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Rocky Flats Plant – Occupational Environmental Dose

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PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
01/06/2004	00	New technical basis document for the Rocky Flats Plant – Occupational Environmental Dose. Incorporates NIOSH and ORAU formal review comments. First approved issue. Initiated by Robert Meyer.
06/29/2004	01	Approved issue of Revision 01. NIOSH material added to Introduction; Table 4-1 columns corrected; Table 4-2 values corrected; page 50 table list change; Section B.2 ^{3}H dose factor discussion added; page 52 table ID changed. Averaged two 1973 Pu and Am intake values to provide one annual estimate; changed FEIS reference citation on page 8, and corrected reference list for 2 citations. Incorporates additional NIOSH formal review comments. Initiated by Robert Meyer.
04/23/2007	02	Approved Revision 02 initiated to update Section 4.1.1 with standard NIOSH text, updated intakes to reflect sitewide maximum intakes which required obtaining location-specific model output as opposed to location-averaged output. Converted intake units to Bq, changed recommended AMAD to 0.3 µm for some years, clarified that resuspension was addressed in modeling. Added CDPHE data for 1999–2005, other clarifications re SC&A comments. Attachment B updated to incorporate ICRP 68 organ dose factors. Worker Outreach comments pertaining to CT-0202 and CT-0208 have been addressed in a revision of Section 4.2.3. Revised to incorporate attribution per ORAU direction. Incorporates NIOSH formal review comments and an additional comment, changing recommendation of 0.3 µm to 1.0 µm AMAD for certain years. Constitutes a total rewrite of document. This revision results in an increase in assigned dose and a PER is required. Training required: As determined by the Task Manager. Initiated by Robert Meyer.
09/01/2020	03	Revision initiated to address Advisory Board comments and items associated with SEC-00192. Added discussion of Federal Bureau of Investigation raid in 1989. Updated internal intakes to be consistent with external because the sources of the data are the same. Updated onsite ambient values. Deleted Attachment B. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of document. Training required: As determined by the Objective Manager. Initiated by Mutty M. Sharfi.

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ACRONYMS AND ABBREVIATIONS

AED	aerodynamic equivalent diameter
AMAD	activity median aerodynamic diameter
AWE	atomic weapons employer
Bq	becquerel
CDPHE	Colorado Department of Public Health and Environment
Ci	curie
CL	confidence limit
cm	centimeter
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
EPA	U.S. Environmental Protection Agency
fCi	femtocurie
FEIS	final environmental impact statement
FGR	Federal Guidance Report
ft	foot
g	gram
GM	geometric mean
HEPA	high-energy particulate air
hr	hour
ICRP	International Commission on Radiological Protection
ID	identification
km	kilometer
m	meter
mCi	millicurie
mi	mile
mrem	millirem
NIOSH	National Institute for Occupational Safety and Health
ORAU	Oak Ridge Associated Universities
PER	Program Evaluation Report
RATCHET	Regional Atmospheric Transport Code for Hanford Environmental Tracking
RFETS	Rocky Flats Environmental Technology Site
RFP	Rocky Flats Plant
s	second
SC&A	S. Cohen & Associates
SEC	Special Exposure Cohort
SRDB Ref ID	Site Research Database Reference Identification (number)

Sv	sievert
TBD	technical basis document
TLD	thermoluminescent dosimeter
TLLα	total long-lived alpha
U.S.C.	United States Code
yr	year
χ/Q	maximum predicted normalized air concentration
µCi	microcurie
µm	micrometer
µrem	microrem
§	section or sections

4.1 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular Department of Energy (DOE) or Atomic Weapons Employer (AWE) facilities or categories of DOE or AWE facilities. They will be revised in the event additional relevant information is obtained about the affected DOE or AWE facility(ies). These documents may be used to assist NIOSH staff in the evaluation of Special Exposure Cohort (SEC) petitions and the completion of the individual work required for each dose reconstruction.

In this document the word "facility" is used to refer to an area, building, or group of buildings that served a specific purpose at a DOE or AWE facility. It does not mean nor should it be equated to an "AWE facility" or a "DOE facility." The terms AWE and DOE facility are defined in sections 7384l(5) and (12) of the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA), respectively. An AWE facility means "a facility, owned by an atomic weapons employer, that is or was used to process or produce, for use by the United States, material that emitted radiation and was used in the production of an atomic weapon, excluding uranium mining or milling." 42 U.S.C. § 7384l(5). On the other hand, a DOE facility is defined as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located … in which operations are, or have been, conducted by, or on behalf of, the [DOE] (except for buildings, structures, premises, grounds, or operations … pertaining to the Naval Nuclear Propulsion Program);" and with regard to which DOE has or had a proprietary interest, or "entered into a contract with an entity to provide management and operation, management and integration, environmental remediation services, construction, or maintenance services." 42 U.S.C. § 7384l(12). The Department of Energy (DOE) determines whether a site meets the statutory definition of an AWE facility and the Department of Labor (DOL) determines if a site is a DOE facility and, if it is, designates it as such.

Accordingly, a Part B claim for benefits must be based on an energy employee's eligible employment and occupational radiation exposure at a DOE or AWE facility during the facility's designated time period and location (i.e., covered employee). After DOL determines that a claim meets the eligibility requirements under EEOICPA, DOL transmits the claim to NIOSH for a dose reconstruction. EEOICPA provides, among other things, guidance on eligible employment and the types of radiation exposure to be included in an individual dose reconstruction. Under EEOICPA, eligible employment at a DOE facility includes individuals who are or were employed by DOE and its predecessor agencies, as well as their contractors and subcontractors at the facility. Unlike the abovementioned statutory provisions on DOE facility definitions that contain specific descriptions or exclusions on facility designation, the statutory provision governing types of exposure to be included in dose reconstructions for DOE covered employees only requires that such exposures be incurred in the performance of duty. As such, NIOSH broadly construes radiation exposures incurred in the performance of duty to include all radiation exposures received as a condition of employment at covered DOE facilities in its dose reconstructions for covered employees. For covered employees at DOE facilities, individual dose reconstructions may also include radiation exposures related to the Naval Nuclear Propulsion Program at DOE facilities, if applicable. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction.

NIOSH does not consider the following types of exposure as those incurred in the performance of duty as a condition of employment at a DOE facility. Therefore these exposures are not included in dose reconstructions for covered employees (NIOSH 2010):

- Background radiation, including radiation from naturally occurring radon present in conventional structures
- Radiation from X-rays received in the diagnosis of injuries or illnesses or for therapeutic reasons

4.1.1 Purpose

The preparation of this technical basis document (TBD) used available information about the Rocky Flats Plant (RFP) environment dating back to 1952.

Occupational environmental dose is the radiation dose received in the course of work duties outside Plant buildings but on the RFP site. Internal and external exposures to radionuclides in the outdoor environment are considered separately here in calculating this dose. Estimated occupational environmental dose can be used when a worker did not have the potential for routine exposures.

4.1.2 Scope

Section 4.2 presents information necessary to estimate internal environmental dose. It identifies the radionuclides of concern. Development of the list of radionuclides relied on work done for the Historical Public Exposure Studies on Rocky Flats (ChemRisk 1994a; Rood, Grogan, and Till 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, were developed for the site. The reports summarize external dose 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.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 4.5.

4.1.3 Special Exposure Cohort

The Secretary of the U.S. Department of Health and Human Services has designated the following class of employees for addition to the SEC (Sebelius 2013):

> *All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Rocky Flats Plant in Golden, Colorado, from April 1, 1952 through December 31, 1983, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees included in the Special Exposure Cohort.*

NIOSH determined that doses to unmonitored RFP workers from neptunium, thorium, and ^{233}U (and its associated ^{232}U and ^{228}Th progeny) cannot be reconstructed from April 1, 1952, through December 31, 1983. The class includes all workers during the SEC period. Because of the identified dose reconstruction infeasibility, all dose reconstructions for monitored workers during the SEC period are considered partial dose reconstructions. If monitoring data are available for workers in the SEC, dose is to be assigned as appropriate based on that data. However, such dose reconstructions are still considered partial dose reconstructions because of the determination that exposure to neptunium, thorium, and ^{233}U (and its associated ^{232}U and ^{228}Th progeny) during the SEC period cannot be bounded (NIOSH 2013). In addition, reconstruction of unmonitored neutron doses before 1967 has been identified as infeasible and therefore cannot be bounded. It is not feasible to reconstruct

unmonitored neutron doses, in a bounding manner, before 1967. Therefore, this TBD does not provide dose reconstruction guidance for unmonitored neutron doses before 1967.

4.1.4 Federal Bureau of Investigation (FBI) Raid of RFP in 1989

On October 24 and 25, 1989, a former RFP worker made allegations in a set of interviews with the U.S. Environmental Protection Agency's National Enforcement Investigation Center's Office of Criminal Investigations and the FBI. The interviews resulted from a telephone call by the interviewee to the FBI Rocky Flats Hotline on June 16, 1989, alleging safety violations and manipulation of laboratory samples at RFP.

Attachment I of ORAUT-TKBS-0011-5, *Rocky Flats Plant – Occupational Internal Dose*, provides a detailed assessment of these allegations (ORAUT 2020). The following is a summary of the conclusions of this assessment:

- No scientific basis for concluding that the issues raised about environmental samples would compromise radiological count results, and the reviewed information does not corroborate a link between the environmental and occupational radiological programs.

- There were no identified situations in which falsification or invalidation of data would affect the ability to perform dose reconstruction under EEOICPA.

- The charges against Rockwell were specific to the impact to the environment under environmental laws such as the Resource Conservation and Recovery Act of 1980 and the Clean Water Act; the charges did not specifically call out a data falsification, data validity issues, or a data quality violation.

- One worker provided information about personal involvement in shredding documents. While those documents could have been some kind of field surveys, there is no indication that those surveys have an effect on the ability to bound or reconstruct dose for the class, as long as adequate personnel monitoring data exist. These records do appear in the associated files and are available to the Project.

- An issue of "penciling-in" information on radiological field survey records was raised. The primary source of radiological information for individual dose reconstruction is the individual TLD dosimetry and bioassay information. These are performed in a laboratory and not documented in the field.

- Concerns about bioassay sample analysis results (false positives and statistical variations); bioassay sample handling and processing; personnel contamination and contamination incidents; and issues related to tritium bubblers, neptunium, Mg-Th alloy, and the Criticality Laboratory. The dose reconstruction process accounts for the potential for missed doses and incorporates methods that are favorable to claimants. All of the issues in relation to tritium bubblers, neptunium, Mg-Th, and the Criticality Laboratory are addressed in other sections of this TBD.

- A contention was made that there was an additional August 1989 aerial Multispectral Scanner Survey (MSS) at RFP in addition to one in June and July of 1989, and that the flyover data indicate the presence of the isotopes ^{137}Cs and ^{90}Sr, which was used to imply that an unreported criticality occurred. No evidence of either a second flyover in August 1989 or evidence supporting a criticality event could be found in the available RFP records. Based on interviews, document reviews, and other files, no evidence or information was identified that disputes the ability to bound RFP worker dose under the EEOICPA program.

- A review of the RFP Technical Safety Appraisal concluded that there was no identified effect on the radiological personnel monitoring data NIOSH uses to support bounding or reconstruction of dose for RFP workers.

- A review of an allegation of record falsification involving mislabeling waste for shipment off site concluded that none of this information affects the ability to reconstruct radiation dose with sufficient accuracy.

- A review of excerpts from the notebooks of an RFP manager who made notes on the radiological program did not support a data falsification issue that would affect the ability to reconstruct dose for RFP workers.

- The ORAU Team conducted a review of the quantity of available personnel radiological monitoring data for this assessment. It concluded that sufficient individual monitoring data exist to support the assessment of worker doses.

- A review of the Colorado Federal District Court Report of the Federal District Special Grand Jury (Unknown 1992) indicates that RFP had a number of violations. These included a deficient ground-water monitoring system, failure to notify government agencies of environmental law violations, storage issues, and chemical violations. However, the grand jury review of the FBI's findings did not identify any deficiencies with the radiological monitoring programs (i.e., no deficiencies are assumed to exist with the external ambient data and/or environmental air data).

Based on this information, the ORAU Team concluded this information has no effect on its ability to perform onsite ambient or environmental dose reconstructions with sufficient accuracy.

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 from 1953 to 1989, although component assembly operations actually began in 1952 and all production operations formally ceased in 1992 (ChemRisk 1992, 1994a; Voillequé and Till 1999a, 1999b, 1999c; Weber et al. 1999).

The Phase I study identified several radionuclides as potentially significant releases: ^{3}H, natural thorium, enriched and depleted uranium, $^{239/240}$Pu, ^{241}Pu, and ^{241}Am. The results of the Phase I study identified plutonium as the primary material of concern in relation to offsite exposures. The Phase II study focused the air pathway exposure assessment on the isotopes of plutonium. The potential importance of ^{3}H, enriched and depleted uranium, and ^{241}Am to onsite exposures was reevaluated here 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, p. 135).

To evaluate the potential significance of radionuclides other than isotopes of plutonium, an estimate was developed of committed inhalation dose from a 1-yr intake using airborne stack emission estimates from the Phase I study, the air dispersion modeling results from Phase II, an assumed inhalation rate of 3,000 m^{3}/yr (2,500 hr/yr × 1.2 m^{3}/hr; breathing rate is based on guidance in ICRP (1994) Publication 66). Attachment A contains a summary of the atmospheric dispersion model. Briefly, the maximum organ dose due to inhalation [assuming an activity median aerodynamic

diameter (AMAD) of 0.3 µm] in any given year since 1953 for any of these radionuclides was estimated to be that for ^{241}Am, on the order of 34 mrem for the stack releases. The maximum predicted committed doses for ^{3}H, depleted uranium, and enriched uranium for a 1-yr intake were 0.06 mrem, 1.2 mrem, and 0.65 mrem, respectively. The Phase I study estimated that ^{241}Am was potentially present in excess of 0.23 times the alpha activity of $^{239/240}$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 (ChemRisk 1994b, Appendix H). Therefore, ^{241}Am associated with soil resuspended from the 903 Area or emitted with plutonium isotopes from stacks was considered a contributor to dose. Stack releases of ^{3}H and the uranium isotopes were excluded as potentially significant contributors.

