

<p>ORAU Team NIOSH Dose Reconstruction Project</p> <p>Technical Basis Document for the Rocky Flats Plant – Occupational Environmental Dose</p>	<p>Document Number: ORAUT-TKBS-0011-4 Effective Date: 06/29/2004 Revision No.: 01 Controlled Copy No.: _____ Page 1 of 56</p>
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RECORD OF ISSUE/REVISIONS

ISSUE AUTHORIZATION DATE	EFFECTIVE DATE	REV. NO.	DESCRIPTION
Draft	09/19/2003	00-A	Technical Basis Document for the Rocky Flats Plant – Occupational Environmental Dose. Initiated by Robert Meyer.
Draft	11/11/2003	00-B	Incorporates OCAS and ORAU comments. Initiated by Robert Meyer.
Draft	11/26/2003	00-C	Incorporates second set of OCAS comments. Initiated by Robert Meyer.
Draft	12/17/2003	00-D	Incorporates OCAS comment from 12/17/2003. Initiated by Robert Meyer.
1/06/2004	01/06/2004	00	First approved issue. Initiated by Robert Meyer.
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Draft	05/07/2004	01-B	Averaged two 1973 Pu and Am intake values to provide one annual estimate; changed FEIS reference citation on p. 8, and corrected reference list for 2 citations. Initiated by Robert Meyer
Draft	05/24/2004	01-C	Includes changes recommended by reviewers Naimon and McGolerick. Initiated by Robert Meyer.
06/29/2004	06/29/2004	01	Approved issue of Revision 01. Initiated by Robert Meyer.

ACRONYMS AND ABBREVIATIONS

α	alpha particle
AED	Aerodynamic Equivalent Diameter
AMAD	Activity Median Aerodynamic Diameter
Am	americium
CDH	Colorado Department of Public Health and Environment (previously Colorado Department of Health)
Ci	curie
DOE	Department of Energy
EEOICPA	Energy Employees Occupational Illness Compensation Program Act
EPA	Environmental Protection Agency (U.S.)
H	hydrogen
hr	hour
ICRP	International Commission on Radiological Protection
μm	micrometer
m^3	cubic meter
mrem	millirem
NIOSH	National Institute for Occupational Safety and Health
pCi	picocurie
Pu	plutonium
RAC	Risk Assessment Corporation
RATCHET	Regional Atmospheric Transport Code for Hanford Environmental Tracking
RFETS	Rocky Flats Environmental Technology Site
RFP	Rocky Flats Plant
TBD	Technical Basis Document
TLD	Thermoluminescent Dosimeter
TLL α	total long-lived alpha
yr	year

4.1 INTRODUCTION

4.1.1 Purpose

Technical Basis Documents and Site Profile Documents are general working documents that provide guidance concerning the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH in the completion of the individual work required for each dose reconstruction.

In preparing this report, available information about the RFP environment dating back to 1952 was used. The user is strongly advised to consider the remarks in the text on appropriate use and interpretation of the material.

In this document the word "facility" is used as a general term for an area, building or group of buildings that served a specific purpose at a site. It does not necessarily connote an "atomic weapons employer facility" or a "Department of Energy facility" as defined in the Energy Employee Occupational Illness Compensation Program Act of 2000 (42 U.S.C. § 7384I (5) and (12)).

4.1.2 Content

The term *occupational environmental dose* refers to the radiation dose received in the course of work duties, outside plant buildings, but on the Rocky Flats Plant (RFP) site. Internal and external exposures to radionuclides in the outdoor environment are considered separately here in calculating this dose. Estimated occupational environmental dose may be utilized when a worker was not monitored adequately to develop a reliable individual dose.

Section 4.2 presents information necessary to estimate internal environmental dose. First it identifies the radionuclides of concern. Screening the list of radionuclides relied on work done for the Historical Public Exposure Studies on Rocky Flats (ChemRisk 1994a; Rood and Grogan 1999). Section 4.2 discusses the resulting source terms (release rates) for radionuclides considered potentially significant to internal environmental dose and provides estimated annual inhalation intake activities of radionuclides.

Section 4.3 contains information necessary for estimating external environmental dose. Ambient doses, taken from annual environmental reports for RFP published from 1975 through 1995, were developed for the site. The reports summarize external dose as measured by thermoluminescent dosimeters (TLDs) at the Plant, its general environs, and selected nearby communities.

Section 4.4 considers uncertainties in the information provided for estimating occupational environmental dose. The discussion addresses sources of uncertainty, and provides quantitative information where possible.

4.2 INTERNAL DOSE FROM ONSITE ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS

4.2.1 Radionuclides of Concern

The Historical Public Exposures Studies on Rocky Flats were conducted in the 1990s as part of a 1989 Agreement in Principle between DOE and the State of Colorado to identify potential health effects in nearby communities exposed to past releases. Phase I and Phase II of the studies provided comprehensive analyses of releases of radionuclides from RFP during its operational period, which ran from 1952 to 1994 (ChemRisk 1994a; Voillequé 1999a; Voillequé 1999b; Voillequé 1999c; Weber et al. 1999). The releases analyzed are believed to be the best estimates available, and are used here to estimate air concentrations on the plant site.

The Phase I study identified several radionuclides as potentially significant releases – Hydrogen-3, natural thorium, enriched and depleted uranium, $^{239/240}\text{Pu}$, ^{241}Pu , and ^{241}Am . The results of the Phase I study identified plutonium as the primary material of concern with respect to off-site exposures.

The Phase II study focused the air-pathway exposure assessment on the isotopes of plutonium. The potential importance of ^3H , enriched and depleted uranium, and ^{241}Am to onsite exposures was reevaluated to determine if previous determinations of insignificance were relevant for occupational exposures. Releases of natural thorium were difficult to quantify, but were probably insignificant based on a review of processes and ventilation filter use (ChemRisk 1994a, Task 5, page 123).

To evaluate the potential significance of radionuclides other than isotopes of plutonium, an estimate was developed of annual inhalation dose using airborne stack emission estimates from the Phase I study, the air dispersion modeling results from Phase II, an assumed inhalation rate of 2,400 m³/yr (ICRP 1975), and dose factors from the International Commission on Radiological Protection (ICRP) Publication 72 (ICRP 1996). Attachment 4A contains a summary of the atmospheric dispersion model. Attachment 4B discusses the methods and results of this screening evaluation. Briefly, the maximum inhalation dose in any given year since 1953 for any of these radionuclides was estimated to be that for ^{241}Am , on the order of 2 mrem for the stack releases. The maximum predicted annual doses for ^3H , depleted uranium, and enriched uranium were 0.05 mrem, 0.5 mrem, and 0.3 mrem, respectively. The Phase I study estimated that ^{241}Am is present, on average, at up to 0.30 times the alpha activity of $^{239,240}\text{Pu}$ in the 903 Area due to ingrowth of ^{241}Am from ^{241}Pu in the cutting oil stored in that area and in soils contaminated with the cutting oil (Appendix H, ChemRisk 1994b). Therefore, ^{241}Am associated with soil resuspended from the 903 Area was considered a contributor to dose, but stack releases of ^{241}Am , ^3H , and the uranium isotopes were excluded as contributors.

The Final Environmental Impact Statement for the RFP (DOE 1980) indicates an isotopic composition of Pu by weight as follows:

Isotope	Percent by weight	Percent of Pu alpha activity ^a
Pu-238	0.01	2.33
Pu-239	93.79	79.62
Pu-240	5.80	18.04
Pu-241	0.36	(beta emitter)
Pu-242	0.03	.00161

a. From ChemRisk 1994a (p. 105, Table 2-34).

Plutonium-238 and ^{242}Pu are minor contributors to the alpha activity of plutonium expected in the RFP environment. Therefore, the following sections report source terms and intakes for $^{239,240}\text{Pu}$.

4.2.2 Source Terms

The discussion of atmospheric source terms for isotopes of $^{239,240}\text{Pu}$ and ^{241}Am released to the RFP environment addresses two phases: the pre-1993 phase, when production activities were ongoing, and the post-1992 phase, when production activities had ceased, and releases were more likely to occur as a result of past contamination or decontamination and decommissioning activities.

4.2.2.1 Pre-1993 Source Terms

Between 1953 and 1993, fabrication and recycling activities at RFP resulted in radionuclide releases to the onsite atmospheric environment as a result of "routine" operations (continuous releases), and "nonroutine incidents" (discrete events). The term "routine" was used in the Radiological Assessments Corporation (RAC) Phase II dose reconstruction reports to distinguish continuous releases from the release spikes that occurred during the 1957 and 1969 fires and during 903 Area high-wind events.

The most significant discrete release occurred during and shortly after September 11, 1957. A glovebox fire in Building 71 (now called Building 771) resulted in a release of plutonium estimated at 21 curies (50th-percentile estimate, Voillequé 1999a). The next most significant discrete release location was the 903 Drum Storage Area (also called the 903 Pad or 903 Area). An estimated 3.1 curies (50th percentile) of plutonium (<30 μm size fraction) were released from the 903 Pad over several years, mainly as a result of mechanical disturbance and wind action (Weber et al. 1999). This estimate is comprised of the 24-hour integrated release quantities for 24 identified discrete events during the 1964-to-1969 period. An asphalt pad placed over the area in 1970 decreased the source term dramatically, although resuspension of downwind contaminated soil continued to disperse plutonium to the air in later years. The third most significant nonroutine contaminant release occurred during the May 11, 1969, fire, when approximately 0.037 Ci (37 mCi) of plutonium were released from the Building 779 stack (Voillequé 1999b).

Total routine (non-discrete-event) plutonium emissions from 1953 to 1989 are estimated to be on the order of 0.12 Ci (Voillequé 1999c). This estimate does not include releases due to resuspension of contaminated soil downwind of the 903 Pad.

4.2.2.2 Post-1992 Source Terms

After 1992, production operations at the RFP officially ceased. However, source terms of plutonium to the environment still exist due from contaminated soils and due to the presence of plutonium contamination within buildings at the site. Air monitoring continues to be carried out by the RFP contractor and the Colorado Department of Public Health and Environment (CDH). It is reasonable to expect that atmospheric concentrations will not increase unless decommissioning activities mobilize previous contamination.

4.2.3 Annual Intake of Radioactivity

To calculate annual intake of $^{239,240}\text{Pu}$ and ^{241}Am , the estimated annual average air concentrations of these isotopes in the RFP environment were multiplied by an annual inhalation rate. The assumed intake rate was 2,400 m^3/yr (ICRP 1975), corresponding to an hourly rate of 1.2 m^3 , for light activity and a 2,000-hr work year. Intake can be scaled for increased inhalation rates or other than 2,000 hours of exposure.

Air concentrations of $^{239,240}\text{Pu}$ and ^{241}Am were estimated as described in Attachment 4A. Onsite air monitoring data are the preferred source of air concentrations for $^{239,240}\text{Pu}$. After 1964, suspension or resuspension (Rood and Grogan 1999, p. 57) of contaminated soil was the main source of plutonium to onsite air. Thus, activity concentrations of ^{241}Am can be estimated after 1964 by assuming that the concentration of ^{241}Am is 30% of the $^{239,240}\text{Pu}$ concentration (ChemRisk 1994b, p. H-1).

Attachment 4A describes the use of atmospheric dispersion modeling results from the Phase II study to estimate air concentrations and intake between 1953 and 1964. During this period, air monitoring results were not readily available and usually did not allow derivation of $^{239,240}\text{Pu}$ concentrations. After 1964, air monitoring data provided either total long-lived alpha (TLL α) concentrations, from which $^{239,240}\text{Pu}$ values could be derived, or actual measurements of $^{239,240}\text{Pu}$. The annual environmental reports (Dow 1972 through 1975; Rockwell 1976 through 1989; EG&G 1989 through 1993; Kaiser-Hill 1994) were often especially useful in providing annual summaries of air concentrations by sampler location, as well as maximum site concentrations based on monthly reporting.

Table 4-1 lists estimated annual intakes of $^{239,240}\text{Pu}$ between 1953 and 1964, based on atmospheric modeling described in Attachment 4A. The values are expressed in pCi/yr. One pCi/yr is equivalent to 10^{-12} Ci/year. The calculated intakes represent an average for six computational nodes in the RFP industrial area, and are exclusive of the buffer zone (Figure 4-1). Table 4-1 includes the 50th- and 95th-percentile estimates associated with particles less than 15 μm aerodynamic equivalent diameter (AED) which is an upper limit for respirable particles according to Rood and Grogan (1999, page iv). Since this likely includes particles that are larger than ICRP 66 (1994) default respirable size of 5 μm AMAD, use of the intakes in Table 4-1 are likely to be claimant-favorable.

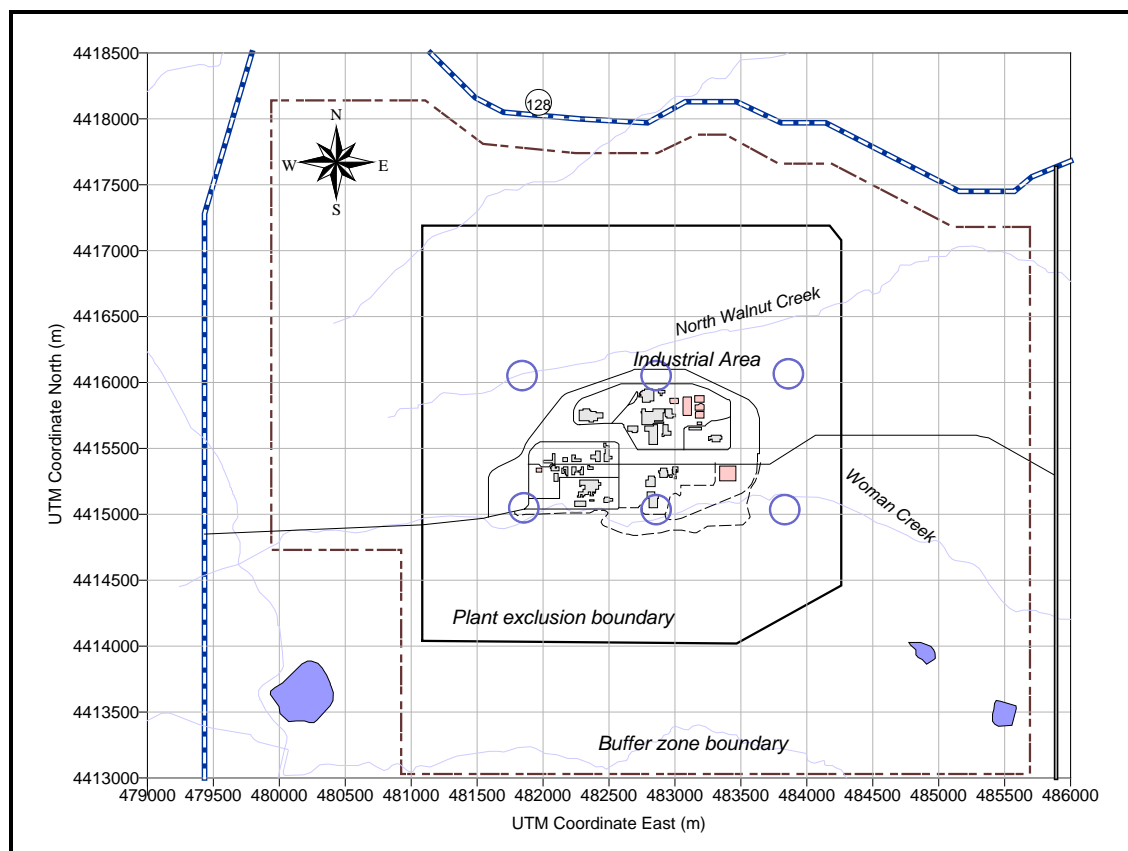


Figure 4-1. Grid map used in atmospheric modeling, showing locations of six computational nodes representing the RFP Industrial Area.

Table 4-1 lists two results for 1957. Results for "1957" include the September 11 fire, which caused the annual intakes to be substantially higher than the years before and after. If a worker was known to have been present during that year, and that individual might have been present in September, it is appropriate to use this value. The results for "1957, w/o fire" can be used for individuals who worked a partial year that did not include the month of September. Elevated releases of $^{239,240}\text{Pu}$ attributable to the 1957 fire occurred over several hours after the fire began at approximately 10:00 p.m. on September 11. Exposure to the direct releases would have occurred during that month (the air concentrations associated with soil-deposited $^{239,240}\text{Pu}$ from the fire are included in both 1957 values by considering resuspension of contaminated soil in the dispersion modeling).

