 μm or less. These particles are small enough to be transported by air, but they are not all respirable. The bias in estimating the respirable particle concentrations would be less, because those particles are more efficiently collected by the sampler inlet.

These examples show how an air sampler count might be corrected for three different conditions. Weber et al. (1999) explains the actual corrections used for S-8 sampler data in the 903 Area source term development.

Example #1. Rocky Flats Low-Volume Sampler, Daily Alpha Count
Moderately High Count Rate, Moderate Winds, Equal Activity Distribution Among Four Particle Sizes

Input assumptions:

Net count: 20 cpm
Counting error: coefficient of variation, 15% (Table III-21)

Fraction of activity associated with particle sizes:

< 5 μm 0.25
10 μm 0.25
15 μm 0.25
20 μm 0.25

Sampler inlet collection efficiency (Table III-23):

< 5 μm 95–100%
10 μm 80–90%
15 μm 60–75%
20 μm 45–55%

Filter collection efficiency:

99–100%

Volume of air sampled:

81.5 m^3, standard deviation 10%

Total efficiency factor for alpha counting:

best estimate, 0.21 cpm per dpm
minimum, 0.15; maximum, 0.25

Uncorrected activity concentration:

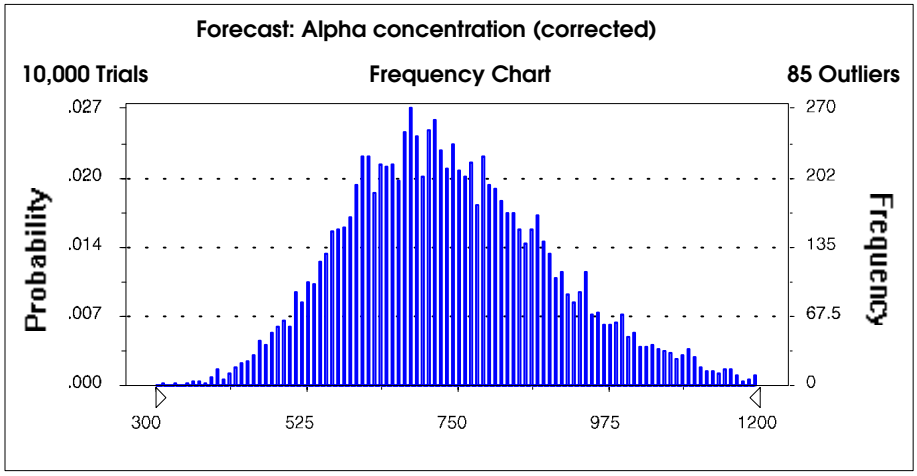
$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / [(0.21 \text{ cpm dpm}^{-1}) \times (2.22 \text{ dpm pCi}^{-1}) (10^{-3} \text{ pCi fCi}^{-1}) (81.5 \text{ m}^3)]$$

or,

$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / 0.038 .$$

Therefore,

$$\text{TLLa activity concentration} = 20 \text{ cpm} / 0.038 = \mathbf{530 \text{ fCi m}^{-3}}$$



Corrected air concentration
Percentiles of uncertainty distribution:

	(fCi m^{-3})
5%	510
10%	550
50%	730
90%	950
95%	1030

Spread in uncertainty distribution:
$5^{th}/50^{th}$ percentile = 0.70
$95^{th}/50^{th}$ percentile = 1.3