The *Final Environmental Impact Statement for the Rocky Flats Plant, Golden, Jefferson County, Colorado* (DOE 1980) indicates an isotopic composition of plutonium by weight as follows:

Table 4-1. Isotopic composition of RFP plutonium.

Isotope	Percent by weight[a]	Percent of Pu alpha activity
Pu-238	0.01	2.34
Pu-239	93.79	79.78
Pu-240	5.80	17.88
Pu-241	0.36	(beta emitter)
Pu-242	0.03	0.00165

a. Source: DOE (1980, Table 2.7.2-2).

As indicated in the table, ^{238}Pu and ^{242}Pu are minor contributors to the alpha activity of plutonium expected in the RFP environment [1]. Therefore, the following sections report source terms and intakes only for $^{239/240}$Pu and ^{241}Am.

4.2.2 Source Terms

The evaluation of atmospheric source terms for $^{239/240}$Pu and ^{241}Am that were released to the RFP environment addresses two periods: the operational period from 1952 to 1992 when production activities were ongoing and the post-operational period (1993 to 2006), 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 Operational Period Source Terms, 1952 to 1992

In 1952, the only operational activities at RFP were in Building 991 (Putzier 1982), in which components manufactured at other locations were assembled (ChemRisk 1992). No significant emissions were estimated to have occurred during this year. From 1953 through part of 1992, fabrication and recycling activities at RFP resulted in radionuclide releases to the onsite atmospheric environment as a result of “routine” operations (continuous releases), and “non-routine incidents” (discrete events). The term “routine” was used in the Radiological Assessments Corporation 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. However, routine releases “included discharges that were due to a variety of unplanned events and conditions that arose during facility operations,” including “small fires involving plutonium metal,” the “peroxide tank explosion in Building 771 in 1957, and the glovebox drain fire in Building 776 in 1965,” among other events (Voillequé and Till 1999c).

The most significant discrete release occurred during and shortly after September 11, 1957. A glovebox fire in Building 71 (also known as Building 771) resulted in a release of plutonium estimated at 21 Ci (50th-percentile estimate; Voillequé and Till 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 Ci (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 consists of the 24-hr integrated release quantities for 24 identified discrete events from 1964 to 1969. 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 non-routine contaminant release occurred during the May 11, 1969, fire, when approximately 0.037 Ci (37 mCi) of plutonium was released from the Building 779 stack (Voillequé and Till 1999b).

Total routine (nondiscrete) plutonium emissions from 1953 to 1989 are estimated to be on the order of 0.12 Ci (Voillequé and Till 1999c). This estimate does not include releases due to resuspension of contaminated soil downwind of the 903 Pad or resuspension of contaminated soil in other areas of the Plant due to deposition from the primary sources. Although the release of plutonium due to resuspension is not included in this estimate of routine emissions, it is addressed in this TBD as a contributor to exposure.

4.2.2.2 Post Operational Period Source Terms, 1993 to 2006

After 1992, production operations at RFP officially ceased. However, releases of plutonium to the environment could still occur due to contaminated soils. Average and maximum annual median ambient concentrations, provided in Table A-2 (Attachment A), are consistent and do not appear to have increased since operations ceased.

4.2.3 Annual Intake of Radioactivity

To calculate intakes of $^{239/240}$Pu and ^{241}Am, the estimated sitewide maximum annual median [2] air concentrations of these isotopes in the RFP environment were multiplied by an annual inhalation rate. The assumed breathing rate was 3,000 m^3/yr, corresponding to an hourly rate for light activity of 1.2 m^3 (based on ICRP Publication 66; 1994), and a 2,500-hr work year. Intake rates should be scaled to account for occupancy times other than 2,500 hr of exposure.

Air concentrations of $^{239/240}$Pu and ^{241}Am were estimated as described in Attachment A. Onsite air monitoring data are the preferred source of air concentrations (see Attachment A) for $^{239/240}$Pu, but in the early years (until 1964) such data were not sufficiently descriptive or complete to allow reliable estimates. Therefore, dispersion modeling results were used to estimate air concentrations of $^{239/240}$Pu for these years. During these early years, stack or building vent emissions were the main source of plutonium to onsite air, and measurements of these releases are available. Resuspension of previously deposited isotopes also contributed to onsite air concentrations. The model for this analysis (described in Attachment A) addressed contributions from the primary sources in the Phase I study (ChemRisk 1992, 1994a) as well as resuspension. The ratios of ^{241}Am activity to $^{239/240}$Pu activity in effluents were not measured until the 1980s; however, between 1985 and 1989, this ratio averaged 0.22 (ChemRisk 1994a). This ratio likely overestimates the fractional release in earlier years, when less ^{241}Am had grown in to the plutonium available for processing Therefore, the use of this ratio to estimate ^{241}Am concentrations from 1953 to 1964 is considered a favorable assumption.

After 1964, suspension or resuspension (Rood, Grogan, and Till 1999, p. 72) of contaminated soil was the main source of plutonium releases to onsite air. Air monitoring data provided either total long-lived alpha (TLLα) concentrations, from which $^{239/240}$Pu values could be derived, or actual measurements of $^{239/240}$Pu. The annual environmental reports (see the Environmental Reports section of the References) were useful in providing summaries of air concentrations by sampler location based on monthly reporting through 1994. After 1994, Rocky Flats Environmental Technology Site (RFETS) monitoring reports and the CDPHE monitoring reports provided quarterly summaries of

monitoring results. Activity concentrations of ^{241}Am were estimated after 1964 by assuming that the concentration of ^{241}Am was 30% of the $^{239/240}$Pu concentration based on measurements of the ^{241}Am/^{239}Pu activity in RFP soil by three separate researchers (Poet and Martell 1972; Krey et al. 1976; Litaor and Allen 1996). This assumption is favorable to claimants because the average measured activity ratios in soil were found to be less than 0.20 by these three groups of researchers [3].

Table 4-2 lists estimated annual intakes of $^{239/240}$Pu and ^{241}Am from 1953 to 1964, based on the atmospheric modeling described in Attachment A. The values are expressed in becquerels per year. The calculated intakes represent a maximum annual median (50th percentile) of the six computational nodes evaluated in the RFP industrial area for 500 Monte Carlo model realizations simulated for each year, and are exclusive of the buffer zone (Figure 4-1). The location of the maximum concentrations was the north-central, northeast, or southeast node, which was expected based on the locations of the primary sources and the general west-to-east wind direction. The median intakes in Table 4-2 correspond to isotopes associated with particles smaller than 15-µm aerodynamic equivalent diameter (AED), which is an upper limit for respirable particles according to Rood, Grogan, and Till (1999, p. 6) [4].

Table 4-2. Sitewide maximum annual median inhalation intakes (Bq/yr) of $^{239/240}$Pu and ^{241}Am based on atmospheric modeling, 1953 to 1964 (AED <15 µm; assume respirable).[a]

Year	Pu-239/240	Am-241[b]
1953	4.50E−05	9.75E−06
1954	2.13E−04	4.63E−05
1955	2.13E−04	4.50E−05
1956	6.88E−04	1.50E−04
1957	8.50E−02	1.88E−02
1957 w/o fire	2.63E−02	5.88E−03
1958	1.14E−02	2.50E−03
1959	1.01E−02	2.25E−03
1960	6.25E−03	1.38E−03
1961	3.63E−03	8.00E−04
1962	6.75E−03	1.50E−03
1963	1.75E−02	3.75E−03
1964	7.63E−02	1.63E−02

a. Atmospheric modeling described in Attachment A.
b. Am-241 intake is assumed to be 22% of the Pu-239/240 intake (Section 4.2.3).

Table 4-2 lists two results for 1957. Results for “1957” include the September 11 fire, which caused the annual intakes to be substantially higher than during 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” should be used for individuals who worked a partial year that did not include the month of September. Elevated releases of $^{239/240}$Pu and ^{241}Am 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 $^{239/240}$Pu from the fire are included in both 1957 values by considering resuspension of contaminated soil in the dispersion modeling).

Table 4-3 lists estimated annual intakes of $^{239/240}$Pu and ^{241}Am between 1965 and 2005. The values for $^{239/240}$Pu in this table are based on sitewide maximum annual median measured concentrations at samplers across the site, and therefore represent concentrations at the locations of higher

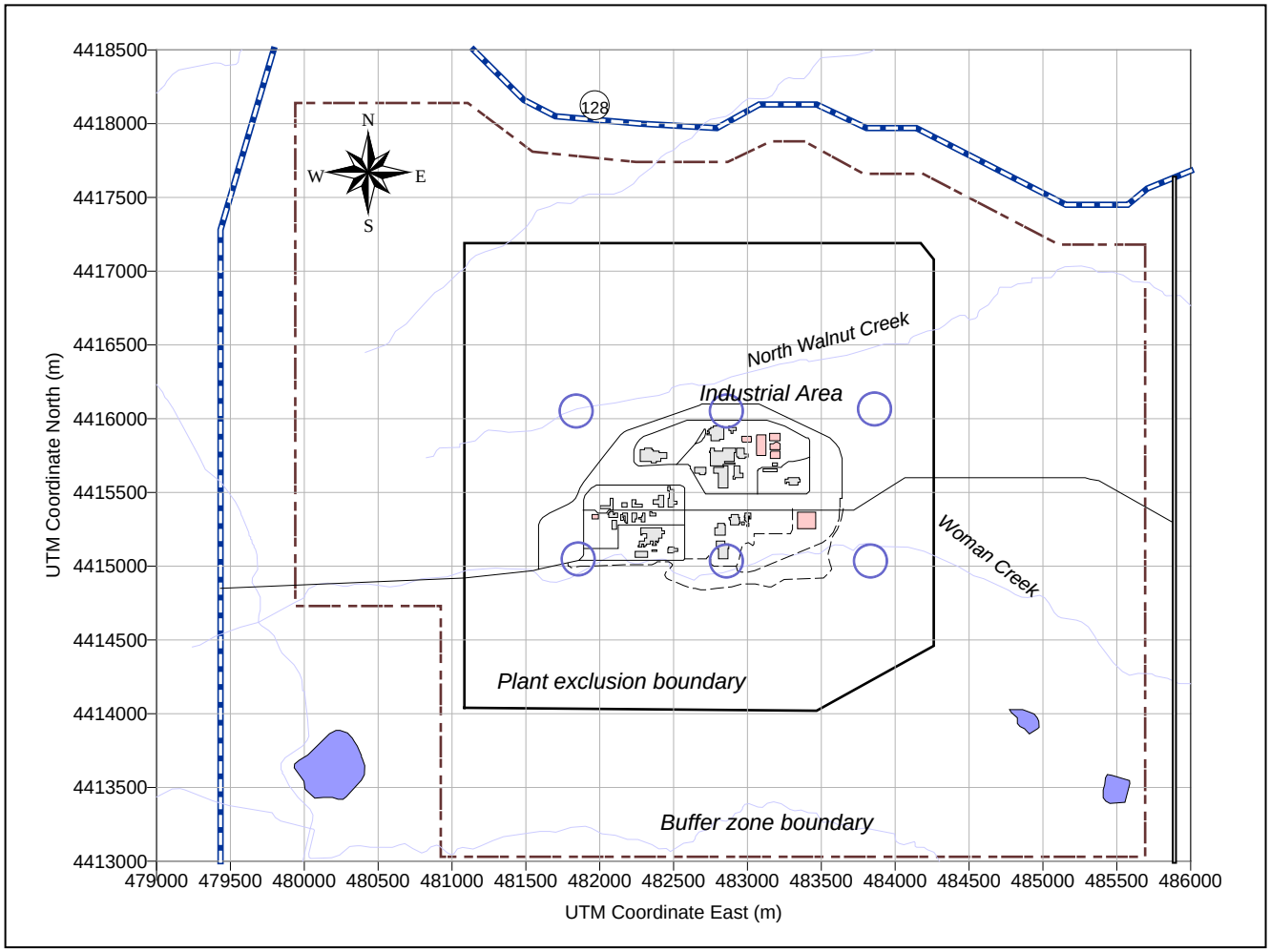

Figure 4-1. Grid map used in atmospheric modeling, showing locations of six computational nodes representing the industrial area.

concentration (typically near the 903 Area). Therefore, these values are inherently favorable to claimants when applied as an estimate of environmental exposure for the industrial area. The $^{239/240}$Pu air concentrations supporting the intakes in Table 4-3 are described in Attachment A and summarized in Table A-2. Intake rates 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. 144–146). Low- and high-volume air samplers at RFP collected particles larger than respirable (Rope et al. 1999) to varying degrees as samplers were replaced over the years. Because the respirable fraction of sampled particles is not accurately known, a respirable fraction of 1.0, which is favorable to claimants, was assumed.

The values reported for ^{241}Am in Table 4-3 (after 1970) were calculated by multiplying the $^{239/240}$Pu intake values by 0.30, in accordance with the rationale described previously. Again, a respirable fraction of 1.0, which is favorable to claimants, was assumed.

Annual intakes estimated in Tables 4-2 and 4-3 are based on a 3,000-m^3/yr (2,500 hr/yr × 1.2 m^3/hr; breathing rate, based on guidance in ICRP Publication 66 inhalation rate (ICRP 1994), but can be scaled to account for different occupancy times. Furthermore, the values can be scaled for partial year exposures, with the following cautionary note. For 1957, approximately 70% of the intake of $^{239/240}$Pu can be attributed to the September 11 fire. Therefore, 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 the 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.