In Table 4-1, the 50th and 95th percentiles represent the median and 95th percent estimates of concentration for 500 Monte Carlo model realizations simulated for each year. The distribution associated with the percentiles is not readily described as either normal or lognormal. The proper interpretation of the 95% confidence limits about the "average" (50th percentile) and "maximum" (95th percentile) in Table 4-1 is discussed in Section 4.4.1.1. Briefly, these limits describe the degree of confidence that the percentile value of the sampled distribution represents the true percentile value of the underlying distribution.

Table 4-2 lists estimated annual intakes of $^{239,240}\text{Pu}$ and ^{241}Am between 1965 and 2002. The average values for $^{239,240}\text{Pu}$ in this table are based on measured concentrations at samplers across the site, but often are biased toward the highest concentrations, typically near the 903 Area after 1970 because concentrations are more often reported for samplers in that area and not for samplers in other RFP areas. Therefore, the average values are inherently claimant-favorable when used as an average for the industrial area. The maximum values listed in Table 4-2 represent the largest monthly concentrations identified for the same samplers. The $^{239,240}\text{Pu}$ air concentrations supporting the intakes in Table 4-2 are described in Attachment 4A and summarized in Table 4A-2. For 1999 and later, intakes are assumed to be the maximum values over the last 20 years, due to the lack of onsite monitoring data readily available (see Attachment 4A). Intakes were calculated from air concentrations by assuming a respirable fraction of 1.0, despite the reported values of this parameter ranging from 0.2 to 0.4 in a review of the subject in Rope et al. 1999, pp. III-79 to III-81). Low- and high-volume air samplers at the RFP collected particles larger than respirable (Rope et al., 1999, pp. III-140 to 105), but to varying degrees as samplers were replaced over the years. Since a defensible respirable fraction is not readily identifiable, a claimant-favorable respirable fraction of 1.0 was assumed.

The values reported for ^{241}Am in Table 4-2 (after 1970) were calculated by multiplying the $^{239,240}\text{Pu}$ intake values by 0.30, in accordance with the rationale described above in this section. Again, a claimant-favorable respirable fraction of 1.0 was assumed.

Annual intakes estimated in Tables 4-1 and 4-2 are based on a 2,400-m³/year inhalation rate, but can be scaled to a different rate. Furthermore, the values can be scaled for partial-year exposures, with the following cautionary note. For 1957, 90% or more of the intake of $^{239,240}\text{Pu}$ can be attributed to the September 11 fire. Thus, if a worker was present on the site for only a portion of 1957, but during September, the entire annual intake should be assumed. If a worker was present on site in 1957, but not during September, the intake designated "1957 w/o fire" should be assumed, and scaling of this value for a partial-year exposure can be done.

The assumed solubility of inhaled $^{239,240}\text{Pu}$ and ^{241}Am considers the following information. Plutonium in metal-working operations and involved in fires is generally insoluble (Class S or slowly transportable in the lungs) or highly insoluble (super Class S). Exceptions, such as plutonium metal associated with solvents, may exist. Plutonium in chemical processing operations can be either

soluble (Class M or moderately transportable in the lungs) or insoluble. A claimant-favorable approach is to assume insoluble plutonium (Class S) for qualifying cancers associated with the respiratory system, and soluble plutonium (Class M) for all other cases.

4.3 EXTERNAL EXPOSURE TO ONSITE ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS

4.3.1 Radionuclides of Concern

As described in Section 4.2.1, ^3H , natural thorium, enriched and depleted uranium, $^{239/240}\text{Pu}$, ^{241}Pu , and ^{241}Am were identified as having been released to the air during the RFP operational period. However, since none of the isotopes of concern are strong gamma emitters, there is little external dose.

4.3.2 Exposure Rates

Prior to 1975, external exposure of workers was determined by the use of film badges, not TLDs (see Section 6.2, Occupational External Dosimetry). Film badges are not well suited to environmental monitoring, and environmental measurements prior to 1975 are not readily available. The determination concerning this lack of data is consistent with other site evaluations. Rope et al. (1999) point out that prior to the early 1970s, environmental data were fewer and of lower quality than later data. In his review of RFP (covering 1952 to 1982), Putzier (1982) spends little time discussing external occupational exposure measurements and none describing outdoor measurements. This is largely because the concern regarding environmental exposures to workers or the public was centered on plutonium. Rope et al. (1999) describe external gamma exposures, but the information is limited to an analysis of aerial survey data that target plutonium by measuring ^{241}Am .

Data that can be used to estimate external environmental dose exist in annual environmental reports for 1975 through 1994 (Dow 1972 through 1975; Rockwell 1976 through 1989; EG&G 1989 through 1993; Kaiser-Hill 1994). Gamma exposure rates were routinely measured at 12 to 15 locations on the site. Those data are plotted in Figure 4-2 and summarized in Table 4-3. The locations of the monitoring stations were not available; however, because the estimate of the potential external gamma dose to workers is based on the annual average for all locations and all measurement periods, specific locations are not essential to the analysis.

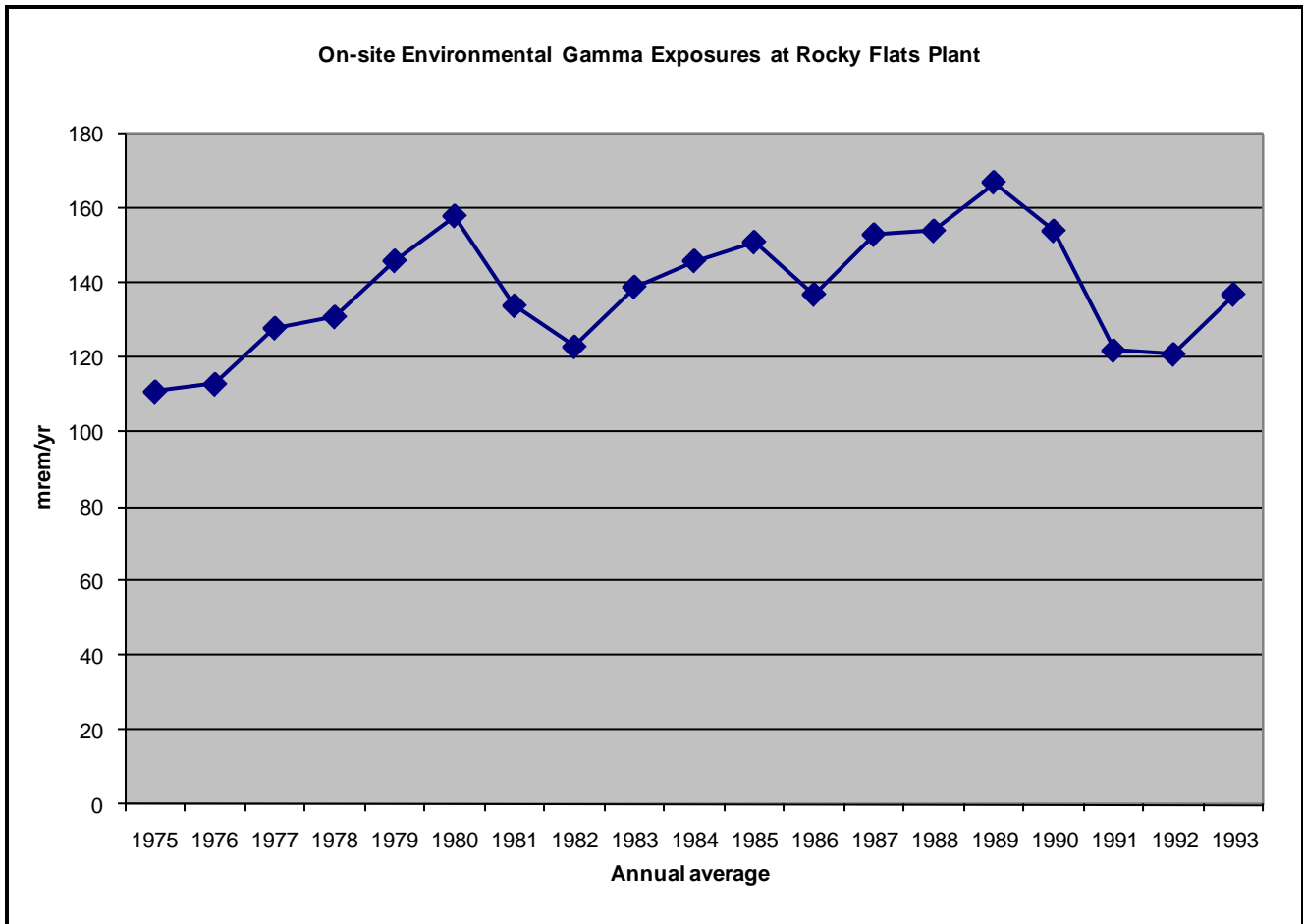


Figure 4-2. External dose (mrem/yr) at the RFP as a function of year..

For the 19-year period from 1975 through 1993, the number of independent measurements ranged from 87 to 176 per year. Table 4-3 lists the standard deviation of the measured samples for these years with the exception of 1977. For 1977, the table lists the standard deviation of the 19 annual measurements. There is no explanation available for the relatively large standard deviations in 1975, 1976, and 1978. The listed exposures are for an entire year (8,760 hours). For claimants whose work history encompasses 1975 through 1993, the annual value in Table 4-3 should be prorated by the number of hours worked during the year.

There is no significant trend ($\alpha = 0.05$) to the means plotted in Figure 4-2. As mentioned above, effluents from RFP were largely alpha-emitting radionuclides with no substantial gamma component, with the exception of ^{241}Am . Releases attributable to incidents that might have vented outside buildings, or due to suspension or resuspension of contaminated soil, would not tend to increase the gamma exposure rate appreciably, due to the relatively large contribution of naturally-occurring radioactivity to exposure rates measured. Workers subject to environmental doses from such incidents would be affected to a far greater extent by internally deposited radionuclides. Therefore, it is reasonable to assign the average value of 138 ± 32 mrem per year (8,760 hours) to years not represented on this table. This dose rate represents the total dose, including background, so use of these values in estimating individual worker doses is claimant-favorable.

Ambient on-site gamma is significantly larger ($p < 0.05$) than ambient gamma measured at "perimeter" stations, but only by an average of 9%. This would seem to indicate that about 10 mrem/yr may be

contributed by contamination inside the exclusion zone boundary. Thus, increases in airborne contamination will not necessarily indicate significant increases in total external dose.

Between 1965 and 1970, the large increase in inhalation intake of Pu is due to soil contamination of the 903 Area, prior to paving of that area. It is likely that the external dose rate at that location may have been elevated compared to other locations at the site due to the Am-241 present. Unfortunately, soil concentrations in the barrel storage location are not available, and few soil concentration data are available for the site prior to 1969 (Rood and Grogan, 1999). In an attempt to estimate external dose to an on-site worker from contaminated soils in the vicinity of the 903 storage area, a Pu soil contamination value of 26 uCi/m² is assumed, based on the 1973 site environmental report which reports this value as the maximum observed in 1970 just downwind of the 903 Pad. Using FGR 12 dose conversion factors for surface contaminated soils, assuming that the maximum observed concentration was constant over the entire site, and using the same isotopic ratios as Krey et al. (1976), we calculate doses at 1 m above ground surface of approx. 4 mrem/yr (for year-round exposure).

Another approach taken to estimate the soil concentration in the barrel storage area was based on estimates of the total Pu-239 released. According to Weber et al. (1999), the highest release estimate for Pu-239 in the 903 storage area was on the order of 1000 g, which corresponds to 62 Ci. If one assumes that this is spread over the 550-ft by 475-ft contamination zone of the 903 Area originally described (Weber et al. 1990), a maximum estimate of external dose of 18 mrem/yr for Am-241 is obtained (assuming Krey's isotopic ratio of 4% for Am-241, the FGR 12 dose conversion factor, a depth of contamination of 15 cm, and year-round exposure).

Finally, we attempted to ascertain what other sources of external exposure might be present. Using dose factors in Federal Guidance Report No. 12, we calculated external dose from submersion in air containing Am-241. The Am-241 levels were calculated using isotopic ratios published by Krey et al. (1976) and modified by RAC when developing soil action levels for the RFETS. Doses from Am-241 in that scenario were negligible, less than one µrem/yr. For these reasons, we feel that use of the ambient gamma values for external environmental dose is reasonable for unmonitored workers throughout the time period from 1953 to the present.

4.4 UNCERTAINTY

Uncertainties in estimates of plutonium intake and external dose to an individual employed at the RFP are associated with natural variations in environmental concentrations, lack of precise information about locations and durations of exposures, and limitations of monitoring data. Not all of these uncertainties can be quantified.

4.4.1 Uncertainty in Internal Exposure Estimates

4.4.1.1 Intakes Estimated for 1953 to 1964

The following discussion on uncertainties in the atmospheric dispersion model predictions used to estimate intakes for the period from 1953 to 1964 interprets the pertinent text from a Phase II document by Rood and Grogan (1999). Uncertainties in model calculations of integrated air concentration and intake (Section 4.2.3) arise from model and parameter uncertainty. Model uncertainty arises from the inability of the computational algorithms to describe rigorously and precisely physical processes that govern the behavior of the system, due either to insufficient knowledge of the processes or inability to measure isolated mechanisms driving the processes.

Model uncertainty is often evaluated in a process called "model validation," which compares model predictions to measured parameters that have not been used to calibrate the model.

Parameter uncertainty arises because of lack of knowledge about, or inability to measure accurately, a parameter's true value. A parameter uncertainty analysis requires the specification of probability distributions describing the value of a parameter considered to be uncertain. The assigned distribution of a parameter characterizes the degree of belief that the true but unknown value lies within a specified range of values.

The Phase II modeling effort performed parameter uncertainty analysis and model validation. Input distributions characterizing the source term, fate and transport calculations, and risk coefficients were developed for the model.

The Phase II dispersion modeling for the identified continuous and discrete sources of plutonium to the air at the RFP used Monte Carlo simulations combined with simple random sampling to propagate uncertainty through the model. In simple random sampling, a random value is taken from the distribution specified for each uncertain model parameter, and a single estimate of the desired endpoint is calculated. This process is repeated for a specific number of model realizations. The result is an empirical approximation of the probability distribution of the model output.

In the current application of the Phase II dispersion model, model output (in pCi/yr annual intake) was developed from 500 model realizations each year and categorized into a percentile ranking. The ordered statistics for the 5th and 95th percentiles for 500 model realizations are 25 and 475, respectively. That is, if the output values for 500 realizations are sorted in ascending order, the 5th percentile represents the 25th highest value and the 95th percentile represents the 475th highest value.

A confidence interval can be defined around percentiles of the output distribution using ordered statistics to define an interval where the true value of a given percentile is at a specified level of confidence. The 95% confidence interval around the 5th percentile in terms of the ordered statistics is 15 to 35. The 95% confidence interval around the 95th percentile in terms of the ordered statistics is 465 to 485. This means we are 95% percent confident that 90% of the model output lies between the ordered statistics 15 and 485. The range of values represented by these ordered statistics will vary depending on the distribution. For example, from Table 4-1, the estimated 95th percentile intake for the year 1957 (with the fire) is 3.46E+02, and the 95% confidence intervals around that value range between 1.36E+02 and 4.92E+02. However, while these intervals are useful in understanding the confidence in the percentile values provided, they make for a complicated table, and thus are not provided in the interests of clarity.

The components of uncertainty that appear in the annual intake estimates reflect uncertainty only in the source terms and environmental transport. Such components of uncertainty are real in the sense they can be derived from measured quantities or inferred from historical records. Uncertainty related to exposure scenarios (i.e., location and duration of exposure) were not included in the calculated statistics.