Average bias: 530 (uncorrected) / 730 (corrected) = 73%

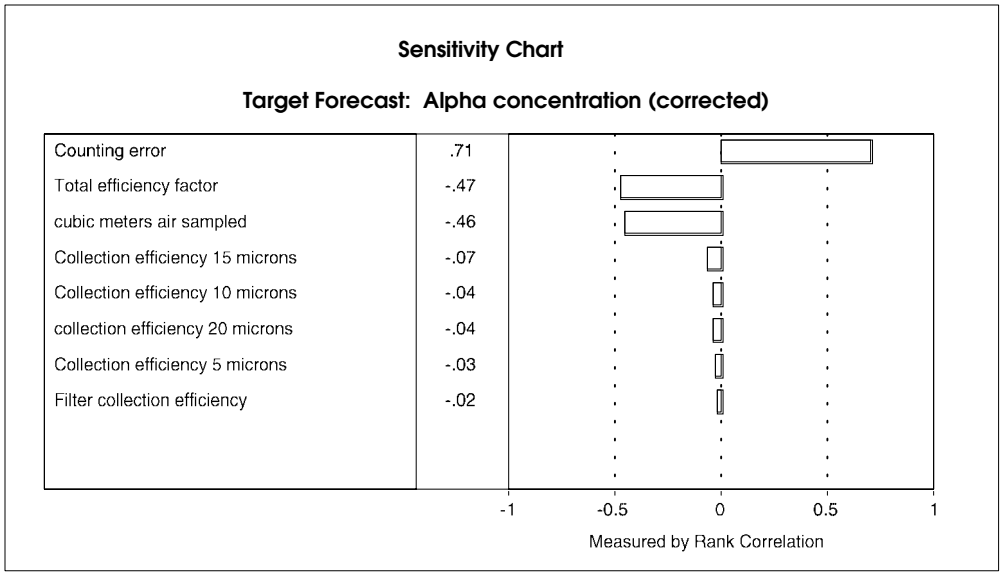


Figure III-61. Example #1, correction of alpha count for sources of bias and uncertainty, and sensitivity of input parameter uncertainty.

Example #2. Rocky Flats Low-Volume Sampler, Daily Alpha Count
High Count Rate, High Winds, Filter Dust Loading, Activity on Predominantly Large Particles

Input assumptions:

Net count: 500 cpm
Counting error: coefficient of variation, 2% (Table III-21)

Fraction of activity associated with particle sizes:

< 5 μm 0.10
10 μm 0.10
15 μm 0.10
20 μm 0.70

Sampler inlet collection efficiency (Table III-23):

< 5 μm 95–100%
10 μm 50–55%
15 μm 10–25%
20 μm 0%

Filter collection efficiency:

99–100%

Volume of air sampled:
In this example, high dust conditions result in filter clogging, which reduces the volume of air sampled below the nominal value of 81.5 m^3. In this simulation, the distribution was triangular with a best estimate of 60 m^3, and minimum and maximum volumes of 40 and 85 m^3.

Total efficiency factor for alpha counting:

best estimate, 0.21 cpm per dpm
minimum, 0.15; maximum, 0.25

Uncorrected activity concentration:

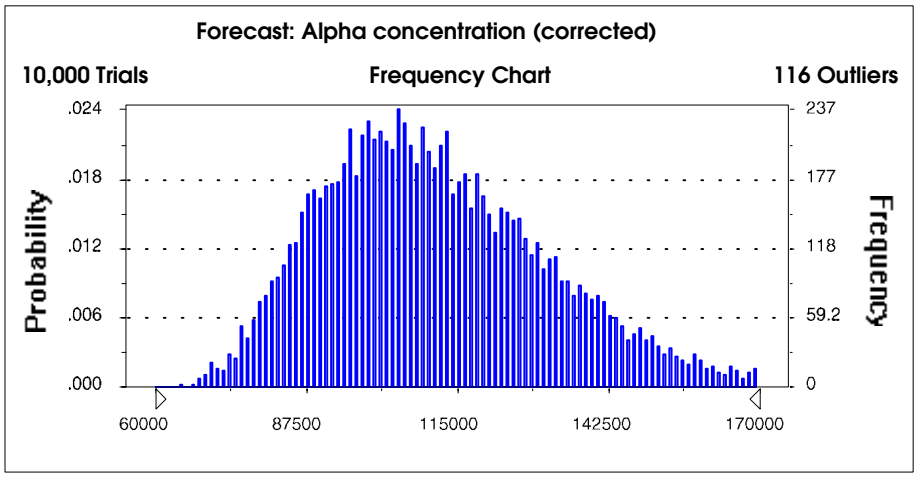
$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / [(0.21 \text{ cpm dpm}^{-1}) \times (2.22 \text{ dpm pCi}^{-1}) (10^{-3} \text{ pCi fCi}^{-1}) (81.5 \text{ m}^3)]$$

or,

$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / 0.038 .$$

Therefore,

$$\text{TLLa activity concentration} = 500 \text{ cpm} / 0.038 = \mathbf{13{,}200 \text{ fCi m}^{-3}}$$