Table 4-3. Sitewide maximum annual median inhalation intakes (Bq/yr) of $^{239/240}$Pu and ^{241}Am based on monitoring data, 1965 to 2005 (respirable fraction of sampled particles = 1.0).

Year	Pu-239/240[a]	Am-241[b]	Year	Pu-239/240[a]	Am-241[b]
1965	2.22E−01	6.66E−02	1986	5.55E−02	1.67E−02
1966	4.44E−01	1.33E−01	1987	1.33E−01	4.00E−02
1967	1.33E+00	4.00E−01	1988	7.77E−02	2.33E−02
1968	4.22E+00	1.27E+00	1989	5.55E−02	1.67E−02
1969	7.44E+00	2.23E+00	1990	9.99E−02	3.00E−02
1970	3.33E−01	9.99E−02	1991	1.33E−01	4.00E−02
1971	4.44E−01	1.33E−01	1992	6.66E−02	2.00E−02
1972	4.33E−01	1.30E−01	1993	4.44E−02	1.33E−02
1973	7.22E−01	2.16E−01	1994	4.44E−02	1.33E−02
1974	3.55E−01	1.07E−01	1995	2.22E−02	6.66E−03
1975	1.11E−01	3.33E−02	1996	8.88E−03	2.66E−03
1976	1.55E−01	4.66E−02	1997	7.77E−03	2.33E−03
1977	6.66E−02	2.00E−02	1998	1.11E−02	3.33E−03
1978	9.99E−02	3.00E−02	1999	1.11E−02	3.33E−03
1979	5.55E−02	1.67E−02	2000	1.11E−02	3.33E−03
1980	5.55E−02	1.67E−02	2001	2.22E−02	6.66E−03
1981	5.55E−02	1.67E−02	2002	5.55E−03	1.67E−03
1982	5.55E−02	1.67E−02	2003	2.22E−02	6.66E−03
1983	4.44E−02	1.33E−02	2004	3.33E−03	9.99E−04
1984	6.66E−02	2.00E−02	2005	1.11E−02	3.33E−03
1985	4.44E−02	1.33E−02			

a. Calculated from maximum air concentration data in Table A-2.
b. Calculated by multiplying Pu-239/240 intake by 0.30 (see Section 4.2.3).

The assumed solubility of inhaled $^{239/240}$Pu should consider the following information. Plutonium in metal-working operations and involved in fires is generally insoluble (type S or slowly transportable in the lungs), but can be highly insoluble (type SS). Exceptions, such as plutonium metal associated with solvents, might exist. Plutonium in chemical processing operations can be either soluble (type M or moderately transportable in the lungs) or insoluble (type S). The solubility of plutonium should be selected based on what is most favorable to the claimant for the organ of interest [5].

The AMAD of airborne $^{239/240}$Pu and ^{241}Am varies according to the source and therefore according to location on the Plant site. For routine releases from stacks and vents in which high-energy particulate air (HEPA) filters were operating as designed, the particle size of stack-effluent particles was reported to be on the order of 0.3 µm AMAD (Rood and Till 1999; Grogan et al. 2000). The AMADs of particles from the two larger fires (1957 and 1969) were not assessed, but there was evidence that the HEPA filtration systems did not remain intact throughout these events (Voillequé and Till 1999a, 1999b). A study of aerosols from another RFP fire involving plutonium and solvents indicated a mass median diameter on the order of 0.32 µm for this incident (Mann and Kirchner 1967), which corresponds to an AMAD of approximately 1-µm using the correlation from Hayden (1975) for RFP environmental plutonium and stack effluent air. However, the sitewide maximum intakes, based on the highest annual average concentration for any given sampler location, in Table 4-3 often reflect the air concentrations at the eastern edge of the site that resulted from suspension and resuspension of soil contaminated with $^{239/240}$Pu and ^{241}Am from the 903 Area. The AMAD of airborne particles containing plutonium in this area of RFP has been characterized as ranging from 2 to 7 µm (Grogan et al. 2000). Although the physical diameters of plutonium particles in soil were found to be on the order of 0.3- to 1-µm AMAD, it appears that the airborne plutonium particles in this area were attached to soil

particles, resulting in an aerosol most appropriately characterized by an AMAD of approximately 5 µm (Grogan et al. 2000).

Therefore, the AMAD is both time- and location-dependent for airborne radionuclides at RFP. Unfortunately, the relative contributions of the various sources of airborne plutonium at specific outdoor work locations are not known. Most of the work locations at RFP were upwind of the 903 Area plutonium source because the predominant wind direction at RFP is from the northwest (Daugherty 1989). Measurements of soil concentrations of plutonium in 1988 around RFP, at distances of 1 and 2 mi from the center of the Plant, indicated dramatically larger concentrations east and southeast of the 903 Area than in other areas (Daugherty 1989). However, the degree to which the concentrations upwind of the 903 Area were influenced by resuspended plutonium is not known. Outdoor workers in the vicinity of the buildings from which routine releases occurred, and upwind of the 903 Area, could have been exposed to particulates with AMADs on the order of 0.3 µm, although resuspended particles would also be present at those locations. The following recommendations are made based on the information given above. For estimating intakes before 1965 (Table 4-2), an AMAD of 1.0 µm should be assumed for airborne ambient $^{239/240}$Pu and ^{241}Am because worker intakes largely resulted from routine releases, the 1957 fire, and any resuspension of deposited $^{239/240}$Pu and ^{241}Am that occurred in the worker's vicinity. An AMAD of 1.0 µm should also be assumed for intakes occurring during 1965, 1966, and 1970 to 1993 [6]. The disturbance of the 903 Area as part of an attempt to remove leaking barrels, along with subsequent high-wind events that served as the main dispersal mechanism for plutonium-contaminated soils in the 903 Area, occurred between 1967 and 1969. During 1967, 1968, and 1969, and after 1993, an AMAD of 5 µm should be assumed [7]. The sitewide maximum intakes, based on the highest annual average concentration for any given sampler location, from 1967 to 1969 (Table 4-3) are dominated by the suspended plutonium that originated from the 903 Area before placement of the asphalt pad. After 1993, when production had ceased, resuspension of plutonium is the mostly likely source of airborne plutonium.

The recommendation to assume an AMAD of 1.0 µm from 1970 through 1993 is based on the fact that air concentrations downwind of the 903 Area, which are better characterized by an AMAD of 5.0 µm, tended to be only slightly (less than an order of magnitude) higher than other onsite areas, but not always. In 1972, the onsite airborne concentrations downwind of the 903 Area were 2 to 3 times higher than in other areas of RFP (Dow 1972–1973). However, in 1990 to 1992, the air concentrations in the main production areas of RFP (northern section of the industrial zone) exceeded those downwind of the 903 Area.

These assumptions are favorable to the claimant in the following respects: (1) for most organs, assuming an AMAD of 1.0 µm increases the dose by about a factor of 1.5 to 1.9 over the dose calculated by assuming an AMAD of 5.0 µm; and (2) using the sitewide maximum intakes, based on the highest annual average concentration for any given sampler location, often implicitly assumes the worker was exposed to the air concentrations downwind of the 903 Area, which was not the location of most exposures. The extrathoracic airways dose factor is a factor of 1.5 to 1.9 times higher for the AMAD assumption of 5 µm (the particle size more appropriate for resuspended plutonium) versus 1.0 µm (ICRP 2001). Therefore, for cases in which the extrathoracic airways [ET, ET1, ET2, LN(ET)] dose is of most interest, the AMAD of 5.0 µm should be assumed for all years.

4.3 EXTERNAL EXPOSURE TO ONSITE ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS

4.3.1 Radionuclides of Concern

As described in Section 4.2.1, ^{3}H, natural thorium, enriched and depleted uranium, $^{239/240}$Pu, ^{241}Pu, and ^{241}Am were identified as having been released to the air during the RFP operational period.

However, because none of the isotopes of concern is a strong gamma emitter, there is little external dose.

4.3.2 Exposure Rates

Before 1975, external exposure of workers was determined by the use of film badges. Film badges are not well suited to environmental monitoring, and environmental measurements before 1975 are not readily available. Rope et al. (1999) pointed out that before 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) spent little time discussing external occupational exposure measurements and none describing outdoor measurements. This is largely because the concern about environmental exposures to workers or the public was centered on plutonium. Rope et al. (1999) described external gamma exposures, but the information is limited to an analysis of aerial survey data that targeted plutonium by measuring ^{241}Am.

Data that can be used to estimate external environmental dose are available in the annual environmental reports for 1975 to 1994 (see the Environmental Reports section of the References). Gamma exposure rates were routinely measured at 12 to 15 locations on the site. Those data are plotted in Figure 4-2 for continuous exposures during a year (8,760 hr) and summarized in Table 4-4 for 2,500 hr/yr exposures. The locations of the monitoring stations were not available, but a sitewide maximum value was estimated by adding the recommended standard deviation (described below), reflecting both spatial and temporal variation, to the sitewide mean value [8].

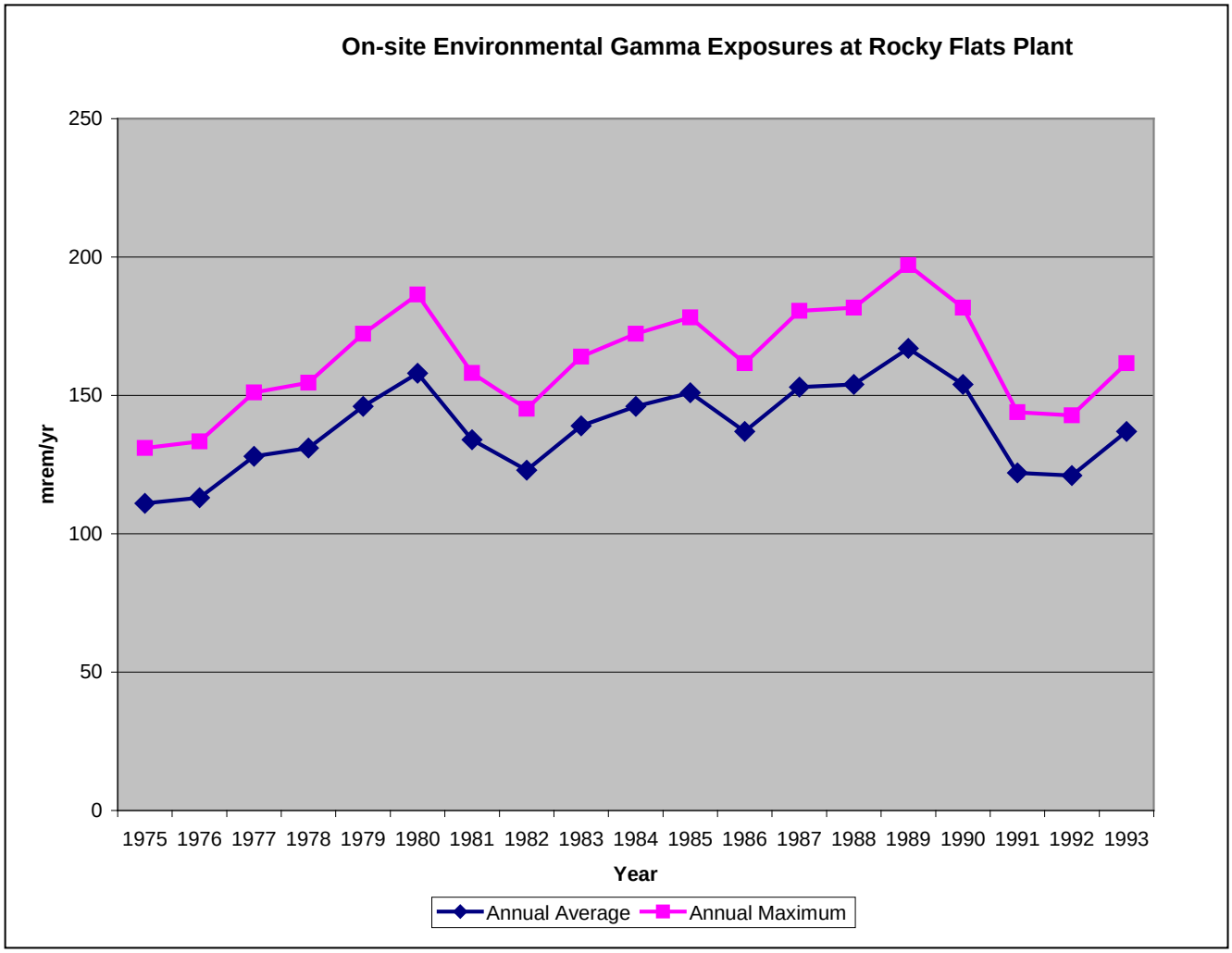

Figure 4-2. External dose (mrem/yr) as a function of year.

For the 19-yr period from 1975 to 1993, the number of independent measurements ranged from 87 to 176 per year. Table 4-4 lists the reported means and standard deviations (2σ) of the measured samples for these years with the exception of 1977. For 1977, only the mean was reported. There is no explanation available for the relatively large standard deviations in 1975, 1976, and 1978. Due to this large variation in reported standard deviations, a recommended standard deviation value (1σ) was developed based on the largest reported standard deviation. The recommended standard deviation in Table 4-4 is based on the 1975 value, which is calculated to be 18% of the mean. This recommendation is favorable to claimants because it results in higher calculated maximum dose rates [9].