4.4.1.2 Intakes Estimated for 1965 to Present

Uncertainties associated with the reported average annual ^{239,240}Pu air concentrations for the site are provided for 1972 to 1976 and 1989 to 1994 in the annual environmental reports (Dow 1972 through 1975; Rockwell 1976 and 1989; EG&G 1989 through 1993; Kaiser-Hill 1994). For these years, error terms representing two standard deviations about the mean (at the 95% confidence level) are

calculated assuming the measurements were normally distributed. In 1977 and 1978, no error terms are reported because “they can be misleading and (their use) is considered inappropriate for the data in this report” (Rockwell 1978, 1979). These latter two reports state that the sampling methods would result in “nonrandom variations in the measured concentrations”, and the “distribution ... would not be normal.” From 1979 to 1988, error terms are reported, but only by sampler location and not normalized to the site average. The error terms, when reported, are based on counting error alone, and thus represent the minimum error that can be associated with the measurements. Although counting error does contribute to data uncertainty, the uncertainty associated with a given average value for the site will also be a function of variability in environmental conditions as well as a function of both time and location. Thus, the reported confidence limits are not useful in estimating uncertainty associated with a site average because they would undoubtedly underestimate that uncertainty.

4.4.2 Uncertainties in External Dose Estimate

Unquantifiable uncertainty in external exposures results from the lack of readily available data prior to 1975 or after 1993. However, onsite measurements made during that 19-year period indicate no significant upward trend to indicate that plant operations are increasing background. Published data for environmental external measurements have normal statistics associated with them. The data published in Table 4-3 vary considerably in the expression of uncertainty presented in the annual environmental reports. For years prior to 1977, no uncertainty data are available. For other years, the uncertainty varies from as much as 23% (1σ) for 1981, to less than 2% for 1983. We recommend using the maximum uncertainty available in the literature, 23%, which is represented by 1981 data. The annual recommended values of uncertainty, expressed in terms of 1σ, are shown in fourth column of Table 4-3.

Note: The values in Tables 4-1 and 4-2 represent the potential intake for a worker spending 2,000 hours during the year outdoors on the site. The intake should be prorated for exposure times less than 2,000 hours in a particular year. Based on the values in Table 4-1, the 95th percentile intake due to the 1957 fire alone would have been $3.43 \times 10^{+2}$ pCi.

Table 4-1. Annual inhalation intake of ^{239,240}Pu for 1953 to 1964, based on atmospheric modeling (aerodynamic equivalent diameter <15 μm; assume respirable).^a

Year	^{239,240} Pu Intake (pCi/year) ^a	
	Average (50 th Percentile)	Maximum (95 th Percentile)
1953	6.56E-04	5.68E-03
1954	3.10E-03	2.14E-02
1955	2.89E-03	1.83E-02
1956	1.00E-02	6.01E-02
1957	3.33E+00	3.46E+02
1957 w/o fire	4.20E-01	3.21E+00
1958	1.74E-01	1.96E+00
1959	1.14E-01	1.09E+00
1960	8.24E-02	8.01E-01
1961	5.42E-02	4.65E-01
1962	9.69E-02	8.09E-01
1963	1.65E-01	1.14E+00
1964	6.02E-01	2.95E+00

- a. Atmospheric modeling described in Attachment 4A.
- b. A pCi is equivalent to 10⁻¹² Ci.

Table 4-2. Annual inhalation intake of ^{239,240}Pu and ²⁴¹Am (1965 to 2002), based on monitoring data (respirable fraction of sampled particles = 1.0)

Year	Average ^{239,240} Pu intake (pCi/yr) ^a	Maximum ^{239,240} Pu intake (pCi/yr) ^b	Average ²⁴¹ Am intake (pCi/yr) ^c	Maximum ²⁴¹ Am intake (pCi/yr) ^d
1965	2.40E+00	1.20E+01	7.20E-01	3.60E+00
1966	7.20E+00	2.40E+01	2.16E+00	7.20E+00
1967	9.60E+00	8.64E+01	2.88E+00	2.59E+01
1968	1.92E+01	3.10E+02	5.76E+00	9.29E+01
1969	2.16E+01	1.32E+03	6.48E+00	3.95E+02
1970	4.80E+00	5.04E+01	1.44E+00	1.51E+01
1971	4.32E+00	2.35E+02	1.30E+00	7.06E+01
1972	5.04E+00	1.46E+02	1.51E+00	4.39E+01
1973	4.08E+01	3.70E+02	1.22E+01	1.11E+01
1974	1.46E+00	3.84E+01	4.39E-01	1.15E+01
1975	4.80E-01	1.51E+01	1.44E-01	4.54E+00
1976	4.56E-01	9.36E+00	1.37E-01	2.81E+00
1977	6.48E-01	6.72E+00	1.94E-01	2.02E+00
1978	6.00E-01	8.40E+00	1.80E-01	2.52E+00
1979	4.08E-01	3.84E+00	1.22E-01	1.15E+00
1980	3.60E-01	5.28E+00	1.08E-01	1.58E+00
1981	4.32E-01	4.80E+00	1.30E-01	1.44E+00
1982	6.24E-01	3.36E+00	1.87E-01	1.01E+00
1983	5.52E-01	3.12E+00	1.66E-01	9.36E-01
1984	6.24E-01	5.28E+00	1.87E-01	1.58E+00
1985	5.76E-01	3.12E+00	1.73E-01	9.36E-01
1986	6.24E-01	2.64E+00	1.87E-01	7.92E-01
1987	1.54E+00	1.01E+01	4.61E-01	3.02E+00
1988	1.15E+00	3.36E+00	3.46E-01	1.01E+00
1989	8.88E-01	6.24E+00	2.66E-01	1.87E+00
1990	1.73E-01	7.44E+00	5.18E-02	2.23E+00
1991	1.75E-01	7.68E+00	5.26E-02	2.30E+00
1992	2.38E-01	5.52E+00	7.13E-02	1.66E+00
1993	1.34E-01	2.04E+00	4.03E-02	6.12E-01
1994	1.37E-01	2.64E+00	4.10E-02	7.92E-01
1995	7.44E-02	7.68E-01	2.23E-02	2.30E-01
1996	1.85E-01	1.99E-01	5.55E-02	5.97E-02
1997	1.22E-01	1.01E+00	3.66E-02	3.03E-01
1998	1.92E-01	1.75E+00	5.76E-02	5.25E-01
1999 ^e	1.54E+00	1.01E+01	4.61E-01	3.02E+00
2000 ^e	1.54E+00	1.01E+01	4.61E-01	3.02E+00
2001 ^e	1.54E+00	1.01E+01	4.61E-01	3.02E+00
2002 ^e	1.54E+00	1.01E+01	4.61E-01	3.02E+00

a. Calculated from air concentration data in Table 4A-2.

b. Refers to maximum measurement averaged over samplers for a given month during the year.

c. Calculated by multiplying annual average ^{239,240}Pu intake by 0.30 (see Section 4.2.3).

d. Calculated by multiplying maximum ^{239,240}Pu intake for a given month by 0.30 (see Section 4.2.3).

e. Assumed higher values from 1987 in lieu of onsite data (perimeter monitoring information only available – see appendix 4A)

Table 4-3. Onsite TLD measurements for listed years

Year	Mrem per year			N
	Mean	2 σ	Recommended value (1 σ)	
before 1975	138		32	
1975	111	40	26	87
1976	113	36	26	134
1977	128	32*	30	98
1978	131	36	30	126
1979	146	8.8	34	133
1980	158	8	35	131
1981	134	62	31	125
1982	123	2	28	120
1983	139	2	32	135
1984	146	8	34	142
1985	151	6	35	93
1986	137	4	32	147
1987	153	4	35	100
1988	154	6	36	109
1989	167	5	39	132
1990	154	4	36	143
1991	122	4	28	108
1992	121	3	28	176
1993	137		32	176
After 1993	138	32	32	
Mean 1975 – 1993	138		32	
2 S.D. (2 σ)	32			

* Because there was no value available for 1977, the 95% confidence limits (i.e. $\pm 2\sigma$) were set equivalent to two standard deviations of the 19 annual means.

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GLOSSARY

alpha particles

Positively charged particles of discrete energies emitted by certain radioactive materials; alpha particles usually expend their energy in short distances and will not usually penetrate the outer layer of skin; they are a significant hazard only when taken into the body where their energy is absorbed by tissues.

beta dose

A designation (i.e., beta) on some external dose records referring to the dose from less-energetic beta, X-ray and/or gamma radiation (SEE ALSO *open window*, or *shallow dose*).

beta radiation

Radiation consisting of charged particles of very small mass (i.e., the electron) emitted spontaneously from the nuclei of certain radioactive elements. Most (if not all) of the direct fission products emit beta radiation. Physically, the beta particle is identical to an electron moving at high velocity.

curie

A special unit of activity. One curie equals 3.7×10^{10} nuclear transitions per second.

dosimeter

A device used to measure the quantity of radiation received. A holder with radiation-absorbing elements (filters) and an insert with radiation-sensitive elements packaged to provide a record of absorbed dose or dose equivalent received by an individual. (SEE *albedo dosimeter*, *film dosimeter*, *neutron film dosimeter*, *thermoluminescent dosimeter*.)

dosimetry

The science of assessing absorbed dose, dose equivalent, effective dose equivalent, etc., from external or internal sources of radiation.

dosimetry system

A system used to assess dose equivalent from external radiation to the whole body, skin, or extremities. This includes the fabrication, assignment, and processing of dosimeters as well as interpretation and documentation of the results.

exposure

As used in the technical sense, exposure refers to a measure expressed in roentgens (R) of the ionization produced by photons (i.e., gamma and X-rays) in air.

film

In general, a "film packet" that contains one or more pieces of film in a light-tight wrapping. When developed, the film has an image caused by radiation that can be measured using an optical densitometer.

film dosimeter

A small packet of film within a holder that attaches to a wearer.

fission

The splitting of a heavy atomic nucleus, accompanied by the release of energy.

fissionable

Material capable of undergoing fission.

gamma rays

Electromagnetic radiation (photons) originating in atomic nuclei and accompanying many nuclear reactions (e.g., fission, radioactive decay, and neutron capture).

Physically, gamma rays are identical to X-rays of high energy, the only essential difference being that X-rays do not originate in the nucleus.

HEU

Highly enriched uranium

isotope

Elements having the same atomic number but different atomic weights; identical chemically but having different physical and nuclear properties

neutron

A basic particle that is electrically neutral weighing nearly the same as the hydrogen atom.

neutron film dosimeter

A film dosimeter that contains a Neutron Track Emulsion, type A, film packet.

operating area

Designation of major onsite operational work areas.

radiation

Alpha, beta, neutron, and photon radiation.

radioactivity

The spontaneous emission of radiation, generally alpha or beta particles, gamma rays, and neutrons from unstable nuclei.

radionuclide

A radioactive isotope of an element, distinguished by atomic number, atomic weight, and energy state

rem

A unit of dose equivalent equal to the product of the number of rad absorbed and the quality factor.

thermoluminescence

Property of a material that causes it to emit light as a result of being excited by heat.

thermoluminescent dosimeter (TLD)

A holder containing solid chips of material that when heated will release stored energy as light. The measurement of this light provides a measurement of absorbed dose.

**ATTACHMENT 4A.
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS
AT THE ROCKY FLATS PLANT**

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ATTACHMENT 4A. ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS AT THE ROCKY FLATS PLANT

Occupational environmental dose reconstruction for the Rocky Flats Plant (RFP) requires that $^{239,240}\text{Pu}$ air concentrations be estimated for onsite locations as a function of time. Actual measurements of $^{239,240}\text{Pu}$ in air for the RFP onsite environment are generally preferable to modeled concentrations, because modeling relies on measurements of parameters related to source terms and meteorology that are often spatially and temporally averaged, thus adding considerable uncertainty to estimates for particular locations or points in time. Further, models often cannot address all the processes or source terms that affect the air concentration. However, there are periods in the RFP operational history when useful measurements of $^{239,240}\text{Pu}$ in air are not available. This attachment describes the availability of monitoring data and modeled estimates and addresses the suitability of each type of estimate for use in occupational dose reconstruction for particular historical time periods. In addition, it provides the technical basis of atmospheric transport calculations supporting the estimated $^{239,240}\text{Pu}$ intakes.

A.1 MEASUREMENT OF TOTAL LONG-LIVED ALPHA ACTIVITY AND PLUTONIUM IN AIR

Task 4 of the Phase II Historical Public Exposures Studies on Rocky Flats (Rope et al. 1999) contains an in-depth historical review and compilation of RFP air monitoring data. This attachment relies on conclusions from that review to determine the usefulness of onsite air monitoring data in estimating $^{239,240}\text{Pu}$ intake from environmental (outdoor) exposures.

According to Rope et al. (1999), $^{239,240}\text{Pu}$ concentrations in air were not measured at RFP until 1969, when the Colorado Department of Health began reporting onsite air concentrations. Between the start of operations in 1952 and 1969, gross alpha activity in air was measured by the RFP contractor, but earlier data during this period (up to 1964) were considered by Rope et al. (1999, pp. III-33-34) to be of limited value for assessing concentrations of long-lived alpha-emitters such as $^{239,240}\text{Pu}$. Prior to 1960, measurements were not made in a way that enabled estimation of the long-lived alpha component of the gross alpha measurements. Thus, there is no reliable way to estimate $^{239,240}\text{Pu}$ in air prior to 1960 from air monitoring data. From 1960 to 1964, counts of gross alpha activity were made near the time of collection (4 hours after) and 1 week later, such that Total Long-Lived alpha (TTL α) activity concentrations could be estimated. Estimates of $^{239,240}\text{Pu}$ concentrations can be made from TTL α activity concentrations in a manner described later in this attachment. However, data collected between 1960 and 1964 was of limited value to the Task 4 studies because they were reported as one onsite number, with the maximum and minimum individual count values shown. Results from individual samplers could not be obtained. Thus, data before 1964 were not suitable or readily available for this TBD. Due to these inadequacies in monitoring data during the period between 1952 and 1964, this attachment relied on atmospheric dispersion modeling conducted for the Phase II study for developing estimates of $^{239,240}\text{Pu}$ concentrations in onsite air. Section 4A.2 describes the Phase II model and application for the present purposes.

From October 1964 until December 1971, the RFP contractor reported daily TTL α activity concentrations in air for individual onsite samplers (Rope et al. 1999, p. III-34). The samplers included S-1 through S-10, S-50, and S-51. The locations of the onsite samplers during this period are shown in the upper "1974" drawing in Figure 4A-1 (reproduced from Figure B-6 in Rope et al. 1999). Monthly average concentrations derived from these measurements, in fCi m^{-3} (10^{-15} Ci m^{-3}), are reproduced from Table B-6 in Rope et al. in Table 4A-1.

In 1969, the Colorado Department of Health (CDH), now called the Colorado Department of Public Health and Environment, began monitoring $^{239,240}\text{Pu}$ in air at the RFP (Rope et al. 1999, p. III-21) near

the east security fence. Rope et al. (1999, p. III-23) used data from the CDH air monitoring stations to evaluate the predicted impact of contaminated soil after placement of the asphalt pad in the 903 Area. These measurements complement the air monitoring conducted by the RFP contractor and the Health and Safety Laboratory (HASL), which began monitoring air in the same vicinity (on the east security fence downwind of the 903 Area) in 1970 (Rope et al. 1999, p. III-12).

In 1970, the Rocky Flats environmental monitoring program began to include routine monitoring of $^{239,240}\text{Pu}$ in air (Rope et al., 1999, p III-63). However, results for $^{239,240}\text{Pu}$, rather than $\text{TLL}\alpha$, were not reported in RFP annual environmental reports until 1973 (Dow 1972 through 1975; Rockwell 1976 through 1989; EG&G 1989 through 1993; Kaiser-Hill 1994). Table 4A-2 lists annual air concentrations, averaged over the various onsite samplers and compiled from annual environmental reports through 1994 and the monthly data in Table 4A-1, along with the maximum onsite monthly measurement corresponding to that year. The units have been converted to fCi m^{-3} for consistency with Table 4A-1. Sampling locations are specified, and can be visualized from Figure 4A-1. The samplers were renumbered in 1975, as indicated in this figure. The annual average was not provided in the annual report for 1977 to 1988, but was calculated from the monthly volume-weighted averages for each sampling location. Beginning in 1977, measurements were not reported for all samplers, but for sampling stations with the greatest potential for elevated airborne radioactivity. Thus, results after 1976 are not as representative of an average for the RFP industrial area, but for the onsite areas likely to be highest in concentrations of $^{239,240}\text{Pu}$ in air.