Corrected air concentration
Percentiles of uncertainty distribution:

	fCi m^{-3}
5%	81,600
10%	86,700
50%	109,000
90%	140,000
95%	150,000

Spread in uncertainty distribution:
$5^{th}/50^{th}$ percentile = 0.75
$95^{th}/50^{th}$ percentile = 1.4

Average bias: 13,200 (uncorrected) / 109,000 (corrected) = 12%

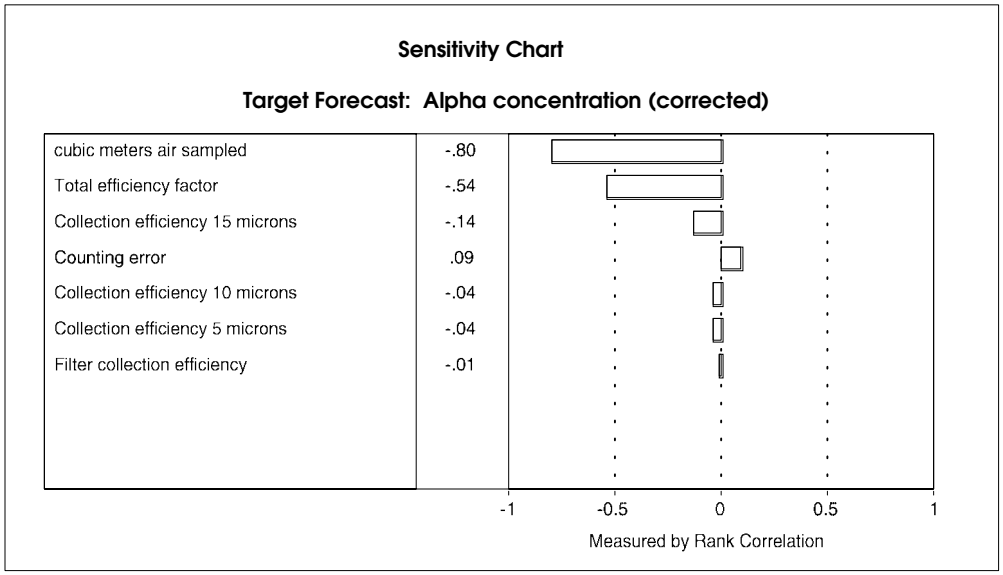


Figure III-62. Example #2, correction of alpha count for sources of bias and uncertainty, and sensitivity of input parameter uncertainty.

Example #3. Rocky Flats Low-Volume Sampler, Daily Alpha Count
Low Count Rate, Low Winds, and Predominantly Small Particles

Input assumptions:

Net count: 0.8 cpm
Counting error: coefficient of variation, 40% (Table III–21)

Fraction of activity associated with particle sizes:

<5 µm 0.50
10 µm 0.25
15 µm 0.15
20 µm 0.10

Sampler inlet collection efficiency (Table III-23):

<5 µm 95–100%
10 µm 95–100%
15 µm 95–100%
20 µm 95–100%

Filter collection efficiency:

99–100%

Volume of air sampled:

81.5 m^3, standard deviation 10%

Total efficiency factor for alpha counting:

best estimate, 0.21 cpm per dpm
minimum, 0.15; maximum, 0.25

Uncorrected activity concentration:

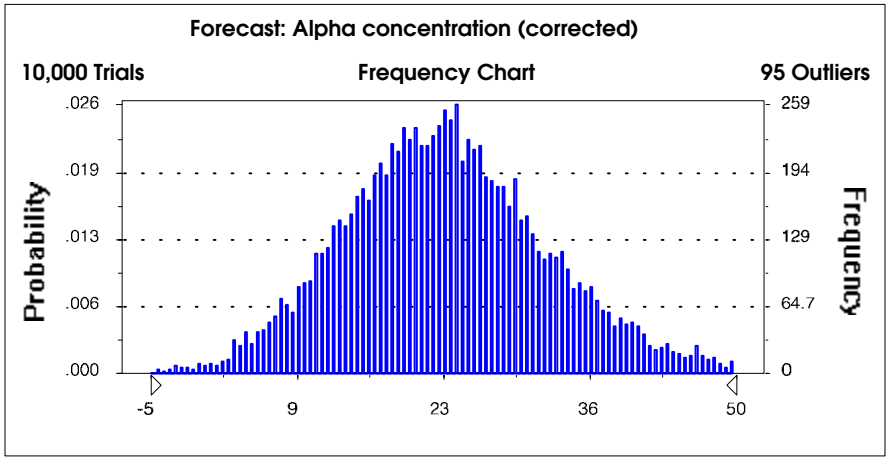
$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / [(0.21 \text{ cpm dpm}^{-1}) \times (2.22 \text{ dpm pCi}^{-1}) (10^{-3} \text{ pCi fCi}^{-1}) (81.5 \text{ m}^3)]$$

or,

$$\text{TLLa activity concentration (fCi m}^{-3}) = (\text{TLLa cpm}) / 0.038 .$$