Table 4-4. External gamma radiation (mrem/yr).[a]

Year	Number of measurements	Reported mean dose rate	Reported standard deviation (2σ)	Recommended standard deviation (σ)	Maximum dose rate[b,c]
1953–1964	No data	39[c]	No data	7	53
1965–1970	No data	56[c,d]	No data	10	76
1971–1974	No data	39[c]	No data	7	53
1975	87	32	11	6	44
1976	134	32	10	6	44
1977	98	37	9	7	51
1978	126	37	10	7	51
1979	133	42	3	8	58
1980	131	45	2	8	61
1981	125	38	2	7	52
1982	120	35	1	6	47
1983	135	40	1	7	54
1984	142	42	2	8	58
1985	93	43	2	8	59
1986	147	39	1	7	53
1987	100	44	1	8	60
1988	109	44	2	8	60
1989	132	48	1	9	66
1990	143	44	1	8	60
1991	108	35	1	6	47
1992	176	35	1	6	47
1993	176	39	1	7	53
After 1993	No data	39[c]	No data	7	53
Mean 1975–1993	Not applicable	**39[c]**	Not applicable	**7**	Not applicable

a. Dose rate corresponding to an exposure time of 2,500 hr/yr.
b. Calculated by adding the recommended 2 x standard deviation to the mean dose rate, which is 18% of the mean.
c. Average of means reported for 1975 through 1993.
d. The calculated contribution from americium in barrel storage area before placement of asphalt pad was determined to be 17 mrem/yr.

There is no significant trend to the means and maximums 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 measured exposure rates. 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 39 ± 7 mrem/yr (2,500 hr) to years not listed in this table. This value represents the average of means corresponding to the 19 yr for which measurements were reported. The assumed standard deviation associated with this average value is based on the assumed 18% standard deviation. This dose rate represents the total dose, including background, so use of these values in estimating individual worker doses is favorable to the claimant.

The listed mean dose rate, in Table 4-4, should be assigned as a normal distribution with the error as twice the recommended standard deviation as a best estimate for a 2,500-hour work year exposure. Dose rates should be scaled to account for occupancy times other than 2,500 hr of exposure.

Ambient onsite gamma is significantly larger ($p < 0.05$) than ambient gamma measured at “perimeter” stations, but only by an average of 9% for years for which data were reported (see the Environmental Reports section of the References). This would seem to indicate that about 3 mrem/yr (for a

2,500-hr/yr exposure) could be contributed by contamination inside the exclusion zone boundary for these years. Therefore, increases in airborne contamination do not necessarily indicate significant increases in total external dose. However, between 1965 and 1970, the large increase in inhalation intake of plutonium is due to soil contamination of the 903 Area before paving of that area. It is likely that the external dose rate at that location might have been elevated in comparison with other locations at the site due to the ^{241}Am present. Unfortunately, soil concentrations in the barrel storage location are not available and few soil concentration data are available for the site before 1969 (Rood, Grogan, and Till 1999). In an attempt to estimate external dose to an onsite worker from contaminated soils in the vicinity of the 903 storage area, a plutonium soil contamination value of 26 µCi/m^2 (9.6 × 10^5 Bq/m^2) was assumed based on the 1973 site environmental report, which was the maximum observed value in 1970 just downwind of the 903 Pad. Using Federal Guidance Report (FGR) No. 12 (Eckerman and Ryman 1993) dose conversion factors for surface-contaminated soils, assuming that the maximum observed concentration was constant over the entire site, and using the mean isotopic ratio reported by Krey et al. (1976) for ^{241}Am/^{239}Pu in RFP soil of 0.13, doses can be calculated at 1 m above ground surface of approximately 3 mrem/yr (for 2,500-hr/yr exposure).

Another approach taken to estimate the soil concentration in the barrel storage area before paving was based on estimates of the total ^{239}Pu released. According to Weber et al. (1999), the highest release estimate for ^{239}Pu in the 903 storage area was on the order of 1,000 g, which corresponds to 62 Ci. If one assumes that this is spread over the 550- by 475-ft contamination zone of the 903 Area originally described (Weber et al. 1999), a maximum estimate of external dose of 17 mrem/yr for ^{241}Am was obtained [assuming the Krey et al. (1976) mean isotopic ratio of 13% for ^{241}Am:^{239}Pu, the appropriate FGR 12 dose conversion factor, a depth of contamination of 15 cm, and 2,500-hr/yr exposure].

Finally, this TBD analysis attempted to ascertain what other sources of external exposure might have been present. Using dose factors in FGR No. 12 (Eckerman and Ryman 1993), the analysis calculated external dose from submersion in air containing ^{241}Am. The ^{241}Am levels were calculated using isotopic ratios published by Krey et al. (1976) and modified by Radiological Assessments Corporation when developing soil action levels for RFETS. Doses from ^{241}Am in that scenario were negligible, less than 1 µrem/yr.

The results of these considerations suggest that use of the ambient gamma values for external environmental dose is reasonable for unmonitored workers throughout the lifetime of the site, with the possible exception of the period after plutonium began leaking from the barrel storage area and before placement of the asphalt pad in that area. The maximum estimated increment to the measured ambient gamma levels was calculated to be 17 mrem/yr for a 2,500-hr/yr exposure. Therefore, this quantity should be added to the measured mean gamma level (39 mrem/yr) applied for 1965 to 1970.

4.4 UNCERTAINTY

Uncertainties in estimates of plutonium intake and external dose to a workers at 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 Estimated Intakes, 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, Grogan, and Till (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 the physical processes that govern the behavior of the system, due either to insufficient knowledge of the processes or inability to measure isolated mechanisms that drive 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 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 application of the Phase II dispersion model, model output (in curies per year of annual intake and converted to becquerels per year of intake for Table 4-2) 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. The distribution associated with the percentiles is not readily described as either normal or lognormal.

The components of uncertainty reflected in the annual intake estimates reflect uncertainty only in the source terms and environmental transport. Such components of uncertainty are real in the sense that 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 Estimated Intakes, After 1964

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 (see the Environmental Reports section of the References). For these years, error terms representing two standard deviations about the mean (at the 95% confidence level) were calculated assuming the measurements were normally distributed. In 1977 and 1978, no error terms were reported because "they can be misleading and (their use) is considered inappropriate for the data in this report" (Barker et al. 1978; Bokowski et al. 1979). These 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 were reported, but only by sampler location and not normalized to the site average. The error terms, when reported, were based on counting error alone and therefore 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

is also be a function of variability in environmental conditions as well as a function of both time and location. Therefore, the reported confidence limits (CLs) 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 before 1975 or after 1993. However, onsite measurements made during that 19-yr period indicate no significant upward trend to indicate that Plant operations were increasing onsite exposures. Published data for environmental external measurements have normal statistics associated with them. The data in Table 4-4 vary considerably in the expression of uncertainty from the annual environmental reports. For years before 1977, no uncertainty data are available. For other years, the uncertainties vary from as much as 18% (1σ) for 1975 to less than 2% for 1983. This TBD recommends using the maximum uncertainty available in the records, 18%, which is represented by 1981 data. The annual recommended values of uncertainty, expressed in terms of 1σ, are listed in Table 4-4.

4.5 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify the source and justification for each associated item. Conventional References, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database (SRDB).

[1] McDowell-Boyer, Laura M. Oak Ridge Associated Universities (ORAU) Team. Environmental Engineer. July 2006.
The recommendation to ignore contributions of ^{238}Pu and ^{241}Pu is based on the facts that (1) their total alpha activity contributions are less than 5% of those of ^{239}Pu and ^{240}Pu based on the reported isotopic composition of plutonium at RFP, and (2) the maximum inhalation organ dose factors for ^{238}Pu and ^{241}Pu are equal to or less than those for ^{239}Pu and ^{240}Pu. Therefore, ^{238}Pu and ^{241}Pu do not contribute more than a few percent of the dose from plutonium.

[2] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
ORAUT-PROC-0031, *Site Profile and Technical Basis Document Development* (ORAUT 2012), provides guidance in TBD preparation, and specifies that maximum sitewide annual median intakes via inhalation will be provided.

[3] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
The measurements by Poet and Martell (1972) indicated that the mean activity ratio in soil ranged from 0.03 to 0.17, but the authors acknowledged that low recovery yields associated with their laboratory procedures resulted in large errors for their ^{241}Am results. Krey et al. (1976) measured the ^{241}Am/^{239}Pu ratios in soil, and found a range from 0.10 to 0.20 with a mean of 0.13 and standard deviation of 0.03. Both authors acknowledged that the ratio would increase over time and peak 70 to 80 yr after plutonium purification (around 2030). Assuming the initial isotopic composition of plutonium from Section 4.2.1, with ^{241}Pu at 0.36%, the resulting peak ratio would be about 0.18 (Poet and Martell claimed this peak was 0.54, but had assumed an isotopic ratio of 1% for ^{241}Pu, which is almost 3 times too high). In 1996, Litaor and Allen (1996) found an average onsite ratio of 0.19, but they indicated difficulties related to high analytical errors with ^{241}Am. Therefore, due to uncertainty in the relative movement of the two isotopes in soil, a higher value of 0.30 is recommended.

[4] Rood, Arthur S. K-Spar Inc. President. July 2006.
The puff trajectory model, Regional Atmospheric Transport Code for Hanford Environmental Tracking (RATCHET), was adapted by Arthur S. Rood from its use for the Phase II Historical Public Exposures Studies for RFP for use in estimating onsite inhalation intakes from 1953 to 1964. The model and adaptations are described in Attachment A, Section A.2.

[5] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
Due to variations in the original source of particles (fires, cutting oil, etc.) and environmental factors, it is recommended that the dose reconstructor always make a selection of dose factor that is favorable to claimants in relation to plutonium solubility.

[6] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. January 2007.
The recommendation to assume the default ICRP Publication 66 AMAD of 1.0 µm ("recommended for exposure in the general environment"; ICRP 1994) during certain periods results in using inhalation organ dose factors that are higher than those for larger respirable particles (those on the order of 5 µm). The cited literature indicates that intakes from plutonium released routinely or from fires during the 1953-to-1966 and 1970-to-1993 periods are possibly smaller than the 5-µm ICRP Publication 66 default recommended for "occupational exposure settings" due in part to the presence of HEPA filter banks at the emission points.

[7] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
Plutonium-contaminated particles suspended or resuspended from soil and other surfaces are known to contribute a significant portion of the airborne activity of plutonium in certain areas of the RFP site, and many of these particles are better characterized by an AMAD of 5 µm. From 1967 to 1969, a number of high-wind events occurred during and after removal of leaking barrels containing plutonium-contaminated cutting oil in the 903 Area, which resulted in dispersal of significant quantities of contaminated soil particulates, mainly in the easterly to southeasterly direction from the 903 Area. The sitewide maximum air concentrations, and resulting estimated intakes, are associated with this region of RFP. The airborne plutonium in this region is best characterized by an AMAD of 5 µm. After 1993, production had ceased, and it is therefore reasonable to expect that the main source of airborne plutonium was resuspension from soil or surfaces and that HEPA-mediated emissions would no longer have occurred.

[8] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
ORAUT-PROC-0031, *Site Profile and Technical Basis Document Development* (ORAUT 2012), provides guidance in TBD preparation and specifies that a site average value be provided along with the standard deviation and a maximum onsite dose rate (derived by adding the standard deviation to the average).

[9] Little, Craig A. ORAU Team. Radioecologist/Health Physicist. 2006
The average of the reported standard deviations (2σ) without the 1975-to-1978 values is approximately 5. The recommended value of 1σ is approximately 10 times higher than most reported values due to the uncertainty posed by the unexplained larger variabilities in 1975, 1976, and 1978. This is an upper bound on the uncertainty, and the resulting estimates of the maximum dose rate are therefore favorable to claimants.

[10] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
Rope et al. (1999, pp. 108–109) did an extensive review of environmental data available for the RFP site and based this least-squares, best-fit estimate of the plutonium contribution to TLLα on data from the Health and Safety Laboratory and Dow, making standard temperature and pressure corrections as necessary. The results were very close to an estimate made by

another researcher who, rather than using a best-fit analysis, assumed a value between 33% and 50% based on a log-log plot of data from CDPHE. The Rope et al. value appears to carry more scientific weight, but it is not in conflict with the other results. Therefore, the value of 0.36 is considered the best estimate available for the period of interest.

[11] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
An electronic presentation addressing special monitoring during decommissioning and demolition projects at RFETS indicates that impacts from various demolition projects undertaken in 1999 (Building 779 glovebox strip-out, Building 729 stack removal, Building 788 demolition, Building 729 demolition, Building 729 clarifier tank removal) were not significant in terms of measured isotopic air concentrations.

[12] Rood, Arthur S. K-Spar Inc. President. July 2006.
Mr. Rood developed the RATCHET application for the Phase II Historical Public Exposure Studies project. He made modifications to the model for this study using the same source terms identified in the earlier work.

[13] Rood, Arthur S. K-Spar Inc. President. July 2006.
The Building 776 stack conforms to the U. S. Environmental Protection Agency (EPA) definition of good engineering practice design. According to the EPA 1995 Building Profile Input Program, building wake effects extend out to 5 times the lesser of the building height or the building perimeter width, such that Building 771 is the only building within the sphere of influence of the 776 stack. The 776 stack must be at least 29 m high (since the Building 771 height is 11.6 m) to conform to good engineering practice and therefore be free of the effects of building wakes. Because the 776 stack is 44 m high, building wake effects are not a concern.

[14] Rood, Arthur S. K-Spar Inc. President. July 2006.
The other modeled elevated sources were the Building 776 roof vents. Because some of the roof vents were an inverted “J” type, effluent was directed down toward the roof, thereby distributing the source across the roof surface. To account for this, Building 776 was modeled not as a point source but as an elevated area source and initial dispersion coefficients were assigned. The coefficients implicitly account for the effects of building wakes by driving the plume initially down to the ground surface.

