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Health Studies on
ROCKY FLATS

PHASE I:
HISTORICAL
PUBLIC
EXPOSURES

PROJECT TASK 5

REPORT

Estimating Historical Emissions From Rocky Flats 1952-1989

ChemRisk

Colorado Department of Public Health and Environment

Prepared by ChemRisk for the Colorado Department of Public Health and Environment

PROJECT TASK 5 REPORT
ESTIMATING HISTORICAL EMISSIONS FROM ROCKY FLATS

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PROJECT BACKGROUND

The U.S. Atomic Energy Commission (AEC) announced its decision on March 23, 1951 to build the Rocky Flats Plant. The plant was built to increase the quantity and quality of the nation's nuclear arsenal and has played an important role in the U.S. nuclear weapons complex in the years that have followed. Early plant operations were for the most part kept behind a "cloak of secrecy," with the main off-site concern being centered around two fire incidents in 1957 and 1969 that received public attention, an inadvertent release of tritium to surface waters in 1973, and a waste storage practice (waste oil drum storage at the site of the 903 Pad) that resulted in the spread of contamination to nearby soil during the late sixties. After the 1969 fire, the public learned for the first time that plutonium had been released routinely and accidently from the plant. In 1984, the site was proposed to be a Superfund site, and in 1989, it was included on the National Priorities List for cleanup of environmental contamination.

Public concern came to a high point in June 1989 when approximately 100 FBI and U.S. Environmental Protection Agency (EPA) agents raided the plant seeking documentation of alleged criminal acts and mismanagement. The Department of Energy (DOE) subsequently suspended plutonium processing to review and upgrade the plant's safety systems. Following the raid, Colorado's Governor Roy Romer negotiated with Energy Secretary Admiral James Watkins to secure funding for closer scrutiny of the plant's activities by the state and for health studies to address the public's concern of potential adverse health effects.

In June 1989, an Agreement in Principle was signed by Governor Romer and Secretary Watkins that included DOE funding for increased environmental surveillance and oversight, remediation, emergency preparedness measures, accelerated cleanup in areas of imminent threat, and health studies. This report is one of the products of Phase I of the health studies known as the Rocky Flats Toxicologic Review and Dose Reconstruction Project, which is being conducted by ChemRisk under contract to the Colorado Department of Health.

The Rocky Flats Toxicologic Review and Dose Reconstruction Project

The primary purpose of this project is to reconstruct potential doses of the contaminants of concern which might have been received by off-site individuals as a result of past Rocky Flats Plant operations. Two points should be emphasized regarding the project scope. First, this project is designed to address exposures from historical operations, not to estimate doses from present and future operations or anticipate future exposure potentials. Second, this project is concerned with doses to individuals off the plant site, as opposed to occupational exposures to plant workers. Information pertaining to workplace exposures or control devices will in general only be considered if it is also relevant to prediction of off-site releases or exposures. The

period of interest for this study begins in 1953 when production related emissions began and covers the period through 1989.

The technical tasks associated with the Phase I Health Studies are listed in Figure 1. The first several tasks focus on the development of an understanding of potential health impacts of contaminants released from the Rocky Flats Plant through a comprehensive look at all the materials and their quantities used at the plant since 1952.

1. Identify Chemicals & Radionuclides Used
2. Select Materials of Concern
3. Reconstruct History of Operations
4. Identify Release Points
5. Estimate Releases
6. Select and Model Exposure Pathways
7. Characterize Land Uses and Demographics
8. Perform Dose Assessment

FIGURE 1: TASKS OF THE ROCKY FLATS TOXICOLOGIC REVIEW AND DOSE RECONSTRUCTION PROJECT

Task 1 involved identification of chemicals and radionuclides used on the Rocky Flats site. Unlike some similar dose reconstruction studies that have been undertaken for federal nuclear facilities, this project is concerned with not only radionuclide emissions, but also releases of hazardous chemicals and mixed wastes containing both radioactive and non-radioactive components. To identify materials used on the site, the ChemRisk team first reviewed radioactive source registries and inventories and chemical inventories produced by plant staff. Chemical inventories listed thousands of chemicals present in very small quantities and some chemicals used in very large quantities. Examples range from 4 milliliters of vinyl chloride kept in a laboratory refrigerator to over 400,000 pounds of nitric acid used at the plant each year. Classified and unclassified records were also reviewed for evidence of other materials used on the Rocky Flats site. The result of Task 1 was a list of over 8,000 materials used on the site (ChemRisk, 1991a).

The objective of **Task 2** was to select chemicals and radionuclides most likely to have posed an off-site human health hazard under historical routine plant operations. Radionuclides that have been included as contaminants of potential concern are those that were handled in substantial

quantity, were associated with production activities, were found in forms that were likely to be released, or were found to be present in plant effluents or in the environment.

For chemicals, a three-stage screening process was developed to narrow down the list of contaminants of potential concern. In the first stage, 629 compounds were identified for further, more refined screening based on their known toxicologic properties, Rocky Flats release histories, or reported inventory quantities. A second stage of screening was performed to roughly estimate if the quantity of a chemical on-site was sufficient to pose an off-site health hazard. Forty-six potential chemicals of concern emerged from Stage 2 Screening. In the final stage of screening, these chemicals were individually evaluated to determine the likelihood of their release, potential quantity of release based on actual storage and usage practices, likely routes of release, and known behavior in the environment.

Using both qualitative and quantitative screening criteria, and taking into account preliminary knowledge of actual storage and usage practices, 32 contaminants of potential concern were identified in Task 2 that could have been associated with off-site health impacts from normal operations of the Rocky Flats Plant (ChemRisk, 1991b). The initial list of contaminants of potential concern was subject to continuing review. As the work progressed, newly identified compounds were evaluated for possible addition to the list of contaminants of concern.

Concurrent with the identification of materials used on the Rocky Flats site, **Task 3** activities sought to document the history of operations at the facility as it might relate to off-site exposures, and **Task 4** activities sought to characterize emission points for radionuclide and chemical releases to the environment (ChemRisk, 1992a). Tasks 3 and 4 of the Rocky Flats Toxicologic Review and Dose Reconstruction Project involved extensive investigation and collection of information describing past operations of the Rocky Flats Plant. The objectives of the historical investigations were to:

- Document the basic history of the Rocky Flats facility, outlining its physical development and its historical mission,
- Document the nature of historical uses of the contaminants of potential concern identified in Task 2,
- Identify any significant historical uses of materials not evaluated as part of the Task 2 selection of contaminants of potential concern,
- Identify potential points of significant releases of materials of concern to air, surface water, or soil,

- Support work in Tasks 5 and 6 by characterizing the potential for significant uncontrolled radionuclide emissions from normal operations in the past that may have gone undetected by effluent monitoring systems, and,

- Identify any accidents, incidents, or waste disposal practices that resulted in contaminant releases with significant potential for off-site transport, also in support of Tasks 5 and 6.

Tasks 3 and 4 investigations consisted of an extensive campaign of document reviews and personnel interviews targeting active and retired Rocky Flats employees, local citizens, and other interested parties. The major outcomes of the investigations are an understanding of the historical uses of the contaminants of potential concern, identification of accidents that warrant detailed evaluation, and documentation of the nature of associated emission points.

The objective of Task 5 was to develop historical release estimates for the routine releases and events selected for detail study. This task was divided into the following categories:

- Routine radioactive airborne emissions,

- Routine nonradioactive airborne emissions,

- Routine surfacewater emissions, and

- Nonroutine contaminant releases.

Historical investigations carried out in Tasks 3, 4 and 5 resulted in the identification of the contaminants listed in Table 1 as the subject of quantitative evaluation.

The historical airborne radioactive effluent monitoring program at the plant was reviewed and evaluated to establish the utility of the data for dose reconstruction. Uncertainties in the monitoring data associated with the sampling and analytic practices at the plant were carefully characterized. The review indicated that the effluent monitoring data reported by the plant provided a good basis for estimating airborne releases from the facility provided the identified uncertainties were incorporated in the estimates. One notable exception was the plant's data for uranium emissions prior to 1961, which are underreported in plant summary documents. The uranium emissions for this period were recalculated using raw data from plant log books. The effluent monitoring data were used as the basis for establishing quantitative annual estimates of routine releases of the radioactive materials of concern.

TABLE 1: CONTAMINANTS IDENTIFIED FOR QUANTITATIVE EVALUATION

SOLVENTS	METALS	OTHERS
Carbon Tetrachloride	Americium-241	Tritium
Chloroform	Beryllium	
Methylene Chloride	Plutonium-239/240	
Tetrachloroethylene	Uranium-234/235 (enriched)	
1,1,1-Trichloroethane	Uranium-238 (depleted)	
Trichloroethylene		

Monitoring data for routine airborne emissions of nonradioactive materials are available only for beryllium, and these data served as the basis for the release estimates of this metal. Routine monitoring for organic solvents of concern was not performed by the plant. Estimates of the plausible ranges of historical emissions for these materials were developed using various types of documentation (e.g., Air Pollution Emissions Notifications, special studies conducted by the plant, and inventory quantities) and information obtained from personnel interviews.

Review of information regarding routine contaminant releases to surface water from the plant indicated relatively limited availability of data to directly quantify the releases. Those data that are available were used to examine whether plant releases measurably increased the radioactivity present in water from potentially impacted reservoirs and drinking water. While the data review suggested that it is plausible that plant-related releases may, during some periods of time, have measurably increased gross alpha radioactivity in the waters of the receiving reservoirs, the resulting measured levels were similar to levels found in other, unaffected reservoirs in the area. For tritium, some measured increases were clearly attributable to Rocky Flats.

Information and data associated with releases of contaminants from the plant for major nonroutine release events (1957 and 1969 fires and 903 Pad) were identified in the Task 5 report. The data and information on these accidental events are very limited. As a result, the analysis of these events requires the use of a number of estimates that introduce uncertainties that are accounted for in the final results. The product of Task 5 efforts is historical contaminant release estimates for contaminants routinely released by the plant and information and data regarding accidental releases requiring further analyses in Task 6 to predict historical contaminant concentrations in environmental media.

Project **Task 6** began the process of evaluating how plant releases traveled off-site and could have resulted in exposure of the public by predicting the concentrations of the contaminants in environmental media such as air, soil and foodstuffs (ChemRisk, 1992c). Based on the nature of contaminant releases, physical properties of the contaminants, local hydrogeology and land-use information, the following exposure pathways were identified to be important in Task 6:

- Inhalation of airborne contaminants due to direct release or soil resuspension,
- Incidental ingestion of contaminated soil,
- Consumption of contaminated vegetables, milk and beef, and
- Ingestion of contaminated drinking water.

An exposure model capable of evaluating these exposure pathways is also developed in Task 6.

One of the primary objectives of Task 6 was to predict the concentrations of contaminants in the air in areas around the plant site so that the amount of contaminant that could have been inhaled by people, deposited on the ground that people could come in contact with, and taken up by vegetation or grazing animals that could be eaten by people could be estimated. Air concentrations were estimated for routine releases of contaminants by using computer models, information on the meteorological conditions at the site (such as wind speed and direction) and the conditions and size of the contaminant release (such as height of the stack, temperature of the exhaust air, and amount of contaminant).

A somewhat different technical approach was required to predict contaminant concentrations in off-site areas that resulted from accidental releases. To evaluate these accidental releases, since there were no or incomplete direct measurements of the releases, information was pieced together from the conditions that were reported during the accident and from monitoring data in the form of air, soil or vegetation samples taken during or shortly after the event. Computer air dispersion models were used to determine, under the estimated conditions of the accidental release, the size of the release that would have been necessary to produce the contamination that was measured at the few locations where air, soil or vegetation samples were taken. The model could then be used to predict the likely concentrations of contaminants at other locations where the public could have been exposed. In some cases, model predictions were compared to environmental sampling data that were not used in the initial estimate of the size of the release. These comparisons test the accuracy of the models in predicting environmental concentrations and add to the confidence that can be placed in the modeling.

The results of Task 6 provided the basis for making estimates of the environmental concentrations of contaminants released from the plant from routine operations, accidents, and resuspension to the air from the soil and the identification of the pathways that these contaminants were most likely to have traveled in reaching the public. These are critical pieces of information needed to calculate the doses of contaminants that the public in the vicinity of Rocky Flats would have received as a result of past plant activities. This information was used in Task 8 to calculate doses to the public.

Areas within several miles of Rocky Flats have changed over time in terms of land use and development since the plant first began operations in 1953. The objective of Project **Task 7** was to identify land uses and populations near the Rocky Flats plant during the period of operations from 1953 to 1989 (ChemRisk, 1992d). The task emphasized identification of the locations of nearby residents that would be most highly exposed and the approximate sizes of populations living near the plant. The use of the lands and waters surrounding the plant were also examined, because this can influence the pathways through which contaminants can migrate and ultimately reach people. Typically, land uses of interest include the raising of crops for human consumption, grazing land and hay produced as feed for cattle consumed by people, or the presence of dairies and drinking water or irrigation reservoirs.

A relatively limited amount of detail about land uses and populations was collected for this first phase of the health studies through personal interviews with long-term landowners and review of census data, historical topographical maps produced by the United States Geological Survey, aerial photographs, deed books and county assessor files to establish land ownership and land use, and county and local government records. A number of other types of federal, state and local agency records were also explored for useful information.

The Task 7 work provides preliminary population information that would be required for the purposes of an epidemiological study, but additional information would ultimately be needed for such studies. The work does not provide detail agricultural production information, based on the contaminants released by the plant, food-related pathways are known to have made only a minor contribution to the total exposure of the public to contaminants released by the plant.

The last technical task is Project **Task 8**. This task combines the information produced in the preceding tasks on the amount of contaminants that were either estimated to be present or measured in the environment from plant releases with the exposure model developed in Task 6 to estimate radiation and chemical doses potentially received by the public. Dose estimates and the uncertainty in these estimates are provided for each of the contaminants listed in Table 1. While the endpoint of the Phase I studies are these dose estimates, and one of the purposes of Phase II is to thoroughly examine what these doses mean in terms of health risk, Task 8 provides some initial interpretations of the doses in terms of health risk in order to provide some perspective on the meaning of the results of Phase I.

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EXECUTIVE SUMMARY

The purpose of Task 5 of the Toxicologic Review and Dose Reconstruction project is to develop historical release estimates for those contaminants and events selected for detailed study in previous project tasks. This report discusses the development of estimates for the following major categories of releases:

- Routine radioactive airborne emissions,
- Routine nonradioactive airborne emissions,
- Routine surface-water-borne emissions, and
- Nonroutine contaminant releases.

The historical airborne radioactive effluent monitoring program at the plant is reviewed and evaluated to establish the utility of the data for dose reconstruction. The data review indicates that the effluent monitoring data reported by the plant provide a good basis for estimating airborne releases from the facility with the notable exception of uranium emissions prior to 1961 which are underreported in plant summary documents. The effluent monitoring data is used as the basis for establishing quantitative annual estimates of routine releases of the radioactive materials of concern. Estimates of uncertainty in the release estimates are based on the review of program practices.

Quantitative estimates of airborne release are derived for each of the radioactive materials of concern, with the exception of Th-232, using the effluent monitoring program data. Th-232 has not been specifically monitored and only saw limited use at the plant and is not believed to have been associated with significant emissions historically.

The nature of the data generated by the plant's historical airborne effluent monitoring program and the similarity in the dose factors for the radioisotopes of concern argue for consideration of the use of composite dose factors and emission estimates that would combine emissions for a number of individual isotopes, as opposed to evaluating all isotopes individually.

Monitoring program data for routine airborne emissions of nonradioactive materials are available only for beryllium, and these data serve as the basis for the release estimates of this metal. Routine monitoring for the organic solvents of concern was not performed by the plant. Estimates of the plausible ranges of historical emissions for these materials were developed using various types of documentation and information obtained from personnel interviews. In many cases, these emission estimates are based on very limited information. However, the identified

range of emissions are believed to bound the actual plausible emissions from the plant for these materials.

Review of information regarding surface-water-borne contaminant releases from the plant indicate relatively limited availability of data to directly quantify the release of materials of concern. Those data that are available are used to examine whether plant releases measurably increased the radioactivity present in potentially impacted reservoirs and drinking water. While the review of the data suggests that it was plausible that plant related releases may, during some periods of time, have measurably increased radioactivity in the waters of the receiving reservoirs, the resulting measured levels of radioactivity were similar to levels found in other unaffected reservoirs in the area.

A review of information and data associated with nonroutine releases of contaminants from the plant provides the basis for further modeling of major release events (1957 fire and 903 Pad), and for evaluation of the relative magnitude of lesser events in comparison to routine emissions. The information presented in this report relative to the major events will be employed in contaminant transport modeling efforts in Task 6 to finalize an emission estimate and to provide the basis for estimating off-site exposures from these events.

1.0 INTRODUCTION

Efforts on Tasks 1 through 4 of the Rocky Flats Toxicologic Review and Dose Reconstruction Project have resulted in the identification of contaminants for reconstruction of releases from historical operations of the Rocky Flats Plant. The contaminants that are the subject of further quantitative evaluation with regard to historical emissions are listed in Table 1-1. The list includes six organic solvents, one nonradioactive metal, four radioactive metallic elements and their isotopes, and tritium, the radioactive form of hydrogen.

TABLE 1-1: MATERIALS FOR SOURCE TERM DEVELOPMENT AS SELECTED IN TASKS 3 AND 4

SOLVENTS	METALS	OTHERS
Carbon Tetrachloride	Americium-241	Tritium
Chloroform	Beryllium	
Methylene Chloride	Plutonium-238,239,240,241,242	
Tetrachloroethylene	Thorium-232	
1,1,1-Trichloroethane	Uranium-233, 234, 235, 238	
Trichloroethylene		

A number of techniques can be employed to quantify releases from operations or facilities. The most effective method typically involves the use of release (effluent) measurement data. This technique uses information from samples collected at release points during process operations. The measured contaminant concentration in combination with established discharge flow rates or volumes is required to establish a source term. There are a variety of questions about the completeness of the sampling record and the adequacy of the methods that must be addressed prior to the use of release measurement data. However, the use of monitoring data in establishing source terms requires the fewest number of assumptions and estimates, thereby reducing the degree of uncertainty in the release quantity. Unfortunately, the monitoring record is often incomplete or absent for the contaminants of interest and other approaches must be used. Alternative approaches can be grouped into three broad categories:

- Material mass balance calculations,
- Analogy to similar, well-characterized processes with established emission factors, and
- Process measurements/engineering calculations.

The mass balance calculation approach involves detailed accounting for each material of interest at a facility. Information on contaminant use such as amount purchased, amount used, amount of material that is incorporated in the product, amount disposed of, and the amount recycled during a defined period of operation must be established so that the amount lost from a process can be calculated. In processes where emissions are passed through contaminant control devices such as some type of filtration, further estimates of the efficiency of the control devices are necessary to finally calculate emissions to the environment. The mass balance calculation approach requires a considerable amount of operations information that is often unavailable. Where the available operations information is limited, the mass balance calculation approach may be effective only in providing relatively crude estimates of environmental release.

In the absence of effluent monitoring data for a specific process of interest, another approach that may be employed involves making analogies to other similar processes that have been well studied at other facilities. Published emission factors are available to estimate emissions of numerous materials from a wide variety of processes and operations. These emission factors may be based on emission point sampling, product analysis, engineering estimation, mass balance analysis, or a combination of these methods. Published emission factors are commonly given in units of contaminant emitted per unit of product throughput or some other process variable. Emission factors are most widely available for commonly used industrial processes, and their availability for many of the unique processes associated with a nuclear weapons plant is rather limited. As with any attempt to generalize complex or variable operations, emission estimates developed using this approach are subject to considerable error.

The third option identified for developing emission estimates involves the use of engineering calculations that may also use information from process measurements. Emission estimation using engineering calculations involves the use of standard principles of chemistry and physics in conjunction with information about process equipment design and operation to predict plausible rates of release of a particular contaminant from a given process. Again, emission estimates based on such calculations are subject to considerable error.

Given the approaches available to quantify contaminant releases, the effluent measurement approach is generally subject to the least error and uncertainty. The error and uncertainty introduced by any inadequacies in the sampling and analytic methods employed in obtaining measurement data are often far smaller than those associated with alternative methods. Therefore, carefully evaluated effluent measurement data have been used whenever possible in reconstructing emission estimates for the contaminants at the Rocky Flats Plant in this report. The extent of the effluent monitoring record at Rocky Flats varies greatly. The record is quite extensive for radioactive materials. The airborne release record for radionuclides is the most complete, while a far more limited record is available for waterborne radioactive releases. On the other hand, the chemical release effluent monitoring record is extremely limited for airborne effluents, and virtually nonexistent for the waterborne releases. To effectively review, evaluate, and present these records and emission estimates, this report has been organized into separate sections addressing airborne releases for both radioactive and nonradioactive materials and waterborne releases of the same materials.

As a general rule, we have presented release estimates to only two significant figures. However, data from original records may also be presented as they appeared in the source documents, which often reported more significant figures.

1.1 Routine Radioactive Airborne Emissions

A significant portion of the Task 5 investigation efforts involved the collection and review of records that characterize historical radioactive effluents from the Rocky Flats facility. The plant operated an extensive airborne effluent monitoring program that generated data that is directly relevant to the development of emission estimates for use in dose reconstruction. This report describes the characteristics of the plant's airborne effluents and provides a review of the historical sampling and analytic practices that were employed to measure these effluents. This review includes the evaluation of sampling system design, analytic method and data treatment practices affecting the accuracy of the reported effluent data. Emission estimates based on the sampling data generated from this monitoring program are presented for use in dose reconstruction, and the uncertainties associated with these estimates are also quantified.

This report also describes methods for estimating the isotopic composition of effluents for those periods during which the plant's sampling and analytic programs did not produce isotopic-specific release quantities. Estimates of potential releases of thorium, which was identified as a material of concern but which saw limited use at the facility, are also presented. Finally, based on the interest expressed by the public as reflected in comments received on previous project reports, a discussion is also included on the possible airborne releases associated with the performance of criticality experiments at the plant.

1.2 Routine Nonradioactive Airborne Emissions

In contrast to the airborne radioactive emissions, the nonradioactive emissions were not subject to routine monitoring, with the exception of beryllium. The beryllium monitoring program review that was performed is described in this report, and the emission data are summarized for use in dose reconstruction. Release estimates for the remaining non-radioactive materials, all organic solvents, are provided in terms of a plausible range of airborne release for various historical periods based on plant documents and other sources of data. The types of documentation and information used to develop these release estimates are described and summarized for each of the organic solvents. The estimated ranges of these releases will be used to reconstruct estimates of the potential off-site doses the public may have received of these organic solvents.

1.3 Uncertainty in Airborne Emission Estimates

Uncertainties in emission estimates can arise from a number of sources, many of which are described in early sections of this report. In Section 4, the methods that were used to quantify the uncertainties in the emission estimates of radioactive and nonradioactive materials that were based on airborne effluent monitoring data are described.

1.4 Routine Surface-Water-Borne Emissions

The plant operated a very limited monitoring program of waterborne effluents. The data that are available from these monitoring programs are not adequate to develop complete historical release estimates for the materials of concern, nor is sufficient information available to estimate releases using other estimating techniques. However, a considerable amount of data have been collected by the plant and the Colorado Department of Health relative to the presence of radioactive materials in reservoirs and drinking water in the vicinity of the plant. The various types of surface water data that are available are presented and evaluated for the purpose of determining whether the reservoirs or drinking waters that have historically received plant effluents and runoff have demonstrated measurably higher amounts of radioactivity than other waters in the area unaffected by the plant. In addition, the limited plant release data that are available are evaluated for the purposes of determining whether recorded releases were associated with increases in measured radioactivity in the receiving reservoirs.

1.5 Nonroutine Contaminant Releases

The final sections of this report provide a discussion of contaminant releases associated with accidents or incidents that were identified as a result of Task 3 and Task 4 efforts. The major release events, the 1957 fire and the events associated with the 903 Pad, require the application of extensive contaminant transport modeling to develop source terms for these events. This report describes the approach that is being employed to develop the source terms, and provides release estimates that have been reported by others for these major events. However, the release estimates that will be used for the purposes of dose reconstruction for these events will be finalized in the Task 6 report, which will address contaminant transport issues.

Other smaller accidental releases are described, and release estimates, which have primarily been developed by others, are presented to put these releases into perspective relative to other releases associated with routine operations and the major accidents.

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2.0 ROUTINE AIRBORNE EMISSIONS OF RADIOACTIVE MATERIALS

The Rocky Flats Plant has engaged in manufacturing efforts related to the production and recycling of components for nuclear weapons since beginning operation in 1952. These efforts include various chemical processing, metalworking and machining, and assembly operations that result in the release of radioactive materials via normal ventilation exhaust from plant buildings. Although the ventilation exhaust systems have included multistage Chemical Warfare Service and, in more recent years, high efficiency particulate air (HEPA) filters to remove the bulk of the radioactive material, some material was continually released to the environment even when these filtering systems were working as intended. The integrity and efficiency of the filtration systems has been the subject of study and debate for many years, and many plant operational documents (e.g., Hornbacher, 1975-1982; and Dow, 1972) identify various problems associated with the operation of these filtration systems.

Previous project investigations (ChemRisk 1991a, 1991b, 1992) identified five elements with various radioactive isotopes as potentially being associated with routine releases from the Rocky Flats Plant. The radioactive elements and their isotopes that have been the subject of detailed investigation for the purposes of quantifying historical releases at the plant are as follows:

- Americium-241
- Plutonium-238, plutonium-239, plutonium-240, plutonium-241, and plutonium-242
- Thorium-232
- Tritium (H-3)
- Uranium-233, uranium-234, uranium-235, and uranium-238

Source term development methods for this study have relied heavily on the use of effluent monitoring data. While other emissions estimating methods that do not rely on the effluent monitoring data (e.g., mass balance and engineering calculations) could be employed for some limited emission reconstructions for some specific operations based on available inventory or engineering information, such approaches would not be successful in estimating facility-wide emissions since the needed historical information for all processes is not available to perform the necessary calculations. Any mass balance or engineering calculations that were performed would be subject to significant uncertainties as a result of the need to estimate a variety of process losses, including losses in ventilation ducts and the need to address the historical efficiency of the HEPA filtration systems. Project investigations have therefore focused on the

evaluation of the effluent monitoring data for use in estimating routine radionuclide emissions from the facility.

One of the primary advantages of the monitoring record is that it not only records releases resulting from normal process operations, but it also reflects releases resulting from upset conditions within a building where the monitoring system continued to operate. In addition, a monitoring system measures contaminants in the airstream after, or downstream of, the HEPA filtration systems, eliminating the need to address filter system efficiency in quantifying emissions.

The monitoring record begins in 1953 and documents the fact that some form of airborne radioactive effluent monitoring was conducted historically whenever processing activities involving radioactive substances were initiated in one of the plant buildings. However, the sampling and analytic methods employed and the type of record vary over time.

The review of the airborne effluent monitoring data record at the plant for the purposes of dose reconstruction included the following activities to identify biases in the sampling and analytic programs that needed to be considered as sources of error or uncertainty:

- Characterization of the basic properties of the radionuclides released from Rocky Flats influencing effluent sampling and environmental transport (i.e., particle size, tritium chemical form, and plutonium and uranium compound solubility),

- Characterization of the radiological effluent sampling and analytic systems and practices that have historically been in place at the plant and the appropriateness of the methods, and

- Characterization of the available monitoring records and the accuracy of data reduction and reporting practices based on the review of raw data and summary-level data reporting documents.

The extensive volume of effluent monitoring data collected by the plant represents one of the best sources of information for quantifying airborne radioactive emissions of the primary production-related radionuclides: plutonium and uranium. Effluent monitoring for tritium covers a somewhat limited time period, but still offers the best source of information for quantifying airborne tritium effluents from the plant. Since americium was specifically quantified in effluent samples to only a limited extent and thorium was not routinely quantified, other emissions estimating approaches must be used to develop source term estimates for these materials.

An issue that was also considered in evaluating routine releases from the facility is whether a significant potential existed for the release of contaminants through unmonitored openings in the buildings or process equipment. Such releases are frequently termed fugitive emissions. An evaluation of the potential for fugitive emissions at Rocky Flats is relevant for the radioactive contaminants and beryllium that were routinely monitored and controlled in plant exhausts. However, it has been assumed that the solvents, which were not routinely monitored, may have left the facility through plant exhausts and in fugitive emissions from various storage and process equipment.

The evaluation of the potential for fugitive emissions to contribute to the routine release of monitored materials began with a review of the general design of the ventilation systems in the production buildings. The review of records and building design information indicates that production buildings at Rocky Flats have historically been designed and were routinely operated at a negative pressure relative to the outside atmosphere, that is, more air is exhausted than is directly supplied as makeup air. Such negative pressure designs are intended to result in air flow from the outside, through different areas or zones of the building to the areas of highest potential contamination, which are typically glove-box areas. The air flow in the building is designed to flow from the outside to what is termed building zone III (typically a corridor), and from there to the process areas (zone II), and finally to the glove-boxes (should there be a leak in the glove-box) which are zone I. All of the air pulled through the process buildings and the glove-boxes is subject to HEPA filtration and monitoring before being exhausted to the atmosphere. As a result, fugitive emissions of the controlled and monitored materials would only be likely to occur during ventilation system failures or major upsets of normal operations.

Historical investigations focused on the identification of such upset conditions associated with accidents and incidents that could result in the unmonitored release of contaminants, and these are discussed in the report of project tasks 3 and 4 (ChemRisk, 1992). Accidents and incidents believed to be associated with off-site releases of contaminants are addressed further in this report and in the task 6 report (ChemRisk, 1993).

The following sections discuss the relevant characteristics of the plant effluents and the sampling and analytic systems and practices employed by the plant to measure these effluents. This is followed by a description of the critical review and data reconstructions that were performed to evaluate the quality of the effluent data summaries published by the plant for plutonium and uranium and the uncertainties associated with these data. These discussions are then followed by individual sections covering issues relevant to source term development for specific alpha-emitting radionuclides, tritium, and thorium, and criticality experiments.

2.1 Characteristics of Rocky Flats Airborne Radioactive Effluents

The mission of the Rocky Flats Plant during the period of interest to this study was production of weapons components by various operations such as chemical separation and processing, machining, and metalworking. Five radioactive elements of concern are being addressed in this evaluation: americium, plutonium, thorium, tritium, and uranium. As described in detail in the Tasks 3 and 4 report (ChemRisk, 1992), the principal forms of radioactive materials associated with Rocky Flats processes and products were enriched uranium, depleted uranium, and weapons-grade plutonium. As such, the routine airborne effluents from the Rocky Flats Plant contained primarily uranium and plutonium particulates entrained in building air that pass through the control or filtering devices and are discharged to the environment.

Americium, tritium, and thorium have been present at Rocky Flats, albeit in quantities much smaller than those associated with routine production of components from plutonium and uranium metals. Americium has been present as a decay product of weapons-grade plutonium and is normally present in plant effluents as a particulate. Thorium has been used to a very limited extent by the plant for production of parts and in several minor applications supporting production of parts from uranium and plutonium. Thorium is also normally present in plant effluents as a particulate.

Tritium is the only radioactive element selected for detailed study that is likely to be present initially in the form of a gas or vapor as opposed to a solid in the effluent airstream. Tritium has been present at the plant as a contaminant of components returned to the site, has been used in standards and in special order work, and has been associated with disassembly of certain weapons components.

Experiments that have been conducted at the Rocky Flats Critical Mass Laboratory since 1965 have also generated gaseous fission products (ChemRisk, 1992). These relatively low power, short duration criticality tests were conducted with uranium and plutonium, primarily in the furtherance of criticality safety in production processes.

A key issue that must be addressed in order to accurately sample and also to model emissions from a facility is what are the physical characteristics of the contaminant, for example, is it in a gas or vapor phase, or is it a solid particulate? Gaseous or vaporous emissions, with a few exceptions, will tend to readily mix with, remain in, and be transported by the airstream. Solid or particulate emissions will be carried in an airstream when released; however, they will also tend to drop out of the atmosphere or be deposited. The rate at which a particle is deposited from the atmosphere and a number of sampling efficiency issues are directly related to the size and density of the particle. Therefore, a key physical characteristic of interest in accurate sampling or modeling all of the radioactive contaminants of concern, with the exception of

tritium, is particle size. Information on the sizes and densities of particulates in Rocky Flats effluent airsteams are summarized in this section.

The chemical form of tritium emitted from an operation is also an important factor in sampling associated effluents. Airborne water vapor samplers most commonly used for tritium sampling will collect tritium oxide (tritiated water vapor, HTO) but will not effectively capture elemental tritium gas (HT or T_2).

The solubility of the chemical forms in which plutonium and uranium have been released from the Rocky Flats Plant is also an important consideration. The ability of a particular compound to dissolve in body fluids and enter the bloodstream from the lungs or the digestive tract is an important factor in determining the nature of the retention of the compound and the magnitude of the resultant dose.

Airborne Effluent Particle Sizes

Historical documentation shows that airborne effluents from Rocky Flats processes involving radioactive materials were always subject to some type of filtration prior to release to the environment. Building air is passed through multiple stages of HEPA filtration prior to exhaust. As a result, the vast majority of particulates remaining in the effluent are typically extremely small in size.

A number of studies have been performed at Rocky Flats to characterize plutonium particle size in effluent air. Identified studies are summarized in Appendix A. These studies all indicate that radioactive particulates routinely released from the facility were predominately composed of extremely small, submicron-sized particles. Because no particle size information for beryllium or uranium was located, it was assumed that the results from the plutonium particle size studies described in Appendix A were representative of uranium and beryllium particle sizes from Rocky Flats HEPA-filtered effluents.

Tritium Chemical Forms

Tritium in the atmosphere exists in three chemical forms: oxide (HTO), elemental (HT or T_2), and organic (e.g., CH_3T). Special sampling projects designed to characterize the chemical forms of tritium present in Rocky Flats airborne effluents have been conducted at various times since 1978 (Hornbacher, 1975-1982). In a 1978 study, tritium was collected from Building 771 main exhaust over a 92-day period using a sampler based on the Östlund technique of oxidizing HT to HTO (Hurley, 1979). The study indicated that:

- During periods when tritium-containing oxide material was being processed and the plutonium recovery incinerator was in operation (25 days over the 92 days studied), the data indicated an average HTO to HT ratio of 11.89 $\pm$ 0.37.

- During nonoperating periods, an average HTO to HT ratio of 1.95 $\pm$ 0.47 was indicated.

The investigators concluded that it was not essential to monitor for HT in addition to HTO at Rocky Flats based on the fact that the maximum permissible concentration for public exposure was 200 times higher for HT than HTO and the observation that, during periods of highest total tritium content, effluents contained mostly HTO.

Dose factors for tritium are often tabulated with separate values for tritiated water and organically bound tritium (ICRP, 1989). For the purposes of this project, dose coefficients for tritiated water are most applicable.

Plutonium and Uranium Compound Solubility

While the chemistry of plutonium is complicated, the nature of operations at the Rocky Flats Plant favored the presence of some plutonium compounds in airborne emissions. Over essentially the entire history of operations at Rocky Flats, plutonium chemical purification and recovery were performed in addition to the machining of plutonium metal and fabrication of weapon components (ChemRisk, 1992). In moist air or moist argon, plutonium metal rapidly oxidizes, producing a mixture of oxides and hydrides (Faust *et al.*, 1988). Thermodynamically, PuO_2 is the most stable plutonium compound. Because moisture in the air greatly accelerates oxidation, fine particles of metal which became airborne from plutonium machining operations likely rapidly oxidized. Because finely divided plutonium is pyrophoric, common practice is to convert turnings to oxide as soon as convenient (Faust *et al.*, 1988).

Important compounds in the chemical processing of plutonium are $Pu(NO_3)_4$, PuO_2, and PuF_4 (Faust *et al.*, 1988; ChemRisk, 1992). Plutonium nitrate is readily soluble in aqueous solutions, and was an intermediate step in chemical processing of plutonium with nitric acid. Plutonium dioxide ("green cake") was an intermediate step in conversion of plutonium to metal at Rocky Flats. PuO_2 is chemically stable and relatively inert, and was the preferred form for shipping and storing plutonium. PuO_2 was hydrofluorinated to form plutonium fluoride in another step in the conversion process to produce plutonium metal; the usual form was PuF_4 ("pink cake"), which is a solid at room temperature. In a 1982 study of airborne particles sampled from a major ventilation plenum (FU2B) in Building 771, collected particles were presumed to be nitrates and fluorides of plutonium (Langer, 1984).

Depleted uranium operations at Rocky Flats were limited to metal forming and fabrication of weapon components (ChemRisk, 1992). Chemical purification and recovery of depleted uranium were not performed. Finely divided uranium, such as fines or chips from machining, readily oxidizes upon contact with air (ChemRisk, 1992). Depleted uranium in Rocky Flats airborne emissions was most likely in the relatively insoluble oxide and dioxide forms rather than the more soluble forms (such as uranyl nitrate) that are frequently encountered when uranium is chemically processed.

From 1954 to 1962, enriched uranium was chemically processed at Rocky Flats using methods modeled after those used at the Oak Ridge Y-12 Plant. Compounds important in the chemical processing of enriched uranium include relatively soluble uranyl nitrate, moderately soluble uranium trioxide and uranium tetrafluoride, and relatively insoluble uranium dioxide (ChemRisk, 1993; USDOE, 1988). Effluents from enriched uranium processing areas likely contained a mixture of these compounds. Before 1954 and after 1962, processing of enriched uranium at Rocky Flats was primarily limited to metal forming and component fabrication operations. Enriched uranium in Rocky Flats airborne emissions during these periods was most likely in the relatively insoluble oxide and dioxide forms.

2.2 Historical Effluent Sampling and Analytic Practices

The following sections provide a review of the sampling and analytic systems employed at Rocky Flats primarily for quantifying the release of radioactive particulates and gases. While airborne particle sampling systems that were in place since the time of initial operations in the early 1950s collected particulates in the exhaust stream, only plutonium and uranium were routinely reported throughout the history of the plant's operations. Routine continual sampling of water vapor in building exhausts for tritium analysis began in early 1974 and has continued to the present day. Pilot operation of an iodine sampling method for Critical Mass Laboratory exhaust in the fall of 1977 (Hornbacher, 1975-1982) did not result in the addition of iodine sampling to the routine exhaust sampling program. The iodine pilot sampling operation was an attempt to develop monitoring capabilities in the event of an accidental criticality (ChemRisk, 1991c, Interview No. 26, 32).

2.2.1 Sampling Systems Review

Rocky Flats radioactive effluent sampling systems were reviewed in the following key areas to evaluate the potential for major inadequacies or biases in the sampling results:

- System designs,
- Sampling flow rates and isokinetic sampling issues,

- Sample loss due to deposition of particulates in sample lines,
- Sampling apparatus filter efficiency, and
- Quantification of stack or vent effluent flow rates and total emission volume.

Much of the information that describes techniques used to collect and measure airborne effluent samples was obtained through a review of Rocky Flats laboratory records and through interviews of present and past plant employees.

2.2.1.1 Sampling System Designs

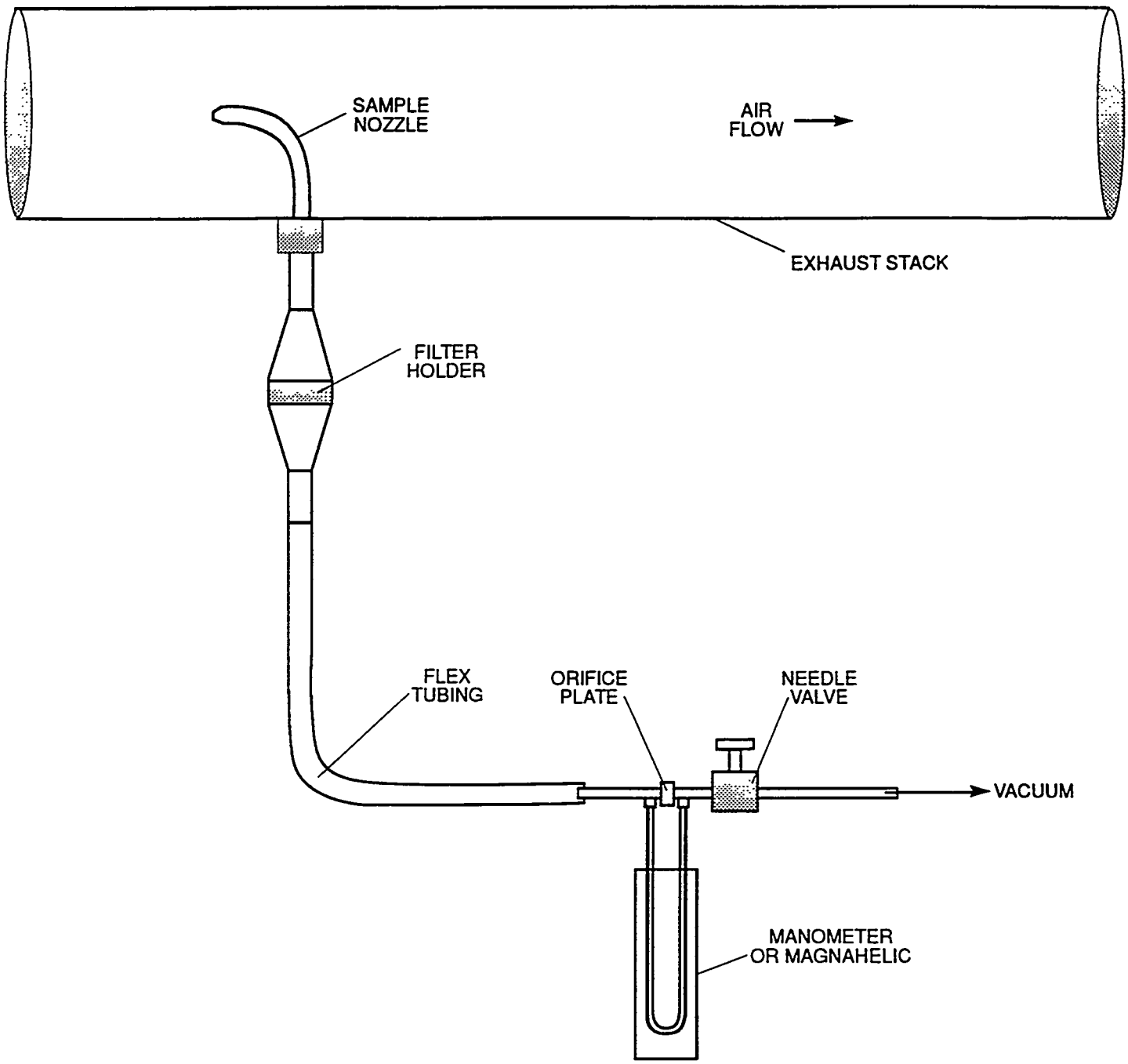
Sampling systems have historically been designed and operated to sample two types of routine radioactive effluent at Rocky Flats. One sampling system is designed to collect airborne radioactive particulates and the other is designed to sample airborne water vapor for tritium analysis. Each of these systems is described in the following sections.

Sampling of Airborne Particulate Matter

The particle sampling systems currently in use at the Rocky Flats Plant are not materially different than those used when the plant commenced operation in 1953. The design of these sampling systems is described in various Rocky Flats Plant internal reports (e.g., Unknown, 1955, 1985a, 1985b) and is described here. Depending on the size and flow rate of the ventilation exhaust duct or stack, one or more (up to six) locations were chosen for sample nozzle placement. Figure 2-1 illustrates the generic effluent sampling system design utilized at Rocky Flats.

Sample nozzles were fabricated on-site by machining a knife-edge nozzle on the end of stainless-steel sample tubing. The sample lines were commercially available stainless-steel tubing with inside diameters of between about 0.2 and 0.9 inch. The sample tubing was bent gradually so that the nozzle faced the airstream. Large radius bends were used to minimize the deposition of particulates in the sample tubing due to impaction.

Filter paper holders that were designed and fabricated on-site were generally installed along the sample tubing just on the outside of the duct, so that the length of sample line between the nozzle and the filter paper was kept to a practical minimum. Sample line lengths were reported to vary between about 14 inches and 10 feet. While the ideal design suggests the placement of filter holders immediately outside the duct, in some cases filter holders were placed at a distance from the duct because of the configuration of the duct or to allow for personnel access



ChemRisk
A Division of McLaren/Hart

FIGURE 2-1
GENERIC EFFLUENT SAMPLING
SYSTEM DESIGN

for exchange of filter papers. Appropriate sample flow rates were established through the use of orifice meters and flow control valves.

Sampling of Airborne Water Vapor for Tritium

Before 1974, sampling for tritium in airborne effluents was not routinely conducted. As early as 1962, Rocky Flats maintained instruments for detection of tritium gas in particular work areas of the plant because operations have sometimes resulted in the storage of tritium containers (Merril, 1965). Portable tritium survey instruments were maintained, some areas were routinely sampled, some operations were monitored with "sniffers," and starting around 1973, air from selected rooms or glove boxes was passed through liquid nitrogen cold traps to collect moisture for liquid scintillation analysis (Dow, 1973c).

Continuous sampling for tritium commenced in four ventilation systems in Buildings 771 and 774 in January 1974 (Putzier and Kittinger, 1975). Beginning in 1974, annual airborne tritium release totals from Rocky Flats were reported in annual environmental reports. In 1974, data were reported for 12 vents, and the program expanded to about 18 in 1977 and 23 in 1981 (EG&G, 1991a). In September 1988, continuous sampling for tritium was decreased to include only several exhaust systems that continued to have a potential for contribution to the tritium release site total (Rockwell, 1989).

Since 1973, Rocky Flats has used an in-house design Greenberg-Smith impinger (RFGSI) containing water for collection of water vapor from ventilation exhausts for the purpose of tritium analysis (Hurley, 1979). The tritium sampling train consists of a single extraction nozzle located within the exhaust duct, a calibrated rotameter for determining the sample flow rate, tandem impinger samplers containing distilled water, and a flow control valve for adjusting the sample flow rate (Campbell, 1985). Motive force is supplied by a centrally located vacuum system, with the sample flow rate adjusted to 50 cm^3 min^{-1}. Samples have typically been collected on Monday, Wednesday, and Friday of each week at the same time that particulate filter papers are exchanged. The RFGSI is only capable of monitoring for tritium oxide (HTO) and gives no indication of the total tritium found in stack emissions. Additional special tritium sampling studies are described below.

In 1976, an initiative to use ethylene glycol instead of water in the tritium samplers was documented. By April 1977, tests had shown that counting efficiencies using ethylene glycol were low and instrument adjustments would be necessary, making it difficult to analyze both water and ethylene glycol samples on one instrument. Problems with air flow restrictions caused the experimental ethylene glycol samplers to become inoperative in May 1980 (Hornbacher, 1975-1982). As water was still in use in the impingers in 1985 (Campbell, 1985), it appears that use of ethylene glycol was never adopted.

In 1978, a pilot test was performed whereby the RFGSI was modified to analyze for both tritium oxide as well as elemental tritium in Building 771 main exhaust (Hurley, 1979). The Östlund-design sampler initially collected HTO on a type 4A molecular sieve, then passed the airstream through a palladium-impregnated molecular sieve that catalyzed the oxidation of HT to HTO, which then adsorbed onto the molecular sieve. Heating then drove water off the molecular sieves for scintillation counting.

During periods when tritium-containing oxide material was being processed and the plutonium recovery incinerator was in operation (25 days over the 92 days studied), reported average concentrations in the Building 771 main exhaust corresponded to release rates of 4.9 $\pm$ 0.053 millicuries per day of HTO and 0.41 $\pm$ 0.012 millicuries per day of HT. These values are based on an exhaust rate of 184,000 cubic feet per minute ($ft^3\ min^{-1}$). The highest daily-average concentrations corresponded to release rates of 22 $\pm$ 0.068 millicuries per day of HTO and 6.1 $\pm$ 0.030 millicuries per day of HT. During nonoperating periods, average concentrations corresponded to release rates of 0.088 $\pm$ 0.17 millicuries per day of HTO and 0.045 $\pm$ .023 millicuries per day of HT.

Because of the relatively low biological hazard of HT compared to HTO and the observation that the ratio of elemental tritium to tritium oxide during periods of highest tritium content in effluents was quite low, use of the modified sampling device never became standard practice. Consequently, the majority of tritium collection was performed with water-based impinger systems.

The 1978 tritium species sampling study did, however, demonstrate apparent differences in the HTO collection efficiencies of the special sampler compared to the RFGSI system in routine use (Hurley, 1979). The special sampler HTO values were significantly higher than values from concurrent impinger samples: RFGSI values averaged 48 $\pm$ 27 percent of the tritium species sampler results. The differences were thought to most likely have stemmed from differences in the HTO collection efficiencies of the two systems, and further investigation was advised.

The special studies suggested that the efficiency of the RFGSI samplers may have been poor, resulting in the collection, on average, of only 48 percent of the tritium present. Although tritium sampler results have been corrected for sampler collection efficiencies of between 85 and 97 percent (Link, 1992), no evidence that the plant had investigated this efficiency issue further was located.

While more details on the range of collection efficiencies experienced at Rocky Flats have not been located, efficiencies for tritium (HTO) collection in water-filled washing bottles are "high and predictable," with efficiencies ranging from 90 to 98 percent reported (NCRP, 1976). A study performed at Los Alamos found that collection efficiencies in water-filled bubbler tritium

samplers ranged from 91 to 95 percent for tritiated water vapor and from 0.019 to 0.024 percent for tritium gas at sampling rates from 8 to 10 liters per minute (Valentine, 1968).

Tritium sampling efficiency is a source of uncertainty in the tritium emission estimates. Based on the limited special studies indicating a collection efficiency of 48 $\pm$ 27 percent, actual tritium emissions to the air are estimated to have ranged from factors of 1.3 (i.e., $(0.48 + 0.27)^{-1}$) to 4.8 (i.e., $(0.48\text{-}0.27)^{-1}$) times the reported amounts.

Summary:	Tritium sampling efficiency is a source of uncertainty in effluent sampling data. Based on data from a limited special sampling study for tritium, efficiency of the standard sampling system is estimated to be 48 $\pm$ 27 percent, meaning actual emissions to the air ranged from 1.3 to 4.8 times those reported.

Sampling of Airborne Iodine

Because of the potential for accidental airborne release of fission product radioiodine generated in criticality experiments that have been conducted in the Critical Mass Laboratory since 1965, an activated charcoal sampler was designed, fabricated, and installed on the Building 875 exhaust plenum in August 1977 (Hornbacher, 1975-1982). Building 875 houses the exhaust filter plenum for Building 886, which contains the Critical Mass Laboratory. Laboratory calibration of an activated charcoal sampling tube was performed, with analysis being conducted by direct counting of the I-131 gamma rays on a gamma spectrometer. The method yielded I-131 collection efficiencies of 86 percent to 91 percent. The pilot program was conducted to provide a basis for evaluation of this type of sampler and to establish the ability to monitor any releases of radioiodine that could occur as a result of a criticality accident in Building 886; however, no evidence of routine use of iodine samplers for airborne effluents from the Rocky Flats Plant has been located.

2.2.1.2 Sample Apparatus Flow Rates and Velocities

The following sections address the accuracy of the quantification of sample volumes and the importance of sampling velocities with regard to sampling bias.

Particle Sampling Flow Rates

Sample flow rates have historically been maintained at approximately 2 $ft^3\ min^{-1}$ for particulate sampling (Campbell, 1985). Orifice plates were installed just downstream of the filter paper holders in the sample line, with pressure taps just upstream and downstream of each orifice.

These pressure taps were connected to a U-tube manometer or a magnahelic pressure gauge in a manner that allowed differential pressure across the orifice to be measured as an indicator of sample flow. Sample lines were then connected to a plant vacuum source, which provides the motive force for drawing a sample. The amount of vacuum supplied by the central vacuum system is adjusted using the needle-type flow control valves, and measurement of the pressure drop across the orifice plate allowed calculation of the corresponding sample flow rate.

In air, if the pressure downstream of an orifice is less than 53 percent of the pressure upstream of the orifice, and the opening comprises less than 4 percent of the area of the orifice plate, that orifice is said to be critical: a sonic velocity is produced at the orifice exit, and constant flow is obtained (ACGIH, 1983). The orifices in the Rocky Flats sampling trains, although they may have sometimes been called critical orifices, were used not as flow regulators but rather as orifice meters to allow estimation of sample flow rates.

Calibration graphs were prepared that show the relationship between pressure drop across the orifice and volumetric flow rate ($ft^3\ min^{-1}$) flowing through the sampler for various types of filter papers (Boss, 1972-1974). The periodic air effluent calibration procedure includes recording the "as found" sample flow rate, adjusting the manometer zero and adding manometer oil as needed, and recording the manometer or magnahelic reading that was in place at the end of calibration (EG&G, 1991b). The measured sampling rates were then used in the calculation of emissions.

It is apparent from documentation of the calculations performed to estimate radioactive material emission quantities that, at least since mid-1974, sample flow rates used for emission total calculation were based on the length of the sampling period and an assumed flow rate of 2 $ft^3\ min^{-1}$, which is equivalent to 56.6 liters per minute (Haynes, 1975, and Unknown, 1976a). Calibration worksheets (EG&G, 1991b) that were reviewed indicated that "as found" sample flow rates varied as much as 38 percent from the assumed 2 $ft^3\ min^{-1}$. It is important to note that the record search yielded only a few calibration worksheets. This documentation provides an extremely limited record of sample flow rates. Flow rates recorded on these few calibration worksheets were far more commonly below the assumed value than above it, which would tend to lead to nonconservative errors (underestimates) in the calculated concentrations. A frequency distribution of observed sample flow rates from this data set divided by 56.6 L min^{-1} has a mean of 0.92 and a standard deviation of 0.11 (Figure 2-2). As described in Section 4, a correction factor, which is assumed to be normally distributed, will be used to describe sample flow rate fluctuations as a source of uncertainty in emission estimates.

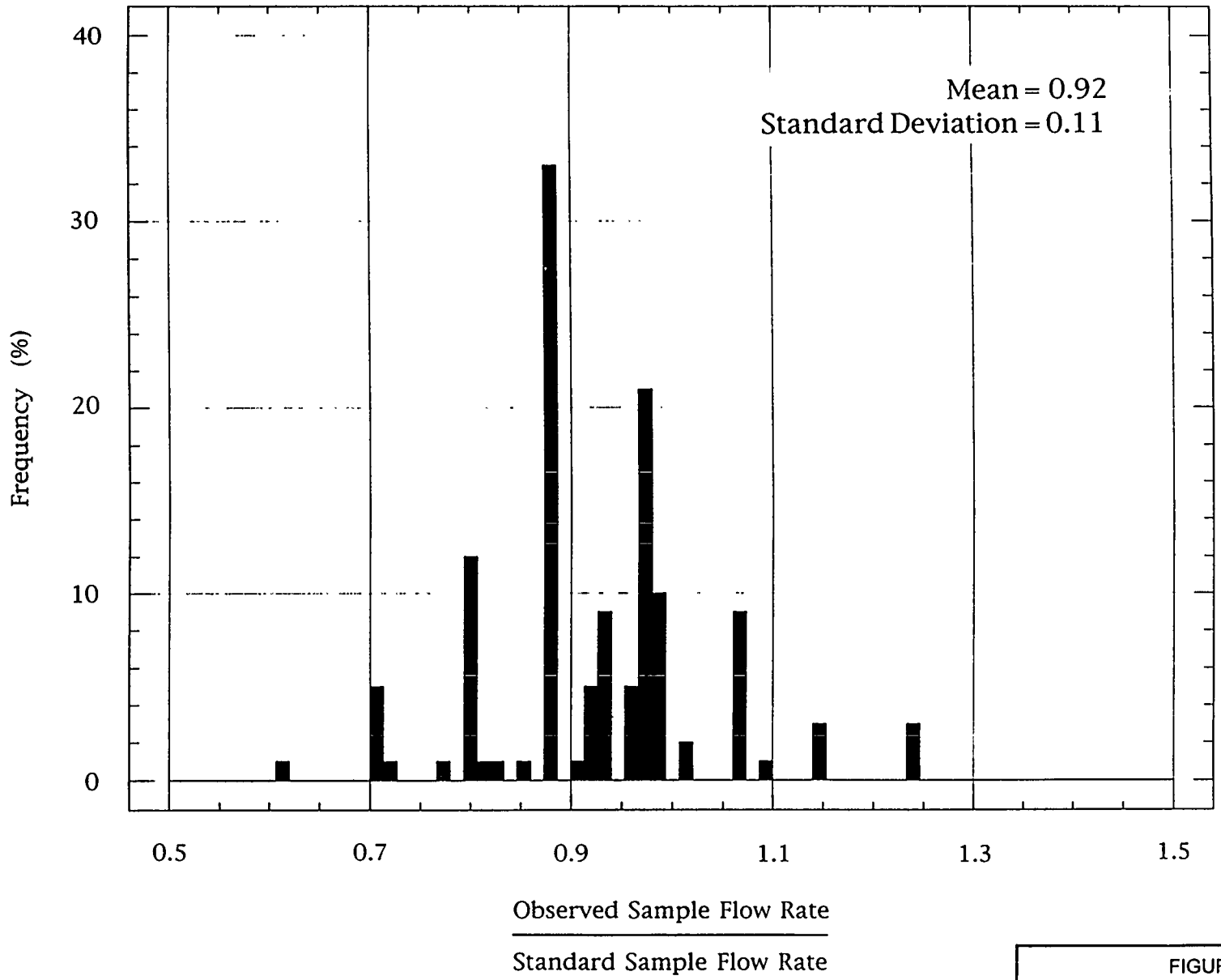


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FIGURE 2-2
A FREQUENCY DISTRIBUTION OF RATIOS OF OBSERVED SAMPLE FLOW RATES TO THE STANDARD SAMPLE FLOW RATE

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Summary: Particle sampling flow rates are a source of uncertainty in effluent data from 1974 to the present day. Data indicate relatively wide uncertainty bounds on this factor, with a bias toward underestimating flow rates. A limited worksheet review has led to the establishment of a correction factor distribution to reflect this uncertainty. The factor is assumed normally distributed and has a mean of 0.92 and a standard deviation of 0.11.

Particle Sampling Velocities

Sample probe diameters were selected from among seven commonly available stainless-steel tubing sizes of 1/4" to 1" in 1/8" outside-diameter increments (Rockwell, 1984). Probe sizes were reportedly selected so that the standard 2 ft^3 min^{-1} ($\pm$ 20 percent) flow rate would result in a flow velocity matching as closely as possible the expected flow velocity in the duct being sampled (Freiberg and Haynes, 1974). When the linear velocity of the extracted sample is equal to the linear velocity of the air in the duct at the sampling point, the sample extraction is said to be isokinetic. Isokinetic sampling is performed in order to prevent the collected sample from being nonrepresentative of the particle size distribution present in the sampled gas. Due to the momentum associated with "large" particles in the effluent, nonrepresentative sampling of the large particles may occur if sampling is not isokinetic. Large particles may not follow flow streamlines, hence they may bypass the sample nozzle if the sample flow rate is too high relative to the effluent flow rate (super-isokinetic).

Conversely, if the sample flow rate is too low relative to the effluent flow rate (sub-isokinetic), large particles outside the flow streamlines of the sampled volume may enter the nozzle. In either case, a sampling error has occurred relative to the large particles. Depending on the desired endpoint of the measurement (e.g., mass, gross activity, concentration), the error introduced by non-isokinetic sampling can occur in either direction, i.e., a positive or a negative bias to the result. Note that since "small" particles more readily follow flow streamlines, sampling isokinetically is not particularly important if the bulk of the contaminant in the effluent air is small particles.

Review of emission point and sampling system parameters for 54 exhaust systems monitored at the Rocky Flats Plant (Rockwell, 1984 and Unknown, RE-1029) indicates that many of the samples deviate significantly from isokinetic. The worst-case velocity mismatch among the data reviewed indicated sub-isokinetic flow by a factor of 17 (Unknown, RE-1029). However, this velocity mismatch occurred in Building 371, a building that never became fully operational. The greatest mismatches in sampling lines for operational buildings where plutonium was handled involved a factor of approximately six (sub-isokinetic and super-isokinetic) in Building 707. It

appears that either probe diameters were not as carefully selected as was indicated or that duct flow rates changed over the years and probes were not correspondingly altered.

The most commonly recognized reference on isokinetic sampling in nuclear facilities is ANSI N13.1-1969 (ANSI, 1969). Relative to "large" versus "small" particles, ANSI N13.1-1969 states:

> "Except in very unusual situations, particles smaller than an aerodynamic diameter of about five microns are able to follow the streamlines of the air, and the fractionation error is not great."

Regardless of effluent particle size distribution, it is simply good engineering practice to design sampling systems to be isokinetic at design process flow rates. As discussed previously in this report, the particle size distributions in the effluents (downstream of filtration devices) at Rocky Flats Plant are quite small, exhibiting mass median diameters of a few hundredths to a few tenths of a micron. An analysis of sampling errors associated with anisokinetic sampling at Rocky Flats is provided in Appendix C of this report. The results indicate that the errors introduced by anisokinetic sampling in the estimation of radionuclide releases from Rocky Flats were less than 5 percent.

An issue that is somewhat related to the issue of isokinetic sampling involves the placement of the sampling nozzles. Good practice dictates that sampling intakes be placed near the point at which the exhaust stream leaves the building and at some distance from bends or changes in the diameters of ductwork to ensure that samples are not taken from areas of flow disturbance that may not have a uniform distribution of contaminants across the airstream. Based on information reviewed to date, however, the sampling errors produced by suboptimum placement of sampling probes in effluent streams should be small given the relatively small particle size of contaminants at Rocky Flats and are not considered to be a significant source of error or uncertainty.

Summary:	Particle sampling velocities are not believed to be a significant source of bias or uncertainty in the effluent monitoring data.

Tritium Sampling Flow Rates

Sample flow rates have historically been maintained at approximately 50 cm^3 min^{-1} (0.002 ft^3 min^{-1}) for tritium samplers (Campbell, 1985). Sample trains include a rotameter for determining sample flow rate and a flow control valve for adjusting the flow rate. For tritium sampling, maintenance of isokinetic extraction velocities is not critical, since the contaminant is in the form

of a gas or vapor rather than a particle and the flow rate can be selected for optimal collection of water vapor (HTO) by the sampler.

As described earlier, sample flow rates have historically been set at approximately 50 cm^3 min^{-1} for tritium samplers. As was the case with particulate sampling, the documentation suggests that the plant assumed that samplers operated at the design flow rate for the purposes of calculating emissions. However, the actual average sampling flow rate is likely to be larger than this value because water was used as the trapping medium. As sampled air was bubbled through the medium, a portion of water was lost by evaporation. This typically results in a drop in resistance to air flow and an increase of sampling flow rate. Since data to characterize the variability of the actual sample flow rates were not located, a reasonable upper bound on this source of uncertainty in emission estimates is established at 1.5 times the estimate, with a lower bound at 0.9 times the estimate.

Summary:	The range of uncertainty in tritium emission estimates due to lack of knowledge of sample flow rates is estimated to be between 0.9 and 1.5 times the estimated emissions.

2.2.1.3 Sample Line Losses

An investigation was performed in 1973 to determine the amount of particulate deposition in sample lines used for effluent sampling systems in Buildings 707, 771, and 776 (Mossoni and Kittinger, 1973). These sample lines were between 0.305 and 0.430 inches in inside diameter and 10 to 14 inches and 9 feet (108 inches) in length. Sample flow rates varied between about 1.4 and 2.0 ft^3 min^{-1}. After being in service for at least one year, sample lines were physically removed and taken to the laboratory for washing with nitric acid and distilled water.

The wash solutions were then analyzed for plutonium by liquid scintillation counting. Results of this study indicated that plutonium deposition in sample lines accounted for about 10 percent or less of the total activity sampled. Due to the small particle size of the effluent sampled, inertial losses in the sample lines were probably quite small, thus the low observed deposition.

The only other document that was located that addressed concerns regarding the potential for sample line losses examined sampling systems used for glove-box exhausts and alpha continuous air monitors (CAMs) in the vicinity of the supercompactor (Newton, 1991). The memorandum stated concerns about the convoluted aerosol transport path associated with the deployment of the CAMs and the potential for intolerable particle losses. However, the document did not provide any estimate of the potential loss, nor was the described system associated with direct measurement of releases to the environment.

To evaluate Mossoni and Kittinger's claims, line deposition losses for representative sampling lines at Rocky Flats were calculated using a computer program based on the method described in Appendix G of Voillequé *et al.* (1991). An outline of the approach is provided in Appendix B. Results of the calculations are summarized below.

Line deposition losses were calculated for:

- A 108-inch sampling line, with inside diameter of 0.43 inches and a flow rate of 688 cubic centimeters per second (cm^3 sec^{-1}). This corresponds to the longest lines tested by Mossoni and Kittinger.

- Lines with inside diameters of 0.402 inches, with lengths and flow rates of 31.75 inches and 950 cm^3 sec^{-1}, 25.75 inches and 967 cm^3 sec^{-1}, and 14 inches and 983 cm^3 sec^{-1}. These parameters correspond to the lines where Mossini and Kittinger measured the greatest deposition.

Particle deposition increases with particle size. Almost all of the particles in Rocky Flats filter plenum exhaust effluent have physical particle diameters smaller than 0.3 micrometer (μm), and calculated deposition losses of 0.3 μm particles in the sampling lines were less than 0.1 percent in all the above cases. This is fully consistent with the claim by Mossoni and Kittinger that errors due to sampling line losses were less than 10 percent of the measured concentrations.

Summary:	Sample line losses should not be a significant source of bias or uncertainty in the effluent monitoring data, based on the small particle size of the effluent and results of a study on line losses.

2.2.1.4 Sampling Apparatus Filter Efficiency

The particle collection efficiency of the filter in a sampling device can be an important determinant in the accuracy of the sampling results. One study was identified that addressed the efficiency of the filter media used to monitor radioactive particulates in process streams and in building air at Rocky Flats (Langer, 1984). The study involved sampling of particles from Building 771 ventilation plenum FU2B, the plenum which historically was found to be challenged by the highest concentrations of radioactive aerosols at the Rocky Flats site. The special sampling effort used four sets of two Whatman EPM 1000 glass fiber filters in tandem over a 10-week period to evaluate the effectiveness of the filter papers in collecting radioactive effluent that had passed through four HEPA filter stages. One set of filters was exposed to nuclear track film for autoradiographic determination of plutonium particle size. Each filter was analyzed for gamma emitting radionuclides and then specifically for plutonium isotopes.

The study concluded that measured filter efficiencies were consistent with values of between 99.7 percent and 99.9 percent, and that the particles incident on the filters had an average size near 0.3 μm, or an average aerodynamic equivalent diameter of 0.5 μm. Standard calculation methods used with these data indicate an average aerodynamic diameter of about 0.6 μm. These results indicate that uncertainties associated with sample collection efficiencies are relatively small for the filter paper that was tested.

Filter paper manufacturers routinely test and report the efficiencies of their filter papers in collecting particles with diameters of 0.3 μm. This particle size has been adopted as the standard for testing because, as shown in Figure 2-3, it is the particle size associated with the lowest efficiency of collection by the various filtration collection mechanisms (i.e., impaction, interception, and diffusion). Therefore, collection efficiencies of particles either larger or smaller than 0.3 μm should be greater than that measured for 0.3 μm.

The plant has used a number of filter papers historically:

Year	Filter
1953 to 1973	Hollingsworth & Vose HV-70 — cellulose-asbestos filter
1973 to 1978	Gellman AE — glass fiber filter
1978 to 1990	Whatman EPM 1000 — glass fiber filter
1990 to present	Whatman EPM 2000 — glass fiber filter

Particle collection efficiencies for these filters reported by the manufacturers and also summarized in ANSI N13.1 (ANSI, 1969) all exceed 99 percent for 0.3 μm particles at the sampling velocities employed by the plant.

Summary: Sampling apparatus filter efficiency should not be a significant source of bias or uncertainty in the effluent monitoring data based on well-documented filter efficiencies and the results of a plant study.