To estimate the $^{239,240}\text{Pu}$ air concentration based on $\text{TLL}\alpha$ activity concentration for data from 1965 to 1973 in Table 4A-2, a conversion factor was adopted from the review of available information on this topic provided in Rope et al. (1999, beginning on p. III-42). Rope et al. evaluated data collected in the early 1970s, and derived a least squares best fit to the data result of 36% plutonium contribution to the $\text{TLL}\alpha$. Therefore, a conversion factor of 0.36 was assumed for converting $\text{TLL}\alpha$ activity concentrations from the annual environmental reports to $^{239,240}\text{Pu}$ activity concentrations.

The annual environmental reports (Dow 1972 through 1975; Rockwell 1976 through 1989; EG&G 1989 through 1993; Kaiser-Hill 1994) provide 95% confidence limits for the average annual $^{239,240}\text{Pu}$ air concentration for RFP for 1972 to 1976 and 1989 to 1994. For these years, the standard deviation was reported as the 95% confidence limits, assuming the measurements were normally distributed. In 1977 and 1978, no standard deviations were reported because "they can be misleading and (their use) is considered inappropriate for the data in this report" (Rockwell 1978, 1979). These latter two reports state that the sampling methods would result in "nonrandom variations in the measured concentrations," and the "distribution ... would not be normal." Standard deviations are reported for 1979 to 1988, but only by sampler location, and not normalized to the site average. The confidence limits, when reported, are based on counting error alone, and thus represent the minimum error that can be associated with the measurements. Although counting error does contribute to uncertainty in the data, the uncertainty associated with a given average value for the site will also be a function of variability in environmental conditions as a function of time and location. Thus, the reported confidence limits are not useful for estimating uncertainty associated with a site average because they would undoubtedly underestimate that uncertainty.

Annual environmental reports were discontinued after 1994 as a result of the change in the RFP mission, so sitewide measurements of $^{239,240}\text{Pu}$ in air after 1994 are not as readily accessible. Between 1995 and July 1998, onsite ambient air concentrations of $^{239,240}\text{Pu}$ are reported in Rocky Flats Environmental Technology Site (RFETS) Monthly and Quarterly Monitoring Reports (EG&G 1995, RMRS 1995a,b; RFETS 1996a-d; RFETS1997a-d; and RFETS1998a-c). In 1995, an onsite average air concentration can be calculated from the reported data for approximately 20 sampling locations, and a maximum corresponding to the sampler at the location of highest concentration is

also readily obtained. From 1996 to July 1998, air concentrations for only two samplers are reported, those samplers being located at the point of highest air concentration based on monitoring in recent years. Thus, the air concentrations reported for those years tend to be more representative of maximum concentrations rather than site-wide averages. Annual average air concentrations, averaged over all sampling locations, were calculated for 1995 through July of 1998, and provided in Table 4A-2. Maximum concentrations in Table 4A-2 correspond to the highest quarterly concentration for any given sampler.

For the period from August 1998 to the present, onsite air concentrations are no longer reported directly in the Quarterly Reports (RFETS 1999a-d, 2000a-c, 2001a-c, 2002a-c, 2003a,b). Rather, the calculated percent of the 10 mrem/y NESHAPs (40 CFR 61) annual average effective dose limit corresponding to measurements made at RFP perimeter monitoring stations are reported. For 1999 through 2002, the estimated percent of the 10 mrem/y dose rate realized at these locations is reported as less than 0.3% for radionuclides other than ^{234}U and ^{238}U . The contributions of uranium isotopes were subtracted from the calculated dose rates since the majority of the observed uranium appears to be due to natural soil contributions (RFETS 2003b). Thus, the annual average effective dose rate estimated at the perimeter locations is 0.03 mrem/year. This can be converted to a $^{239,240}\text{Pu}$ air concentration, by assuming an effective dose factor, a breathing rate, and a fractional contribution to the dose by various radionuclides. For the purposes of this report, the following claimant-favorable assumptions are made: All of the dose arises from $^{239,240}\text{Pu}$ and ^{241}Am ; 70% of the calculated dose rate is due to plutonium; and 30% of the calculated dose rate is due to ^{241}Am . Thus, the dose rate attributable to plutonium is estimated to be 0.023 mrem/year, and to ^{241}Am is estimated to be 0.007 mrem/year. Assuming the NESHAPs dose rate calculation was done by RFETS with a 2400 m^3/y inhalation rate, and effective dose rates of 1.16e-4 Sv/Bq (for $^{239,240}\text{Pu}$) and 1.20e-4 Sv/Bq (for ^{241}Am), from EPA's Federal Guidance Report 11 (EPA, 19**), air concentrations can be calculated that correspond to the perimeter dose rates. These are concentrations are given as annual average values in Table 4A-2, with no corresponding maximum values available (i.e., NA).

Beginning in the last quarter of 2001, performance monitoring was begun to monitor specific demolition and remediation activities. Onsite monitoring is done and evaluated under these circumstances. However, only gross alpha activity is reported in readily-available reports, making it difficult to estimate the fraction of that due to the plutonium and americium isotopes of concern here.

The CDH and RFP contractor continue to monitor air around the RFP periphery and onsite. Although not readily available for this report, it may be possible to acquire more of this information upon specific request.

A.2 ATMOSPHERIC TRANSPORT MODELING

The atmospheric transport modeling results used to develop estimates of $^{239,240}\text{Pu}$ intake by workers between 1953 and 1964 relied on application of a model developed for the offsite risk assessment (Rood and Grogan 1999). Application of this model for onsite predictions was not the original intent, so simulated concentrations are likely to be less reliable than measured concentrations for estimates of $^{239,240}\text{Pu}$ intake. The application of this model for the Phase II Historical Public Exposures Studies focused on offsite rather than onsite concentrations, which are of interest here. Thus, the model did not incorporate building wake effects in predicting air concentrations. Rood and Grogan (1999) described atmospheric transport modeling as follows:

Atmospheric transport modeling performed in the Phase II studies used the Regional Atmospheric Transport Code for Hanford Environmental Tracking (RATCHET) model (Ramsdell et al. 1994). Selection of RATCHET was based on a model comparison study performed for

Phase II (Rood 1999b). In this study, five models, ranging in complexity from a simple straight-line Gaussian plume model (Industrial Source Complex Short Term Version 2 [EPA 1992]) to a complex terrain model (Terrain-Responsive Atmospheric Code [Hodgin 1991]), were compared to tracer measurements taken during the 1991 Winter Validation Tracer Study (Brown 1991). The results of this evaluation indicated no one model clearly outperformed the others. However, the puff trajectory models (RATCHET, TRIAD [Hicks et al. 1989], and INPUFF2 [Petersen and Lavdas 1986]) generally had lower variability and higher correlation to observed values compared to the other models. The RATCHET model was chosen for these calculations because it incorporates spatially varying meteorological and environmental parameters. Additionally, the model includes modules that perform random sampling of the meteorological parameters, allowing for Monte Carlo analysis of uncertainty.

Atmospheric transport simulations were performed differently for discrete and continuous events. For discrete events (Rood and Grogan 1999a, 1999b, 1999c), meteorological data for the specific days of the event were available. RATCHET was run using its Monte Carlo sampling features that sampled from distributions of basic transport parameters for each Monte Carlo trial. Transport parameters that were considered stochastically included wind speed, wind direction, mixing height, precipitation, and Monin-Obukhov scaling length. Uncertainty in the source term was also included in the simulation. Output consisted of n realizations of time-integrated concentration (TIC) and deposition at each of the 2295 computational nodes in the model domain (Figure 3).

Continuous events were modeled somewhat differently. Meteorological data from Rocky Flats for most of the assessment period were lacking. Therefore, we relied on a technique often used in prospective analysis and in retrospective analysis when historical records are lacking. This technique uses compilations of recently acquired meteorological data as a surrogate for past or future conditions and typically only applies to assessments of long-term (>1 year) dispersion conditions. We employed this technique for estimating annual average plutonium concentrations from routine releases and continuous 903 Area suspension releases (Rood 1999a; Rood and Grogan 1999b) using a 5-year data set from 1989 to 1993. Uncertainty was represented using several multiplicative correction factors that accounted for uncertainty in the dispersion process, meteorology, and deposition-plume depletion.

The model domain (Figure 3) encompasses a 2200 km² area (50 km north-south x 44 km east-west). The domain extends 28 km south, 12 km west, 22 km north, and 32 km east from the RFP. Most of the Denver metropolitan area and the city of Boulder are included in the domain.

The domain was limited in its western extent because few receptors were present there during the RFP operations (ChemRisk 1994e) and most of the contaminant plumes traveled east and southeast of the plant.

Figure 3 from Rood and Grogan (1999) is not reproduced here because of the irrelevance of much of the model domain to the present study. Rather, Figure 4A-2 shows the model domain of relevance, which is the RFP industrial area. There are six nodes in the industrial area, providing the ability to estimate six different air concentrations for these regions.

Discrete events in the Phase II studies were defined as those that led to releases of plutonium, which occurred over a relatively short period, and included releases from glovebox fires in 1957 and 1969, and suspension of plutonium-contaminated soil from the 903 Area during unusually high wind events in 1968 and 1969. Continuous releases included "routine" releases from the Building 771 stack and Building 776 roof vents, and suspension of ^{239,240}Pu-contaminated soil from the 903 Area from 1964 to 1969, before the asphalt pad was in place, but not including identified high wind events. Resuspension of ^{239,240}Pu deposited on soil from 903 Area suspension releases was included in the comprehensive evaluation of exposure to ^{239,240}Pu released from RFP (Rood and Grogan 1999). A

time-dependent factor approach was used to address the resuspension of soil contaminated by released $^{239,240}\text{Pu}$ and deposited as a result of the continuous and discrete events. Rood and Grogan (1999, p. 17) describe the source term characterization.

Aerodynamic diameter of $^{239,240}\text{Pu}$ particles released is important in predicting dispersion and deposition; and ultimately in estimating the respirable fraction of the $^{239,240}\text{Pu}$ in air. The source terms characterized for the Phase II studies considered potential size distributions, and the predictive modeling addressed differential dispersion and deposition according to particle size. Thus, results of the modeling enabled prediction of the $^{239,240}\text{Pu}$ concentration in air associated with respirable particles.

Comparisons of model output for the RFP onsite and perimeter regions to air monitoring data from onsite and perimeter samplers, respectively, were made to determine the adequacy of the predictive results in representing concentrations to which RFP workers might have been exposed. Figure 4A-3 plots the annual average $^{239,240}\text{Pu}$ concentrations in air as a function of time in the RFP industrial area from 1953 to 1990. Model-predicted concentrations represent the average of the six onsite computational nodes shown in Figure 4A-2, and are for particles less than 30- μm Aerodynamic Equivalent Diameter (AED). Figure 4A-4 plots annual average $^{239,240}\text{Pu}$ concentrations in air as a function of time for the perimeter area of the plant from 1953 through 1989. For perimeter locations, model-predicted concentrations represent the average of 27 computational nodes, shown in Figure 4A-5. The 5th-, 50th-, and 95th-percentile concentrations for 500 realizations for each of these model applications are shown for the period from 1953 to 1989.

Measured data shown in Figure 4A-3 represent the average concentrations across the onsite samplers listed in the legend. Samplers S-1 through S-10, S-50, and S-51 were maintained by an RFP contractor throughout the period of interest. The annual average TLL α data from 1965 through 1972, calculated from the data in Table 4A-1, were converted to $^{239,240}\text{Pu}$ concentrations for comparison to the model predictions. A conversion factor of 0.36 (0.36 Ci $^{239,240}\text{Pu}$ per Ci TLL α) reported in Figure III-23 in Rope et al. (1999) was used. Average concentrations from samplers S-5 thru S-9 were obtained from Table B-10 in Rope et al. (1999), which is reproduced in Table 4A-3. The onsite values in this table tend to be slightly higher than the averages in Table 4A-2 because they are restricted to samplers S-5 through S-9 only for all years. Figure 4A-3 also plots the arithmetic average of measurements obtained by CDH samplers D-3, D-4, and AP-56, located at the eastern security fence of the RFP (Figure 4A-6). The data from which averages were derived for these CDH samplers are listed in Table 4A-4, which was reproduced from Table III-4 in Rope et al. (1999).

From Figure 4A-3, it is clear that the modeled 50th-percentile concentrations can underpredict the measured concentrations, especially after about 1970. Before 1970, agreement between model predictions and measured concentrations is fairly good. One possible explanation for underprediction that does not necessarily render modeling as an underprediction of $^{239,240}\text{Pu}$ intake at RFP is that reported $^{239,240}\text{Pu}$ concentrations are often at locations of expected elevated concentrations, while the modeled concentrations are averages for the entire site. After 1970, the main source of plutonium to onsite air was resuspension of contaminated soil (Rood and Grogan 1999, p. 57); thus, one would expect air concentrations in the area of highest soil plutonium (downwind of the 903 Area) to be higher than the site average. Because the measured data predominantly arose from that area (samplers S-5 through S-9), it is likely that measured concentrations would exceed the site average. It is not necessarily likely, however, that an exposed individual would spend a significant amount of time in an area of higher $^{239,240}\text{Pu}$ concentration in air. Thus, the model might be a more reasonable predictor of average dose for an individual exposed to average concentrations across the site, rather than in the areas of higher $^{239,240}\text{Pu}$ concentration.

Other reasons predicted concentrations might deviate from measured onsite concentrations include the influence of building wakes, which can increase or decrease concentrations in the proximity of buildings. Further, it is possible, and likely, that modeled concentrations do not include potential sources that might have been important to onsite exposures.

Measured data shown in Figure 4A-4 for the perimeter concentrations were obtained from the 1971 through 1990 annual environmental reports prepared by RFP contractors and represent average concentrations from RFP contractor perimeter samplers, shown in Figure 4A-5 as triangular symbols. These data were corrected for contributions from background $^{239,240}\text{Pu}$, because the measured values are low enough that fallout concentrations can contribute a significant portion of the total measurement. Yearly measurements of $^{239,240}\text{Pu}$ in air in Denver (Rope et al. 1999, Table B-14, reproduced in Table 4A-5) were assumed to be representative of RFP background levels. These levels were subtracted from the perimeter measurements in Table 4A-3 to obtain the average net perimeter $^{239,240}\text{Pu}$ concentrations listed in Table 4A-6 and plotted in Figure 4A-4. From Figure 4A-4, it is clear that the model predictions and measured concentrations are in good agreement for these perimeter locations.

The results of these comparisons of model predictions with measured $^{239,240}\text{Pu}$ concentrations support the following conclusions. Onsite application of the atmospheric dispersion model developed for the Phase II historical offsite exposure studies must be done with caution. Predicted average onsite concentrations might not adequately represent actual concentrations of interest, due to the large spatial variation in soil and, thus air, contamination at the site. While it is desirable to use measured concentrations from the many air samplers across the site to derive average and maximum values, some of the historical data to support this are not readily available or of sufficient quality. Therefore, it is reasonable, especially prior to 1970, to use the Phase II model application for estimating onsite average concentrations in lieu of measurement data.