[15] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
Rope et al. (1999, pp. 108–109) did an extensive review of environmental data available for the RFP site and based this least-squares, best-fit estimate of the plutonium contribution to TLLα on data from the Health and Safety Laboratory and Dow, making standard temperature and pressure corrections as necessary. The results were very close to an estimate made by another researcher who, rather than using a best-fit analysis, assumed a value between 33% and 50% based on a log-log plot of data from CDPHE. The Rope et al. value appears to carry more scientific weight, but it is not in conflict with the other results. Therefore, the value of 0.36 is considered the best estimate available for the period of interest.

[16] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
There is no evidence that background radioactivity in the Denver area would be significantly different from that in the RFP area; therefore, this assumption is reasonable.

[17] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
Radionuclides other than $^{239/241}$Pu and ^{241}Am that have been released from RFP and contribute less than 1-mrem/yr dose to any organ in the year of maximum release are not expected to contribute more than 5% of the total environmental dose in comparison with the maximum

dose contributions of $^{239/240}$Pu and ^{241}Am in the same year. This was verified by comparing the maximum estimated doses for these other radionuclides (Attachment B, Tables B-1 to B-3) and the sitewide maximum doses associated with the $^{239/240}$Pu and ^{241}Am inhalation intake values in Tables 4-2 and 4-3.

[18] McDowell-Boyer, Laura M. ORAU Team. Environmental Engineer. July 2006.
This statement is supported by the text following it, which demonstrates considerable effort by ChemRisk to review effluent monitoring data, including raw data when necessary. It is important to recognize that ChemRisk reevaluated the uncertainty associated with the reviewed data and proposed larger uncertainties due to possible measurement and recording errors.

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GLOSSARY

alpha emitter
Radioisotope that decays by emitting an alpha particle.

alpha particle (α)
See *alpha radiation*.

alpha radiation
Positively charged particle emitted from the nuclei of some radioactive elements. An alpha particle consists of two neutrons and two protons (a helium nucleus) and has an electrostatic charge of +2.

becquerel
International System unit of radioactivity equal to 1 disintegration per second; 1 curie equals 37 billion (3.7×10^{10}) Bq.

beta radiation
Charged particle emitted from some radioactive elements with a mass equal to 1/1,837 that of a proton. A negatively charged beta particle is identical to an electron. A positively charged beta particle is a positron.

curie
Traditional unit of radioactivity equal to 37 billion (3.7×10^{10}) becquerels, which is approximately equal to the activity of 1 gram of pure ^{226}Ra.

depleted uranium
Uranium with a percentage of ^{235}U lower than the 0.7% found in natural uranium.

dosimeter
Device that measures the quantity of received radiation, usually a holder with radiation-absorbing filters and radiation-sensitive inserts packaged to provide a record of absorbed dose received by an individual. See *thermoluminescent dosimeter*.

dosimetry
Measurement and calculation of internal and external radiation doses.

enriched uranium
Uranium in which processing has increased the proportion of ^{235}U to ^{238}U to above the natural level of 0.7% by mass. Reactor-grade uranium is usually about 3.5% ^{235}U; weapons-grade uranium contains greater than 90% ^{235}U.

exposure
(1) In general, the act of being exposed to ionizing radiation. (2) Measure of the ionization produced by X- and gamma-ray photons in air in units of roentgens.

film
In the context of external dosimetry, radiation-sensitive photographic film in a light-tight wrapping. See *film dosimeter*.

film dosimeter
Package of film for measurement of ionizing radiation exposure for personnel monitoring purposes. A film dosimeter can contain two or three films of different sensitivities, and it can

contain one or more filters that shield parts of the film from certain types of radiation. When developed, the film has an image caused by radiation measurable with an optical densitometer. Also called film badge.

gamma radiation

Electromagnetic radiation (photons) of short wavelength and high energy (10 kiloelectron-volts to 9 megaelectron-volts) that originates in atomic nuclei and accompanies many nuclear reactions (e.g., fission, radioactive decay, and neutron capture). Gamma photons are identical to X-ray photons of high energy; the difference is that X-rays do not originate in the nucleus.

gamma ray, particle, or photon (γ)

See *gamma radiation*.

isotope

One of two or more atoms of a particular element that have the same number of protons (atomic number) but different numbers of neutrons in their nuclei (e.g., ^{234}U, ^{235}U, and ^{238}U). Isotopes have very nearly the same chemical properties.

nuclide

Stable or unstable isotope of any element. Nuclide relates to the atomic mass, which is the sum of the number of protons and neutrons in the nucleus of an atom. A radionuclide is an unstable nuclide.

operating area

Usually refers to a major operational work area at a site.

radiation

Subatomic particles and electromagnetic rays (photons) with kinetic energy that interact with matter through various mechanisms that involve energy transfer.

radioactivity

Property possessed by some elements (e.g., uranium) or isotopes (e.g., ^{14}C) of spontaneously emitting energetic particles (electrons or alpha particles) by the disintegration of their atomic nuclei. See *radionuclide*.

radionuclide

Radioactive nuclide. See *nuclide*.

rem

Traditional unit of radiation dose equivalent that indicates the biological damage caused by radiation equivalent to that caused by 1 rad of high-penetration X-rays multiplied by a quality factor. The sievert is the International System unit; 1 rem equals 0.01 sievert. The word derives from roentgen equivalent in man; rem is also the plural.

thermoluminescent dosimeter (TLD)

Device for measuring radiation dose that consists of a holder containing solid chips of material that, when heated, release the stored energy as light. The measurement of this light provides a measurement of absorbed dose. TLDs replaced film dosimeters at essentially all U.S. Department of Energy sites beginning in the 1960s.

ATTACHMENT A
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS

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ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

A.1 INTRODUCTION

Occupational environmental dose reconstruction for RFP requires that $^{239/240}$Pu air concentrations be estimated for onsite locations as a function of time. Actual measurements of $^{239/240}$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, which adds 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 $^{239/240}$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 periods. In addition, it provides the technical basis of atmospheric transport calculations that support the estimated $^{239/240}$Pu intakes.

A.2 MEASUREMENT OF TOTAL LONG-LIVED ALPHA ACTIVITY AND PLUTONIUM IN AIR

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

According to Rope et al. (1999), $^{239/240}$Pu concentrations in air were not measured at RFP until 1969, when CDPHE 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. 96–98) to be of limited value for assessing concentrations of long-lived alpha emitters such as $^{239/240}$Pu. Before 1960, measurements were not made in a way that enabled estimation of the long-lived alpha component of the gross alpha measurements. Therefore, there is no reliable way to estimate $^{239/240}$Pu in air before 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, so that TLLα activity concentrations could be estimated. Estimates of $^{239/240}$Pu concentrations can be made from TLLα activity concentrations in a manner described later in this attachment. However, data from 1960 to 1964 were 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. Therefore, data from before 1964 were either not suitable or not readily available for this TBD. Due to these inadequacies in monitoring data during the period between 1952 and 1964, this TBD relied on atmospheric dispersion modeling for the Phase II study for developing estimates of $^{239/240}$Pu concentrations in onsite air. This section describes the Phase II model and application for the present purposes.

From October 1964 until December 1971, the RFP contractor reported daily TLLα activity concentrations in air for individual onsite samplers (Rope et al. 1999, p. 99). 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 A-1 (reproduced from Figure B-6 in Rope et al. 1999). Monthly average concentrations derived from these measurements, in fCi/m^3 (1×10^{-15} Ci/m^3), are reproduced from Table B-6 in Rope et al. in Table A-1. (Note: Figures and tables appear at the end of this attachment.)

In 1969, CDPHE began monitoring $^{239/240}$Pu in air at the RFP (Rope et al. 1999, p. 86) near the east security fence. Rope et al. (1999, p. 88) used data from the CDPHE air monitoring stations to

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ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

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, 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. 77).

In 1970, the RFP environmental monitoring program began to include routine monitoring of $^{239/240}$Pu in air (Rope et al. 1999, p. 128). However, results for $^{239/240}$Pu, rather than TLLα, were not reported in RFP annual environmental reports until 1973 (see the Environmental Reports section of the References). Table A-2 lists annual air concentrations, averaged over the onsite samplers and compiled from annual environmental reports through 1994 and the monthly data in Table A-1, along with the maximum onsite monthly measurement corresponding to that year. The units have been converted to femtocuries per cubic meter for consistency with Table A-1. Sampling locations are specified, and can be visualized from Figure A-1. The samplers were renumbered in 1975, as indicated in this figure. The annual average was not provided in the reports 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. Therefore, results after 1976 are not representative of an average for the RFP industrial area, but rather for the onsite areas likely to have the highest concentrations of $^{239/240}$Pu in air.

To estimate the $^{239/240}$Pu air concentration based on TLLα activity concentration for data from 1965 to 1973 in Table A-2, a conversion factor was adopted from the review of available information on this topic in Rope et al. (1999, p. 107). Rope et al. evaluated data from the early 1970s and derived a least-squares best fit to the data result of 36% plutonium contribution to the TLLα. Some of the TLLα activity in onsite air was due to naturally occurring alpha emitters and fallout plutonium. The remainder could be attributed to plutonium, americium, and uranium from RFP (Rope et al. 1999). A conversion factor of 0.36 was assumed for converting TLLα activity concentrations from the annual environmental reports to $^{239/240}$Pu activity concentrations [10].

The annual environmental reports provide 95% CLs for the average annual $^{239/240}$Pu air concentration for RFP for 1972 to 1976 and 1989 to 1994. For these years, the standard deviation was reported as the 95% CLs, 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” (Barker et al. 1978; Bokowski et al. 1979). These 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 were reported for 1979 to 1988, but only by sampler location, and not normalized to the site average. The CLs, when reported, were based on counting error alone and therefore 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 was also a function of variability in environmental conditions as a function of time and location. Therefore, the reported CLs 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 $^{239/240}$Pu in air after 1994 are not as readily accessible. Between 1995 and July 1998, onsite ambient air concentrations of $^{239/240}$Pu were reported in RFETS and other monthly and quarterly monitoring reports (EG&G 1995; RMRS 1995a to 1998d). For 1995, an onsite average air concentration could be calculated from the reported data for approximately 20 sampling locations, and a sitewide maximum annual average corresponding to the sampler at the

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

After July 1998, onsite air concentrations have not been reported directly in the quarterly reports. However, CDPHE reported onsite concentrations in the eastern area of the industrial zone, where air concentrations tended to be highest (downwind of the contaminated soil area), after 1998. The concentrations CDPHE reported were used to estimate sitewide maximum air concentrations for years after 1998 and are included in Table 4-3. Special project monitoring has been carried out to monitor specific demolition and remediation activities. Formal reporting of this monitoring activity has been requested [11].

A.3 ATMOSPHERIC TRANSPORT MODELING

The atmospheric transport modeling results that were used to develop estimates of $^{239/240}$Pu intake by workers between 1953 and 1964 relied on application of a model developed for the offsite risk assessment (Rood, Grogan, and Till 1999) [12]. Application of this model for onsite predictions was not the original intent, and simulated concentrations are considered less reliable than measured concentrations for estimates of $^{239/240}$Pu intake for reasons stated in the introductory paragraph to this attachment. The application of this model for the Phase II Historical Public Exposures Studies focused on offsite rather than onsite concentrations, the latter being of interest here. However, the model application is such that building wake effects, a potential concern for areas close to the sources, are not significant for two reasons. First, releases from the Building 771 stack were not likely to be affected by building wakes because the 44-m stack is sufficiently high in relation to nearby buildings so that the plume was not significantly affected [13]. Second, all other elevated sources in the model were treated as area sources, such that initial dispersion was assigned, which implicitly accounts for the effects of building wakes [14].

Rood, Grogan, and Till (1999) described atmospheric transport modeling as follows (literature citations removed from quotation):

> *Atmospheric transport modeling performed in the Phase II studies used the Regional Atmospheric Transport Code for Hanford Environmental Tracking (RATCHET) model. Selection of RATCHET was based on a model comparison study performed for Phase II. In this study, five models, ranging in complexity from a simple straight-line Gaussian plume model (Industrial Source Complex Short Term Version 2) to a complex terrain model (Terrain-Responsive Atmospheric Code), were compared to tracer measurements taken during the 1991 Winter Validation Tracer Study. The results of this evaluation indicated no one model clearly outperformed the others. However, the puff trajectory models (RATCHET, TRIAD, and INPUFF2) 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.*

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Atmospheric transport simulations were performed differently for discrete and continuous events. For discrete events, 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 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 × 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 and most of the contaminant plumes traveled east and southeast of the plant.

Figure 3 from Rood, Grogan, and Till (1999) is not reproduced here because of the irrelevance of much of the model domain. Rather, Figure A-2 shows the model domain of relevance, which is the RFP industrial area. There are six nodes in the industrial area, which provides 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 that occurred over a relatively short period, and these 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 those did not include the above-noted high-wind events. Resuspension of $^{239/240}$Pu deposited on soil from 903 Area suspension releases and other routine and non-routine events was included in the comprehensive evaluation of exposure to $^{239/240}$Pu released from RFP (Rood, Grogan, and Till 1999). A time-dependent factor approach was used to address the resuspension of soil contaminated by released $^{239/240}$Pu and deposited as a result of the continuous and discrete events. Rood, Grogan, and Till (1999, p. 32) described the source term characterization.

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The aerodynamic diameter of released $^{239/240}$Pu particles is important in predicting dispersion and deposition and ultimately in estimating the respirable fraction of the $^{239/240}$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. Therefore, the results of the modeling enabled prediction of the $^{239/240}$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 A-3 plots the annual average $^{239/240}$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 A-2, and are for particles less than 30-µm AED. Figure A-4 plots annual average $^{239/240}$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 A-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.