Therefore,

$$\text{TLLa activity concentration} = 0.8 \text{ cpm} / 0.038 = \mathbf{21 \text{ fCi m}^{-3}}$$



Corrected Air Concentration
Percentiles of uncertainty distribution:

	(fCi m^{-3})
5%	8
10%	11
50%	23
90%	36
95%	40

Spread in uncertainty distribution:
$5^{th}/50^{th}$ percentile = 0.35
$95^{th}/50^{th}$ percentile = 1.7

Average bias: 21 (uncorrected) / 23 (corrected) = 91%

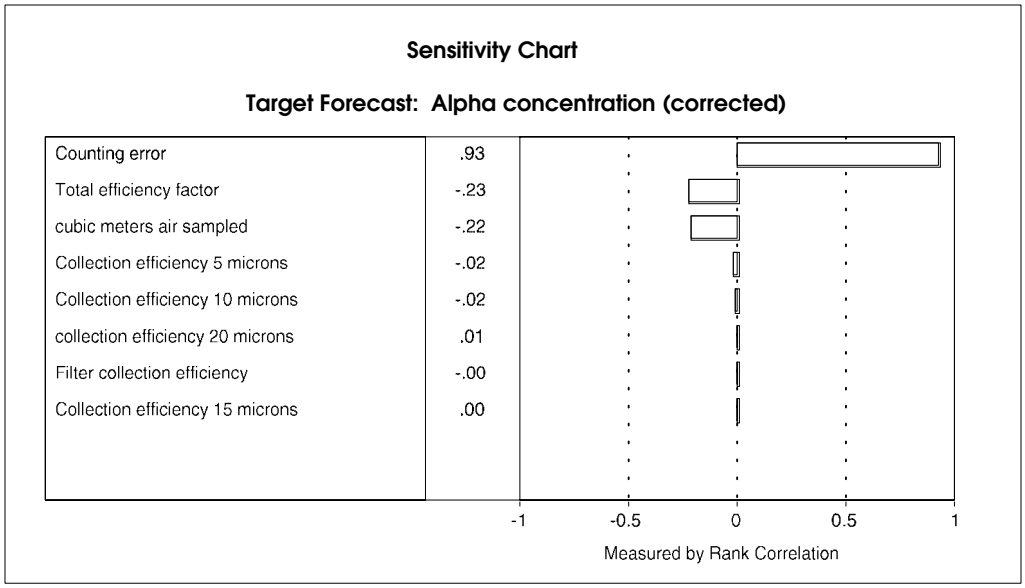


Figure III-63. Example #3, correction of alpha count for sources of bias and uncertainty, and sensitivity of input parameter uncertainty.

SUMMARY

Based on the framework outlined in Chapter I, a number of sets of air monitoring data were identified as likely to be useful to the historical public exposures study. These data were evaluated in this report, previous drafts of which served as a reference for other source term and risk reports produced in Phase II. Those data sets that were ultimately used are summarized in Table III-26.

Table III-26. Application of Air Monitoring Data in the Historical Public Exposures Studies on Rocky Flats, Phase II

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

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

In Table III-26, onsite air sampling refers to locations within the industrial area. Note that there are no useful data offsite during the time of greatest interest (before 1970) that could be used to compare with predicted concentrations. Comparison with predictions was possible in the 1970s and 1980s using data from several agencies (RF contractor, CDH, and HASL). These comparisons were made on an annual average time resolution, with an estimate of fallout background subtracted from the measured values. The data used in this comparison are tabulated in Appendix B, Table B-15. Some data were not used quantitatively in source term development or risk assessment, but do illustrate fundamental points that influenced our decisions in those areas. Those data are listed in Table III-26 as well.

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