2.2.1.5 Stack or Vent Exhaust Flow Rates and Volume Quantification

Effluent sampling systems are designed to quantify the concentration of contaminants in an exhaust flow by collecting or measuring all the contamination in some known fraction of the total exhaust volume. The total amount of a contaminant released by an exhaust flow can then be calculated by multiplying the concentration of contamination detected by the sampling device by the total volume of air that was exhausted over the sampling period. Therefore, in addition to measuring the contaminant concentration in an effluent stream (e.g., microcuries of contaminant present per cubic meter of sampled air, μCi m^{-3}), the total volume of air exhausted

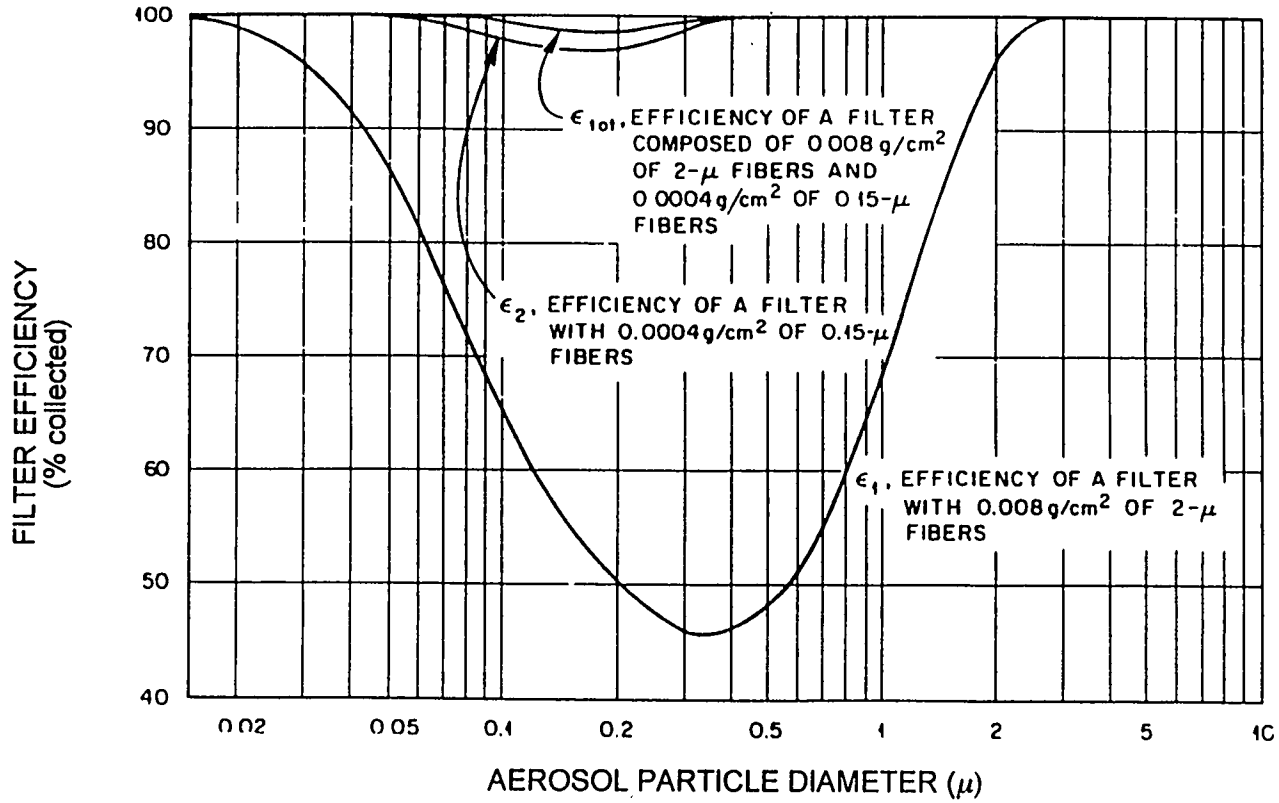

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FIGURE 2-3
CALCULATED FILTER EFFICIENCIES
ASSOCIATED WITH
THREE AIR FILTERS

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(e.g., total volume of contaminated air in cubic meters, m^3) must also be established in order to quantify the total volume of contaminant released:

Total Contaminant Released (μCi)	=	Concentration of Contaminant in Airstream (μCi m^{-3})	×	Total Volume of Air Exhausted (m^3)

The historical records indicate that two different methods were used to calculate contaminant release totals using the contaminant concentration measurements and total exhaust volume estimates. That the early Rocky Flats Plant was divided into discrete A, B, C, and D "plants," each with a large degree of operational independence, likely contributed to this difference in health physics practices in calculating release totals. In the first method of calculating release totals, the sampled concentrations were multiplied by the measured or estimated discharge flow for the sampling period for each stack to obtain the total amount of radioactivity released. These total contaminant release values were then summed to arrive at monthly release totals.

In the second method, the sampled concentrations were averaged for a month for each stack or duct, and this result was multiplied by the total flow discharged for the month. Although this second method is not as accurate as the first method, it will yield acceptable results as long as the flow rates are not excessively variable.

Throughout the series of records reviews and personnel interviews that were conducted with regard to radioactive effluent measurements, there were few indications of the origins of the exhaust flow rates or integrated exhaust volumes historically used in release total calculations.

The flow rates used in calculations during the 1950s and 1960s were apparently based on one or more of the following:

- Engineering design flow rates, such as from design specifications of fans.
- Original measurements made to balance flow in heating, ventilation, and air conditioning (HVAC) ductwork.
- Installed differential pressure flow elements and gauges, such as the commonly used magnahelic pressure gauge.
- Periodic measurements performed specifically for assisting in the quantification of effluents.

It is clear from the review of effluent calculations from the 1950s and 1960s that the effluent flows used were commonly standard flow rates multiplied by the number of hours of work that occurred in the building during the sampling period. The fans were apparently started at the beginning of a work period, usually for one or two shifts, and shut down at the end of the work period. As a result, the effluent volume was calculated using standard flow rates (measured or estimated) and the number of hours of fan "on" time.

Beginning around 1970, Rocky Flats personnel began to conduct tests of ventilation exhaust flow rates in various ducts and stacks using standard pitot tube type measurement techniques (Boss, 1978; Unknown, 1985a). In addition, flow rate measurement sensors and totalizers were installed in selected ducts and stacks around 1975 (Boss, 1991).

The practices associated with exhaust volume quantification are a source of uncertainty for the emission estimates.

In the interest of quantifying the potential variability in exhaust flow rates for buildings at the plant, available data quantifying exhaust volumes have been plotted. Data are plotted for Building 771 (Figure 2-4), which is the primary building handling and releasing plutonium, and for Building 883A (Figure 2-5), which was the only uranium building for which more than a couple of flow rate data points could be established from the documentation. The data points on Figures 2-3 and 2-4 during the 1980s have been obtained from computerized data sheets that listed the actual data that is assumed to be derived from flow totalizers at the plant.

The effluent volume data plotted for 1969 and the 1970s was reported on a number of work sheets that were found in plant files, while the volume data plotted for the 1950s and 1960s was back-calculated using information contained in effluent data calculation sheets. The basis for these data could not be established and could well have included any of the identified estimating techniques or measurement methods used by the plant.

The fluctuation of the effluent volume for Building 771 (Figure 2-4) is relatively small, with the highest volume, reported in 1987, being about 1.6 times greater than the lowest volume, reported in 1986. The data for earlier periods (the basis of these data could not be established) suggest that effluent volumes may have reached a level that was relatively consistent once the basic nature of the operations performed in Building 771 stabilized in the late 1950s. These data suggest that errors in the range of 50 percent (i.e., uncertainty range of 0.5 to 1.5 times emission estimate), and on the high end certainly no more than 100 percent (i.e., 2 times emission estimate), could plausibly be associated with the use of fixed effluent volume assumptions that may have been used in the 1950s, 1960s, and early 1970s.

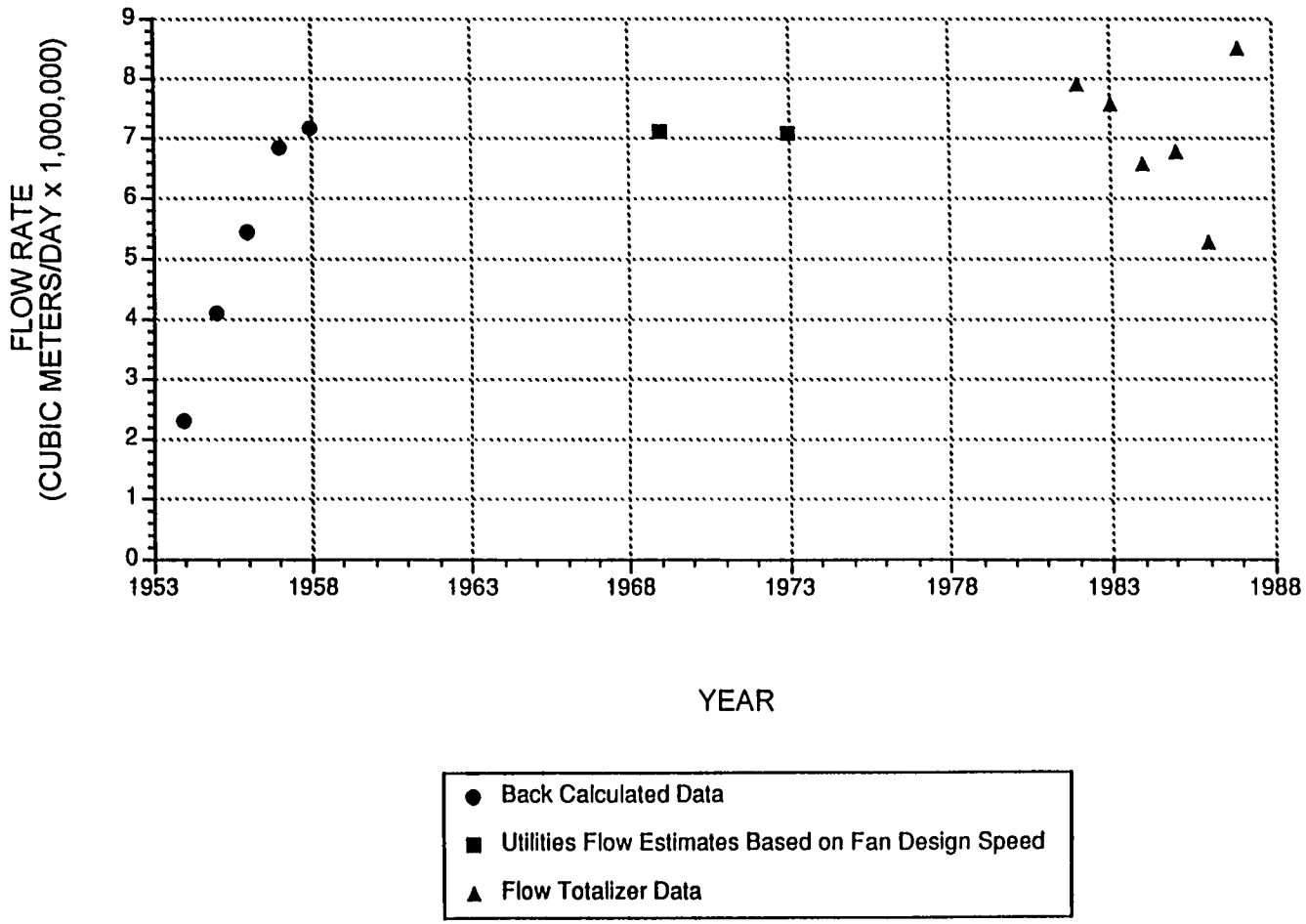

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FIGURE 2-4
FLOW RATE FOR MAIN EXHAUST
IN BUILDING 771

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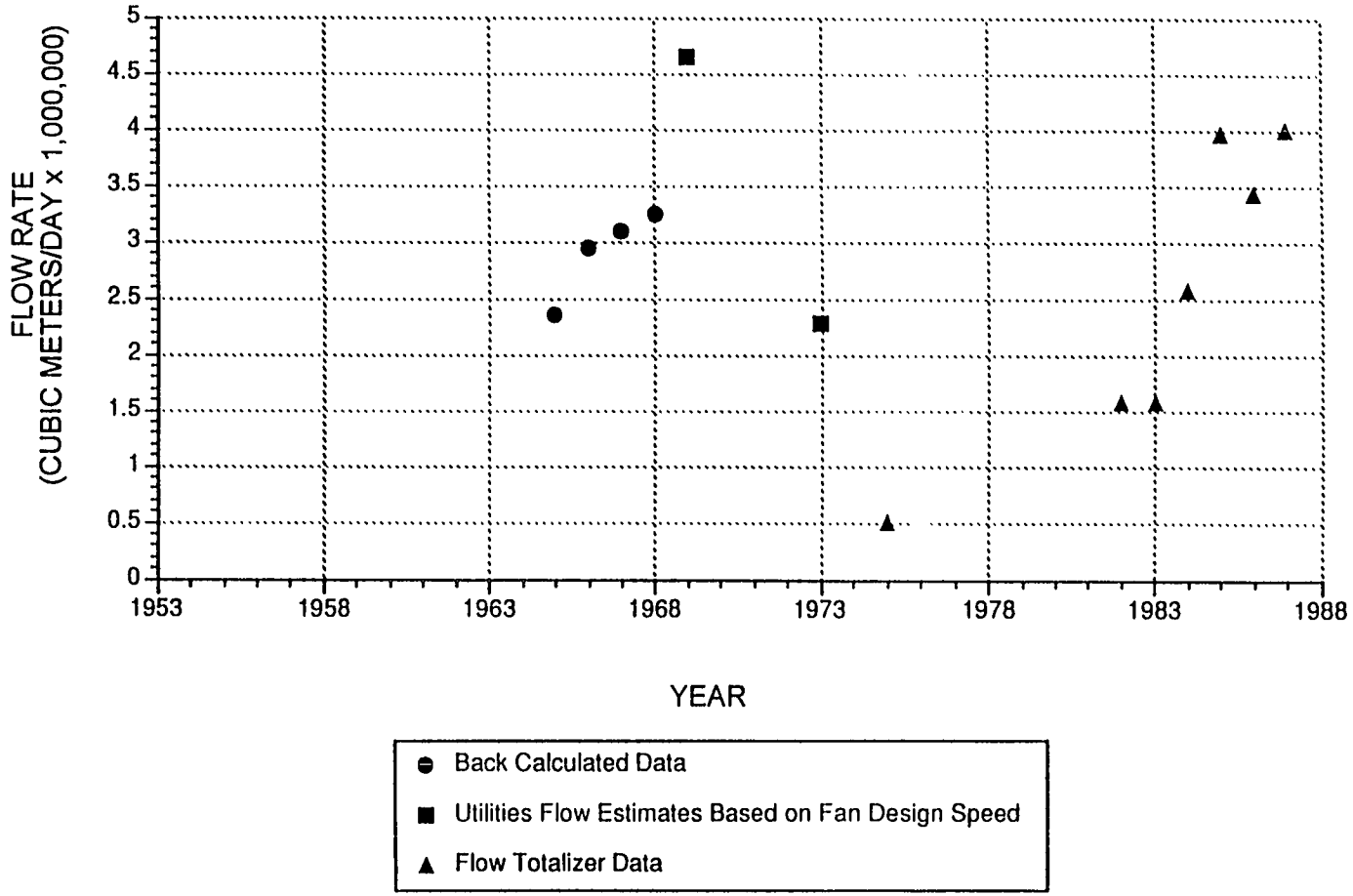

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FIGURE 2-5
FLOW RATE FOR EXHAUST
IN BUILDING 883A

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The fluctuation in effluent volumes for Building 883A (Figure 2-5) appears to be much greater; however, much of this fluctuation can likely be attributed to efforts to save energy in the early 1970s as part of the plant's response to the energy crisis. Plant documents indicate that the plant made efforts to reduce energy consumption during the early 1970s by turning ventilation systems down or off when there were no activities in a building (Boss, 1972-74). If it is assumed that the lowest values recorded in the 1970s and early 1980s were due to plant practices specific to this particular time period, the variability in effluent volumes for other time periods would again fall in the range identified for Building 771.

The Rocky Flats plant completed a number of studies in 1992 related to the performance of the ventilation exhaust monitoring systems. One of the investigations included the measurement of exhaust flow rates using a reference method and comparing the results to those produced by the standard measurement devices and flow totalizers that were in place in the ducts. Plant personnel performing the study have indicated that the recent-day totalizer results frequently overstated exhaust volumes (and therefore emissions), and that totalizer calibrations were generally last performed in the early 1980s (Osborne, 1992 a, b).

Measurements taken in 1992 in many Rocky Flats exhaust ducts indicated that at that time, flow rates reported by the standard measurement devices or flow meters used in the ducts on average over-stated the flow rates measured by the more accurate reference method by 120 percent, or a factor of two over reference-method flow rates for all ducts combined. Measurement errors in individual ducts ranged from under-estimates of flows by 79 percent to over-estimates of flows by 531 percent (Osborne, 1993). These measurements characterize errors associated with the performance of measurement systems over a relatively small period of time. There were no series of measurements in the individual ducts to further characterize the accuracy of measurements from the standard flow meters or the variability of flows in the ducts to evaluate the effect on the annual average estimates of exhaust volumes. However, these preliminary results suggest that effluent volume quantification has continued as a potential source of error even after the installation of flow-totalizers.

This limited review of data, along with the recognition of the fact that operation of a ventilation system at more than twice its design volume for an extended period is unlikely, supports the establishment of an upper bound on this source of uncertainty at two times the estimate. It is more likely that a ventilation system would be turned down or off, thus reducing effluents. A reasonable lower bound of uncertainty associated with an assumed volume is estimated to be 0.5 times the value used.

Summary: The range of uncertainty in emission estimates due to lack of documentation of the exhaust flow volumes is estimated to be 0.5 and 2 times the estimated emissions for plutonium, uranium, and tritium.

2.2.2 Analytical Methods

The radioactive effluents that sample analyses must be able to quantify consist primarily of depleted uranium, enriched uranium, and weapons-grade plutonium. Rocky Flats effluents also include, to a lesser extent, americium and tritium. Thorium was used in very limited applications, and, as will be discussed later, was likely associated with only very limited emissions. The isotopes uranium-233 and plutonium-238 each were used to a limited extent in a few special projects.

Depleted uranium has lower levels of the fissile uranium-235 isotope than the approximately 0.7 percent found in naturally occurring uranium. As a result, depleted uranium consists primarily of the uranium-238 isotope. The enriched uranium used at Rocky Flats is enriched in uranium-235 content to approximately 93 percent by weight. On an activity basis, however, uranium-234 constitutes most of the radioactivity of enriched uranium due to its much higher specific activity.

Weapons-grade plutonium is a mixture of isotopes in the following approximate percentages by weight (EG&G, 1989):

Plutonium-239	93.79	percent
Plutonium-240	5.80	percent
Plutonium-241	0.36	percent
Plutonium-242	0.03	percent

A significant contaminant in plutonium is americium-241, a radioactive daughter of plutonium-241. At the time plutonium is either first produced in a reactor or purified by chemical separation (e.g., molten salt extraction), the americium content is essentially zero. As the plutonium ages (decays), the americium-241 daughter activity increases. The average age of weapons-grade plutonium is about 10 years, and americium-241 accounts for about 10 percent to 20 percent of the total radioactivity (USDOE, 1980).

During the first two decades of plant operation, it was assumed that the radioisotope responsible for the total long-lived alpha radioactivity measured in a sample was the radioisotope that was primarily processed in the building that the sample came from. In other words, all releases from enriched uranium, depleted uranium, and plutonium buildings were assumed to be enriched uranium, depleted uranium, and plutonium, respectively. Subsequently, analytic methods have

been used to quantify the specific elements and their isotopes found in plant effluents. However, in some cases accurate reporting of the specific isotopes in plant effluents did not take place until the late 1980s.

2.2.2.1 Analytical Practices for Alpha-Emitting Particulates

During initial plant operations in 1953, air effluent samples were counted for long-lived gross alpha radioactivity with gas-flow proportional counters manufactured by a commercial supplier. Within the first year of operations, spurious electrical problems led to modification of these counters by installing photomultiplier tubes coupled with zinc sulfide detectors in place of the gas-flow component of the counters. These modified counters were called "SC counters" and were also known as "scintillation counters." These early counting systems were located in the individual production facilities throughout the Rocky Flats Plant.

In 1956, Rocky Flats replaced the earlier model of the alpha scintillation counter with a commercially available counter called the MAC 5. This instrument consisted of a bank of alpha scintillation counters used for gross alpha counting of the effluent samples collected in the production facilities. Each production or processing facility at the Rocky Flats Plant was equipped with one of these instruments to provide direct field support for measurement of airborne effluent samples collected in the ventilation and exhaust systems.

Due to the difficulty in obtaining spare parts for the MAC 5 counters, Rocky Flats decided in 1964 to replace the MAC 5 counters with similar alpha scintillation counters that were designed in-house and fabricated commercially. They became known as "Sharene counters" (Tyree, 1992). As with previous counters, these instruments were used for gross alpha counting.

In 1974, all of the alpha scintillation counters used for gross alpha counting of effluent samples were moved to a central laboratory location in Building 123. This move was to address concerns with potential contamination problems and electrical power fluctuations associated with the production facilities (Haynes, 1975), and the possibility that laboratories in production facilities might become inaccessible in cases of emergency, as occurred with the 1969 fire in Building 776/777 (Tyree, 1992).

Beginning in July 1973, samples from each building were composited weekly and analyzed for specific isotopes. The procedure involved ion exchange chromatography followed by alpha spectroscopy (Dow, 1974a). Alpha spectroscopy had been performed on environmental samples since at least 1960 and probably earlier. In the early alpha pulse-height analysis of environmental samples, samples were digested and evaporated onto stainless-steel disks and placed inside a Frisch grid ionization chamber with counting gas of 90 percent argon and 10 percent methane.

Also starting in 1973, solid state surface barrier (SSSB) detectors were employed by the airborne effluent monitoring program. These detectors provided specific isotopic analysis capabilities through alpha pulse-height spectrometry. Up until 1979, these surface barrier detectors were used primarily to measure samples that were above background based on gross alpha count results. In 1979, counter procedures changed so that all air samples were measured for specific plutonium isotopes exclusively with an alpha spectroscopy system. Gross alpha counting of air samples for quick screening continued to be performed on the Sharene counters up until 1985, when they were phased out and alpha spectrometry became the primary mode for analyzing air samples.

A historical summary of the counting equipment used to measure airborne effluent samples for the Rocky Flats Plant is presented in Table 2-1.

Detection Efficiency and Self-Absorption Factor for Particulates

From 1953 to 1974, Rocky Flats measured gross alpha radioactivity collected on air filters to estimate routine stack emissions of plutonium and uranium from production buildings. As previously discussed in this report, radioactivity on air filters was measured with alpha scintillation radiation detection equipment. Since plutonium and uranium process operations were typically conducted in separate buildings, gross alpha measurement results were attributed to either plutonium or uranium depending upon the main functions of each building.

There were two correction factors applied to gross alpha counting results obtained with the alpha scintillation detectors to account for inefficiencies in the detector and errors that arose from absorption of alpha particles emitted by radioactive particles buried in filter paper or obscured by other material such as ambient dust collected on the filter. Monitoring records indicate that Rocky Flats routinely applied such correction factors to all gross alpha radioactivity analyses of effluent samples (Haynes, 1975). Other references indicate these same factors were first used in the 1950s and continued to be used for the same application until the mid-1970s, when alpha spectrometry became the primary method for quantifying emissions of alpha-emitting radionuclides (Boss, 1982).

The following discussion describes the method in which correction factors were applied to gross alpha radioactivity measurements to determine the quantity of radioactivity contained on individual air particulate filters. Both active and retired Rocky Flats personnel stated that values of 0.30 and 0.70, combined into a "total" counting efficiency factor of 0.21, were historically used to correct sample counting results for detector efficiency and alpha particle self-absorption (Haynes, 1992). The value of 0.30 meant that the counting instrument measured only 30 percent of the radioactivity present on the air filter, and was commonly referred to as the counting

TABLE 2-1

ROCKY FLATS INSTRUMENTATION FOR COUNTING AIRBORNE EFFLUENT SAMPLES

TIME PERIOD	INSTRUMENT TYPE	TYPE OF ANALYSIS	PLANT LOCATION(S)	COUNTING EFFICIENCY[3] (%)	CALIBRATION SOURCE	SELF-ABSORPTION CORRECTION	FILTER PAPER
1953 - 56	alpha scintillation counter, "SC" (converted from gas-flow type)	gross alpha, Pu-239, U-235, and Am-241	771, 881, 774, 991, 441	30 ± 3	evaporated and electroplated pure Pu-239 and U-235	included in calculations but not listed separately	Hollingsworth and Vose HV-70
1956 - 64	MAC 5 counter, alpha scintillation counter	gross alpha, Pu-239, U-235, and Am-241	444, 447, 771, 776, 774, 777, 778, 881, 883, possibly others	30 ± 3	electroplated pure Pu-239 and U-235	included in calculations but not listed separately	Hollingsworth and Vose HV-70
1964 - 85	"Sharene"alpha scintillation counter	gross alpha, Pu-239, U-235, and Am-241	771, 776, 777, 778, 881, 883, & 123[2]	30 ± 3	electroplated pure Pu-239 and U-235	0.30[4]	Hollingsworth and Vose HV-70, Gellman AE[5]
1973 - present	surface barrier detector, ND 6600, Tennelec detector (New Canberra system, 1991)	alpha spectroscopy and gross alpha[1]	123	30 ± 3	electroplated pure Pu-239 and U-235	N/A	Gellman AE, Whatman EPM 1000, Whatman EPM 2000[6]
1973 - present	liquid scintillation counter	tritium	123	84 to near 100 based on quench curves	internal tritium standards	N/A	RFGSI Impinger

1 In 1973, Rocky Flats began to perform specific isotopic analyses, i.e., alpha spectroscopy, on all air samples that were above background levels. In 1979, surface barrier detectors were used for gross alpha counting only for air samples collected in plutonium processing buildings.

2 In 1974, all instruments were moved to a centralized location in Building 123.

3 The ± 3 percent is not a statistically derived standard deviation, rather a percent range that each instrument must fall within based on daily measurements of an electroplated calibration source. They assumed that the quoted activity on the calibration source was within +/- 10 percent of the true activity. The error associated with counting efficiencies needs to be included in the error propagation of the overall measurement uncertainty.

4 The self-absorption factor was adopted in 1971 as a separate factor used to calculate gross alpha activity in air filters. This factor does not apply to alpha spectroscopy.

5 Gellman AE filters were adopted by the sampling program in 1973.

6 1973 - 1978 Gellman AE
1978 - 1990 Whatman EPM 1000
1990 - present Whatman EPM 2000

NA = Not Applicable

efficiency. This efficiency factor was based on counts collected from calibration sources with known radioactivity content.

Counting Efficiency:

$$Eff = \frac{c\ min^{-1}}{dis\ min^{-1}} = 0.30$$

Where:

Eff	=	counting efficiency of alpha scintillation detector (counts/minute per disintegration/minute; c min^{-1}/dis min^{-1})
c min^{-1}	=	count rate indicated by detector (counts/minute)
dis min^{-1}	=	radioactivity of calibration source (disintegrations/minute)

It was assumed that 30 percent of the alpha particles emitted by the radioactivity imbedded in each filter paper would not penetrate the fibers of the paper or the accumulated dust layer. Only 70 percent were assumed to be emitted from the sample, and of those that did, only 30 percent were assumed to be detected. A combined correction factor resulted:

Combined Correction Factor (Counting Efficiency and Self-Absorption):

$$CCF = Eff \times Abs$$

Where:

CCF	=	combined correction factor (c min^{-1} per dis min^{-1})
Abs	=	self-absorption factor (dimensionless)

To correct for the effects of incomplete detection and self-absorption, plant personnel would divide measured counting results by 0.21 as depicted below.

Calculated Activity of Plutonium or Uranium:

$$A = \frac{c\ min^{-1}}{(0.21)(2.22)}$$

Where:

A	=	radioactivity in the sample (picocuries, pCi; 1 pCi = 1×10^{-12} Ci)
2.22	=	conversion factor (dis min^{-1} per pCi)

The fact that the counting efficiency/self-absorption correction factor was assumed to be independent of alpha particle energy could be one source of the discrepancies that were observed when results of total long-lived alpha analyses were compared to isotope-specific analyses.

Measurements obtained with the MAC 5 instruments (used initially in 1956) and Sharene counters (which replaced the MAC 5 units in 1964) were both routinely adjusted with the reported "total efficiency" value of 0.21. Plant personnel verified counting efficiencies by performing daily measurements with electroplated standard alpha radiation sources that were traceable to the National Bureau of Standards. The values remained reasonably consistent over the years, due in large part to plant procedures which required technicians to adjust each instrument's amplifier gain in order to maintain 30 percent counting efficiency (Haynes, 1975; Tyree, 1992). Based on review of the available information, the 30 percent counting efficiency value appears to be reasonable for the type of detector and measurement application used at the Rocky Flats plant for gross alpha counting.

Documents that describe the technical basis for the self-absorption correction factor were not located during this investigation. Interviewees referred to a study that derived or confirmed the technical basis for the self-absorption correction factor. However, documentation of such a study was never located. The study reportedly used a gamma ray counter to measure the conversion X rays emitted from the decay of plutonium deposited on a filter paper. Assuming negligible loss of conversion X rays emitted from the paper, the measurement could then be used to quantify the total alpha radioactivity contained on the filter paper. Converting the conversion X ray value to disintegrations per minute (dis/min) and then comparing this to the direct alpha counting results yielded a burial loss value that reportedly agreed quite well with the 30 percent value used for the HV-70 filter paper (Putzier, 1982).

The type of filter paper used for sampling airborne particulates in effluent streams changed over the years; however, there was no corresponding change in the self-absorption correction factor used when quantifying gross alpha radioactivity. It is reported that the factor was used because it had always been used, and because plant personnel believed that it led to conservative

estimates, tending to over-estimate total plutonium or uranium released to the atmosphere. Although lacking documentation that describes the origin of the self-absorption factor, it appears that the value used was a reasonable approximation of counting losses encountered with the types of alpha counting instruments in use at the time. The variability and uncertainty associated with this factor is expected to be significant due to the high variability of dust-loading conditions that were likely to have existed in the air streams that were sampled. Documentation of the uncertainty associated with the correction factor was not identified during this investigation. Without supportive evidence documenting the variability of this factor, it is difficult to quantify what influence it had on the overall uncertainty of historical airborne plutonium, americium, and uranium release estimates.

One record was located that documented periodic determinations of counting efficiency in the early years of plant operation; it was confirmed through interviews with several active and retired employees that daily calibration checks were performed on counting instruments through the use of standards traceable to the National Bureau of Standards (Haynes, 1975; Haynes, 1992; Tyree, 1992). This same practice to validate counting efficiencies was continued into at least the late 1970s (Boss, 1978). Since extensively documented quality control programs for laboratory instrumentation are a fairly recent development, this lack of documentation in the earlier years was not inconsistent with standard practices employed by many nuclear facilities at the time. The correction factors that were used are considered to be consistent with those normally used for the type of instrumentation in question, and, although the uncertainty associated with the self-absorption factor does not appear to be documented, the factors described in this discussion appear to be the best estimates that are available for this study. A further discussion of the uncertainties associated with airborne radionuclide release estimates is found later in this report.

Summary:	The plant has routinely applied reasonable correction factors for detection efficiency and filter paper self-absorption. Therefore, these factors should not be a source of significant bias or uncertainty in the emission estimates.

Analytical Sensitivity

The sensitivity of effluent analysis techniques is important when sampling results are reported as non-detects, meaning that radioactive materials were not present in the samples at levels that could be positively detected at a stated level of confidence. The lower limit of detection (LLD) is a measure of the sensitivity of a specific analytical technique for radioactive materials. Although the terms have slightly different meanings, LLD is commonly used interchangeably with another parameter called minimum detectable activity (MDA).

The LLD is a statistical measure that defines an activity (or concentration) of radioactive material that can be detected above background for a specific sampling and analysis scenario. It is by definition an "a priori" measure, that is, it is a characteristic of the specific technique rather than an after-the-fact determination for a particular sample and analysis. For analysis of alpha-emitting radioactive materials in airborne effluents collected on filter paper samples, LLD is defined as shown in Figure 2-6.

The analysis of alpha emitting particulates of uranium and plutonium collected on filter paper samples requires a relatively high sensitivity when applied to effluent samples. This sensitivity is achieved by relatively long counting times and low backgrounds. Sample count times used at the Rocky Flats Plant were typically about 60 minutes. Background count rates of the alpha scintillation detectors were on the order of 0.1 count per minute.

Based on review of raw data for effluent sampling and analysis at the Rocky Flats Plant, the above values are typical. This level of sensitivity would apparently have met the needs of airborne effluent monitoring at Rocky Flats, since the LLD corresponds to a small fraction (less than one percent for most buildings) of the concentrations typically emitted from release points at the plant.

Representative LLDs for the sampling and analytic methods used for quantifying airborne effluent at the Rocky Flats Plant can be calculated using data from 1976 (Rockwell, 1976a) and the equation found in Figure 2-6:

$$LLD = \frac{4.66\sqrt{\frac{0.1}{60}}}{0.30 \times 1 \times 0.70 \times 200 \times (2.22\times10^{6})} = 2\times10^{-9}\ \mu Ci\ m^{-3}$$

Summary:	The analytic methods historically employed by the plant were sufficiently sensitive to detect contaminants in the effluent stream.

$$LLD\ (\mu Ci\ m^{-3}) = \frac{4.66 \times S_b}{Eff \times C \times Abs \times F \times (2.22 \times 10^6)}$$

Where:

4.66	=	$(2)(\sqrt{2})(k)$, where k = 1.645, representing a 5 percent chance of incorrectly determining radioactivity is present when it is absent and 95 percent confidence that radioactivity will be detected when present (Watson *et al.*, 1980).
S_b	=	standard deviation of background count rate, c min^{-1}

$$S_b = \sqrt{\frac{N_b}{t_b}}$$

N_b	=	the background count rate, c min^{-1}
t_b	=	the background count time, minutes
Eff	=	counting efficiency, c min^{-1} per dis min^{-1}
C	=	collection efficiency, dimensionless
Abs	=	self-absorption correction factor, dimensionless
F	=	volume of air sampled, cubic meters
2.22×10^6	=	the number of dis min^{-1} per microcurie (μCi)

FIGURE 2-6: LIMIT OF DETECTION RELATIONSHIPS

Correction for the Presence of Short-lived Alpha Emitters

Another potential source of analytic error that was evaluated as part of Task 5 investigations included practices associated with adjusting analytic results for the contributions of short-lived alpha emitters. The analytical technique used to count air samples collected on filter papers must take into consideration the contribution to the observed result from relatively short-lived daughters of naturally occurring radioactive materials such as radon and thoron (radon-220). A common way of accounting for these contributions is to calculate a correction factor using equations such as those derived by Koval (Koval, 1945; Moe *et al.*, 1972).

As summarized in Figure 2-7, the Koval method requires that the sample be counted twice. Results of these measurements are used in an equation that estimates the long-lived alpha activity by correcting for the short-lived activity. The Rocky Flats Plant has used the Koval method throughout its entire history of operation (Rockwell, 1976b; Boss, 1978).

In applying the Koval method, a particulate sample was counted first about 4 hours after it was removed from the sampling location. After this decay period, the contribution of the short-lived daughters of radon-222 to the alpha activity of the sample became negligible (the longest-lived daughter has a half-life of about 27 minutes). The sample was then counted again, between one day and one week later. The time between counts at Rocky Flats was normally 24 hours (Haynes, 1975). Results of both counts were then used to determine and correct for the contribution of alpha-emitting daughter products of lead-212 and calculate the concentration of total long-lived alpha activity in the sample. Samples that exceeded 0.020 pCi m^{-3} were retained for a third count to verify the presence of long-lived radioactivity (Unknown, 1985b).

Summary:	The plant routinely employed appropriate methods to account for the presence of short-lived alpha emitters in samples.

Specificity of Analysis

The analysis method for radioactive effluents from the Rocky Flats Plant has historically been counting total long-lived alpha emissions and expressing the result in terms of enriched uranium, depleted uranium, or plutonium, depending on which material was processed in the particular building sampled. Since gross alpha analysis is not specific to any radionuclide, the plant made attempts during the 1970s (once routine isotope-specific analytic methods became available) to determine the accuracy of this practice based on measurements of the actual plutonium content of releases from plutonium processing buildings. This section of the report briefly discusses the findings of these studies.

$$CLL = \frac{(A \times C_2) - (B \times C_1)}{Abs \times Eff \times F}$$

$$A = \frac{1}{1-e^{-\lambda t}}$$

$$B = \frac{e^{-\lambda t}}{1-e^{-\lambda t}}$$

where:

CLL	=	concentration of long-lived alpha activity, disintegrations/minute (dis min^{-1}) per cubic meter
C_1	=	count rate from 1st count, counts/minute (c min^{-1})
C_2	=	count rate of sample from 2nd count, c min^{-1}
λ	=	decay constant for Pb-212, .0655 $hour^{-1}$
t	=	time between C_1 and C_2 counts, hours
Abs	=	self-absorption correction, dimensionless
Eff	=	counting efficiency, c min^{-1} per dis min^{-1}
F	=	volume of air sampled, cubic meters

FIGURE 2-7: THE KOVAL EQUATION

During May 1973, particulate filter samples obtained from various plutonium buildings were counted for total long-lived alpha emissions and subsequently analyzed specifically for plutonium (Hobbs, 1973a). The observed ratio of plutonium to total long lived-alpha ranged from less than 0.01 to 1.03 with a mean value of 0.23. This ratio was intended to indicate the portion of total long-lived alpha activity that plutonium represented. If all the sampled long-lived alpha radioactivity was plutonium, the Koval method accurately subtracted the contributions of short-lived radionuclides, and the laboratory procedure used for plutonium separation was performed perfectly, the result would be expected to be very near one.

The same type of experiment was conducted during June 1973 (Hobbs, 1973b). The observed ratio of plutonium to total long-lived alpha in individual samples varied between less than 0.01 and 35. The observed ratio of plutonium to total long-lived alpha based on weekly releases ranged from 0.23 to 1.01 with a mean value for the month of 0.71. These studies clearly call into question the validity of making assumptions with regard to the source of measured gross alpha activity in effluent. However, an internal plant memo in 1975 (Hornbacher, 1975-1982) indicated that calculations made to determine the correlation between total long-lived alpha results for plutonium buildings and the results of analysis specifically for plutonium were strongly affected by the data used in the analysis. If all data were considered, the correlation was poor; however, if only samples with activity above 0.002 pCi m^{-3} were evaluated, the correlation was very good. The report suggests that errors likely occurred when emissions were low. Such errors could very well have been a result of the analytic error associated with low count rate samples.

Airborne long-lived alpha emissions for 1973 were independently calculated as part of this study. Because emissions from plutonium areas for the second half of that year were reported in terms of radiochemically determined Pu-239 (Dow, 1974a), the gross alpha and Pu-239 values for these months can be compared. When the Pu-239 value for November (<30.68 μCi) is excluded and absolute values of reported "less than" values are used, reported Pu-239 emissions average 97 percent of independently calculated gross alpha release totals over the five-month period. All monthly release totals for 1973 were reported as "less than" values. This is apparently reflection of a practice at that time to represent totals as less than values when one or more individual results were below detectability at the stated confidence level. The possible elevation of Pu-239 releases indicated by the high "less than" value for November is not reflected in the reconstructed monthly emissions, which varied between 3.9 and 6.3 μCi over the period of interest as shown in Table 2-7. When the elevated November value is included, reported Pu-239 emissions average 1.7 times the independently calculated gross alpha release totals over the six-month period.

Another plant memo in 1977 pointed out that the correlation between total long-lived alpha and plutonium measurements was erratic (Hornbacher, 1975-1982). As a result, the memo suggested the need for increased counting time in the total long-lived alpha analysis.

These observations suggest the need for some caution in making assumptions regarding the quantification of isotopes using long-lived alpha data. However, it should be recognized that they also suggest that the errors introduced were likely greatest when emissions were low, and the emissions were generally not low during the period when only long-lived alpha measurements were made. During the 1970s and 1980s, when emissions were dramatically lower, isotope-specific effluent quantification methods came into use. It should also be recognized that, in view of the similarity of the dose factors between the isotopes of plutonium and uranium, any errors in apportioning gross alpha radioactivity to specific radionuclides are not expected to be associated with significant errors in off-site dose estimates. For the purpose of this evaluation, it is estimated that the uncertainty associated with this practice is ± 20 percent.

Summary: The use of nonspecific long-lived alpha measurements and assumptions regarding the composition of the associated radioactivity represent a source of uncertainty. The importance of this source of uncertainty is believed to be small since the errors were likely smallest during periods of higher emissions when gross alpha measurements were relied on, and because of the similarity of dose factors for plutonium and uranium nuclides.

Alpha Spectral Analysis

Although alpha spectroscopy was practiced on environmental samples as early as the late 1950s or 1960 (Ray and Hammond, 1960), routine isotopic analysis of effluent sample filters did not start until around 1973. All samples submitted for specific radiochemical analysis are currently quantified using the alpha spectral analysis counting system.

In current practice, the gross alpha counting system is used for screening environmental, effluent, and room air filters to identify samples that are outside of normal ranges before submitting them to the longer process of specific radiochemical analysis. The gross alpha counting system consists of a PDP 11/10 minicomputer, two 50-channel counting systems, 100 scintillation detectors, two removable disk drives, a magnetic tape drive, and a teletype (Rockwell, 1982).

Individual airborne particulate sample filter papers are subjected to nonspecific alpha screening counts, using the methods described, in order to judge the effectiveness of effluent controls. The particulate filters are then composited by exhaust system and subjected to monthly radiochemical separation, purification, and alpha pulse-height analysis. Effluent air samples exceeding a predetermined action level are not included in the exhaust system composite; they are processed individually, by sampling period, through the chemical separation and purification process.

The chemical separation process involves dissolving the glass-fiber filters in a mixture of acids, to which are added known amounts of nonindigenous tracer radionuclide solutions that are traceable to the National Bureau of Standards and enable tracking of the chemical recovery of the analysis. Tracers used include plutonium-236, plutonium-242, uranium-232, uranium-236, americium-243, and curium-244 (Rockwell, 1989). An aliquot of the original sample is retained for backup purposes. Samples exhibiting chemical recovery below 10 percent or above 110 percent are automatically scheduled for reanalysis.

Plutonium, uranium, and americium are separated using a triisooctylamine (TIOA) solvent extraction (Campbell, 1985). The separated elemental fractions are then purified in an appropriate ion-exchange column. An aliquot of the separated and purified sample is submitted for electrodeposition on a stainless-steel planchet, which is then submitted for alpha pulse-height analysis. An aliquot of the dissolved filters is also submitted for beryllium analysis.

The alpha spectrometric analyses are performed using solid-state surface barrier detectors. The alpha spectral system consists of 128 surface barrier silicon detectors, each with an aluminum window. The detectors are connected to a Nuclear Data 6600 Analyzing System and an Environmental Data Handler (Rockwell, 1982).

Routine samples are counted for at least 16 hours; special samples may be counted for as long as a week. Detector backgrounds are counted once per month; standards traceable to the National Bureau of Standards are counted weekly for determination of detection efficiencies. Analytical or reagent blanks are included in each batch of samples for quality control purposes. The average of the last five blanks for each particular sample type and isotope is used for blank correction of the sample data (Rockwell, 1982).

While the alpha spectroscopy procedure used for particulate filter papers separated plutonium, uranium, and americium content into separate specimens for analysis, americium-241 emissions were not reported until 1985 because of problems with performance of the laboratory method. These problems were associated with the addition of plutonium-236 as a tracer in the chemical separation, which led to calculated americium recoveries that were too high. Decay products of plutonium-236 that were chemically similar to americium smeared spectra, which interfered with

efficient identification of characteristic alpha peaks and resulted in analyses of control samples that did not yield expected results (Hornbacher, 1975-1982; Bokowski, 1994).

Detection limits for radionuclides of interest via alpha spectral analysis, in terms of minimum detectable amounts per milliliter of sampled air, are as follows (Rockwell, 1989):

- Americium-241 2×10^{-11} μCi m^{-3}
- Plutonium-239, 240 5×10^{-11} μCi m^{-3}
- Uranium-233, 234, 238 8×10^{-11} μCi m^{-3}

The above values represent 1988 detection limits, however, they are typical of detection limit values throughout the 1980s.

Summary:	The introduction of alpha spectral analysis in the 1970s permitted the quantification of isotope-specific releases, but analytic difficulties prevented the accurate reporting of americium until the mid-1980s.

2.2.2.2 Analytical Practices for Tritium

Since initiation in January 1974 of routine sampling of airborne water vapor for the purposes of tritium analysis, beta radioactivity of associated samples has been quantified by liquid scintillation analysis. Airborne tritium oxide (HTO) is collected in the distilled water contained in the bubbler-type samplers historically used for routine monitoring at Rocky Flats. The impinger water sample is distilled and an aliquot is added to a commercial scintillation cocktail in a plastic vial; a portion of the distillate is retained for backup purposes (Campbell, 1985). Descriptions of the milliliter ratios of sample to cocktail vary among references as 5:27 (Rockwell, 1988), 5:5 (Rockwell, 1982), and 4:21 (Hurley, 1979). Samples are counted in a liquid scintillation analyzer, which measures the light given off when tritium's weak beta particles transfer their energy to the scintillator in solution. Count times are documented to be 10 minutes (Rockwell, 1989) and 4-20 minutes (Rockwell, 1982).

The sensitivity of the routine procedure for liquid scintillation counting corresponds to approximately 1 pCi m^{-3} of sampled air (Hurley, 1979; EG&G, 1991d). Another procedure has been used that can yield a sensitivity of 0.01 pCi m^{-3}. In that special procedure, 2 to 3 milliliters of sample are decomposed by passing over hot magnesium turnings. The resultant hydrogen is purified by passage through a palladium thimble and is counted in an evacuated proportional counter for approximately 1000 minutes (Hurley, 1979).

Calibration of the liquid scintillation counter is accomplished using a series of "quenched" standards to derive an efficiency curve (Campbell, 1985). Quenching is any reduction in the energy transfer process in the scintillation solution. With increased quenching, the beta spectrum of a standard will shift to lower energies. By counting a series of samples containing known amounts of radioactivity and varying amounts of quenching material, an efficiency curve for the analyzer is generated. By observing ratios of counts produced in various regions of spectra, a quench-indicating parameter for each sample is calculated and a counting efficiency determined.

Annual environmental reports issued by the Rocky Flats Plant since 1974 have included results of the Health and Environment Laboratories Interactive Measurement Evaluation and Control Program. That program, which is internal to Rocky Flats, involves preparation of standards containing known amounts of various analytes for measurement by normal laboratory methods for quality control purposes. The number of control analyses in this program ranged from 52 to over 1,000 per year for each analyte. Rocky Flats also participated in "crosscheck" programs in which several control samples were received each year from the USEPA and the USDOE. The Rocky Flats program summaries provide, in part, values of the relative error associated with tritium measurements by liquid scintillation analysis. These relative error values are based on numerous analyses of water samples containing known amounts of tritium by the same analytical procedures used for effluent samples. The reported relative error values can be viewed as indicators of the total error associated with the analytical portion of the effluent sampling and analysis programs and can be used to correct the sampling results.

Summary: Routine, specific analytic practices have been used to quantify tritium in airborne effluents since 1974. The method employed was sufficiently sensitive and subject to regular calibration. The procedure included the quantitation of errors, which can be used to correct analytic results.

2.3 Airborne Radioactive Effluent Data — Plutonium and Uranium

The preceding sections have provided a review of the historical sampling and analytic practices employed by the plant and potential sources of error that could result from these practices. This section provides a review of the data that were generated as a result of the airborne effluent monitoring program and again searches for potential sources of error in the reporting of the effluent data.

Several key sources of effluent data were identified early in the investigation phase of Task 5, including the Legal/Environmental File, the Environmental Master File (EMF), and the Denver Federal Records Center. These records repositories and the investigative processes applied to them are extensively discussed in the Tasks 3 and 4 report (ChemRisk, 1992). The EMF was the

source of numerous plant technical reports, internal memos, and to a lesser extent raw data that were used during Task 5. Records available at the Federal Records Center are mostly original data sheets, including raw sample data results from effluent and other air sampling activities and, to a lesser extent, effluent calculation work sheets.

In addition to these sources of information, summaries of effluent data are also presented in the Final Environmental Impact Statement (USDOE, 1980), the Omnibus Environmental Assessment for the Rocky Flats Plant (USERDA, 1975), and the USDOE Effluent Information System, which is a computerized database that is currently operated by EG&G Idaho, Inc. for DOE. These effluent data were intensively examined for the purpose of establishing radioactive contaminant release estimates.

The purpose of the effluent data review was to determine the accuracy of summary-level effluent data reported by the plant. If the basis for the compilation of the summaries was not clear or was shown to be unreliable or inaccurate, then additional emphasis would be placed on obtaining and reviewing raw data for the relevant period or periods of concern.

Investigations at the Rocky Flats Plant and the Denver Federal Records Center resulted in identification of many of the historical monitoring raw data records. These records, in combination with information obtained in interviews of active and retired Rocky Flats Plant personnel, were used as the basis for the evaluation of Rocky Flats radioactive effluent reporting practices in this report.

Initial efforts to review the effluent sampling data were aimed at establishing the feasibility of reconstructing summary-level airborne emission estimates from raw data records retained by the plant, since it was not clear whether the necessary records were available to perform such a task. Once it was established that data reconstruction was feasible, the review program was expanded to cover a range of time periods and all measured radioactive effluents for the selected periods. The results of initial reconstruction efforts and the subsequent expanded review are described in the following sections.

2.3.1 Initial Reconstruction of Summary-Level Effluent Data — Plutonium and Uranium

Based on initial reviews of available records, it became clear that the Final Environmental Impact Statement represented one of the most comprehensive compilations of airborne emissions reported by the plant for the period from the early 1950s through the 1970s. During the independent review of raw data, investigators were able to trace much of the process that was used to compile the data to produce the effluent summaries found in the Final Environmental Impact Statement, which was to a large part based on work performed for the Omnibus Environmental Assessment (USERDA, 1975).

In order to evaluate the accuracy of reported summaries of Rocky Flats effluent data, several periods of time were selected for complete reconstruction of release totals from raw data when available, or data from as close to the raw data level as feasible. Methods and results of these activities are described in this section.

In order to gain further understanding of the methods the Rocky Flats Plant used to compile effluent data and to perform a preliminary evaluation of the accuracy of effluent summary documents, three initial reconstructions of effluent data were performed. Initial reconstructions were performed for the following effluent monitoring data sets:

- Plutonium effluents for 1963
- Uranium effluents for 1969
- Plutonium effluents for 1973

The initial reconstruction efforts required the use of estimates of ventilation exhaust flow rates to calculate monthly and annual releases. Throughout the following reconstruction discussions, flow rate estimates are often required since the effluent data used were in the form of raw filter paper analysis results. These raw results were simply gross long-lived alpha radioactivity observed on the collected sample divided by the total sample volume, that is, disintegrations per minute (dis min^{-1}) of long-lived alpha activity per cubic meter of air passed through the filter. Table 2-2 presents the ventilation exhaust flow rate data used in the reconstructions, as well as the reference from which each estimate was derived.

Plutonium effluents for 1963 were reconstructed based upon archived records that contained the results of analyses of daily effluent samples from Buildings 771 and 774 (Dow, 1963a). The monthly average concentration of plutonium was calculated from these data for each building in dis min^{-1} per cubic meter. Building 774 did not have its own ventilation system until October 23, 1963. Ventilation exhaust flow rates were estimated from 1973 summary data (Dow, 1973a). The monthly average effluent concentrations were multiplied by the flow rate estimates and summed to yield an annual plutonium release estimate.

Uranium effluents for 1969 were reconstructed based upon archived records that contained the monthly data summaries of uranium releases from Buildings 331, 444, 447, 881, 883, 886, and 889 (Dow, 1969a). These monthly summary data were used to calculate total releases by building by stack for the year, and these results were summed to yield an annual uranium release estimate for 1969.

TABLE 2-2

EXHAUST FLOW RATES USED TO RECONSTRUCT EMISSIONS

Building	Duct	Average Volumetric Flow Rate ($m^3\ d^{-1}$)	Reference
331	-	0.19×10^6	Dow (1969c)
444	2	4.9×10^6	Dow (1969c)
444	3	2.9×10^6	Dow (1969c)
447	-	2.6×10^6	Dow (1969c)
447	-	1.0×10^6	Dow (1973b)
559	Small	0.088×10^6	Dow (1973a)
559	Large	1.0×10^6	Dow (1973a)
771	Main	7.1×10^6	Dow (1969b)
771	Main	7.1×10^6	Dow (1973a)
774	-	0.59×10^6	Dow (1969b)
774	-	0.58×10^6	Dow (1973a)
776/777	Main	7.8×10^6	Dow (1973a)
776	Booster 1	0.70×10^6	Dow (1973a)
776	Booster 2	0.32×10^6	Dow (1973a)
776	Booster 3	0.32×10^6	Assumption
779-779A	-	0.63×10^6	Dow (1973a)
865	E & W	2.1×10^6 Total	Dow (1973b)
881[1]	1,2,3	8.5×10^6 Total	Dow (1969c)
881[1]	1,2,3	8.7×10^6 Total	Dow (1973b)
881[1,2]	4	2.9×10^6	Boss (1983)
881[1]	5	1.1×10^6	Dow (1969c)
881[1]	5 & 5A	0.97×10^6 Total	Dow (1973b)
883	A	4.6×10^6	Dow (1969c)
883	A	2.3×10^6	Dow (1973b)
883	B	4.6×10^6	Dow (1969c)
883	B	1.1×10^6	Dow (1973b)
886	-	0.17×10^6	Dow (1973b)
991	-	0.37×10^6	Dow (1973b)

1 Building 881 enriched uranium operation ceased in 1962. It is possible that ventilation exhaust flow rates were higher in 1957 than estimated by 1969 and 1973 data.

2 The flow rate estimate for Building 881 Duct 4 assumes the flow rate in Duct 4 was equivalent to that of Duct 1, 2 or 3 (based on reference Boss, 1983). The Duct 1, 2, and 3 total flow rate was therefore divided by 3 to estimate Duct 4 flow.

Plutonium effluents for 1973 were reconstructed based upon archived records that contained the monthly data summaries of plutonium releases from Buildings 559, 707, 771, 774, 776, 777 and 779 (Dow, 1973a). These monthly summary data were used to calculate total releases by building by stack for the year, and these results were summed to yield an annual plutonium release estimate for 1973.

The results of these initial reconstructions are presented in Table 2-3. Included in this table are the corresponding reported releases from the Rocky Flats Plant Final Environmental Impact Statement (USDOE, 1980). This initial comparison suggested good agreement between the reconstructed releases and reported releases and provided evidence that the summary data presented in the Final Environmental Impact Statement had been reconstructed using methods very similar to those employed in the initial reconstructions performed for this study.

Based on the initial reconstructions, the following course of action was adopted:

- Perform complete effluent data reconstructions for the initial test years chosen (1963, 1969, and 1973) based on available gross alpha activity data.

- Perform reconstructions of additional test years: 1957, 1984, and 1986 were chosen. The year 1957 was chosen because it represented both a year of relatively significant routine effluents and because it was the year of a serious fire in Building 771. Calendar years 1984 and 1986 were selected to represent years in which isotope-specific analyses were used to quantify airborne emissions.

If the results of these reconstructions were consistent with the summary data, then summary data would be considered valid for use in defining airborne radioactive source terms for the facility for the purpose of dose reconstruction. Appropriate uncertainty estimates would then be applied to the summary data for use in estimating off-site exposure to these contaminants.

2.3.2 Expanded Reconstruction of Effluent Data — Plutonium and Uranium

Complete summary-level data reconstructions were performed for each of the test years for plutonium and uranium. The reconstruction process that was used is described in this section. Results of the reconstructions are presented with two significant figures.

TABLE 2-3

INITIAL AIRBORNE EMISSION RECONSTRUCTION RESULTS

	Total Alpha-Emitting Radioactivity Released (μCi)	
	Reported in DOE's 1980 Final Environmental Impact Statement	**Results of Initial ChemRisk Reconstructions**
1963 Plutonium Areas	3,900	3,500
1969 Uranium Areas	220	220
1973 Plutonium Areas	77	62

2.3.2.1 Plutonium Emissions

The following discussions summarize the process that was employed to reconstruct the summary-level airborne emission data for plutonium released from the Rocky Flats Plant during 1957, 1963, 1969, 1973, 1984, and 1986.

1957 Reconstruction

Records of plutonium effluent measurements from 1957 were found at the Rocky Flats Legal/Environmental File (Ray, 1957). Plutonium operations were limited exclusively to Building 771 in 1957; therefore, all gross alpha results from effluent samples in this building were attributed to plutonium effluents. As discussed earlier, these data are based on samples collected from plutonium building exhaust systems and analyzed by gross alpha scintillation counting. Alpha spectrometric (radionuclide specific) analytical techniques were not in use at this time.

Plutonium records were in the form of Rocky Flats Plant internal monthly Health Physics reports for Building 771 (Ray, 1957). These reports contained the average and maximum concentrations (gross alpha) based on the effluent samples, as well as the calculated mass of material released from the building in micrograms (μg).

Results of plutonium analyses in 1957 were expressed in units of disintegrations per minute per cubic meter (dis min^{-1} m^{-3}). These results were averaged over each month and this average was combined with flow rate data and a specific activity of 0.0613 μCi μg^{-1} (USDHEW, 1970) to calculate the total micrograms of material released. Results listed in the monthly reports for September are for before the fire on the 11th and after restart of the sampling system on the 19th.

The results of the reconstruction of plutonium emissions for 1957 are presented in Table 2-4. The review of the raw data suggests that the releases of plutonium in October 1957 were quite high, and that the data were inconsistent with the Final Environmental Impact Statement summary. As a result, the data were further analyzed by back-calculating the average vent flow volumes that would have been required to produce the stated release. The back-calculated monthly-average vent flow volumes for 1957 are listed in Table 2-4 along with monthly plutonium emission totals.

The required monthly-average ventilation flow volume back-calculated from the reported results for the month of October was 4.8×10^9 m^3, which is significantly higher than the average flows determined for other months. This result could be explained by a number of alternatives, such as; 1) the building ventilation was dramatically increased, 2) a calculational error was made in

TABLE 2-4

SUMMARY OF 1957 AIRBORNE PLUTONIUM EMISSIONS FOR BUILDING 771

Month	Total Long-Lived Alpha-Emitting Radioactivity Released (μg)	Average Radioactivity Concentration (dis min^{-1} m^{-3})	Maximum Radioactivity Concentration (dis min^{-1} m^{-3})	Corresponding Back-Calculated Exhaust Volume ($m^3 \times 10^8$)
January	610	0.36	3.0	2.3
February	110	0.082	1.1	1.8
March	630	0.46	14	1.9
April	110	0.078	0.94	2.0
May	73	0.043	0.35	2.3
June	9,400	6.5	170	2.0
July	670	0.40	2.0	2.3
August	140	0.09	0.40	2.1
September:[1] *Before fire* *After fire*	 Not Available Not Available	 3.7 75	 15 2,100	 Not Available Not Available
October	230,000	6.6	34	48
November	830	0.50	1.5	2.3
December	3,500	2.0	15	2.4
Total	250,000[2] (15,000 μCi)			

NOTES:

Source of Raw Data: Ray, 1957.

1 Results listed for September are for before the September 11th fire and after the September 19th restart of the sampling system.

2 Includes no release data from September. Emissions from the 1957 fire are characterized separately as part of Task 6.

October, or 3) ventilation flow rates and contaminant concentrations varied widely over the month, making the use of average values for the measurements to reconstruct the monthly release highly inaccurate. Building ventilation may have been increased to reduce the high airborne contamination levels resulting from the fire. However, based on interviews with presently employed and past Rocky Flats Plant personnel (Coles, 1991; Putzier, 1991), ventilation flow rates of nearly 20-times the norm were impossible to achieve, therefore the stated results would more likely represent a calculational error.

It is even more likely that exhaust flow rates were highly variable during the time period following the fire, with the ventilation fans potentially being turned down during some periods to accommodate some cleanup efforts, or turned up to high speeds at other times. At the same time, radioactivity release rates could also have varied widely in the post-fire period. Given the relatively large difference between the average measured radioactivity and the maximum reported radioactivity, variations in the ventilation flow rates could significantly affect the calculated release total. The total release of 230,000 micrograms reported in the Health Physics Report for Building 771 was chosen for use as the October release value.

The total emission of plutonium for 1957 of 250,000 μg presented in Table 2-4 does not include any release data from September when the fire rendered the sampling system inoperable. The reconstructed release estimate, equivalent to 15,000 μCi, is considerably higher than the 1,595 μCi attributed to routine emissions in 1957 in the Final Environmental Impact Statement (USDOE, 1980). One explanation for this rather large discrepancy may be that, in calculating the October 1957 release for the FEIS, plutonium releases were bounded by assuming that the maximum flow rate observed throughout the year (2.4×10^8 m^3 per month) occurred during October. The plutonium releases from normal operations in October calculated by multiplying the average concentration for the month by this assumed maximum flow rate, that is, 6.6 dis min^{-1} m^{-3} times 2.4×10^8 m^3, correspond to a release total of 720 μCi. If the plutonium release estimate obtained using the assumed maximum flow rate for October (720 μCi) is used, the routine release total for 1957 becomes 1,700 μCi, which agrees well with the Final Environmental Impact Statement.

1963 Reconstruction

Records of plutonium in airborne effluents from Rocky Flats during 1963 were located in the Denver Federal Records Center (Dow, 1963a-l). Plutonium records were found for Buildings 771, 774, and 776/777. These records were in the form of daily typewritten reports titled "Above MPC Air Sample Results" for each building. These reports contained the analytic results for work room air samples that were above the maximum permissible concentration and the daily stack sample results. The results were reported in dis min^{-1} m^{-3}. The results from the daily reports were averaged over each month and then for the year; results are shown in Table 2-5.

The formula used to convert the results from dis min^{-1} m^{-3} to μCi is also presented in Table 2-5. The results of the reconstruction indicate that 3600 μCi of plutonium were released from Rocky Flats during 1963, compared to the Final Environmental Impact Statement reported value of 3,903 μCi. The difference in estimates is relatively small (about 8 percent) and most likely the result of the use of different exhaust flow rate assumptions. As discussed earlier, exhaust volumetric flow rates for the early years of plant operation are poorly documented, and it was necessary to assume the exhaust flow rates were the same as those documented in the 1960s and 1970s.

1969 Reconstruction

Both raw data and compiled data for plutonium were located in the Denver Federal Records Center for calendar year 1969. The compiled data were in the form of monthly work sheets for Buildings 559, 771, 774, 776/777, and 779. Approximately 5 percent of the results reported on the work sheets were spot-checked against the values presented on the raw data sheets. Through this spot-check, it was determined that the compiled data and the raw data were in agreement; therefore, the compiled data were used for 1969 release data reconstruction. The reconstruction of 1969 effluent data was conducted in a manner similar to that described above for 1963 plutonium data. Results of the reconstruction of plutonium effluent data are presented in Table 2-6. These results indicate that 1,400 μCi of plutonium were released from Rocky Flats during 1969, compared to the Final Environmental Impact Statement reported release of 1,660 μCi. The difference between the estimate from the raw data reconstruction and the Final Environmental Impact Statement estimate is about 16 percent.

1973 Reconstruction

Compiled data for plutonium in Rocky Flats airborne effluents during 1973 were located in the Denver Federal Records Center. The results were reported as microcuries of plutonium released from each building by month. Compiled plutonium data were located for Buildings 559, 707, 771, 774, 776/777, and 779. In addition, raw data were located for Buildings 559, 776/777, 778, and 779. The existence of raw data enabled spot-checking of approximately 10 percent of the compiled data for accuracy. Through this spot-checking, it was determined that the compiled data for these buildings were accurate. The reconstruction results for the plutonium buildings are presented in Table 2-7. The absolute values of the "less than" values presented in Table 2-7 were used in the summation of yearly totals. The data reconstruction resulted in a release estimate of 62 μCi of plutonium during 1973, which is approximately 19 percent less than the 77 μCi plutonium release reported in the Final Environmental Impact Statement.

TABLE 2-5

SUMMARY OF 1963 AIRBORNE PLUTONIUM EMISSIONS

Month	Average Daily Concentration of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})		
	Building 771 Main Duct	Building 774 Stack	Buildings 776/777 Main Duct
January	7.0	*	0.073
February	8.3	*	0.073
March	3.6	*	0.10
April	5.9	*	0.083
May	4.3	*	0.13
June	1.1	*	0.069
July	2.7	*	0.064
August	1.5	*	0.022
September	0.65	*	0.067
October	0.61	0.025	0.030
November	0.14	0.070	0.018
December	0.22	0.13	0.010
1963 Average Daily Concentration	3.0	0.019	0.062
Release Totals (μCi)	3500	1.8	80
Grand Total (μCi)	3600		

NOTES:

Sources of Raw Data: Dow, 1963b and c.

* Building 774 sampling commenced October 23, 1963.

Values are calculated means of raw data points from air sample results log (Dow, 1963c).

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

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TABLE 2-6

SUMMARY OF 1969 AIRBORNE PLUTONIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})				
	Building 771 Ducts			Building 559 Ducts	
	Main 1	Main 2	Main 3	Large	Small
January	0.29	0.41	0.35	0.0071	0.028
February	0.71	0.59	0.77	0.012	0.030
March	0.19	0.19	0.25	0.022	0.066
April	0.36	1.1	0.71	0.021	0.024
May	0.33	0.35	0.44	0.023	0.029
June	0.54	0.53	0.47	0.016	0.085
July	0.38	0.43	0.50	0.023	0.023
August	0.25	0.24	0.32	0.0082	0.022
September	0.21	0.30	0.31	0.015	0.021
October	0.18	0.27	0.26	0.0076	0.021
November	0.35	0.43	0.34	0.0069	0.0038
December	0.29	0.24	0.34	0.011	0.0088
Total	4.1	5.1	5.1	0.17	0.36
1969 Average Daily Concentration	0.34	0.43	0.43	0.014	0.030
Total μCi Released	470			2.3	0.43

NOTES:

Sources of Raw Data: Dow, 1969a and b

All results are calculated means of raw data points from referenced air sample results log.

Building 771 release is based on average concentration in Main 1, 2, and 3 Ducts (0.40 dis min^{-1} m^{-3}).

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

(continued on next page)

TABLE 2-6 (Continued)

SUMMARY OF 1969 AIRBORNE PLUTONIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters in Building 776 Ducts (dis min^{-1} m^{-3})			
	Main	Booster 1	Booster 2	Booster 3
January	0.012	0.026	4.8	N/A
February	0.014	0.022	0.10	N/A
March	0.008	0.008	0.37	N/A
April	0.007	0.011	0.36	N/A
May	1.4	0.021	0.17	N/A
June	1.0	N/A	N/A	N/A
July	3.4	N/A	4.1	N/A
August	1.7	N/A	1.3	N/A
September	0.12	N/A	0.098	0.35
October	0.079	N/A	0.15	0.23
November	0.29	N/A	0.25	0.36
December	0.14	N/A	0.046	0.36
Total	8.2	0.083	12	1.4
1969 Average Daily Concentration	0.68	0.017	1.1	0.34
Total μCi Released	870	0.86	58	6.0

NOTES:

Sources of Raw Data: Dow, 1969a and b

All results are calculated means of raw data points from referenced air sample results log.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

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TABLE 2-6 (Continued)

SUMMARY OF 1969 AIRBORNE PLUTONIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters in Building 774 Ducts (dis min^{-1} m^{-3})			
	Stack	Booster 1	Booster 2	Water
January	0.034	0.050	0.13	0.16
February	0.014	0.16	0.16	0.12
March	0.028	0.047	0.25	0.11
April	0.062	0.15	0.31	0.19
May	0.097	0.099	0.15	0.099
June	0.074	0.11	0.15	0.058
July	0.033	0.072	0.17	0.11
August	0.068	0.021	0.088	0.24
September	0.043	0.093	0.060	0.95
October	0.018	0.11	0.078	0.45
November	0.034	0.30	0.23	0.17
December	0.025	0.044	0.064	0.34
Total	0.53	1.3	1.8	3.0
1969 Average Daily Concentration	0.044	0.11	0.15	0.25
Total Building 774 Release (μCi)	13			
1969 Release Total (μCi)	1400			

NOTES:

Sources of Raw Data: Dow, 1969a and b

All results are calculated means of raw data points from referenced air sample results log.

Building 774 release is based on average in Stack, B1, B2, and Water (0.14 dis min^{-1} m^{-3}).