The measured data in Figure A-3 represent the average concentrations across the onsite samplers listed in the legend. Samplers S-1 to S-10, S-50, and S-51 were maintained by an RFP contractor throughout the period of interest. The annual average TLLα data from 1965 through 1972, calculated from the data in Table A-1, were converted to $^{239/240}$Pu concentrations for comparison to the model predictions. A conversion factor of 0.36 (0.36 Ci $^{239/240}$Pu per Ci TLLα) reported in Figure III-23 in Rope et al. (1999) was used [15]. Average concentrations from samplers S-5 to S-9 were obtained from Table B-10 in Rope et al. (1999), which is reproduced in Table A-3. The onsite values in this table tend to be slightly higher than the averages in Table A-2 because they are restricted to samplers S-5 to S-9 only for all years. Figure A-3 also plots the arithmetic average of measurements from CDPHE samplers D-3, D-4, and AP-56 at the eastern security fence of RFP (Figure A-6). The data from which averages were derived for these CDPHE samplers are listed in Table A-4, which was reproduced from Table III-4 in Rope et al. (1999).

From Figure A-3, it is clear that the modeled 50th-percentile concentrations could 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 $^{239/240}$Pu intake at RFP is that reported $^{239/240}$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, Grogan, and Till 1999, p. 7). Therefore, air concentrations in the area of highest soil plutonium (downwind of the 903 Area) would be likely 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 exceeded 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 $^{239/240}$Pu concentration in air. Therefore, 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 $^{239/240}$Pu concentration.

The measured data in Figure A-4 for the perimeter concentrations were from the 1971 to 1990 annual environmental reports prepared by RFP contractors and represent average concentrations from RFP contractor perimeter samplers (shown in Figure A-7 as triangular symbols). These data were

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corrected for contributions from background $^{239/240}$Pu because the measured values are low enough that fallout concentrations can contribute a significant portion of the total measurement. Yearly measurements of $^{239/240}$Pu in air in Denver (Rope et al. 1999, Table B-14, reproduced in Table A-5) were assumed to be representative of RFP background levels [16]. These levels were subtracted from the perimeter measurements in Table A-3 to obtain the average net perimeter $^{239/240}$Pu concentrations listed in Table A-6 and plotted in Figure A-4. From Figure A-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 $^{239/240}$Pu concentrations support the following conclusions. Onsite application of the atmospheric dispersion model that was 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 therefore 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 before 1970, to use the Phase II model application for estimating onsite average concentrations in lieu of measurement data.

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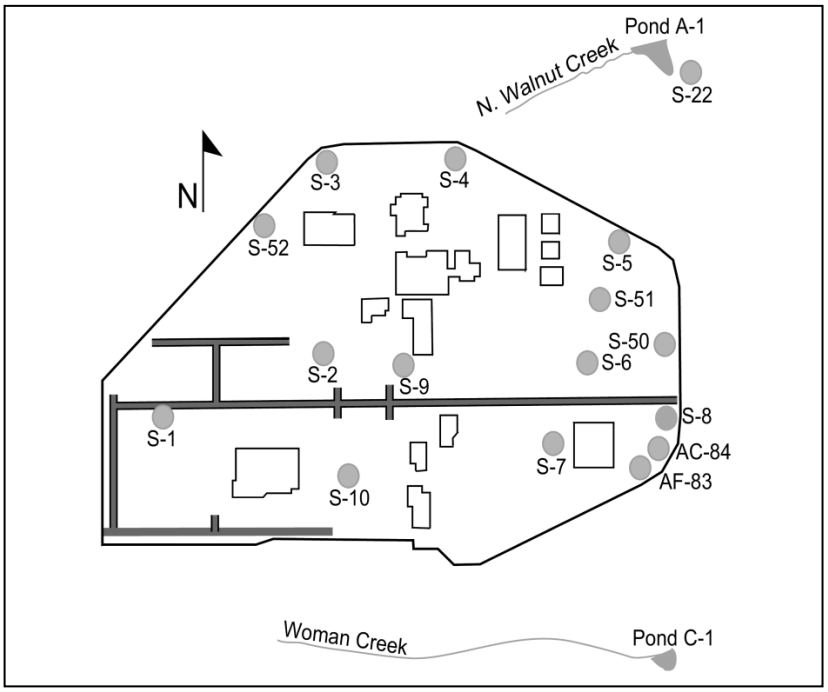
1974



1975

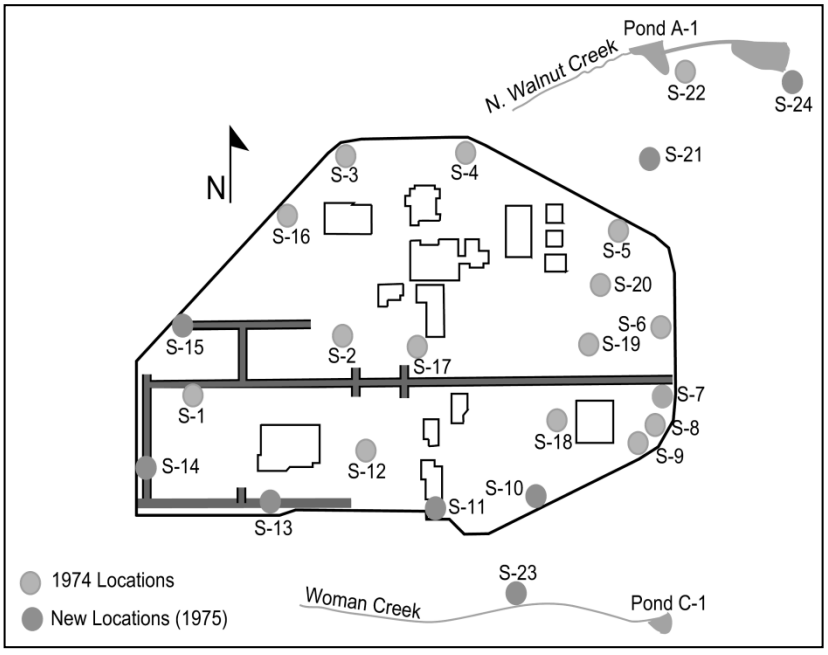


Figure A-1. Locations of samplers operated by contractor (Rope et al. 1999, Figure B-6). In 1975, samplers were renumbered and new samplers were added.

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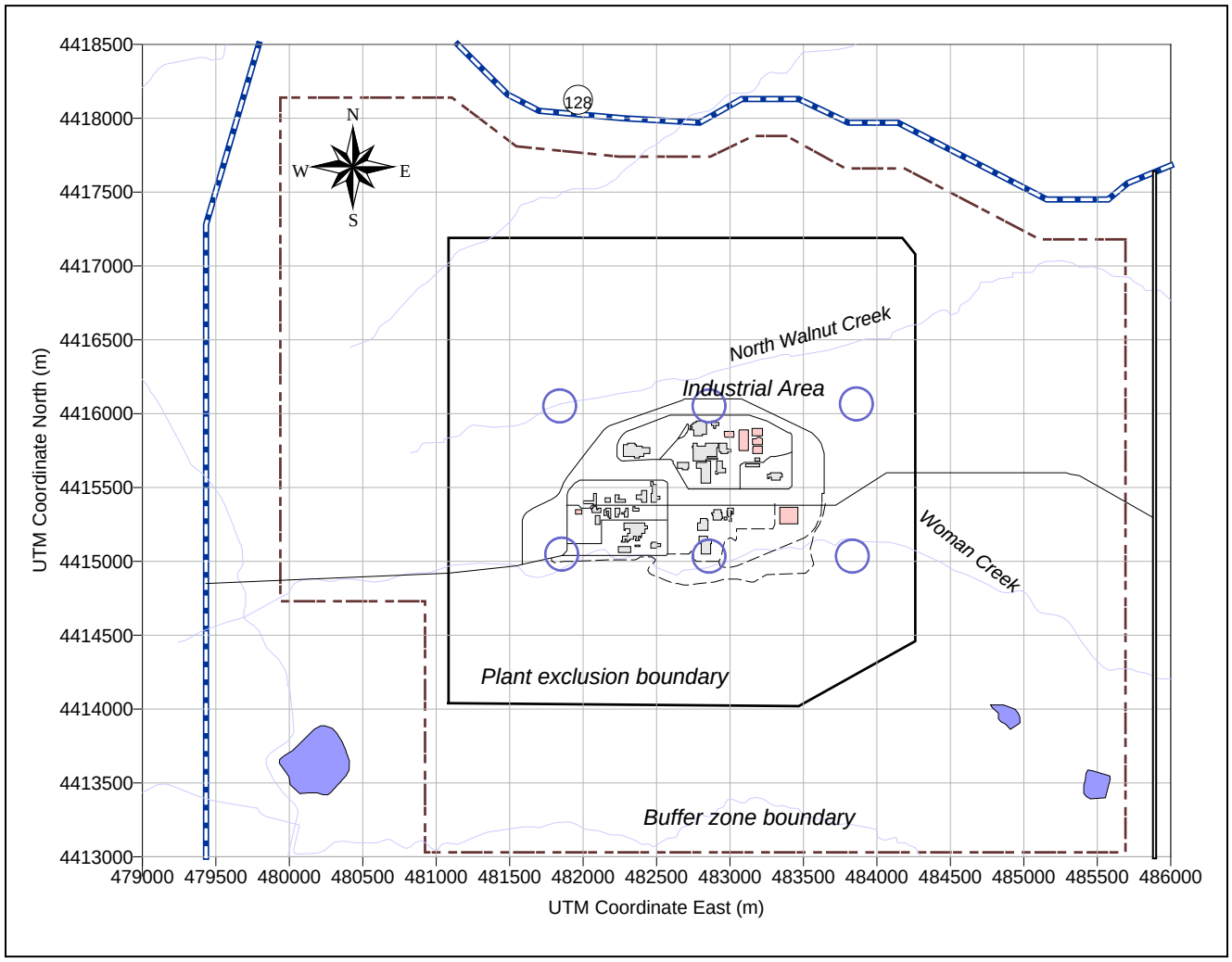

Figure A-2. Location of computational nodes (circles) used to predict average air concentrations for industrial area.

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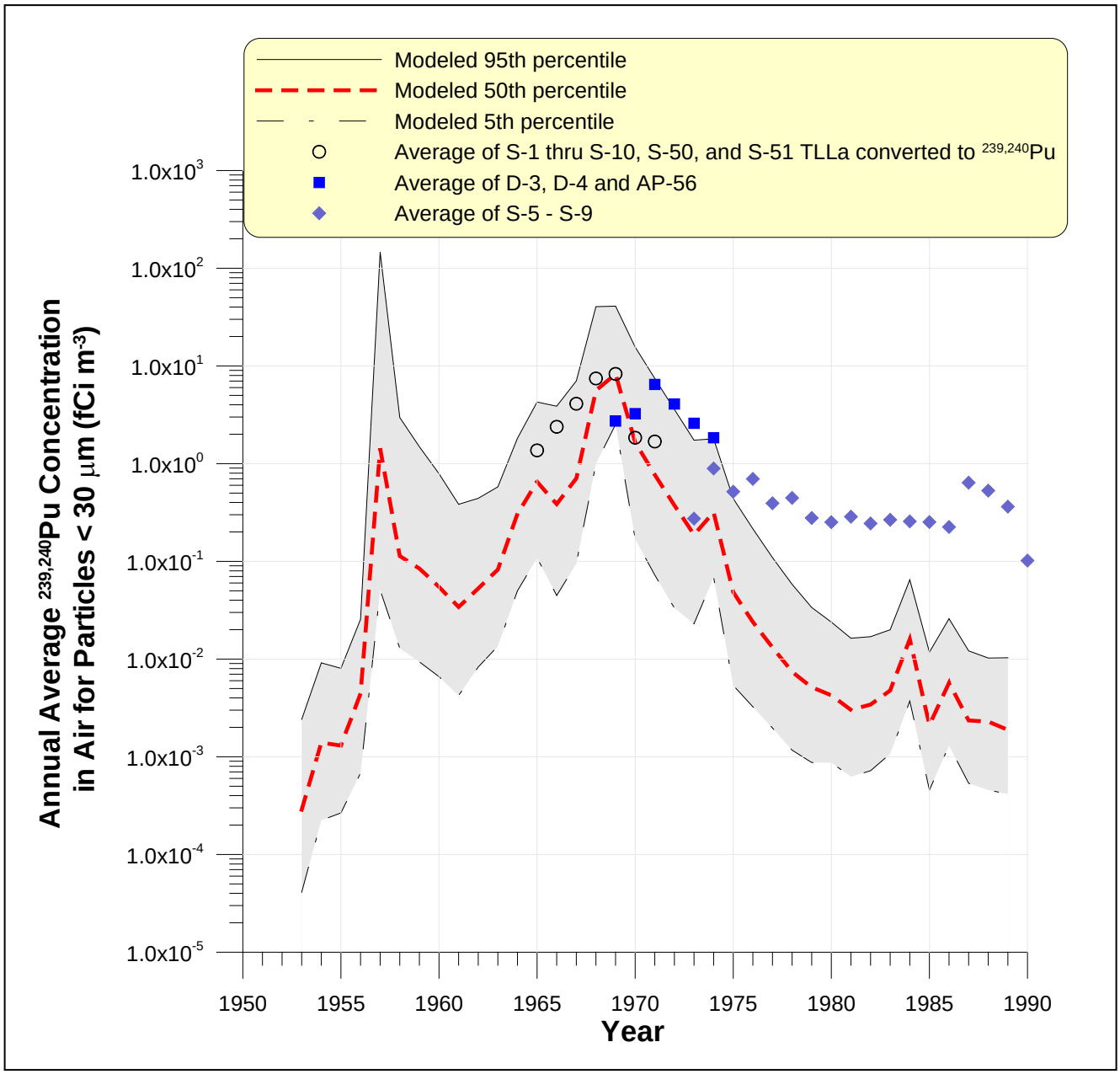

Figure A-3. Annual average $^{239/240}$Pu concentrations in air as a function of time for particles <30-μm AED in industrial area. Model-predicted concentrations represent the average of six onsite computational nodes as shown in Figure A-3 using the model described in Rood, Grogan, and Till (1999).