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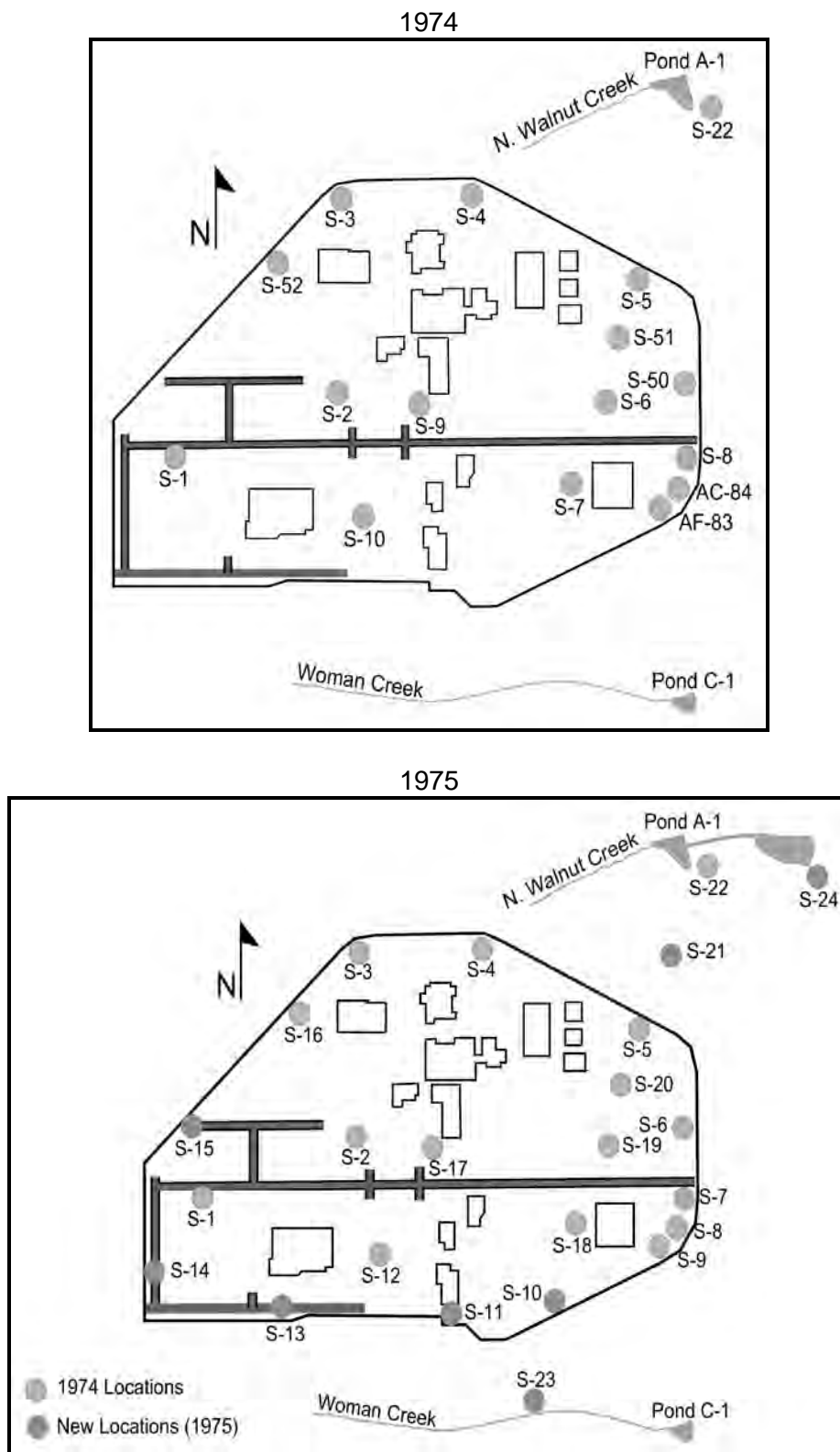


Figure 4A-1. Locations of samplers operated by RFP contractor. In 1975, samplers were renumbered and new samplers were added. (Reproduced from Rope et al. 1999, Figure B-6).

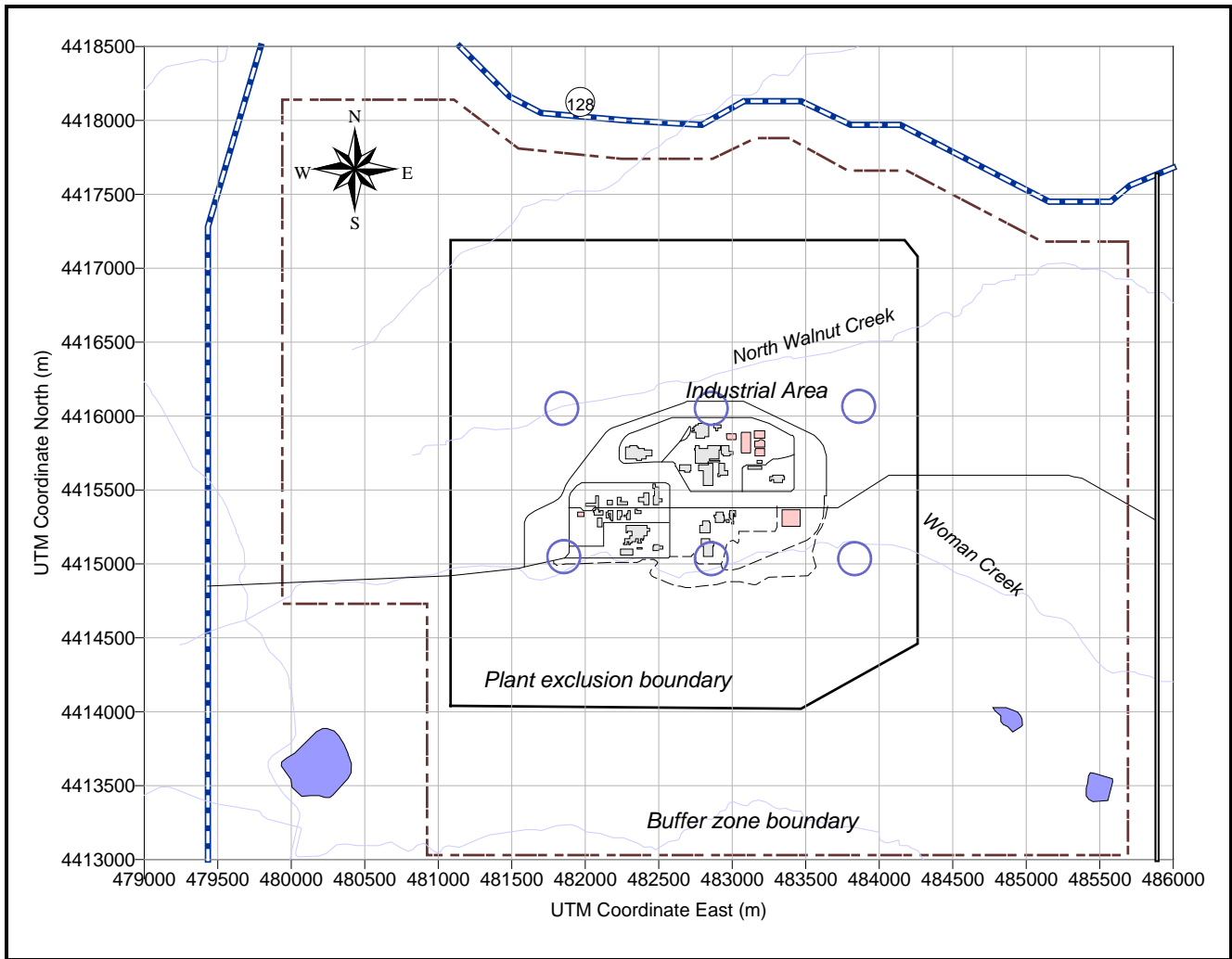


Figure 4A-2. Location of onsite (industrial area) computational nodes (blue circles) used to predict average onsite air concentrations for RFP Industrial Area.

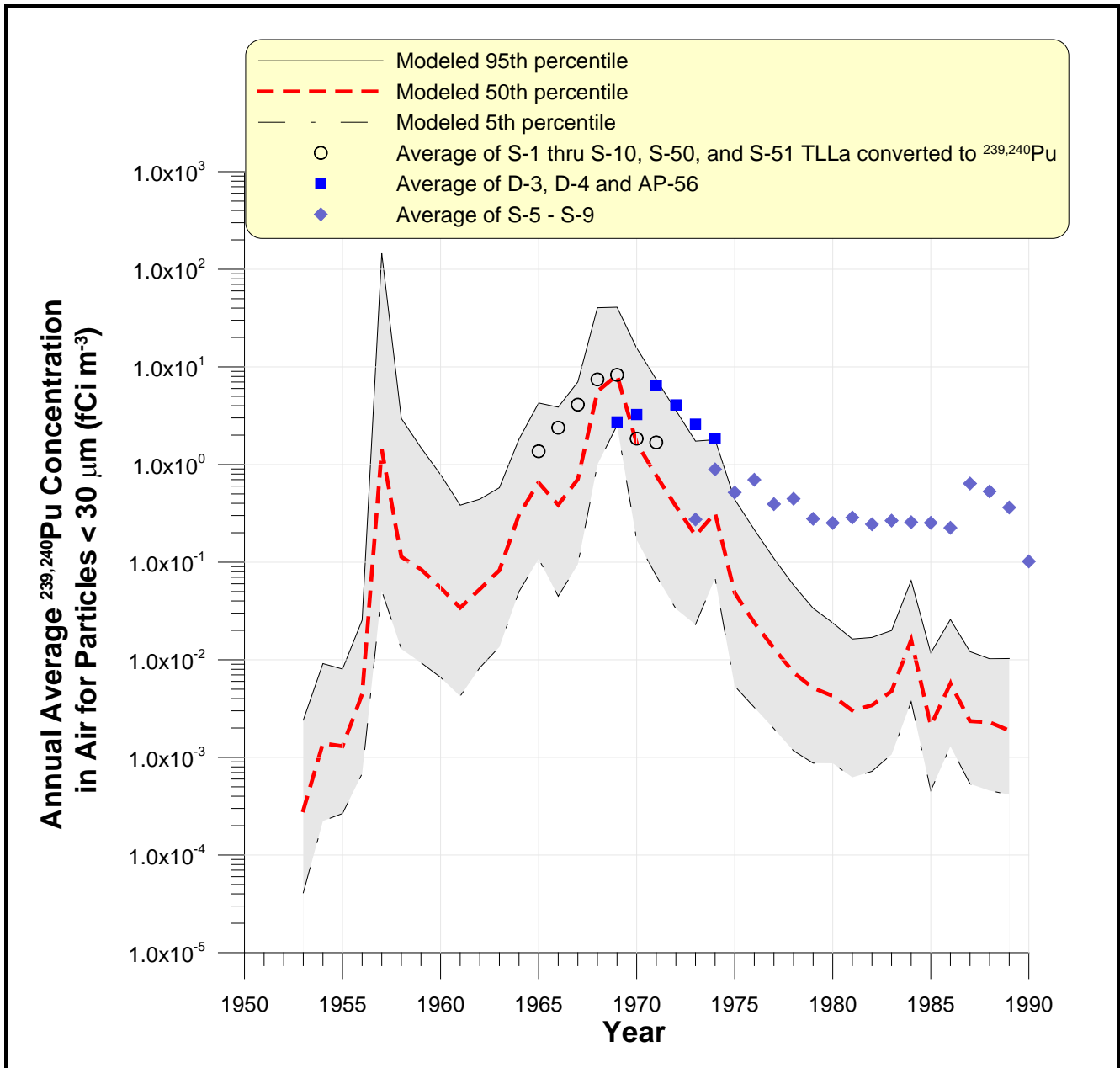


Figure 4A-3. Annual average $^{239,240}\text{Pu}$ concentrations in air as a function of time for particles $< 30\ \mu\text{m}$ Aerodynamic Equivalent Diameter (AED) in RFP industrial area. (Model-predicted concentrations represent the average of six onsite computational nodes as shown in Figure 4A-3 using the model described in Rood and Grogan (1999).)

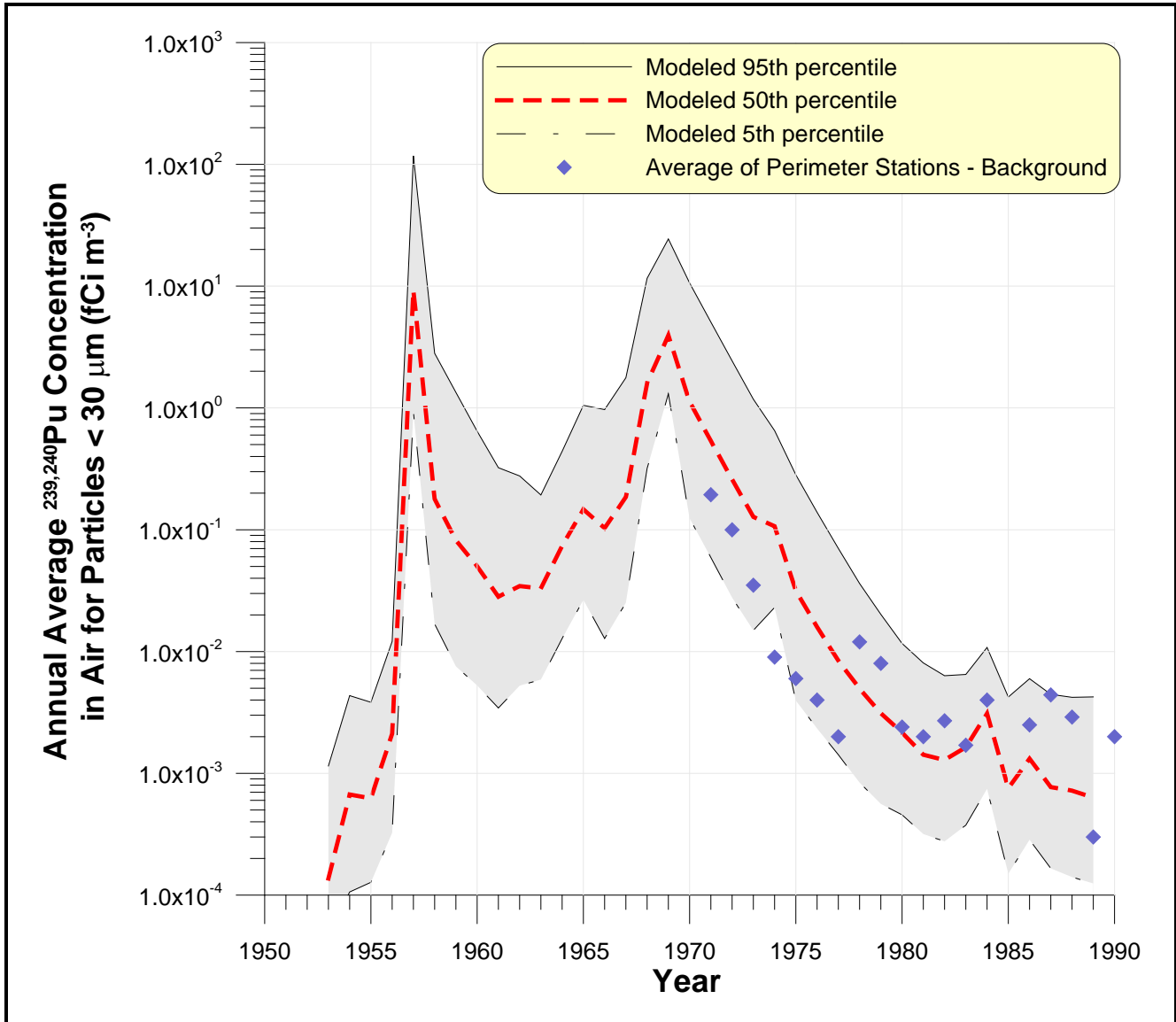


Figure 4A-4. Annual average $^{239,240}\text{Pu}$ concentrations in air as a function of time for particles $< 30\ \mu\text{m}$ Aerodynamic Equivalent Diameter (AED) in the perimeter area surrounding RFP. (Model-predicted concentrations represent the average of 27 computational nodes as shown in Figure 4A-7 using the model described in Rood and Grogan (1999).)