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-7

SUMMARY OF 1973 AIRBORNE PLUTONIUM EMISSIONS

Month	Total Long-Lived Alpha-Emitting Radioactivity Released (μCi)							
	Buildings 776/777	Building 559	Building 778	Building 771	Building 774	Building 771A	Building 779	Building 707
January	8.5	0.14	0.018	2.4	0.29	0.042	0.03	0.29
February	1.6	0.096	0.012	1.2	0.17	0.043	0.016	0.24
March	0.75	0.082	0.019	1.3	0.096	0.032	0.024	0.16
April	2.7	0.040	0.014	1.1	0.10	0.049	0.069	0.23
May	1.2	0.063	0.025	1.2	0.07	0.035	0.029	0.20
June	0.62	0.14	0.035	0.8	0.089	0.025	7.0	0.35
July	0.76	0.079	0.027	2.1	1.5	0.033	0.049	0.16
August	1.8	0.091	0.021	1.7	0.16	0.02	1.6	0.17
September	2.3	0.062	0.023	1.2	0.19	0.037	0.03	0.19
October	1.4	0.031	0.027	1.8	<0.23	0.052	0.12	0.30
November	<3.0	<0.085	<0.032	<2.1	<0.35	<0.054	<0.24	<0.088
December	<1.3	<0.094	<0.021	<1.7	<0.28	<0.057	<0.33	<0.14
1973 Release Totals (μCi)	26	1.0	0.27	19	3.5	0.48	9.5	2.5
Grand Total Release (μCi)	62							

Reference: Dow, 1973a

A possible explanation for the difference between the results may be the way in which the "less than" values were handled by plant personnel. Information collected during this investigation does not indicate how less than values were used by plant personnel in generating emission totals.

2.3.2.2 Uranium Emissions

The following discussions summarize the data reconstruction process that was employed to reconstruct the summary-level airborne emission data for enriched uranium released during 1957, 1963, 1969, 1973, 1984, and 1986.

1957 Reconstruction

Records of uranium effluent measurements were found in the Denver Federal Records Center (Dow, 1957a,b). Enriched uranium operations were ongoing in Building 881 and a portion of Building 883; hence, all gross alpha results from effluent samples collected in these buildings were attributed to enriched uranium effluents. Depleted uranium operations were ongoing in Buildings 444, 447, and a portion of Building 883; hence, all gross alpha results from effluent samples collected in these buildings were attributed to depleted uranium. Uranium records consisted of analytical result log sheets for the uranium buildings in 1957. The observed activity concentration, in dis min^{-1} per cubic meter of sampled air, was reported on the log sheets for samples collected from effluent ducts and stacks during 1957. Depleted and enriched uranium results in these records were averaged for each month and duct. Monthly results were then averaged and the release total calculated using flow rate data from Table 2-2.

Records indicate that Building 881 Duct 5 commenced operation on July 30, 1957 and that Building 883 Ducts A and B commenced operation on August 12, 1957. All other uranium discharge ducts appeared to be in operation for the entire year. Results of these calculations, including total uranium effluents for each duct for 1957, are presented in Tables 2-8 and 2-9.

Results indicate that approximately 730 μCi of depleted uranium were released from the Rocky Flats Plant in 1957, compared to a documented release of 38 μCi (USDOE, 1980). The reconstructed release estimate, while not a large fraction of total activity released during the history of Rocky Flats operations, was more than twenty times larger than that reported in the Final Environmental Impact Statement for 1957. This discrepancy led to detailed investigations that ultimately determined that data summary activities conducted by the plant for the purposes of the Omnibus Environmental Assessment for the Rocky Flats Plant and the Final Environmental Impact Statement did not include the contributions of all buildings that processed depleted uranium during the early years of operation. The documentation of calculations performed by plant personnel in support of the data summary effort suggest that the preparers

TABLE 2-8

SUMMARY OF 1957 AIRBORNE DEPLETED URANIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})			
	Building 883	Building 444		Building 447
	Duct A	Duct 2	Duct 3	
January	*	0.024	0.23	0.073
February	*	0.021	0.36	0.092
March	*	0.012	0.29	0.11
April	*	0.067	0.49	0.040
May	*	0.060	0.23	0.14
June	*	0.018	0.35	0.15
July	*	0.060	1.0	0.15
August	0.25	0.11	0.34	8.2
September	0.24	0.069	0.15	0.11
October	0.37	0.13	0.43	1.9
November	0.45	0.072	0.37	0.092
December	0.38	0.17	0.34	0.048
1957 Average Daily Concentration	0.13	0.068	0.38	0.93
Release Totals (μCi)	98	55	180	400
Grand Total (μCi)	730			

NOTES:

Sources of Raw Data: Dow, 1957a and b.

* Building 883 sampling commenced August 12, 1957.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-9

SUMMARY OF 1957 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})					
	Building 883	Building 881				
	Duct B	Duct 1	Duct 2	Duct 3	Duct 4	Duct 5
January	*	0.077	0.12	0.064	0.17	**
February	*	0.062	0.080	0.057	0.088	**
March	*	0.065	0.074	0.040	0.071	**
April	*	0.089	0.15	0.048	0.054	**
May	*	0.064	0.13	0.078	0.045	**
June	*	0.16	0.22	0.12	0.094	**
July	*	0.096	0.15	0.087	0.14	0.073
August	0.49	0.083	0.12	0.091	0.10	0.13
September	0.35	0.11	0.12	0.093	0.088	0.15
October	0.45	0.95	0.13	0.13	0.15	0.19
November	0.39	0.11	0.11	0.16	0.13	0.16
December	0.45	0.13	0.10	0.14	0.15	0.17
Total	2.1	2.0	1.5	1.1	1.3	0.87
1957 Average Daily Concentration	0.16	0.17	0.12	0.092	0.11	0.073
Release Totals (μCi)	120	79	56	43	52	13
Grand Total (μCi)	360					

NOTES:

Source of Raw Data: Dow, 1963h.
* Building 883 sampling commenced August 12, 1957.
** Building 881 Duct 5 sampling commenced in July 1957.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

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erroneously believed that effluent sample data were not available for Buildings 444, 447, and duct A of Building 883 prior to 1961. As a result, the summary data reconstruction activities for depleted uranium were extended to include all the years prior to 1961.

The results indicate that approximately 360 μCi of enriched uranium were released from Rocky Flats in 1957. This was slightly more than 1.5 times the reported release of 230 μCi. Investigation revealed that the preparers of the early data summaries were not aware of enriched uranium effluent monitoring results for Building 881 for the period from 1954 through 1956. As a result, the independent reconstruction of summary data for enriched uranium emissions from Building 881 was expanded to include the years 1954 through 1956.

Pre-1961 Reconstruction

Enriched uranium emissions for years of Rocky Flats operation prior to 1957 and depleted uranium emissions for years before 1961 were reconstructed using raw data found in the Denver Federal Records Center. The data reconstruction is presented for both enriched and depleted uranium.

Enriched Uranium — Logbooks on file at the Denver Federal Records Center contain radionuclide release results, assumed to be enriched uranium, for Building 881 for the time period 1954-1956 (Dow, 1963h) that were not reported in the Final Environmental Impact Statement. Despite an extensive search, effluent monitoring data for 1953 were not located. Building 881 did not start operations until the latter part of 1953 (ChemRisk, 1991c, Interview No. 39), and, additionally, the enriched uranium operations were limited and primarily involved equipment testing and personnel training. Given the limited nature of the 1953 enriched uranium operations, it is likely that the operational releases did not exceed those reported for 1954. Therefore, the 1953 release total is assumed to be the same as the 1954 release total. The reconstruction required the entry of all data contained in the logbooks into a spreadsheet program. The data were then reduced to monthly averages by duct, and yearly averages were calculated. The 1954, 1955, and 1956 yearly average enriched uranium concentrations in airborne effluents for Ducts 1, 2, 3, and 4 are presented in Tables 2-10, 2-11, and 2-12.

The total release for each year was then calculated using the following equation:

$$\text{Release } (\mu\text{Ci}) = \frac{(\text{Average concentration, dis min}^{-1}\ \text{m}^{-3}) \times (\text{Exhaust flow rate, m}^3\ \text{d}^{-1}) \times (365\ \text{d y}^{-1})}{2.22 \times 10^6\ \text{dis min}^{-1}\ \mu\text{Ci}^{-1}}$$

Actual ventilation flow rates for this time period could not be located. Therefore, exhaust flow rate data from Table 2-2 were again used in the reconstruction.

TABLE 2-10

SUMMARY OF 1954 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})			
	Building 881 Duct 1	Building 881 Duct 2	Building 881 Duct 3	Building 881 Duct 4
January	*	*	*	*
February	*	*	*	*
March	*	*	*	*
April	*	*	*	*
May	*	*	*	*
June	0.031	0.0040	0.032	0.0080
July	0.012	0.0060	0.0020	0.010
August	0.0050	0.013	0.011	0.0070
September	0.0020	0.0070	0.013	0.016
October	0.0080	0.0060	0.0005	0.0080
November	0.0070	0.0020	0.0015	0.0050
December	0.0020	0.0020	0.0020	0.0050
1954 Average Daily Concentration	0.0056	0.0033	0.0052	0.0049
Release Totals (μCi)	2.7	1.6	2.5	2.3
Grand Total (μCi)	9.1			

NOTES:

Source of Raw Data: Dow, 1963h.

* No data were located for this time period.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-11

SUMMARY OF 1955 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})			
	Building 881 Duct 1	Building 881 Duct 2	Building 881 Duct 3	Building 881 Duct 4
January	0.0020	0.014	0.002	0.0060
February	0.010	0.037	0.044	0.046
March	0.0080	0.11	0.044	0.024
April	0.0090	0.011	0.021	0.023
May	0.020	0.033	0.040	0.022
June	0.022	0.012	0.013	0.011
July	0.014	0.020	0.036	0.0050
August	0.16	0.21	0.18	0.098
September	0.018	0.033	0.10	0.046
October	0.031	0.026	0.066	0.032
November	0.016	0.025	0.054	0.019
December	0.018	0.016	0.039	0.026
1955 Average Daily Concentration	0.028	0.046	0.053	0.03
Release Totals (μCi)	13	22	25	14
Grand Total (μCi)	74			

NOTES:

Source of Raw Data: Dow, 1963h.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-12

SUMMARY OF 1956 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})			
	Building 881 Duct 1	Building 881 Duct 2	Building 881 Duct 3	Building 881 Duct 4
January (excluding January 24th)	0.015	0.079	0.065	0.028
February	0.055	0.040	0.055	0.023
March	0.061	0.22	0.11	0.085
April	0.12	0.12	0.21	0.14
May	0.13	0.24	0.10	0.15
June	0.10	0.18	0.093	0.14
July	0.063	0.23	0.16	0.22
August	0.11	0.11	0.049	0.11
September	0.086	0.16	0.087	0.13
October	0.067	0.14	0.053	0.14
November	0.064	0.22	0.075	0.16
December	0.052	0.15	0.069	0.12
Average Daily Concentration (excluding January 24th)	0.077	0.16	0.092	0.12
Total Released (μCi) (excluding January 24th)	37	76	44	57
Average Daily Concentration on January 24, 1956	540	130	6.5	25
Total Released (μCi) on January 24th, 1956	700	170	8.5	33
1956 Release Totals (μCi)	740	250	53	90
Grand Total (μCi)	1100			

NOTES: Source of Raw Data: Dow, 1963h.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

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The results of the reconstruction for 1954-1956 are presented in Table 2-13. This table also presents a comparison of the data to values reported by the Rocky Flats Plant in the Final Environmental Impact Statement, which reported no enriched uranium releases for each of the years during this time period.

The 1956 release is approximately 20 percent of the 1953-1989 total release of enriched uranium reported by the plant. It should be noted that the majority of this release (910 μCi) occurred on a single day, January 24, 1956 (See Table 2-12). An effort was made to find an explanation for this apparent anomaly; however, there was no documentation or individual that could provide an explanation for this relatively large release of enriched uranium. The elevated release estimate could not be readily attributed to a simple calculational or reporting error, since elevated emissions were noted in all sampled ducts for Building 881, and this elevated data set on the log sheet was surrounded by data for other sampling periods with values that are many orders of magnitude smaller. At this point in time, it must be assumed that these documented releases in 1956 are accurate; however, no explanation for the magnitude of the reported release has been offered.

Depleted Uranium - The depleted uranium effluent releases estimates presented in the Final Environmental Impact Statement did not include data from Buildings 444 and 447 for the time period 1954 through August 1960. The effluent sampling data from 1954-1960 were located at the Denver Federal Records Center and used to reconstruct the effluent releases from Buildings 444 and 447. The data were in the form of air sample logbooks, and results were reported in dis min^{-1} m^{-3}. These data were reduced in a manner similar to the data reduction techniques described for enriched uranium. Tables 2-14 through 2-19 present the yearly averages calculated for Buildings 444 and 447. These data were further reduced to provide release estimates in microcuries using the exhaust flow rate data in Table 2-2.

Table 2-20 presents the results of the depleted uranium effluent reconstruction for 1954 to 1960 and provides a comparison to the values reported in the Final Environmental Impact Statement. Based on the reconstructed emissions of depleted uranium for the period of 1954-1960, it appears that significantly larger amounts of this material were emitted by the plant during this period than was previously reported.

1963 Reconstruction

Records of enriched and depleted uranium in airborne effluents from Rocky Flats during 1963 were located in the Denver Federal Records Center (Dow, 1963a-l). Like the 1957 data, uranium records for 1963 were in the form of log sheets of analytical results for the uranium buildings. These records for each month were averaged by duct and by building. The yearly average for each building was calculated, and the total activity released was calculated using

TABLE 2-13

AIRBORNE ENRICHED URANIUM RELEASE ESTIMATES, 1954-1956

Year	Raw Data Reconstruction (μCi)	FEIS Data*
1954	9.1	None Reported
1955	74	None Reported
1956	1,100	None Reported

* Final Environmental Impact Statement (USDOE, 1980)

TABLE 2-14

SUMMARY OF 1954 AIRBORNE DEPLETED URANIUM EMISSIONS FROM ROCKY FLATS BUILDING 444

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})	
	Building 444 Duct 2	Building 444 Duct 3
January	0.0068	2.1
February	0.031	0.79
March	0.22	1.1
April	0.19	1.5
May	0.10	1.8
June	0.19	1.7
July	0.24	1.9
August	0.18	3.9
September	0.38	2.5
October	0.12	4.4
November	0.15	5.6
December	0.077	2.1
1954 Average Daily Concentration	0.16	2.4
Total Released (μCi)	130	1100

NOTES:

Sources of Raw Data: Dow, 1954-1956.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-15

SUMMARY OF 1955 AIRBORNE DEPLETED URANIUM EMISSIONS FROM ROCKY FLATS BUILDING 444

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})	
	Building 444 Duct 2	Building 444 Duct 3
January	0.13	2.1
February	0.24	6.0
March	0.31	2.2
April	0.39	4.0
May	0.32	2.7
June	0.35	3.4
July	0.26	3.6
August	0.080	2.3
September	0.11	3.4
October	0.66	4.5
November	0.68	6.4
December	0.22	4.3
1955 Average Daily Concentration	0.32	3.8
Total Released (μCi)	260	1800

NOTES:

Sources of Raw Data: Dow, 1954-1956.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-16

SUMMARY OF 1956 AIRBORNE DEPLETED URANIUM EMISSIONS FROM ROCKY FLATS BUILDINGS 444 AND 447

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})		
	Building 444		Building 447
	Duct 2	Duct 3	
January	0.25	1.9	*
February	0.19	3.6	*
March	0.32	1.6	*
April	0.16	1.4	*
May	0.09	1.1	*
June	0.19	1.4	*
July	0.24	1.8	*
August	0.59	1.8	*
September	0.29	1.3	*
October	0.55	1.4	0.2
November	0.28	2.2	0.082
December	**	**	0.13
1956 Average Daily Concentration	0.26	1.7	0.035
Total Released (μCi)	210	810	15

NOTES:

Sources of Raw Data: Dow, 1954-1956.

* Building 447 monitoring apparently commenced in October 1956.

** Data were not located for this time period.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

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TABLE 2-17

SUMMARY OF 1958 AIRBORNE DEPLETED URANIUM EMISSIONS FROM ROCKY FLATS BUILDINGS 444 AND 447

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})		
	Building 444		Building 447
	Duct 2	Duct 3	
January	0.055	0.18	0.057
February	0.055	0.28	0.040
March	0.088	0.29	0.50
April	0.043	0.15	0.088
May	0.014	0.21	3.1
June	0.19	0.29	0.19
July	17	0.38	0.44
August	0.050	0.27	0.46
September	0.038	0.35	0.028
October	0.11	0.39	0.04
November	0.20	0.30	0.065
December	0.35	0.38	0.070
1958 Average Daily Concentration	1.5	0.29	0.42
Total Released (μCi)	1200	140	120

NOTES:

Source of Raw Data: Dow, 1963h.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-18

SUMMARY OF 1959 AIRBORNE DEPLETED URANIUM EMISSIONS FROM ROCKY FLATS BUILDINGS 444 AND 447

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})		
	Building 444		Building 447
	Duct 2	Duct 3	
January	0.20	0.34	0.054
February	0.45	0.33	0.045
March	0.031	0.38	0.033
April	0.028	0.30	0.035
May	0.045	0.20	0.035
June	0.0065	0.33	0.025
July	0.013	0.34	0.036
August	0.11	0.20	0.036
September	0.046	0.30	0.031
October	0.068	0.26	0.064
November	0.093	0.25	0.038
December	0.034	0.23	0.051
Total	1.4	3.4	0.48
1959 Average Daily Concentration	0.12	0.28	0.040
Total Released (μCi)	95	130	17

NOTES:

Source of Raw Data: Dow, 1963h.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-19

SUMMARY OF 1960 AIRBORNE DEPLETED URANIUM EMISSIONS FROM ROCKY FLATS BUILDINGS 444 AND 447

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})		
	Building 444		Building 447
	Duct 2	Duct 3	
January	0.0065	0.26	0.084
February	0.0065	0.15	0.13
March	0.054	0.11	0.029
April	0.017	0.15	0.028
May	0.0060	0.24	5.2
June	0.010	0.27	0.025
July	0.084	0.25	0.031
August	0.097	0.29	0.031
September	*	*	*
October	*	*	*
November	*	*	*
December	*	*	*
1960 Average Daily Concentration	0.035	0.21	0.69
Total Release (μCi)	19	68	200

NOTES:

Source of Raw Data: Dow, 1963h.

* Emissions from Buildings 444 and 447 were not independently reconstructed for these months. Emissions for these buildings were included in the calculated totals prepared for the Final Environmental Impact Statement.

Release quantities for January through August 1960 were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 243 d, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-20

AIRBORNE DEPLETED URANIUM RELEASE ESTIMATES, 1954-1960

Year	Independently Reconstructed (μCi)	Reported in the FEIS[1] (μCi)
1954	1,200	None Reported
1955s	2,100	None Reported
1956	1,000	None Reported
1957	730	38
1958	1,600[2]	51
1959	270[2]	34
1960	350[2]	58

Source of Raw Data: Dow, 1957b

1 Final Environmental Impact Statement (USDOE, 1980). The FEIS only reflects Building 883 emissions during this time period; contributions from Buildings 444 and 447 were not included from 1957 through August 1960.

2 Emissions from Buildings 444 and 447 were independently reconstructed and added to the reported depleted uranium emission totals for other buildings.

flow rate data from Table 2-2. Results of these calculations are presented in Tables 2-21 and 2-22. The data reconstruction resulted in a release estimate of 490 μCi of depleted uranium for 1963, compared to the reported release of 340 μCi of depleted uranium in the Final Environmental Impact Statement. These estimates are considered to be in relatively good agreement, with the difference likely being due to the exhaust flow rate assumptions used. The independent reconstruction yielded an enriched uranium release estimate of 330 μCi for 1963, compared with a release of 280 μCi that was reported in the FEIS.

1969 Reconstruction

Raw and compiled data for uranium in effluents from Buildings 331, 444, 447, 881, 883, 886, and 889 were found in the Denver Federal Records Center. As was done with the 1969 plutonium data, raw data were spot-checked against the compiled summary data for accuracy. The raw data were found to be in agreement with the compiled data; therefore, the compiled data were used to reconstruct the 1969 uranium releases. The results of the 1969 data reconstruction are presented in Tables 2-23 and 2-24. The reconstruction resulted in release estimates of 160 μCi and 50 μCi of depleted and enriched uranium during 1969, respectively, which was in good agreement with the releases of 170 μCi and 51 μCi of depleted and enriched uranium reported in the FEIS.

1973 Reconstruction

Compiled data for uranium in Rocky Flats airborne effluents during 1973 were located in the Denver Federal Records Center for Buildings 881, 883, 865, 886, 889, 991, 444, and 447. The results were reported as microcuries of uranium released from each building by month. No raw data were located for any of these buildings. The reconstruction results using this compiled data for the uranium buildings are presented in Tables 2-25 and 2-26. The 1973 data reconstruction resulted in release estimates of 54 μCi and 12 μCi of depleted and enriched uranium, respectively, which is in good agreement with the emissions of 63 μCi and 12 μCi of depleted and enriched uranium reported in the FEIS.

2.3.2.3 1984 Reconstructions

Effluent data for calendar year 1984 were located among a retired employees files. The data were contained in a three-ring binder labelled "1984." The data consisted of computer generated summary sheets of monthly isotope specific effluent releases in µCi (Pu-238, Pu239/40, U-238, U235, and Am-241) for each monitored release point at the Rocky Flats plant and monthly handwritten isotope-specific analytical reports for each monitored release point (Boss, 1984).

TABLE 2-21

SUMMARY OF 1963 AIRBORNE DEPLETED URANIUM EMISSIONS

Month	Average Daily Concentration of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})				
	Building 883	Building 444		Building 447	Building 331
	Duct A	Duct 2	Duct 3		
January	0.087	0.018	0.047	0.13	0.052
February	0.089	0.048	0.040	0.19	0.058
March	0.046	0.41	0.36	0.38	0.0068
April	0.076	0.059	0.19	1.9	0.062
May	0.053	0.15	0.32	2.2	0.012
June	0.017	0.045	0.077	0.81	0.073
July	0.10	0.088	0.27	0.16	N/A
August	0.059	0.12	0.49	0.31	N/A
September	0.44	0.029	0.30	0.035	0.002
October	N/A	0.15	0.079	0.78	0.0063
November	N/A	0.12	0.17	0.081	0.016
December	N/A	0.050	0.097	0.14	0.0058
Total	0.97	1.3	2.4	7.1	0.29
1963 Average Daily Concentration	0.11	0.11	0.20	0.59	0.029
Release Totals (μCi)	62	85	97	250	0.91
Grand Total (μCi)	490				

NOTES:

Sources of Raw Data: Dow, 1963e-l.

N/A = Not Available

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-22

SUMMARY OF 1963 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Average Daily Concentrations of Long-Lived Alpha Emitters (dis min^{-1} m^{-3})						
	Building 883	Building 881					
	Duct B	Duct 1	Duct 2	Duct 3	Duct 4	Duct 5	Duct 5A
January	0.15	0.10	0.11	0.20	0.24	0.19	0.30
February	0.11	0.070	0.096	0.046	0.10	0.21	0.066
March	0.061	0.056	0.15	0.096	0.12	0.24	0.090
April	0.042	0.086	0.098	0.10	0.15	0.21	0.080
May	0.063	0.15	0.10	0.14	0.20	0.18	0.039
June	0.050	0.15	0.17	0.14	0.16	0.25	0.095
July	0.11	0.14	0.13	0.089	0.084	0.17	1.6
August	0.059	0.093	0.13	0.11	0.12	0.17	6.2
September	0.13	0.13	0.10	0.091	0.11	0.14	0.084
October	N/A	0.053	0.062	0.074	0.068	0.13	0.058
November	N/A	0.072	0.050	0.037	0.052	0.080	0.078
December	N/A	0.050	0.049	0.058	0.13	0.10	0.11
1963 Average Daily Concentration	0.086	0.096	0.10	0.095	0.13	0.17	0.73
Release Totals (μCi)	49	47	49	44	62	16	58
Grand Total (μCi)	330						

NOTES:

Source of Raw Data: Dow, 1963h.

Annual release quantities were calculated by multiplying:

1. The average concentration (dis min^{-1} m^{-3}) for each duct or stack from above,
2. The average duct or stack flow rate (m^3 d^{-1}) from Table 2-2,
   (The Table 2-2 total value for Bldg 881 Ducts 1, 2, and 3 was divided by 3 for application to individual ducts. The Table 2-2 total value for Bldg 881 Ducts 5 & 5A was divided by 2 for application to individual ducts.)
3. 365 d y^{-1}, and
4. 4.50×10^{-7} μCi dis^{-1} min.

TABLE 2-23

SUMMARY OF 1969 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Total Long-Lived Alpha-Emitting Radioactivity Released (μCi)		
	Building 881		Building 883
	Ducts 1, 2, 3	Duct 5	Duct B
January	4.9	0.41	1.1
February	2.0	0.45	0.81
March	1.7	0.27	0.95
April	1.7	0.23	0.91
May	3.2	0.27	0.92
June	2.9	0.38	3.5
July	4.2	0.42	1.4
August	3.6	1.7	2.7
September	1.3	0.26	0.87
October	1.5	0.26	2.2
November	0.67	1.0	0.28
December	0.88	0.20	0.28
1969 Release Totals (μCi)	28	5.9	16
Grand Total (μCi)	50		

NOTES:

Source of Raw Data: Dow, 1969c.

N/A = Not Available

TABLE 2-24

SUMMARY OF 1969 AIRBORNE DEPLETED URANIUM EMISSIONS

Month	Total Long-Lived Alpha-Emitting Radioactivity Released (μCi)						
	Building 883	Building 444		Building 447	Building 886	Building 331	Building 889
	Duct A	Duct 2	Duct 3				
January	1.0	0.75	4.2	20	0.030	0.020	N/A
February	0.45	0.52	0.77	7.3	0.040	0.014	N/A
March	0.32	0.54	1.0	4.2	0.040	0.010	N/A
April	0.64	0.24	0.94	1.3	0.040	0.020	N/A
May	0.65	0.73	1.0	0.78	0.030	0.20	N/A
June	1.4	0.65	0.46	1.4	0.050	0.10	N/A
July	0.81	0.90	1.1	1.9	0.050	0.20	N/A
August	1.7	0.47	6.7	1.1	0.040	0.040	N/A
September	0.69	0.96	0.27	1.1	0.040	0.020	0.018
October	1.2	0.35	2.3	0.73	0.090	0.040	NA
November	0.86	0.7	85	1.4	0.050	0.0090	0.009
December	1.2	0.55	0.79	1.1	0.12	0.030	0.05
1969 Release Totals (μCi)	11	7.40	100	42	0.62	0.70	0.077
Grand Total (μCi)	160						

NOTES:

Source of Raw Data: Dow, 1969c.

N/A = Not Available

TABLE 2-25

SUMMARY OF 1973 AIRBORNE ENRICHED URANIUM EMISSIONS

Month	Total Long-Lived Alpha-Emitting Radioactivity Released (μCi)		
	Building 881		Building 883
	Ducts 1, 2, and 3	Duct 5	Duct B
January	0.56	0.050	0.063
February	0.21	0.027	0.050
March	1.4	0.032	0.032
April	0.71	0.12	4.7
May	0.34	0.072	0.081
June	0.13	0.059	0.054
July	0.21	0.014	0.19
August	0.45	0.11	0.068
September	0.55	0.063	0.032
October	0.40	0.054	0.11
November	0.23	0.036	0.036
December	0.47	0.31	0.10
Release Totals (μCi)	5.7	0.95	5.5
Grand Total (μCi)	12		

Source of Raw Data: Dow, 1973b.

TABLE 2-26

SUMMARY OF 1973 AIRBORNE DEPLETED URANIUM EMISSIONS

Month	Total Long-Lived Alpha-Emitting Radioactivity Released (μCi)							
	Building 883 Duct A	Building 865	Building 886	Building 889	Building 991	Building 444		Building 447
						Duct 1	Ducts 2, 3	
January	1.1	0.099	0.0090	0.027	0.0050	0.059	0.17	1.5
February	0.55	0.032	0.0020	0.014	0.0010	0.018	0.73	0.61
March	0.40	0.054	0.0090	0.005	0.0090	0.12	0.18	0.21
April	0.69	0.045	0.0090	0.018	0.0050	0.0050	0.32	0.31
May	0.56	0.045	0.013	0.027	0.0090	0.027	0.17	0.50
June	0.64	0.054	0.0090	0.023	0.0050	0.018	0.27	1.3
July	0.67	30	0.0090	0.018	0.0050	0.023	0.14	0.33
August	0.88	0.78	0.0090	0.0090	0.0050	0.145	0.095	0.58
September	0.27	0.20	0.0050	0.0050	0.0010	0.027	0.095	0.28
October	0.41	0.099	0.0050	0.041	0.0050	0.036	0.086	0.57
November	0.22	0.0050	0.032	0.014	0.059	0.14	0.25	1.4
December	0.58	0.13	0.014	0.023	0.027	0.51	0.17	3.0
Release Totals (μCi)	7.0	32	0.13	0.22	0.14	1.1	2.7	11
Grand Total (μCi)	54							

Source of Raw Data: Dow, 1973b.

The monthly analytical data were compiled by release point and entered into a database. The results were reported as concentrations (fCi m^{-3}). To convert these concentrations to a total release in μCi, the effluent flow rate values were required. The effluent flow rate for each release point was found in the typewritten monthly summary sheets, which contained effluent flow rate data for each release point. The effluent flow rate was multiplied by the concentrations reported in the analytical data sheets and a factor to convert fCi m^{-3} to μCi m^{-3} to arrive at a total release in μCi. A summary of the results of the reconstruction of the 1984 effluent data is provided in Table 2-27.

The reconstruction resulted in a total release of 78 μCi of Pu-239/240, 2.3 μCi of Pu-238, 20 μCi of U-233/234, and 5.5 μCi of U-238. In comparison, the 1984 Annual Environmental Monitoring Report (Setlock, 1985) reports releases of 13 μCi of Pu-239/240, 17 μCi of U-233/234, 18 μCi of U-238, and 3 μCi of Am-241. It is important to note that the analytical data (located by ChemRisk) did not contain results for the Am-241 isotope, however, the Environmental Monitoring Report did report a value for Am-241 and the analytical data contained results for the Pu-238 isotope but the Environmental Monitoring Report did not report a value for Pu-238. It is not known why this discrepancy occurred.

The reconstructed results for U-238, U-233/34, and Pu-239/40 are 18 percent greater, 69 percent smaller, and 500 percent greater, respectively, than the results reported in the 1984 Annual Environmental Monitoring Report.

2.3.2.4 1986 Reconstructions

Like the calendar year 1984 effluent data, data for the 1986 reconstruction were located among a retired employees files. The data were contained in a three ring binder labelled "1986." These data were compiled in a database similar to the database created for the 1984 data. The total release by effluent release point was calculated and is presented in Table 2-28.

The reconstruction resulted in a total release of 29 μCi of Pu-239/240, 0.77 μCi of Pu-238, 9.1 μCi of Am-241, 11 μCi of U-233/234, and 2.9 μCi of U-238. In comparison the 1986 Annual Environmental Monitoring Report (Setlock, 1987) reports a release of 14 μCi of Pu-239/240, 0.90 μCi of Pu-238, 5.0 μCi of Am-241, 4.0 μCi of U-233/234, and 17 μCi of U-238.

The reconstructed results for Pu-239/240, Pu-238, Am-241, U-233/234, and U-238 were 110 percent greater, 14 percent smaller, 82 percent greater, 82 percent smaller, and 175 percent greater, respectively, then the results reported in the 1986 Annual Environmental Monitoring Report.

TABLE 2-27

SUMMARY OF 1984 AIRBORNE RADIONUCLIDE EMISSIONS

Building/Stack Identifier	Plutonium-239/240 Released (μCi)	Plutonium-238 Released (μCi)	Uranium-233/234 Released (μCi)	Uranium-238 Released (μCi)
371-n01	0.068	0.016	0.25	0.045
371-n02	0.048	0.017	0.41	0.035
371-sss	0.053	0.019	0.13	0.069
374-mai	0.11	0.023	0.69	0.11
374-spd	0.058	0.016	0.11	0.017
444-d01	ND	ND	0.42	0.061
444-d02	ND	ND	1.2	0.16
444-d05	ND	ND	0.093	0.015
447-mai	ND	ND	3.6	0.014
559-561	0.084	0.033	0.43	0.10
707-101	0.015	0.0013	0.0096	0.0016
707-102	0.015	0.0041	0.093	0.015
707-105	0.036	0.0084	0.22	0.035
707-106	0.018	0.011	0.19	0.030
707-107	0.038	0.014	0.25	0.045
707-108	0.040	0.017	0.18	0.035
771-cma	0.62	0.023	0.034	0.0088
771-crm	0.45	0.019	0.035	0.010
771-ma2	4.8	0.24	0.41	0.20
771-mai	58	1.0	1.2	0.28
774-202	0.19	0.0088	0.092	0.0052
776-201	0.0089	0.0033	0.024	0.0044
776-202	0.027	0.0053	0.071	0.016
776-204	0.11	0.024	0.43	0.081
776-205	0.0019	0.0014	0.028	0.0044
776-206	0.066	0.014	0.82	0.12
776-251	0.045	0.018	0.20	0.069
776-50c	0.011	0.0055	0.12	0.035
776-50d	0.091	0.011	0.096	0.022
778-ldy	0.26	0.019	0.78	0.089
779-729	0.096	0.047	0.27	0.044
779-782	0.063	0.017	0.18	0.051
865-eee	ND	ND	0.39	0.13
865-www	ND	ND	0.34	0.045
881-d12	0.056	0.055	0.17	0.63

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TABLE 2-27 (Continued)

SUMMARY OF 1984 AIRBORNE RADIONUCLIDE EMISSIONS

Building/Stack Identifier	Plutonium-239/240 Released (μCi)	Plutonium-238 Released (μCi)	Uranium-233/234 Released (μCi)	Uranium-238 Released (μCi)
881-d34	12	0.36	0.14	0.69
881-d56	0.10	0.072	0.17	0.99
883-b123	ND	ND	2.8	0.55
883-b465	0.0072	0.00078	2.3	0.49
886-875	0.015	0.0078	0.12	0.031
889-mai	0.013	0.091	0.29	0.050
991-985	0.032	0.010	0.25	0.043
991-mai	0.019	0.012	0.13	0.036
Total	78	2.3	20	5.5

ND = No value given. Raw data indicated result equal to zero. It appears that monitoring was not performed during these periods.

TABLE 2-28

SUMMARY OF 1986 AIRBORNE RADIONUCLIDE EMISSIONS

Building/Stack Identifier	Plutonium-239/240 Released (μCi)	Plutonium-238 Released (μCi)	Americium-241 Released (μCi)	Uranium-233/234 Released (μCi)	Uranium-238 Released (μCi)
371-n01	0.15	0.0088	0.25	0.13	0.032
371-n02	0.12	0.0049	0.051	0.11	0.025
371-sss	2.6	0.054	0.25	0.26	0.056
374-mai	0.77	0.039	0.22	0.25	0.052
374-spd	0.064	0.0024	0.045	0.040	0.0085
444-d05	ND	ND	ND	0.064	0.0017
444-mai	ND	ND	ND	0.82	0.16
447-mai	ND	ND	ND	0.52	0.090
559-561	0.11	0.0086	0.034	0.11	0.033
707-101	0.0014	0.000087	0.00040	0.0026	0.00085
707-102	0.013	0.00078	0.0014	0.010	0.0038
707-105	0.021	0.0018	0.014	0.033	0.0097
707-106	0.083	0.0019	0.0030	0.032	0.0060
707-107	0.096	0.0029	0.0088	0.031	0.0093
707-108	0.082	0.0043	0.0071	0.038	0.015
707-R21	0.014	0.00095	0.0066	0.0022	0.00018
707-R22	0.0090	0.0011	0.0016	0.0033	0.000058
707-R23	0.012	0.00098	0.00096	0.0019	0.00037
707-R24	0.0049	0.00026	0.0012	0.0018	0.00015
707-R25	0.023	0.0014	0.0013	0.0021	0.00040
707-R26	0.0054	0.00014	0.00068	0.0024	0.000046
707-R27	0.0046	0.000085	0.00012	0.0013	0.000094
707-R45	0.0038	0.00019	0.00032	0.00094	0.000025
707-R46	0.025	0.00062	0.0013	0.0013	0.000035
771-cma	0.29	0.0080	0.034	0.021	0.0034
771-crm	0.44	0.013	0.11	0.026	0.0041
771-mai	12	0.27	1.6	0.35	0.068
771-ma2	10	0.24	6.3	0.39	0.091
774-202	0.11	0.0029	0.020	0.0080	0.0015
776-201	0.0041	0.00043	0.00053	0.0050	0.0011
776-202	0.012	0.00083	0.0029	0.0099	0.0017
776-204	0.10	0.00054	0.022	0.059	0.022
776-205	0.057	0.0020	0.0040	0.022	0.0059

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TABLE 2-28 (Continued)

SUMMARY OF 1986 AIRBORNE RADIONUCLIDE EMISSIONS

Building/Stack Identifier	Plutonium-239/240 Released (μCi)	Plutonium-238 Released (μCi)	Americium-241 Released (μCi)	Uranium-233/234 Released (μCi)	Uranium-238 Released (μCi)
776-206	0.022	0.0015	0.0037	0.029	0.0064
776-207	0.0093	0.00014	0.0018	0.00070	ND
776-251	0.0088	0.00046	0.0020	0.013	0.0049
776-252	0.034	0.00099	0.0038	0.044	0.0072
776-50c	0.027	0.0016	0.0050	0.036	0.013
776-50d	0.050	0.0025	0.0021	0.023	0.0096
778-ldy	0.077	0.0030	0.010	0.28	0.044
779-782	0.14	0.0095	0.015	0.16	0.056
779-729	0.045	0.0044	0.0012	0.043	0.0084
865-eee	ND	ND	ND	0.21	0.050
865-www	ND	ND	ND	0.21	0.054
881-d12	1.0	0.030	0.0067	0.17	0.36
881-d34	0.41	0.024	0.0081	0.23	0.42
881-d56	0.062	0.0055	0.0033	0.28	0.11
883-b123	ND	ND	ND	2.2	0.35
883-b465	ND	ND	ND	2.1	0.35
883-ccc	ND	ND	ND	1.5	0.26
886-875	0.017	0.0017	0.0032	0.089	0.022
889-mai	0.016	0.00081	0.00070	0.40	0.058
991-985	0.021	0.0012	0.014	0.037	0.011
991-mai	0.016	0.0029	0.0015	0.079	0.016
Total	29	0.77	9.1	11	2.9

ND = No value given. Raw data indicated result equal to zero. It appears that monitoring was not performed during these periods.

2.3.3 Results of the Data Reconstruction Efforts

The results of efforts to reconstruct emissions of plutonium and uranium from raw or partially compiled data are summarized and compared to those reported in the Final Environmental Impact Statement by the plant in Tables 2-29, 2-30, and 2-31. The reconstructed plutonium emissions (Table 2-29) were in good agreement with the totals reported in the Final Environmental Impact Statement for each reconstructed year, with the notable exception of the release in 1957 related to the period after the 1957 fire. The reconstructed uranium emissions (Table 2-30) are not in agreement with those reported in the Final Environmental Impact Statement from 1954 through 1960 due to the omission of data for a number of buildings in the summaries presented in the Final Environmental Impact Statement.

Airborne radioactivity release estimates that have been determined based on reconstructed or reported effluent measurements are summarized in Tables 2-32 and 2-33. The uncertainties associated with these measurements must be specified before they can be used in dose reconstruction. However, before the uncertainties in these numbers are specified, the quantification of the various isotopes of each radioactive element that was emitted will be addressed. The following section addresses this step.

2.4 Isotopic Composition of Alpha-Emitting Effluents

Development of useful airborne emission estimates for americium, plutonium and uranium requires the quantification of their individual isotopes. The preceding sections of this report have addressed quantification of radioactive materials that have been emitted from the Rocky Flats Plant based on historical measurements. For a large portion of the operational history of the Rocky Flats Plant (the time period that saw the largest quantities of radioactivity released from the site), emissions were measured in terms of long-lived alpha radioactivity. These measurements are the basic data available for reconstruction of plutonium and americium emissions for 1953 to 1973 and for uranium emissions for 1953 through 1977.

2.4.1 Plutonium and Americium Isotopes

Rocky Flats plutonium has contained the isotopes Pu-238, Pu-239, Pu-240, Pu-241 and Pu-242 in proportions reflected in Table 2-34 (USDOE, 1980). These values are averages over a two-year period from July 1976 to July 1978 and are useful for translating nonspecific or multiple isotope effluent measurements to releases of specific radionuclides. As shown in the table, Pu-238 and Pu-242 together contribute less than 3 percent to the total plutonium alpha activity. In this report, individual source terms were not developed for these two isotopes. Instead, it was assumed that the total plutonium alpha activity is equal to long-lived alpha activities associated with Pu-239/240.

TABLE 2-29

REPORTED AND RECONSTRUCTED AIRBORNE PLUTONIUM RELEASE ESTIMATES

Year	1980 Final Environmental Impact Statement (μCi)	Reconstructed (μCi)
1957	1,600*	15,000*
1963	3,900	3,600
1969	1,700	1,400
1973	77	62

* Does not include 1957 fire releases.

TABLE 2-30

REPORTED AND RECONSTRUCTED URANIUM RELEASE ESTIMATES

Year	Depleted Uranium Areas (μCi)		Enriched Uranium Areas (μCi)	
	FEIS[1]	Reconstructed	FEIS[1]	Reconstructed
1954	Not Reported	1,200	Not Reported	9.1
1955	Not Reported	2,100	Not Reported	74
1956	Not Reported	1,000	Not Reported	1,100
1957	38[2]	770[3]	230	360
1958	51[2]	1,600[3]	310	Not Reconstructed
1959	34[2]	270[3]	540	Not Reconstructed
1960	58[2]	350[3]	860	Not Reconstructed
1963	340	490	280	330
1969	1702	160	51	50
1973	63	54	11	12

1 Final Environmental Impact Statement (USDOE, 1980)
2 Reported values did not include emissions from Buildings 444 and 447.
3 Emissions from Buildings 444 and 447 were independently reconstructed and added to the reported depleted uranium emission totals for other buildings.

TABLE 2-31

REPORTED AND RECONSTRUCTED AIRBORNE RADIONUCLIDE RELEASE ESTIMATES

Data Period and Data Source	Plutonium-238 Release (μCi)	Plutonium-239/240 Release (μCi)	Americium-241 Release (μCi)	Uranium-238 Release (μCi)	Uranium-233/234 Release (μCi)
1984 per DOE Effluent Information System	Not Reported	13	3	18	17
1984 per ChemRisk	2.3	78	Not Reconstructed	5.5	20
1986 per DOE Effluent Information System	0.90	14	5.0	17	4.0
1986 per ChemRisk	0.77	29	9.1	2.9	11

TABLE 2-32

ESTIMATED ROCKY FLATS AIRBORNE RADIOACTIVITY EMISSIONS, 1953-1977*

Numbers in Shaded Boxes Were Independently Generated

Year	Total Long-Lived Alpha Radioactivity Released (μCi)		
	Plutonium Areas	Depleted Uranium Areas	Enriched Uranium Areas
1953	2.0	1,200**	9.1**
1954	65	1,200	9.1
1955	72	2,100	74
1956	230	1,000	1,100
1957	15,000	770	360
1958	3,100	1,600	310
1959	1,400	270	540
1960	1,300	350	860
1961	1,500	520	480
1962	3,000	370	250
1963	3,600	490	330
1964	2,800[1]	240	190
1965	6,500[2]	280	190
1966	320	140	230
1967	400	140	110
1968	490	140	160
1969	1,400[3,4]	160	50
1970	380[5]	190	64
1971	74[6,7,8]	58	41
1972	61[9,10]	42	4
1973	62	54	12
1974	*Based on Isotope-Specific Measurements (See Table 2-33)*	9	27
1975		28	28
1976		12	16
1977		19	21

* Totals include accidental releases as noted:
[1] Chemical explosion in glove box (10 μCi).
[2] Glove-box drain plug fire (1,200 μCi).
[3] Plutonium glove-box and building fire in production building (856 μCi). Monitored portions of 1969 fire emissions are included in this value.
[4] Plutonium fire in tunnel between buildings (20 μCi).
[5] Contamination release from spill caused by cleaning plugged drain line (25 μCi).
[6] Contamination spread from reduction furnace explosion (4 μCi).
[7] Plutonium can explosion, with fire and contamination (<4 μCi).
[8] Contamination from spill through hole in barrel liner (<4 μCi).
[9] Incinerator glove-box explosion and fire (<2 μCi).
[10] Incinerator fire and contamination (<2 μCi).
** No data were found for 1953. Uranium releases for the year are assumed to equal those for 1954.

TABLE 2-33

ESTIMATED ROCKY FLATS AIRBORNE RADIONUCLIDE EMISSIONS, 1974-1989

Numbers in Shaded Boxes Were Independently Generated

Year	Plutonium-239/240 (μCi)	Americium-241 (μCi)	Uranium-238* (μCi)	Uranium-233/234* (μCi)
1974	960[1]	*Not Reported for this Period (See Table 2-38)*	*Based on Total Long-Lived Alpha Measurements (See Table 2-32)*	
1975	10			
1976	4.0			
1977	4.0			
1978	2.8		33	21
1979	5.5		26	9.2
1980	12		15	15
1981	8.2		18	12
1982	20		19	12
1983	78		31	20
1984	78		5.5	20
1985	9.2	2.0	39	7.9
1986	29	9.1	2.9	11
1987	15	3.2	12	4.6
1988	15	2.0	9.3	2.6
1989	4.5	1.1	2.5	5.2

* Uranium emissions for 1981 through 1984 were reported as total uranium. Fractions of the totals that were enriched and depleted were estimated as described in Section 2.4.

[1] Includes release from control valve failure (934 μCi).

TABLE 2-34

TYPICAL ISOTOPIC COMPOSITION OF ROCKY FLATS PLUTONIUM

Isotope	Percentage by Mass[1]	Range of Mass Percentage[2]	Percentage of Plutonium Alpha Activity[1]	Percentage of Pu-239/240 Alpha Activity[3]
Pu-238	0.01	0.03 - 0.05	2.33	2.39
Pu-239	93.79	92.84 - 93.84	79.62	81.53
Pu-240	5.80	5.5 - 6.5	18.04	18.47
Pu-241	0.36	Not Given	508.5* (beta activity)	520.7** (beta activity)
Pu-242	0.03	Not Given	0.00161	0.00165

* Pu-241 is a beta emitter. It does not emit alpha particles. In Rocky Flats plutonium it emits beta particles at 3.82 to 4.51 times the rate that alpha-emitting nuclides present emit alpha particles.

** Pu-241 present in Rocky Flats plutonium emits beta particles at 5.21 times the rate that the Pu-239 and Pu-240 present emit alpha particles.

REFERENCES/SOURCES:

[1] Rockwell, 1989.
[2] Del Pizzo *et al.*, 1970.
[3] Calculated from the specific activities of the individual nuclides in Column 4.

Total long-lived alpha activity measured in effluents from a plutonium building included contribution from Pu-239/240 and Am-241. Am-241 is a decay product of Pu-241 and exists as an undesirable contaminant in weapons-grade plutonium. Plutonium like that used at Rocky Flats contained about 0.3 mass percent Pu-241 (Rockwell, 1985) and initially contained about 0.0001 percent Am-241 (Krey *et al.*, 1976). The Pu-241, however, decayed relatively quickly (with a 14.4-year half-life) to form Am-241 as time passed after purification. Based on an average age of plutonium handled at Rocky Flats (time since removal from the reactor or purification), of ten years (USDOE, 1980), the Am-241 to Pu-239/240 ratio should be somewhat lower than 12 percent, based on the decay schemes of the radionuclides alone. The Final Environmental Impact Statement for the Rocky Flats Plant states that the americium to plutonium activity ratio has ranged from 10 percent to 20 percent, but also states that this ratio can change during processing that separates americium from plutonium. Based on data from the DOE Effluent Information System (EG&G, 1991a) and independently reconstructed emissions for 1986, the average airborne Am-241 emissions for each year from 1985 to 1989 were between 13 and 31 percent of the plutonium alpha activity release total for the same year (Table 2-35). This information was used to develop release estimates of total plutonium alpha activity and Am-241.

Over the operation history of the Rocky Flats Plant, plutonium and americium isotopes were monitored and reported in a number of ways. Before 1973, only total long-lived alpha activity in effluents was monitored. From 1974 through 1984, only Pu-239/240 was measured; no monitoring record of Am-241 was found. After 1985, Pu-239/240 and Am-241 were routinely measured and reported. Because the identities of contaminants reported for the three time periods are different, different approaches were used to develop source terms for total plutonium alpha activity for the period between 1953 and 1973, Pu-239/240 for 1974 to 1989, and for Am-241.

2.4.1.1 Plutonium and Americium Isotopes During 1953 - 1973

During this period, only total long-lived alpha activity in effluents was monitored and reported. The americium content of airborne effluents was not specifically measured. In order to translate measurements of total long-lived alpha radioactivity from plutonium facilities to emission totals of Pu-239/240, Am-241 and Pu-241, a number of relationships must be characterized:

- The fractions of total long-lived alpha radioactivity from plutonium facilities that were actually plutonium-239/240 and americium-241 must be estimated.
- The ratio of relatively short-lived beta-emitting plutonium-241 (14.4-year half-life) to total plutonium alpha activity must be determined.

TABLE 2-35

ACTIVITY RATIOS OF AM-241 TO PU-239/240
IN MONITORED ROCKY FLATS EFFLUENTS

Calendar Year	Ratio of Airborne Am-241 Released to Pu-239/240 Released
1985	22%
1986	31%
1987	21%
1988	13%
1989	24%
Average	22%

References:

Independent reconstruction for 1986, DOE Effluent Information System (EG&G, 1991a) for other years.

Based on the monitoring data in Table 2-35, it was estimated that the average ratio of Am-241 emissions to Pu-239/240 emissions is 22 percent. Since the reported total long-lived alpha activity is essentially equal to the sum of the Pu-239/240 and Am-241, release estimates of Pu-239/240 and Am-241 can be calculated by solving the following two equations:

$$F_{Am} + F_{Pu} = 1$$

$$F_{Am} / F_{Pu} = 0.22$$

Where:

F_{Am} = Fraction of total long-lived alpha activity associated with Am-241, dimensionless; and

F_{Pu} = Fraction of total long-lived alpha activity associated with Pu-239/240, dimensionless.

F_{Pu} and F_{Am} were calculated to be 0.82 and 0.18, respectively. Table 2-36 presents the calculated release estimates of plutonium alpha activity and Am-241 between 1953 and 1973; they were estimated by multiplying the reported total long-lived alpha activity by 0.82 and 0.18, respectively.

Based on the result of a two-year study conducted by the RFP from July 1976 to July 1978, it was estimated that the ratio of short-lived beta-emitting Pu-241 (14.4-year half-life) to measured total plutonium alpha activity is 508.5 percent (Table 2-34). Using this relationship, source terms of Pu-241 for 1953 through 1973 were calculated from the release estimates of the total plutonium alpha (Pu-239/240) data of the same period and are also shown in Table 2-36.

2.4.1.2 Plutonium and Americium Isotopes During 1974 - 1984

From 1974 through 1984, Pu-239/240 releases through routine operations were monitored by analytical technique that is specific for the radionuclides. However, Am-241 was not included in the monitoring scheme and its release information is not available. In order to translate measurements of Pu-239/240 to emissions of Am-241 and Pu-241, a number of relationships must be characterized:

- The ratio of Am-241 to Pu-239/240 in airborne routine releases must be estimated.

TABLE 2-36

PLUTONIUM ALPHA ACTIVITY, PU-241, AND AM-241 RELEASE ESTIMATES, 1953-1973
Derived from Total Long-Lived Alpha Measurements

Year	Plutonium Alpha (μCi)	Americium-241 (μCi)	Plutonium-241 (μCi)
1953	1.6	0.36	8.3
1954	53	12	270
1955	59	13	300
1956	190	41	960
1957	12,000	2,700	63,000
1958	2,500	560	13,000
1959	1,100	250	5,800
1960	1,100	230	5,400
1961	1,200	270	6,300
1962	2,500	540	13,000
1963	3,000	650	15,000
1964	2,300	500	12,000
1965	5,300	1,200	27,000
1966	260	58	1,300
1967	330	72	1,700
1968	400	88	2,000
1969	1,100	250	5,800
1970	310	68	1,600
1971	61	13	310
1972	50	11	250
1973	51	11	260

- The ratio of relatively short-lived beta-emitting plutonium-241 to Pu-239/240 must be determined.

The approach used to develop release estimates of Am-241 for 1953 - 1973 was also used for the years between 1974 and 1984. Based on the Am-241 measurements reported for 1985 through 1989, the average ratio of Am-241 emission to Pu-239/240 emission is 22 percent (Table 2-35). Assuming this relationship has not changed significantly over time, source terms of Am-241 in this period can be calculated by multiplying Pu-239/240 release estimates by 0.22.

Table 2-37 presents the calculated release estimates of Am-241 between 1974 and 1984; they were determined by multiplying the reported Pu-239/240 release estimates by 0.22.

Table 2-34 shows that the ratio of short-lived beta-emitting Pu-241 to Pu-239/240 alpha activity is 520.7 percent. This relationship was used to calculate source terms of Pu-241 from 1974 through 1984, which are also shown in Table 2-37.

2.4.1.3 Plutonium and Americium Isotopes During 1985 - 1989

During this period of time, Pu-239/240 and Am-241 emissions were routinely sampled and monitored by alpha spectrometry following radiochemical separation. The reported values were used as the source terms of these two radionuclides from 1985 through 1989. However, Pu-241 was not included in the monitoring scheme and its release information is not available. In order to translate measurements of Pu-239/240 to emissions Pu-241, it is necessary to know the ratio of relatively short-lived beta-emitting plutonium-241 to Pu-239/240 alpha.

The approach used in the section above was also used to develop source terms for Pu-241 from 1985 through 1989. Release estimates of Pu-241 shown in Table 2-38 were calculated by multiplying the Pu-239/240 alpha activity by 5.207.

2.4.2 Uranium Isotopes

Uranium has historically been processed at Rocky Flats in two forms: enriched and depleted. Tables 2-39 and 2-40 show the reported activity fractions of the different radionuclides present in Rocky Flats enriched and depleted uranium (USDOE, 1980). These fractions are useful for developing source terms for radionuclides that were not monitored from those that were actually measured.

For the purpose of this evaluation, release estimates were developed for enriched and depleted uranium. Taking the source term analyses to the level of individual uranium isotopes is not likely to improve the result of dose assessment because of the following reasons:

TABLE 2-37

PU-241 AND AM-241 RELEASE ESTIMATES, 1974-1984
Derived from Pu-239/240 Measurements

Year	Plutonium-241 (μCi)	Americium-241 (μCi)
1974	5,000	210
1975	52	22
1976	21	0.88
1977	21	0.88
1978	15	0.62
1979	29	1.2
1980	62	2.6
1981	43	1.8
1982	100	4.4
1983	410	17
1984	410	17

TABLE 2-38

PU-241 RELEASE ESTIMATES, 1985-1989
Derived from Pu-239/240 Measurements

Year	Plutonium-241 (μCi)
1985	48
1986	150
1987	78
1988	78
1989	23

TABLE 2-39

ACTIVITY FRACTIONS OF ROCKY FLATS ENRICHED URANIUM

Nuclide	Percentage by Weight	Percentage of Alpha Activity	Percentage of Beta Activity
Th-231		-	99.1
Th-234		-	0.892
U-234		96.5	-
U-235	over 93%	3.11	-
U-236		0.389	-
U-238		0.0280	-

TABLE 2-40

ACTIVITY FRACTIONS OF ROCKY FLATS DEPLETED URANIUM

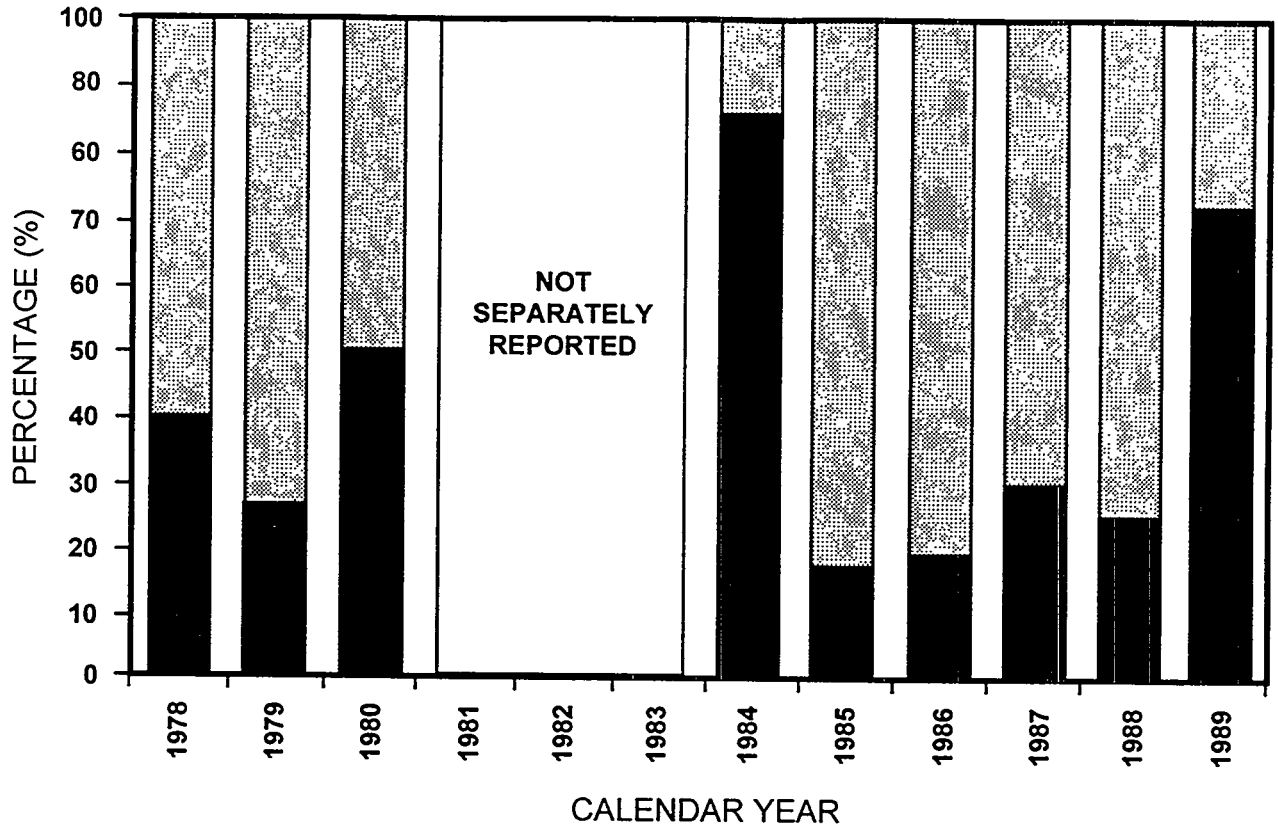
Nuclide	Percentage by Weight	Percentage of Alpha Activity	Percentage of Beta Activity
Th-231		-	1.42
Th-234		-	98.6
U-234		9.69	-
U-235	less than 0.7%	1.28	-
U-238		89.0	-

- both enriched and depleted uranium are dominated by one isotope; U-234 constitutes over 96 percent of alpha activity of enriched uranium and U-238 constitutes about 89 percent of alpha activity of depleted uranium,
- physical and toxicological properties of U-234 and U-235 are similar, and
- estimation of isotopic composition of the source terms of enriched and depleted uranium will introduce additional uncertainty into the dose assessment.

Routine isotopic analyses of effluent sample filters did not start until 1973, and reporting of total long-lived alpha activity continued for uranium facilities until approximately 1977. Therefore, for the period before 1978, total long-lived alpha radioactivity measured from facilities which process depleted uranium was assumed to represent depleted uranium and total long-lived alpha radioactivity measured from facilities which process enriched uranium is assumed to represent enriched uranium.

Even after 1978, not all uranium isotopes were routinely measured. For example, only U-238 and U-233/234 emissions were reported for 1978-1980 and 1984-1989. And for 1981-1983, only total uranium emissions were reported. For the purpose of the dose reconstruction, depleted uranium emissions were calculated by multiplying the reported or independently reconstructed U-238 emissions times a factor of 1.12 (1/0.89; where 0.89 is from Table 2-40) for the periods 1978-1980 and 1984-1989. Similarly, source terms of enriched uranium for the same periods of time were estimated by multiplying the reported U-233/234 by a factor of 1.03 (1/0.97; where 0.97 is from Table 2-39). For calendar years 1981-1984, Rocky Flats uranium emissions were reported only as total uranium emissions, based on alpha spectral measurements of U-233/234 and U-238. Separate depleted and enriched results were not reported. However, separate values were obtained from the independent reconstruction of 1984 release totals. The historical fractions of airborne depleted and enriched uranium emissions reported or independently reconstructed for 1978-1980 and 1984-1989 are depicted in Figure 2-8. It shows during these periods enriched fraction varied from 17 percent to 78 percent with an average of 40 percent of total uranium alpha activity. During the same time, depleted fraction varied from 22 percent to 83 percent with an average of 60 percent of total uranium alpha activity. These average composition values were used to estimate the emissions of enriched and depleted uranium from the plant for 1981 through 1983.

Estimated source terms of enriched and depleted uranium from the plant between 1953 and 1989 are presented in Table 2-41. They will be used in Task 8 to evaluate radiation doses associated with exposures to enriched and depleted uranium.



LEGEND

ENRICHED URANIUM

DEPLETED URANIUM

ChemRisk
A Division of McLaren/Hart

FIGURE 2-8
ENRICHED AND DEPLETED
FRACTIONS
OF RFP URANIUM EMISSIONS

RKY194T5/2-8 REV:03-02-94

TABLE 2-41

ESTIMATED AIRBORNE ENRICHED AND DEPLETED URANIUM RELEASES, 1978-1989
Derived from U-233/234 and U-238 Measurements

Year	Enriched Uranium (μCi)	Depleted Uranium (μCi)
1978	22	37
1979	9.5	29
1980	15	17
1981	12	18
1982	12	19
1983	20	31
1984	21	6.2
1985	8.1	44
1986	11	3.2
1987	4.7	13
1988	2.7	10
1989	5.4	2.8

2.5 Airborne Radioactive Effluent Data — Tritium

As discussed in Section 2.2, tritium, the only radionuclide of concern likely to be emitted in forms other than particulate, has been routinely monitored in airborne effluents since 1974. While effluent monitoring data for tritium are limited, they remain the best information for use in estimation of historical emissions.

2.5.1 The Nature of Tritium Association with Rocky Flats

As evidenced by the effluent monitoring performed since 1974, the Rocky Flats Plant is a source of routine, low-level tritium emissions. At Rocky Flats, tritium has been handled in standards, special order work, and contaminated components returned to the site. Tritium has been associated with some nuclear weapon trigger "pits." Extremely low levels of tritium are also generated by interactions of neutrons, alpha particles, and gamma rays with materials handled at Rocky Flats, including plutonium, boron, beryllium, and nitrogen (Dow, 1973a).

Some aspects of tritium handling at Rocky Flats are matters of national security and are considered classified information, and therefore cannot be discussed in this report. Classified documentation has been reviewed by members of the ChemRisk investigation team. The classified nature of certain aspects of tritium handling does not interfere with development of tritium release estimates that will serve to bound the magnitude of potential health risks from historical tritium emissions.

As discussed in the Tasks 3 and 4 report, there have been a small number of incidents reported in which significantly larger amounts of tritium than usual were released from the Rocky Flats Plant. These include tritium release incidents in 1968, 1973, and 1974, which are discussed in Section 5 of this report.

2.5.2 Availability of Monitoring Data

Beginning in 1974, annual airborne tritium release totals from Rocky Flats were reported in Rocky Flats Plant annual environmental reports. In 1974, data were reported for 12 vents, and the program expanded to 18 in 1977 and 23 in 1981 (EG&G, 1991a). In September 1988, continuous sampling for tritium was decreased to include only several exhaust systems that continued to have a potential for contribution to the site tritium release total (Rockwell, 1989).

2.5.3 Reconstruction of 1974 Airborne Tritium Releases

In order to verify the accuracy of reported tritium emission estimates, effluent data from a one-year period were assembled and release totals were independently verified. The year chosen was

1974, the first year that tritium sampling and analysis were routinely performed. This section presents the methods and results of reconstruction of tritium emissions for 1974.

Compiled records of tritium airborne effluents from Rocky Flats during 1974 were located in the Denver Federal Records Center (Dow, 1974b). These data were in the form of monthly worksheets for Buildings 774, 776/777, 561, 707, and 779. The results were reported as curies of tritium released from each building by month. No raw data were located for any of these buildings. The reconstruction results using the compiled tritium data are presented in Table 2-42.

The reconstruction of a tritium release total for 1974 based on available data resulted in a value of 8.4 curies, which is slightly lower than the emission of approximately 10 curies reported in the Final Environmental Impact Statement (USDOE, 1980). It should be noted, however, that the reconstructed emission total does not include the contributions of several months for which data could not be located for several buildings. The periods of missing data include August and September for Building 774; September, November, and December for Building 779; and March for Building 776. In order to develop an emission value that could be compared with site totals reported by Rocky Flats, tritium releases were estimated for the months for which data were not located.

Estimates were derived for Buildings 774 and 779 by averaging the tritium releases for each building for the months of 1974 for which data were available. This approach assumes that releases were at similar level to those in other months. These values are marked as estimates in Table 2-42. The Building 776 estimate was based on an average of available data from the months of 1974 other than September. September was not included in the average because the majority of the tritium released during that month was due to an incident that is discussed in Section 5 of this report.

When the estimates are substituted for the missing monthly data, the independently calculated site tritium release increases to 9.8 curies. This value is in good agreement with the 1974 airborne tritium release totals of approximately 10 curies reported in the Final Environmental Impact Statement and 10.2 curies reported in the Effluent Information System (EG&G, 1991a).

2.5.4 Results of Tritium Source Term Review

Based on the review of sampling, analytical, and reporting practices for Rocky Flats airborne tritium emissions since 1974 and information describing the nature of tritium handling at Rocky Flats prior to 1974, the emission estimates that will provide the foundation for the reconstruction of off-site doses resulting from airborne tritium effluents will come from the following sources:

TABLE 2-42

SUMMARY OF 1974 AIRBORNE TRITIUM EMISSIONS

Month	Airborne Tritium Released (curies)				
	Building 774	Building 776/777	Building 561	Building 707	Building 779
January	0.82	0.079	-[3]	-[3]	0.13
February	0.67	0.24	-	-	0.024
March	0.81	0.075[2]	-	-	0.11
April	0.80	0.032	-	-	0.12
May	0.93	0.13	0.0025	-	0.018
June	0.44	0.063	0.0013	-	0.032
July	0.40	0.022	0.0024	-	0.024
August	0.54[1]	0.015	0.0025	-	0.10
September	0.54[1]	1.5	0.040	-	0.075[1]
October	0.24	0.057	0.0018	0.0008	0.11
November	0.17	0.043	0.005	0.022	0.075[1]
December	0.10	0.068	0.0042	0.001	0.075[1]
1974 Totals	6.5	2.3	0.060	0.024	0.89
Grand Total	9.8				

[1] Estimated because data were not located for this month. The value presented is based on the average of available release totals for the other months of the year.

[2] Estimated because data were not located for March. The value presented is based on the average of available release totals for the other months of the year excluding September, which was dominated by an incident.

[3] Tritium sampling and analysis began in Building 561 in May 1974 and in Building 707 in October 1974.

- Tritium emissions from 1974 through 1989 will be based on the record of tritium emissions as contained in Rocky Flats Plant annual environmental reports and the Effluent Information System for these years, and the identified uncertainties will be incorporated in these measurement results.

- Tritium emissions before 1974 will be bounded based on available information concerning the nature of tritium handling at Rocky Flats during that period. Approximations will be made of how tritium releases in time spans during this period might possibly have varied compared to emissions of the monitored period from 1974 to 1989.

Airborne tritium emission data for 1953 through 1989 are shown in Table 2-43. Release estimates for the period from 1953 through 1973 are discussed in the following section. The estimates for this earlier period are far less certain than for the period for which monitoring data are available, but the following estimates are not likely to have been exceeded and should allow bounding of the potential health risks from historical tritium releases.