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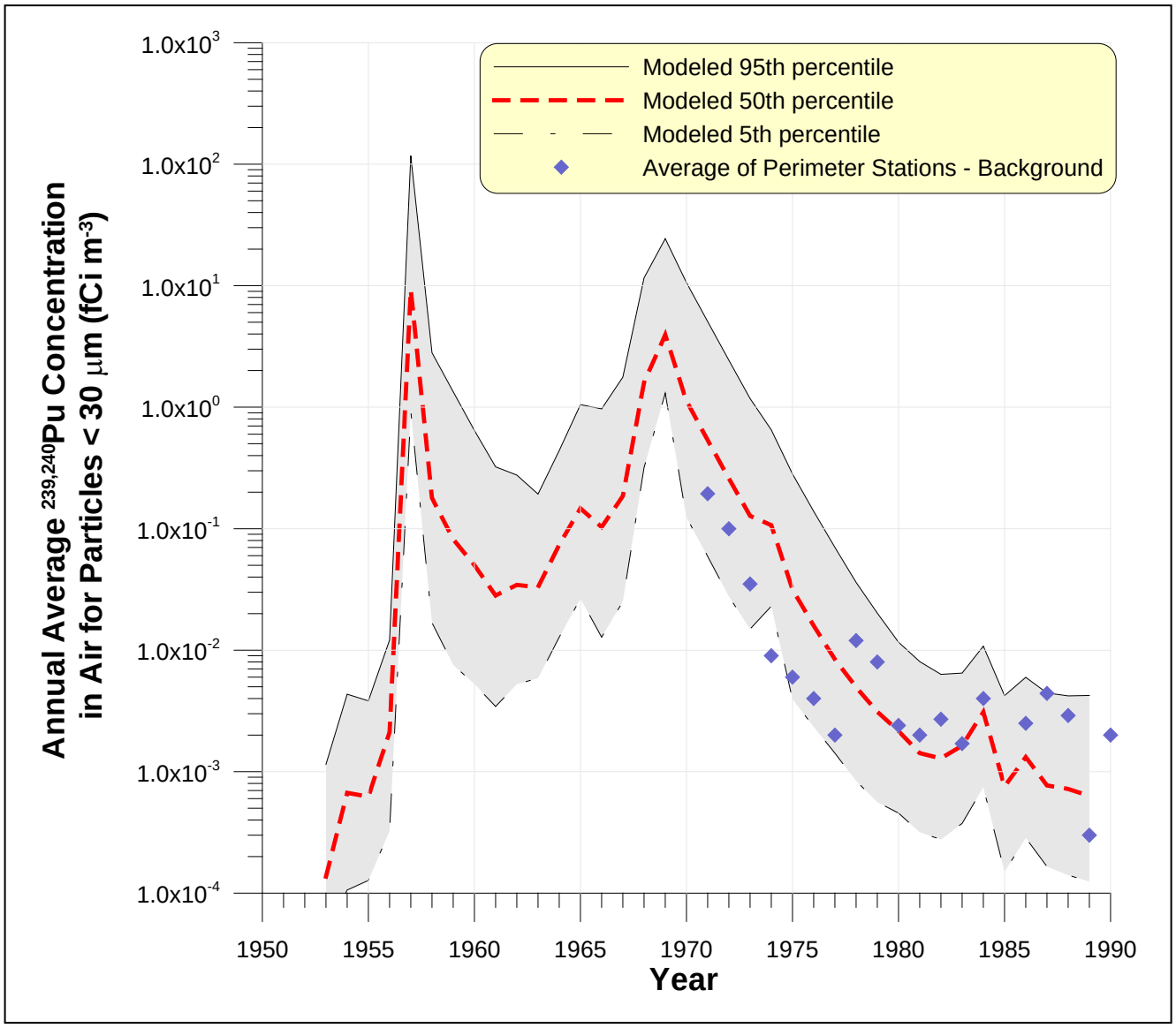

Figure A-4. Annual average $^{239/240}$Pu concentrations in air as a function of time for particles <30-μm AED in the perimeter area. Model-predicted concentrations represent the average of 27 computational nodes as shown in Figure A-7 using the model described in Rood, Grogan, and Till (1999).

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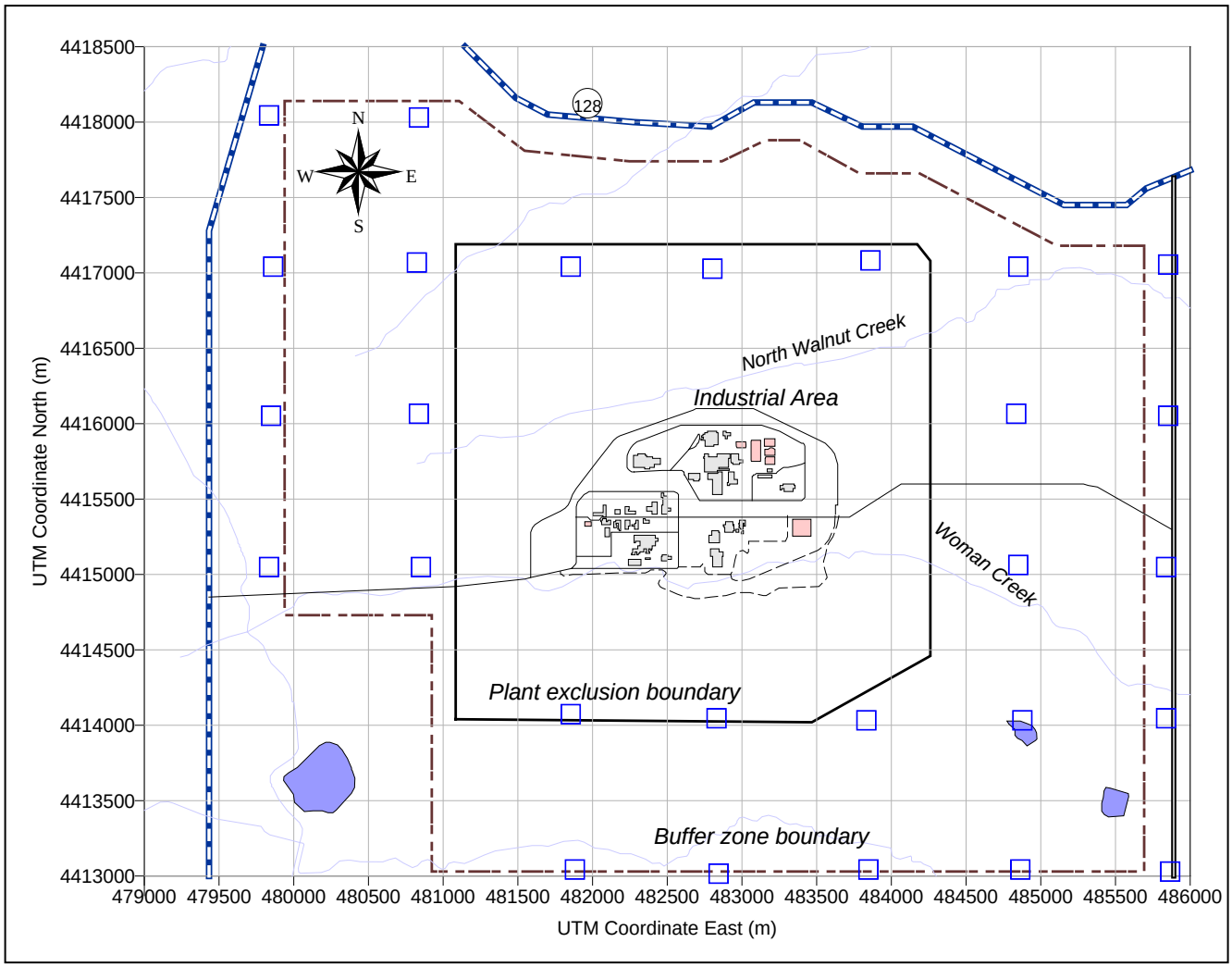

Figure A-5. Locations of computational nodes (squares) used to compute annual average perimeter concentrations shown in Figure A-6.

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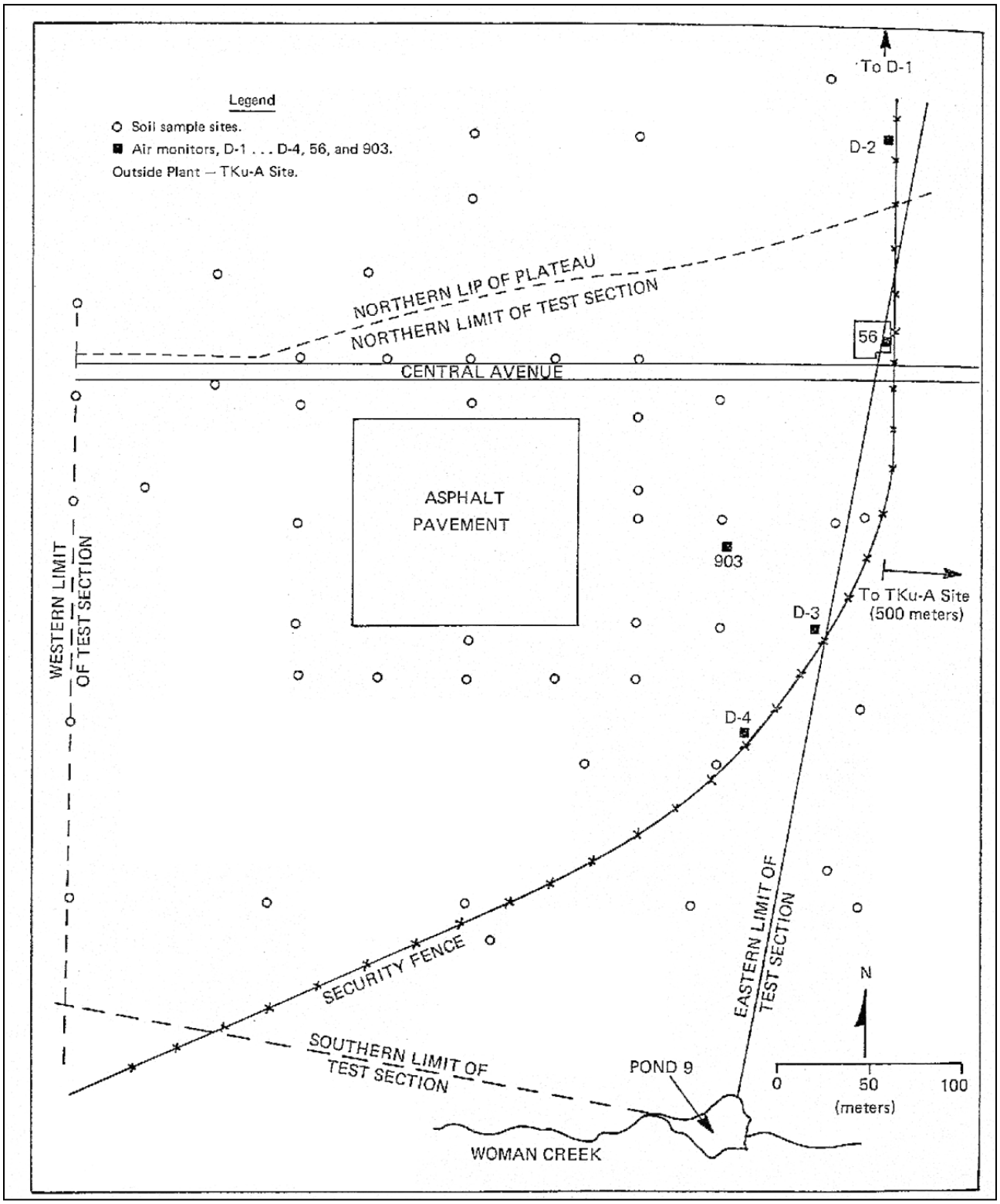

Figure A-6. Eastern part of industrial area, showing locations of CDPHE air monitoring stations D-1 to D-5 (Rope et al. 1999, Figure III-13). Sampler AP-56 is believed to be north of Central Avenue near the security fence.