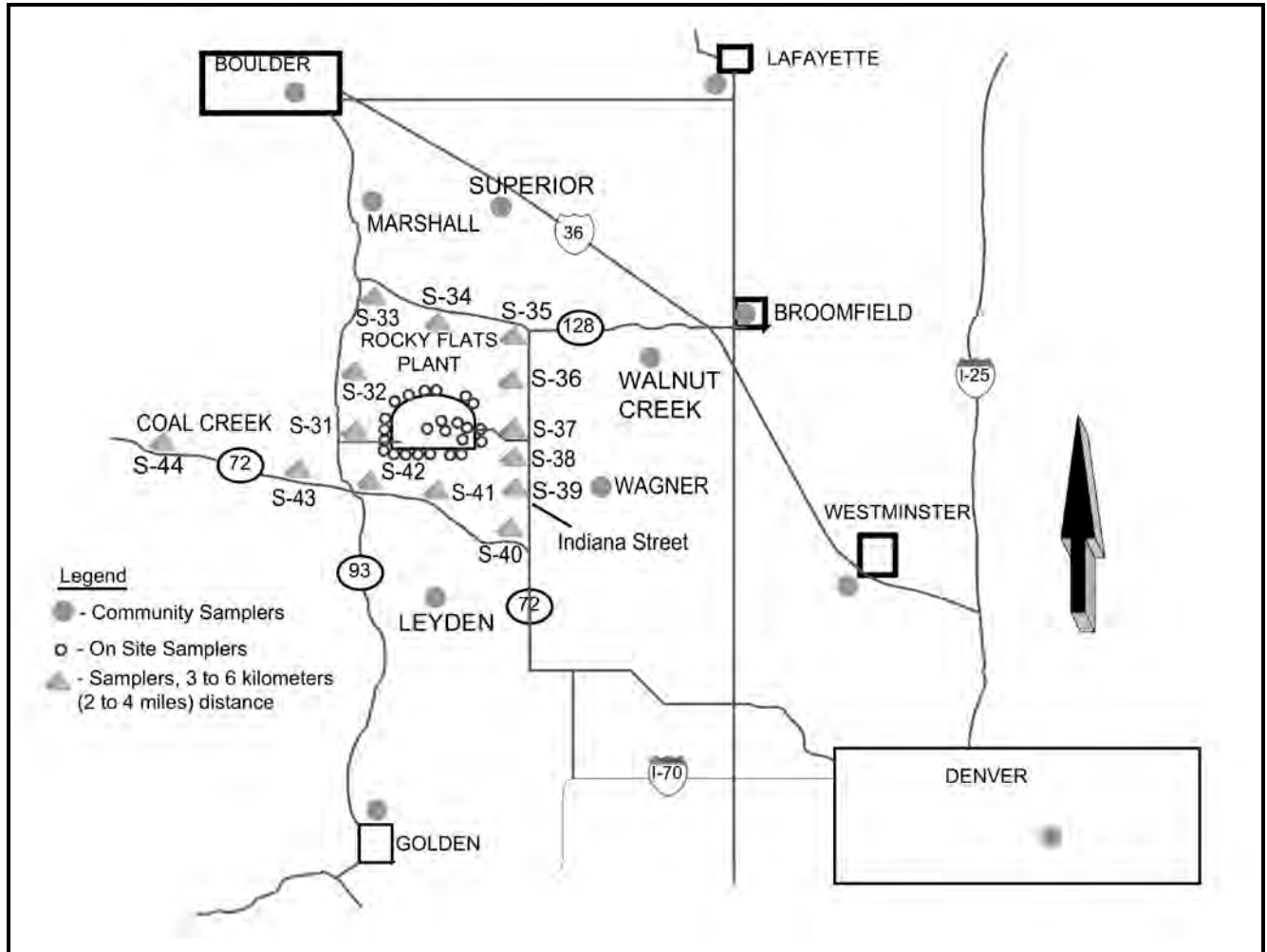


Figure 4A-5. Locations of offsite air samplers in 1975 (Figure B-4 from Rope et al., 1999).

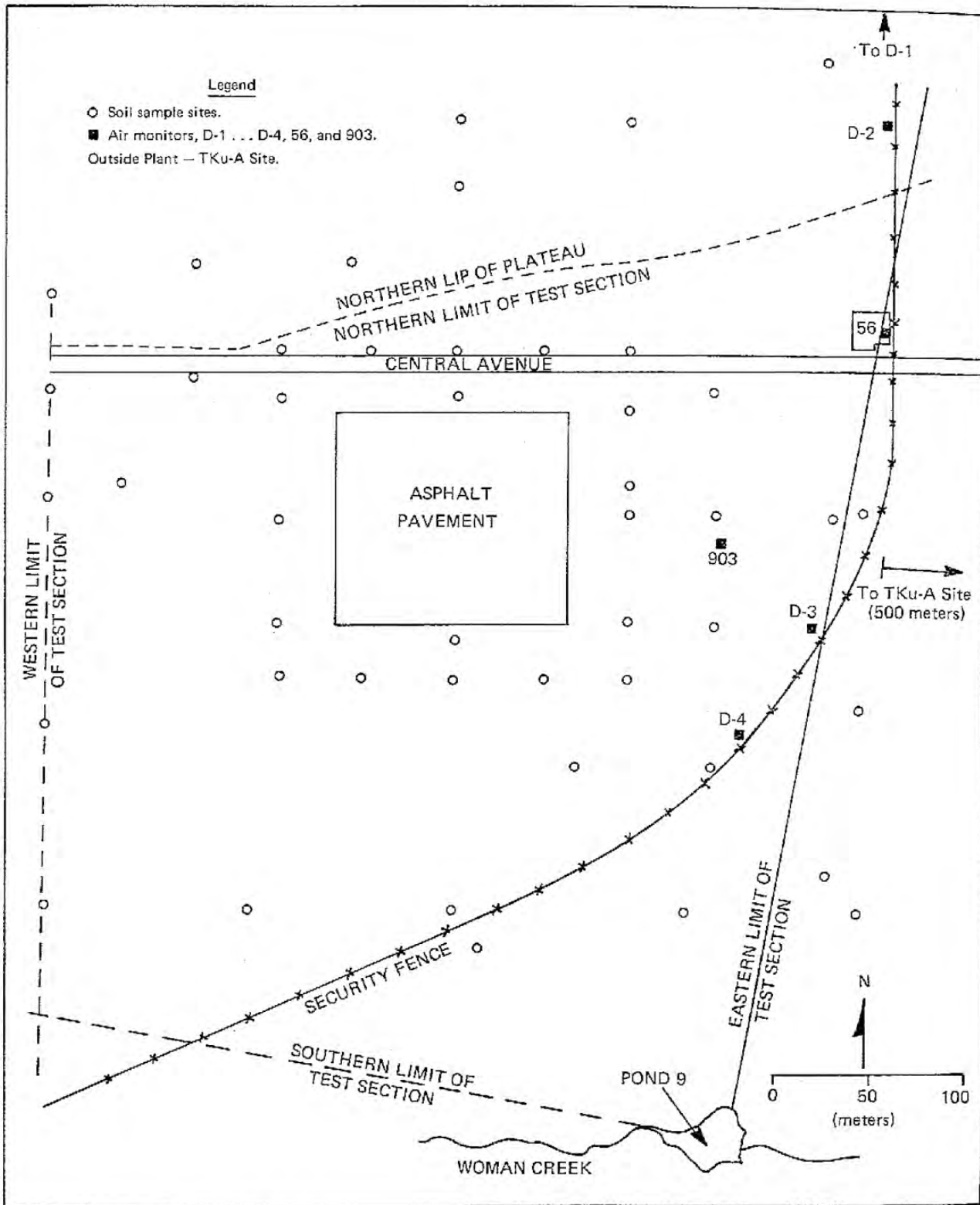


Figure 4A-6. Eastern part of RFP industrial area, showing locations of CDH air monitoring stations D-1 through D-5. Sampler AP-56 is believed to be north of Central Avenue near the security fence. (Reproduced from Figure III-13 of Rope et al. (1999).)

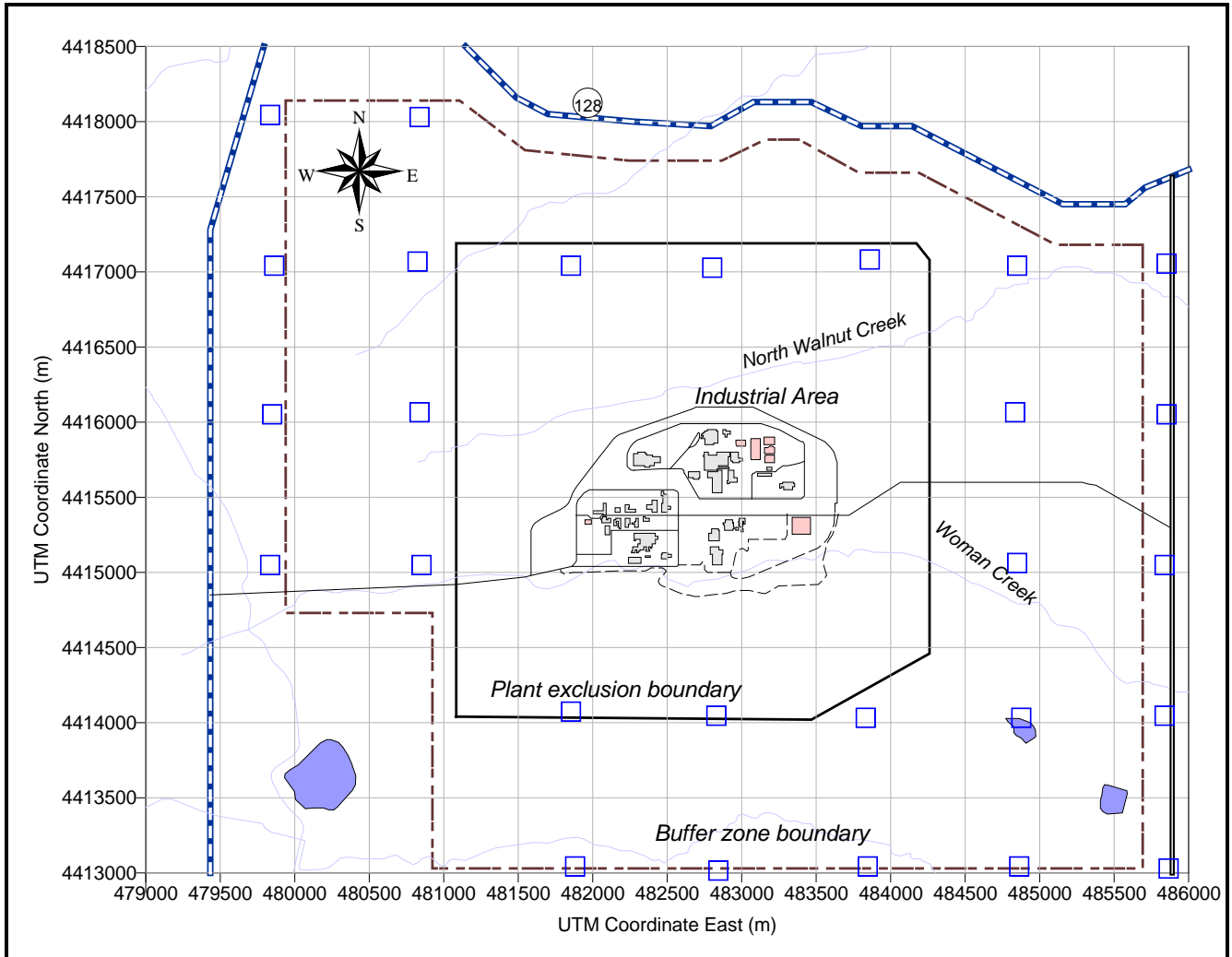


Figure 4A-7. Locations of computational nodes (blue squares) used to compute annual average perimeter concentrations shown in Figure 4A-6.

Table 4A-1. Monthly average concentrations (fCi m⁻³) of total long-lived alpha activity in onsite air samples between October 1964 and December 1971 (reproduced from Table B-6 in Rope et al. (1999); reconstructed from daily measurements).

	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-50	S-51
Oct-64	2	6	2	4	4	3	3	11	6	4		
Nov-64	1	2	6	1	2	2	2	16	3	2		
Dec-64	2	3	4	2	7	4	3	79	4	3		
Jan-65	2	2	11	3	7	2	2	8	3	2		
Feb-65	3	2	2	4	4	4	3	4	2	3		
Mar-65	5	3	3	5	3	5	2	2	3	3		
Apr-65	5	3	2	2	2	3	3	5	2	2		
May-65	3	13	1	3	4	5	3	4	2	3		
Jun-65	4	2	2	2	4	5	2	8	2	2		
Jul-65	4	2	2	2	3	6	2	9	1	3		
Aug-65	4	1	2	3	3	4	2	5	2	3		
Sep-65	5	3	3	3	7	5	0	5	2	4		
Oct-65	3	3	3	1	2	4	2	3	3	2		
Nov-65	10	3	3	2	4	4	2	9	3	4		
Dec-65	4	7	7	6	14	13	5	7	4	9		
Jan-66	11	6	4	2	8	4	3	12	6	14		
Feb-66	3	2	3	5	6	2	1	11	3	4		
Mar-66	4	2	4	3	3	3	2	11	5	3		
Apr-66	3	2	3	4	4	4	2	6	3	2		
May-66	4	4	4	3	6	5	5	5	7	7		
Jun-66	7	5	4	2	3	13	8	12	6	8		
Jul-66	7	6	4	4	6	6	5	11	7	6		
Aug-66	15	7	4	12	7	11	8	13	8	6		
Sep-66	28	8	9	12	18	16	5	10	7	8		
Oct-66	11	4	4	7	8	8	9	10	7	4		
Nov-66	7	2	3	16	8	6	6	13	5	3		
Dec-66	7	4	5	14	10	12	4	8	8	6		
Jan-67	8	4	7	15	13	7	6	22	23	8		
Feb-67	5	6	6	22	8	8	13	38	6	4		
Mar-67	6	1	5	11	7	8	5	9	5	3		
Apr-67	3	3	6	6	10	7	3	11	5	7		
May-67	9	4	7	8	9	12	8	9	6	5		
Jun-67	10	5	8	13	14	11	17	18	5	5		
Jul-67	15	5	7	8	11	14	28	19	9	3		
Aug-67	27	6	15	10	11	17	28	26	10	12		
Sep-67	5	3	3	6	4	14	14	22	4	5		
Oct-67	5	3	7	7	5	15	24	99		7		
Nov-67	6	5	10	6	4	9	8	49		7		
Dec-67	9	7	17	7	3	3	9	92		4		
Jan-68	8	4	12	5	6	3	8	29		5		
Feb-68	15	9	23	12	7	6	10	33		11		
Mar-68	14	18	22	8	9	17	11	116		28		
Apr-68	16	11	13	9	6	27	31	182		10		
May-68	8	9	17	10	7	21	70	155		8		
Jun-68	6	6	8	12	4	51	68	110		18		
Jul-68	5	4	7	4	4	14	14	31		3		
Aug-68	9	6	8	5	7	9	10	17		4		

Table 4-A-1. (Continued)

	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-50	S-51
Sep-68	6	6	7	5	5	4	9	50		3		
Oct-68	12	7	8	6	4	11	19	33		3	7	11
Nov-68	7	4	6	3	7	9	6	168		4	4	2
Dec-68	11	6	4	8	6	6	20	357		3	11	3
Jan-69	10	6	9	7	10	15	127	1525		3	7	5
Feb-69	6	4	8	5	5	4	23	129		3	4	3
Mar-69	7	4	3	5	4	2	4	208		2	5	3
Apr-69	7	3	5	3	6	3	22	148	4	4	5	6
May-69	5	11	9	9	9	17	21	28	10	6	9	6
Jun-69	4	4	7	4	4	7	9	68	4	4	7	3
Jul-69	15	7	7	7	4	20	7	20	4	4	11	6
Aug-69	4	6	5	6	5	7	7	22	5	3	6	9
Sep-69	6	6	5	5	6	5	6	19	4	3	5	5
Oct-69	6	3	5	18	3	5	3	12	2	3	3	3
Nov-69	3	2	4	5	4	3	2	12	2	3	4	4
Dec-69	5	4	5	7	4	2	4	32	7	5	4	4
Jan-70	3	2	4	4	4	3	3	12	3	4	2	3
Feb-70	2	4	5	4	3	4	13	33	3	6	2	7
Mar-70	3	4	4	15	3	5	4	10	4	4	3	5
Apr-70	10	5	5	7	9	7	5	7	2	3	5	3
May-70	9	4	5	3	4	4	2	10	1	4	4	4
Jun-70	14	7	12	9	57	13	7	10	4	13	7	10
Jul-70	7	5	1	3	2	2	2	4	3	3	4	4
Aug-70	3	4	2	3	2	4	10	4	3	4	2	5
Sep-70	2	6	6	3	3	3	3	5	3	4	4	4
Oct-70	5	3	5	4	3	4	2	5	3	4	3	4
Nov-70	2	6	3	3	5	4	1	3	2	2	4	2
Dec-70	3	4	3	3	3	3	6	5	2	3	4	2
Jan-71	4	4	3	2	3	4	7	5	4	4	4	4
Feb-71	3	4	2	4	4	3	6	7	4	5	3	5
Mar-71	5	4	4	5	3	3	8	8	4	5	8	4
Apr-71	4	4	5	7	4	3	3	26	4	5	5	5
May-71	5	4	5	3	4	6	4	9	4	5	5	4
Jun-71	4	3	4	5	5	4	4	10	4	7	7	4
Jul-71	4	3	5	4	4	12	3	12	3	2	4	2
Aug-71	4	4	6	4	3	3	4	8	4	3	9	4
Sep-71	5	3	6	3	3	3	2	6	3	4	6	3
Oct-71	5	4	4	3	6	3	2	12	2	5	7	5
Nov-71	3	4	6	3	5	5	3	10	2	4	5	4
Dec-71	4	5	4	3	3	4	4	6	3	4	5	4

Table 4A-2. Estimated annual average concentrations (fCi m⁻³) of ^{239,240}Pu in onsite air samples between 1965 and 1994 based on measurement data provided in Table 4A-1, RFP annual environmental reports^a, and Rocky Flats Environmental Technology Site Monthly and Quarterly Monitoring Reports.