2.5.5 Estimation of Tritium Releases from 1953 through 1973

The largest documented tritium releases from the plant have been those associated with accidents or incidents taking place during the period from 1968 through 1973 when hundreds to thousands of curies of tritium were estimated to have been released to the air. These events are described in Section 6 of this report. Routine emissions, as measured after 1974, were extremely small in comparison, the range typically being one curie or less annually. The incidents that occurred during the period of 1968 through 1974 involved a "special project," a number of occasions of processing of tritium-contaminated materials received from off-site sources, and apparent "special assembly" work. Such activities were likely to have taken place throughout the 1960s, and accidents similar to those reported in the late 1960s and early 1970s may have gone undetected and unreported during earlier times. Investigators were unable to establish whether tritium was present at the plant prior to the early 1960s.

Based on the limited information available regarding tritium release during the period from 1953 to 1967, it is reasonable to assume that releases may have been similar to those identified during the period from 1968 through 1973. The reported release of tritium over the six-year period from 1968 through 1973 has been estimated to be between 840 and 2,340 curies, or about 140 to 390 curies per year. If one made the conservative assumption that such releases occurred each year from 1953 through 1967, the total tritium released over this period would range as high as 5,800 curies. On the other hand, given the lack of information regarding tritium presence and use during this period, it may be equally likely that releases for some years totaled

TABLE 2-43

ESTIMATED ROCKY FLATS AIRBORNE TRITIUM EMISSIONS, 1953-1989
Numbers in Shaded Boxes Were Independently Generated

Calendar Year	Estimated Tritium Release (curies)
1953-1967	15-12,000 (1-800 annually)
1968-1973	840-2,340 (140-390 annually)
1974	9.8
1975	1.5
1976	1.2
1977	0.53
1978	0.90
1979	0.84
1980	0.78
1981	0.44
1982	0.23
1983	0.16
1984	0.14
1985	0.16
1986	0.22
1987	0.17
1988	0.026
1989	0.17
Total	870 to 14,000

no more than approximately one curie annually, consistent with routine releases measured after 1973.

Annual tritium emissions during the period of 1953 through 1967 are therefore likely to have ranged between 1 and 390 curies. To further account for the lack of any quantitative information for the period prior to 1968, an additional uncertainty factor of two will be applied, doubling the range of possible emissions. Annual tritium emissions during the period of 1953 through 1967 will therefore be represented as ranging between 1 and 800 curies with equal likelihood (uniform distribution) for the purposes of estimating off-site exposures. Annual average releases for the period of 1968 through 1973 are estimated to have ranged with equal likelihood between 140 and 390 curies. Tritium releases for the period from 1974 through 1989 are those measured by the tritium effluent monitoring system.

2.6 Airborne Radioactive Effluent Data — Thorium-232

As discussed in the Tasks 3 and 4 report, thorium has been used in relatively small quantities for various applications at Rocky Flats since 1952. Thorium is a metallic element of the actinide series, with several isotopic forms. The thorium-232 isotope is a naturally occurring radioactive form of thorium that decays through a series of alpha, beta, and gamma emissions to finally yield stable lead (lead-210). More than 99.99 percent of natural thorium is thorium-232; the remainder is thorium-230 and thorium-228.

2.6.1 The Nature of Thorium Association with Rocky Flats

Thorium has been used in the following ways at Rocky Flats (Unknown, 1976b):

- The principal use has been fabrication of metal parts from natural thorium metal as well as various alloys of thorium specified by the "customers."
- Thorium oxide, also known as thoria, may have been used as a mold-coating compound in limited experiments.
- Thorium compounds were used in various laboratory analytical procedures and in development programs.
- Twice during the period from 1964 to 1969, "thorium strikes" were performed to remove gamma-emitting thorium-228 from uranium-233 metal needed for fabrication of test devices. The "strikes" involved a fluoride precipitation and filtration process. Natural thorium was used (Putzier, 1982).

- Thorium has also been used as a stand-in or replacement for the more expensive uranium or plutonium components in certain phases of development programs.

- A project involved thorium component production in Building 881 over several years in the late 1950s to early 1960s. Processes used were the same as those used for enriched uranium. The thorium chemical recovery associated with this project occurred at Savannah River or Oak Ridge (ChemRisk, 1991c; RE-891, Interview No. 48).

2.6.2 Possible Magnitudes of Thorium Releases

Review of available information supports the statement in the Rocky Flats Final Environmental Impact Statement (USDOE, 1980) that thorium has been handled in such small quantities as to preclude a significant release. Over the period from 1952 to 1976, which saw the majority of thorium applications at Rocky Flats, the quantity of thorium that was present varied from none to approximately 238 kg in any one month (Unknown, 1976b). Most thorium used was metallic; however, other forms such as oxide or nitrate are known to have been used.

Thorium oxide was never used as a mold-coating compound on a production scale. Thorium compounds used in laboratory analytical procedures and in development programs were in quantities too small to show up on material accountability records. Concentrate from "thorium strikes" were packaged and shipped for burial in Idaho. Quantities of thorium used as a stand-in for uranium or plutonium were too small to show up on material accountability records, but in 1976 the total amount used in stand-in roles was estimated to approach 7 kg (Unknown, 1976b). Special projects involving thorium component production were subjected to containment measures above and beyond those used for uranium because of the added gamma radiation exposure associated with thorium (ChemRisk, 1991c, Interview No. 48).

Inventory control was maintained on thorium compounds when any operating entity within the plant possessed 500 g or more. Quantities less than 500 g were not reportable, and quantities above 500 g were rounded to the nearest kilogram. There are two terms that are useful in bounding quantities of thorium that might have been released:

- Normal Operational Loss (NOL) — the quantity of a source or special nuclear material, in any chemical or physical form, determined by measurement or estimated on the basis of measurement, that has been discarded.

- Material Unaccounted For (MUF) — the difference between a physical inventory of a material and its book inventory after all known removals (such as accidental

losses, normal operational losses, and authorized write-offs) have been reflected in the book inventory.

Between the start-up of the Rocky Flats Plant and 1976, inventory records indicated a cumulative NOL of 32 kg of thorium, with a MUF total of 8 kg (Unknown, 1976b). Given that the primary use of Rocky Flats thorium was in metalworking processes, a large fraction of the material discarded (i.e., Normal Operational Loss) would likely have been in solid wastes or particulates trapped by ventilation exhaust filters. It is therefore likely that less than 32 kg of thorium were released in airborne effluents from the Rocky Flats Plant.

2.6.3 Results of Thorium Release Review

The information reviewed in the course of this project indicates that thorium-232 has not been a significant component of airborne effluents from the Rocky Flats Plant. Because thorium would most likely have been emitted in particulate form, and thorium-232 emits an alpha particle with each decay, thorium emissions are reflected in results of measurements of total long-lived alpha radioactivity that were performed since the early 1950s. Thorium operations have been insignificant relative to the primary production activities centered around plutonium and uranium, and little data exist to support the quantification of release. Therefore, efforts to attribute a portion of total long-lived alpha activity measurements to thorium-232 will not be made.

Although alpha spectrometric analysis procedures were modified around 1976 to include specific identification of thorium nuclides (Hornbacher, 1975-1982), thorium content of airborne effluents has not been routinely reported. Because of the apparent diminishing of applications of thorium compounds since the 1970s, development of source term estimates for thorium-232 during the 1970s and 1980s is not considered to be warranted.

2.7 Potential Airborne Effluents from Criticality Experiments

The presence of large quantities of fissionable materials in numerous forms at Rocky Flats has made it necessary to maintain an active criticality safety program, which was described in some detail in the Tasks 3 and 4 report. The Nuclear Safety Group has conducted its work in the Building 886 Critical Mass Laboratory, which went into operation in 1965. The group performs experiments and calculations to identify container or vessel geometries or arrays of nuclear material that have the potential to lead to fissioning of associated materials. Experiments and calculations are conducted to evaluate the potential for criticality under varying conditions and to validate computer programs used for criticality safety analysis (EG&G, 1991c). Since 1965, the Nuclear Safety Group has conducted about 1600 critical mass experiments using uranium and plutonium. The experiments have involved fissionable materials in solutions (800 tests), compacted powder (300), and metallic forms (500) (Rothe, 1992). Since 1983, criticality

experiments have not been conducted with solid materials. They are now conducted primarily with uranyl nitrate solutions, which are reused (ChemRisk, 1991c).

Airborne effluents from Building 886 have been sampled for radioactive particulates since 1965. Over the period from 1971 through 1989, reported plutonium effluents from Building 886 were at most 5 percent of the site total (in 1978), and enriched uranium emissions were at most 10 percent of the site total (in 1976) (EG&G, 1991a). Releases from Building 886 are included in the assessment of routine effluents from the Rocky Flats site.

Approximately half of the 1600 criticality experiments conducted in Building 886 actually achieved criticality. The experiments were conducted in a manner to control the level of fissioning, for example, by varying distance between pieces of metal and depths of solutions, and only very rarely were the radiation levels such that it was not possible to directly touch the reaction vessels immediately after the experiments. The experiments conducted in the Rocky Flats laboratory generally involved power levels of no more than 10 milliwatts for no more than one hour (ChemRisk, 1991c, Interview No. 53). There were approximately six "high-power" experiments that were taken to between 10 and 100 times the power of typical tests (Rothe, 1992). Using a conversion factor of 3×10^{16} fissions per megawatt-second (Thomas, 1978), this power level and duration correspond to a maximum of 1×10^{12} fissions from a typical Rocky Flats Plant criticality experiment and a maximum of 1×10^{14} fissions from a high-power experiment. For purposes of comparison, the documented criticality accidents in the U.S. (Stratton, 1967) yielded from about 3,000 to 40 million times more fissions than the maximum from a typical Rocky Flats Plant experiment.

Radioactivity possibly released from the Critical Mass Laboratory included enriched uranium and plutonium and fission products formed in fission of these materials. Fission products in the Rocky Flats solutions have been reported to be nearly unmeasurable; there has been no need for monitoring of fission product levels, administrative limitation of concentrations, or purification treatment of the solutions because fission product buildup has been insignificant (Rothe, 1992). While fission products are generally liberated from test solutions, they largely remain trapped in metal and compacted powder test specimens. The power levels of the Rocky Flats experiments have been much lower than those required to vaporize metals (Rothe, 1992).

Calculations have been performed to estimate the releases of fission products that would result from the presumed maximum credible criticality accidents at the Rocky Flats Plant (USDOE, 1980). These calculations addressed the consequences of criticality accidents with plutonium metal ingots and with plutonium solution. While criticality tests at Rocky Flats have not used plutonium solutions (Rothe, 1992), the plutonium solution accident assessments should serve to bound the potential impacts of solution criticalities. In each of the cases analyzed for the safety analysis, transmission of 0.0002 percent of liberated particulates through HEPA filters was

assumed. It was also assumed that 0.05 percent of the plutonium in solution becomes airborne and that 25 percent of halogen and 100 percent of noble gas fission products are released. The total number of fissions was calculated to be 8×10^{18} for the plutonium metal accident and 2.2×10^{20} for the solution criticality case (USDOE, 1980).

Releases from historical Rocky Flats criticality experiments were conservatively estimated by scaling down the calculated releases for the maximum credible criticality accidents by the ratio of the maximum number of fissions for Rocky Flats tests (1×10^{12} per typical test and 1×10^{14} for each high-power test) to the number of fissions calculated for each maximum credible criticality accident. Based on the reported frequencies of experiment types (Rothe, 1992), it was assumed that 800 tests have achieved criticality and that three-quarters involved solutions and one-quarter involved fissionable metal. Release estimate totals are presented in Table 2-44 and include halogen (bromine and iodine) and noble gas (krypton and xenon) fission products and residual quantities of the fissionable radionuclides and the nuclides that accompany them in weapons grade special nuclear material. Radionuclides with half-lives of less than one hour have been omitted from this table.

The estimated plutonium release from criticality experiments totals 0.1 microcurie, which made an insignificant contribution to the reported normal operational release of over 8,000 microcuries from plutonium facilities during the period of interest from 1965 through 1989 that the Critical Mass Laboratory was active (USDOE, 1980; EG&G, 1991a). Noble gases are chemically inert and would have resulted in off-site exposure primarily by immersion in the cloud as it passed by any individuals; once the cloud passed by, dose was no longer delivered to the individuals. Inhalation exposure to noble gases is generally considered to be insignificant; inhalation dose factors are typically not provided for noble gas nuclides due to the relative insignificance of inhalation exposure. The listed bromine isotopes can result in inhalation doses to the lung and other organs, but their half-lives are 35 hours or less, and inhalation dose factors (committed effective dose equivalent per unit intake) are at least a factor of 4,000 less than those for isotopes of plutonium (USEPA, 1988).

Radioactive iodine isotopes, which concentrate in the thyroid gland when inhaled or ingested, have half-lives of eight days or less and inhalation dose factors that are at least a factor of 1,800 less than those for isotopes of plutonium.

Summary:	Criticality experiments were not likely to have been the source of significant releases of radionuclides from the plant.

TABLE 2-44

ESTIMATED RADIONUCLIDE RELEASES
FROM ROCKY FLATS CRITICALITY EXPERIMENTS

Radionuclide[1]	Release from 600 Solution Criticality Tests (Ci)	Release from 200 Metal Criticality Tests (Ci)	Release from 6 High-Power Solution Tests (Ci)
Bromine-80m	8.6e-09[2]	0.0	2.3e-14
Bromine-82	9.4e-07	0.0	2.6e-12
Bromine-83	1.1e-03	5.2e-05	2.9e-09
Krypton-83m	4.1e-03	2.0e-04	1.1e-08
Krypton-85m	4.0e-03	1.8e-04	1.1e-08
Krypton-87	2.5e-02	1.0e-03	6.8e-08
Krypton-88	1.6e-02	6.5e-04	4.4e-08
Iodine-130	4.0e-06	7.1e-08	1.1e-11
Iodine-131	1.7e-04	7.3e-06	4.6e-10
Iodine-132	2.0e-02	8.2e-04	5.4e-08
Iodine-133	2.9e-03	1.1e-04	7.9e-09
Iodine-135	8.2e-03	3.5e-04	2.2e-08
Xenon-131m	3.4e-04	0.0	9.2e-10
Xenon-133	1.4e-03	5.6e-05	3.8e-09
Xenon-133m	1.2e-04	5.3e-06	3.4e-10
Xenon-135	1.8e-02	7.1e-04	4.9e-08
Plutonium-238	1.1e-13	4.6e-11	3.1e-19
Plutonium-239	3.8e-12	1.6e-09	1.1e-17
Plutonium-240	8.9e-13	3.6e-10	2.4e-18
Plutonium-241	2.9e-11	1.2e-08	8.0e-17
Plutonium-242	7.8e-17	3.2e-14	2.1e-22
Americium-241	9.7e-13	3.9e-10	2.6e-18

[1] Radionuclides with half-lives of less than 1 hour have been excluded.
[2] 8.6e-09 equals 8.6×10^{-9}, or 0.0000000086.

2.8 Summary of Radionuclide Release Estimates

The emission estimates that will provide the foundation for the reconstruction of off-site doses from Rocky Flats airborne radioactive effluents are shown in Table 2-45. These estimates are based on the following sources:

- Plutonium emissions for calendar years 1957, 1963, 1969, 1973, 1984, and 1986 are based on independent calculations of release totals performed in this study. Plutonium emissions for other years are based on the annual effluent estimates presented in the Final Environmental Impact Statement (USDOE, 1980) for the years 1953 to 1977 and the DOE Effluent Information System for 1978 to 1989. As described in Section 2.4, airborne emissions of plutonium alpha activity, Am-241, and Pu-241 were estimated based on total long-lived alpha measurements for 1953 through 1973. Pu-239/240 measurements were the bases for estimation of plutonium alpha activity emissions for 1974 through 1989, Am-241 and Pu-241 emissions for 1974 through 1984, and Pu-241 emissions from 1985 through 1989. Americium-241 measurements were reported beginning in 1985.

- Enriched uranium emissions for 1954 through 1957, 1963, 1969, 1973, 1984, and 1986 are based on independent data reconstructions performed as part of this study. Enriched uranium emissions for other years are based on annual effluent estimates presented in the Final Environmental Impact Statement (USDOE, 1980) for the years 1958 to 1977, and the DOE Effluent Information System for the years 1978 to 1989. No monitoring data were located for 1953. Enriched uranium operations in 1953 were limited, and it is assumed that 1953 uranium emissions are equal to reconstructed values for 1954. As described in Section 2.4, measurements of total long-lived alpha activity in effluents from enriched uranium areas were the bases for enriched uranium emission estimates for 1953 through 1977. Enriched uranium emission estimates for 1978 through 1989 were based on U-233/234 measurements reported or reconstructed for that time period.

- Depleted uranium emissions for 1954 through 1960, 1963, 1969, 1973, 1984, and 1986 are based on independent data reconstructions performed as part of this study. Depleted uranium emissions for other years are based on the annual effluent estimates presented in the Final Environmental Impact Statement (USDOE, 1980) for the years 1961 to 1977, and the DOE Effluent Information System for the years 1978 to 1989. Uranium emissions from 1953 are assumed to equal 1954 reconstructed values. As described in Section 2.4, measurements of total long-lived alpha activity in effluents from depleted uranium areas were the bases for depleted uranium emission estimates for 1953 through 1977. Depleted uranium emission estimates for 1978 through 1989 were based on U-238 measurements reported or reconstructed for that time period.

TABLE 2-45

ESTIMATED ROCKY FLATS AIRBORNE RADIOACTIVITY EMISSIONS, 1953-1989 (μCi)

Year	Plutonium Alpha	Depleted Uranium	Enriched Uranium	Plutonium-241	Americium-241	Tritium
1953	1.6	1,200	9.1	9.3	0.36	1.5×10^{7} to 1.2×10^{10} (1-800 Ci y^{-1})
1954	53	1,200	9.1	270	12	
1955	59	2,100	74	300	13	
1956	190	1,000	1,100	960	41	
1957	12,000	730	360	63,000	2,700	
1958	2,500	1,600	310	13,000	560	
1959	1,100	270	540	5,800	250	
1960	1,100	350	860	5,400	230	
1961	1,200	520	480	6,300	270	
1962	2,500	370	250	13,000	540	
1963	3,000	490	330	15,000	650	
1964	2,300	240	190	12,000	500	
1965	5,300	280	190	27,000	1,200	
1966	260	140	230	1,300	58	
1967	330	140	110	1,700	72	
1968	400	140	160	2,000	88	8.4×10^{8} to 2.3×10^{9} (140-390 Ci y^{-1})
1969	1,100	160	50	5,800	250	
1970	310	190	64	1,600	68	
1971	61	58	41	310	13	
1972	50	42	4	250	11	
1973	51	54	12	260	11	
1974	960	9	27	5,000	210	9,800,000
1975	10	28	28	52	22	1,500,000
1976	4.0	12	16	21	0.88	1,200,000
1977	4.0	19	21	21	0.88	530,000
1978	2.8	33	21	15	0.62	900,000
1979	5.5	26	9.2	29	1.2	840,000
1980	12	15	15	62	2.6	780,000
1981	8.2	18	12	43	1.8	440,000
1982	20	19	12	100	4.4	230,000
1983	78	31	20	410	17	160,000
1984	78	5.5	20	410	17	140,000
1985	9.2	39	7.9	48	2.0	160,000
1986	29	2.9	11	150	9.1	220,000
1987	15	12	4.6	78	3.2	170,000
1988	15	9.3	2.6	78	2.0	26,000
1989	4.5	2.5	5.2	23	1.1	170,000

- Airborne tritium emissions for 1974 are based on an independent data reconstruction performed as part of this study. Tritium emissions for other years beyond 1973 are based on the records of tritium emissions contained in Rocky Flats Plant annual environmental reports and the Effluent Information System. Tritium emissions before 1974 have been bounded based on available information concerning the nature of tritium handling at Rocky Flats during that period.

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3.0 ROUTINE AIRBORNE EMISSIONS OF NONRADIOACTIVE MATERIALS

The preceding sections of this report have focussed on the radioactive materials of concern and the extensive airborne effluent monitoring program that has been in operation to quantify the release of these materials. As part of initial project efforts, materials of concern were identified based on their potential to cause off-site health impacts (ChemRisk 1991a, 1991b, 1992). These materials include both radioactive and nonradioactive materials. The nonradioactive materials of concern that were identified as warranting the development of emission estimates for routine release include:

- Beryllium
- Carbon Tetrachloride
- Chloroform
- Methylene Chloride
- Tetrachloroethylene
- 1,1,1-Trichloroethane
- Trichloroethylene

With the exception of beryllium, all of these materials are organic solvents that are liquids with relatively high vapor pressures, so that they will readily volatilize in air. Beryllium is a metallic element that is commonly found in solid form.

A beryllium effluent monitoring program was routinely conducted throughout most of the plant's operating history. However, there was no routine sampling of the organic solvents of concern. The following sections review the beryllium sampling program, provide a summary of the beryllium emission estimates, and also describe the approach to estimating emissions of each of the organic solvents based on available data and information.

3.1 Beryllium

The history of beryllium use at Rocky Flats has been summarized in the Tasks 3 and 4 report (ChemRisk, 1992). Beryllium has nearly always been present at Rocky Flats, but it was not actually used in full-scale production operations until 1958. Beryllium operations were not part of the manufacturing process in the first years of plant operation, but beryllium was being handled by the production engineering group in preparation for application to a new weapons concept. These early beryllium operations took place in Building 444 in preparation for regular pit production, which began in 1958. Beryllium manufacturing operations in Building 444 included casting (foundry), cutting, heat-treating, rolling, and machining. Beryllium foundry operations ceased in 1975.

Historically, beryllium has been present and monitored in a number of plant buildings. Some of these other buildings include the "B" side of Building 883, which in 1964 was converted to beryllium rolling and forming operations. In 1970, Building 865 began operations, serving as a research and development facility primarily for the manufacturing processes using uranium and beryllium. Recent-day uses of beryllium documented in Air Pollution Emission Notice reports from the late 1980s also include vapor deposition to coat metal parts in Building 705.

The Tasks 3 and 4 report described the nature of the ventilation systems used to control beryllium emissions. When manufacturing started in 1958, the system consisted of "Aero-Tech" cyclone separator units (to remove the larger particles from the exhaust stream) exhausting to the main building exhaust serving the uranium operations, which was subject to HEPA filtration. Upgrades in 1964 and 1974 improved on the cyclone separator units, and in 1986 Building 444 exhaust filtration was upgraded to include two stages of HEPA filtration.

Beryllium has been monitored in plant exhaust systems since at least 1960. It is currently monitored in 50 vents, although it is actually processed in only six of the associated areas according to plant reports (EG&G, 1990). According to the 1980 Final Environmental Impact Statement (USDOE, 1980), beryllium was among eleven elements analyzed by atomic absorption in waterborne effluents.

As a metallic element present in particulate form in airborne plant effluents, beryllium is the only nonradioactive material of concern in this study that would have been controlled by HEPA filtration that was installed at the plant for the purpose of controlling radioactive effluents. While some concerns have been raised about the control of beryllium in workroom air, virtually all airborne effluents discharged from plant facilities that processed beryllium were subject to HEPA filtration. One exception may have been Building 441 during the early 1960s. An industrial hygiene report from 1963 suggests that beryllium discharges from Building 441 were relatively high and recommended that exhaust hood filters be installed in the system (Hammond, 1963). Exhaust hood filters were not installed in Building 441 until sometime in 1964. The beryllium effluent monitoring data for Building 441 reflect the effect of increased filtration in reduced emissions. Interviewees have indicated that Building 441 housed a beryllium analytical laboratory. The following sections describe the plant program for monitoring beryllium in airborne plant effluents and summarize the emission data generated as a result of that program.

3.1.1 Historical Effluent Sampling and Analytic Practices — Beryllium

The beryllium monitoring program was the responsibility of the industrial hygiene group at Rocky Flats. The records searches for this study identified few documents pertaining to beryllium sampling and analysis, particularly for the 1950s and 1960s. A Rocky Flats retiree indicated that information relating to beryllium sampling was summarized in the industrial

hygiene monthly progress reports; however, a comprehensive source of these documents could not be located. While it has been reported that Rocky Flats operated a beryllium sampling program during the 1950s, no documentation of such a program or sampling data have been located for this period.

The sampling practices for beryllium are reported to be very similar to those employed for radionuclides, which is logical given the similarity in the physical characteristics of the beryllium, plutonium, and uranium effluents. The sampling system for each of these materials must be able to collect very small metal particles remaining in the effluent airstream after HEPA filtration. Documentation of the sampling programs from the late 1970s and 1980s indicates that in many cases individual samples collected from ducts or buildings were analyzed for both radioactive contaminants and beryllium. During the 1980s, beryllium analysis was being done on a monthly basis from a ventilation system composite filter sample. However, the plant used a number of different beryllium analytic techniques over time that may have resulted in separate sample collection during some periods of plant operation. The historical radioactive effluent sampling practices relative to sampling system design for particulates, sample apparatus flow rates and velocities, sample line losses, and stack or vent exhaust volume quantification discussed earlier in this report are generally applicable to beryllium sampling, as are the uncertainties in the final emission estimates.

A number of analytic practices were employed historically by Rocky Flats to quantitate beryllium in effluent samples. While the exact periods during which the various practices were used could not be established from plant documentation, a number of published reports and plant procedures describe the beryllium analytic procedures and personnel interviews at the plant that aided in identifying the periods during which the various methods were used. The first reference to beryllium analytic procedures was located in an internal Dow Chemical Company document from January 1964, which provided instrument operation instructions for beryllium quantification using emission spectroscopy (Dow Chemical, 1964). The document described the operation of a spectrograph that utilized photomultiplier tubes that replaced photographic plates as a detector and that was capable of quantitating beryllium in any form in the range of 0.005 to 1000 μg on Whatman 41 filter paper. Plant personnel indicated that emission spectrographs were used during the period from approximately 1958 to 1965.

The next discussion of analytic methods for beryllium related to Rocky Flats was located in a journal article from the scientific literature (Bokowski, 1968). The paper describes the application of direct-reading atomic absorption spectrophotometry (flame) to the analysis of beryllium in air on Whatman 41 filter paper as well as other types of samples. The paper documented good accuracy and precision for the method and an ability to quantitate beryllium in aqueous solution as low as 0.003 micrograms per milliliter. The approximate period of use of this method was 1965 to 1971.

The third significant document addressing beryllium analysis described a study of the use of nonflame (graphite furnace) atomic absorption spectrometry for quantifying submicron amounts of beryllium (Hurlbut and Bokowski, 1974). The discussion of the nonflame method examined the use of both Whatman No. 41 filters and Gelman Type E glass filters and found some analytic interference with the use of the glass filters. The study reported that comparison of results obtained by standard flame atomic absorption analysis and nonflame atomic absorption analysis was good, but the nonflame technique has the advantage that as little as 0.002 micrograms of beryllium per paper filter can be detected, whereas the direct flame technique is limited to about 2 micrograms of beryllium per filter paper. This method has been in use at Rocky Flats from approximately 1971 to the present.

In addition to these documents generated by Rocky Flats employees describing various analytic methods, a fairly lengthy review paper on beryllium prepared by a Rocky Flats employee was also located during records searches. The paper provides a literature review of the history, uses, occurrences, analytic chemistry, and biochemistry of beryllium (Hurlbut, 1974).

While the documents do not specifically describe the programs that were in place for the collection and analysis of routine air samples at Rocky Flats, they do demonstrate that Rocky Flats scientists were actively developing and evaluating accurate and precise methods for quantitating beryllium in environmental samples. This record suggests that some care was taken in the sampling and analysis of airborne plant effluents for beryllium.

3.1.2 Airborne Effluent Data — Beryllium

As described earlier, the industrial hygiene group was responsible for the beryllium sampling program at Rocky Flats and generated documentation of the program. A relatively complete record of the annual beryllium emissions was compiled from detailed sample data logbooks for 1960 through 1970 and annual beryllium releases reported in the Annual Environmental Monitoring Reports for 1971 through 1989. No sampling data was located for the period prior to 1960.

The logbooks from the period of 1960 through 1970 contained daily sample results for both workroom air and building effluents. The building effluent data was entered onto a computerized spreadsheet to calculate the monthly average beryllium concentrations for each stack as well as the annual averages for 1960 through 1970, which are presented in Table 3-1. In the case where less-than values were reported in the logbook, one-half the reported value was used to calculate the averages. The method of replacing less-than values with one-half the limit of detection is supported by the USEPA (1989) and Gilbert (1987). Gilbert states:

TABLE 3-1

ANNUAL AVERAGE BERYLLIUM CONCENTRATIONS
($\mu g\ m^{-3}$)

Building and Release Point	Calendar Year										
	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970
331	ND	0.0022 (7 mo.)	0.022	0.0025	0.0044	0.020	0.00075 (4 mo.)	NA	NA	NA	NA
441	0.77 (3 mo.)	0.16	0.067 (11 mo.)	0.092	0.018	0.028 (11 mo.)	0.039 (4 mo.)	NA	NA	NA	NA
444 Duct 2	ND	ND	0.00039 (7 mo.)	0.00099	0.0024	0.0083	0.0065	0.0049	0.013	0.0039	0.0036
444 Duct 3	0.000060	0.0000055	0.00047	0.0018	0.0026	0.0062	0.011	0.0073	0.0067	0.0031	0.0027
447	ND	ND	ND	0.0019 (11 mo.)	0.0029	0.0060	0.0076	0.0052	0.0025	0.0033	0.0020
774	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.00014
779	ND	ND	ND	ND	ND	ND	ND	0.0015 (11 mo.)	0.00088	0.0012	0.00034
883-A	NA	NA	NA	NA	NA	NA	0.0012 (11 mo.)	0.0069	0.0024	0.0078	0.0017 (11 mo.)
886	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.0010
991	ND	ND	ND	ND	0.0050 (5 mo.)	0.0097	0.00050 (1 mo.)	ND	ND	ND	ND

NA = No Beryllium Processing Activity
ND = No Beryllium Monitoring Data Located

> The method of replacing less-than values by one-half the limit of detection is unbiased for the mean, but not for the variance, if the analytical measurement technique cannot result in negative measurements, and if all measurements between zero and the Limit of Detection are equally likely to occur (a uniform distribution). Kushner (1976) studied this method when aerometric data below the detection limit are lognormal. For his application (pollution data) he concluded that biases using the mid-point would be overshadowed by measurement error.

In order to evaluate the impact of replacing less-than values with one-half the limit of detection on the release estimates for beryllium, a more detailed analysis was performed on the 1962 and 1968 beryllium monitoring data. Although about 30 percent of the air monitoring data reported in 1962 were below the applicable detection limit (0.0001 $\mu g\ m^{-3}$ in most instances), their impact on the release estimate for the year was relatively small. This was demonstrated by calculating annual averages setting less-than values equal to the detection limit and then setting them to zero. The difference between beryllium release estimates for 1962 calculated by these two methods was less than 2 percent. In 1968, less than 1 percent of beryllium air monitoring results were below detection limits, and the difference between release estimates calculated by the two methods described above was less than 1 percent.

As was the case with quantifying radioactive effluents, exhaust flow rates or total exhaust volume must be known to calculate total emissions based on the sampling data. Again, the estimates of these values presented in Table 2-2 had to be used since there is little documentation of this information from the 1960s. An additional problem was encountered in that there was no documentation of ventilation flow rates or volumes for Building 441 from the sources used to compile Table 2-2 since Building 441 was converted from production use to office use in the mid-1960s. In order to estimate beryllium emissions from Building 441, it was necessary to assume that exhaust ventilation rates were equivalent to those of a building of approximately the same size, Building 331. Building 331 is somewhat larger at approximately 23,000 versus approximately 18,000 square feet for Building 441. Both buildings served as research and analytic laboratories in the early 1960s. Given Building 331's larger size, ventilation volumes may have been larger than those of Building 441; however, this would lead to conservative emission estimates (i.e., would result in an overestimate of emissions rather than an underestimate).

The annual beryllium emission estimates for the period from 1960 through 1970 calculated from data compiled from sample data logbooks and using exhaust volume estimates are presented in Table 3-2. As indicated in the table, beryllium data are not consistently available for the identified buildings, and in most cases this is a result of changes in building uses such as:

TABLE 3-2

ANNUAL BERYLLIUM RELEASES BY BUILDING (grams)

Building	1960	1961	1962	1963	1964	1965	1966	1967	1968	1969	1970
331	ND	0.09	1.5	0.1	0.3	1.4	0.02	NA	NA	NA	NA
441	13	11	4.3	6.4	1.2	1.8	0.9	NA	NA	NA	NA
444	0.064	0.01	0.9	3.7	6.9	21	23	17	31	9.3	9.3
447	ND	ND	ND	1.6	2.8	5.6	7.2	4.9	2.3	2.8	1.9
774	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.03
779	ND	ND	ND	ND	ND	ND	ND	0.03	0.2	0.3	0.08
883	NA	NA	NA	NA	NA	NA	1.8	11	4.1	13	2.6
886	ND	ND	ND	ND	ND	ND	ND	ND	ND	ND	0.06
991	ND	ND	ND	ND	0.3	1.3	0.005	ND	ND	ND	ND
YEARLY TOTALS	13	11	6.7	12	12	31	33	33	38	25	14

NA = No Beryllium Processing Activity
ND = No Beryllium Monitoring Data Located

- Building 883 — beryllium operations moved in around 1965, and
- Building 441 — beryllium operations moved out around 1966.

It is believed that the plant initiated a broader sampling program for beryllium in 1970 that led to the inclusion of Buildings 886 and 774. The broader sampling program was likely a result of greater environmental awareness. Beryllium was not machined in either building, but may have been present in wastes in Building 774 or parts in Building 886. No data for beryllium emissions were located for the period prior to 1960. While it is likely that beryllium was present on-site during initial years of operation, it was not used in the manufacturing process until 1958, and this use was only in Buildings 441 and 444. In the absence of any data, it is believed reasonable to assume that emissions from these buildings during 1958 and 1959 were approximately the same as those reported in 1960.

Beryllium releases for the period from 1971 through 1989 have been documented in the Annual Environmental Monitoring Reports issued by the plant. In many cases, the Annual Environmental Monitoring Reports reported beryllium release totals as less-than values. The 1975 annual report stated that samples with concentrations below the minimum detectable concentrations (MDC) were considered to be at the MDC for averaging purposes. Averages calculated with below-MDC results were identified with a less-than sign (<). In addition, the annual emission total for 1983 was reported as a negative number. The explanation in the text of the document was that this indicated that the air sampled for the year could not be distinguished from the background level associated with the analysis. A summary of the annual beryllium release totals for this period is presented in Table 3-3.

A beryllium release total for 1984 was independently calculated by ChemRisk as part of this study. In order to perform the calculations, a data base of the analytical results of beryllium emissions was created. The calculated airborne beryllium release total for 1984 is 0.31 grams. Release totals for individual stacks for 1984 are contained in Table 3-4. The value of 0.31 grams is in good agreement with the value of 0.3 grams reported in the Rocky Flats plant 1984 Annual Environmental Report and presented in Table 3-3.

Concern has been identified for potential beryllium release during fires. Three reports of fires in beryllium areas at Rocky Flats were located. Two of the fires, one on June 25, 1962 and one on February 14, 1964 were confined to workrooms in Building 444. Any beryllium released as a result of the fires would have passed through the plenums and been monitored by the stack sampling equipment (Reyland and Rogers, 1964; Boatman, 1962). The third fire reported in Rocky Flats literature occurred on February 23, 1978 in the plenum building serving Building 444. Based on witness accounts the report indicates that the HEPA filters continued to function until water was applied. However, the fire did burn through the plenum prefilters. The stack

TABLE 3-3

REPORTED ANNUAL BERYLLIUM RELEASES

Year	Release (grams)
1971	16
1972	<2.0
1973	<7.1
1974	<10
1975	<5.2
1976	<3.7
1977	<4.9
1978	<17
1979	<1.5
1980	<1.1
1981	0.2
1982	0.1
1983	- 0.1
1984	0.3
1985	0.5
1986	0.1
1987	0.2
1988	0.1
1989	0.6

Sources of Data: Rocky Flats Annual Environmental Reports

TABLE 3-4

1984 BERYLLIUM EMISSIONS DATA

Location	Total 1984 Release (grams)
371-NO1	6.9×10^{-4}
371-NO2	1.6×10^{-3}
371-SSS	1.0×10^{-2}
374-MAI	5.2×10^{-4}
374-SPD	6.8×10^{-4}
444-D02	9.3×10^{-2}
444-DO1	2.1×10^{-2}
444-DO5	7.3×10^{-3}
447-MAI	3.0×10^{-3}
559-561	1.7×10^{-2}
707-101	9.4×10^{-5}
707-102	8.2×10^{-4}
707-105	2.1×10^{-3}
707-106	1.1×10^{-4}
707-107	6.7×10^{-4}
707-108	2.7×10^{-3}
771-C Roo	7.5×10^{-4}
771-C Main	4.2×10^{-4}
771-Main 2	5.2×10^{-3}
771-Main	1.2×10^{-2}
774-202	5.5×10^{-4}
776-201	1.2×10^{-3}
776-202	8.2×10^{-4}
776-204	6.6×10^{-3}
776-205	3.8×10^{-4}
776-206	3.4×10^{-3}
776-251	2.5×10^{-3}

(continued on next page)

TABLE 3-4 (Continued)

1984 BERYLLIUM EMISSIONS DATA

Location	Total 1984 Release (grams)
776-252	3.4×10^{-3}
776-50C	1.7×10^{-3}
776-50D	2.8×10^{-3}
778-LDY	1.2×10^{-2}
779-729	1.8×10^{-2}
779-782	9.1×10^{-3}
865-EEE	6.7×10^{-3}
865-WWW	2.3×10^{-3}
881-D12	3.6×10^{-3}
881-D34	5.2×10^{-3}
881-D56	7.4×10^{-3}
883-AAA	1.5×10^{-2}
883-BBB	1.8×10^{-2}
886-875	2.7×10^{-3}
889-MAI	1.4×10^{-3}
991-985	5.8×10^{-3}
991-MAI	2.9×10^{-3}
TOTAL	**0.31 grams**

air monitoring equipment operated throughout the incident and the report quantifies the beryllium released to ambient air at 14.5 grams (Hess, 1978). The water used to fight the fire was impounded in areas south and east of Building 444.

The beryllium concentrations in this water were 4.3 mg L^{-1} and 1.6 mg L^{-1}, respectively. Because the stack monitoring equipment was operating during these fires, the beryllium routine release totals for 1962, 1964 and 1978 include any releases associated with these fires.

The beryllium release summaries suggest extremely low environmental emissions of beryllium averaging in the tens of grams or less annually. A 1980 plant internal letter indicated that, based on an evaluation by the plant's General Service Laboratory, use of the minimum detectable amount (MDA) value for beryllium at each effluent measurement location would result in a calculated minimum beryllium discharge per month of 0.4 gram (Hornbacher, 1980). This would lead to a reported yearly minimum discharge of about 4 to 5 grams even if none of the samples had a positive analysis result. The information that was reviewed suggests that the beryllium data handling practices may have led to the reporting of annual emissions that were higher than the actual releases. However, given the low magnitude of the reported emissions, the uncertainty introduced by this practice has not been characterized.

The sources of uncertainty related to the collection of samples and the quantification of exhaust volumes discussed for plutonium and uranium measurements also apply to beryllium measurements. Therefore, the range of beryllium emissions was characterized using the same approach described for quantifying uncertainties associated with plutonium and uranium release estimates. This approach included uncertainties in exhaust flow rate estimates, in sampling flow rate estimates, and in analytical results. As described in Appendix G, Monte Carlo simulation was used to combine the different sources of uncertainty and calculate the overall uncertainty factor of airborne beryllium emissions.

3.2 Organic Solvents — Airborne Emissions

The organic solvents that have been identified as being of potential concern at the Rocky Flats Plant have been used in a variety of applications. The nature of recent-day use (late 1980s) of these materials has been exhaustively evaluated in reports prepared by EG&G Rocky Flats titled Air Pollution Emission Notices (APEN). APEN project work involved more than thirty full-time project personnel and ten part-time personnel devoted to the project for over a year and a half preparing reports for essentially every Rocky Flats building. Each APEN report documents the configurations of the modern-day air handling systems, the processes conducted in each building, vents and/or stacks associated with emissions, and assumptions and factors used by EG&G and its subcontractors to calculate process emissions. The APEN reports describe modern-day plant processes and activities. While these reports provide an excellent resource

for establishing current-day releases of the organic solvents of interest, they are generally not a useful source of historical information.

The extent and quality of historical documentation of the use and release of the organic solvents are meager at best in comparison to the documentation provided by the APEN reports. In many cases, the actual nature of the historical use of the material is poorly documented, and in all cases there is insufficient historical information on material purchases, uses, disposal, and recycling to apply any of the estimating techniques such as mass balance calculations or use of emission factors that were employed to prepare the APENs. The historical information that is available consists of some limited historical inventory information, limited documentation of specific usage, reports of special effluent sampling or quantification efforts, and information obtained as a result of personnel interviews. Due to the limited availability of historical data, plausible ranges of historical emissions were estimated for screening purposes only, to establish a basis for determining level of concern.

Unlike radionuclides and beryllium, there were no efforts at Rocky Flats to control organic solvent emissions by use of exhaust system recovery devices, scrubbers, or traps. Because of this fact, significant fluctuations due to failure of control devices did not occur. Emissions were more directly related to the quantities of each solvent that were historically used and the fractions of the quantities that were used that became airborne.

The following sections review the information that was identified as being useful for the development of emission estimates for each of the organic solvents of concern. In cases where more historical data were available, an estimate of the probable range of the historical annual airborne release is provided. The estimated range of release has been established following the review of all the identified data. The data often only provide a rough guide to the actual possible emissions, but the estimated range of emissions derived from the data is believed to encompass the actual emission rates. In cases where no data or information is available for a given period of time, it has been assumed that emissions were the same as those estimated during the nearest point in time for which some type of information is available. Where data and information indicate trends in release rates over time, a simplifying assumption of linear change over time is made in the absence of information to the contrary.

Detailed documentation of all available sources of information on emission estimates for the organic solvents of concern can be found in Appendix E.

3.2.1 Carbon Tetrachloride

As described in the Tasks 3 and 4 report, carbon tetrachloride was used at Rocky Flats to clean glove-box walls, furnaces, product components, metal chips, machinery, and instruments. Estimated historical emission rates of carbon tetrachloride have been based on airborne concentration measurements, estimates of quantities of the material used or kept on hand, and statements made by retired and active workers during interviews. The time period of interest from 1953 to 1989 can be broken into three distinct time spans relative to estimates of annual emission quantities based on the available information and factors discussed below.

1953 to 1957

Investigations have indicated that carbon tetrachloride was used extensively in plutonium operations, but not in uranium, beryllium, or stainless steel operations. Large-scale plutonium production did not begin until approximately 1957, when a change in weapon design concept called for increased use of plutonium in fissionable weapon components. In these early years, plutonium was machined in a "dry" state, that is without use of cutting oils. Carbon tetrachloride was used as a coolant, sometimes being applied with a squeeze bottle (ChemRisk, 1991-1992, Interview No. 78). Uranium machining during these years was typically performed with a 15:1 oil and tetrachloroethylene mixture (Navratil and Miner, 1984).

Plant personnel were cognizant of the hazards of carbon tetrachloride use as early as 1953, but documented efforts at solvent substitution in these early years were limited to non-production operations, such as cleaning of typewriters and desks (Hicks and Langell, 1952). Tetrachloroethylene and trichloroethylene were recommended in these applications.

It is estimated that carbon tetrachloride emissions prior to 1958 were approximately one-tenth of emissions from 1958 to 1970, or ranged between 4 to 20 tons per year. Given the documented, albeit relatively minor, uses of the solvent during this period, it is unlikely that annual uses or emissions were below about 600 gallons or 4 tons or that emissions exceeded 10 percent of those experienced after the significant increase in plutonium machining that occurred around 1957.

1958 to 1970

The period from 1958 to 1970 was the period of maximum use of carbon tetrachloride at Rocky Flats. Because many major uses of carbon tetrachloride were tied to routine maintenance activities, for example glove-box cleaning during regularly scheduled inventories, emissions of the solvent are thought to have been relatively constant during this period rather than following any ups and downs of production rates. Based on available information, it is believed that the

transition to large-scale plutonium production may have taken up to three years, from 1958 to 1961. By 1961, carbon tetrachloride emissions had reached levels similar to those of the early 1970s before solvent substitution began to decrease carbon tetrachloride use. Based on documentation of early 1970s use, measurement, and inventory data described below, estimates of carbon tetrachloride emissions range between 40 and 200 tons per year for 1961 to 1970.

The upper bound of 200 tons per year is approximately 30 percent above the release rate (153 tons per year) that corresponds to peak levels of carbon tetrachloride measured around 1974/1975 by Hobbs (1982). The upper bound is above the estimated 1974 peak emission rate to account for some decrease in carbon tetrachloride use beginning around 1972. The fact that carbon tetrachloride usage was in a downward trend was likely offset somewhat by the fact that the 153 tons per year estimate reflects peak measurements rather than average emissions over long periods of time. The elimination of uses of carbon tetrachloride at Rocky Flats was neither immediate nor comprehensive- significant uses remained in 1970. In fact, at the time of plant shutdown in 1989, procedures still called for use of carbon tetrachloride in plutonium facilities.

Between 1958 and 1968, approximately 3,500 drums containing plutonium-contaminated oil were stored at the Building 903 drum storage area (Seed *et al.*, 1971). These drums included carbon tetrachloride in varying proportions with straight-chain hydrocarbon mineral oil (Shell Vitrea) and other liquids. Assuming that each 55-gallon drum contained 20 percent carbon tetrachloride, the amount stored over the 10-year period is estimated to have been approximately 256 tons. This is equivalent to about 26 tons per year or about 13 percent of the estimated annual emission of that period of time. Since only a fraction of the stored carbon tetrachloride was released into the environment through leakage of storage drums, the assumption of complete volatilization of the carbon tetrachloride used at Rocky Flats that is the basis of various historical emission estimates apparently results in the estimates being conservative in this regard.

1971 to 1989

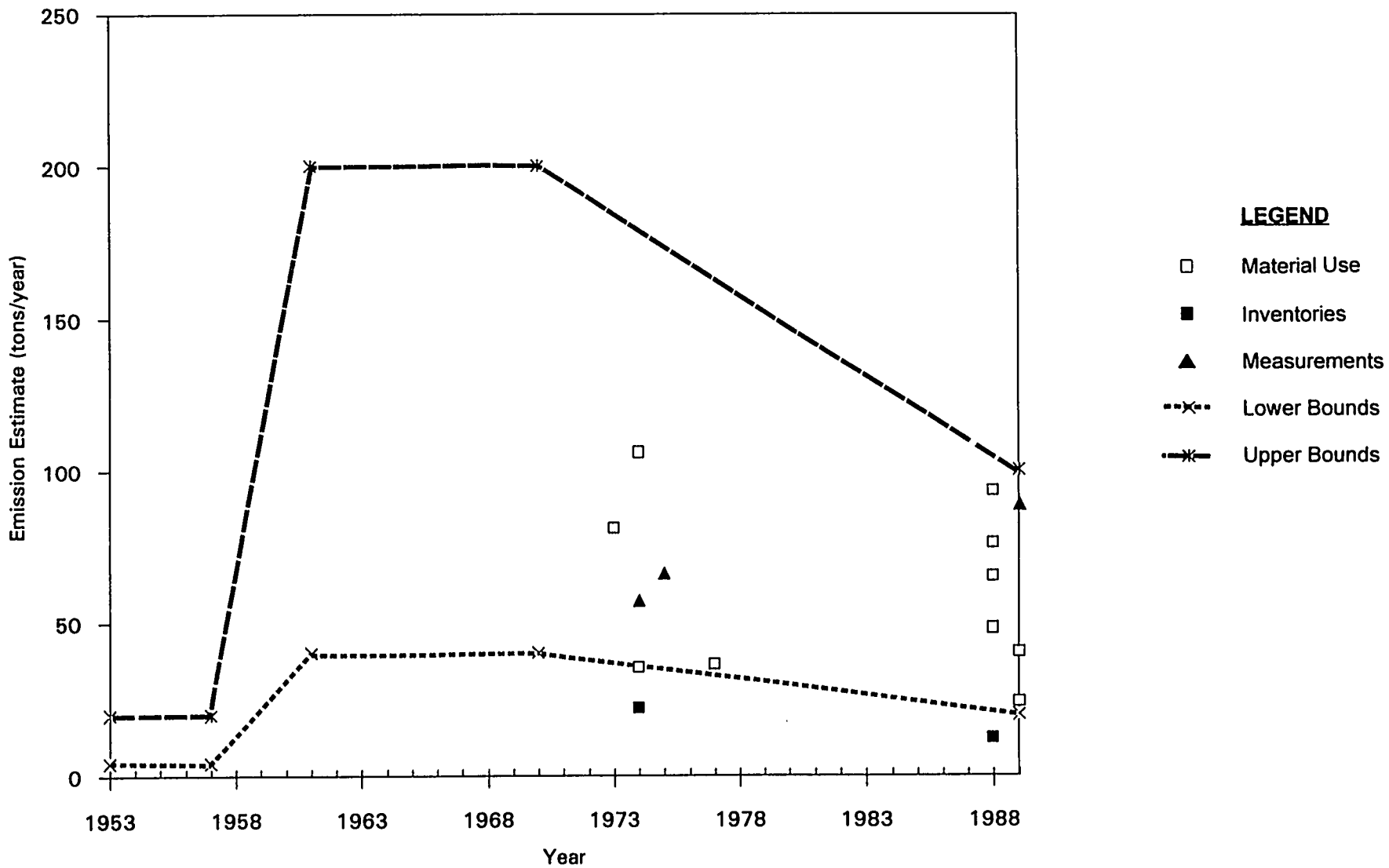
Starting around 1972, attempts were made to eliminate carbon tetrachloride from some production operations, due in part to measurements of carbon tetrachloride emissions from building stacks that were occasionally high enough to exceed permissible exposure levels at ground level (Musgrave, 1975). Carbon tetrachloride was used during and after plutonium machining to remove coolant oil from parts, in ultrasonic cleaners prior to inspection, and in dip tanks to degrease lathe turnings prior to briquetting (Musgrave, 1975). In 1974, 1,1,1-trichloroethane (TCA) was recommended to replace carbon tetrachloride in ultrasonic cleaning in Building 707 (Musgrave, 1975).

Various studies and projects involving estimation of carbon tetrachloride use at Rocky Flats or measurement of airborne concentrations of the solvent are available for the period from 1971 to 1989. Emission estimates based on these resources are shown as points on Figure 3-1. Results of past studies addressing carbon tetrachloride are summarized in Table 3-5 and are also described in Appendix E.

Hazardous material records indicate that there was a 44 percent decrease (from 22 to 12 tons per year) in carbon tetrachloride inventory from 1974 to 1989 (Barrick, 1974; Setlock, 1990). The Final Environmental Impact Statement for the Rocky Flats Plant site (USDOE, 1980) includes a tabulation of consumption rates of the "major chemicals" for fiscal year 1977. The reported annual usage of carbon tetrachloride is 5,334 gallons (36 tons). The same report included an estimated Rocky Flats plant airborne discharge rate of 4.73 grams per second (66 tons per year) in 1975. An estimate of carbon tetrachloride inventory in production areas for 1973 is 81 tons per year (Kruehauf and Richter, 1974). Release estimate based on material balance estimates for 1974-1975 ranged from 34 to 106 tons per year (Hobbs, 1982). Monitoring during the same period indicated average carbon tetrachloride emissions of 56 tons per year and maximum emission of 153 tons per year (Hobbs, 1982). Based on the available use, monitoring, and inventory data points, it is estimated that carbon tetrachloride emissions ranged between 40 and 200 tons per year in 1970 and decreased linearly to between 20 and 100 tons per year in 1989. These estimates are not inconsistent with other estimates of carbon tetrachloride use from 1988 to 1990, ranging from 40 to 93 tons per year (EG&G, circa 1990; EG&G, 1990-1991; Hamilton and Moser, 1990; USDOE, 1989).

3.2.2 Chloroform

As described in the Tasks 3 and 4 report, historical uses of chloroform at Rocky Flats are not well documented. Activities that have involved use of chloroform include laboratory analyses of gallium content of plutonium samples, as a "glue" used by carpenters to join Plexiglas, and for dissolving of plastics and photoresists. The estimated historical emission rates of chloroform have been based on reports of quantities of the material used or kept on hand, limited warehouse purchasing records, and statements made by retired and active workers during interviews. The period from 1952 to 1989 has been divided into two time spans based on the available information and factors discussed below.



ChemRisk
A Division of McLaren/Hart

FIGURE 3-1
CARBON TETRACHLORIDE EMISSION ESTIMATES
(TONS PER YEAR)

RKY294TS/3-1; REV:02-22-94

TABLE 3-5

CARBON TETRACHLORIDE EMISSION AND INVENTORY ESTIMATES

Information Source	Basis for Value(s)	Time Period of Estimate	Inventory (tons)	Resulting Carbon Tetrachloride Emission Rate (tons/yr)
Report on Handling and Monitoring of CCl_4 (Fruehauf & Richter, 1974)	Estimates of quantities used, with complete evaporation assumed.	1973		81
Stack Emission Monitoring, CCl_4 in B-776/777 Booster 1 (Johnson, 1973)	Gas chromatograph monitoring in Building 776/777 exhaust. Daily emissions are given in pounds and gallons.	June 4 through July 9, 1973		12
Potentially Harmful Materials Inventory (Barrick, 1974)	Estimates of inventories of hazardous materials kept on hand in 1974.	1974	22	
Report of CCl_4 Emissions from Production Areas (Hobbs, 1982)	Monitoring and material balance methods.	1974-1975		Monitoring: 56 Material Balance: 34 & 106
Final Environmental Impact Statement (USDOE, 1980)	"Normal average" release rate given for 1975 based on monitoring. Estimates of 1977 quantities used, with complete evaporation assumed.	1975 1977		66 36
USDOE Environmental Team Audit (USDOE, 1989)	Estimates of quantities used, with complete evaporation assumed.	1988		93
Air Stack Release Tabulation (EG&G, circa 1990)	Estimates of quantities used, with complete evaporation assumed.	1988 and 1989		1988: 65 1989: 24
Chemical Inventory Database (Setlock, 1990)	Estimates of inventories of hazardous materials kept on hand in 1988-1989.	1988-1989	12	
Report of Building 707 Halogenated Solvent Usage (Ferrera, 1988)	Estimates of quantities used, with complete evaporation assumed.	July 1988 to July 1989		Average: 48 Maximum: 76
Air Pollution Emission Notices (EG&G, 1990-1991)	Use estimates, with assumed complete evaporation. Some use of USEPA emission factors.	1989		40
Volatile Organic Emission Monitoring (Hamilton and Moser, 1990)	Duct sampling at six points.	1989		89

1953 to 1974

A 1974 Harmful and Potentially Harmful Materials Inventory (Barrick, 1974) indicated a chloroform inventory of 5,513 liters, or 8.9 tons. It is estimated that chloroform supplies were replenished at a rate such that approximately two-times the indicated inventory quantity (or about 18 tons per year) were used and released each year. Based upon the reported nature and extent of chloroform uses, it was unlikely that the replenishment rate for the solvent was significantly greater than this or that annual emissions were less than approximately one-quarter of the quantity kept on hand. Chloroform emissions of between 2 and 20 tons per year from 1953 to 1974 are estimated for screening purposes.

1975 to 1989

A number of data points relating to chloroform inventories, purchases, and usage rates at Rocky Flats after 1972 are shown in Figure 3-2 and summarized in Table 3-6. Additional details regarding emissions are presented in Appendix E. The Final Environmental Impact Statement for the Rocky Flats Plant Site (USDOE, 1980) includes a tabulation of consumption rates of the "major chemicals" for fiscal year 1977. No mention of chloroform use is included. Purchasing records from the Rocky Flats warehouse (EG&G, 1974-1988) indicate that chloroform orders totaled 360 gallons (2.2 tons) in 1985 and 300 gallons (1.8 tons) in 1986.

APEN calculations indicate a site-total chloroform emission rate of 0.84 ton per year based on 1986 usage data (EG&G, 1990-1991). A chemical inventory database containing estimates of quantities of hazardous materials kept on hand in 1988-1989 indicated that approximately 0.55 ton of chloroform was at the Rocky Flats facility (Setlock, 1990). It is estimated that chloroform emissions decreased linearly from a range of 2 to 20 tons per year in 1974 to a range of 0.5 and 5 tons per year in 1989. The 1989 value that defines the lower bound of plausible chloroform emissions for this period (.5 ton per year) is approximately 25 percent below the APEN chloroform emission estimate and slightly below the 1988/1989 inventory quantity for chloroform. In the later years of Rocky Flats operations, uses of chloroform were relatively minor. As a result, there was an increasing likelihood that minor operations contributing significant proportions to the site emission total went unidentified. While the APEN assumption of complete volatilization likely tended to cause emissions to be overstated, it is also possible that unidentified minor chloroform emission sources existed and that inventories reported in 1988/1989 underwent some replenishment during each year.

Review of available information indicates that it is unlikely that emissions of chloroform in the late-1980s exceeded the 1988/1989 inventory quantity by a factor of ten. As a result, the 1989 chloroform emission rate that defines the upper bound of plausible chloroform emissions was estimated to be 5 tons per year.

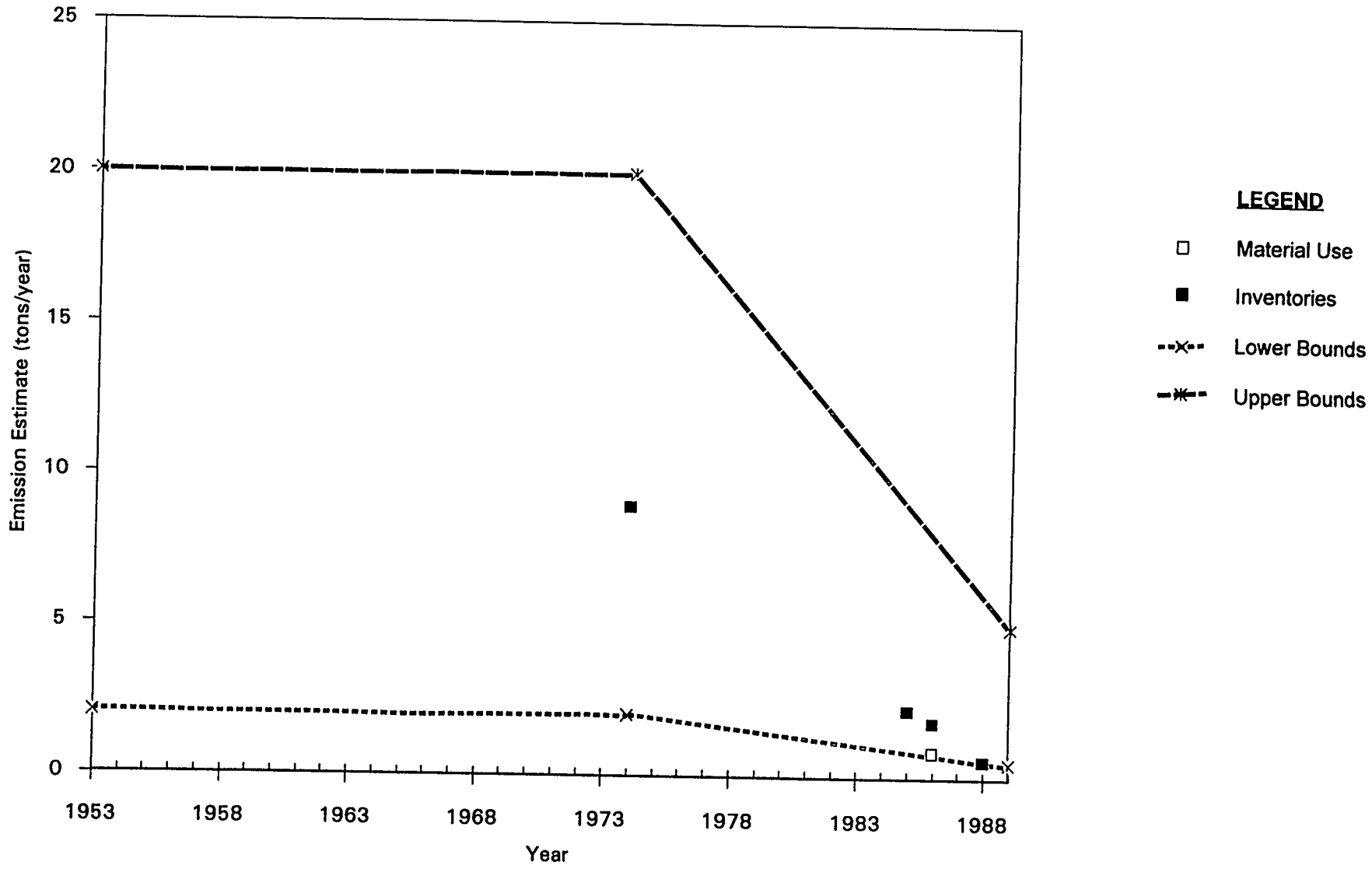


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FIGURE 3-2
CHLOROFORM EMISSION ESTIMATES
(TONS PER YEAR)

TABLE 3-6

CHLOROFORM EMISSION AND INVENTORY ESTIMATES

Information Source	Basis for Value(s)	Time Period of Estimate	Inventory (tons)	Resulting Chloroform Emission Rate (tons/yr)
Harmful and Potentially Harmful Materials Inventory (Barrick, 1974)	Estimates of inventories of hazardous materials kept on hand in 1974.	1974	8.9	
Rocky Flats Plant Warehouse Purchasing Records (EG&G, 1974-1988)	Records of dates and quantities of purchases of certain chemicals.	1985-1986	1985: 2.2 1986: 1.8	
Air Pollution Emission Notices, Buildings 559/561, 881, 374, and 460 (EG&G,1990-1991)	Estimates of quantities used, with assumed complete evaporation.	1986		0.84
Chemical Inventory Database (Setlock, 1990)	Estimates of inventories of hazardous materials kept on hand in 1988-1989.	1988-1989	0.55	

3.2.3 Methylene Chloride

As described in the Tasks 3 and 4 report, historical uses of methylene chloride at Rocky Flats are not well documented. Methylene chloride has been present in paints and paint strippers used at the plant, it is an ingredient in the Cee Bee® solution used in aqueous component cleaning in Building 460, and has been used in several laboratories and process areas for sample preparation and analysis. Historical emission rates of methylene chloride have been established based on estimates of quantities of the material used or kept on hand, limited warehouse purchasing records, and statements made during interviews of retired and active workers. The time period from 1952 to 1989 has been divided into two time spans based on the available information and factors discussed below.

1953 to 1974

A 1974 Harmful and Potentially Harmful Materials Inventory (Barrick, 1974) indicated a methylene chloride inventory of 1,502 liters, or 2.2 tons. A range of between 3 and 15 tons per year is estimated for screening purposes for the period from 1953 to 1974. Based upon the 1974 inventory quantity of 2.2 tons, purchasing record indications of replenishment during the year, and the indication in worker interviews that methylene chloride usage was significant before 1974, it is unlikely that emissions from 1953 to 1974 were less than 3 tons per year. The plausible upper bound of annual methylene chloride emissions is consistent with an ordering frequency of between five and seven times per year observed in warehouse purchasing records (EG&G, 1974-1988) and the 1974 inventory of 2.2 tons.

1975 to 1989

A number of data points relating to methylene chloride inventories, purchases, and usage rates at Rocky Flats after 1974 are shown in Figure 3-3 and summarized in Table 3-7. Additional details regarding emissions are presented in Appendix E. The Final Environmental Impact Statement for the Rocky Flats Plant Site (USDOE, 1980) includes a tabulation of consumption rates of the "major chemicals" for fiscal year 1977. No mention of methylene chloride use is included. Purchasing records from the Rocky Flats warehouse (EG&G, 1974-1988) indicate that methylene chloride orders totaled about 825 gallons (4.6 tons) in 1980 and about 600 gallons (3.4 tons) in 1984.

APEN calculations indicate a site-total methylene chloride emission rate of 3.3 tons per year (EG&G, 1990-1991). A chemical inventory database containing estimates of quantities of hazardous materials kept on hand in 1988-1989 indicated that approximately 0.31 ton of methylene chloride was at the Rocky Flats facility (Setlock, 1990). Another report of Rocky

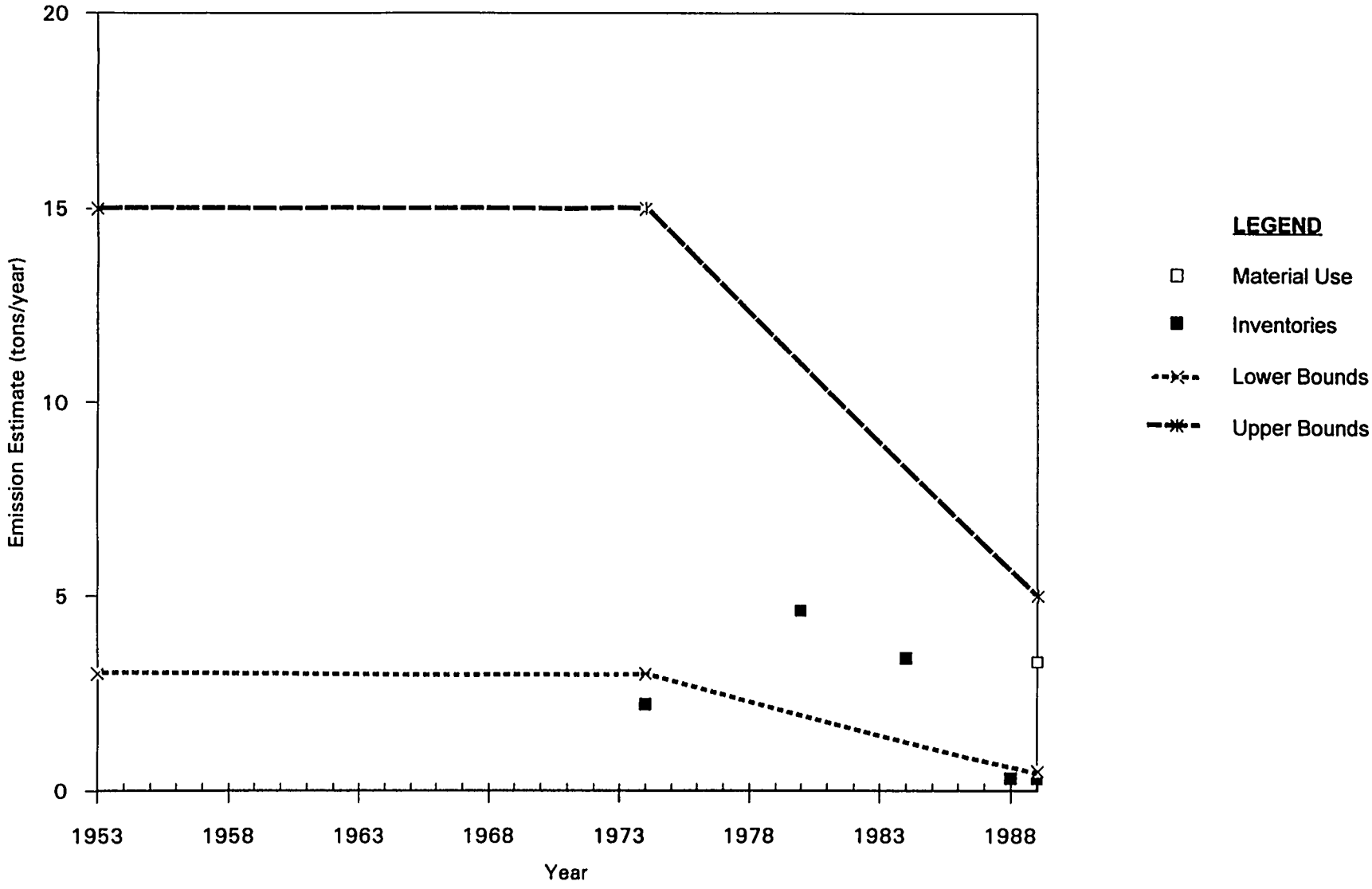

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FIGURE 3-3
METHELYNE CHLORIDE EMISSION ESTIMATES
(TONS PER YEAR)

RKY294T5/3-3; REV:02-22-94

TABLE 3-7

METHYLENE CHLORIDE EMISSION AND INVENTORY ESTIMATES

Information Source	Basis for Value(s)	Time Period of Estimate	Inventory (tons)	Resulting Methylene Chloride Emission Rate (tons/yr)
Harmful and Potentially Harmful Materials Inventory (Barrick, 1974)	Estimates of inventories of hazardous materials kept on hand in 1974.	1974	2.2	
Rocky Flats Plant Warehouse Purchasing Records (EG&G, 1974-1988)	Records of dates and quantities of purchases of certain chemicals.	1980, 1984	1980: 4.6 1984: 3.4	
Chemical Inventory Database (Setlock, 1990)	Estimates of inventories of hazardous materials kept on hand in 1988-1989.	1988-1989	0.3	
Report of Rocky Flats Usage of Methylene Chloride (Grocki, 1989a)	Estimates of inventories in 1989; present in Buildings 123, 440, 559, 771, 881, T452B.	1989	0.3	
Air Pollution Emission Notices, Buildings 776/777, 771, 881, 451, 460, 374, and 228A/B (EG&G, 1990-1991)	Estimates of quantities used, with assumed complete evaporation.	around 1989		3.3

Flats methylene chloride usage in 1989 listed a total of 616 pounds (0.3 ton) of methylene chloride (Grocki, 1989a).