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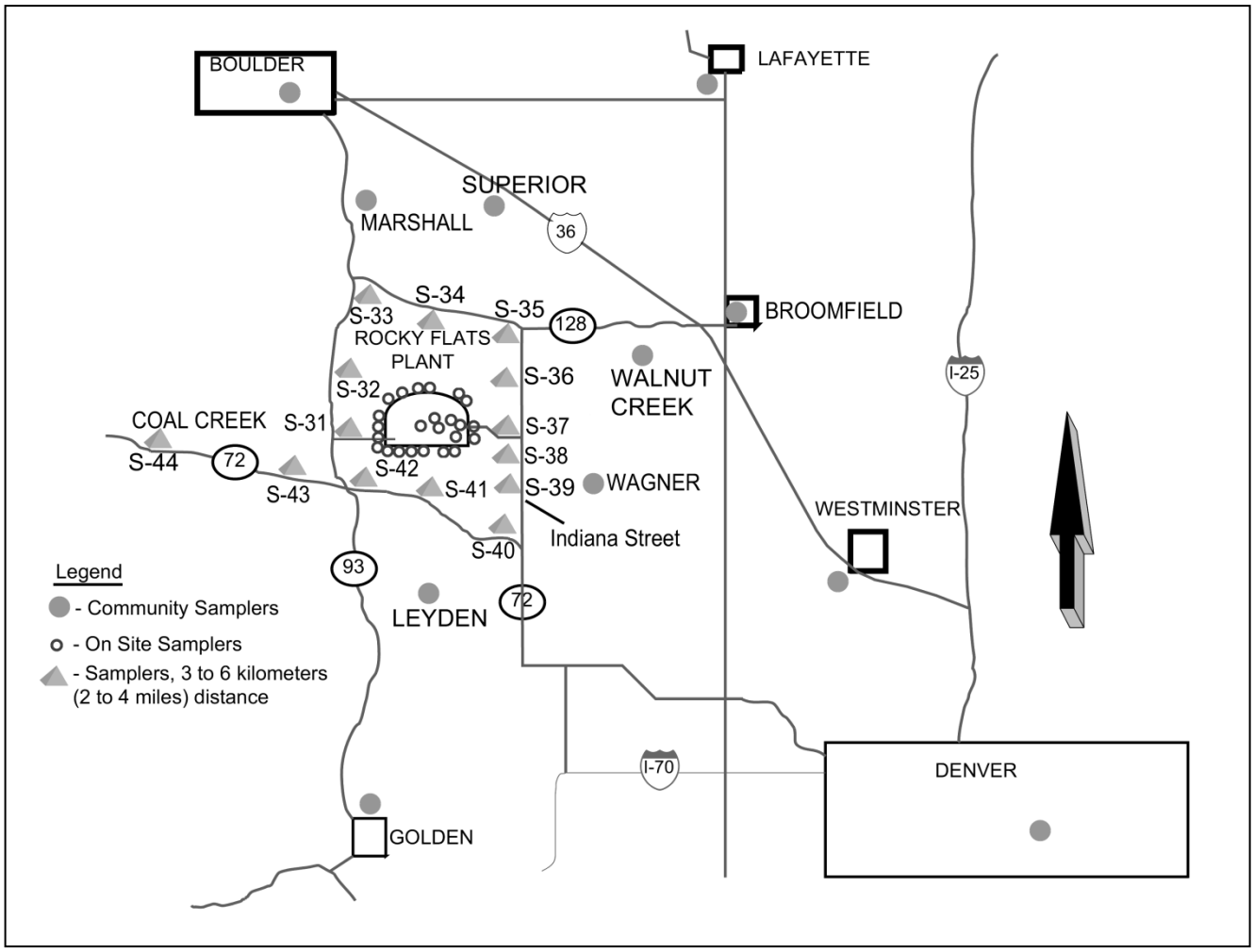

Figure A-7. Locations of offsite air samplers in 1975 (Rope et al. 1999, Figure B-4).

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Table A-1. Monthly average concentrations (fCi/m^3) of TLLα activity in onsite air samples, October 1964 to December 1971.[a]

Month	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	No data	No data
Nov 64	1	2	6	1	2	2	2	16	3	2	No data	No data
Dec 64	2	3	4	2	7	4	3	79	4	3	No data	No data
Jan 65	2	2	11	3	7	2	2	8	3	2	No data	No data
Feb 65	3	2	2	4	4	4	3	4	2	3	No data	No data
Mar 65	5	3	3	5	3	5	2	2	3	3	No data	No data
Apr 65	5	3	2	2	2	3	3	5	2	2	No data	No data
May 65	3	13	1	3	4	5	3	4	2	3	No data	No data
Jun 65	4	2	2	2	4	5	2	8	2	2	No data	No data
Jul 65	4	2	2	2	3	6	2	9	1	3	No data	No data
Aug 65	4	1	2	3	3	4	2	5	2	3	No data	No data
Sep 65	5	3	3	3	7	5	0	5	2	4	No data	No data
Oct 65	3	3	3	1	2	4	2	3	3	2	No data	No data
Nov 65	10	3	3	2	4	4	2	9	3	4	No data	No data
Dec 65	4	7	7	6	14	13	5	7	4	9	No data	No data
Jan 66	11	6	4	2	8	4	3	12	6	14	No data	No data
Feb 66	3	2	3	5	6	2	1	11	3	4	No data	No data
Mar 66	4	2	4	3	3	3	2	11	5	3	No data	No data
Apr 66	3	2	3	4	4	4	2	6	3	2	No data	No data
May 66	4	4	4	3	6	5	5	5	7	7	No data	No data
Jun 66	7	5	4	2	3	13	8	12	6	8	No data	No data
Jul 66	7	6	4	4	6	6	5	11	7	6	No data	No data
Aug 66	15	7	4	12	7	11	8	13	8	6	No data	No data
Sep 66	28	8	9	12	18	16	5	10	7	8	No data	No data
Oct 66	11	4	4	7	8	8	9	10	7	4	No data	No data
Nov 66	7	2	3	16	8	6	6	13	5	3	No data	No data
Dec 66	7	4	5	14	10	12	4	8	8	6	No data	No data
Jan 67	8	4	7	15	13	7	6	22	23	8	No data	No data
Feb 67	5	6	6	22	8	8	13	38	6	4	No data	No data
Mar 67	6	1	5	11	7	8	5	9	5	3	No data	No data
Apr 67	3	3	6	6	10	7	3	11	5	7	No data	No data
May 67	9	4	7	8	9	12	8	9	6	5	No data	No data
Jun 67	10	5	8	13	14	11	17	18	5	5	No data	No data
Jul 67	15	5	7	8	11	14	28	19	9	3	No data	No data
Aug 67	27	6	15	10	11	17	28	26	10	12	No data	No data
Sep 67	5	3	3	6	4	14	14	22	4	5	No data	No data
Oct 67	5	3	7	7	5	15	24	99	No data	7	No data	No data
Nov 67	6	5	10	6	4	9	8	49	No data	7	No data	No data
Dec 67	9	7	17	7	3	3	9	92	No data	4	No data	No data
Jan 68	8	4	12	5	6	3	8	29	No data	5	No data	No data
Feb 68	15	9	23	12	7	6	10	33	No data	11	No data	No data
Mar 68	14	18	22	8	9	17	11	116	No data	28	No data	No data
Apr 68	16	11	13	9	6	27	31	182	No data	10	No data	No data
May 68	8	9	17	10	7	21	70	155	No data	8	No data	No data
Jun 68	6	6	8	12	4	51	68	110	No data	18	No data	No data
Jul 68	5	4	7	4	4	14	14	31	No data	3	No data	No data
Aug 68	9	6	8	5	7	9	10	17	No data	4	No data	No data
Sep 68	6	6	7	5	5	4	9	50	No data	3	No data	No data
Oct 68	12	7	8	6	4	11	19	33	No data	3	7	11

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Month	S-1	S-2	S-3	S-4	S-5	S-6	S-7	S-8	S-9	S-10	S-50	S-51
Nov 68	7	4	6	3	7	9	6	168	No data	4	4	2
Dec 68	11	6	4	8	6	6	20	357	No data	3	11	3
Jan 69	10	6	9	7	10	15	127	1525	No data	3	7	5
Feb 69	6	4	8	5	5	4	23	129	No data	3	4	3
Mar 69	7	4	3	5	4	2	4	208	No data	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

a. Source: Rope et al. (1999, Table B-6), reconstructed from daily measurements.

ATTACHMENT A
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

Table A-2. Estimated annual average concentrations (fCi/m^3) of $^{239/240}$Pu in onsite air samples, 1965 to 2005.[a]

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	TLLα	0.36	1[e]	2[e]
1966	S1–S10	TLLα	0.36	2[e]	4[e]
1967	S1–S10	TLLα	0.36	4[e]	12[e]
1968	S1–S8, S10	TLLα	0.36	7[e]	38[e]
1969	S1–S10, S50–S51	TLLα	0.36	8[e]	67[e]
1970	S1–S10, S50–S51	TLLα	0.36	2[e]	3[e]
1971	S1–S10, S50–S51	TLLα	0.36	2[e]	4[e]
1972	S1–S10, S50–S51	TLLα	0.36	<2.1	3.9
1973	S1–S10, S50–S52	TLLα	0.36	<2.2	<3.9
1973	S1–S10, S50–S52	Pu-239/240	1	<1.2	<6.5
1974	S1–S10, S50–S52	Pu-239/240	1	0.6	3.2
1975	S1–S10, S22, S50–S52, Af-83, Ac-84	Pu-239/240	1	<0.2	1.0
1976	S1–S24	Pu-239/240	1	0.2	1.4
1977	S1–S24	Pu-239/240	1	0.3[f]	0.6
1978	S5–S9, S19–S21	Pu-239/240	1	0.3[f]	0.9
1979	S2, S5–S9,S15–S16, S19–S21	Pu-239/240	1	0.2[f]	0.5
1980	S5–S9, S16, S19–S21	Pu-239/240	1	0.2[f]	0.5
1981	S5–S9, S16, S19–S21	Pu-239/240	1	0.2[f]	0.5
1982	S5–S9, S16, S19–S21	Pu-239/240	1	0.3[f,g]	0.5
1983	S5–S9	Pu-239/240	1	0.2[f,g]	0.4
1984	S5–S9	Pu-239/240	1	0.3[f,g]	0.6
1985	S5–S9	Pu-239/240	1	0.2[f,g]	0.4
1986	S5–S9	Pu-239/240	1	0.3[f,g]	0.5
1987	S5–S9	Pu-239/240	1	0.7[f,g]	1.2
1988	S5–S9	Pu-239/240	1	0.5[f,g]	0.7
1989	S5–S9	Pu-239/240	1	0.4	0.5
1990	S1–S24, S88	Pu-239/240	1	0.1	0.9
1991	S1–S25	Pu-239/240	1	0.1	1.2
1992	S1–S25	Pu-239/240	1	0.1	0.6
1993	S3–S25	Pu-239/240	1	0.1	0.4
1994	S3–S25	Pu-239/240	1	0.1	0.4
1995[h]	S005–S007, S009, S101-104, S106, S107, S109, S110, S112, S116, S119, S121, S123, S202–S206, S208, S211	Pu-239/240	1	0.02	0.2
1996[h]	S007, S107	Pu-239/240	1	0.08	0.08
1997[h]	S007, S107	Pu-239/240	1	0.05	0.07
1998[h,i]	S007, S107	Pu-239/240	1	0.080	0.1
1999[j,k]	D-1,D-3	Pu-239/240	1	0.07	0.1
2000[j]	D-1,D-3	Pu-239/240	1	0.08	0.1
2001[j]	D-1,D-3	Pu-239/240	1	0.08	0.2
2002[j]	D-1,D-3	Pu-239/240	1	0.03	0.05
2003[j]	D-1,D-3	Pu-239/240	1	0.09	0.2

ATTACHMENT A
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

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)
2004[j]	D-1	Pu-239/240	1	0.03	0.03
2005[j,l]	D-1	Pu-239/240	1	0.1	0.1

a. Based on measurement data provided in Table A-1, RFP annual environmental reports, RFETS monthly and quarterly monitoring reports, and CDPHE quarterly environmental surveillance reports (Environmental Reports section of the References), unless otherwise noted.
b. Refer to Figure A-1 for samplers through 1994; after 1994, see monthly and quarterly monitoring reports for maps.
c. Based on Rope et al. (1999, pp. 107–111).
d. The maximum monthly concentration at any one sampling location.
e. Calculated from data in Table A-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. Sources: Quarterly environmental monitoring reports (EG&G 1995; RMRS 1995a, 1995b, 1996a, 1996b, 1996c, 1998b, 1998c).
i. Based on data collected January through July 1998.
j. Sources: CDPHE quarterly environmental surveillance reports (CDPHE 2000 to 2005b).
k. Based on last 3 quarters only.
l. Based on first 2 quarters only.

Table A-3. Annual average concentrations (fCi/m^3) of plutonium in air for three location groups, contractor monitoring, 1971 to 1990[a].

Year	Onsite[b]	Perimeter	Community
1971	No sampler	0.26	No sampler
1972	No sampler	0.14	No sampler
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 RFP contractor annual reports by Rope et al. (1999, Table B-10); 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.

ATTACHMENT A
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

Table A-4. Comparison of $^{239/240}Pu$ concentrations (fCi/m^3) in onsite and offsite air as measured by CDPHE.[a]

Onsite

Location	1969	1970	1971	1972	1973	1974
D-1	No data	0.29	0.31	1.44	0.74	0.44
D-2	No data	0.37	0.34	0.69	0.62	0.73
D-3	No data	6.29	10.29	4.47	2.87	3.77
D-4	No data	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)

Location	1969	1970	1971	1972	1973	1974
D-5 (SE boundary)	No data	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	No data	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	No data	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)

Location	1969	1970	1971	1972	1973	1974
APC-29 Durango	No data	No data	No data	<0.12	<0.02	<0.05
APC-42 Fort Collins	No data	No data	No data	<0.04	<0.08	0.14
APC-81 Walsenburg	No data	No data	No data	0.07	<0.06	0.38
APC-108 Rangely	No data	No data	No data	0.1	<0.08	0.13
Offsite (remote) average[c]	No data	No data	No data	**0.08**	**0.06**	**0.18**

a. Source: Rope et al. (1999, Table III-4).
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-# stations and 0.03 for D-# stations).

ATTACHMENT A
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

Table A–5. Annual average concentrations (fCi/m^3) of $^{239/240}$Pu in Denver air, 1971 to 1989.[a]

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

a. Source: Rope et al. (1999, Table B-14).
b. M = average of values measured by the Public Health Service or EPA in Denver for that year. See Rope et al. (1999, Figure III-7) and associated text for additional discussion.

ATTACHMENT A
ESTIMATION OF PLUTONIUM AIR CONCENTRATIONS (continued)

Table A-6. Corrected annual average concentrations of $^{239/240}Pu$ in air at perimeter monitoring stations (fCi/m^3).

Year	Pu-239/240 concentration[a]	Net Pu-239/240 concentration[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. Source: Table A-3.
b. The net concentration is the concentration in the second column minus the corresponding average background value reported in Table A-5.
c. Average background measurement (Table A-5) exceeded measured perimeter value.