Year	Sampling location ^b	Type of measurement	Conversion factor to ^{239,240} Pu ^c	Annual average air concentration (fCi m ⁻³)	Maximum air concentration ^d (fCi m ⁻³)
1965	S1-S10	Total long-lived alpha	0.36	1 ^e	5 ^e
1966	S1-S10	Total long-lived alpha	0.36	3 ^e	10 ^e
1967	S1-S10	Total long-lived alpha	0.36	4 ^e	36 ^e
1968	S1-S8, S10	Total long-lived alpha	0.36	8 ^e	129 ^e
1969	S1-S10, S50-S51	Total long-lived alpha	0.36	9 ^e	549 ^e
1970	S1-S10, S50-S51	Total long-lived alpha	0.36	2 ^e	21 ^e
1971	S1-S10, S50-S51	Total long-lived alpha	0.36	1.8	98.
1972	S1-S10, S50-S51	Total long-lived alpha	0.36	2.1	61.
1973	S1-S10, S50-S52	Total long-lived alpha	0.36	<2.2	262.
1973	S1-S10, S50-S52	^{239,240} Pu	1	<1.2	46.
1974	S1-S10, S50-S52	^{239,240} Pu	1	0.61	16.
1975	S1-S10, S22, S50-S52, Af83, Ac84	^{239,240} Pu	1	<0.20	6.3
1976	S1-S24	^{239,240} Pu	1	0.19	3.9
1977	S1-S24	^{239,240} Pu	1	0.27 ^f	2.8
1978	S5-S9, S19-S21	^{239,240} Pu	1	0.25 ^f	3.5
1979	S2, S5-S9, S15-S16, S19-S21	^{239,240} Pu	1	0.17 ^f	1.6
1980	S5-S9, S16, S19-S21	^{239,240} Pu	1	0.15 ^f	2.2
1981	S5-S9, S16, S19-S21	^{239,240} Pu	1	0.18 ^f	2.0
1982	S5-S9, S16, S19-S21	^{239,240} Pu	1	0.26 ^{f,g}	1.4
1983	S5-S9	^{239,240} Pu	1	0.23 ^{f,g}	1.3
1984	S5-S9	^{239,240} Pu	1	0.26 ^{f,g}	2.2
1985	S5-S9	^{239,240} Pu	1	0.24 ^{f,g}	1.3
1986	S5-S9	^{239,240} Pu	1	0.26 ^{f,g}	1.1
1987	S5-S9	^{239,240} Pu	1	0.64 ^{f,g}	4.2
1988	S5-S9	^{239,240} Pu	1	0.48 ^{f,g}	1.4
1989	S5-S9	^{239,240} Pu	1	0.37	2.6
1990	S1-S24, S88	^{239,240} Pu	1	0.072	3.1
1991	S1-S25	^{239,240} Pu	1	0.073	3.2
1992	S1-S25	^{239,240} Pu	1	0.099	2.3
1993	S3-S25	^{239,240} Pu	1	0.056	0.85
1994	S3-S25	^{239,240} Pu	1	0.057	1.1
1995	S005-S009, S101-104, S106-S107, S109-S110, S112, S116, S119, S121, S123, S202-S206, S208, S211	^{239,240} Pu	1	0.031	0.32
1996	S007, S107	^{239,240} Pu	1	0.077	0.083
1997	S007, S107	^{239,240} Pu	1	0.051	0.42
1998 ^h	S007, S107	^{239,240} Pu	1	0.080	0.73
1999	perimeter	^{239,240} Pu	1	0.022 ⁱ	NA
2000	perimeter	^{239,240} Pu	1	0.022 ⁱ	NA
2001	perimeter	^{239,240} Pu	1	0.022 ⁱ	NA
2002	perimeter	^{239,240} Pu	1	0.022 ⁱ	NA

- a. Compiled from measurement data provided in Dow (1972 through 1975); Rockwell (1976 through 1989); EG&G (1989 through 1993); and Kaiser-Hill (1994).
- b. Refer to Figure 4A-1 for samplers through 1994; after 1994, see respective Monthly and Quarterly Monitoring Reports for maps.
- c. Based on Rope et al. (1999, pp. III-42 – III-46).
- d. The maximum monthly concentration at any one sampling location.
- e. Calculated from data in Table 4A-1.
- f. Calculated by averaging reported annual volume-weighted average concentration for each sampler.
- g. Results only reported for samplers in area of higher measured plutonium air concentrations for RFP.
- h. Based on data collected January through July, 1998.
- i. Estimated from reported percent of NESHAPs dose (10 mrem/yr) at perimeter stations (see text in section 4A.1).

Table 4A-3. Annual average concentrations (fCi m⁻³) of plutonium in air for three location groups, Rocky Flats contractor monitoring (1971–1990)^a [reproduced from Table B-10 in Rope et al. (1999)]

	Onsite^b	Perimeter	Community
1971		0.26	
1972		0.14	
1973	0.274	0.05	0.26
1974	0.892	0.058	0.34
1975	0.517	0.037	0.031
1976	0.698	0.015	0.013
1977	0.393	0.038	0.037
1978	0.446	0.06	0.06
1979	0.278	0.02	0.02
1980	0.252	0.01	0.01
1981	0.287	0.018	0.019
1982	0.244	0.005	0.006
1983	0.226	0.003	0.007
1984	0.257	0.005	0.005
1985	0.235	0.002	0.002
1986	0.225	0.005	0.003
1987	0.639	0.005	0.003
1988	0.529	0.003	0.002
1989	0.363	0.001	0.001
1990	0.102	0.002	0.001

- a. Obtained from Rocky Flats Plant contractor annual reports. Data are plotted as a line chart in Chapter III.
- b. Onsite samples are the average of locations S-5, S-6, S-7, S-8, and S-9, as numbered in 1975.

Table 4A-4. Data from Table III-4 in Rope et al. (1999) showing a comparison of ^{239,240}plutonium concentrations (fCi m⁻³) in onsite and offsite air, measured by CDH.^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,b,c).

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).

Table 4A-5. Data from Table B-14 in Rope et al. (1999) showing annual average concentrations (fCi m⁻³) of ^{239,240}Pu in Denver air, 1971–1989.

Year	Average	5th percentile	95th percentile	Method of determination ^a
1971	6.6×10^{-2}	5.3×10^{-2}	7.9×10^{-2}	M
1972	4.0×10^{-2}	3.2×10^{-2}	4.8×10^{-2}	M
1973	1.5×10^{-2}	1.2×10^{-2}	1.8×10^{-2}	M
1974	4.9×10^{-2}	3.9×10^{-2}	5.9×10^{-2}	M
1975	3.1×10^{-2}	2.5×10^{-2}	3.7×10^{-2}	M
1976	1.1×10^{-2}	9.0×10^{-3}	1.3×10^{-2}	M
1977	3.6×10^{-2}	2.8×10^{-2}	4.3×10^{-2}	M
1978	4.8×10^{-2}	3.8×10^{-2}	5.7×10^{-2}	M
1979	1.2×10^{-2}	9.3×10^{-3}	1.4×10^{-2}	M
1980	7.6×10^{-3}	3.8×10^{-3}	1.1×10^{-2}	M
1981	1.6×10^{-2}	3.8×10^{-3}	1.1×10^{-2}	N
1982	2.3×10^{-3}	8.0×10^{-3}	2.4×10^{-2}	N
1983	1.3×10^{-3}	6.5×10^{-4}	2.0×10^{-3}	M
1984	1.0×10^{-3}	5.0×10^{-4}	1.5×10^{-3}	M
1985	4.5×10^{-3}	2.3×10^{-3}	6.8×10^{-3}	M
1986	2.5×10^{-3}	1.2×10^{-3}	3.7×10^{-3}	M
1987	6.0×10^{-4}	3.0×10^{-4}	9.0×10^{-4}	M
1988	1.0×10^{-4}	5.0×10^{-5}	1.5×10^{-4}	M
1989	7.0×10^{-4}	3.5×10^{-4}	1.1×10^{-3}	M

- a. M = average of values measured by the Public Health Service or Environmental Protection Agency in Denver for that year; N = no data for Denver, EPA data from New York City used. See Figure III-7 and associated text in Rope et al. (1999) for additional discussion.

Table 4A-6. Corrected annual average concentrations of ^{239,240}Pu in air at perimeter monitoring stations (from Table B-10 in Rope et al. 1999)

Year	^{239, 240} Pu Concentration (fCi m ⁻³) ^a	Net ^{239, 240} Pu Concentration (fCi m ⁻³) ^b
1971	2.60E-01	1.94E-01
1972	1.40E-01	1.00E-01
1973	5.00E-02	3.50E-02
1974	5.80E-02	9.00E-03
1975	3.70E-02	6.00E-03
1976	1.50E-02	4.00E-03
1977	3.80E-02	2.00E-03
1978	6.00E-02	1.20E-02
1979	2.00E-02	8.00E-03
1980	1.00E-02	2.40E-03
1981	1.80E-02	2.00E-03
1982	5.00E-03	2.70E-03
1983	3.00E-03	1.70E-03
1984	5.00E-03	4.00E-03
1985	2.00E-03	0.00E+00 ^c
1986	5.00E-03	2.50E-03
1987	5.00E-03	4.40E-03
1988	3.00E-03	2.90E-03
1989	1.00E-03	3.00E-04
1990	2.00E-03	2.00E-03

- a. From Table 4A-3.
- b. The net concentration is the concentration in the second column minus the corresponding average background value reported in Table 4A-5.
- c. Average background measurement (Table 4A-5) exceeded measured perimeter value.

ATTACHMENT 4B. SCREENING OF RADIONUCLIDE SOURCE TERMS FOR ENVIRONMENTAL DOSE EVALUATION

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B.1 SCREENING METHODOLOGY

To evaluate the potential importance of the radionuclides other than plutonium isotopes reported to have been released to the air at the Rocky Flats Plant (RFP) since operations began in 1952, information from Phase I and Phase II of the Historical Public Exposure Studies on Rocky Flats (ChemRisk 1994; Rood and Grogan 1999) were used to estimate historical onsite yearly inhalation doses for the radionuclides identified. Radionuclides were considered insignificant contributors to dose if estimated maximum annual dose associated with the release was on the order of 1 millirem or less for the period of interest.

The source terms described in Phase I were assumed to be reasonable estimates of the releases of tritium, depleted uranium, enriched uranium, and ²⁴¹Am. For Phase I, effluent monitoring data reported by the RFP was reviewed, and determined to provide a good basis for estimating airborne releases of all radionuclides with the exception of believed underreporting of uranium emissions prior to 1961 (ChemRisk 1994, p. 9). For the period before 1978, "total long-lived alpha radioactivity measured from facilities which process depleted uranium was assumed to represent depleted uranium and total long-lived alpha radioactivity measured from facilities which process enriched uranium was assumed to represent enriched uranium" (ChemRisk 1994, p.113). It is important to note that the release estimates reported here do not appear to include emissions from the 903 Area, which occur as a result of suspension of the primary contamination (i.e., oil-contaminated soil), or resuspension of the windblown material deposited downwind. Thus, this component of the emissions of ²⁴¹Am is not fully evaluated here, and is addressed in the Occupational Environmental Dose section.

To calculate an estimated maximum annual dose from these releases, the release rates were multiplied by a normalized air concentration (χ/Q), in s/m^3 , to obtain maximum onsite air concentration for each radionuclide. The χ/Q value was obtained from atmospheric dispersion modeling done in *Phase II Comprehensive Assessment of Exposure and Lifetime Cancer Incidence Risk from Plutonium Released from the Rocky Flats Plant, 1953-1989* (Rood and Grogan 1999), where all identified sources of plutonium were considered together to develop plutonium air concentration profiles of the RFP vicinity. The maximum χ/Q value for the industrial area and buffer zone was selected for estimating annual air concentrations. The model predicted a maximum χ/Q of 6.28×10^6 s/m^3 at a computational node in the north-central portion of the industrial area of the site.

Calculated annual air concentrations were multiplied by an inhalation rate of $2,400 \text{ m}^3/\text{yr}$, representing a 2,000-hr work year and 1.2 m^3 per hour inhalation rate to obtain an estimate of annual radionuclide intake. The annual intake for each radionuclide was multiplied by its respective inhalation dose factor from International Commission on Radiological Protection (ICRP) Publication 72 (ICRP 1996) for tritium and ^{241}Am , and from a Phase II report (Voillequé 1999) for uranium, to obtain an estimate of maximum annual dose.

B.2 TRITIUM

Tritium is known to have been released both during routine RFP operations and accidents. Routine releases were reported in annual reports after 1973 (ChemRisk 1994), and were estimated to be less than 10 Ci per year. Several larger releases of tritium were documented and reviewed in the Phase I study. The three most significant incidents with respect to tritium release occurred in 1968, 1973, and 1974. The highest estimated release of tritium cited in the Phase I study occurred in the 1973 accident, where it was reported that up to 350 to 1,600 Ci of tritium were released in exhausted air from Building 779A.

The estimated annual releases (Q) of tritium, in Ci/yr, from the Phase I study are listed in Table 4B-1. This table includes the inhalation dose factor assumed for tritium (ICRP 1996) and the calculated dose for each year. For screening purposes, this inhalation factor corresponds to the highest value reported for all forms of tritium compounds (ICRP 1996), representing Type S particulate tritium compounds, and is approximately 14 times higher than the value for tritiated water vapor. While this is an overestimate of the dose factor for the most likely form of tritium (water vapor), the χ/Q used may somewhat underestimate the true normalized air concentration for tritiated water vapor, as it assumes the emitted compounds are particulate, and thus that deposition occurs. From Table 4B-1, the maximum dose was calculated for 1953 to 1989 to be less than 1 mrem/yr. Thus, tritium was an insignificant contributor to inhalation dose based on the criterion put forth in the first paragraph of this appendix, and is not addressed further in this Technical Basis Document (TBD).

B.3 DEPLETED AND ENRICHED URANIUM

Uranium was processed at RFP in two forms: depleted and enriched. Release estimates in the Phase I studies (ChemRisk 1994) were developed for both forms. Until 1978, estimated release of both forms rely on measurements of total long-lived alpha (TLL α), because routine isotopic analyses of effluent sample filters from uranium facilities were not performed until that time. After 1978, not all uranium isotopes of interest were routinely measured. Thus, the Phase I analysis based estimates on known alpha activity fractions for the two types of uranium.

Approximately 15 mCi of depleted uranium and 7 mCi of enriched uranium were released to the RFP air environment between 1953 and 1989. According to the ChemRisk (1994) analysis, more than

90% of the activity released is estimated to have occurred before 1970, with the highest releases in the 1950s.

The estimated annual releases (Q) of depleted and enriched uranium, in $\mu\text{Ci}/\text{yr}$, from the Phase I study are listed in Tables 4B-2 and 4B-3. These tables include the inhalation dose factors assumed for the two forms of uranium, taken from a Phase II study (Voillequé 1999), and the calculated doses for each year. The dose factors were converted from effective dose, in rem per μg inhaled (Table 3 in Voillequé 1999), using specific activities for enriched and depleted uranium (Table 1 in Voillequé 1999), as listed below.