It is assumed that methylene chloride emissions decreased linearly, ranging from between 3 to 15 tons per year in 1974 to between 0.5 and 5 tons per year in 1989. Based upon the 1989 inventory of 0.31 tons of methylene chloride, documented replenishment rates during the 1980s, and the APEN emission estimate of 3.3 tons per year, it is unlikely that annual emissions of methylene chloride were less than 0.5 ton per year.

The 1989 point (5 tons per year) that defines the upper bound of methylene chloride emissions is based upon the APEN emission estimate of 3.3 tons per year and the possibility that; due to the continued widespread U.S. use of methylene chloride in paint strippers, pesticides, and certain aerosol products (ATSDR, 1991); unidentified minor methylene chloride emission sources existed. It is unlikely that actual emissions significantly exceeded the APEN estimate, however, due to the conservative assumption in the APEN assessment that all methylene chloride that was used was lost to the atmosphere.

3.2.4 Tetrachloroethylene

As described in the report of Tasks 3 and 4, tetrachloroethylene (PCE) was widely used for uranium and plutonium part cleaning and degreasing in Buildings 881, 444, 883, 771, and 776 (ChemRisk, 1991-1992, Interview Nos. 39,48,53,78) for a large portion of Rocky Flats history. The estimated range of historical emissions of PCE is based on estimates of quantities of material used or kept on hand and statements made by retired and active workers during interviews. The period of interest from 1953 to 1989 can be broken into the following time spans based on the available information summarized below.

1953 to 1961

Uranium machining during the early years of Rocky Flats operation was typically performed with a 15:1 Shell Vitrea 923-8D oil and tetrachloroethylene mixture (Navratil and Miner, 1984). Because PCE has a relatively low volatility, it was not widely used on plutonium. PCE was substituted for carbon tetrachloride for about four months in 1966, but residue built up on inspection devices and on the plutonium, and its use was abandoned (Musgrave and Hornbacher, 1973). According to a former Building 881 worker, about 25 drums (assumed to contain 55 gallons each) per month of PCE were used in enriched uranium operations, with about 10 percent recovered (ChemRisk, 1991-1992, Interview No. 39). PCE was distilled from oil in Building 881 (Navratil and Miner, 1984).

Based upon the reported quantity of PCE used in Building 881 and the reported but unquantified used of the solvent in four other Rocky Flats buildings, an upper bound of plausible PCE emission was set at 300 tons per year. It is unlikely that site-total emissions exceeded three times the Building 881 estimate because PCE was used mainly in fabrication of uranium weapon components, and the assumption of complete evaporation of the PCE that was used is quite conservative given the relatively low volatility of the solvent.

Even with the relatively low volatility of PCE, the reported quantity of the solvent used in Building 881 alone during the period (100 tons per year) indicates that it is unlikely that annual emissions were less than 50 tons per year. A lower bound of plausible PCE emission was set at 50 tons per year for the period from 1953 through 1961.

1962 to 1966

Around 1962, enriched uranium operations (the oralloy line) were lost to the Oak Ridge Y-12 Plant as the Department of Defense moved to eliminate functional redundancy among the nation's weapon production facilities. Over the period from 1962 to 1966, use of PCE decreased due to phase-out of enriched uranium recovery and machining activities. PCE emissions are estimated to have decreased to about one-third of their peak values over the four-year period between when oralloy operations were phased out of Building 881 and when they were replaced by J-line stainless steel operations in 1966. PCE emissions are estimated to have decreased linearly, ranging from between 50 and 300 tons per year in 1961 to between 20 and 100 tons per year in 1966.

1967 to 1977

A few data points relating to PCE inventories and usage rates at Rocky Flats after 1972 are shown in Figure 3-4 and in Table 3-8. Additional details regarding emissions are presented in Appendix E. Beginning around 1973, solvent substitution efforts saw PCE being replaced by TCA for plutonium component degreasing. A 1974 Harmful and Potentially Harmful Materials Inventory (Barrick, 1974) indicated a PCE inventory of only 8 tons. The Final Environmental Impact Statement for the Rocky Flats Plant site (USDOE, 1980) includes a tabulation of consumption rates of the "major chemicals" for fiscal year 1977. No mention of PCE use is included. Based on these few data points, it is assumed for screening purposes that PCE emissions decreased linearly from a range of 20 to 100 tons per year in 1966 to between 0.000010 and 1.0 ton per year in 1977.

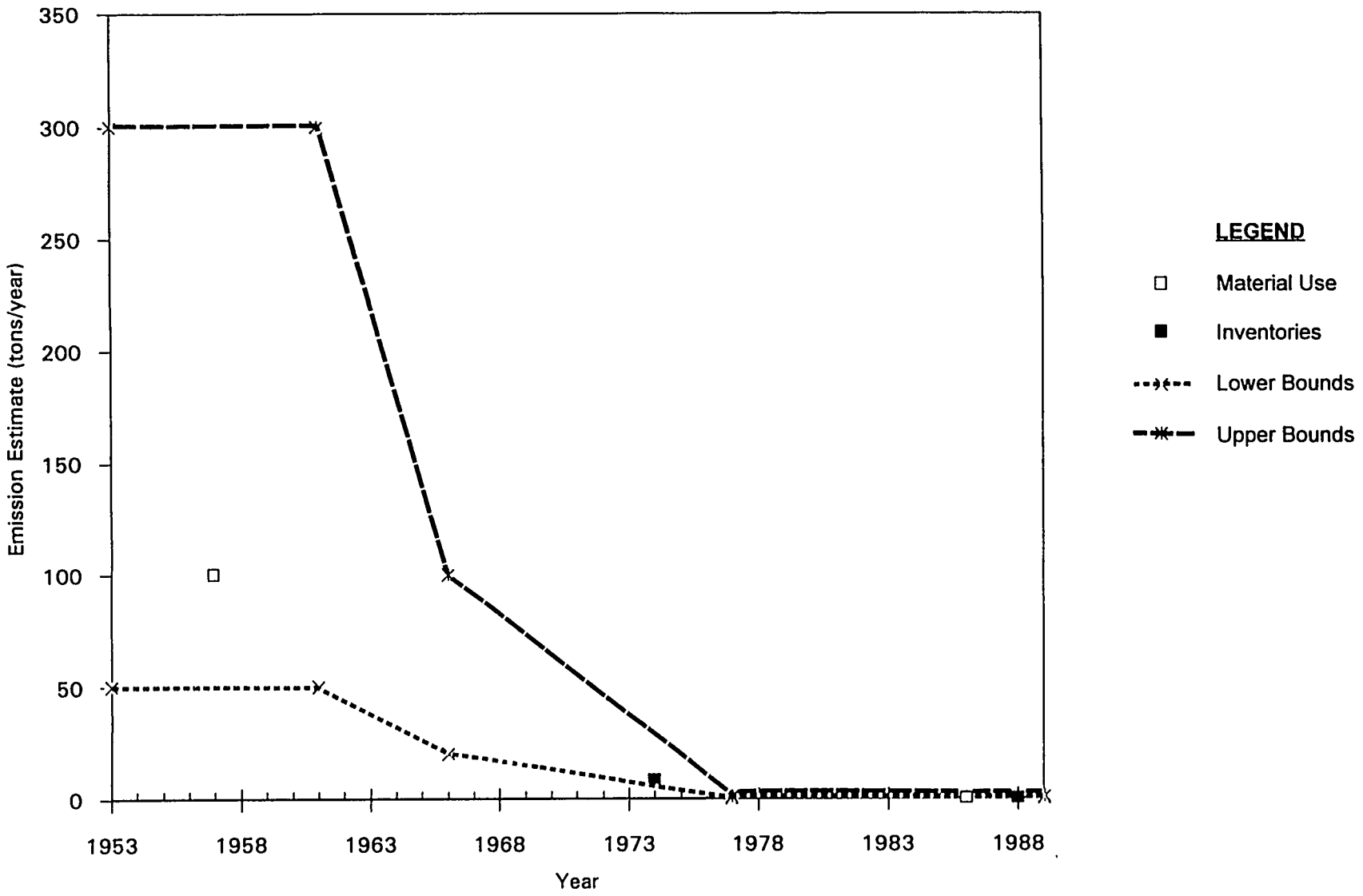

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FIGURE 3-4
TETRACHLOROETHYLENE EMISSION ESTIMATES
(TONS PER YEAR)

RKY294T5/3-4; REV:02-22-94

TABLE 3-8

TETRACHLOROETHYLENE EMISSION AND INVENTORY ESTIMATES

Information Source	Basis for Value(s)	Time Period of Estimate	Inventory (tons)	Resulting PCE Emission Rate (tons/yr)
Worker Interview (ChemRisk, 1991-1992; Interview No. 39)	Personal recollection of PCE usage rates in oralloy processing.	1952-1962		100 (25 drums/month, with 10% recycled)
Harmful and Potentially Harmful Materials Inventory (Barrick, 1974)	Estimates of inventories of hazardous materials kept on hand in 1974.	1974	8.0	
Air Pollution Emission Notices (EG&G, 1990-1991)	Estimates of quantities used, with complete evaporation assumed.	1986-1987		0.00007
Chemical Inventory Database (Setlock, 1990)	Estimates of inventories of hazardous materials kept on hand in 1988-1989.	1988-1989	0.003	
Report of Rocky Flats Usage of Tetrachloroethylene (Grocki, 1989a)	Estimates of 1989 inventories of PCE in Buildings 881, 559, 452B.	1989	0.00004	

1978 to 1989

APEN calculations indicate a site-total PCE emission rate of 0.00007 ton per year for 1986-1987 (EG&G, 1990-1991). A chemical inventory database containing estimates of inventories of hazardous materials kept on hand in 1988-1989 indicated that approximately 0.003 ton of PCE was at the Rocky Flats facility (Setlock, 1990). Another report of Rocky Flats PCE usage in 1989 listed a total of .0882 pound (.00004 ton) of PCE (Grocki, 1989a). Based on this information, PCE emissions are estimated for screening purposes to have ranged from between 0.000010 and 1.0 ton per year during 1978 to 1989.

3.2.5 1,1,1-Trichloroethane

As described in the Tasks 3 and 4 report, 1,1,1-trichloroethane (TCA) was used in cleaning and degreasing of metal parts for a large portion of Rocky Flats history. It is probable that use of the solvent began with the increased use of oils and solvents around 1957, when increased demand for plutonium components brought about the end of "dry" machining of plutonium. Various sources of information concerning TCA usage at Rocky Flats and emissions from 1974 to 1989 are summarized in Table 3-9 and depicted in Figure 3-5. Additional details regarding emissions are presented in Appendix E. The time period of interest from 1953 to 1989 can be broken into the following time spans.

1953 to 1957

There are no indications of TCA use prior to 1957. For screening purposes it is assumed that airborne emissions ranged between 0 and 5 tons per year from 1953 to 1957.

1958 to 1973

Although the earliest documented use of TCA was in 1963, it is probable that use of the solvent began with the increased use of oils and solvents around 1957. Available information suggests that TCA was used to clean parts in plutonium production areas of Buildings 771 and 776 from approximately 1958 to 1973. TCA was initially avoided in plutonium machining applications due to explosion hazard; in at least one instance, an explosion occurred when TCA was inadvertently substituted for TCE in a plutonium machining operation (Hobbs, 1970).

The airborne emissions for this period are assumed to be relatively consistent with the 1974 inventory quantity of 34 tons (Barrick, 1974). It is estimated that 1958 to 1963 was a period of transition to increased use of TCA, and that TCA emissions for the period from 1963 to 1973 ranged from 20 to 60 tons per year.

TABLE 3-9

1,1,1-TRICHLOROETHANE EMISSION AND INVENTORY ESTIMATES

Information Source	Basis for Value(s)	Time Period of Estimate	Inventory (tons)	Resulting 1,1,1-TCA Emission Rate (tons/yr)
Harmful and Potentially Harmful Materials Inventory (Barrick, 1974)	Estimates of inventories of hazardous materials kept on hand in 1974.	1974	34	
Final Environmental Impact Statement (USDOE, 1980)	Estimate of 1977 consumption.	1977		26
Rocky Flats Plant Warehouse Purchasing Records (EG&G, 1974-1988)	Records of dates and quantities of purchases of certain chemicals.	1980-1984		70-80
Monthly Status Report of Halogenated Solvent Use (Ferrera, 1988)	Estimates of quantities used, with complete evaporation assumed.	1987-1988		44
B-707 Chlorinated Solvent Usage Report (Weis, 1988b)	Estimates of quantities used, with assumed complete evaporation.	1988		20
USDOE Environmental Team Audit (USDOE, 1989)	Estimates of quantities used, with complete evaporation assumed.	1988		24
Chemical Inventory Database (Setlock, 1990)	Estimates of inventories of hazardous materials in 1988-1989.	1988-1989	2.6	
Air Stack Release Tabulation (EG&G, circa 1990)	Estimates of quantities used, with complete evaporation assumed.	1988-1989		1988: 24 1989: 23
Halogenated Solvent Usage Update (Church, 1989)	Estimates of quantities used, with complete evaporation assumed.	July 1988 to July 1989		17
Volatile Organic Emission Monitoring (Hamilton and Moser, 1990)	Duct sampling at six points. Highest concentrations used to estimate emission rates.	1989		46
Air Pollution Emission Notices (EG&G, 1990-1991)	Estimates of quantities used, with complete evaporation assumed.	1989		20.8

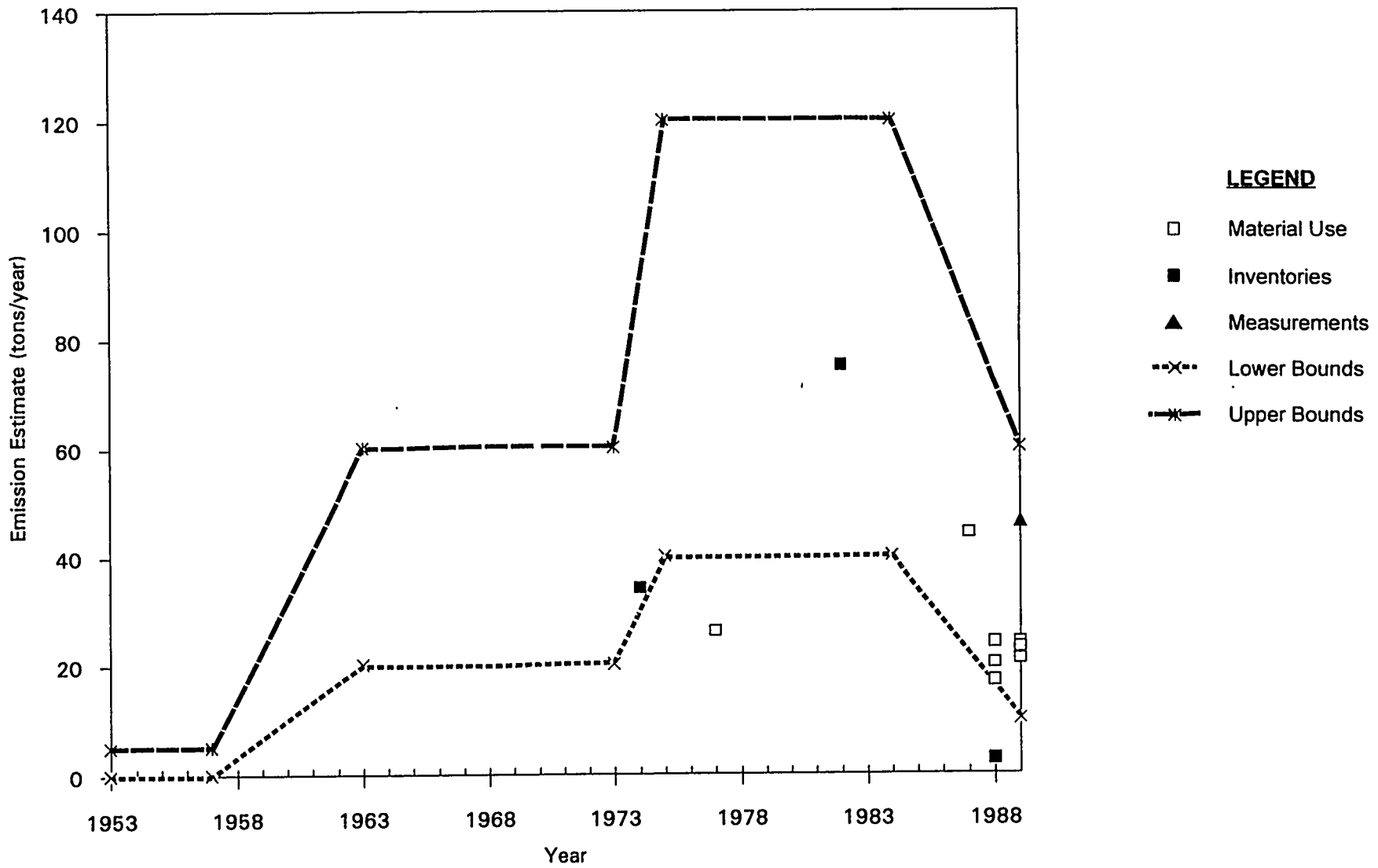

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FIGURE 3-5
1,1,1-TRICHLOROETHANE EMISSION ESTIMATES
(TONS PER YEAR)

RKY294T5/3-5; REV:02-22-94

1974 to 1984

TCA began replacing trichloroethylene (TCE) for vapor degreasing of parts in plutonium areas beginning around 1973. By the end of 1974, TCE remained in use in only one plutonium operation, and was used only in research and analytical activities by February 1975. The Final Environmental Impact Statement (USDOE, 1980) reported annual usage of TCA in 1977 of 4,675 gallons (26 tons).

As a result of the substitution of TCA for TCE, emissions of TCA are estimated to have reached a range of between 40 and 120 tons per year during the late 1970s based on a limited number of warehouse purchasing records from the early 1980s, suggesting purchases of 70 to 80 tons per year (EG&G, 1974-1988).

1985 to 1989

As mentioned above, purchasing records from the Rocky Flats warehouse (EG&G, 1974-1988) indicate that TCA orders totaled between 70 and 80 tons per year in the early 1980s. In the middle to late 1980s, another round of solvent substitution efforts began that included use of isopropyl alcohol and De-Solv-it® in place of TCA in a number of non-plutonium cleaning operations. In 1988, cleaning practices in Building 447 involved submerging parts in a TCA tank to remove oils, grease, or wax, followed by submersion in Oakite® detergent solution and then an aqueous rinse tank. At that time, use of De-Solv-it® instead of TCA was recommended in order to eliminate use of chlorinated solvents (Weis, 1988a). Water and detergent solutions could not be used on plutonium, however, because water rapidly corrodes plutonium (Musgrave and Hornbacher, 1973). Orders of TCA dropped to 33 tons in 1987, and the 1988-1989 inventory of 2.6 tons (Setlock, 1990) represents a 92% decrease from the 1974 value. Based on these factors and the identified data points from studies addressing TCA uses and emissions from 1987 to 1989, it is estimated that TCA emissions decreased linearly from between 40 and 120 tons per year in 1984 to between 10 and 60 tons per year in 1989. These estimates are not inconsistent with other estimates of TCA use in 1988 and 1989 ranging from 17 to 46 tons per year (EG&G, circa 1990; EG&G, 1990-1991; Ferrera, 1988; Church, 1989; Hamilton and Moser, 1990; USDOE, 1989; Weis, 1988b).

3.2.6 Trichloroethylene

As described in the Tasks 3 and 4 report, trichloroethylene (TCE) was used in large quantities at Rocky Flats to clean and degrease beryllium, plutonium, and uranium parts. Historical emissions of TCE have been estimated based on quantities of the solvent used or kept on hand, warehouse purchasing records, some limited effluent monitoring data, and statements from active

and retired workers. The time period of interest from 1953 to 1989 can be broken into the following time spans based on the available information and factors discussed below.

1953 to 1962

According to a former Building 881 worker, about 25 drums (assumed to contain 55 gallons each) per month of TCE were used in enriched uranium operations, with about 10 percent recovered. TCE was used in the Soxhlet extraction process for enriched uranium recovery (ChemRisk, 1991-1992, Interview No. 39) and was widely used in vapor degreasers for part cleaning. TCE was distilled from oil in Building 881 (Navratil and Miner, 1984). Based on this reported level of use of the solvent, the estimated TCE emissions for this period range between 50 and 200 tons per year.

1963 to 1973

During 1962, enriched uranium operations (the oralloy line) were moved to the Oak Ridge Y-12 Plant as the Department of Defense moved to eliminate functional redundancy among the nation's weapon production facilities. Over the period from 1962 to 1963, use of TCE decreased due to the phase-out of enriched uranium recovery and machining activities. The decrease in TCE usage due to loss of oralloy operations was somewhat offset by efforts to substitute TCE for acetone, isopropyl alcohol, and other solvents for cleaning plutonium, beryllium, and uranium parts. An internal Rocky Flats Plant memo indicates that 10,000 gallons (or about 62 tons) of TCE were used plant-wide during 1973 (Musgrave and Hornbacher, 1973). A report on the annual usage of TCE indicated site-total consumption of 60 tons of TCE during 1973 (Dow Chemical, 1972-1974). Based on these reported levels of use of the solvent, and the reported reduction in use during 1962 and 1963, TCE emissions for 1973 are estimated to have ranged between 30 and 90 tons per year.

1974 to 1975

Various data points relating to TCE inventories, purchases, and usage rates at Rocky Flats after 1972 are shown in Figure 3-6 and summarized in Table 3-10. Additional details concerning emissions are presented in Appendix C. In late 1973, the Rocky Flats Area Office of the Atomic Energy Commission (USAEC) made commitments to the Environmental Protection Agency (USEPA) regarding elimination of TCE use in certain Rocky Flats buildings that were estimated to be exceeding TCE emission limits (Thompson, 1973). By the end of 1974, TCE remained in use in only one plutonium operation and was used only in research and analytical activities by February 1975 (Bean, 1975). A 1974 Harmful and Potentially Harmful Materials Inventory (Barrick, 1974) indicated a TCE inventory of 25 tons, while purchasing records from the Rocky Flats warehouse (EG&G, 1974-1988) indicate that TCE orders totaled only 15 drums

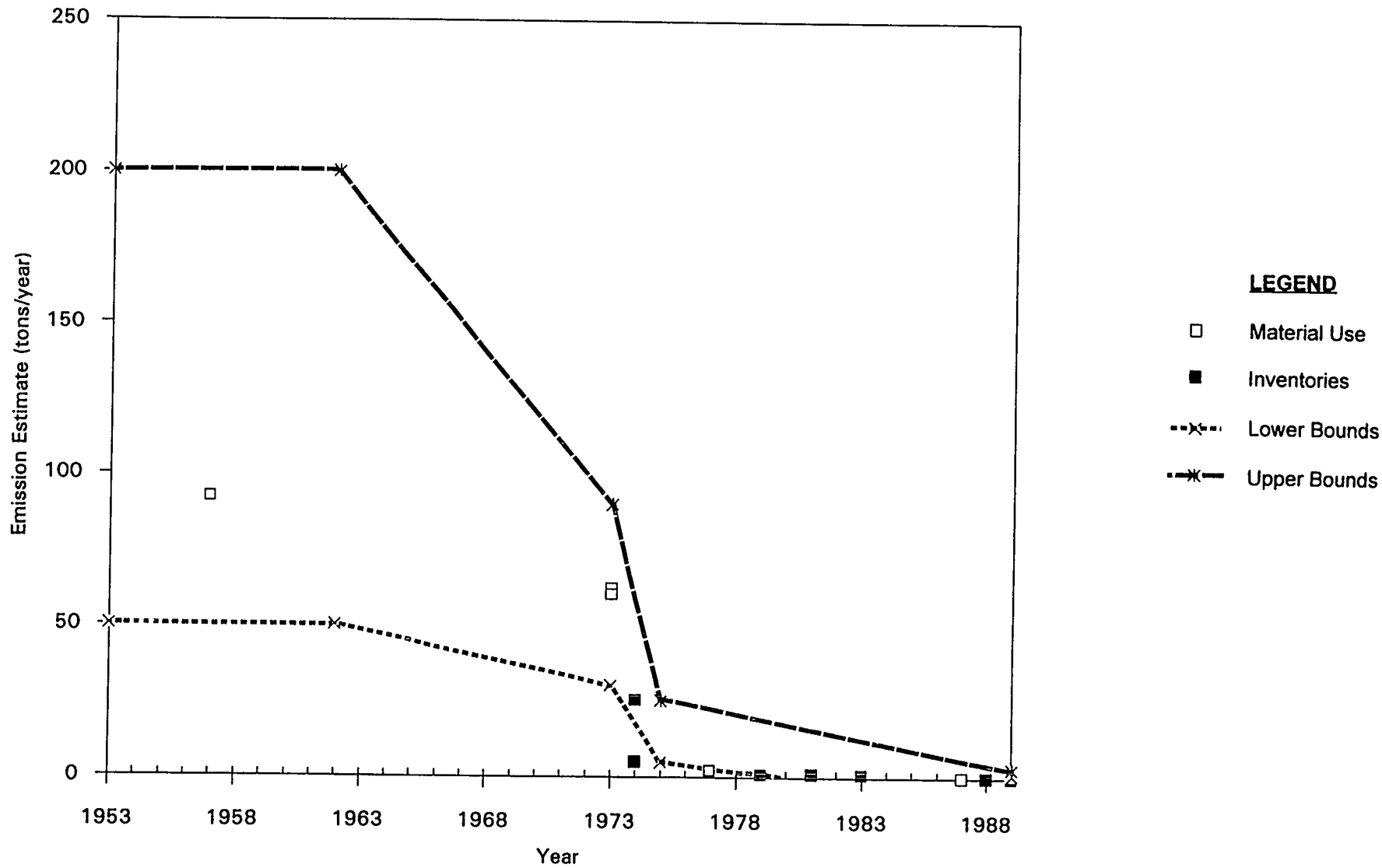

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FIGURE 3-6
TRICHLOROETHYLENE EMISSION ESTIMATES
(TONS PER YEAR)

TABLE 3-10

TRICHLOROETHYLENE EMISSION AND INVENTORY ESTIMATES

Information Source	Basis for Value(s)	Time Period of Estimate	Inventory (tons)	Resulting TCE Emission Rate (tons/yr)
Worker Interview (ChemRisk, 1991-1992; Interview No. 39)	Personal recollection of TCE usage rates in oralloy processing.	1952-1962		92 (25 drums/month, with 10% recycled)
Report on Use of TCE for Degreasing of Be, Pu, and U (Musgrave and Hornbacher, 1973)	Quantity estimate of TCE used for ultrasonic vapor degreasing of metal parts.	1973		62
Dow Chemical Trichloroethylene Folder (Dow Chemical, 1972-1974)	Estimates of quantities used, including by building and user.	1973		60
Stack Emission Monitoring, TCE in B-776/777 Booster 1 (Johnson, 1973) CH-850	Gas chromatograph monitoring in Building 776/777 exhaust. Daily emissions in pounds and gallons.	June 4 through July 9, 1973		5.0 average
Harmful and Potentially Harmful Materials Inventory (Barrick, 1974)	Estimates of inventories of hazardous materials.	1974	25	
Rocky Flats Plant Warehouse Purchasing Records (EG&G, 1974-1988)	Records of dates and quantities of purchases of certain chemicals.	1974, 1979, 1981, 1983		1974: 5 1979: 0.67 1981: 1.0 1983: 0.67
Final Environmental Impact Statement (USDOE, 1980)	Estimates of 1977 quantities used.	1977		2
Chemical Inventory Database (Setlock, 1990)	Estimates of inventories of hazardous materials.	1988-1989	0.15	
Report of Rocky Flats Usage of Trichloroethylene (Grocki, 1989b)	Estimates of inventories of TCE in Buildings 559 and 881.	1989	0.000014	
Air Pollution Emission Notices, Building 374 (EG&G, 1991d)	Estimates of quantities used, with complete evaporation assumed.	1987		1.5×10^{-3}

(about 5 tons) in 1974. It is estimated that TCE emissions decreased linearly from between 30 to 90 tons per year in 1973 to between 5 and 25 tons per year in 1975.

1976 to 1989

TCE was used only for research and analytical activities by February 1975 (Bean, 1975). An Air Pollution Emission Notice for Building 460 erroneously reported the release of TCE in the amount of 0.15 ton per year for 1986-1989, when in fact TCA had actually been used (Costain, 1992). APEN emission estimates for Building 374, the only other APEN reporting TCE release, were extremely minor at 1.5×10^{-3} ton per year (EG&G, 1991d). The Rocky Flats Plant Final Environmental Impact Statement (USDOE, 1980) indicated that 330 gallons (2 tons) per year of TCE were consumed at the plant in 1977. A chemical inventory database containing estimates of inventories of hazardous materials kept on hand in 1988-1989 indicated that approximately 0.15 ton of TCE was at the Rocky Flats facility (Setlock, 1990), while purchasing records from the Rocky Flats warehouse (EG&G, 1974-1988) indicate that TCE orders totaled 2 or 3 drums every two years from 1979 to 1983. Another report of Rocky Flats TCE usage in 1989 listed a total of 13 milliliters of TCE present, all of which was located in laboratory areas (Grocki, 1989b). Based on this documentation, it is estimated for screening purposes that TCE emissions decreased linearly from between 5 and 25 tons per year in 1975 to between 0.0010 and 1.0 ton per year in 1989.

3.2.7 Emission Estimates of Organic Solvents

Unlike airborne releases of radioactive materials and beryllium, there is little or no routine effluent sampling data for organic solvents. In this section, the ranges of plausible emissions of organic solvents are presented based on historical information on material purchases, uses, disposal, and recycling. Upper and lower bounds of emissions for carbon tetrachloride, chloroform, methylene chloride, tetrachloroethylene, 1,1,1-trichloroethane and trichloroethylene from 1953 through 1989 are listed in Table 3-11 through 3-16. The emission bounds have been established with the intent of ensuring that the ranges encompass the actual value for any one year.

TABLE 3-11

UPPER AND LOWER BOUNDS OF CARBON TETRACHLORIDE EMISSIONS (1953-1989)

Year	Lower Bound (tons/year)	Upper Bound (tons/year)
1953-1957	4.0	20
1958	13	65
1959	22	110
1960	31	160
1961-1970	40	200
1971	39	200
1972	38	190
1973	37	180
1974	36	180
1975	35	170
1976	34	170
1977	33	160
1978	32	160
1979	31	150
1980	29	150
1981	28	140
1982	27	140
1983	26	130
1984	25	130
1985	24	120
1986	23	120
1987	22	110
1988	21	100
1989	20	100

TABLE 3-12

UPPER AND LOWER BOUNDS OF CHLOROFORM EMISSIONS (1953-1989)

Year	Lower Bound (tons/year)	Upper Bound (tons/year)
1953-1974	2.0	20
1975	1.9	19
1976	1.8	18
1977	1.7	17
1978	1.6	16
1979	1.5	15
1980	1.4	14
1981	1.3	13
1982	1.2	12
1983	1.1	11
1984	1.0	10
1985	0.90	9.0
1986	0.80	8.0
1987	0.70	7.0
1988	0.60	6.0
1989	0.50	5.0

TABLE 3-13

UPPER AND LOWER BOUNDS OF METHYLENE CHLORIDE EMISSIONS (1953-1989)

Year	Lower Bound (tons/year)	Upper Bound (tons/year)
1953-1974	3.0	15
1975	2.8	14
1976	2.7	14
1977	2.5	13
1978	2.3	12
1979	2.2	12
1980	2.0	11
1981	1.8	10
1982	1.7	9.7
1983	1.5	9.0
1984	1.3	8.3
1985	1.2	7.7
1986	1.0	7.0
1987	0.83	6.3
1988	0.67	5.7
1989	0.50	5.0

TABLE 3-14

UPPER AND LOWER BOUNDS OF TETRACHLOROETHYLENE EMISSIONS (1953-1989)

Year	Lower Bound (tons/year)	Upper Bound (tons/year)
1953-1961	50	300
1962	44	260
1963	38	220
1964	32	180
1965	26	140
1966	20	100
1967	18	91
1968	16	82
1969	15	73
1970	13	64
1971	11	55
1972	9.1	46
1973	7.3	37
1974	5.5	28
1975	3.6	19
1976	1.8	10
1977-1989	0.000010	1.0

TABLE 3-15

UPPER AND LOWER BOUNDS OF 1,1,1-TRICHLOROETHANE EMISSIONS (1953-1989)

Year	Lower Bound (tons/year)	Upper Bound (tons/year)
1953-1957	0	5.0
1958	3.3	14
1959	6.7	23
1960	10	32
1961	13	42
1962	17	51
1963-1973	20	60
1974	30	90
1975-1984	40	120
1985	34	110
1986	28	96
1987	22	84
1988	16	72
1989	10	60

TABLE 3-16

UPPER AND LOWER BOUNDS OF TRICHLOROETHYLENE EMISSIONS (1953-1989)

Year	Lower Bound (tons/year)	Upper Bound (tons/year)
1953-1962	50	200
1963	48	190
1964	46	180
1965	45	170
1966	43	160
1967	41	150
1968	39	140
1969	37	130
1970	35	120
1971	34	110
1972	32	100
1973	30	90
1974	18	58
1975	5.0	25
1976	4.6	23
1977	4.3	22
1978	3.9	20
1979	3.6	18
1980	3.2	16
1981	2.9	15
1982	2.5	13
1983	2.1	11
1984	1.8	9.6
1985	1.4	7.9
1986	1.1	6.1
1987	0.72	4.4
1988	0.36	2.7
1989	0.0010	1.0

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4.0 UNCERTAINTY IN AIRBORNE EMISSION ESTIMATES

Uncertainties in emission estimates can arise from a number of sources, and many of these sources have been described and discussed in the preceding sections and are further elaborated on here. Two general approaches to quantifying the uncertainty in emission estimates have been adopted in this report. The first approach, which is employed when emission estimates are developed primarily from effluent monitoring data, involves the examination and quantification of the potential error introduced as a result of the various elements of the sampling and analytic programs.

The second approach to quantifying uncertainty is used when there is little or no routine effluent sampling data and estimates must be developed based on a variety of documents and information obtained from personnel interviews. In this case, the range of potential emissions for specified periods of the plant's operating history are bounded for a particular material of concern. Upper and lower bounds of estimated solvent emissions were presented in Section 3. The bounds have been established with the intent of ensuring that the range encompasses the actual value for any one year.

The purpose of this section is to quantify on the uncertainties in the emission estimates of radioactive and nonradioactive materials that were based on effluent monitoring data.

4.1 Random and Systematic Errors

Errors associated with a measurement can be either random or systematic. When a measuring instrument is used repeatedly to measure some attribute, the readings or results will typically be distributed around a given value. The fluctuation of readings around a given value is dependent on the sample matrix, the instrument, and the experimental condition. This type of measurement error, also called random error, can be reduced but can never be completely eliminated. Random error may be reduced either by improving the precision of the instrument or by tightening the control of the sampling conditions, or both.

The influence of random errors in estimating some value can also be reduced by taking many measurements and averaging them. When many measurements are taken, readings above the true value are likely to be balanced by readings below the true value. As a result, averaging many measurements will reduce the uncertainty in a value due to random error. It has been shown that the random error of an average is inversely proportional to $\sqrt{n}$, where n is the number of measurements.

On the other hand, systematic error may occur as a result of inaccurate or poorly designed sampling or analytic instruments. In this case, an instrument may give readings consistently higher or lower than the true value. Unlike random error, systematic error cannot be reduced by taking many measurements and calculating the average. However, systematic error in a sampling or analytical method can be detected and quantified by either measuring a standard, or comparing results with another method of known accuracy. Once the systematic error of an instrument or method is quantified, its effect on subsequent measurement results can be compensated for by using a correction factor. It is the purpose of this section to identify and quantify potential sources of systematic errors in developing release estimates of the contaminants of concern.

4.2 Uncertainty in Effluent Monitoring Program Results

As has been described, the sampling of contaminants in airborne effluents at the plant relies on the performance of the following basic calculation:

$$C = \frac{A}{V}$$

Where:

C	=	Concentration of a contaminant in effluent (quantity per cubic meter of air)
A	=	Quantity of contaminant in a sample (e.g., disintegrations per minute or micrograms)
V	=	Volume of sample (cubic meters of air)

Annual emissions from the facility (E) are then calculated by multiplying the concentration of a contaminant in an effluent (C) times the total amount of air released annually from the particular emission point and adding the resulting values up for the entire facility. The total amount of air released is based on the flow rate (Q) and the length of operation (T) of the exhaust system:

$$\mathbf{E} = \mathbf{C} \times \mathbf{Q} \times \mathbf{T}$$

In order to perform these calculations, the plant performed the following activities, each having attendant uncertainties:

1) A fraction of the effluent stream was extracted,

2) Contaminants were collected from the extracted sample on filter paper,

3) Contamination was measured on a sample filter, and

4) The total volume of effluent air associated with the sampled stream was estimated or measured.

One of the objectives of the investigation and review of the sampling and analytic systems used by the plant and described in this report was to identify practices that would have led to systematic errors in the effluent data that were produced. The areas that were examined for systematic error included potential bias arising from the following plant activities:

1) Extraction of a fraction of the effluent stream.

   - Non-isokinetic sampling.
   - Nonrepresentative sampling.

2) Contaminant collection from the extracted sample on filter paper.

   - Sample line loss.
   - Sampling apparatus filter efficiency.

3) Contamination measurement on a sample filter

   - Self-absorption.
   - Insufficient sensitivity of methods.

As described in Section 2.2, most of these has been discussed in some detail and ruled out as significant sources of systematic error in the airborne contaminant monitoring system, in many cases, because of correction factors used by the plant to compensate for these errors.

Two areas that were identified as the largest potential sources of error or uncertainty for particulate sampling and that are also important in tritium sampling involve the first and last steps of the process. As part of the first step involving the extraction of a fraction of the effluent stream, the determination of the size of the fraction (V, or volume of sample) that is

extracted is dependent on knowledge of the flow rate of the sampling device. However, documentation suggests that in mid-1974 the target sampling flow rate (2 ft^3 min^{-1} or 56.6 L min^{-1}) was routinely used as opposed to the actual measured rate (Rockwell, 1976b) for particulate sampling and 50 cubic centimeters per minute for tritium sampling. To the extent that flow rates were regularly readjusted to the target rate, fluctuations around the target resulted in random errors that were unimportant given the thousands of measurements made. However, the limited review of some actual particulate sampling flow rates during the period after 1974 suggests that actual flow rates may frequently have varied significantly from the target rate, more frequently low than high, which would lead to the underestimation of emissions. Based on the result of a special study, a frequency distribution of the measured sampling flow rates divided by 56.6 L min^{-1} is presented in Figure 2-2. This figure represents a distribution of values that can be used to compensate for the bias in sampling flow rate. For the purpose of this analysis, the correction factor of sampling flow rate was assumed to be normally distributed with a calculated mean of 0.92 and a standard deviation of 0.11.

As described in Section 2.2.1.2, sampling flow rates have historically been set at approximately 50 cm^3 min^{-1} for tritium samplers. However, the actual average sampling flow rate is likely to be larger than this value. It is because water was used as the trapping medium for tritium; as sampled air was bubbled through the medium, a portion of water was lost to evaporation. This resulted in a drop in resistance to air flow and an increase of sampling flow rate. Since data to characterize the variability of the actual sample flow rates were not located, it is assumed that the correction factor of tritium sampling has a triangular distribution with a best estimate of 1.2 and an upper and lower bounds of 1.5 and 0.9, respectively.

The second identified source of potential significant error is in the last step of the process, the quantification of the effluent flow rate (Q) needed to establish the total volume of air released from the plant. As discussed in Section 2.2.1.5, there was little documentation of the methods used to establish the flow rates or the actual air volumes used to calculate emissions from the plant during the 1950s, 1960s, and early 1970s. The data review suggested that the basis for the flow rates used by the plant may have ranged from engineering design flow rates to rates based on a variety of different sampling or testing programs. In the mid-1970s, the plant began to employ flow totalizers to quantify flow rates in the exhaust systems, which should have reduced the importance of the flow rate factor as a source of uncertainty; however, indications from a recent study are that this may not be the case. It is estimated that the actual average annual exhaust flow volumes may have ranged from one-half to two times the standard volumes that were used in most calculations. Therefore, the uncertainty in emissions estimated due to the lack of documentation of exhaust flow volumes was represented by treating emission estimates as triangular distributions with lower and upper bounds at 0.5 and 2 times the reported value, with the most probable value equal to the reported value.

Beginning in 1974, the plant also began reporting the average relative error associated with the techniques used to quantify the various contaminants present in effluents. The plant annual environmental reports indicate that these error terms include all random and systematic errors in the standards, analytical chemistry, and measurement process for elements collected on effluent filters. Investigators were unable to clearly establish whether the plant corrected the reported release estimates for these errors; however, it is believed that they did not. In order to compensate for the analytical bias, correction factors based on the relative errors will be applied to the release estimates reported between 1974 and 1989 (Table 4-1).

Information about the accuracy of the long-lived gross alpha data measured before 1974 is not available. As there was a continuous improvement of radiation detection technology in the 1960s and 1970s, it is reasonable to assume that the relative error introduced by the counting procedure had also decreased over time. For the purpose of this evaluation, it was assumed that the uncertainty associated with gross alpha counting before 1974 is about twice as large as that of Pu-239/240 measured between 1974 and 1989. Using this approach, the relative error associated with gross alpha analysis performed between 1953 and 1973 was represented by a normal distribution with a mean of 1.1 and a standard deviation of 0.3.

In the case of tritium sampling, a study (described in Section 2.2.1.1.) was also identified that indicates that poor efficiency with the standard sampling device. Poor tritium collection efficiencies are believed to result in the underestimation of tritium emissions. The uncertainty associated with tritium sampling efficiency was represented by treating emission estimates as triangular distributions with lower and upper bounds at 1.3 to 4.8 times the reported values and a most probable value of 3.0.

4.3 Uncertainty Associated with Estimating Specific Isotopic Releases Based on Nonspecific Monitoring Data

Before 1973, only long-lived gross alpha activities in effluent flows were routinely measured and reported. As described in Section 2.4, release estimates of specific isotopes for this time period were carried out in two steps. First, long-lived gross alpha activity sampled in effluents from buildings in which plutonium was handled was assumed to consist solely of those alpha-emitting nuclides associated with weapons grade plutonium, and activity in effluents from buildings in which uranium was handled was assumed to consist solely of those radionuclides associated with enriched or depleted uranium, depending on the type of uranium handled. Second, based on known ratios of and Am-241 to Pu-239/240 and Pu-241 to Pu-239/240, release estimates of Pu-239/240, Am-241 and Pu-241 were calculated from the gross alpha data. Potential sources of uncertainty associated with this approach are evaluated and estimated in Appendix F.

TABLE 4-1

CORRECTION FACTORS TO COMPENSATE FOR THE ANALYTICAL ERRORS IN THE MONITORING RESULTS REPORTED FROM 1974 THROUGH 1989

Year	Plutonium-239/240	Uranium-234, -235, -238	Americium-241	Tritium	Beryllium
1974	1.34	1.44	1.20	1.03	2.27
1975	1.17	1.35	1.49	1.06	1.41
1976	1.09	1.13	1.16	1.07	1.55
1977	1.09	NA	NA	1.05	1.31
1978	1.16	1.14	NA	0.96	NA
1979	0.88	1.16	1.24	0.93	1.44
1980	1.35	0.97	1.23	0.97	2.16
1981	0.92	0.96	1.08	0.95	0.75
1982	0.95	0.71	1.17	0.97	0.47
1983	1.09	1.13	1.05	0.91	0.95
1984	1.10	1.10	1.11	0.92	0.96
1985	1.12	1.13	1.30	0.90	0.96
1986	1.02	1.07	1.02	0.93	1.05
1987	0.96	0.97	1.10	0.89	0.99
1988	1.06	0.75	0.95	0.76	0.94
1989	0.89	0.87	1.06	0.90	0.95
Mean =	1.07	1.06	1.15	0.95	1.21
Standard Deviation =	0.14	0.20	0.13	0.08	0.49

N/A = Not Available

Even after 1973, airborne releases of beta-emitting Pu-241 were not routinely measured by the plant. Therefore, source terms of Pu-241 from 1974 to 1989 were calculated from release estimates of Pu-239/240 and special study results which quantified the ratio of Pu-241 to Pu-239/240 in effluent releases. Uncertainty associated with this calculation is also estimated in Appendix F.

4.4 Overall Uncertainty Associated with Release Estimates of Contaminants

In summary, review of the sampling and analytic systems used to monitor plant effluent identified a number of potential sources of random and systematic errors. The potential sources of random error are numerous; however, the contribution of these sources of error to the overall uncertainty in the annual emission estimates is small given the large number of samples that were collected to establish the annual estimate. Potential sources of systematic error due to lack of information about the identity of the analyte and estimation of sampling flow rates and exhaust flow rates are identified. Uncertainty factors were developed to represent these potential sources of uncertainty. As described in Appendix G, overall uncertainty factors for the release estimates of the contaminants of concern were calculated using a statistical technique called Monte Carlo simulation. A brief description of Monte Carlo simulation and its application in this evaluation is provided in Appendix H.

The annual emission estimates were then multiplied by the appropriate overall uncertainty factors to generate a probability distribution for the annual emissions. It is believed that the use of these emission probability distributions will bound the actual emissions in a year. If dose assessment results suggest the need, the range of uncertainty in these emissions could probably be reduced following further evaluation of plant practices. The historical emission estimates and their uncertainties that will be used to estimate off-site doses resulting from plant operations are summarized in Table 4-2 through 4-4 and Figures 4-1 through 4-6 for plutonium, uranium, americium-241, and tritium. The lower and upper bounds identified in the tables and figures represent the boundaries of the 95 percent confidence interval centered on the geometric mean.

The emission estimates for tritium prior to 1974 are not based on measurements. The estimated range of annual tritium emissions for this period was treated as a uniform distribution with the identified lower and upper bounds of 1 and 800 for the period of 1953 through 1967 and 140 and 390 for the period of 1968 through 1973.

Table 4-5 and Figure 4-7 provide the release estimates and the uncertainties for beryllium. They are based on the release estimates reported in Section 3.1 and the two overall uncertainty factors of beryllium developed as described in Appendix G. The lower and upper bounds identified in the table and figures represent the boundaries of the 95 percent confidence interval centered on the geometric mean.

TABLE 4-2
AIRBORNE EMISSION ESTIMATES AND UNCERTAINTIES—PLUTONIUM

Year	Plutonium Alpha Activity (μCi)			Plutonium-241 (μCi)		
	95% Lower Confidence Limit	Geometric Mean	95% Upper Confidence Limit	95% Lower Confidence Limit	Geometric Mean	95% Upper Confidence Limit
1953	0.81	2.1	5.3	4.4	11	29
1954	27	69	180	130	320	830
1955	30	77	200	140	360	920
1956	97	250	630	450	1,200	2,900
1957	6,100	16,000	40,000	30,000	76,000	190,000
1958	1,300	3,300	8,300	6,100	16,000	40,000
1959	560	1,400	3,700	2,700	7,000	18,000
1960	560	1,400	3,700	2,500	6,500	17,000
1961	610	1,600	4,000	3,000	7,600	19,000
1962	1,300	3,300	8,300	6,100	16,000	40,000
1963	1,500	3,900	10,000	7,000	18,000	46,000
1964	1,200	3,000	7,700	5,600	14,000	37,000
1965	2,700	6,900	18,000	13,000	32,000	83,000
1966	130	340	870	610	1,600	4,000
1967	170	430	1,100	800	2,000	5,200
1968	200	520	1,300	940	2,400	6,100
1969	560	1,400	3,700	2,700	7,000	18,000
1970	160	400	1,000	750	1,900	4,900
1971	31	79	200	150	370	950
1972	25	65	170	120	300	770
1973	26	66	170	120	310	800
1974	490	1,200	3,200	2,300	6,000	15,000
1975	5.1	13	33	24	62	160
1976	2.0	5.2	13	9.8	25	65
1977	2.0	5.2	13	9.8	25	65
1978	1.4	3.6	9.3	7.0	18	46
1979	2.8	7.2	18	14	35	89
1980	6.1	16	40	29	74	190
1981	4.2	11	27	20	52	130
1982	10	26	67	47	120	310
1983	40	100	260	190	490	1,300
1984	40	100	260	190	490	1,300
1985	4.7	12	31	23	58	150
1986	15	38	97	70	180	460
1987	7.6	20	50	37	94	240
1988	7.6	20	50	37	94	240
1989	2.3	5.9	15	11	28	71

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TABLE 4-3
AIRBORNE EMISSION ESTIMATES AND UNCERTAINTIES—URANIUM

Year	Enriched Uranium (μCi)			Depleted Uranium (μCi)		
	95% Lower Confidence Limit	Geometric Mean	95% Upper Confidence Limit	95% Lower Confidence Limit	Geometric Mean	95% Upper Confidence Limit
1953	4.6	12	30	610	1,600	4,000
1954	4.6	12	30	610	1,600	4,000
1955	38	96	250	1,100	2,700	7,000
1956	560	1,400	3,700	510	1,300	3,300
1957	180	470	1,200	370	950	2,400
1958	160	400	1,000	810	2,100	5,300
1959	270	700	1,800	140	350	900
1960	440	1,100	2,900	180	460	1,200
1961	240	620	1,600	260	680	1,700
1962	130	320	830	190	480	1,200
1963	170	430	1,100	250	640	1,600
1964	96	250	630	120	310	800
1965	96	250	630	140	360	930
1966	120	300	770	71	180	470
1967	56	140	370	71	180	470
1968	81	210	530	71	180	470
1969	25	65	170	81	210	530
1970	33	83	210	96	250	630
1971	21	53	140	29	75	190
1972	2.0	5.2	13	21	55	140
1973	6.1	16	40	27	70	180
1974	14	35	90	4.6	12	30
1975	14	36	93	14	36	93
1976	8.1	21	53	6.1	16	40
1977	11	27	70	9.7	25	63
1978	11	27	70	17	43	110
1979	4.7	12	31	13	34	87
1980	7.6	20	50	7.6	20	50
1981	6.1	16	40	9.1	23	60
1982	6.1	16	40	9.7	25	63
1983	10	26	67	16	40	100
1984	10	26	67	2.8	7.2	18
1985	4.0	10	26	20	51	130
1986	5.6	14	37	1.5	3.8	9.7
1987	2.3	6.0	15	6.1	16	40
1988	1.3	3.4	8.7	4.7	12	31
1989	2.6	6.8	17	1.3	3.3	8.3

1019ALR6

TABLE 4-4
AIRBORNE EMISSION ESTIMATES AND UNCERTAINTIES—AMERICIUM-241 AND TRITIUM

Year	Americium-241 (μCi)			Tritium (Ci)		
	95% Lower Confidence Limit	Geometric Mean	95% Upper Confidence Limit	95% Lower Confidence Limit[1]	Geometric Mean[1]	95% Upper Confidence Limit[1]
1953	0.20	0.50	1.3	1	NA	800
1954	6.6	17	43	1	NA	800
1955	7.1	18	47	1	NA	800
1956	22	57	150	1	NA	800
1957	1,500	3,800	9,700	1	NA	800
1958	310	780	2,000	1	NA	800
1959	140	350	900	1	NA	800
1960	130	320	820	1	NA	800
1961	150	380	970	1	NA	800
1962	300	760	1,900	1	NA	800
1963	360	910	2,300	1	NA	800
1964	270	700	1,800	1	NA	800
1965	660	1,700	4,300	1	NA	800
1966	32	81	210	1	NA	800
1967	39	100	260	1	NA	800
1968	48	120	320	140	NA	390
1969	140	350	900	140	NA	390
1970	37	95	240	140	NA	390
1971	7.1	18	47	140	NA	390
1972	6.0	15	39	140	NA	390
1973	6.0	15	39	140	NA	390
1974	110	290	750	11	25	57
1975	12	31	79	1.7	3.9	8.8
1976	0.48	1.2	3.2	1.4	3.1	7.0
1977	0.48	1.2	3.2	0.61	1.4	3.1
1978	0.34	0.87	2.2	1.0	2.3	5.3
1979	0.66	1.7	4.3	0.97	2.2	4.9
1980	1.4	3.6	9.3	0.90	2.0	4.6
1981	0.98	2.5	6.5	0.51	1.1	2.6
1982	2.4	6.2	16	0.27	0.60	1.4
1983	9.3	24	61	0.18	0.42	0.94
1984	9.3	24	61	0.16	0.36	0.82
1985	1.1	2.8	7.2	0.18	0.42	0.94
1986	5.0	13	33	0.25	0.57	1.3
1987	1.8	4.5	12	0.20	0.44	0.99
1988	1.1	2.8	7.2	0.030	0.068	0.15
1989	0.60	1.5	3.9	0.20	0.44	0.99

[1] Tritium emissions for 1953 through 1973 are treated as uniform distributions with identified lower and upper bounds.

NA = Not Applicable

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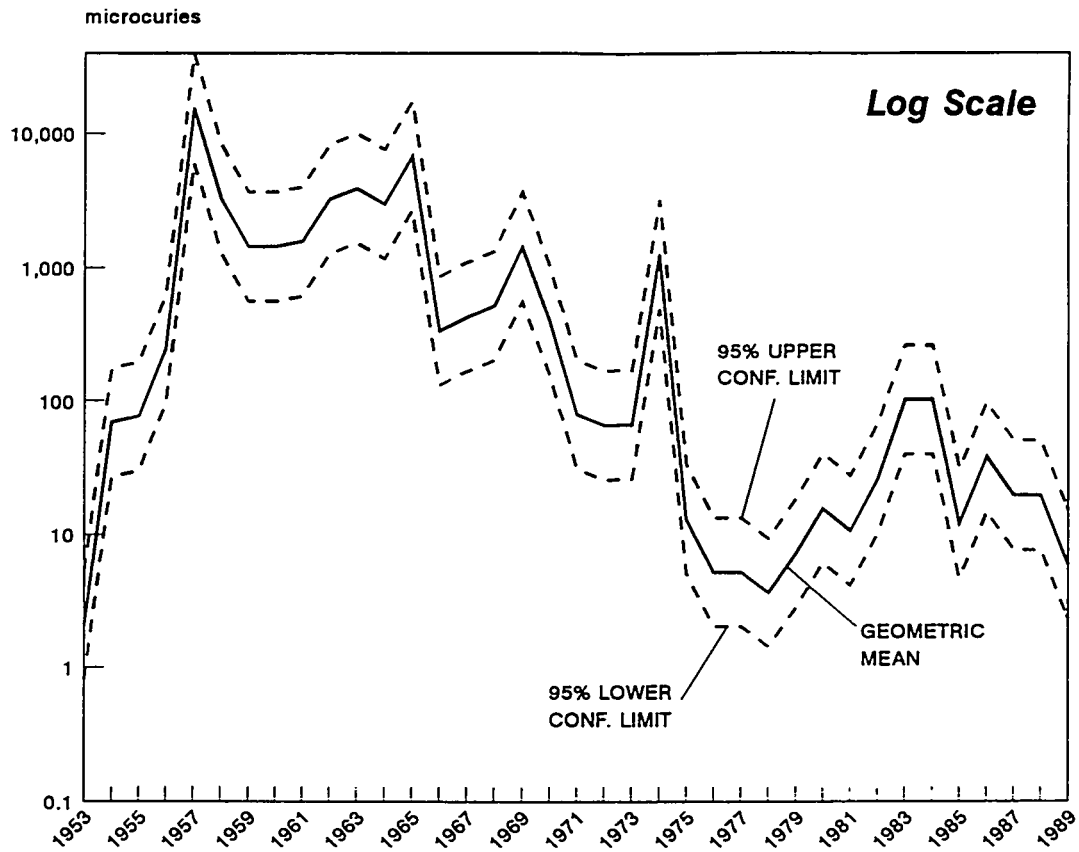

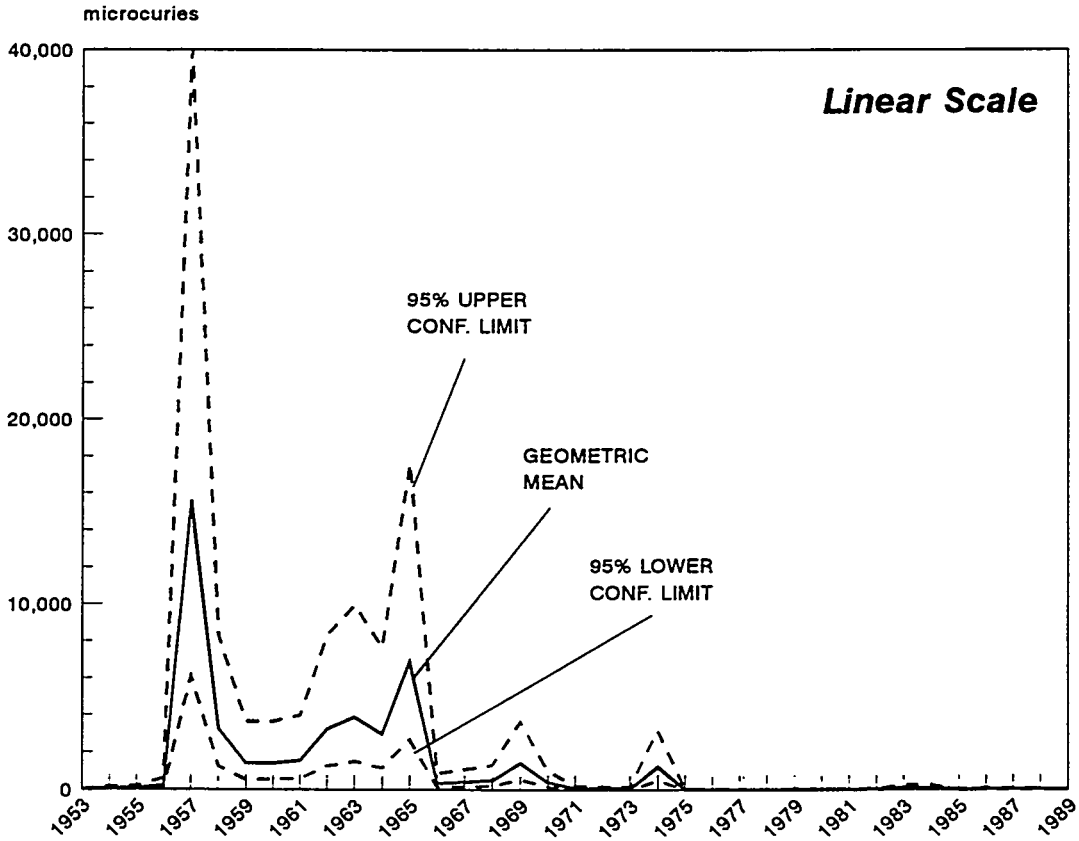

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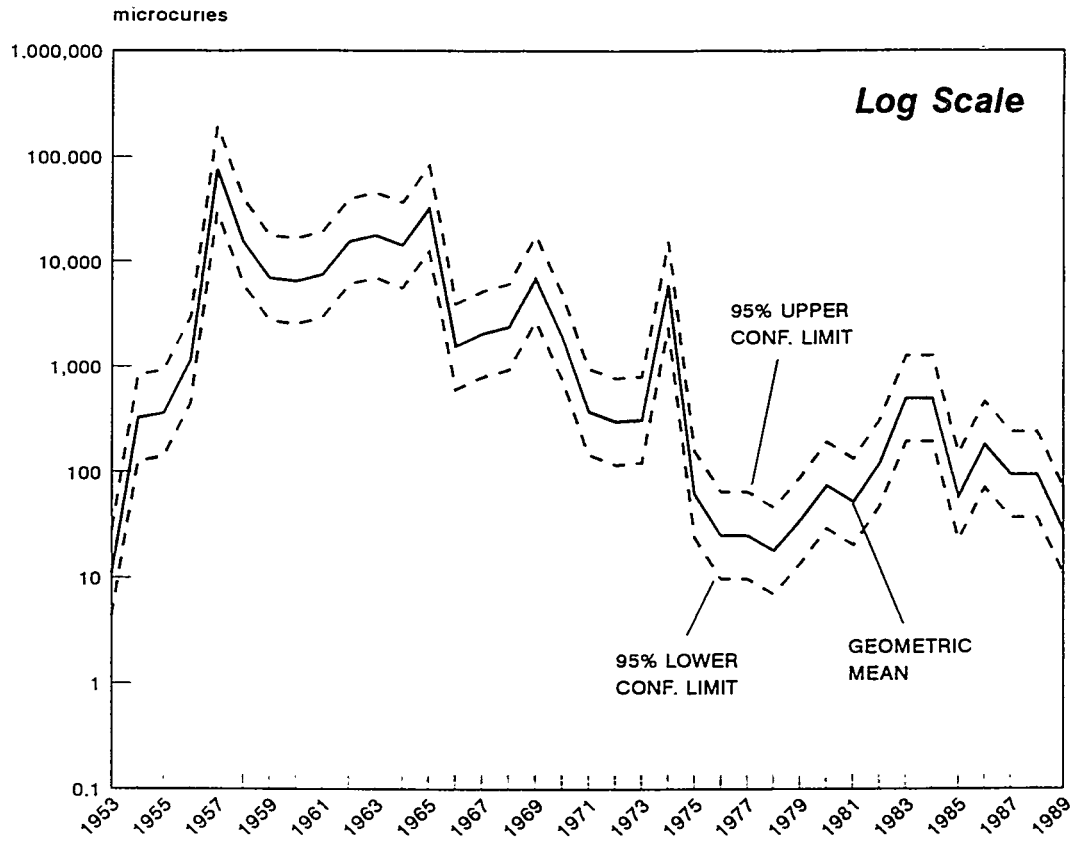
FIGURE 4-1
AIRBORNE RELEASE ESTIMATES AND
UNCERTAINTIES - PLUTONIUM ALPHA
(μCi)

RKY294T5/4-1; REV: 02-22-94



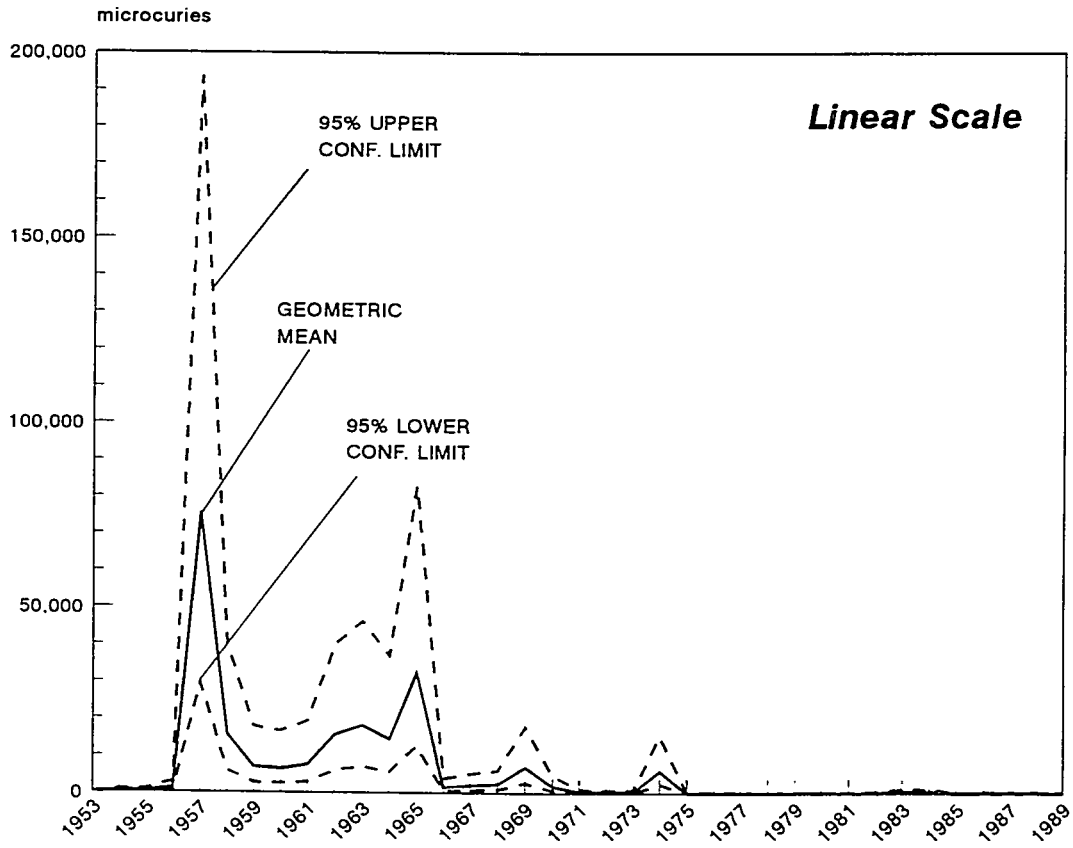


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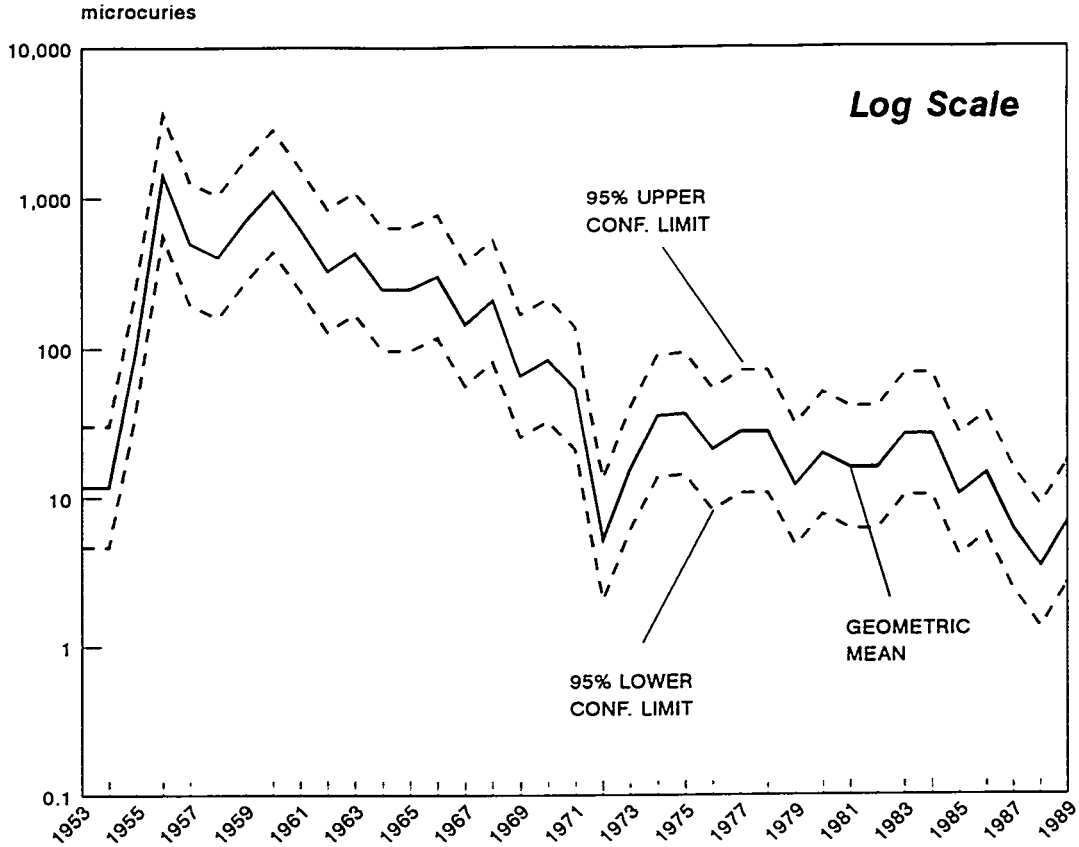
FIGURE 4-2
AIRBORNE RELEASE ESTIMATES AND
UNCERTAINTIES - PLUTONIUM-241
(μCi)



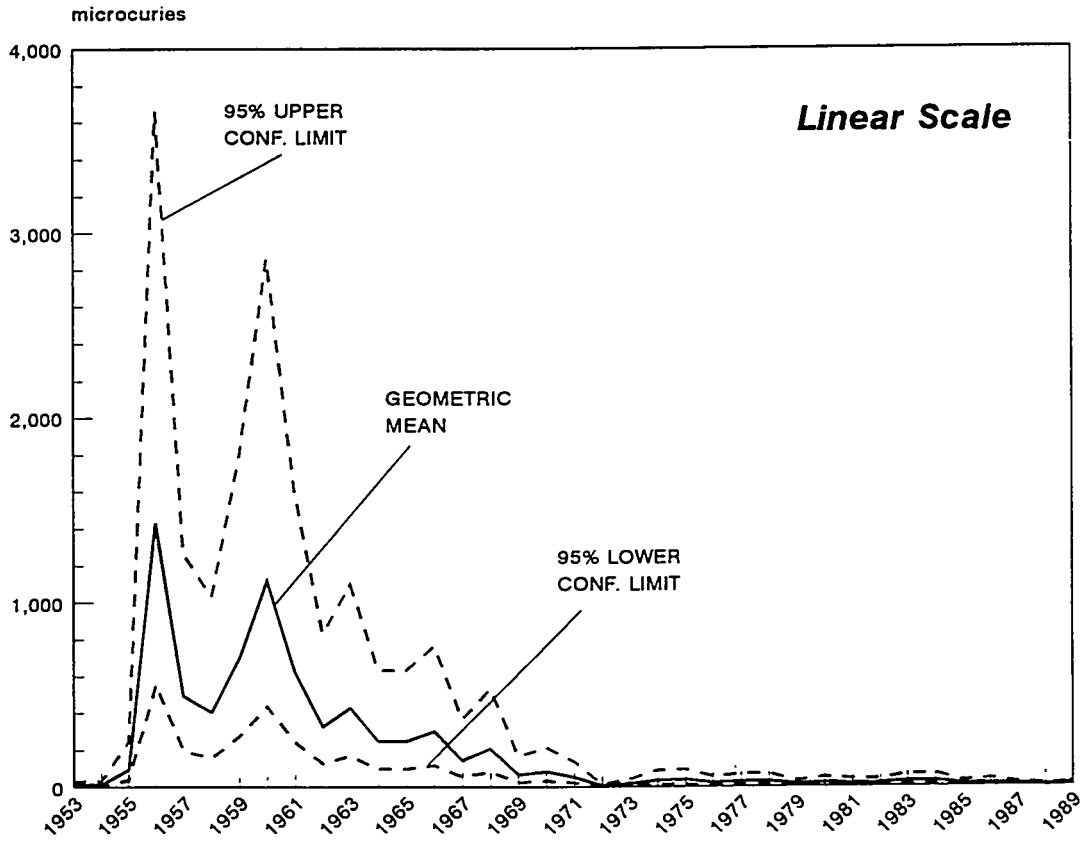


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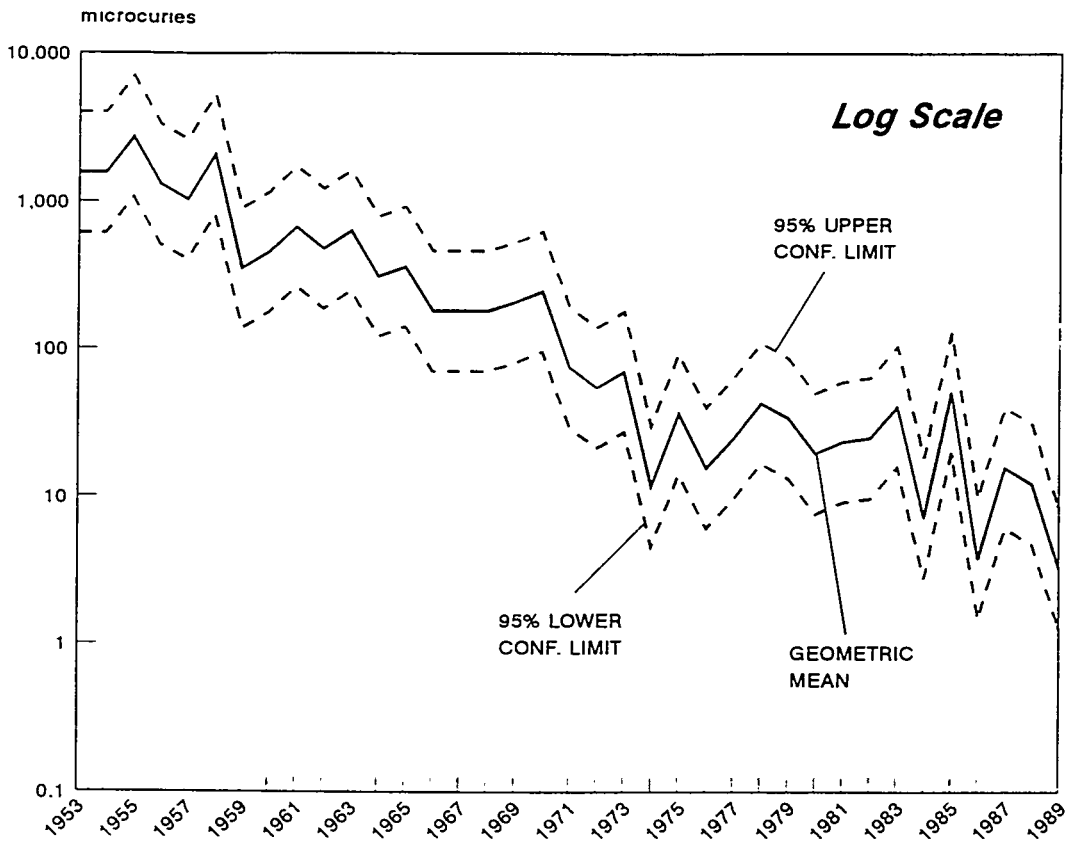
FIGURE 4-3
AIRBORNE RELEASE ESTIMATES AND
UNCERTAINTIES - ENRICHED URANIUM
(μCi)

RKY294T5/4-3; REV: 02-22-94



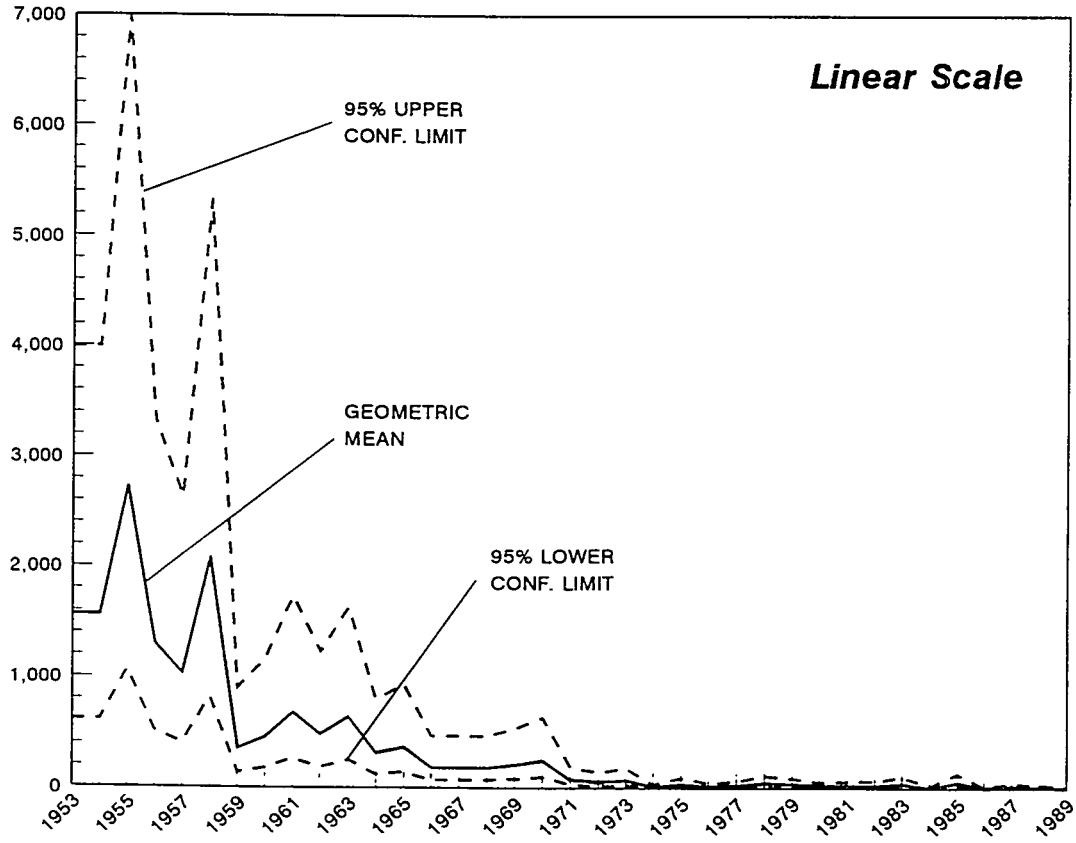


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FIGURE 4-4
AIRBORNE RELEASE ESTIMATES AND
UNCERTAINTIES - DEPLETED URANIUM
(μCi)

RKY294T5/4-4; REV: 02-22-94

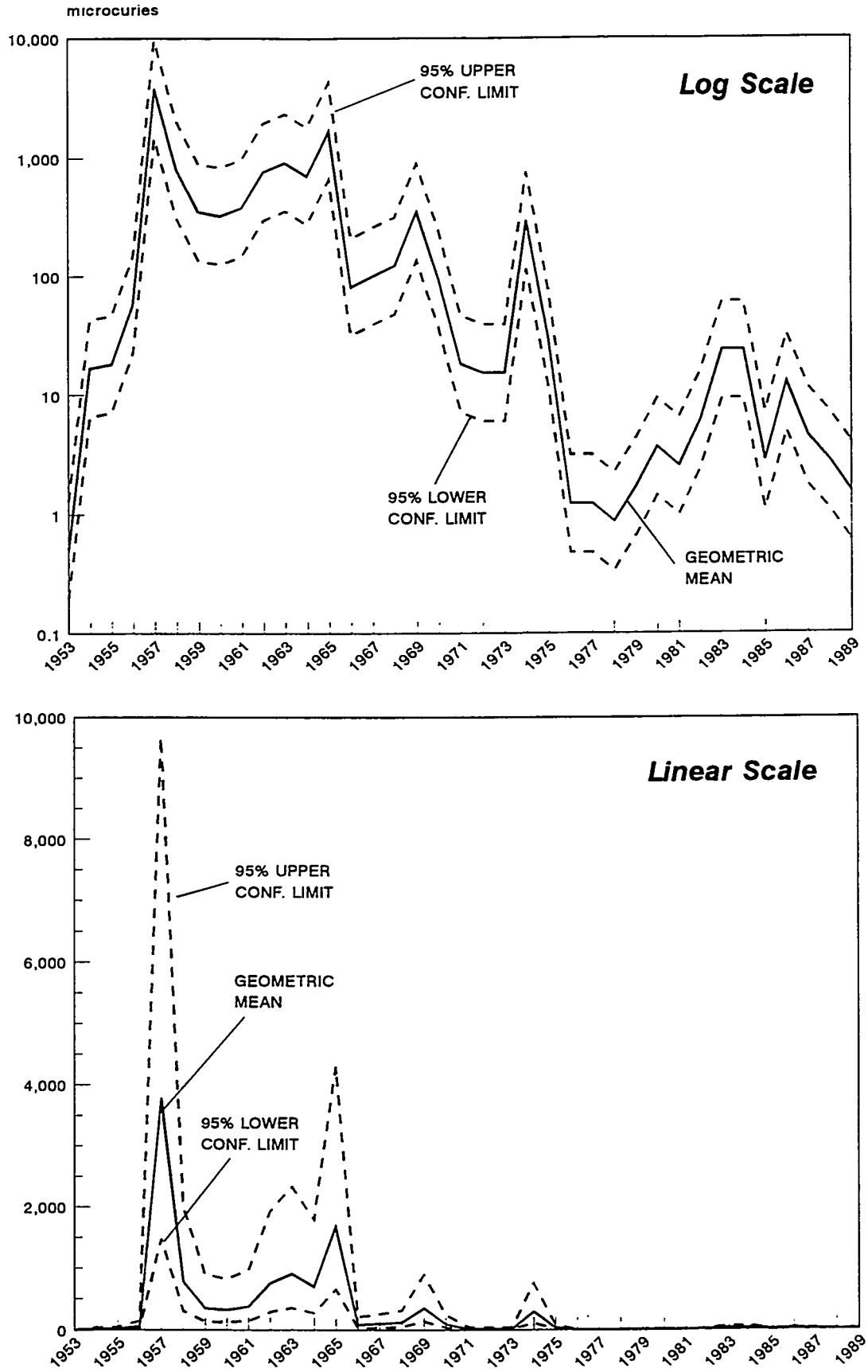

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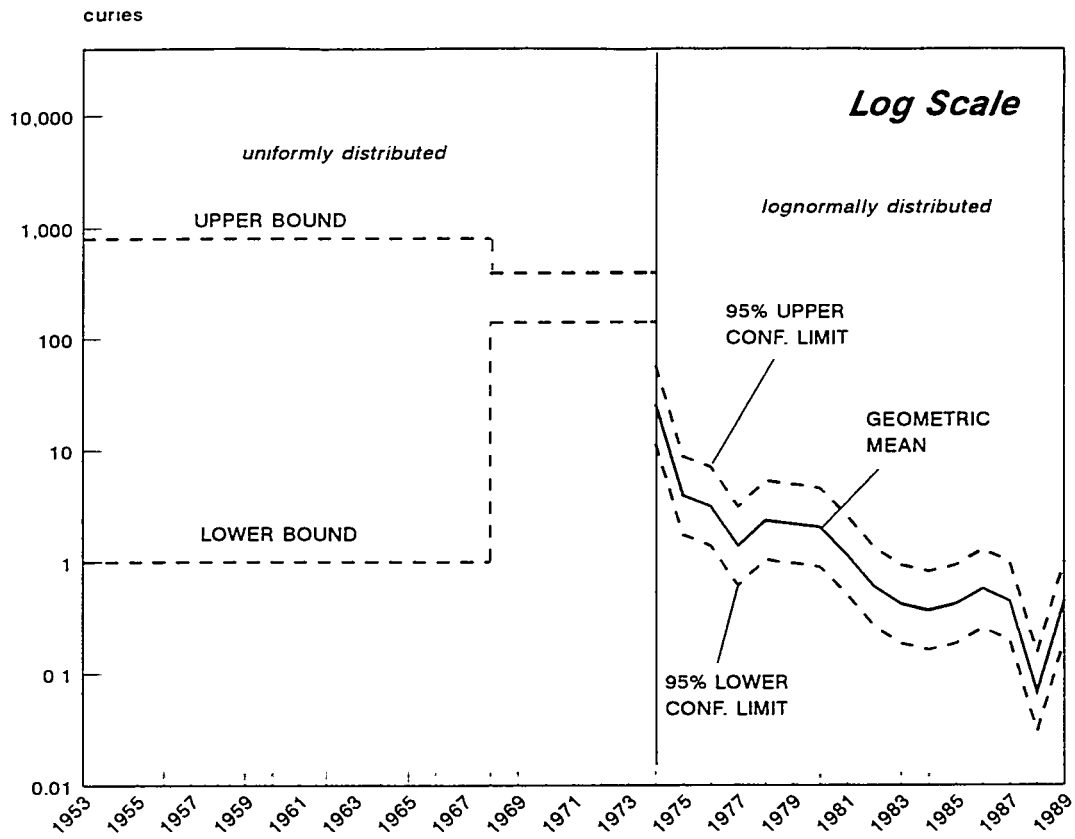
FIGURE 4-5
AIRBORNE RELEASE ESTIMATES AND
UNCERTAINTIES - AMERICIUM-241
(μCi)

RKY294T5/4-5; REV: 02-22-94



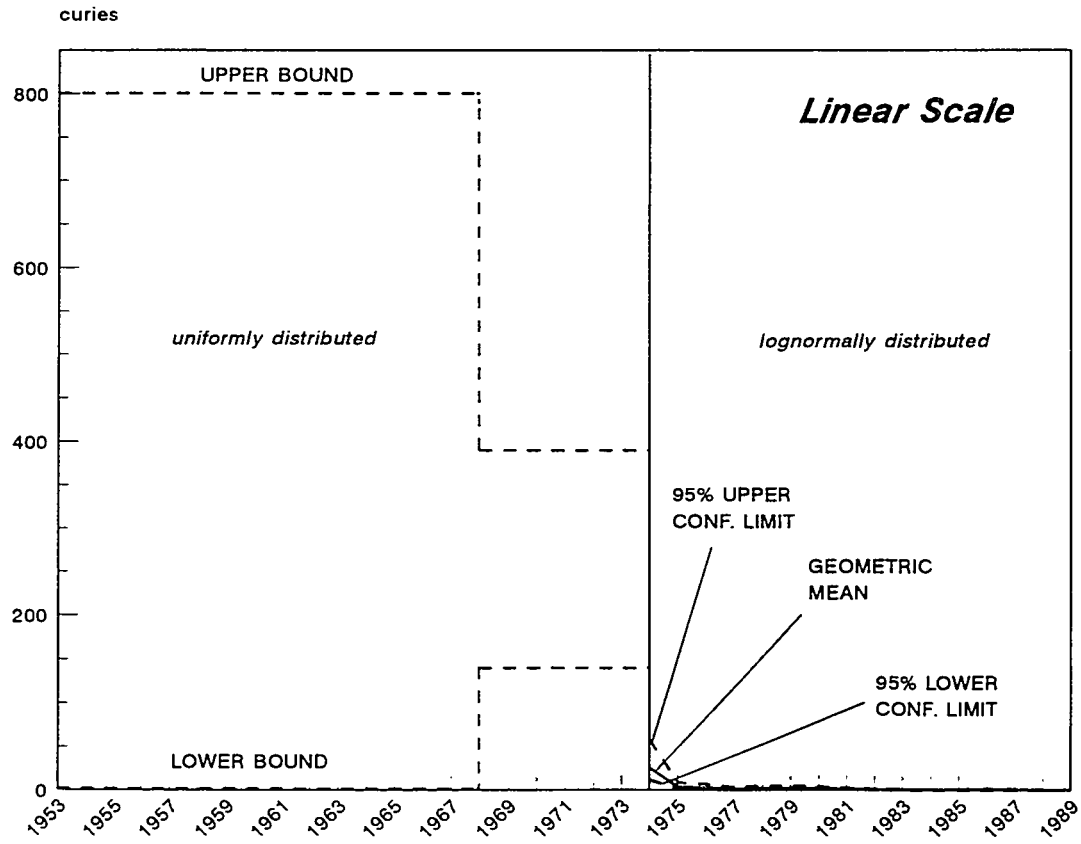


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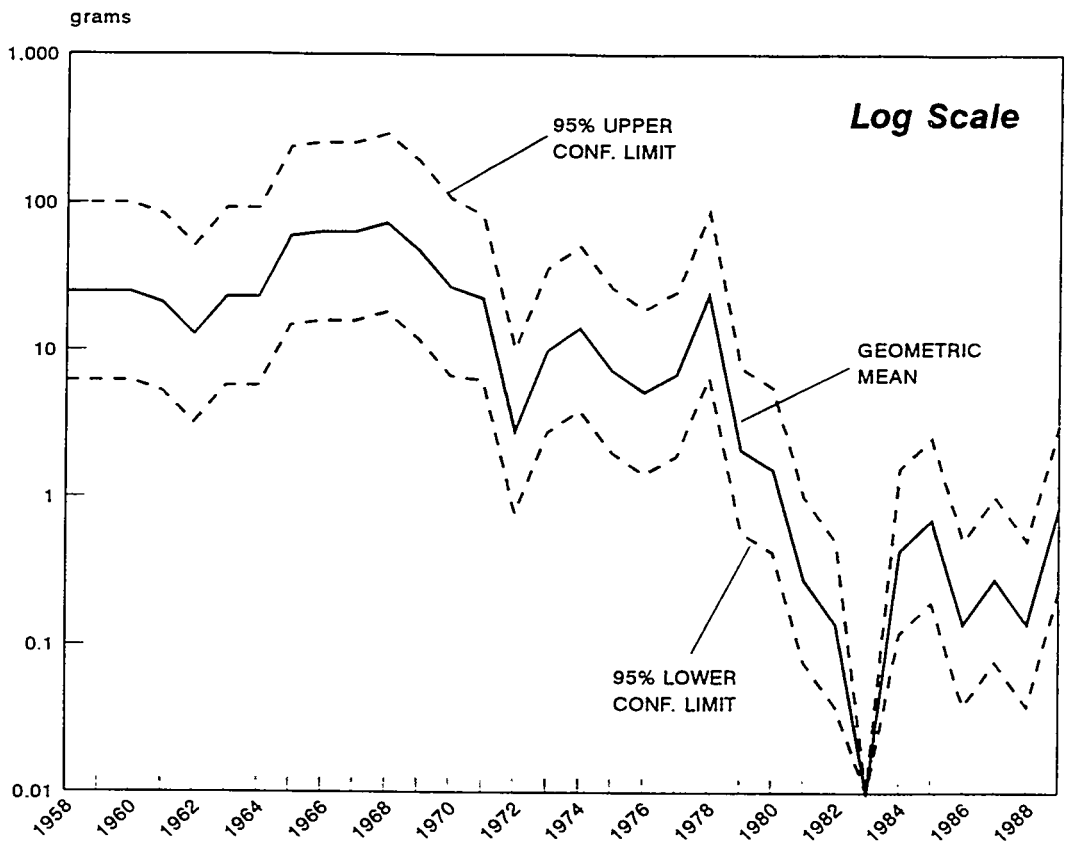
FIGURE 4-6
AIRBORNE RELEASE ESTIMATES AND UNCERTAINTIES- TRITIUM
(Ci)

RKY294T5/4-6; REV: 02-22-94

TABLE 4-5

AIRBORNE EMISSION ESTIMATES AND UNCERTAINTIES— BERYLLIUM (g)

Year	95% Lower Confidence Limit	Geometric Mean	95% Upper Confidence Limit
1958	6.2	25	99
1959	6.2	25	99
1960	6.2	25	99
1961	5.2	21	84
1962	3.2	13	51
1963	5.7	23	91
1964	5.7	23	91
1965	15	59	240
1966	16	63	250
1967	16	63	250
1698	18	72	290
1969	12	48	190
1970	6.7	27	110
1971	6.2	22	81
1972	0.78	2.8	10
1973	2.8	9.9	36
1974	3.9	14	51
1975	2.0	7.3	26
1976	1.4	5.2	19
1977	1.9	6.9	25
1978	6.6	24	86
1979	0.58	2.1	7.6
1980	0.43	1.5	5.6
1981	0.078	0.28	1.0
1982	0.039	0.14	0.51
1983	0	0	0
1984	0.12	0.43	1.6
1985	0.19	0.70	2.5
1986	0.039	0.14	0.51
1987	0.078	0.28	1.0
1988	0.039	0.14	0.51
1989	0.23	0.84	3.0



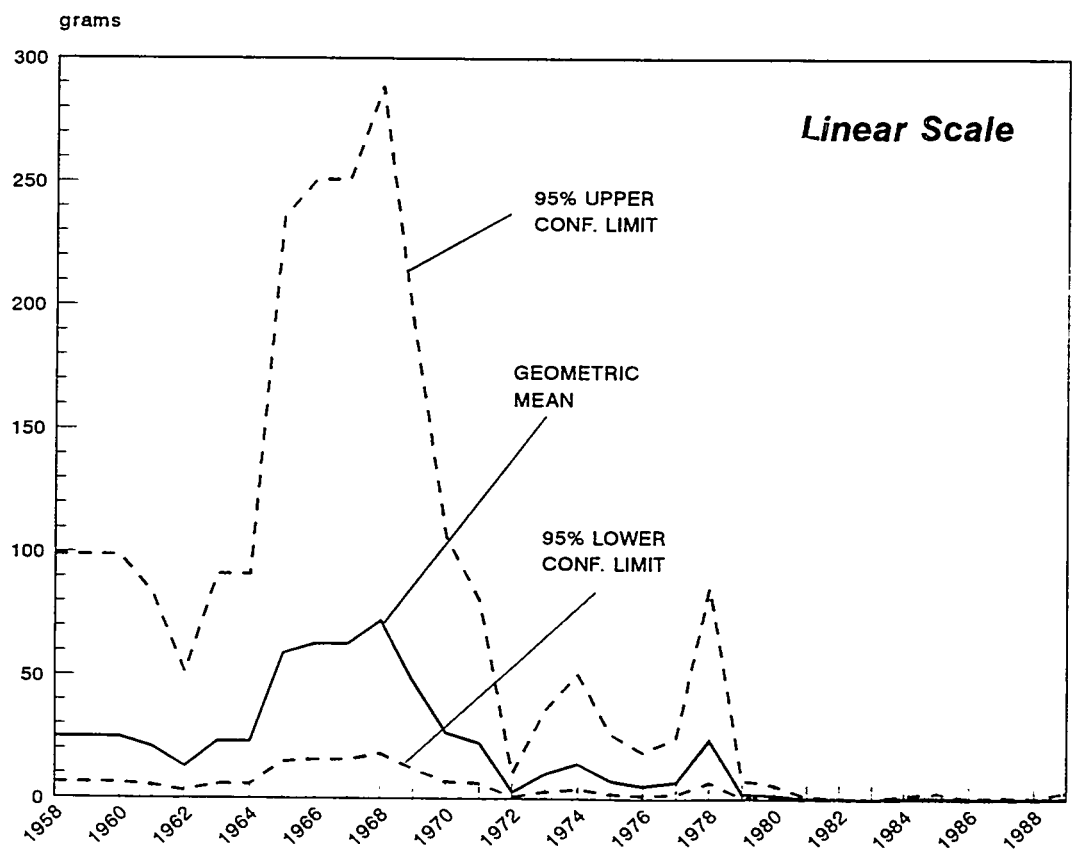


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FIGURE 4-7
AIRBORNE RELEASE ESTIMATES AND
UNCERTAINTIES - BERYLLIUM
(g)

RKY294T5/4-7: REV: 02-22-94

5.0 ROUTINE CONTAMINANT RELEASES TO SURFACE WATERS

The report of Tasks 3 and 4 detailed the history of the system of ponds that have been developed at the site to receive various plant effluents and site runoff. Historical plant operations resulted in the release of contaminants into the pond systems, and in some cases, directly into site drainages. Surface waters provide a means of transporting contaminants off the site where exposures to the public may have occurred. For the purposes of this project, we are interested in characterizing the extent to which the contaminants of interest have traveled off-site in surface waters.

The contaminants of interest for further study on the project include both radioactive and nonradioactive materials. The nonradioactive materials of interest are all volatile solvents, with the exception of beryllium, which is a metal. Studies indicate that the primary removal process of volatile organic compounds from surface water is evaporation. Half-lives for volatile organic compounds can range from minutes to days depending on the physical properties of the compound and mixing condition of the water body (Dilling, 1975; Dilling, 1977; Schwarzenbach, 1979 and Cohen, 1978). Based on this information, volatile contaminants of concern released in liquid effluents by the facility are assumed to be readily evaporated from surface waters. Since the early days of Rocky Flats operations, liquid wastes discharged from the facility were temporarily stored in retention ponds prior to release. As a result, volatile contaminants of concern released with liquid effluents are unlikely to have been transported off-site in surface waters to any significant extent.

With the exception of tritium, the radioactive materials of interest are metals. Tritium is readily transported in surface waters. The metals of concern have low solubility in water and will tend to be adsorbed by soils or sediment. This tendency would have reduced the concentration of dissolved contaminants in water.

The following sections review the history of the surface water bodies in the vicinity of the Rocky Flats Plant and the availability of surface water monitoring data. Additionally, the approach to quantifying contaminant concentrations in surrounding surface waters is described.

5.1 Surface Water History

The Tasks 3 and 4 report discussed the history of the surface waters surrounding the Rocky Flats plant. This section briefly revisits the development of holding ponds and reservoirs.

5.1.1 Retention Ponds

A number of retention ponds were constructed along the creeks that drain the Rocky Flats site for use in management of plant wastes and surface water runoff. The purpose of the retention ponds was to hold wastewater on-site for a period of time to allow any volatiles contained in the wastewater to evaporate and to allow any metals to settle out and/or bind to pond sediments. The ponds that historically received plant releases are now known as the A-series and B-series ponds. They are located on North Walnut Creek and South Walnut Creek. The C-series ponds, which received runoff from the site, are located on Woman Creek. Prior to the pond reconstruction activities that took place in the early 1970s, the ponds were not given letter designators. Plant documents prepared prior to 1973 refer to each pond by number. Table 5-1 presents the original number and current designator of each pond still in operation in 1992. Figure 5-1 depicts the holding ponds as they exist today (1992).

5.1.1.1 North Walnut Creek Drainage

The history of activities related to the North Walnut Creek drainage is depicted on the time line in Figure 5-2. Between 1953 and 1957, the plant reported that approximately 2.23 mCi of low-level contaminated laundry wastes were discharged either directly to North Walnut Creek or to Pond A-1 after it was completed in July 1953 (USDOE, 1991a). In 1957, laundry wastewater was rerouted to Building 774, where it was treated along with process wastes and released to South Walnut Creek. After 1957, Pond A-1 was reported to have received overflow laundry wastewater, cooling tower blowdown, and steam condensate. The laundry wastewater discharges to North Walnut Creek resulted in the accumulation of plutonium in the sediments of Pond A-1 and North Walnut Creek. In the early 1970s, Pond A-1 underwent a major reconstruction and retention Ponds A-2 and A-3 were added to the North Walnut Creek drainage. The reconstruction activities resulted in sediment resuspension from Pond A-1 and likely increased the potential for release of plutonium to Great Western Reservoir. When the reconstruction activities were completed, Ponds A-1, A-2, and A-3 received cooling tower blowdown and steam condensate from process and laboratory facilities until 1974. From 1974 to 1980, Pond A-2 was isolated from North Walnut Creek and Ponds A-1 and A-3 collected surface runoff. In 1980, Pond A-4 was completed. From 1980 to 1989, Ponds A-3 and A-4 collected surface runoff, while Ponds A-1 and A-2, isolated from North Walnut Creek, received water from Pond B-2.

5.1.1.2 South Walnut Creek Drainage

The South Walnut Creek drainage pond history is also depicted in Figure 5-2. Ponds B-1, B-2, and B-3 were constructed prior to the start of production activities at Rocky Flats. The three B-series ponds received decontaminated process wastewater, sewage treatment plant effluent, and laundry wastewater from 1952 to 1970. The water was passed through each holding pond and

TABLE 5-1

RETENTION POND ORIGINAL AND CURRENT DESIGNATIONS

Original Number	Current Designator
Pond 1	Pond A-1
Pond 2	Solar Evaporation Pond
Pond 3	Pond B-1
Pond 4	Pond B-2
Pond 5	Pond B-3
Pond 6	Pond no longer present
Pond 7	Pond no longer present
Pond 8	Pond no longer present
Pond 9	Pond C-1

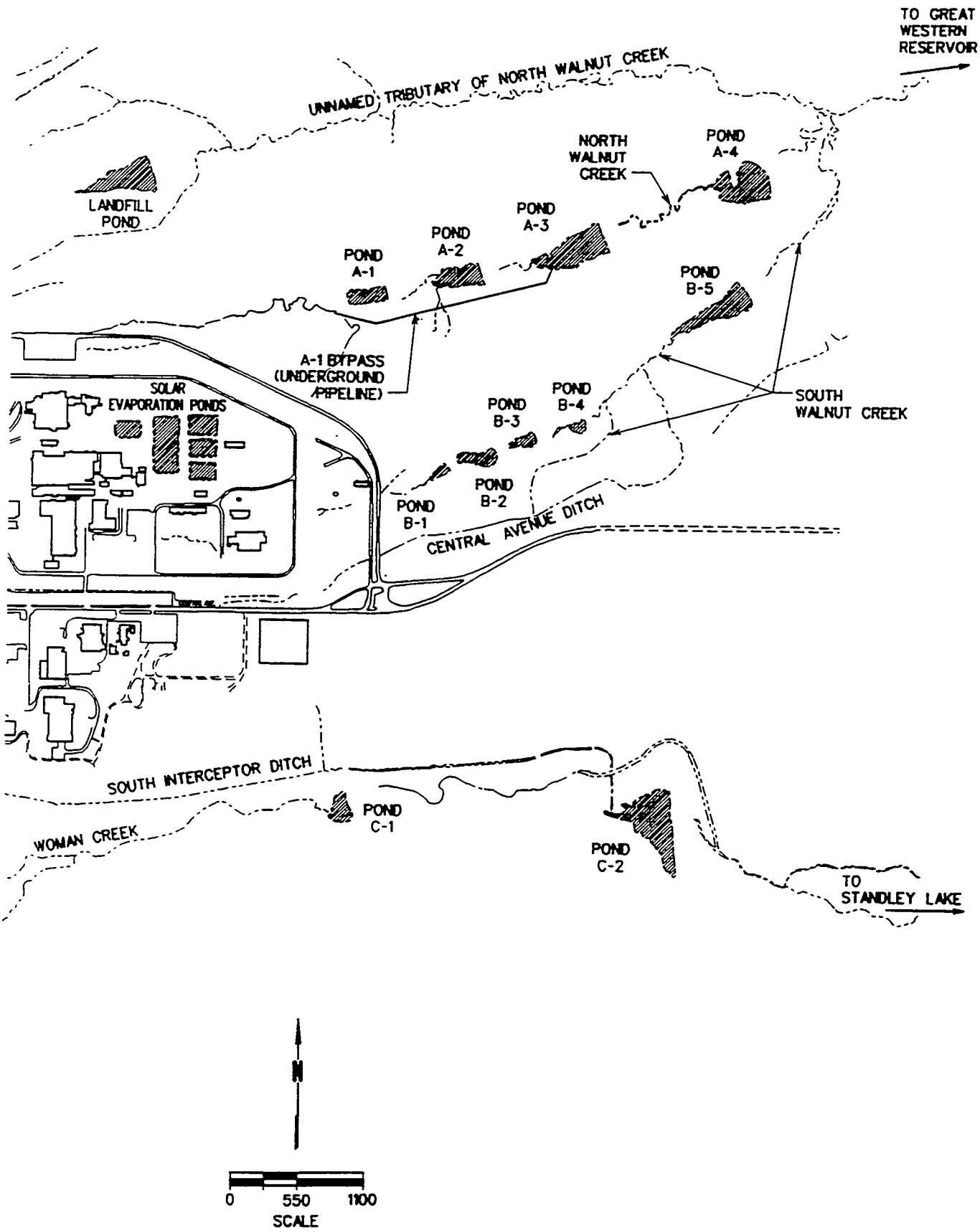

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FIGURE 5-1
ROCKY FLATS HOLDING PONDS
AND CREEKS, 1992

RKY294T5/5-1; REV: 02-22-94

SOUTH WALNUT CREEK

1952

Ponds B-1, B-2, and B-3 completed 1952. The ponds received decontaminated process wastewater and sewage treatment plant (STP) effluent. Pond B-3 was the final holding pond.

1957

Ponds received decontaminated process wastewater, STP effluent, and laundry wastewater.

Pond B-4 completed. Ponds B-1, B-2, and B-3 underwent major reconstruction. This resulted in the resuspention of pond sediments and released radionuclides to Great Western Reservoir. All ponds received wastewater from the secondary treatment facility.

1970

1973

1974

Liquid sanitary wastes discharged to ponds B-1, B-2, and B-4. Pond B-2 isolated — used to impound process wastewater which was pumped to pond A-2.

Pond B-5 completed.

1980

Ponds B-1 and B-2 used as back-up surface water control ponds. Pond B-3 used for spray evaporation. Ponds B-4 and B-5 stored surface runoff from the central portion of the site. Liquid sanitary waste discharged to South Walnut Creek through pond B-4.

1989

NORTH WALNUT CREEK

No Ponds on North Walnut Creek. The creek directly received low-level contaminated laundry wastewater.

1952

Pond A-1 completed in July 1953. Low-level contaminated laundry wastewater discharged to Pond A-1 then to North Walnut Creek.

1957

From time to time Pond A-1 received laundry liquid waste water, cooling tower blowdown, and steam condensate discharges. The pond was also used to control surface water runoff from the northern portion of the site.

Ponds A-2 and A-3 completed. Pond A-1 underwent major reconstruction. This resulted in the resuspension of pond sediments and released bound radionuclides to Great Western Reservoir. Ponds A-1, A-2, and A-3 received cooling water blowdown and steam condensate from process and lab facilities.

1970

1973

1974

Pond A-2 isolated from North Walnut Creek. The pond was used to store process wastewater. Ponds A-1 and A-3 received surface run off.

Pond A-4 completed.

1980

Ponds A-3 and A-4 receive surface runoff. Ponds A-1 and A-2 isolated from North Walnut Creek. Water in Ponds A-1 and A-2 was spray evaporated.

1989

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FIGURE 5-2
TIME LINE OF EVENTS FOR THE NORTH AND SOUTH WALNUT CREEK DRAINAGE

RKY294T5/5-2; REV: 02-22-94

was finally released to South Walnut Creek through an outlet in Pond B-3. Similar to the North Walnut Creek drainage, the South Walnut Creek drainage (Ponds B-1, B-2, and B-3) underwent reconstruction in the early 1970s. Pond B-4 was constructed at that time. This reconstruction of the streambed upstream of Pond B-1 resulted in the disturbance of sediments containing plutonium. As a result of the sediment disturbance, much of the upstream sediment migrated into Pond B-1 and subsequently increased the plutonium inventory of all the B-series ponds. As with the reconstruction activities on North Walnut Creek, resuspended plutonium from the ponds potentially migrated downstream to Great Western Reservoir during this period.

In the mid-1970s, the plant adopted a zero-release policy for process wastewater. Plant documentation suggests that the process wastewater was discharged into Pond B-2 and pumped to Pond A-2 from which it was spray evaporated with the objective of meeting zero-release. In 1980, Pond B-5 was completed to serve as an overflow pond. From 1980 to 1989, Ponds B-1 and B-2 were used for surface water control. Pond B-3 was used for spray evaporation, and Ponds B-4 and B-5 were used to store stormwater runoff from the central portion of the site. Throughout this time period, liquid sanitary wastes were still discharged to South Walnut Creek through Pond B-4. Although no detailed information on pond releases after the adoption of the zero release policy was located, it is clear that the recent admission of Resource Conservation and Recovery Act (RCRA) violations during this period by Rockwell indicate that the zero discharge policy was an objective that was not met.

5.1.1.3 Woman Creek Drainage

The history of the Woman Creek drainage is depicted in Figure 5-3. Pond C-1 was built in 1952 and received surface runoff, cooling tower blowdown, and filter backwash from the water treatment facility. Ponds 6, 7, and 8 were built on the Woman Creek drainage in 1955 (Dow, 1953-1973). Pond 6 was located south of the water treatment plant, Pond 7 was located south of the Building 881 sewage lift station, and Pond 8 was located south of the Building 881 dock area (Dow, 1953-1973). No information regarding the use of these ponds or the time of their closure was found. However, one interviewee indicated that Pond 6 may have been used to capture filter backwash from the water treatment plant (Hill, 1992). From 1973 to 1980 Pond C-1 was used to control and capture surface runoff. In 1980, Pond C-2 was built to detain runoff water from the South Interceptor Ditch, thus isolating Pond C-1 and Woman Creek from any Rocky Flats runoff. After a period of retention in the C-2 pond, captured water was released into Woman Creek until the line to the Broomfield Diversion Ditch was constructed, and may have periodically been diverted via Mower ditch to Mower Reservoir (Brunch, 1993).

WOMAN CREEK

Pond C-1 received filter backwash from the water treatment facility and cooling tower blowdown. Additionally the pond was used to control surface water runoff from the southern portion of the plant.

1952

Ponds 6, 7, and 8 built. Use unknown. Date of backfill unknown.

1955

Pond C-1 continued to receive filter backwash from the water treatment plant and cooling tower blowdown. Additionally, C-1 was used to control surface runoff from the southern portion of the plant.

1973

Pond C-1 was used to control and capture surface runoff.

Pond C-2 built to detain runoff water from the Southern Interceptor Ditch.

1980

Pond C-2 received surface runoff. Pond C-1 isolated from Rocky Flats surface runoff.

1989

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FIGURE 5-3
TIME LINE OF EVENTS FOR THE WOMAN CREEK DRAINAGE

RKY294T5/5-3; REV: 02-22-94

5.1.2 Reservoirs

Three possibly impacted reservoirs are in the immediate vicinity of Rocky Flats. They are depicted in Figure 5-4 and include Great Western Reservoir, Standley Lake, and Mower Reservoir. Each of these reservoirs is briefly described in this section.

5.1.2.1 Great Western Reservoir

Great Western Reservoir is located approximately 0.25 mile east of the Rocky Flats Plant's eastern buffer zone boundary (Indiana Street). The reservoir receives runoff from Clear Creek through Church Ditch, Coal Creek through McKay Ditch, Upper Church Ditch, and Walnut Creek. Originally, the reservoir was 42 feet deep and had a storage capacity of 1,420 acre-feet. In 1955, the Turnpike Land Company bought the reservoir and established the Broomfield Heights Mutual Service Association to own and operate water and sewer utilities for the Broomfield Heights development. In 1958, the reservoir was enlarged to its present storage capacity of 3,250 acre-feet (City of Broomfield, 1991). In 1962, the city of Broomfield bought the water and sewer services from the Turnpike Land Company.

Great Western Reservoir water was used for agricultural irrigation until 1955. Since 1955, the sole water use has been as the city of Broomfield's municipal water supply.

5.1.2.2 Standley Lake

Standley Lake is a large reservoir located approximately one mile from the southeast corner of the Rocky Flats Plant's eastern buffer zone boundary (Indiana Street). It is owned by the Farmers Reservoir and Irrigation Company (FRICO). From 1914 to 1966, water from Standley Lake was used only for irrigation. The city of Westminster first used the lake as a drinking water source in 1966. Presently, the city of Westminster owns 37.3 percent of the shares in the Standley Lake Division, and the cities of Thornton and Northglenn own 13.3 and 17.7 percent of the shares, respectively. The remaining shares (31.7 percent) are still owned by FRICO.

5.1.2.3 Mower Reservoir

Mower Reservoir is a small, privately owned impoundment located just southeast of the Rocky Flats Plant (USDOE, 1991c). The reservoir is fed by Woman Creek via Mower Ditch, an irrigation ditch that originates within the Rocky Flats boundary (USDOE, 1991c). Mower Reservoir is used for stock watering, domestic lawn watering, and irrigation of approximately 80 acres (State of Colorado, 1973).

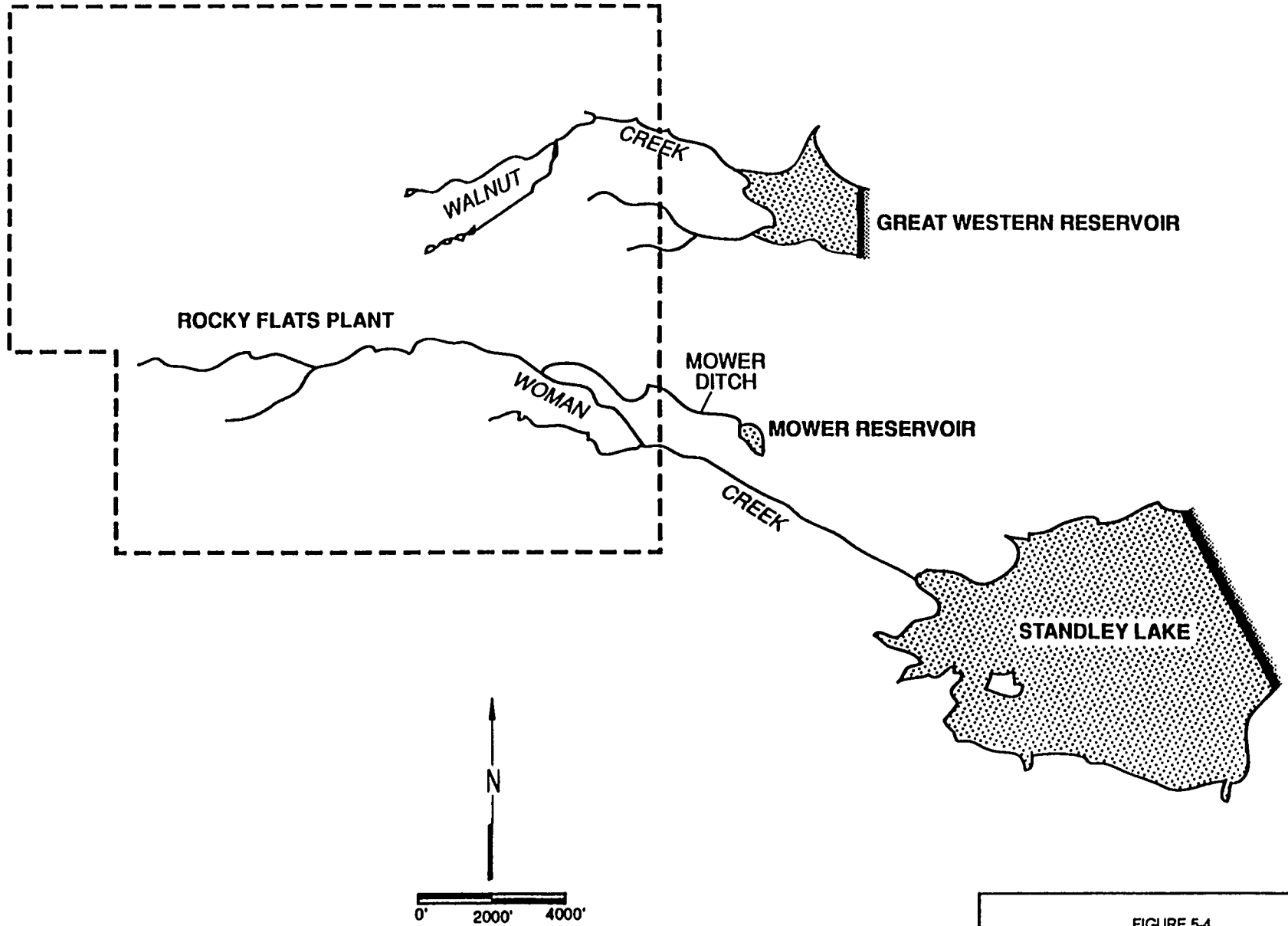

FIGURE 5-4
POSSIBLY IMPACTED RESERVOIRS
NEAR ROCKY FLATS

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RKY294T5/5-4; REV:02-22-94

5.2 Available Monitoring Data

The first step in identifying the most effective approach to estimating surface-water-related contaminant releases from the plant was to determine the availability of contaminant monitoring data for the terminal ponds on North and South Walnut Creek and for the three reservoirs that received plant effluents or runoff. The following sections provide a history of the contaminant monitoring activities associated with the terminal ponds and the reservoirs.

5.2.1 Radionuclide Data

As discussed in Section 2, the plant retained an extensive record of airborne effluent radionuclide data in a relatively consistent format that was generally easily understood and reproducible. However, the plant did not maintain a similar data set for surface water effluents. Unlike the health physics group, the waste disposal group did not have an organized system for records retention. Many of the waste disposal group's monthly reports are available; however, raw data supporting these reports were not located.

Two different types of surface water sampling data were located for use in evaluating plant releases. The first type of data is effluent information. In general, this information was obtained from the waste disposal coordination monthly progress reports (Dow, 1953-1973) and the DOE Off-Site Discharge Information System (ODIS). The information provided in the waste disposal reports is quite comprehensive for effluents discharged to South Walnut Creek from 1953 to 1970. The ODIS also provides information on effluents discharged to South Walnut Creek from 1971-1989. However, information regarding effluents (primarily surface runoff) discharged to North Walnut Creek and Woman Creek was not routinely reported.

The second type of data is reservoir and drinking water monitoring data. Rocky Flats has monitored both surrounding reservoirs and drinking water from various cities from 1952 to the present. Additionally, the Colorado Department of Health (CDH) monitored reservoirs and drinking water from July 1970 to the present. The completeness and usefulness of each of these data sources are described in the following sections.

5.2.1.1 Effluent Information

Data on effluents from the ponds to off-site streams are available in Monthly Waste Disposal Coordination Reports (1952 to 1970), Annual Environmental Monitoring Reports (1971 to 1988), Monthly Environmental Monitoring Reports (December 1986 to July 1990), and the ODIS (1971-1989).

The Monthly Waste Disposal Coordination Reports (1953-1971) contain the following information:

- point of release (e.g., building),
- receiving surface water stream (e.g., South Walnut Creek),
- volume released (in gallons),
- gross alpha radioactivity concentration (Ci L^{-1}), and
- total activity released (Ci).

The data in these reports are not complete in that they do not cover the entire period for all ponds. For example, all of the above information is available for both Pond A-1 and Pond B-3 from July 1953 through September 1955. However, from October 1955 to August 1971, the above information is only available for Pond B-1.

The Annual Environmental Monitoring Reports for 1971 to 1988 quantify contaminant concentrations in the final holding pond in each drainage system. However, no data are available on the volume of releases from the ponds.

The Monthly Environmental Monitoring Reports quantify volumes of waste released from holding ponds A-4, B-5, C-1, and C-2 for December 1986 through July 1990. Release volumes per day and per month are given.

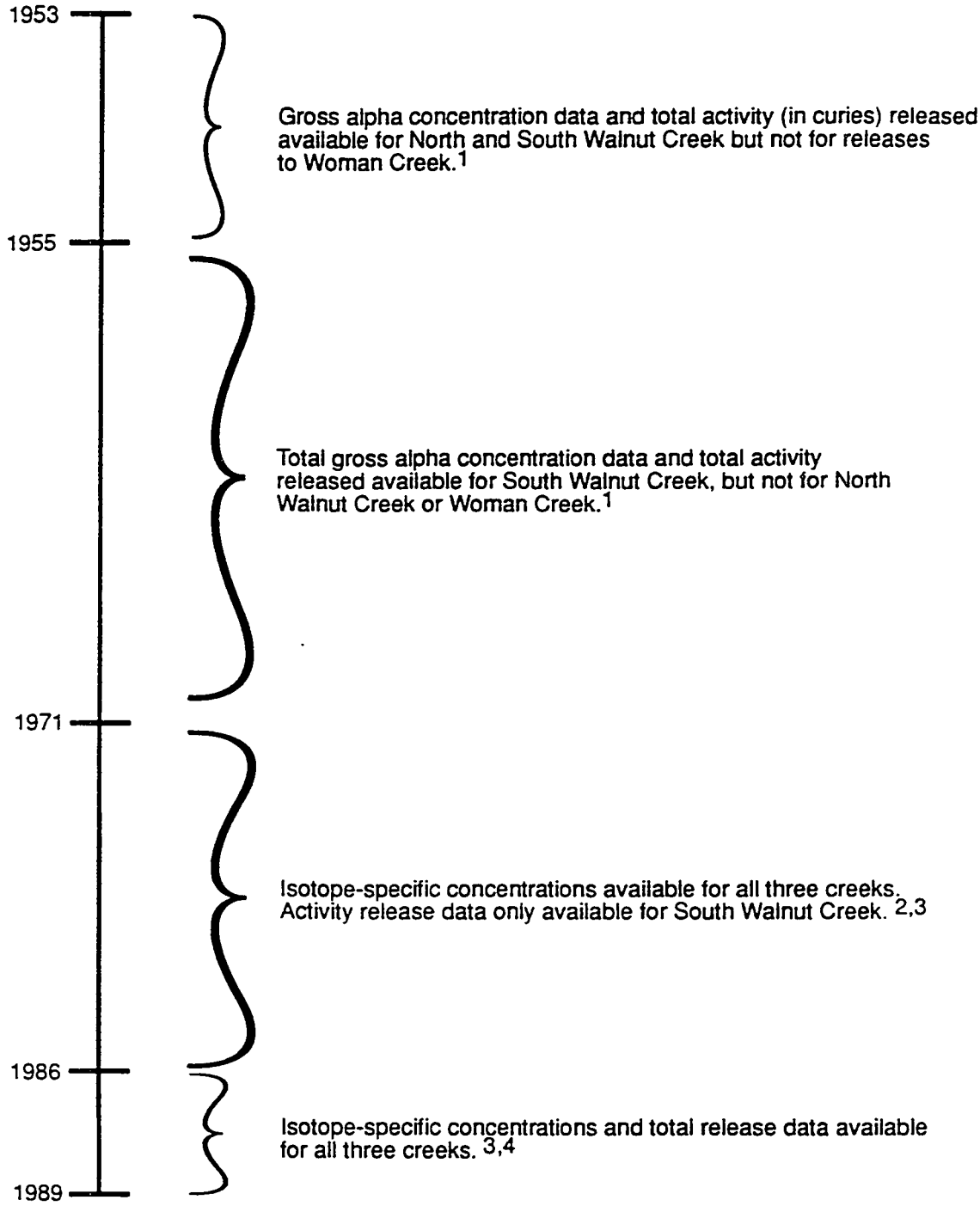
ODIS contains data for 1971-1989 that indicate the total curies of americium, plutonium, uranium, and tritium released to South Walnut Creek from the B-series ponds.

Figure 5-5 summarizes the availability of data on waterborne releases monitored by Rocky Flats.

The absence of data for Woman Creek and North Walnut Creek during the majority of the plant's existence limits our ability to develop site-wide contaminant release estimates. However, the available records can be used to evaluate whether contaminants released to Walnut Creek produced a measurable effect on the radioactivity levels in Great Western Reservoir. This evaluation is discussed in greater detail in subsequent sections of this report.

5.2.1.2 Reservoir and Drinking Water Monitoring

As previously mentioned, there are two sets of reservoir and drinking water monitoring data available. The first data set was collected by the Rocky Flats Site Survey group from 1952 to the present. The second was collected by CDH from July 1970 to the present. For the purposes of this analysis, data generated by the Site Survey group have been examined for the 1952 to



Source:

[1] Monthly Waste Disposal Coordination Reports, 1953-1971

[2] Annual Environmental Monitoring Reports, 1971-1988

[3] On-Site Discharge Information System, 1971 - 1989

[4] Monthly Environmental Monitoring Reports, 1986-1990

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FIGURE 5-5
WATERBORNE EFFLUENT
RELEASE INFORMATION

RKY294T5/5-5; REV: 02-22-94

1970 time period and the data generated by CDH have been examined for the period after 1970. The following sections provide an overview of each data source.

Site Survey Reservoir and Drinking Water Monitoring, 1952-1970

In 1952, prior to the start-up of operations at Rocky Flats, a survey of environmental background gross alpha concentrations was conducted by a group from the Hanford Reservation in Washington (Quimby, 1952). Part of the survey included sampling the reservoirs surrounding the site and sampling the drinking water of nearby cities. Additionally, once operations began at Rocky Flats, the Site Survey group at Rocky Flats continued to sample reservoirs and city drinking water near the facility at intervals ranging from annually to monthly. All samples throughout the 18-year time period were analyzed for gross alpha activity. Over the years, the radioactivity in over 20 lakes and reservoirs remote from the plant was also monitored. However, few were monitored on a consistent basis. ChemRisk contacted USGS, USEPA, CDH and the Northern Colorado Water Conservancy District in an unsuccessful attempt to locate a body of water that is more distant from Rocky Flats and has a gross alpha monitoring record that could serve as a source of "background" measurements for comparison with data collected from surface waters near the plant. Information gathered from the above-mentioned sources indicate that specific radionuclide data were rarely collected prior to 1971. For instance, no monitoring data for plutonium were located for specific water bodies suggested as possible background locations (Carter Lake and Yrandby Lake), and uranium monitoring data are only available from 1990 to the present.

Figure 5-6 depicts monitoring data availability for Great Western Reservoir, Standley Lake, Baseline Reservoir, and Mower Reservoir. Baseline Reservoir is located six miles north of the plant and did not receive waterborne effluents from the plant. For the purposes of our analysis of potential direct releases of contaminants to surface water, the radioactivity levels measured in Baseline Reservoir may be considered unaffected by Rocky Flats or "background" measurements.

Drinking water samples included water from the cities of Arvada, Boulder, Broomfield, Denver, Golden, Lafayette, Louisville, Thornton, and Westminster. Figure 5-7 depicts the availability of drinking water monitoring data for each of these cities. It is important to note that Great Western Reservoir was not a drinking water source for the city of Broomfield until 1955. Standley Lake was not a drinking water source for the city of Westminster until 1965.

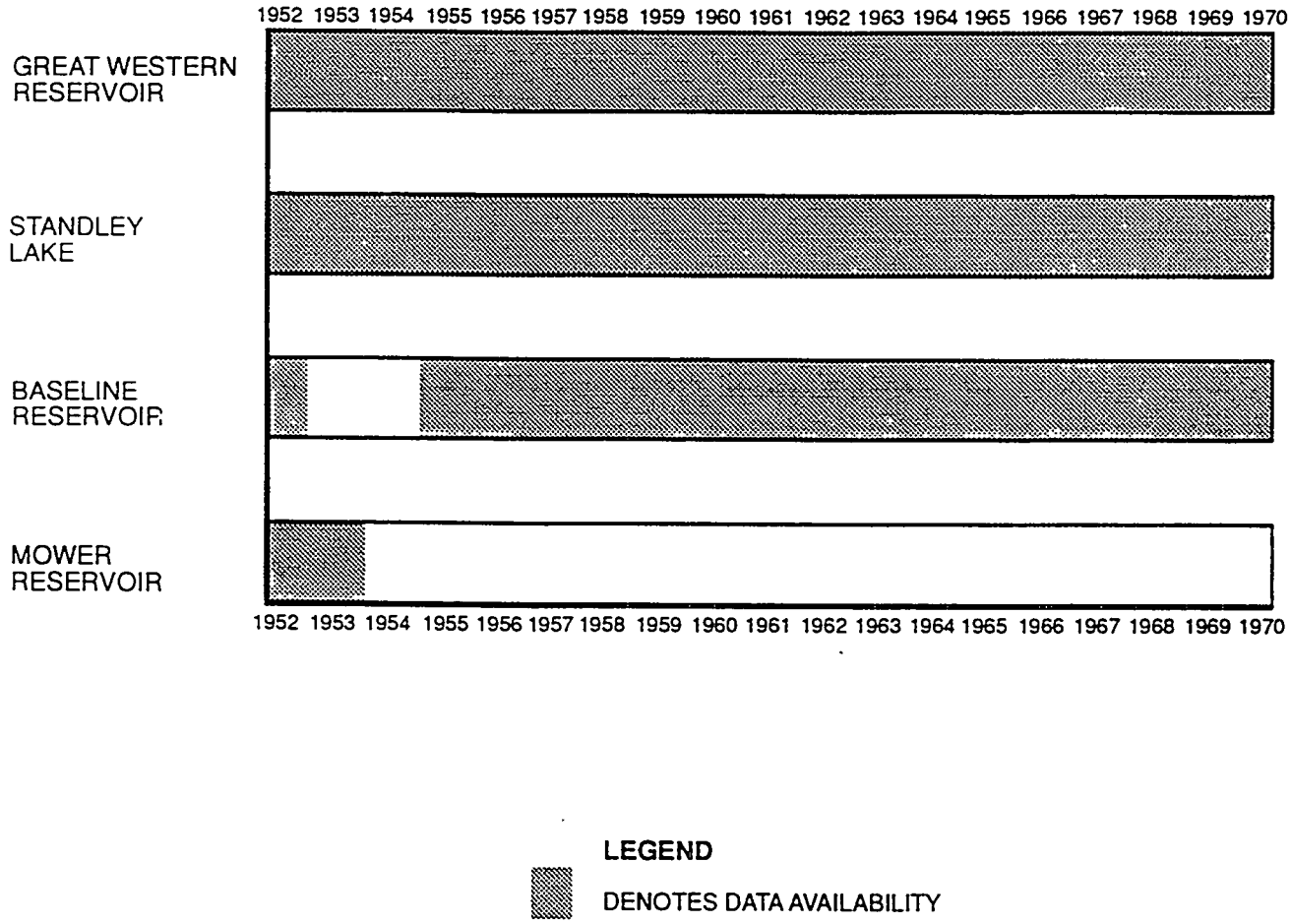

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FIGURE 5-6
AVAILABILITY OF GROSS ALPHA RESERVOIR MONITORING DATA COLLECTED BY THE SITE SURVEY GROUP AT ROCKY FLATS, 1952-1970

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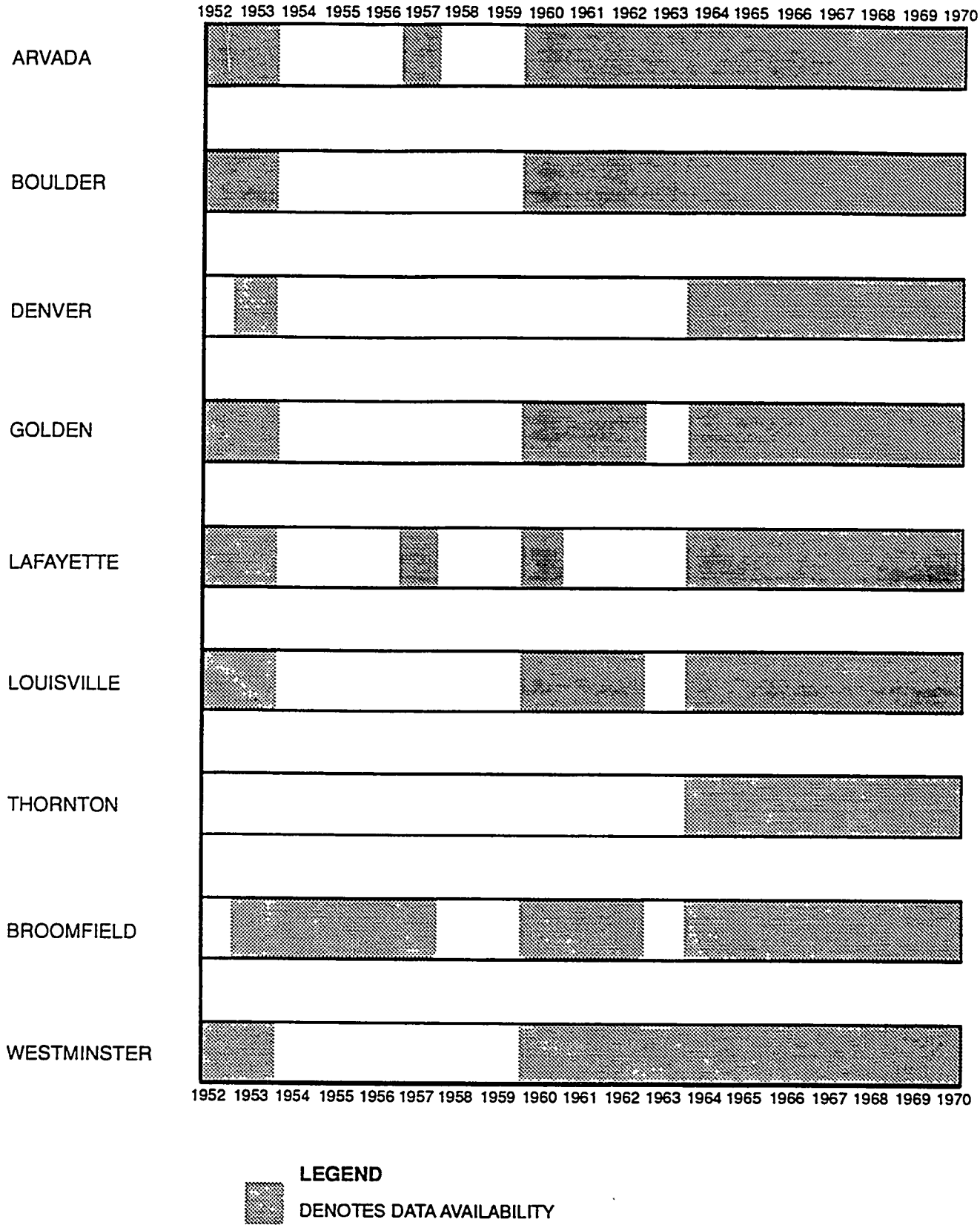

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FIGURE 5-7
AVAILABILTIY OF DRINKING WATER
MONITORING DATA COLLECTED BY
THE SITE SURVEY GROUP AT
ROCKY FLATS, 1952-1970

RKY294T5/5-7; REV: 02-22-94

Colorado Department of Health Reservoir and Drinking Water Monitoring, 1970-1989

In July 1970, CDH began a sampling program monitoring the reservoirs and drinking water for cities surrounding the Rocky Flats plant. The cities included in this analysis are Arvada, Boulder, Broomfield, Golden, and Westminster. The samples were screened for gross alpha and gross beta concentrations and then analyzed for Pu-238, Pu-239/240, natural uranium, and tritium. The drinking water from the cities of Broomfield and Westminster was sampled on a bimonthly basis and the drinking water from Arvada, Boulder, and Golden was sampled either quarterly or biannually. Since July 1970, these data have been reported in monthly Rocky Flats Plant Surveillance reports (CDH, 1970-1991).

The cities of Broomfield (after 1955) and Westminster (after 1965) received a majority of their drinking water from Great Western Reservoir and Standley Lake, respectively, both of which have possibly been impacted by effluents or runoff from Rocky Flats. Until the early 1980s, the city of Arvada received a majority of its water from Ralston Reservoir. The city of Boulder receives the majority of its water from the Arapahoe Glacier in the Rockies. The city of Golden receives all of its water from Clear Creek. The water supplies for the cities of Arvada, Boulder, and Golden do not receive runoff or effluent from Rocky Flats and therefore represent unaffected waters with "background" radioactivity levels in drinking water (Terry, 1992).

In addition to the drinking water samples that were collected by CDH, water samples were occasionally collected directly from Great Western Reservoir, Mower Reservoir, and Standley Lake. As with the drinking water samples, these samples were screened for gross alpha and beta contamination, and then analyzed for Pu-238, Pu-239/240, natural uranium, and tritium.

Some shortcomings of this data set are: 1) the lack of plutonium and uranium drinking water samples from background reservoirs for 1976 and 1977; 2) the lack of plutonium samples for the city of Westminster drinking water in 1977; and 3) the absence of sampling at Mower Reservoir until 1980. These shortcomings notwithstanding, the quantity and isotope-specific nature of the data, in addition to the fact that the data were collected by an organization independent of Rocky Flats, make this data set particularly useful in performing an independent evaluation of potential plant impacts relative to surface water.

Summary: A variety of data sources are available that quantify radionuclides in surface waters. However, many of the data sets are incomplete and are not sufficient for estimating site-wide radionuclide releases. However, the data can be used to identify any major release events and to determine whether there were greater levels of radioactivity in the reservoirs receiving water from the plant than in other reservoirs in the region.

5.2.2 Monitoring of Chemical Contaminants in Surface Waters

Throughout the first 30 years of operations at Rocky Flats, data were not collected on the waterborne effluents for any of the nonradioactive chemicals of concern. Measurements of chemical parameters which are not addressed in this study are available for the 1971-1989 time period. These parameters were measured in effluent at USEPA-NPDES discharge permit points. These data, which include pH, nitrate, phosphate, fluoride, total solids, and hexavalent chromium, are presented in the Annual Environmental Monitoring Reports.

As discussed previously, the volatile chemicals of concern readily evaporate from surface waters and were therefore unlikely to have been transported off-site in surface waters to any significant extent. Beryllium is the only nonradioactive contaminant of concern having the potential to be transported off-site in surface waters. Although the 1980 Final Environmental Impact Statement states that beryllium was among eleven elements analyzed for in plant waterborne effluents, only the 1971 and 1972 annual environmental reports report waterborne beryllium effluent data. However, beryllium is a metal that has low solubility in water and will tend to be adsorbed by surface soils or sediment. This tendency would likely have reduced the extent to which beryllium was transported off-site in surface waters. The only information available for addressing past releases from the plant would be measurements of beryllium in reservoir sediments. To date, only one study to determine beryllium concentrations in the sediments of Great Western Reservoir and Standley Lake has been located (USEPA, 1975). This study did not investigate the beryllium concentrations in background reservoirs and did not draw any conclusions as to whether or not the beryllium concentrations in Great Western Reservoir and Standley Lake were elevated. The analytical results of the study are summarized below:

- Beryllium concentrations in sediment dredge samples from Great Western Reservoir sediments ranged from <0.5 - 1.5 parts per million (ppm), with a mean of 1.0 ppm.
- Beryllium concentrations in core samples from Great Western Reservoir ranged from <0.5 - 1.9 ppm, with a mean of 1.0 ppm.
- Beryllium concentrations in sediment dredge samples from Standley Lake ranged from 0.7 to 1.4 ppm, with a mean of 1.1 ppm.
- Beryllium concentrations in core samples from Standley Lake were all reported as less than 2.0 ppm.