Type of uranium	Specific activity ($\mu\text{Ci}/\mu\text{g}$)	Effective dose (rem) per μg inhaled	Dose factor used in Tables 4-B.3-1 and 4-B.3-2 (mrem/ μCi)
Depleted	3.6×10^{-7}	1.1×10^{-5}	3.06×10^4
Enriched	6.5×10^5	2.3×10^{-3}	3.54×10^4

From Tables 4B-2 and 4B-3, the maximum doses were calculated for 1953 to 1989 to be always less than 1 mrem/yr, and generally less than 0.1 mrem per year. Thus, depleted and enriched uranium were insignificant contributors to inhalation dose based on the criterion put forth in the first paragraph of this attachment, and are not addressed further in this TBD. This conclusion is supported by the conclusions of the analysis by Voillequé (1999) in the Phase II study, which found that releases of enriched and depleted uranium at RFP pose substantially less risk (in terms of radiological dose) than plutonium releases.

B.4 AMERICIUM-241

Americium-241, a decay product of ^{241}Pu , exists as an undesirable contaminant in weapons-grade plutonium. Although RFP plutonium initially contained about 0.0001% ^{241}Am (ChemRisk 1994, p. 106), the ^{241}Am -to- $^{239/240}\text{Pu}$ ratio increases over time due to decay of ^{241}Pu . Not until 1985 was ^{241}Am routinely monitored at RFP. Thus, an estimate of the release rate of this isotope from all possible sources requires assumptions regarding the ratio of its activity to other alpha emitters for which activity was measured. Based on data for 1985 through 1989, the average ratio of ^{241}Am -to- $^{239/240}\text{Pu}$ emissions was about 22% (ChemRisk 1994, p. 110). Therefore, the Phase I work assumed that this ratio existed throughout the entire operational period of the facility to derive estimated releases of ^{241}Am , in $\mu\text{Ci}/\text{yr}$, listed in Table 4B-4. This assumption might not be realistic if efforts were made to separate the ^{241}Am from the plutonium being processed, but it is likely more applicable to resuspension releases of this isotope that occur years after the plutonium was deposited on the soil.

Table 4B-4 estimates show peak ^{241}Am releases occurring in 1957 (because the peak $^{239/240}\text{Pu}$ releases occurred that year from the 1957 fire) and dropping off dramatically since 1976. A total of 11 mCi is calculated to have been released, with 34% of that in 1957.

Table 4B-4 includes the inhalation dose factor assumed for ^{241}Am (ICRP 1996) and the calculated dose for each year. From Table 4B-4, the maximum dose was calculated for 1953 to 1989 to be approximately 2 mrem/yr, in 1957. However, for all other years the estimated maximum dose was less than 1 mrem/yr and orders of magnitude less for many years. Thus, ^{241}Am was an insignificant contributor to inhalation dose based on the criterion in the first paragraph of this attachment, and is not addressed further in this TBD.

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Table 4B-1. Maximum estimated annual doses for tritium.

Year	Annual tritium release ^a		95% CL Calculated air concentration ^b	Maximum tritium dose ^c
	GM ^d (Ci/yr)	95% CL ^d (Ci/yr)	uCi/m ³	mrem/yr
1953	NA	8.00E+02	1.59E-04	3.68E-01
1954	NA	8.00E+02	1.59E-04	3.68E-01
1955	NA	8.00E+02	1.59E-04	3.68E-01
1956	NA	8.00E+02	1.59E-04	3.68E-01
1957	NA	8.00E+02	1.59E-04	3.68E-01
1958	NA	8.00E+02	1.59E-04	3.68E-01
1959	NA	8.00E+02	1.59E-04	3.68E-01
1960	NA	8.00E+02	1.59E-04	3.68E-01
1961	NA	8.00E+02	1.59E-04	3.68E-01
1962	NA	8.00E+02	1.59E-04	3.68E-01
1963	NA	8.00E+02	1.59E-04	3.68E-01
1964	NA	8.00E+02	1.59E-04	3.68E-01
1965	NA	8.00E+02	1.59E-04	3.68E-01
1966	NA	8.00E+02	1.59E-04	3.68E-01
1967	NA	8.00E+02	1.59E-04	3.68E-01
1968	NA	3.90E+02	7.76E-05	1.79E-01
1969	NA	3.90E+02	7.76E-05	1.79E-01
1970	NA	3.90E+02	7.76E-05	1.79E-01
1971	NA	3.90E+02	7.76E-05	1.79E-01
1972	NA	3.90E+02	7.76E-05	1.79E-01
1973	NA	3.90E+02	7.76E-05	1.79E-01
1974	2.50E+01	5.70E+01	1.13E-05	2.62E-02
1975	3.90E+00	8.80E+00	1.75E-06	4.04E-03
1976	3.10E+00	7.00E+00	1.39E-06	3.22E-03
1977	1.40E+00	3.10E+00	6.17E-07	1.42E-03
1978	2.30E+00	5.30E+00	1.06E-06	2.44E-03
1979	2.20E+00	4.90E+00	9.75E-07	2.25E-03
1980	2.00E+00	4.60E+00	9.16E-07	2.11E-03
1981	1.10E+00	2.60E+00	5.18E-07	1.20E-03
1982	6.00E-01	1.40E+00	2.79E-07	6.43E-04
1983	4.20E-01	9.40E-01	1.87E-07	4.32E-04
1984	3.60E-01	8.20E-01	1.63E-07	3.77E-04
1985	4.20E-01	9.40E-01	1.87E-07	4.32E-04
1986	5.70E-01	1.30E+00	2.59E-07	5.98E-04
1987	4.40E-01	9.90E-01	1.97E-07	4.55E-04
1988	6.80E-02	1.50E-01	2.99E-08	6.89E-05
1989	4.40E-01	9.90E-01	1.97E-07	4.55E-04

- a. From Phase I studies (ChemRisk 1994)
- b. Based on maximum χ/Q of 6.28×10^{-6} s/m³
- c. Assuming dose factor of 9.62×10^{-1} mrem/ μ Ci and intake rate of 2,400 m³/year
- d. GM = geometric mean; CL = confidence limits

Table 4B-2. Maximum estimated annual doses for depleted uranium.

Year	Annual depleted uranium release ^a		95% CL Calculated air concentration ^b	Maximum depleted uranium dose ^c
	GM ^d (μCi/yr)	95% CL ^d (μCi/yr)	uCi/m ³	mrem/yr
1953	1.60E+03	4.00E+03	7.96E-10	5.85E-02
1954	1.60E+03	4.00E+03	7.96E-10	5.85E-02
1955	2.70E+03	7.00E+03	1.39E-09	1.02E-01
1956	1.30E+03	3.30E+03	6.57E-10	4.82E-02
1957	9.50E+02	2.40E+03	4.78E-10	3.51E-02
1958	2.10E+03	5.30E+03	1.06E-09	7.75E-02
1959	3.50E+02	9.00E+02	1.79E-10	1.32E-02
1960	4.60E+02	1.20E+03	2.39E-10	1.75E-02
1961	6.80E+02	1.70E+03	3.38E-10	2.49E-02
1962	4.80E+02	1.20E+03	2.39E-10	1.75E-02
1963	6.40E+02	1.60E+03	3.19E-10	2.34E-02
1964	3.10E+02	8.00E+02	1.59E-10	1.17E-02
1965	3.60E+02	9.30E+02	1.85E-10	1.36E-02
1966	1.80E+02	4.70E+02	9.36E-11	6.87E-03
1967	1.80E+02	4.70E+02	9.36E-11	6.87E-03
1968	1.80E+02	4.70E+02	9.36E-11	6.87E-03
1969	2.10E+02	5.30E+02	1.06E-10	7.75E-03
1970	2.50E+02	6.30E+02	1.25E-10	9.21E-03
1971	7.50E+01	1.90E+02	3.78E-11	2.78E-03
1972	5.50E+01	1.40E+02	2.79E-11	2.05E-03
1973	7.00E+01	1.80E+02	3.58E-11	2.63E-03
1974	1.20E+01	3.00E+01	5.97E-12	4.39E-04
1975	3.60E+01	9.30E+01	1.85E-11	1.36E-03
1976	1.60E+01	4.00E+01	7.96E-12	5.85E-04
1977	2.50E+01	6.30E+01	1.25E-11	9.21E-04
1978	4.30E+01	1.10E+02	2.19E-11	1.61E-03
1979	3.40E+01	8.70E+01	1.73E-11	1.27E-03
1980	2.00E+01	5.00E+01	9.95E-12	7.31E-04
1981	2.30E+01	6.00E+01	1.19E-11	8.77E-04
1982	2.50E+01	6.30E+01	1.25E-11	9.21E-04
1983	4.00E+01	1.00E+02	1.99E-11	1.46E-03
1984	7.20E+00	1.80E+01	3.58E-12	2.63E-04
1985	5.10E+01	1.30E+02	2.59E-11	1.90E-03
1986	3.80E+00	9.70E+00	1.93E-12	1.42E-04
1987	1.60E+01	4.00E+01	7.96E-12	5.85E-04
1988	1.20E+01	3.10E+01	6.17E-12	4.53E-04
1989	3.30E+00	8.30E+00	1.65E-12	1.21E-04

- a. From Phase I studies (ChemRisk 1994)
- b. Based on maximum γ/Q of 6.28×10^{-6} s/m³
- c. Assuming dose factor of 3.06×10^4 mrem/μCi and intake rate of 2,400 m³/year
- d. GM = geometric mean; CL = confidence limits

Table 4B-3. Maximum estimated annual doses for enriched uranium.

Year	Annual enriched uranium release ^a		95% CL Calculated air concentration ^b	Maximum enriched uranium dose ^c
	GM ^d (μCi/yr)	95% CL ^d (μCi/yr)	uCi/m ³	mrem/yr
1953	1.20E+01	3.00E+01	5.97E-12	5.07E-04
1954	1.20E+01	3.00E+01	5.97E-12	5.07E-04
1955	9.60E+01	2.50E+02	4.98E-11	4.23E-03
1956	1.40E+03	3.70E+03	7.37E-10	6.26E-02
1957	4.70E+02	1.20E+03	2.39E-10	2.03E-02
1958	4.00E+02	1.00E+03	1.99E-10	1.69E-02
1959	7.00E+02	1.80E+03	3.58E-10	3.04E-02
1960	1.10E+03	2.90E+03	5.77E-10	4.90E-02
1961	6.20E+02	1.60E+03	3.19E-10	2.71E-02
1962	3.20E+02	8.30E+02	1.65E-10	1.40E-02
1963	4.30E+02	1.10E+03	2.19E-10	1.86E-02
1964	2.50E+02	6.30E+02	1.25E-10	1.07E-02
1965	2.50E+02	6.30E+02	1.25E-10	1.07E-02
1966	3.00E+02	7.70E+02	1.53E-10	1.30E-02
1967	1.40E+02	3.70E+02	7.37E-11	6.26E-03
1968	2.10E+02	5.30E+02	1.06E-10	8.96E-03
1969	6.50E+01	1.70E+02	3.38E-11	2.88E-03
1970	8.30E+01	2.10E+02	4.18E-11	3.55E-03
1971	5.30E+01	1.40E+02	2.79E-11	2.37E-03
1972	5.20E+00	1.30E+01	2.59E-12	2.20E-04
1973	1.60E+01	4.00E+01	7.96E-12	6.77E-04
1974	3.50E+01	9.00E+01	1.79E-11	1.52E-03
1975	3.60E+01	9.30E+01	1.85E-11	1.57E-03
1976	2.10E+01	5.30E+01	1.06E-11	8.96E-04
1977	2.70E+01	7.00E+01	1.39E-11	1.18E-03
1978	2.70E+01	7.00E+01	1.39E-11	1.18E-03
1979	1.20E+01	3.10E+01	6.17E-12	5.24E-04
1980	2.00E+01	5.00E+01	9.95E-12	8.46E-04
1981	1.60E+01	4.00E+01	7.96E-12	6.77E-04
1982	1.60E+01	4.00E+01	7.96E-12	6.77E-04
1983	2.60E+01	6.70E+01	1.33E-11	1.13E-03
1984	2.60E+01	6.70E+01	1.33E-11	1.13E-03
1985	1.00E+01	2.60E+01	5.18E-12	4.40E-04
1986	1.40E+01	3.70E+01	7.37E-12	6.26E-04
1987	6.00E+00	1.50E+01	2.99E-12	2.54E-04
1988	3.40E+00	8.70E+00	1.73E-12	1.47E-04
1989	6.80E+00	1.70E+01	3.38E-12	2.88E-04

- a. From Phase I studies (ChemRisk 1994)
- b. Based on maximum γ/Q of 6.28×10^{-6} s/m³
- c. Assuming dose factor of 3.54×10^4 mrem/μCi and intake rate of 2,400 m³/year
- d. GM = geometric mean; CL = confidence limits

Table 4B-4. Maximum estimated annual doses for ²⁴¹Am.

Year	Annual ²⁴¹ Am release ^a		95% CL calculated air concentration ^b	Maximum ²⁴¹ Am dose ^c
	GM ^d (μCi/yr)	95% CL ^d (μCi/yr)	uCi/m ³	mrem/yr
1953	5.00E-01	1.30E+00	2.59E-13	2.21E-04
1954	1.70E+01	4.30E+01	8.56E-12	7.30E-03
1955	1.80E+01	4.70E+01	9.36E-12	7.98E-03
1956	5.70E+01	1.50E+02	2.99E-11	2.55E-02
1957	3.80E+03	9.70E+03	1.93E-09	1.65E+00
1958	7.80E+02	2.00E+03	3.98E-10	3.39E-01
1959	3.50E+02	9.00E+02	1.79E-10	1.53E-01
1960	3.20E+02	8.20E+02	1.63E-10	1.39E-01
1961	3.80E+02	9.70E+02	1.93E-10	1.65E-01
1962	7.60E+02	1.90E+03	3.78E-10	3.22E-01
1963	9.10E+02	2.30E+03	4.58E-10	3.90E-01
1964	7.00E+02	1.80E+03	3.58E-10	3.05E-01
1965	1.70E+03	4.30E+03	8.56E-10	7.30E-01
1966	8.10E+01	2.10E+02	4.18E-11	3.56E-02
1967	1.00E+02	2.60E+02	5.18E-11	4.41E-02
1968	1.20E+02	3.20E+02	6.37E-11	5.43E-02
1969	3.50E+02	9.00E+02	1.79E-10	1.53E-01
1970	9.50E+01	2.40E+02	4.78E-11	4.07E-02
1971	1.80E+01	4.70E+01	9.36E-12	7.98E-03
1972	1.50E+01	3.90E+01	7.76E-12	6.62E-03
1973	1.50E+01	3.90E+01	7.76E-12	6.62E-03
1974	2.90E+02	7.50E+02	1.49E-10	1.27E-01
1975	3.10E+01	7.90E+01	1.57E-11	1.34E-02
1976	1.20E+00	3.20E+00	6.37E-13	5.43E-04
1977	1.20E+00	3.20E+00	6.37E-13	5.43E-04
1978	8.70E-01	2.20E+00	4.38E-13	3.73E-04
1979	1.70E+00	4.30E+00	8.56E-13	7.30E-04
1980	3.60E+00	9.30E+00	1.85E-12	1.58E-03
1981	2.50E+00	6.50E+00	1.29E-12	1.10E-03
1982	6.20E+00	1.60E+01	3.19E-12	2.72E-03
1983	2.40E+01	6.10E+01	1.21E-11	1.04E-02
1984	2.40E+01	6.10E+01	1.21E-11	1.04E-02
1985	2.80E+00	7.20E+00	1.43E-12	1.22E-03
1986	1.30E+01	3.30E+01	6.57E-12	5.60E-03
1987	4.50E+00	1.20E+01	2.39E-12	2.04E-03
1988	2.80E+00	7.20E+00	1.43E-12	1.22E-03
1989	1.50E+00	3.90E+00	7.76E-13	6.62E-04

a. From Phase I studies (ChemRisk 1994)

b. Based on maximum γ/Q of 6.28 x 10⁻⁶ s/m³

c. Assuming dose factor of 3.55 x 10⁵ mrem/μCi and intake rate of 2,400 m³/year

d. GM = geometric mean; CL = confidence limits