In an attempt to determine whether or not the beryllium concentrations in Great Western Reservoir sediment could be considered elevated, the Colorado State University Soils Testing Laboratory at Fort Collins and the Geologic Division of USGS were contacted (Self, 1993; Modreski, 1993). Neither organization was able to provide regional beryllium concentration in soil or sediment information. However, beryllium soil concentrations in Colorado, as reported in a scientific literature, range from not detectable to 5.0 ppm, with a mean of 1.0 ppm and a standard deviation of 1.2 (Dragun and Chiasson, 1991).

Summary: No data were identified that could be used to quantify historical releases of nonradioactive materials to area surface waters. Of the stable materials under study, only beryllium would be likely to remain in surface waters where it would most likely be associated with sediments. Beryllium concentrations measured in the sediments of Great Western Reservoir and Standley Lake by USEPA in 1975 are in the same range as those measured in soil samples from other regions of Colorado.

5.3 Surface Water Data Evaluation Approach

The monitoring of contaminants in plant effluents to off-site waterways has been relatively limited and is inadequate for the purposes of developing reasonably accurate estimates of historical contaminant releases from the plant. Our observations regarding the availability of data for source term development are consistent with a number of statements made in the plaintiff's sentencing memorandum in the criminal prosecution brought by the U.S. Department of Justice against Rockwell International Corporation (USA, 1992). This memorandum indicates that there were periodic releases of various industrial and hazardous wastes through the sewage treatment plant during the 1980s, and that the improper operation of the spray irrigation fields during this same period contributed to off-site dispersal of contaminants. In addition, the memorandum goes on to note that:

> "[T]here was no regular monitoring of other industrial or toxic pollutants either entering or leaving the STP [sewage treatment plant], being sent to the holding ponds or being spray irrigated" (USA v. Rockwell International Corporation, 1992, p. 72).

The focus of the surface water investigations was therefore shifted to the monitoring data for city water supplies that may have been impacted by plant releases. It should be noted, however, that the use of reservoir and drinking water data for the purposes of assessing plant impacts is complicated by the potential presence of contaminants resulting from other sources or from natural background levels of contaminants.

Based on the sources and availability of reservoir monitoring data described previously, the evaluation of surface water contaminants is addressed for two distinct time periods. The first time period, 1952-1970, represents the period for which some limited documentation of effluents and plant-generated monitoring data for reservoirs and drinking water in the form of gross alpha measurements are available. These data were used in the following manner to evaluate whether, on an annual basis, the reservoirs or drinking water were measurably affected by radioactive releases from the plant:

- Direct comparisons were made of pond effluent monitoring data (annual average gross alpha releases) from Walnut Creek to measured gross alpha concentration in Great Western Reservoir to establish whether plant releases were associated with measurable increases in radioactivity in receiving reservoirs.

- Potential theoretical increases in gross alpha concentrations in Great Western Reservoir were estimated based on estimates of annual gross alpha activity releases to Walnut Creek.

- Direct comparison were made of gross alpha activity in possibly impacted drinking water from Broomfield and Westminster to gross alpha activity in "background" drinking water from Arvada, Boulder, and Golden.

- Direct comparison were made of gross alpha activity in Great Western Reservoir (1955-1970) and Standley Lake (1965-1970) to gross alpha activity in a "background" reservoir, Baseline Reservoir.

The second time period, 1971-1989, represents the period in which the plant adopted a "zero release" policy for process waste water. As a result, only a very limited data set on plant effluents is available. However, a rather complete set of isotope-specific reservoir and drinking water monitoring data is available from sampling conducted by the Colorado Department of Health. The Colorado Department of Health data were used to make direct comparisons between isotope-specific concentrations in possibly impacted drinking water (i.e., drinking water from Broomfield and Westminster) and nonimpacted drinking water (i.e., drinking water from Arvada, Boulder, and Golden).

5.4 Surface Water Monitoring Data Evaluation for 1952-1989

The following sections provide the results of comparisons of surface water monitoring data for the period 1952-1989.

5.4.1 Effluent Data Evaluation

Reported annual releases of gross alpha radioactivity to Walnut Creek for 1952-1970 are summarized in Table 5-2. Over this 18-year period, the plant reported the release of 54,400 μCi of gross alpha activity. This release estimate is primarily derived from monitored releases from Pond B-3 into South Walnut Creek; any contaminant release associated with surface runoff is not accounted for in this estimate. Another point that should be noted is that the water the plant receives for use contains natural radioactivity; therefore, a portion of the radioactivity in water released from plant holding ponds is not derived from plant operations. In order to estimate the impact of the release of gross alpha to Walnut Creek on the water quality of the Great Western Reservoir between 1953 and 1970, it is also necessary to know the time history of reservoir volume. As no data on the volume of the Great Western Reservoir prior to 1971 were located, annual rainfall at Rocky Flats was investigated as a possible surrogate of reservoir volume. Associated annual rainfall data are shown in Table 5-3. For the purpose of identifying if there is a correlation between plant releases and gross alpha levels detected in the reservoir water, annual gross alpha activity releases, and annual gross alpha activity releases per unit annual rainfall are plotted with annual average gross alpha concentrations detected in Great Western Reservoir in Figure 5-8. It should be noted that the enlargement of Great Western Reservoir in 1958 might have affected gross alpha concentrations in reservoir water.

A qualitative review of these data suggests that there is no clear relationship between plant releases and gross alpha activity in Great Western Reservoir between the years of 1959-1962, when an approximately twofold increase in the amount of gross alpha activity released from the plant was not associated with a similar increase in measured gross alpha activity in Great Western Reservoir. On the other hand, the data from 1966 to 1970 suggest that plant releases may have influenced the observed activity in Great Western Reservoir, at least during 1966. However, other events, such as the abortive reentry of the plutonium-238-powered SNAP 9-A navigational satellite power supply in the upper atmosphere in 1964 (Eisenbud, 1987), cannot be ruled out as potential sources of the increased gross alpha concentrations in 1966.

A calculation to establish whether there was any correlation among the data sets indicated that the reported annual plant releases of gross alpha activity and the reported annual plant releases of gross alpha activity per annual rainfall are not correlated with radioactivity in Great Western Reservoir (correlation coefficients of -0.04 and 0.21, respectively).

To further evaluate the potential for plant releases of radioactivity to increase observed activity in Great Western Reservoir, some simple dilution calculations were performed. These calculations were used to estimate the possible increase in radioactivity in the reservoir that would result once a plant release was diluted by the water in the reservoir. This simple analysis assumed that plant releases were diluted by a volume of water equal to 75 percent of the total

TABLE 5-2

REPORTED ANNUAL GROSS ALPHA RADIOACTIVITY RELEASES
TO WALNUT CREEK, 1952-1970

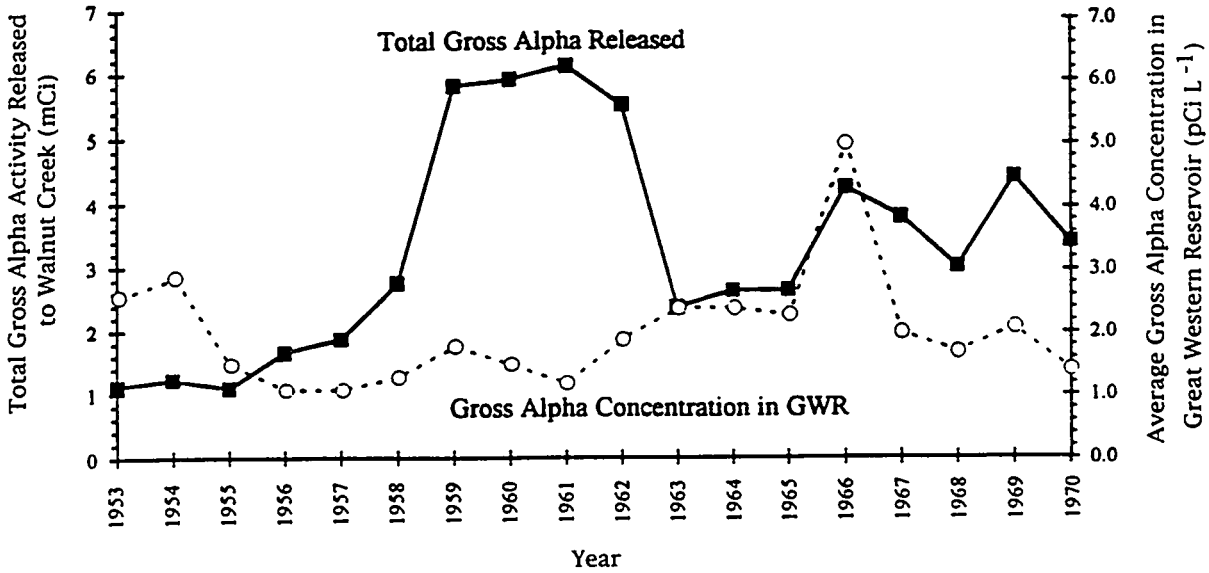
Year	Total (μCi)
1952	No release reported
1953	1,126
1954	1,226
1955	1,099
1956	1,653
1957	1,863
1958	2,736
1959	5,800
1960	5,900
1961	6,110
1962	5,500
1963	2,360
1964	2,620
1965	2,630
1966	4,227
1967	3,765
1968	2,982
1969	4,384
1970	3,369

Source: Boss, M. 1973. Release History Folder. ChemRisk Repository Number RE-357.

TABLE 5-3

ANNUAL RAINFALL AT THE ROCKY FLATS PLANT
(inches)

Year	Rainfall	Year	Rainfall
1953	11.26	1965	18.87
1954	7.76	1966	10.24
1955	14.77	1967	22.54
1956	13.42	1968	12.71
1957	22.67	1969	24.67
1958	18.07	1970	18.56
1959	19.65	1971	14.30
1960	13.72	1972	14.78
1961	16.08	1973	21.55
1962	8.26	1974	13.73
1963	12.23	1975	12.22
1964	8.79	1976	13.51



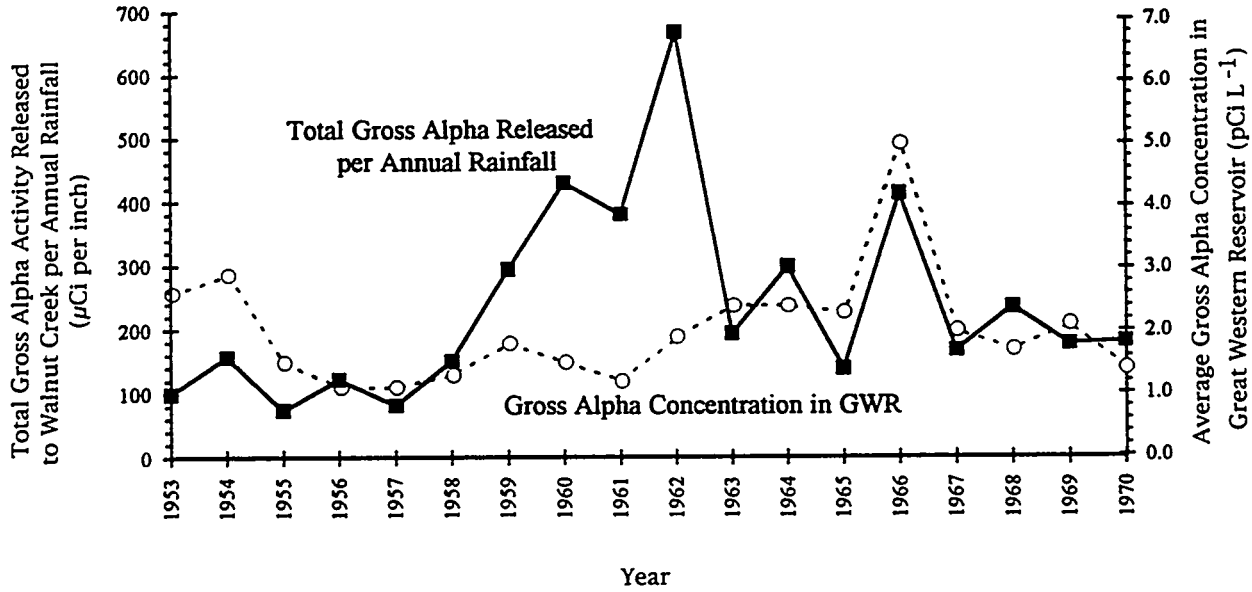


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FIGURE 5-8
TOTAL GROSS ALPHA ACTIVITY RELEASED TO WALNUT CREEK AND ANNUAL AVERAGE GROSS ALPHA CONCENTRATION IN GREAT WESTERN RESERVOIR (1953-1970)

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capacity of Great Western Reservoir. This calculation is believed to be conservative in that it assumes that an annual release quantity is diluted by less than a single reservoir volume and that there was no inflow or outflow during the year that would further dilute the radioactivity. In addition, the analysis assumes that all of the contamination released by the plant reaches Great Western Reservoir, which is more than a mile downstream. The annual total increase in the gross alpha concentration in Great Western Reservoir that would be expected to result from the release of radioactivity to Walnut Creek based on these simplifying assumptions can be calculated as follows:

$$\text{Increase in alpha activity at Great Western Reservoir} = \frac{\text{total alpha activity released (pCi)}}{(0.75) \times (\text{total capacity of Great Western Reservoir, L})}$$

For 1953:

$$\text{Increase in alpha activity at Great Western Reservoir} = \frac{1.13 \times 10^9 \text{ pCi}}{0.75 \times (1.75 \times 10^9 \text{ L})} = 0.86 \text{ pCi L}^{-1}$$

Table 5-4 presents the results of this calculation for each year for which effluent data are available. The estimated increases of gross alpha activity in Great Western Reservoir due to the reported annual effluent releases from the plant range from 0.86 to 2.0 pCi L^{-1}. This should be considered a very conservative estimate of the possible influence of plant releases on Great Western Reservoir.

Examination of the two data sets discussed in this section does not provide any clear evidence that plant releases of contaminants in surface water resulted in measurable increases in radioactivity in Great Western Reservoir. A simple dilution calculation suggests that reported plant releases conservatively had the potential to elevate activity in Great Western Reservoir on average by little more than 1 pCi L^{-1} during the 1960s.

5.4.2 Reservoir Water Monitoring Data Evaluation

Another evaluation was performed to further examine whether radioactivity levels or their fluctuations in Great Western Reservoir or Standley Lake differed from those measured in other reservoirs in the region, in this case Baseline Reservoir. Associated data are shown in Figure 5-9 and Table 5-5. A qualitative review of these data suggests that the gross alpha concentrations and their fluctuations in these three reservoirs appear similar during this time period, with the exception of 1966. As noted previously, an increase in the release of radioactivity from the plant during 1966 appeared to be possibly associated with a corresponding increase in the gross alpha concentration in Great Western Reservoir. The cause of the increase

TABLE 5-4

THEORETICAL INCREASE IN GROSS ALPHA CONCENTRATIONS IN GREAT WESTERN RESERVOIR USING ROCKY FLATS REPORTED RELEASE TOTALS, 1953-1970

Year	Theoretical Increase in Gross Alpha Concentration in Great Western Reservoir at 75% Capacity* (pCi L^{-1})
1953	0.86
1954	0.93
1955	0.83
1956	1.26
1957	1.41
1958	0.93
1959	1.9
1960	2.0
1961	2.0
1962	1.8
1963	0.8
1964	0.87
1965	0.87
1966	1.4
1967	1.3
1968	1.0
1969	1.5
1970	1.1

* Providing no inflow or outflow occurs throughout the year.

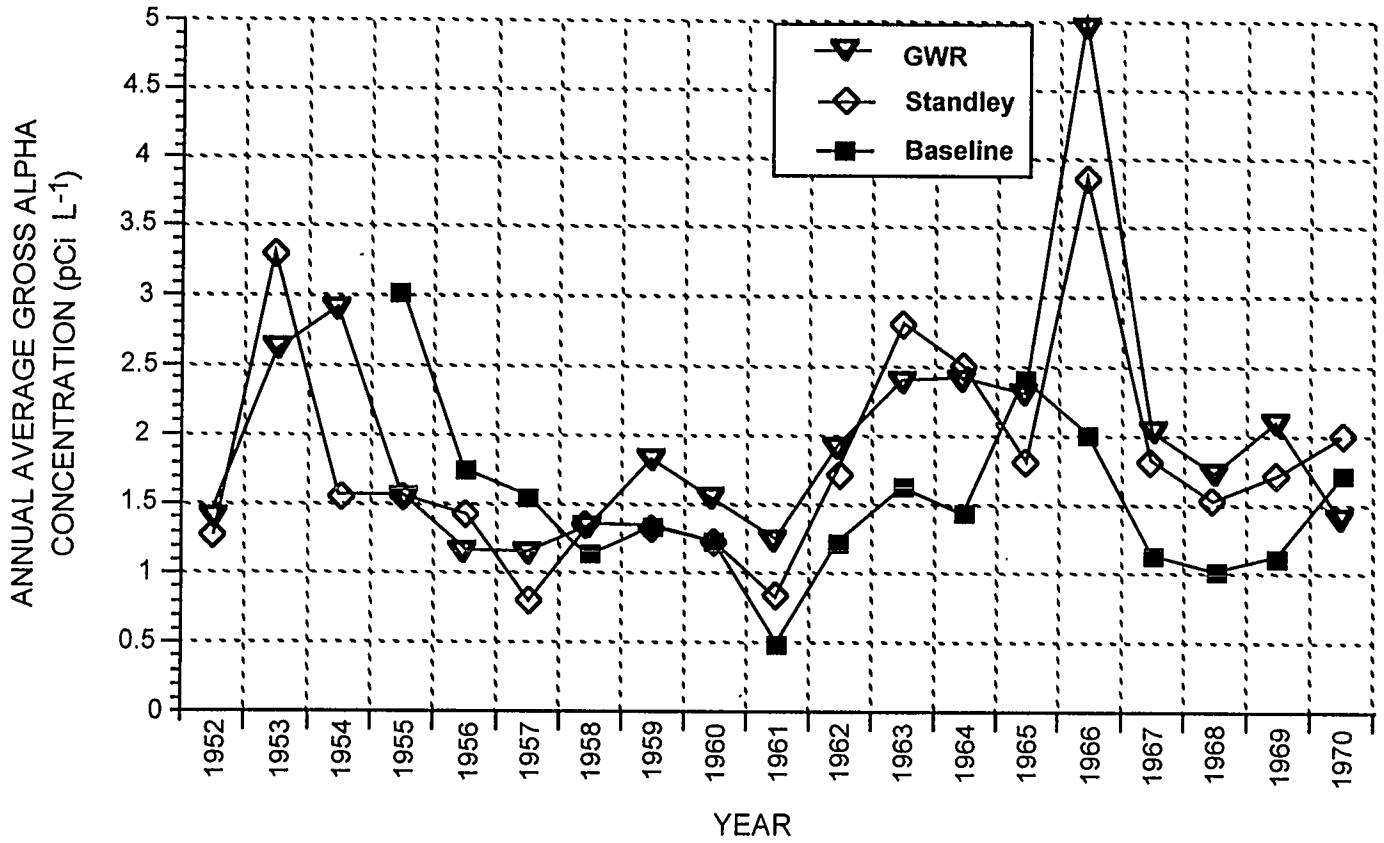

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FIGURE 5-9
ANNUAL AVERAGE GROSS ALPHA CONCENTRATION
FOR THE GREAT WESTERN RESERVOIR,
STANDLEY LAKE, AND BASELINE RESERVOIR
(1952-1970)

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TABLE 5-5

ANNUAL GROSS ALPHA CONCENTRATION IN BASELINE AND GREAT WESTERN RESERVOIRS AND STANDLEY LAKE

(pCi L^{-1})

Year	Baseline Reservoir	Great Western Reservoir Annual Average Gross Alpha Concentration	Standley Lake Annual Average Gross Alpha Concentration
1952	1.22	1.42	1.2
1953	not sampled	2.6	3.3
1954	not sampled	2.9	1.5
1955	3.0	1.5	1.5
1956	1.7	1.1	1.4
1957	1.5	1.1	0.76
1958	1.1	1.3	1.3
1959	1.3	1.8	1.3
1960	1.2	1.5	1.2
1961	0.44	1.2	0.8
1962	1.2	1.9	1.7
1963	1.6	2.4	2.8
1964	1.4	2.4	2.5
1965	2.4	2.3	1.8
1966	2.0	5.0	3.9
1967	1.1	2.0	1.8
1968	0.98	1.7	1.5
1969	1.1	2.1	1.7
1970	1.7	1.4	2.0
1952-1970 Average	1.5	2.0	1.8

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in gross alpha concentration in Standley Lake is not known. No data are available for Baseline Reservoir for 1953 and 1954.

While the concentration of alpha activity in Baseline Reservoir is about the same as Great Western Reservoir and Standley Lake for most years during this period, alpha activity was on average approximately 0.5 and 0.3 pCi L^{-1} greater in Great Western Reservoir and Standley Lake, respectively, than in Baseline. If the correlation between the data from each of these reservoirs is calculated for the years in which data are available, fluctuations in radioactivity in Baseline Reservoir are not highly correlated with either Great Western Reservoir (correlation coefficient = 0.26) or Standley Lake (correlation coefficient = 0.34). However, radioactivity levels in Standley Lake and Great Western Reservoir are closely correlated (correlation coefficient = 0.9).

It is also important to note that although minimal amounts of contaminants were likely released into the atmosphere or creeks from the Rocky Flats Plant in 1952, gross alpha concentrations measured in Great Western Reservoir and Standley Lake in that year are similar to those measured in the later years. The average of gross alpha measurements in Great Western Reservoir in 1952 was 1.4 pCi L^{-1}, about 31 percent lower than the average level measured between 1952 and 1970. Similarly, the average of gross alpha measurements in Standley Lake in 1952 was 1.2 pCi L^{-1}, about 33 percent lower than the average level measured between 1952 and 1970. These data indicate that a major portion of the gross alpha activities measured in Great Western Reservoir and Standley Lake between 1953 and 1970 may be attributed to natural background.

Mower Reservoir Monitoring

From 1952 to 1953, nine samples were collected and analyzed for gross alpha activity from this reservoir. These data are presented in Table 5-6. This data set is too limited to draw conclusions from regarding the long-term impacts plant effluents may have had on the reservoir.

5.4.3 Drinking Water Monitoring Data Evaluation

Comparisons were also made between the concentration of gross alpha activity in drinking water from the cities of Broomfield and Westminster and "background" concentrations in drinking water from Arvada, Boulder, and Golden. The annual average drinking water concentrations from Arvada, Boulder, and Golden are plotted in Figure 5-10. Gross alpha activity concentrations ranged from 0.1 pCi L^{-1} to 4.4 pCi L^{-1}. Similarly, the annual averages of the Broomfield and Westminster data are plotted in Figure 5-11. As noted in Figure 5-11, Broomfield did not begin receiving water from Great Western Reservoir until 1955 and Westminster did not begin receiving water from Standley Lake until 1966. Gross alpha activity

TABLE 5-6

1952-1953 MOWER RESERVOIR MONITORING DATA

Date	Gross Alpha Activity (pCi L^{-1})
5/26/52	1.7
7/17/52	1.0
9/15/52	3.2
11/10/52	1.8
4/6/53	9.0
5/5/53	8.8
5/21/53	9.5
6/24/53	1.2
7/27/53	6.0

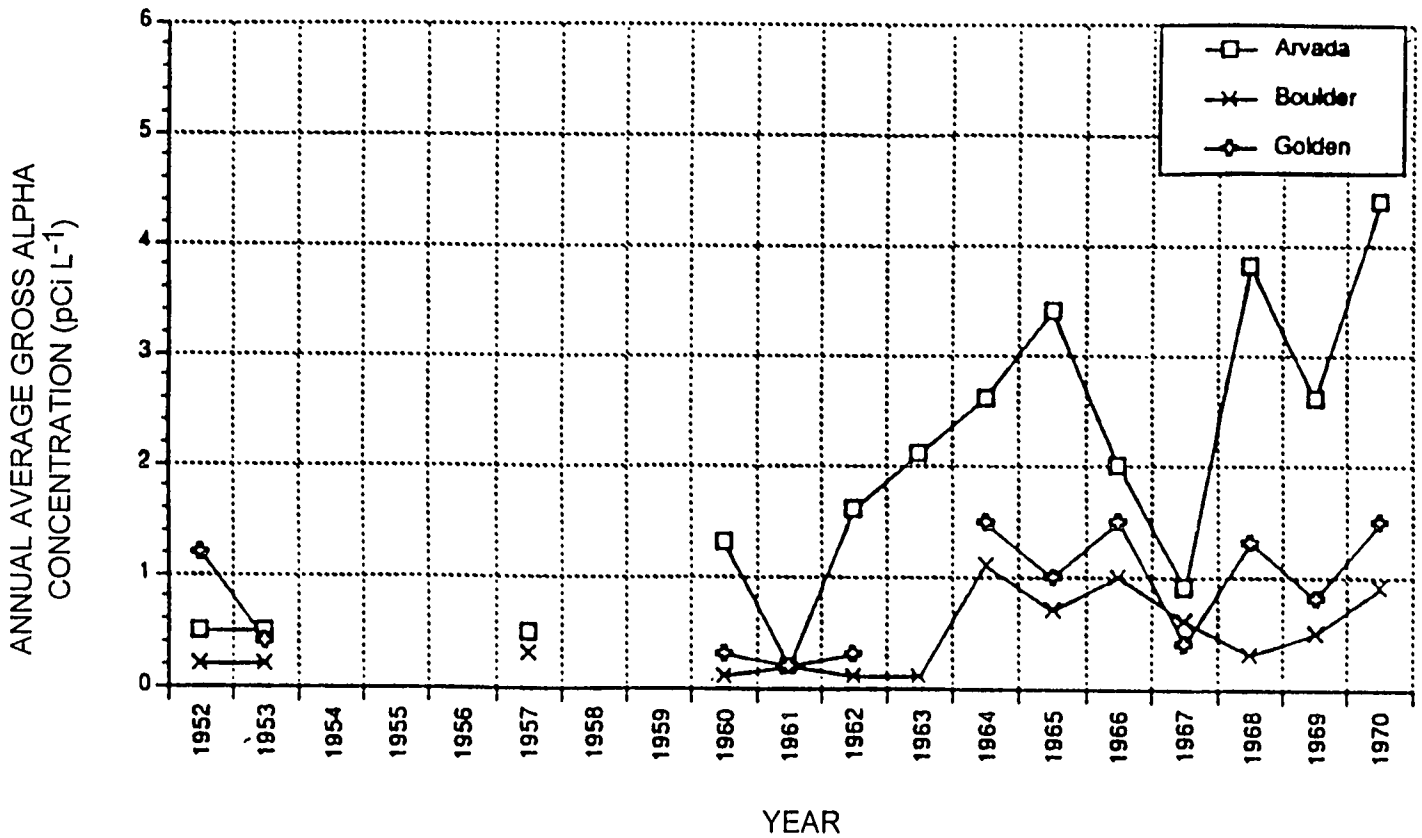

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FIGURE 5-10
ANNUAL AVERAGE GROSS ALPHA CONCENTRATION IN DRINKING WATER FOR THE CITIES OF ARVADA, BOULDER, AND GOLDEN (1952-1970)

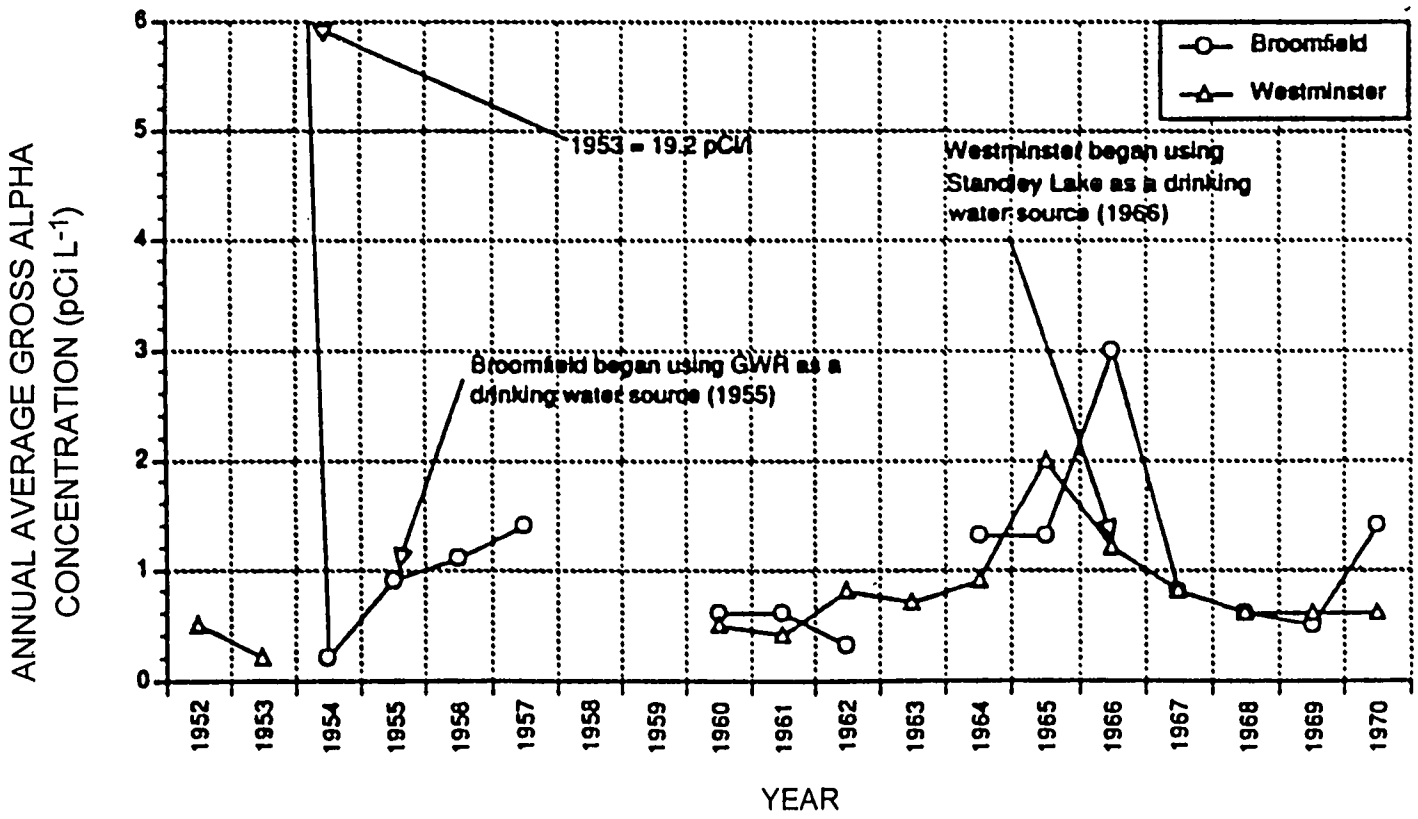

FIGURE 5-11
ANNUAL AVERAGE GROSS ALPHA CONCENTRATION IN DRINKING WATER FOR THE CITIES OF BROOMFIELD AND WESTMINSTER (1952-1970)

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concentrations in Broomfield and Westminster drinking waters ranged from 0.1 pCi L^{-1} to 19.2 pCi L^{-1}. It is important to note that the maximum of 19.2 pCi L^{-1} measured in Broomfield drinking water occurred prior to the use of Great Western Reservoir as a drinking water source. A qualitative evaluation of these data suggest that the gross alpha concentrations in drinking water from Broomfield and Westminster are similar to gross alpha concentrations in drinking water from Arvada, Boulder, and Golden.

As indicated in Table 5-7, the average radioactivity in the "background" cities' drinking water for the period from 1955 to 1970 was slightly higher than that of Broomfield or Westminster during the time in which these cities derived their water from Great Western Reservoir and Standley Lake. However, in 1966 gross alpha activity measured in drinking water of Broomfield was 1.5 pCi L^{-1} higher than that measured in the "background" cities. This single-year elevation of radioactivity may be attributable to the plant; but such fluctuation was not inconsistent with levels often found in "background" drinking water supplies.

5.5 Surface Water Monitoring Data Evaluation for 1970-1989

In the 1970-1989 time period, the water supplies for the cities of Broomfield and Westminster were primarily drawn from Great Western Reservoir and Standley Lake, respectively. As discussed previously, CDH primarily sampled finished drinking water for Broomfield and Westminster, rather than sampling Great Western Reservoir or Standley Lake during this period. CDH also collected samples of drinking water from Arvada, Boulder, and Golden, three cities that receive water from reservoirs at a considerable distance from Rocky Flats and that do not receive runoff or effluents from Rocky Flats (Terry, 1992). The sampling of the distant water supplies during this period provides a reference point for the comparison of the level of radioactivity found in waters possibly impacted by the plant. The CDH reports provide information on concentrations of plutonium, natural uranium, and tritium in the sampled waters.

In order to evaluate the large amount of data collected by Colorado Department of Health from 1970 to 1989, a database was created in which all plutonium, uranium, and tritium data for the cities of Broomfield, Westminster, Arvada, Boulder, and Golden were entered. It is important to note that, on average more than 70 percent of the samples for all water supplies were below limits of detection. A summary of the frequency of detection and the maximum values is presented in Tables I-3 through I-6 in Appendix I of this report.

The first step taken in examining these data sets was to calculate the annual average Pu-239/240, Pu-238, natural uranium, and tritium concentrations for each of the five cities. The annual average radionuclide concentrations in drinking water are tabulated in Tables I-7 through I-10 in Appendix I of this report.

TABLE 5-7

ANNUAL GROSS ALPHA CONCENTRATIONS FOR DRINKING WATER
(pCi L^{-1})

Year	"Background"[1]	Broomfield Drinking Water[2]	Westminster Drinking Water[3]
1955	Not Sampled	0.9	Not Applicable
1956	Not Sampled	1.1	Not Applicable
1957	0.4 (just Arvada & Boulder)	1.4	Not Applicable
1958	Not Sampled	Not Sampled	Not Applicable
1959	Not Sampled	Not Sampled	Not Applicable
1960	0.6	0.6	Not Applicable
1961	0.2	0.6	Not Applicable
1962	0.7	0.3	Not Applicable
1963	1.1 (just Arvada & Boulder)	Not Sampled	Not Applicable
1964	1.7	1.3	Not Applicable
1965	1.7	1.3	2.0
1966	1.5	3.0	1.2
1967	0.6	0.8	0.8
1968	1.8	0.6	0.6
1969	1.3	0.5	0.6
1970	2.3	1.4	0.6
1955-1970 Average	1.2	1.1	1.0

1 The background level is the average of the annual average gross alpha concentrations in the drinking water of Arvada, Boulder, and Golden.
2 Broomfield drinking water derived at least in part from Great Western Reservoir.
3 Westminster drinking water derived at least in part from Standley Lake after 1965.

In order to examine whether releases or runoff from Rocky Flats measurably altered plutonium, uranium, or tritium concentrations in Broomfield or Westminster, concentrations of these contaminants in drinking water from Broomfield and Westminster were plotted and compared to data plots for the cities of Arvada, Boulder, and Golden (Figures 5-12 through 5-19). A qualitative review of these data suggests:

- From 1972 to 1974, Pu-239/240 concentrations in the drinking water of Broomfield and Westminster may have risen as a result of pond reconstruction, which resuspended sediments in the North and South Walnut Creek drainage.

- The accidental release of tritium in 1973 and 1974 clearly elevated the tritium concentrations in Broomfield drinking water.

Tables 5-8 through 5-11 present the average "background" reservoir data and Westminster and Broomfield average radionuclide concentrations in drinking water for the period 1970-1989. As indicated earlier, a large number of samples were below the detection limit. In calculating the annual average values, one-half the detection limit was used whenever non-detect results were reported. When using one-half the limit of detection for non-detect results, calculated annual averages often fell below the limit of detection. It is important to note that because there are so many non-detect results, it is not likely that the adjusted data are normally (or even lognormally) distributed; therefore, the average does not necessarily provide an accurate representation of the data. However, the average likely overestimates the actual contaminant concentration in drinking water. An alternative approach for evaluating the data to that described here is presented in Appendix I. While there are no known direct statistical tests that can be used when so many non-detects are involved, the generalized likelihood-ratio test was used to give an approximate solution to this problem in the case of large sample sizes. The application of this test to the data is discussed in Appendix I. The test results did not suggest that the concentrations of radionuclides in drinking water from Broomfield and Westminster differed from that found in drinking water from Arvada, Boulder, or Golden.

As indicated in Tables 5-8 and 5-9, the Pu-238 and Pu-239/240 concentrations in the "background" cities' drinking water averaged for the entire period from 1970-1989 were slightly higher than the plutonium concentrations in the drinking water of Broomfield and Westminster over the same period. The annual data indicate that the two-year period of pond reconstruction (1972-1974) along the North and South Walnut Creek drainage systems may have elevated levels of plutonium in Great Western Reservoir. The data suggest that from 1972 to 1974, the pond reconstruction may have elevated the plutonium levels in the Broomfield drinking water as much as 0.16 pCi L^{-1}. However, this elevated value is not inconsistent with the year-to-year fluctuations of plutonium in the drinking water from the "background" cities.

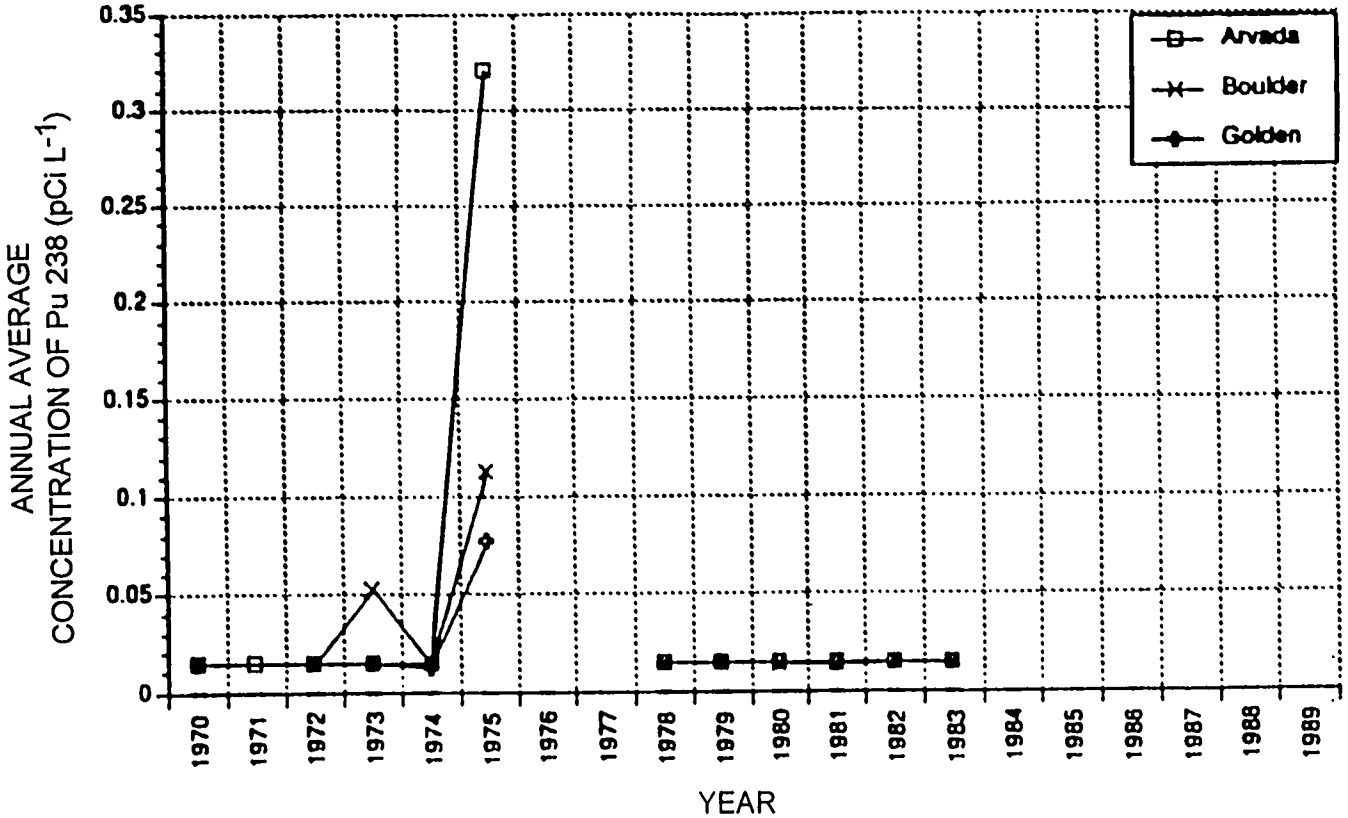

FIGURE 5-12
ANNUAL AVERAGE CONCENTRATION OF Pu 238 IN DRINKING WATER FOR THE BACKGROUND CITIES OF ARVADA, BOULDER, AND GOLDEN (1970-1983)

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RKY294T5/5-12; REV: 02-22-94

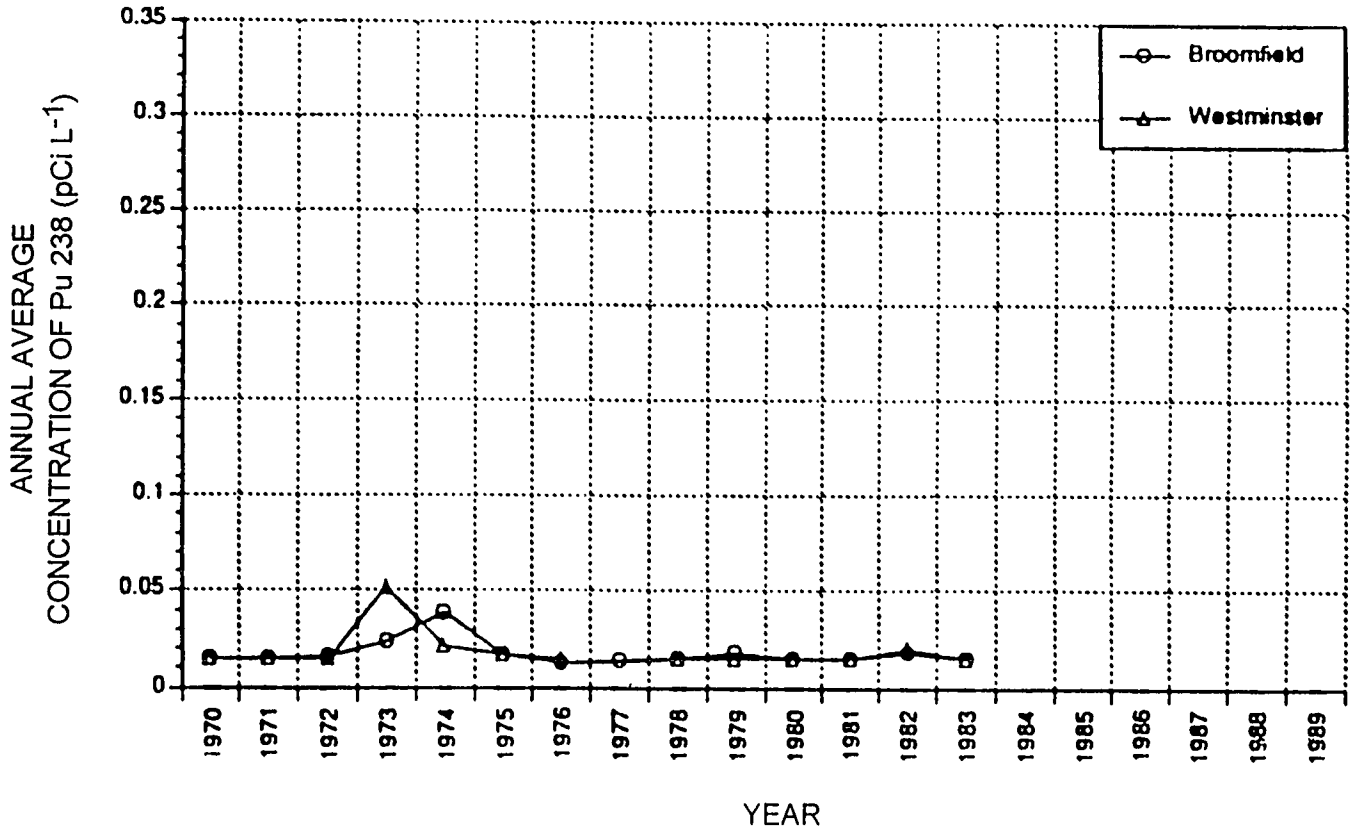

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FIGURE 5-13
ANNUAL AVERAGE CONCENTRATION OF Pu 238
IN DRINKING WATER FOR THE CITIES OF
BROOMFIELD AND WESTMINSTER
(1970-1983)

RKY294T5/5-13, REV 2-14-94

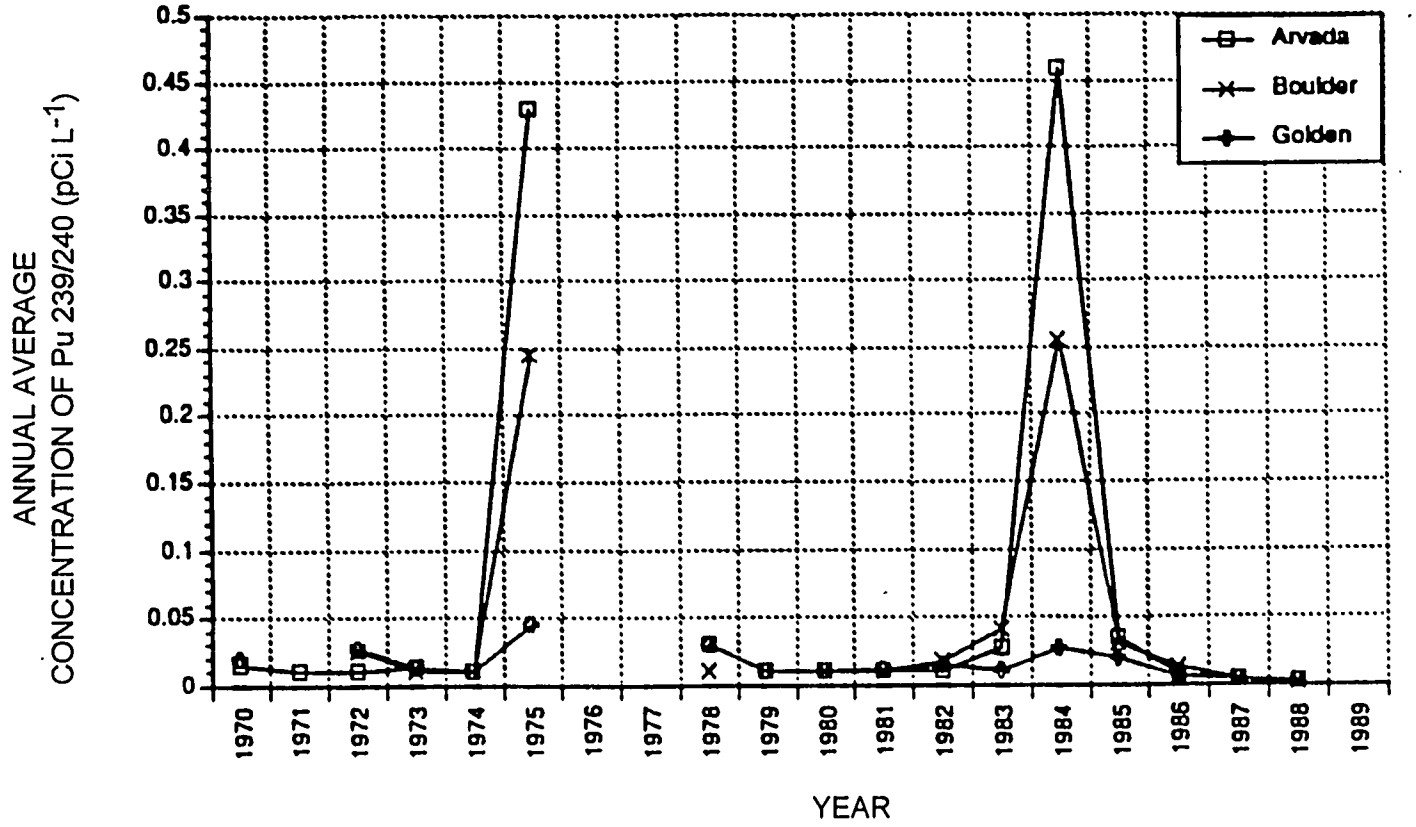

FIGURE 5-14
ANNUAL AVERAGE CONCENTRATION OF Pu 239/240 IN DRINKING WATER FOR THE BACKGROUND CITIES OF ARVADA, BOULDER, AND GOLDEN (1970-1988)

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RKY294T5/5-14, REV 2-14-94

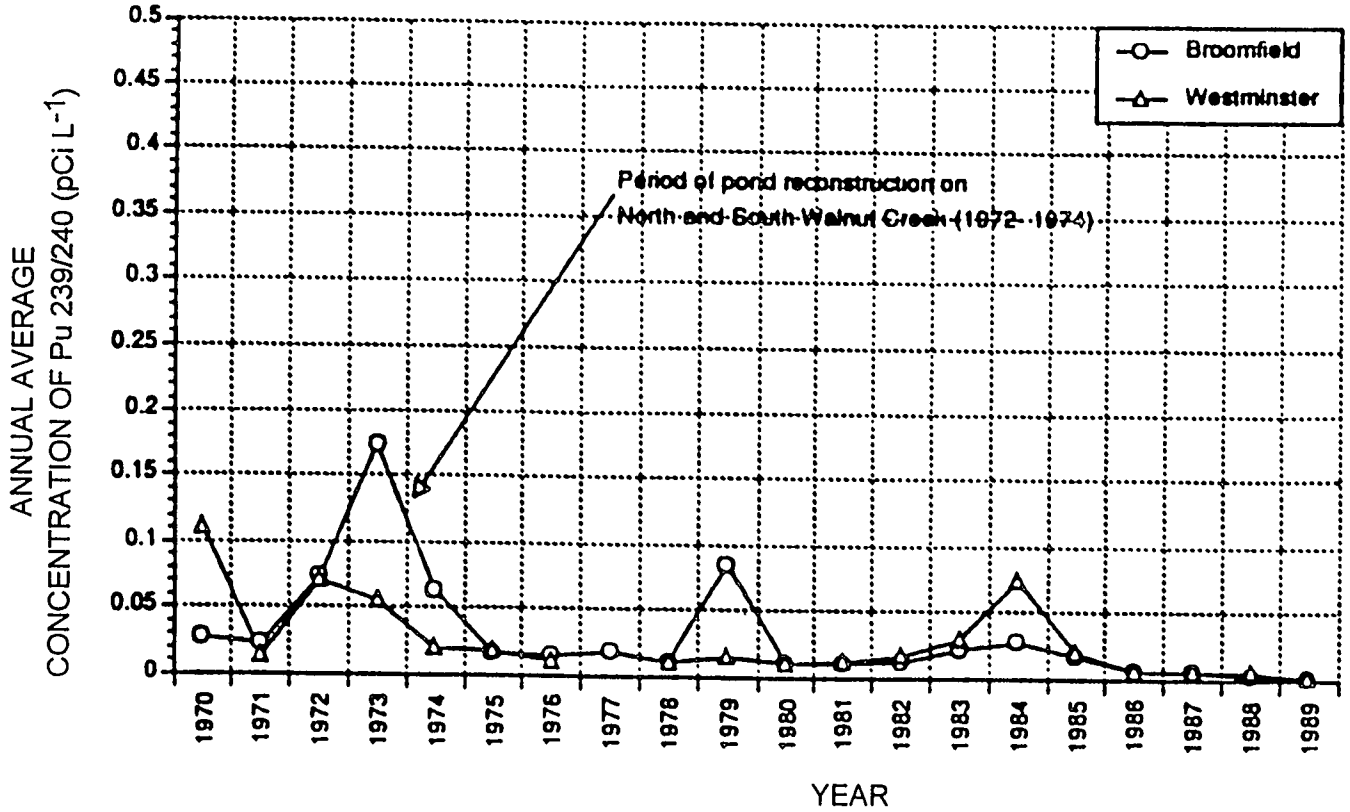

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FIGURE 5-15
ANNUAL AVERAGE CONCENTRATION OF Pu 239/240
IN DRINKING WATER FOR THE CITIES
OF BROOMFIELD AND WESTMINSTER
(1970-1989)

RKY294T5/5-15

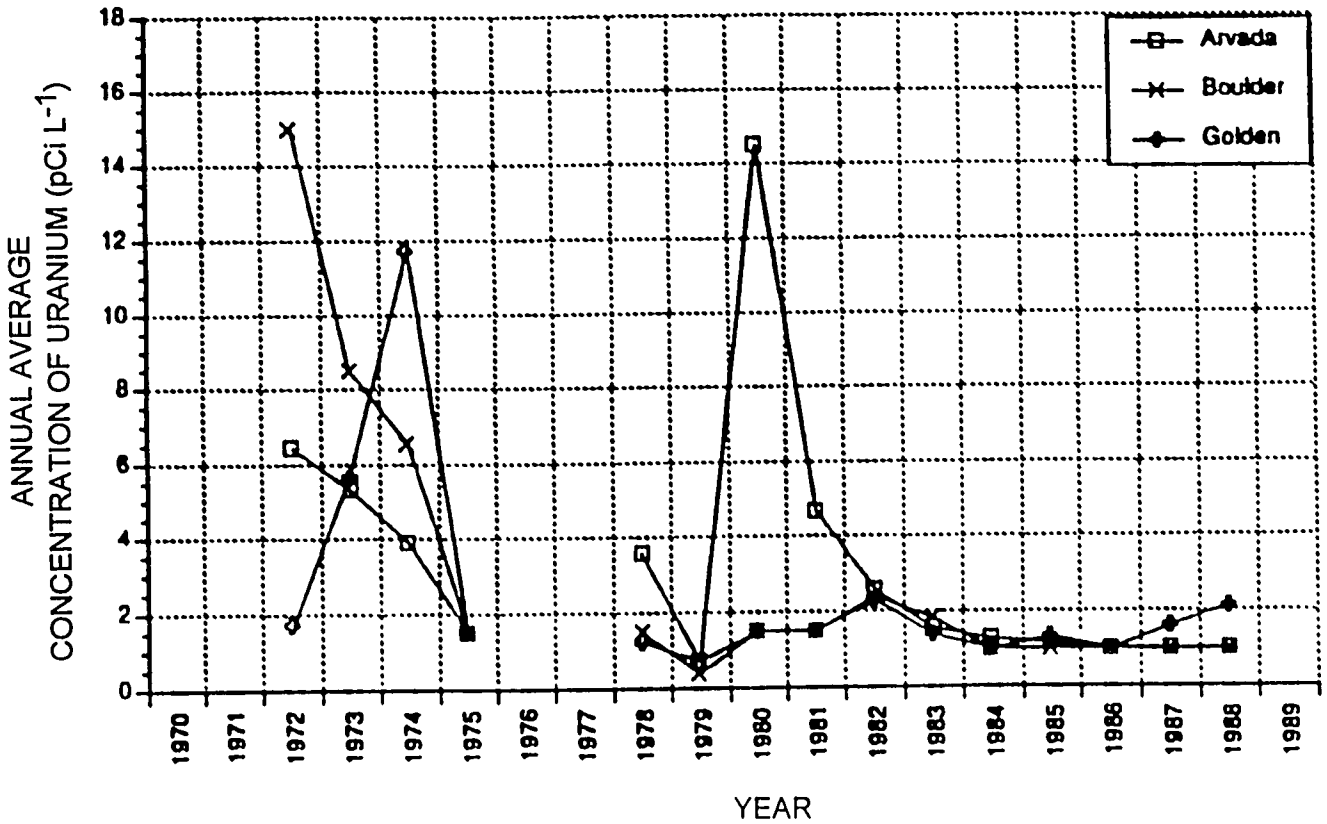

FIGURE 5-16
ANNUAL AVERAGE CONCENTRATION OF URANIUM IN DRINKING WATER FOR THE BACKGROUND CITIES OF ARVADA, BOULDER, AND GOLDEN (1972-1988)

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RKY294T5/5-16, REV 2-14-94

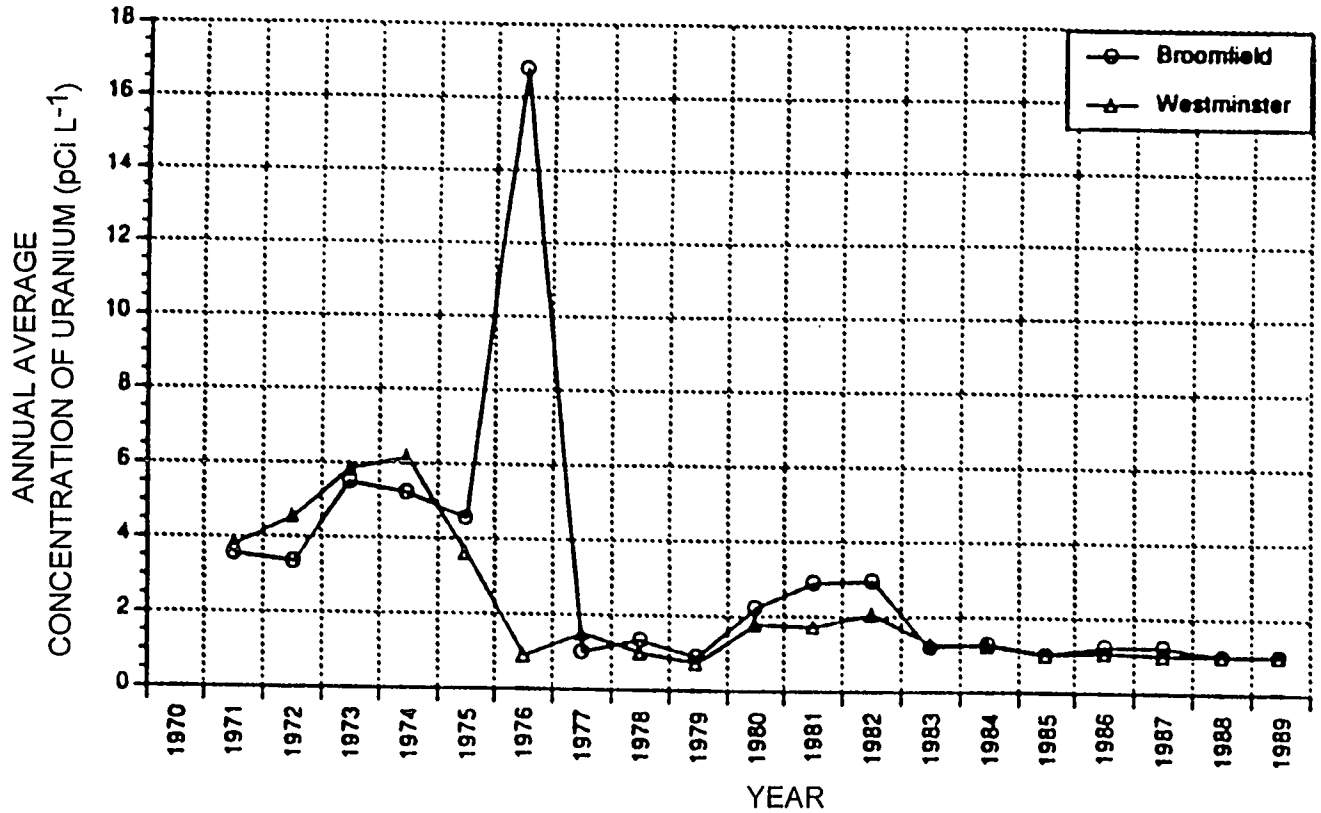

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FIGURE 5-17
ANNUAL AVERAGE CONCENTRATION OF URANIUM
IN DRINKING WATER FOR THE CITIES OF
BROOMFIELD AND WESTMINSTER
(1971-1989)

RKY294T5/5-17; REV: 02-22-94

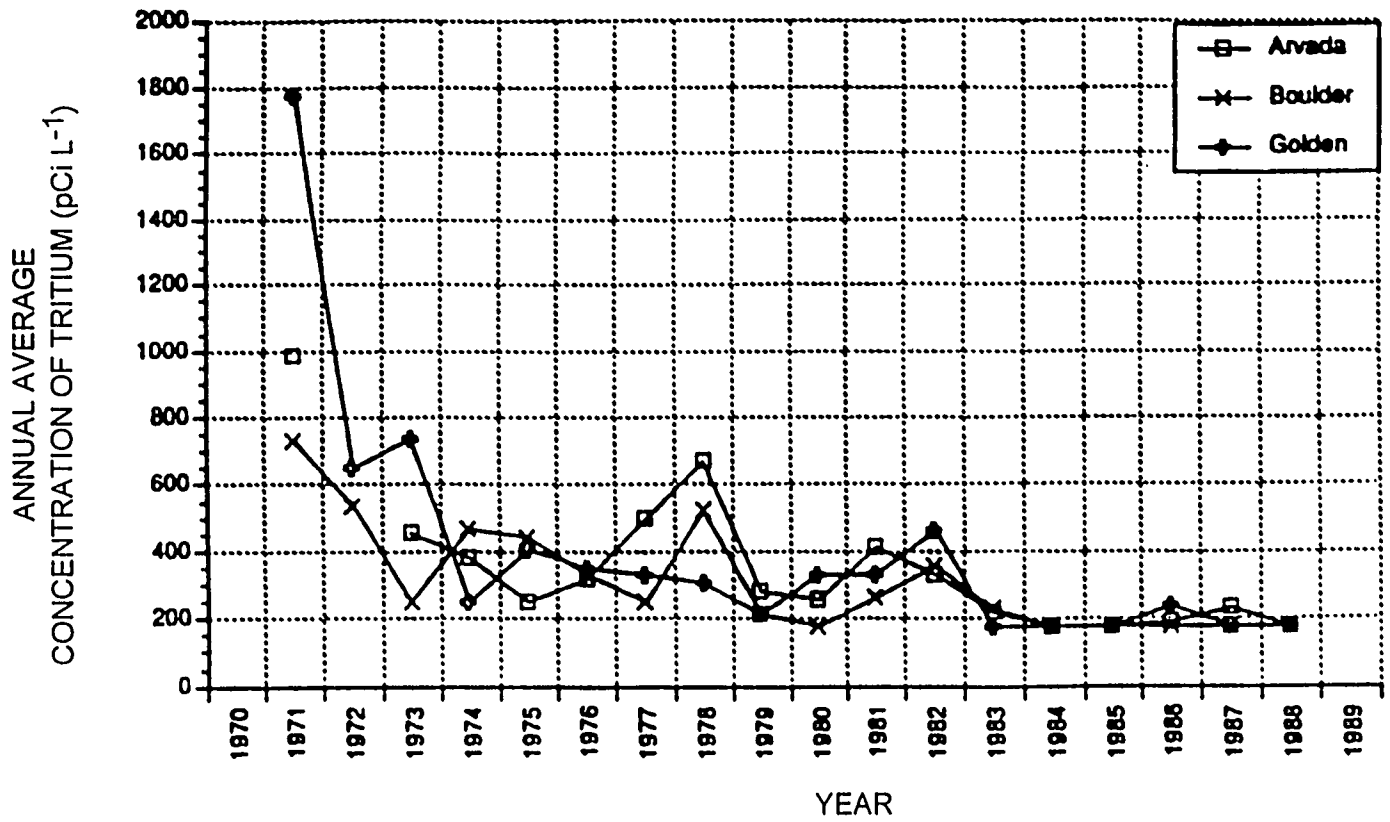

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FIGURE 5-18
ANNUAL AVERAGE CONCENTRATION OF TRITIUM
IN DRINKING WATER FOR THE BACKGROUND CITIES
OF ARVADA, BOULDER, AND GOLDEN
(1971-1988)

RKY294T5/5-18

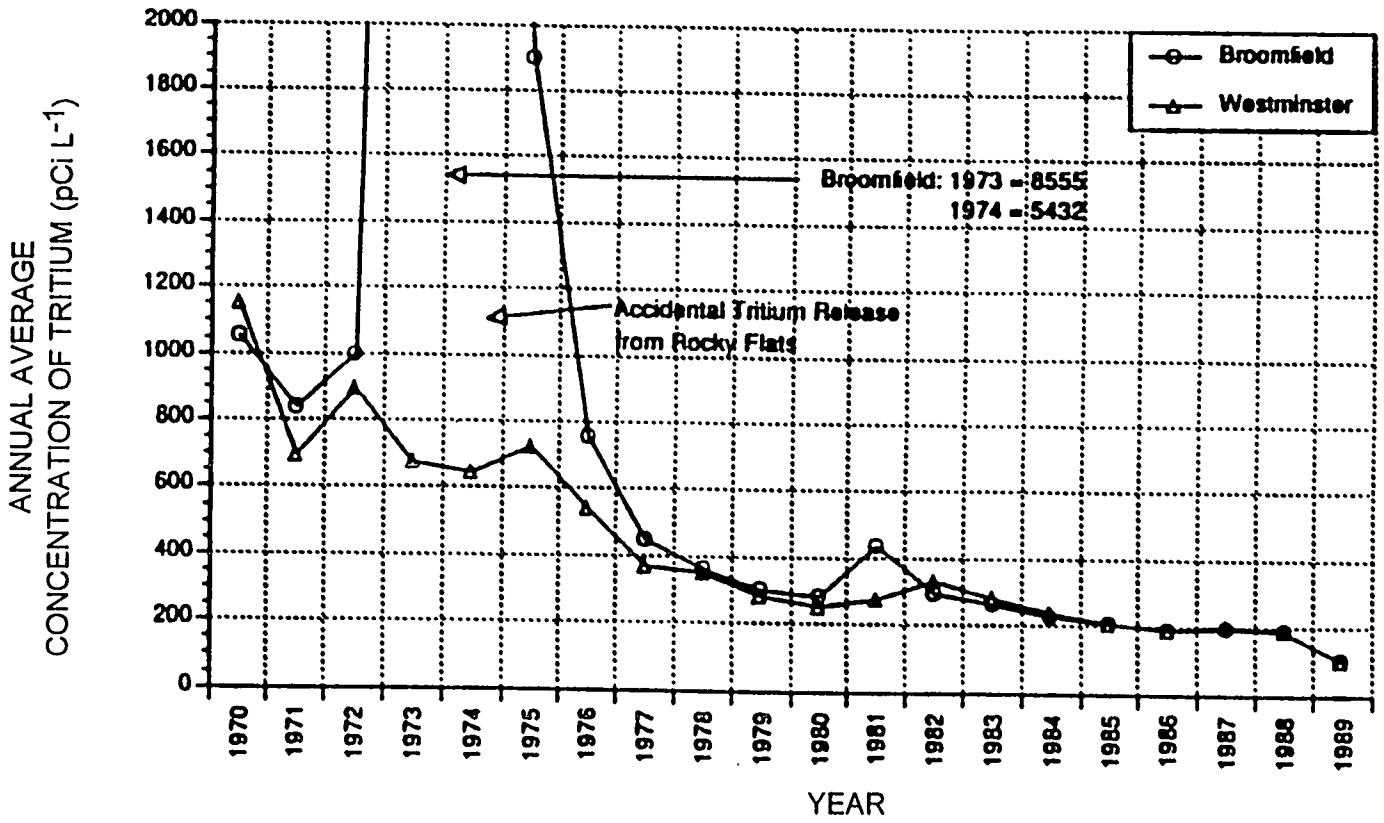

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FIGURE 5-19
ANNUAL AVERAGE CONCENTRATION OF TRITIUM
IN DRINKING WATER FOR THE CITIES OF
BROOMFIELD AND WESTMINSTER
(1970-1989)

RKY294T5/5-19

TABLE 5-8

ANNUAL AVERAGE DRINKING WATER CONCENTRATION OF PU-238

(pCi L^{-1})

Year	"Background"[1,2]	Westminster[2] Value	Broomfield[2]
1970	0.015	0.015	0.015
1971	0.015	0.015	0.015
1972	0.015	0.015	0.016
1973	0.028	0.051	0.02
1974	0.014	0.021	0.039
1975	0.17	0.017	0.02
1976	no samples	0.015	0.013
1977	no samples	no samples	0.014
1978	0.015	0.015	0.015
1979	0.015	0.015	0.018
1980	0.015	0.015	0.015
1981	0.014	0.015	0.015
1982	0.015	0.02	0.02
1983	0.015	0.015	0.015
1984-1989	Did not analyze for Pu-238		

1970-1983 Average	0.03	0.02	0.02

1 Background concentrations were determined by averaging the Pu-238 annual average for drinking water from the cities of Arvada, Boulder, and Golden.

2 Annual averages were calculated by summing all samples and dividing by the number of samples. When a sample result was below the limit of detection, the contaminant was considered to be present at one-half the limit of detection.

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TABLE 5-9

ANNUAL AVERAGE DRINKING WATER CONCENTRATION OF PU-239/240

(pCi L^{-1})

Year	"Background"[1,2]	Westminster[2]	Broomfield[2]
1970	0.018	0.11	0.027
1971	0.010	0.01	0.022
1972	0.021	0.07	0.073
1973	0.021	0.055	0.174
1974	0.012	0.02	0.063
1975	0.24	0.02	0.02
1976	no samples	0.01	0.02
1977	no samples	no samples	0.02
1978	0.023	0.01	0.01
1979	0.010	0.02	0.084
1980	0.010	0.01	0.01
1981	0.010	0.01	0.01
1982	0.014	0.017	0.01
1983	0.026	0.029	0.02
1984	0.028	0.07	0.03
1985	0.21	0.02	0.02
1986	0.010	0.006	0.006
1987	0.04	0.006	0.006
1988	0.02	0.006	0.002
1989	no samples	0.002	0.004
1970-1989 Average	0.041	0.034	0.032

1 Background concentrations were determined by averaging the Pu-239/240 annual average for drinking water from the cities of Arvada, Boulder, and Golden.

2 Annual averages were calculated by summing all samples and dividing by the number of samples. When a sample result was below the limit of detection, the contaminant was considered to be present at one-half the limit of detection.

TABLE 5-10

ANNUAL AVERAGE DRINKING WATER CONCENTRATION
OF NATURAL URANIUM

(pCi L^{-1})

Year	"Background"[1,2]	Westminster[2]	Broomfield[2]
1970	no samples	no samples	no samples
1971	no samples	3.8	3.5
1972	7.7	4.5	3.3
1973	6.5	5.8	5.5
1974	7.4	6.2	5.2
1975	1.5	3.6	4.5
1976	no samples	0.85	16.7
1977	no samples	1.5	1
1978	2.1	0.99	1.3
1979	0.58	0.69	0.85
1980	5.8	1.8	2.2
1981	2.6	1.7	2.9
1982	2.4	2.0	2.9
1983	1.6	1.3	1.1
1984	1.1	1.2	1.2
1985	1.1	1	1
1986	1	1.1	1.2
1987	1.2	1	1.8
1988	1.4	1	1
1989	no samples	1	1
1970-1989 Average	2.9	2.2	3.1

1 Background concentrations were determined by averaging the natural uranium annual average for drinking water from the cities of Arvada, Boulder, and Golden.

2 Annual averages were calculated by summing all samples and dividing by the number of samples. When a sample result was below the limit of detection, the contaminant was considered to be present at one-half the limit of detection.

TABLE 5-11

ANNUAL AVERAGE DRINKING WATER CONCENTRATION OF TRITIUM
(pCi L^{-1})

Year	"Background"[1,2]	Westminster[2]	Broomfield[2]
1970	no samples	1200	1100
1971	1200	NA	840
1972	590	890	1000
1973	480	680	8600
1974	370	650	5400
1975	370	720	1900
1976	330	540	750
1977	360	370	450
1978	500	350	360
1979	240	280	300
1980	250	250	280
1981	340	280	440
1982	380	330	300
1983	210	280	270
1984	180	240	220
1985	180	210	210
1986	200	190	190
1987	200	200	190
1988	180	190	190
1989	no samples	100	100
1970-1989 Average	360	420	1200

1 Background concentrations were determined by averaging the tritium annual average for drinking water from the cities of Arvada, Boulder, and Golden.

2 Annual averages were calculated by summing all samples and dividing by the number of samples. When a sample result was below the limit of detection, the contaminant was considered to be present at one-half the limit of detection.

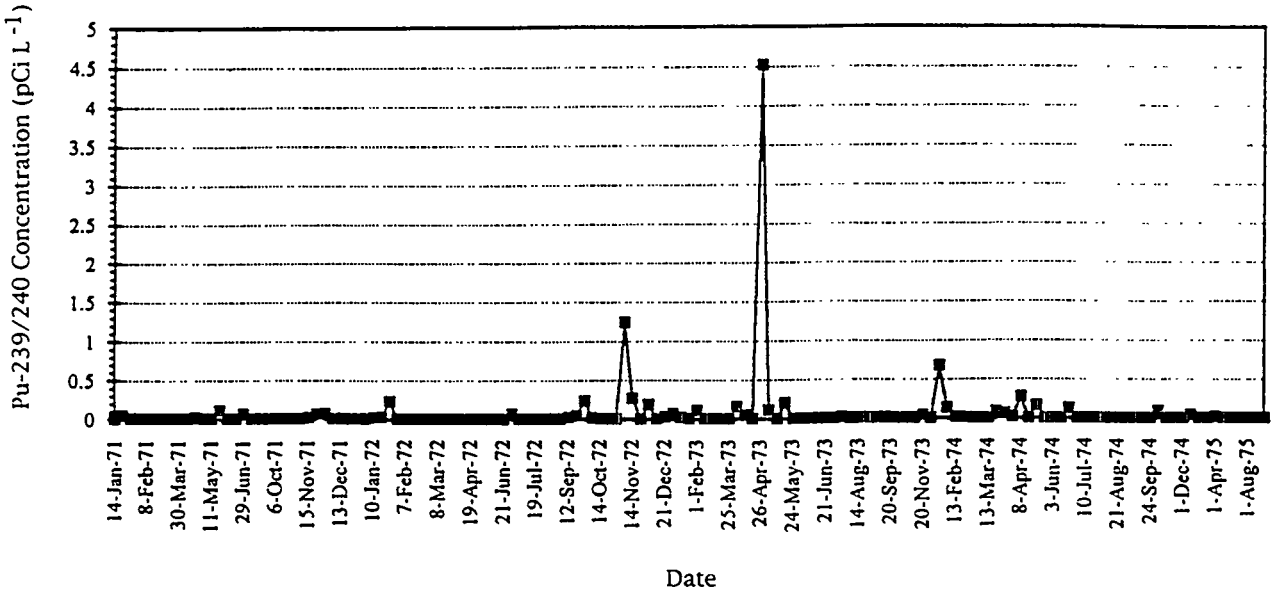
N/A = Not Available

Another way to evaluate the effects of this release event is to compare the levels of plutonium-239/240 in Broomfield water and Westminster water. In the 1970-1989 time period, the water supplies for the cities of Broomfield and Westminster were primarily drawn from Great Western Reservoir and Standley Lake, respectively. Both reservoirs are located downwind from the RFP and were likely to have been approximately equally impacted by airborne emissions from the plant; however only Great Western Reservoir received water directly from the North and South Walnut Creek drainage systems and was likely to have been impacted by contaminants released to the creeks. As plutonium-239/240 contaminated sediment was released into the North and South Walnut Creek drainage systems, drinking water monitoring data for the cities of Broomfield and Westminster taken between 1971 and 1975 with finer resolution than the annual were plotted to evaluate the impact of this release event. As shown Figure 5-20, with the exception of a few data points, levels of plutonium-239/240 in drinking water of Broomfield and Westminster measured during that period of time are about the same, less than 0.2 pCi L^{-1}. The two highest plutonium-239/240 levels measured, 1.3 pCi L^{-1} and 4.5 pCi L^{-1}, are associated with water samples collected from Broomfield in early November 1972 and in late April 1973. No similar elevation of plutonium-239/240 concentrations is observed in the water monitoring data for Westminster. Therefore, the two elevated levels of plutonium-239/240 in Broomfield water between 1972 and 1973 may be attributable to the pond reconstruction activities.

As shown in Table 5-10, the "background" natural uranium average concentration of 2.9 pCi L^{-1} for the entire period from 1970-1989 was below the Broomfield average of 3.1 pCi L^{-1} and above the Westminster average of 2.2 pCi L^{-1}. A review of the annual average Broomfield data indicates that this increased average is primarily due to a single year (1976) in which the average natural uranium concentration was almost three times greater than any other year. As indicated in Figures 5-16 and 5-17, drinking water from the individual "background" cities demonstrated similar episodes of elevated concentrations of natural uranium.

Both the Westminster and Broomfield 1970-1989 average tritium drinking water concentrations exceeded the "background" cities' tritium concentration in drinking water. At 1200 pCi L^{-1}, the long-term average tritium concentration for the Broomfield drinking water is clearly elevated above the "background" tritium concentration of 360 pCi L^{-1}. As mentioned previously, this is primarily due to the accidental release of tritium in 1973 and 1974. At 420 pCi L^{-1}, the 1970-1989 average tritium concentration in Westminster drinking water was slightly elevated above "background." A more detailed analysis of this accident can be achieved by comparing the levels of tritium measured in the Broomfield and Westminster water collected between 1971 and 1975 that are shown in Figure 5-21. As mentioned earlier, because liquid wastes were released into the North and South Walnut Creek drainage systems, only Great Western Reservoir and Broomfield water are expected to be directly impacted by the release of tritium to surface waters. As shown in Figure 5-21, before April 1973, levels of tritium in drinking water of Broomfield and Westminster are about the same, about 1,000 pCi L^{-1}. However, starting from

Pu-239/240 in Broomfield Water (1971-1975)



Pu-239/240 in Westminster Water (1971-1975)

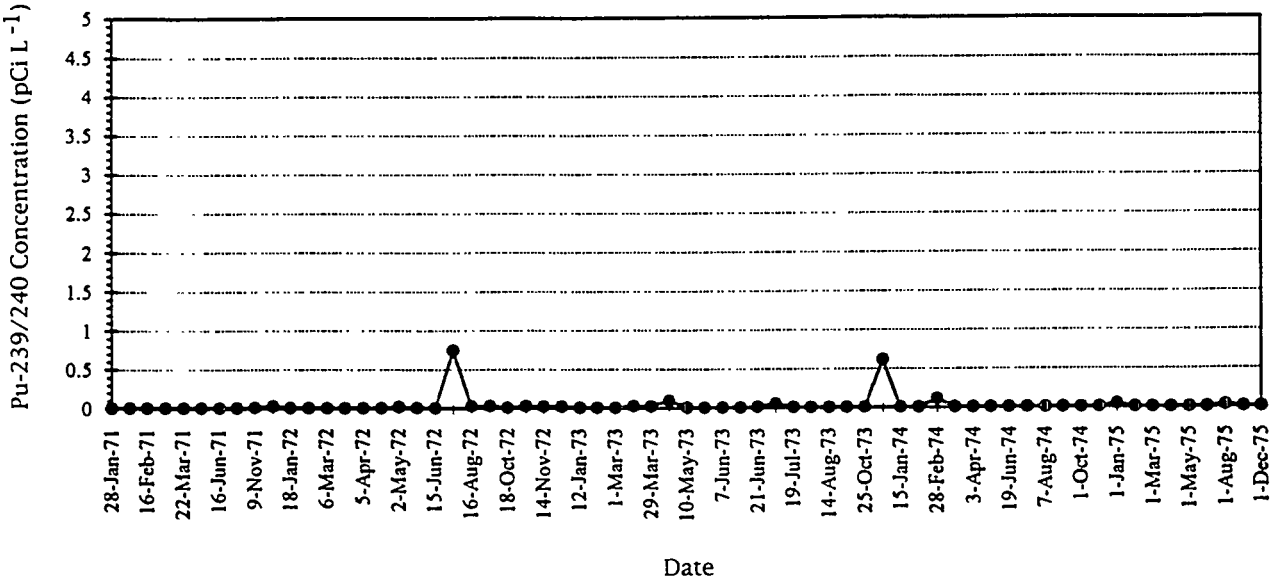


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FIGURE 5-20
LEVELS OF PLUTONIUM - 239/240 IN DRINKING WATER OF BROOMFIELD AND WESTMINSTER BETWEEN 1971 AND 1975

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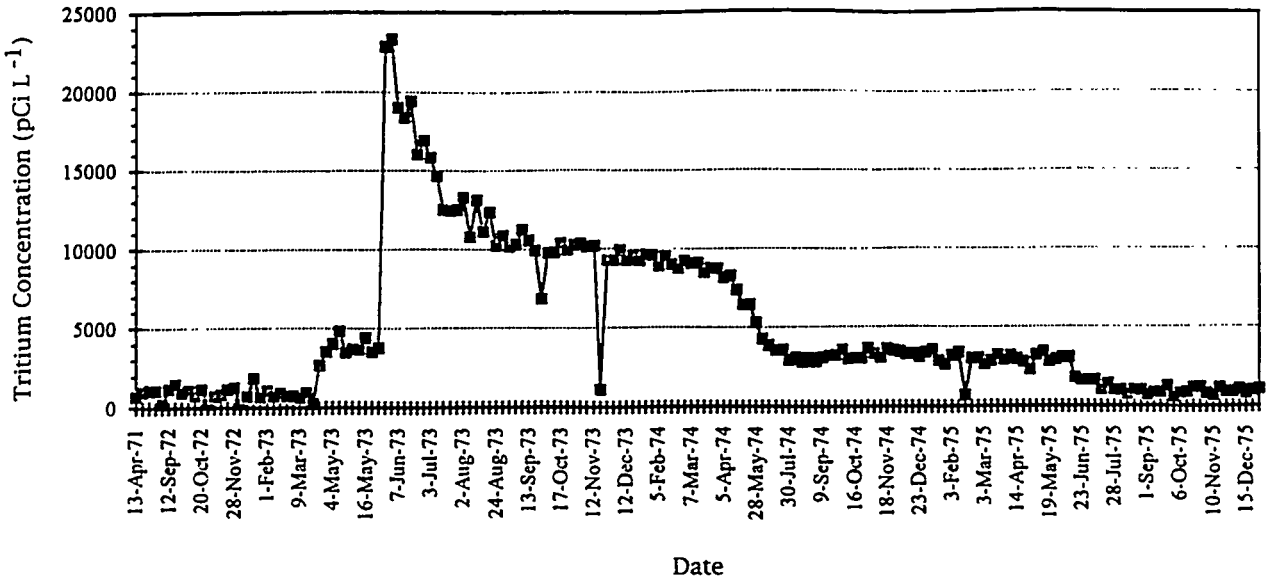
Tritium in Broomfield Water (1971-1975)



Tritium in Westminster Water (1971-1975)

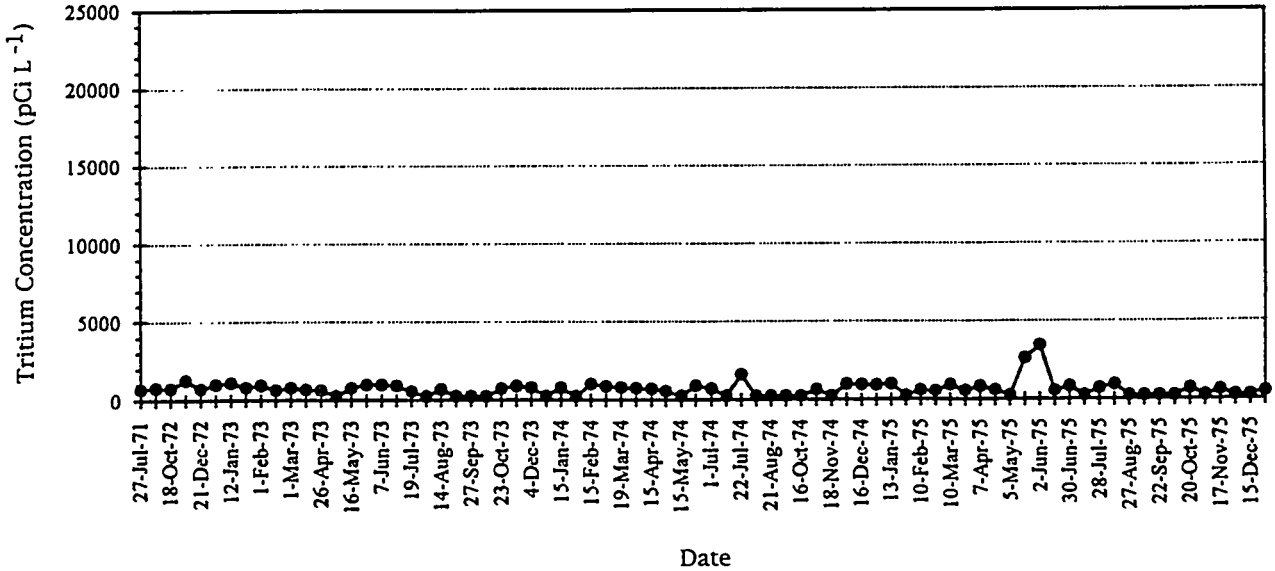


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FIGURE 5-21
LEVELS OF TRITIUM IN DRINKING WATER OF BROOMFIELD AND WESTMINSTER BETWEEN 1971 AND 1975

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April 2, 1973, tritium concentration in Broomfield water increased sharply and peaked at about 23,000 pCi L^{-1} in May 1973; it remained at a relatively high level for another two years before dropped back to the pre-1973 levels. No such changes were observed in the water samples collected from Westminster. It is likely that the accidental release of tritium to the Walnut Creek drainage systems in 1973 caused the observed elevation of tritium concentrations in Broomfield drinking water.

Mower Reservoir

CDH did not monitor Mower Reservoir for plutonium until 1980. During the 1980s, only seven samples were analyzed for Pu-238 and Pu-239/240. Three of the seven samples contained detectable concentrations of Pu-239/240, and no samples contained detectable concentrations of Pu-238. The maximum Pu-239/240 concentration was 0.12 pCi L^{-1}. The limit of detection for Pu-239/240 analyses was 0.02 pCi L^{-1}.

From 1974-1985, nine samples were analyzed for natural uranium. Three of nine samples contained detectable concentrations of natural uranium. The maximum concentration was 3.6 pCi L^{-1}. The limit of detection for the natural uranium analyses was 2.0 pCi L^{-1}.

From 1974-1986, thirteen samples were analyzed for tritium. Four of thirteen samples contained detectable concentrations of tritium. The tritium concentrations ranged from 360 to 770 pCi L^{-1}. The limit of detection for tritium was 350 pCi L^{-1}.

5.6 Results of Surface Water Monitoring Data Evaluations

The results of the examination of surface water monitoring data are presented in this section for 1952 to 1970, for 1970 to 1989, and for Mower Reservoir.

5.6.1 Conclusions for 1952 to 1970

On average, only a slight difference was detected between the gross alpha activity in background reservoirs and drinking water and the alpha activity in Great Western Reservoir, Standley Lake, and the drinking water of Broomfield and Westminster. The data suggest that throughout the 1952-1970 period, some single-year fluctuations of alpha activity in drinking water that may be attributable to the plant could have been as high as 1.5 pCi L^{-1}, but such fluctuations were not inconsistent with levels often found in "background" drinking water supplies.

5.6.2 Conclusions for 1970 to 1989

It is difficult to draw any conclusions regarding relative concentrations of radionuclides in drinking water during this period because of the large number of non-detect values in the data set for plutonium and uranium. Only a slight qualitative difference was observed in the average Pu-238 and Pu-239/240 drinking water concentrations of Broomfield, Westminster, and "background" cities. The pond reconstruction of the early 1970s may have caused an increase of 0.02 pCi L^{-1} and 0.15 pCi L^{-1} for Pu-238 and Pu-239/240, respectively, over a two-year period, but such levels were not inconsistent with levels often found in "background" water supplies.

The long-term average and annual average natural uranium concentrations in drinking water for the cities of Westminster and Broomfield were similar to those observed in the "background" cities.

Tritium concentrations for the city of Broomfield were clearly elevated above background. In single-year fluctuations, this elevation was as much as 8,100 pCi L^{-1}. Over the period from 1970-1989, the average elevation in tritium levels in Broomfield drinking water was 840 pCi L^{-1}.

The statistical analysis provided in Appendix I did not suggest any significant differences in the concentration of the sampled radionuclides in the waters of either Broomfield or Westminster compared to the "background" cities over the long-term with the possible exception of uranium, but even this is only weakly supported. Given the nature of the data set that are available none of these analyses should be construed as being definitive.

5.6.3 Conclusions Regarding Mower Reservoir

There is an insufficient amount of data to draw conclusions regarding the impact the plant may have had on Mower Reservoir. However, the limited sampling data available provide no indication that impacts to Mower Reservoir were significantly different than those discussed for Great Western Reservoir and Standley Lake for the periods sampled. In fact, no Pu-238, Pu-239/240, natural uranium, or tritium concentrations exceeded the long-term average concentrations for the "background" cities.

5.7 Sediment Sampling

Another potential source of information relative to surface water contamination is sediment sampling data. Over the last twenty years, a number of studies have addressed the plutonium concentrations in the bottom sediments of Great Western Reservoir and Standley Lake. An extensive summary of these studies is available in the Historical Information Summary and

Preliminary Health Risk Assessment for Operable Unit No. 3 (USDOE, 1991c). This section will briefly discuss the findings of these studies.

Seven studies have addressed the plutonium concentrations in the bottom sediments of Great Western Reservoir (USEPA, 1971; USEPA, 1973; USEPA, 1975; Dow, 1974; Battelle, 1981; Rockwell, 1979; Rockwell, 1985, as provided in USDOE, 1991c, Appendix D). The range of average plutonium concentrations reported in these seven studies was 0.11-3.5 pCi g^{-1} for surface sediments and 0.24 - 2.7 pCi g^{-1} for sediment cores. It should be noted that three of the seven studies were based on fewer than ten sediment samples (USDOE, 1991c). The results of these studies indicate that on average the plutonium concentrations in Great Western Reservoir sediments were greater than the USEPA estimated baseline level for area surface waters of ≤ 0.1 pCi g^{-1} (USEPA, 1973, as provided in USDOE, 1991c, Appendix D). Five of the seven studies concluded that the elevated plutonium concentrations and deposition patterns could be linked to historical releases from the Rocky Flats Plant.

Seven studies have addressed the plutonium concentrations in the sediments of Standley Lake (USEPA, 1971; USEPA, 1973; USEPA, 1975; Dow, 1974; Battelle, 1981; USDOE, 1978; Rockwell, 1984, as provided in USDOE, 1991c, Appendix D). The range of average plutonium concentrations reported in these seven studies was 0.04 - 1.3 pCi g^{-1} for surface sediments and 0.016 - 0.19 pCi g^{-1} for sediment cores. The results of these studies suggest that the plutonium concentrations in Standley Lake were greater than the estimated baseline of ≤ 0.1 pCi g^{-1} (USEPA, 1973, as provided in USDOE, 1991c, Appendix D). However, the conclusions of these studies are conflicting. Two of the USEPA studies concluded that the elevated plutonium levels resulted from unspecified releases from Rocky Flats (USEPA, 1971; USEPA, 1973; as cited in USDOE, 1991c), while, in 1975, the USEPA study concluded that there was no discernable plutonium contamination in Standley Lake attributable to Rocky Flats releases (USEPA, 1975, as cited in USDOE, 1991c). In 1978, a USDOE study concluded that 70 percent of the plutonium in Standley Lake was attributable to releases from Rocky Flats and speculated that this plutonium was transported both by airborne particulates and by surface water (USDOE, 1978, as cited in USDOE, 1991c).

From this brief review of the available sediment studies, it can be concluded that the plutonium concentrations in the bottom sediments of Great Western Reservoir are greater than the estimated baseline concentration of ≤ 0.1 pCi g^{-1}. It is also well understood that these elevated concentrations are likely due to historical releases from Rocky Flats. In a majority of the studies, the average plutonium concentrations in Standley Lake exceeded the reported baseline concentration of ≤ 0.1 pCi g^{-1}. Although the conclusions in a number of the reports are inconsistent, it is likely that the elevated plutonium concentrations are due to releases from Rocky Flats.

Summary: Available data are not sufficient for reconstructing contaminant releases in surface water from the plant. Except for previously identified accidental releases of plutonium-239/240 and tritium, the reservoir and drinking water monitoring data do not provide a clear indication of other plant contaminant release events. The magnitude of exposures associated with some of the observed fluctuations in radioactivity can be quantified, but we cannot determine whether the plant was the source of these fluctuations.

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6.0 NONROUTINE CONTAMINANT RELEASES

A number of nonroutine events have resulted in the release of contaminants from the Rocky Flats Plant. The events include accidents that have occurred, others represent operating practices that resulted in unintended releases of contaminants, while still others represent waste disposal practices that were associated with some expected release. Nonroutine contaminant releases identified as a result of an extensive search of classified and unclassified records are described in detail in the Tasks 3 and 4 report (ChemRisk, 1992). Documented accidents primarily involve the release of radionuclides, specifically plutonium.

Published records suggest that most of the plutonium released from the plant was released from the 1957 fire, the 1969 fire and the leaking oil drums on the 903 Pad. These events are the main focus of accident source term reconstruction. Other radionuclide releases include the accidental releases of plutonium through the filter plenum exhaust vents in 1965 and 1974, tritium in 1968 and the early 1970s, and depleted uranium in 1965. A well-publicized chemical release that resulted in some degree of on-site contamination is the 1989 release of chromic acid. The circumstances of these accidents as they relate to source term reconstruction will be discussed in some detail in this section.

There have been several attempts to estimate accidental radionuclide releases from Rocky Flats. After the May 11, 1969 fire, inquiries by Dr. E. Martell and the Colorado Committee for Environmental Information led to studies of plutonium deposition in the soils around Rocky Flats as a means of estimating total radionuclide releases from Rocky Flats. Such studies were published by Krey and Hardy (1970), Poet and Martell (1972), and Krey (1976). These studies drew attention to the importance of accidental releases. The 1980 USDOE FEIS contains some information on accidents at Rocky Flats. In 1981, C. W. Barrick, a former long-term Rocky Flats Plant employee, completed a Rocky Flats report summarizing the status of information about accidental radionuclide releases. Attention has also been drawn to accidental releases of radionuclides from Rocky Flats by the work of Dr. Carl Johnson, and by S. Chinn in connection with the Church litigation (see glossary).

This section summarizes available information relevant to source terms for accidental releases of radionuclides to the area surrounding the Rocky Flats Plant. Quantitative data necessary to develop accident source term estimates have been found in many places, including papers in the open scientific literature and in Rocky Flats reports. USDOE and Rocky Flats accident reports, available to the public in the Rocky Flats Reading Room, have also been used. In the case of the 1968 tritium accident, the accident has been acknowledged in the 1980 USDOE FEIS, but additional information is limited.

Since it is not possible to go back in time and duplicate or verify measurements and observations made at the time of the accidents, the approach to developing source terms involves careful review of data for plausibility and internal consistency, and cross-checking estimates with ambient data collected by the Rocky Flats Plant, the Colorado Department of Health, and others whenever possible.

6.1 Major Release Events

Major plutonium accidents with previously estimated releases greater than 1,000 microcuries (μCi) include the 1957 fire, the 1969 fire and the 903 Pad incident. The development of source terms for these major accidents involves the use of various types of documentation of these events, including monitoring and sampling data. The development of source terms for these events also required computer transport modeling of the ambient plutonium concentrations expected to result from different release scenarios consistent with available information about the accidental releases. The predicted plutonium concentrations in air and soil were compared to the available ambient measurement data to identify the source terms most likely to have produced the measured results. Description of this computer modeling activity is covered in the Task 6 report and will not be repeated here. Resulting estimates of source terms for the 1957 fire, the 1969 fire and the 903 Pad releases are included in this section.

6.1.1 1957 Fire

The 1957 fire began in Building 771 at about 10 p.m. on September 11, 1957 when metallic plutonium casting residues spontaneously ignited in a glove-box in Room 180. The fire then spread to an exhaust filter plenum, Rooms 281 and 282, consuming a considerable quantity of filters and damaging the ductwork and fan system. Because the samplers in the 771 Building stack were disabled by the filter plenum explosion during the early stages of the fire, there are no measurements of the release from the 771 stack during the fire. The limited data available from ambient air and vegetation monitoring during and after the fire were used in Task 6 to estimate the amount of plutonium released.

<u>1957 Fire Source Term Estimation</u>

A detailed description of the 1957 fire modeling is provided in the Task 6 report; only a summary of the modeling effort and source term estimation is presented here. Based on the historical information of the accident, modeling of the 1957 fire was divided into two periods: coarse particles released from the filter plenum explosion and fine particles from subsequent unfiltered fire emission. As particles of different size ranges and deposition characteristics were released in these two periods, they are also modeled separately in Task 6.

Coinciding with the explosion in the main filter plenum, a single "puff" of coarse particles is assumed to have been released at 10:40 p.m. on September 1957. The released particles were relatively large and primarily deposited within relatively short distances, leading to the observed plutonium contamination on vegetation. The USEPA INPUFF model was used to predict the dispersion and deposition of released plutonium particles. The amount of plutonium released during this period was estimated by matching the plutonium deposition pattern predicted by the model with the measured vegetation data. Using this approach, it is estimated that 60 μCi of plutonium could have been released by the explosion. The estimates of the upper and lower bounds of the 95 percent confidence interval about the best estimate are 1900 μCi and 1.9 μCi, respectively.

After the explosion, fine particles of plutonium produced by the burning HEPA filter and the fire in Building 771 were released into the atmosphere. The INPUFF model was used to model the average plutonium concentrations in air at several on-site and off-site locations during the 13.5-hour fire event. The appropriate release estimate was determined by matching the predicted air concentrations with those obtained from the routine on-site and off-site air samplers. Once the plutonium release estimate was established, predicted air concentrations at other locations were compared with additional air measurements taken by portable air samplers to increase the confidence level of the predictions. The total estimated release of fine particles from the fire is 0.07 Ci. The estimates of the upper and lower bounds of the source term of the 1957 fire are 2.4 Ci and 0.002 Ci, respectively.

There are considerable uncertainties associated with the release estimates because of the many assumptions that have been made and the generally poor quality of the available sampling and meteorological data. In addition, much of the estimating process for the fine particle release relied upon results at or below the detection limit, which would produce bounding estimates rather than best estimates of the release.

Previous Estimates of 1957 Fire Release

Barrick (1981) provides the largest published estimate of the total release of radioactivity to the environment from the September 11, 1957 fire at Rocky Flats. Barrick estimates that the release was between 0.7 and 1.3 Ci, with a most probable value of 1.0 Ci. At 0.0734 Ci per gram of plutonium, a release of 1 Ci corresponds to 14 grams of plutonium.

The 1980 USDOE FEIS estimated the total release from the 1957 fire as 25,618 μCi, or 0.35 gram of plutonium. Barrick (1981) notes that the FEIS estimate was obtained by subtracting the average of stack sampler readings from those months of 1957 unaffected by the accident from the amount of total alpha activity (assumed to be all plutonium) recorded by the Building 771 stack sampler from September through December 1957. However, this approach neglects the

fact that the Building 771 stack samplers were not operating from one hour after the fire began until eight days later on September 19, 1957. Barrick (1981) says that, for the above reasons, the estimate provided in the USDOE 1980 FEIS cannot be supported nor can an estimate of "one gram of plutonium off-site" often ascribed to Hammond.

Arguments presented during the Church lawsuit suggested that large amounts of plutonium could have been present on HEPA filters, and therefore could have been released during the fire. The estimates of plutonium loading on the filters presented in the trial arguments ranged from 10 to 250 kilograms (760 to 18,400 Ci).

6.1.2 The Building 903 Drum Storage Area

As described in the Tasks 3/4 and 6 reports (ChemRisk, 1992, 1993), the 903 Pad release began when plutonium-contaminated cutting oil and solvents, stored in drums to await recycling for recovery of plutonium, leaked into the soil. Soil particles contaminated with plutonium were subsequently carried off-site by the wind. This release from the 903 Pad is believed to have been the largest release of plutonium from Rocky Flats. The amount of plutonium released from the 903 Pad was estimated in Task 6 by using an air dispersion model and environmental monitoring data collected around the plant. A brief description of the modeling effort and the release estimate obtained in Task 6 are provided in this section.

903 Pad Source Term Estimation

The distribution of soil contamination observed in late 1969 and the early 1970s is the most important source of information regarding the 903 Pad release. However, it represents the cumulative result of releases from the pad and provides no insight into the time history of the release. Many different release scenarios were analyzed in Task 6, ranging from one large single day release to releases over the entire life of the pad. Based on the predictions of the Fugitive Dust Model (FDM) (Winges, 1990), the release scenario that gave the best fit to all the available data and assumptions used in the reconstruction was one that extended over a number of years. Actual releases from the pad were likely to have varied from day to day, being closely related to specific activities that led to disturbance of the pad. However, there is insufficient information on these daily activities to permit the evaluation of short time-span releases. The analysis performed in Task 6 was therefore focussed on the prediction of releases over the entire release period. FDM deposition modeling indicates that a total of 25 Ci were released from the 903 Pad:

- 11.4 Ci were redeposited on the pad,
- 13.6 Ci escaped from the pad,

- 8 Ci escaped from the plant exclusion area boundary, and
- 6.8 Ci escaped from the buffer zone boundary.

There are considerable uncertainties associated with the release estimates because of the many assumptions that have been made and the use of surrogate meteorological data. It was estimated in Task 6 that the uncertainty about the amount of plutonium that escaped from the buffer zone boundary is about a factor of 10.

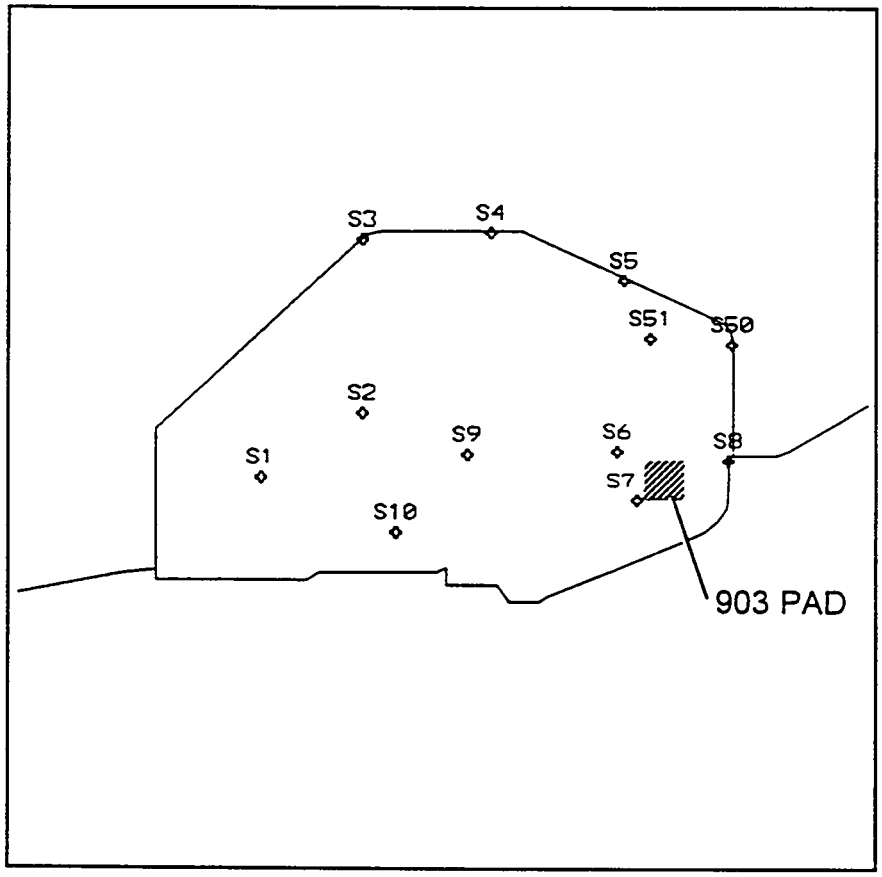
During the 903 Pad releases, air samples were routinely collected at several on-site and off-site locations. As shown in Figure 6-1, three on-site samplers (S-6, S-7 and S-8) are of particular importance. S-6 and S-7 are located near the pad while S-8 is located downwind from the pad. To increase our confidence on the modeling result, the predicted respirable airborne concentrations based on the soil contamination were compared with those measured by the three on-site air samplers. The five-year average S-8 air concentration predicted by FDM is 0.45 pCi/m^3, about 4 times higher than the measured value of 0.12 pCi/m^3. The predicted average for S-6 is 0.040 pCi/m^3, about 3 times higher than the measured value of 0.014 pCi/m^3, and the predicted average for S-7 is 0.12 pCi/m^3, about 6 times higher than the measured value of 0.02 pCi/m^3. The observed values are within the range of predicted uncertainty.

Previous Estimates of Total 903 Pad Release

Krey estimated a total 903 Pad release of 11.4 Ci +/- 20 percent. Assuming that the release was predominantly plutonium, the mass of plutonium released can be calculated by assuming 0.0734 Ci per gram of plutonium. The Krey estimate of 903 Pad releases is based on environmental measurements of soil plutonium concentrations and exceeds (by a factor of 1.8) the estimated release of 86 grams of plutonium (6.3 Ci at 0.0734 Ci per gram of plutonium) referenced by Seed *et al.* (1971). The Seed *et al.* (1971) estimate was based on the total number of leaking barrels and an estimated average plutonium concentration in the oil.

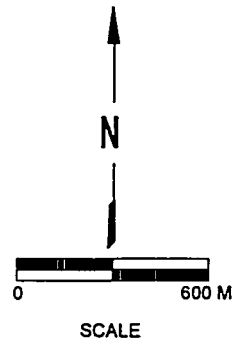
6.1.3 1969 Fire

Similar to the 1957 fire, the 1969 fire started when pressed plutonium briquettes spontaneously ignited in a glove box in Building 776. The fire subsequently spread to other inter-connected glove boxes and caused considerable damage to the building and its equipment. The fire burned for approximately four hours with some relatively minor flare-ups discovered as much as six hours after the first fire alarms.



LEGEND

◇ ON-SITE SAMPLER



ChemRisk
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FIGURE 6-1
ON-SITE AIR SAMPLERS
AT ROCKY FLATS

RKY294T5/6-1 REV. 03-03-94

Fire contaminants were reported to be almost completely contained within the buildings. Some smoke was observed coming out of the west end doors when they were opened and some smoke was observed coming from the roof and roof exhaust vents. One report theorizes that some of the observed smoke at the roof was from thermal decomposition of the roof itself (Willging, 1969). The fire did not breach the building roof.

1969 Fire Source Term Estimation

Because the stack samplers on Building 776 were disabled during the fire, measurements of the releases from the building during the fire are incomplete. In Task 6, the air monitoring data and meteorological data collected during and after the fire were used to estimate releases from the fire. Based on the prediction of INPUFF model, a total release of 2.8 mCi of plutonium is considered consistent with the air monitoring data. Three sources of uncertainty were identified in reconstructing the fire release: air dispersion model, air monitoring data, and time resolution/number of data points. The overall uncertainty of the source term of the 1969 fire was estimated to be about a factor of 20.

Previous Estimates of the 1969 Fire Release

In the 1980 Final Environmental Impact Statement (FEIS), a release of 856 μCi plutonium is attributed to the 1969 Fire (USDOE, 1980). No estimate of the amount of plutonium released to the environment is provided in the U.S. AEC 1969 Fire report (USAEC, 1969b). However, based on the information provided in the report it was estimated in Task 6 that approximately 2.6 to 5.9 mCi of plutonium were deposited on or in the vicinity of Building 776 during the 1969 Fire. This estimate cannot be directly compared to the estimate derived in Task 6, since it is based on activity deposited on-site and the approach employed in the Task 6 report accounts for only the airborne activity that had the potential to travel off-site.

6.2 Other Accidental Releases

Several smaller release incidents involving radionuclides and chromic acid were also identified in the Tasks 3 and 4 report (ChemRisk, 1992). Two plutonium accidents are discussed here; however, these releases have been included in the estimates of routine releases.

6.2.1 Plutonium Accidents

Accidental plutonium releases estimated in the 1980 USDOE FEIS at around 1,000 microcuries (μCi) each include the 1965 glove-box drain fire and the 1974 control valve failure. All of these releases were emitted through the filter plenum exhaust vents and were measured by stack samplers. In comparison, the annual releases of plutonium from normal operations at Rocky

Flats, as estimated in the 1980 USDOE FEIS, exceeded 1,000 μCi in all years from 1957 through 1965. ChemRisk's search of classified and unclassified records at Rocky Flats has not identified any other plutonium accidents likely to have caused releases of this magnitude.

6.2.1.1 1965 Glove-Box Drain Fire

On October 15, 1965, a fire occurred during a maintenance operation that involved unplugging a coolant recirculation line for a tape-controlled turning machine in Building 776/777. A double-bagged center punch was being used to dislodge an obstruction in the coolant recirculation line. Sparking occurring when the punch was struck, and a flash fire resulted. The fire vented to the room air and contaminants were spread throughout the building by the general ventilation system. The fire lasted 30-90 seconds before being extinguished with carbon dioxide.

Emissions from this accident were released through the main exhaust plenum filters. Therefore, this release is incorporated in the normal emission data and does not have to be modeled separately. The 1980 USDOE FEIS provides a release estimate of 1,170 μCi compared to releases from normal operations for 1965 of 5,348 μCi.

6.2.1.2 1974 Control Valve Failure

On April 2, 1974, radioactive particulates escaped through a roof exhaust vent on Building 707 following a glove-box atmosphere control valve accident. The accident was caused by a series of events when the inert atmosphere exhaust valve from the Building 707 storage vault was being closed during a glove-box maintenance procedure. A pressure surge caused contaminated gas to flow upstream through the inert gas supply system and back through the chiller and standby recirculation fan that had been turned off as part of the maintenance procedure. The contaminated gas was then pumped into the atmosphere by the purge exhaust fans through the exhaust vent.

The 1980 USDOE FEIS provides a release estimate of 934 μCi. Based on an interview with a former plant employee (Hornbacher, 1991), it was reported that the release estimate of from 233 to 236 μCi supplied in the Rocky Flats "Report of Investigation of Incident with Inert System No. 2 in Building 707 — April 2, 1974," by Freiberg *et al.* (1974), was later updated to agree with the estimate provided in the FEIS. The radionuclide release for normal operations for 1974 was 22 μCi, significantly lower than pre-1965 years due to the addition of 2 filter stages to plutonium processing building ventilation controls (in 1965) and upgraded filters in 1970 (USDOE, 1980). This release from the control valve failure did not bypass the stack monitors and has been included in the routine release estimates for 1974.

6.2.2 Depleted Uranium Burning

Depleted uranium in several physical forms was historically burned at the Rocky Flats Plant. Oils contaminated with depleted uranium were burned during the late 1950s and early 1960s as a method of waste disposal, until this practice ceased in 1965. Air emissions during burning oils were monitored by Rocky Flats Health Physics personnel. Wooden pallets contaminated with liquid containing depleted uranium were reportedly burned at a single on-site location in 1965. Also in 1965, similar wooden pallets containing sheets of depleted uranium were inadvertently burned at a single on-site location. This section identifies and discusses the data that are available to evaluate exposure of the off-site public as a result of these practices.

6.2.2.1 Open Pit Burning of Oils

The burning of radionuclide contaminated oil is known to have occurred in two on-site areas designated as oil burn pits #1 and #2 during the late 1950s and early 1960s. Airborne releases and any resuspension of contaminated soils are of potential concern for off-site health impacts.

Oil Burn Pit #1

Oil burn pit #1 has been referred to as the "garage oil burning pit" (Putzier, 1970). Approximately ten drums of waste oil containing depleted uranium from Buildings 444 and 881 were burned on August 18, 1956 as a waste disposal experiment (Owen and Steward, 1974). Based on available records, this seems to have been the only burning to have taken place in pit #1 (USDOE, 1992). The radionuclide content of the waste oil was not measured. Burn residue was left in place and covered with backfill (Unknown, 1975). Building 335 was then constructed over the burn pit (Owen and Steward, 1974). According to the above information, the burn residue is confined to the site and no off-site releases of contaminated soil have occurred to date. However, some quantity of depleted uranium was likely released to the air during burning.

Air monitoring was performed as part of the 1956 oil burning experiment. Total alpha radiation readings ranged from 0.1 to 30 dis min^{-1} m^{-3}. The lower reading was taken from the roof of Building 123 and the higher reading was approximately 60 feet south of the burn pit, directly in the visible smoke plume. These readings can be compared with background values of 0.9 and 3.5 dpm/m^3 taken immediately prior to the experiment (Chapman, 1956). This experiment indicated to Rocky Flats personnel that burning oils contaminated with depleted uranium did not pose a health hazard.

The available air monitoring results do not provide sufficient information to estimate total radionuclide emissions from burning the contaminated oil. Air sampling locations are not specified in detail; burn durations, temperatures, and dispersion patterns of emissions are unknown. In addition, all the parameters affecting emissions dispersion, such as wind speed, direction, temperature profiles, are also unknown.

Oil Burn Pit #2

Oil burn pit #2 was actually two trenches excavated parallel to each other. As a result of the 1956 experiment conducted in oil burn pit #1, a total of at least 1354 drums (USDOE, 1992) of waste oil, coolant, and still bottoms contaminated with depleted uranium from Buildings 444 and 881 were burned during the period of 1957-1965 (Freiberg, 1991). The burning was not continuous, but approximately 80 drums of oil were burned in a typical month during some periods (Ryan, 1965). The radionuclide content of the burned waste was not measured.

The burn residue and some flattened drums were originally covered with fill and the area was posted in 1959 to warn of contamination (Hill, 1959). Oil burning was discontinued in 1965 and oil burn residue and some flattened drums were covered with fill (Freiberg, 1991). In 1978, approximately 13,440 cubic feet of contaminated soil was excavated and shipped off-site for disposal. The volume of depleted uranium residue in the area was estimated to be 10,000 cubic feet (Unknown, 1985).

Oil burning in oil burn pit #2 was conducted at night to minimize concern from the smoke. Air monitoring was routinely performed by Rocky Flats Health Physics personnel. Available air monitoring results are summarized in Table 6-1. The results range from 0.0 to 16.7 dis min^{-1} m^{-3} (Dow Chemical, 1955-1965). Based on the evaluation performed in Task 6, it was estimated that a total of about 0.01 Ci could have been released from the oil burns.

6.2.2.2 Pallet Burning

Rocky Flats records indicate that wooden pallets containing depleted uranium were burned on the Rocky Flats Plant site in 1965 in two locations. Pallet burning is not reported to have been a routine practice. It was conducted southwest of oil burn pit #2 in 1965. According to persons interviewed for the Comprehensive Environmental Assessment and Response Program (CEARP) Phase I investigation, pallets believed to have been contaminated with a liquid containing depleted uranium were burned (USDOE, 1986). The quantity of contamination and details of the event are unknown. No documentation was located and long-term employees of the Rocky Flats Plant were not able to verify that wooden pallets were burned at this site (USDOE, 1992). The burn site was reported to have been "removed" in the 1970s (USDOE, 1986). "Removed"

TABLE 6-1

AIR MONITORING RESULTS OIL BURN PIT #2

Date	Location/Comments	Result (dis min^{-1} m^{-3})
6/4/59	50 yards downwind of oil pit #2, south of Building 991	4.9
8/13/59	South of Building 991	3.9
4/25/60	South of Building 991	4.3
5/27/60	50 yards downwind of oil pit #2, south of Building 991	0.0
7/7/60	South of Building 991	0.062
8/11/60	50 yards downwind of oil pit #2, south of Building 991	0.0
1/23/61	30 yards downwind of oil pit #2, south of Building 991	2.6
2/2/61	100 yards downwind of oil pit #2, southeast of Building 991	0.6
2/15/61	30 yards downwind of oil pit #2, southeast of Building 991	4.4
3/13/61	50 yards downwind of oil pit #2, southeast of Building 991	2.74
4/19/61	Unspecified location in "heavy smoke"	7.22
4/28/61 (1)	Unspecified location, at first "very black smoke"	14.6
4/28/61 (2)	Unspecified location, "grey smoke after fire had burned down"	4.35
5/12/61	South of Building 991	7.54
5/18/61	Unspecified location	10.2
6/8/61 (1)	Unspecified location, "very heavy smoke"	2.5

(continued on next page)

TABLE 6-1 (Continued)

AIR MONITORING RESULTS OIL BURN PIT #2

Date	Location/Comments	Result (dis min^{-1} m^{-3})
6/8/61 (2)	Unspecified location, "light smoke"	1.5
6/21/61	Unspecified location	3.2
7/19/61 (1)	Unspecified location, "heavy black smoke"	3.7
7/19/61 (2)	Unspecified location, "light smoke"	1.9
8/3/61	Unspecified location	3.3
8/7/61	Above Building 991	2.1
8/29/61	Unspecified location	7.9
9/13/61 (1)	Above Building 991 "at height of fire"	9.2
9/13/61 (2)	Above Building 991 "near end of fire"	5.8
9/27/61	Above Building 991	1.2
2/7/62	Unspecified location	1.72
5/15/63	Unspecified location	8.6
10/2/63	Unspecified location	4.58
10/3/63 (1)	50 yards to "right of oil pit"	0.33
10/3/63 (2)	30 yards from fire	0.76
10/15/63	"At oil pit"	4.9
1/16/64	Southeast of Building 991	0.449
2/11/64	"At oil pit"	16.7

typically means that contaminated soil was excavated from a site. In conclusion, the occurrence of wooden pallet burning at this location cannot be substantiated. In addition, a source term for a potential release cannot be constructed due to the lack of any information on what was actually burned and in what quantities.

Pallet burning of depleted uranium was also reported to have occurred in a burning pit south of Building 881 in May 1965. Three sheets (60 kg) of depleted uranium were inadvertently burned with the pallets. The depleted uranium sheets were positioned between large pallets for shipment from Medina, Texas, and possibly not well labeled. Records indicate that some portion of the depleted uranium residue remained after the burn because two barrels of contaminated soil were removed for disposal after discovery of the event (Young, 1965). No details of the quantity of residue are available.

If the value of 0.24 percent for the airborne amount of burning plutonium metal presented in the Rocky Flats FEIS is assumed to apply to uranium, the burning of 60 kg of uranium would have liberated 144 grams of uranium or 58 microcuries of alpha radioactivity [based on an alpha specific activity for depleted uranium of 4×10^{-7} Ci g^{-1} (Rich *et al.*, 1988)].

6.2.3 Tritium Release Incidents

Tritium is known to have been released during operations at Rocky Flats. A low level of routine releases was observed and several accidents resulted in emission of larger quantities of tritium to off-site air and surface waters. This section describes source term aspects of three accidental releases of tritium reported from the Rocky Flats Plant.

<u>The 1968 Tritium Release</u>

The 1980 USDOE FEIS acknowledges that several hundred curies of tritium were released from Rocky Flats in an accident during 1968. The accident resulted in the release of 600 Ci of gaseous tritium to the ambient air (USDOE, 1980). The details of this accident are classified, but several classified documents describing the event have been reviewed by ChemRisk project personnel. No additional information could be located to either support or refute the release estimate.

<u>The 1973 Tritium Release</u>

The first evidence of this release was from a routine monthly water sample collected by the Colorado Department of Health from Walnut Creek that indicated 3,000,000 pCi L^{-1} of tritium. This concentration was well above background and equaled the maximum permissible concentration for uncontrolled areas. This anomalous result could not be explained based on

routine Rocky Flats sources. Subsequent Walnut Creek sample results showed decreasing tritium concentrations indicating an isolated release. The 1973 tritium release occurred when contaminated scrap plutonium received from Lawrence Livermore Laboratory was handled in a process not designed for tritium control.

Barrick (1981) says:

> "...an estimated 350 to 1600 Ci of tritium was released in exhausted air and 150 to 400 Ci was located in process and waste waters. Processing of the waste waters...resulted in 100 to 300 Ci tritium in on-site tanks, basins and ponds and 50 to 100 Ci in the Great Western Reservoir (average 0.01 microcurie/liter for 3 months)." (page 29)
>
> "It was calculated that a typical Broomfield resident drinking Great Western Reservoir water would receive less than 1.4 millirem compared to a maximum permissible level of 170 millirem." (page 32)

In comparison, a Rockwell International December 1986 news release claims:

> "...100 to 500 curies of tritium were released in exhausted building air; 150 to 400 curies were located in process and waste water on plant site. An USEPA report indicated 56 curies of tritium migrated into Great Western Reservoir, the drinking water supply for the city of Broomfield."

The USAEC report, "Investigation of the Tritium Release Occurrence at the Rocky Flats Plant" (USAEC, 1973), concludes:

> "In summary, approximately 500-2000 curies of tritium were transferred to Rocky Flats from LLL in the scrap shipment. This tritium was released in large part up the stack of Building 779A where it was dispersed to the environment. Due to the small size of the airborne releases, coupled with the low contamination levels found in the Rocky Flats environs, Dow (Rocky Flats) staff...concluded that no significant off-site contamination or public exposure could have resulted from the atmospheric releases.
>
> "The remaining tritium followed the liquid waste processing flow sheets of the scrap and the associated wastes therefrom, resulting in dispersal to Buildings 779, 771, 774, 881, 444 and other areas which handled the scrap. The treated liquid wastes from these buildings were subsequently discharged to the sanitary sewer

> or to the evaporation ponds, in a time sequence which could account for the calculated accumulation of 100 - 300 curies in the evaporation ponds and the 50 - 100 curies found in Great Western Reservoir.
>
> "Dow (Rocky Flats) analysis of the tritium levels in the Great Western Reservoir indicates that a Broomfield resident drinking water from Great Western Reservoir continually during 1973 would receive less than 1.4 millirem during 1973 or less than 1 percent of the radiation protection guidelines for the public. Elevated trace levels of tritium which were found below the Rocky Flats sanitary landfill could have originated from 'cold wastes' from areas not contaminated by plutonium but which could have been unknowingly contaminated with low levels of tritium or by deposition from the original airborne release." (pp. 27 and 28)

The 1973 findings associated with the tritiated plutonium initiated an investigation of other possible similar shipments and processing of tritiated plutonium. The investigation discovered three other shipments with maximum estimated tritium releases of 57 Ci (April 1969), 40 Ci (March 1971), and 29 Ci (November 1971).

The 1974 Tritium Release

The 1974 tritium release is believed to have been caused by a problem with a pressure cooler operation in Building 776/777. The report "Investigation of a Tritium Release Occurring in Building 777 on September 3-4, 1974" (USAEC, 1974, page 28) estimates this release as 1.5 Ci.

6.2.4 Chromic Acid Spill

On February 22, 1989, waste chromic acid overflowed a tank and a secondary containment berm into the basement of Building 444. The solution leaked into the foundation of the building to the footing drain system, which was piped to the plant's sewage treatment plant. The contamination moved through the treatment plant in about 24 hours and was discharged to retention pond B-3. The B-3 pond water was pumped to the east spray field and spray irrigated onto frozen ground. Consequently, the chromium-contaminated spray water ran off the hillsides adjacent to the spray field and was collected in the water impoundment ponds on-site. Chromium was not identified as a contaminant until February 28 (USDOE, 1989).

Seventy pounds of dry chromic acid (hexavalent chromium) had been placed in the tank just prior to the spill on February 22. An investigation estimated that 750 gallons of chromic acid entered the drain on February 23. The material entering the sewage treatment plant on February 23 was 13 ppm chromium, and the treated effluent exiting the sewage treatment plant was 2 ppm

chromium. Approximately 2,763,000 gallons of B-3 pond water was spray irrigated from February 23 to March 4. On March 4, the B-3 pond was sampled and contained 0.19 ppm total chromium. Most of the B-3 pond water ran into the B-5 and C-2 ponds. The total chromium concentrations of these two ponds were 0.08 and 0.06 ppm, respectively. The spray field runoff water contained from 0.03 to 0.08 ppm total chromium (USA, 1992). The USDOE report on the chromic acid accident (USDOE, 1989) states:

> "No off-site release of the chromium contaminant occurred. An estimated 30 pounds of chromium were released to the sanitary sewer system and approximately 5 pounds passed through the sewage treatment plant and were distributed in the spray fields and ponds.
>
> "After chromium was identified as the contaminant on February 28, soil samples taken at one and six inch depths in the spray fields showed chromium levels to be within site background levels. Daily water samples taken from the impoundment ponds after the ice thawed on the ponds, thus permitting representative sampling, indicated chromium levels below the Clean Water Act drinking water standard of 0.05 ppm. Borehole samples indicated no chromium contamination above background levels in the gravels and soils adjacent to the Building 444 foundation drain system."

No documentation of off-site contamination was located for the event.

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7.0 SUMMARY

This report completes an important step in the process of historical dose reconstruction in providing the emission estimates that will form the basis for subsequent contaminant transport modeling in project Task 6 and exposure and dose assessment in Task 8. The principle elements of this effort have included the following:

- A review and evaluation of the airborne radioactive effluent monitoring program, which has provided the basis for establishing quantitative annual estimates of routine releases of the radioactive materials of concern and the uncertainties associated with these releases.

  Quantitative estimates of airborne releases can be derived for each of the radioactive materials of concern, with the exception of Th-232, using the effluent monitoring program data. Th-232 has not been specifically monitored, saw only limited use at the plant, and is not believed to have been associated with significant emissions historically.

  The nature of the data generated by the plant's historical airborne effluent monitoring program and the similarity in the dose factors for the radioisotopes of concern argue for consideration of the use of composite dose factors and emission estimates that would combine emissions for a number of isotopes, as opposed to evaluating all isotopes individually.

- Routine airborne emissions of nonradioactive materials, which can be developed from monitoring program data only in the case of beryllium. Routine monitoring for the organic solvents of concern was not performed by the plant. Estimates of the plausible ranges of historical emissions for these materials were developed using various types of documentation and information obtained from personnel interviews. In many cases, these emission estimates are based on very limited information. However, the identified range of emissions is believed to bound the actual plausible emissions from the plant for these materials.

- Review of information regarding surface-water-borne contaminant releases from the plant indicating relatively limited availability of data to directly quantify the release of materials of concern. What data were available were used to examine whether plant releases measurably increased the radioactivity present in potentially impacted reservoirs and drinking waters. While the review of the data suggested that it was plausible that plant-related releases may have on some occasions measurably increased radioactivity in the waters of the receiving reservoirs, the

resulting measured levels of radioactivity were similar to levels found in other unaffected reservoirs in the area.

- A review of information and data associated with nonroutine releases of contaminants from the plant providing the basis for further modeling of major release events (1957 fire and 903 Pad) and for evaluation of the relative magnitude of lesser events in comparison to routine emissions. The information presented in this report relative to the major events will be employed in contaminant transport modeling efforts in Task 6 to finalize an emission estimate and to provide the basis for estimating off-site exposures from these events.

APPENDIX A

AIRBORNE EFFLUENT PARTICLE SIZE STUDIES

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AIRBORNE EFFLUENT PARTICLE SIZE STUDIES

Discussions of particle sizes rely on the use of the following terms:

- Aerodynamic equivalent diameter - the diameter of a unit density (1 g cm^{-3}) sphere with the same settling velocity as the particle in question. For PuO_2 particles, aerodynamic equivalent diameters are approximately three times actual particle diameters (Hayden, 1976).

- Mass median diameter - for a distribution of particles of various sizes, the mass median diameter identifies the size for which half the total *mass* of material collected is contributed by smaller particles and half by larger particles.

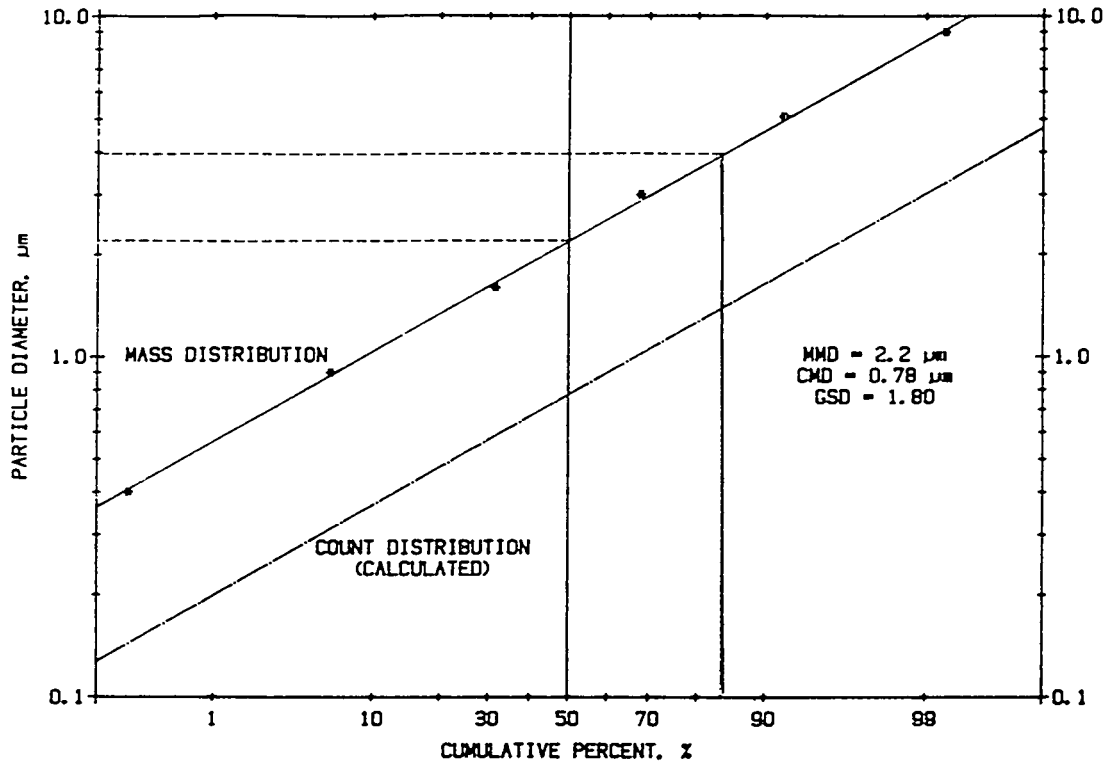
- Count median diameter - for a distribution of particles of various sizes, the count median diameter identifies the size for which half the total *number* of particles collected is contributed by smaller particles and half by larger particles.

- Activity median aerodynamic diameter - the median of the distribution of radioactivity or toxicological or biological activity with respect to aerodynamic diameter (Hinds, 1986). The AMAD is the diameter of a unit density sphere with the same terminal settling velocity in air as that of an aerosol particle whose activity is the median for the entire aerosol (USEPA, 1988).

The methods by which several of the above descriptors of particle size can be determined from sampling data are depicted in Figure A-1.

A number of studies of the particle size distribution of effluents from Rocky Flats were conducted in the early to mid-1970s. Several of these studies were authored by J. A. Hayden and are documented in Rocky Flats internal reports designated "Product and Health Physics Research Service Reports." A number of these studies are summarized in the following discussion and in Table A-1.

Building 776 exhaust was sampled in 1972 using 3 μm membrane filters, which were analyzed for particle size using the fission track method (Hayden *et al.*, 1972a). The results of this study indicated that the count median diameter of particles observed in Building 776 effluent air was between 0.07 and 0.12 μm in general and between 0.12 and 0.15 μm in an exhaust system that appeared to be malfunctioning at the time. A recommendation was made that future collections be made with two filters arranged in tandem in order to allow for better quantitative estimates of material released.



(Fraction of total particle mass contributed by particles less than given diameter)

Mass Median Diameter (MMD) = diameter corresponding to the 50 percentile (median).

For lognormally distributed particles, Geometric Standard Deviation (GSD) and Count Median Diameter (CMD) are found as follows:

$$GSD = \frac{84 \text{ percentile particle size}}{50 \text{ percentile particle size}} = \frac{50 \text{ percentile size}}{16 \text{ percentile size}}$$

$$CMD = MMD\ e^{-3\ (\ln GSD)^2}$$

Reference: Hinds, 1986.

Note: Sample data do not represent Rocky Flats sampling studies.

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FIGURE A-1
LOG PROBABILITY PLOT OF PARTICLE
SIZE DATA WITH RELATED RELATIONSHIPS

TABLE A-1: SUMMARY OF ROCKY FLATS AIRBORNE PARTICLE SIZE STUDY DATA

Study	Sampling Point	Count Median Diameter	Mass Median Diameter	Aerodynamic Equivalent Diameter	Comments
Hayden *et al.*, 1972 (used 3 μm membrane filters, fission track sizing method)	Building 776 exhaust (in general)	0.07 - 0.12 μm			Tandem filters were not used.
Hayden *et al.*, 1972 (used 3 μm membrane filters, fission track sizing method)	Building 776 exhaust (in a malfunctioning exhaust system)	0.12 - 0.15 μm			Similar distributions on the two filters. Significant breakthrough to backup filters.
Hayden, 1972 (used 3 and 0.8 μm membrane filters, fission track sizing)	Building 776 effluent		0.09 μm GSD = 1.6	0.3 μm (if PuO_2) 0.4 μm (if metal)	Particle sizes were consistent with theory of HEPA filter operation.
Hayden, 1974 (used membrane filters, fission track sizing method)	Building 771 effluent	0.09 - 0.19 μm			Leaking HEPAs caused non-lognormal distribution, indicating multiple sources.
Hayden *et al.*, 1976 (continuous air monitor filters; fission track, alpha track, and microscopy sizing techniques)	Inside Building 707 after a plutonium oxide spill	most were < 0.2 μm			Lognormally distributed.
Moss *et al.*, 1961 (used membrane filters, optical and alpha track sizing)	Unfiltered plutonium aerosols from plutonium processing		0.14 to 0.65 μm (SD 1.3 to 1.9)		Size distributions varied little between operations.
Hayden, 1978 (Pu salts, alpha track sizing)	Between HEPA stages, Building 771	0.075 μm		0.25 μm (if PuO_2)	Collected between 5th and 6th stages of HEPA filters.
Elder *et al.*, 1974 (cascade impactor, alpha counting)	Building 707 exhaust prior to final filters.			over 50% 1-5 μm* (3.3 to 4.7 μm* dominates)	Fabrication operations produced largest particle sizes.
Elder *et al.*, 1974 (cascade impactor, alpha counting)	Building 771 exhaust prior to final filters.			over 70% <1.0 μm*	Recovery operations produced highest activity and smallest aerosol sizes.
Nininger and Osborne, 1992 (laser particle analyzer)	Building 559/561 exhaust			<<2 percent >5 μm	Strong bias toward particles smaller than 0.3 μm. All radionuclides unquantifiable.

SD = standard deviation; GSD = geometric standard deviation; *Activity median aerodynamic diameters (AMAD)

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Samples of Building 776 effluent air were again obtained in 1972 over several 48-hour periods using 3 μm and 0.8 μm membrane filters (Hayden, 1972). The filters were again analyzed for particle size using the fission track method. The study concluded that the mass median diameter of particles observed in Building 776 effluent air was about 0.09 μm and that particle size distributions on the two types of filters were similar. However, a number of problems were noted with the sampling apparatus used in the study. For the 48-hour collection periods in this study, significant breakthrough of the millipore filter sampling media (millipore filters are not used for routine effluent sampling) occurred. An average of 50 percent of the total particles collected during 4 tests of the 3 μm filters were on the backup filter. During the one test of the 0.8 μm filter, 46 percent of the particles collected were found on the backup filter. No third filter assembly was used in these tests.

Samples of Building 771 effluent air were obtained in 1974 using membrane filters that were analyzed for particle size by the fission track method (Hayden, 1974). The results of this study indicated that

- The count median diameter of particles observed in Building 771 effluent air was between 0.09 and 0.19 μm.

- Certain upstream HEPA filters were leaking, resulting in a particle size distribution that was not lognormal. This indicated multiple sources for the sampled particles.

A review of Rocky Flats particle size data prepared in 1976 (Hayden, 1976) referred to the earlier particle sizing work done in Building 776 (Hayden, 1972) and stated some further conclusions:

- The measured mass median diameter of plutonium particles of 0.09 μm is equal to an aerodynamic equivalent diameter of 0.3 μm. This equivalency was based on a density of 11.45 g cm^{-3} for PuO_2 spheres. If the effluent was plutonium metal of density 19.8 g cm^{-3}, the aerodynamic equivalent diameter would have been about 0.4 μm.

- The observed aerodynamic equivalent diameter of 0.3 μm was considered to be consistent with the theory of operation of HEPA filters; that is, theory predicts that the filter media will be the least efficient for 0.3 μm particles. Prior to HEPA filtration, the effluent is expected to contain larger particles.

From the above studies, the best estimate of particle size distribution for particulate emissions that passed through HEPA filters (plutonium, uranium, etc.) is given by the size distribution that penetrates HEPA filters. Hayden (1976) reports that plutonium particles in Rocky Flats effluent air have a mean diameter of 0.09 micrometers and a very narrow size range (geometric standard

deviation = 1.6). Taking 0.09 as a geometric mean particle size, and assuming a lognormal size distribution with geometric standard deviation 1.6, 99 percent of the particles are smaller than 0.27 micrometers.

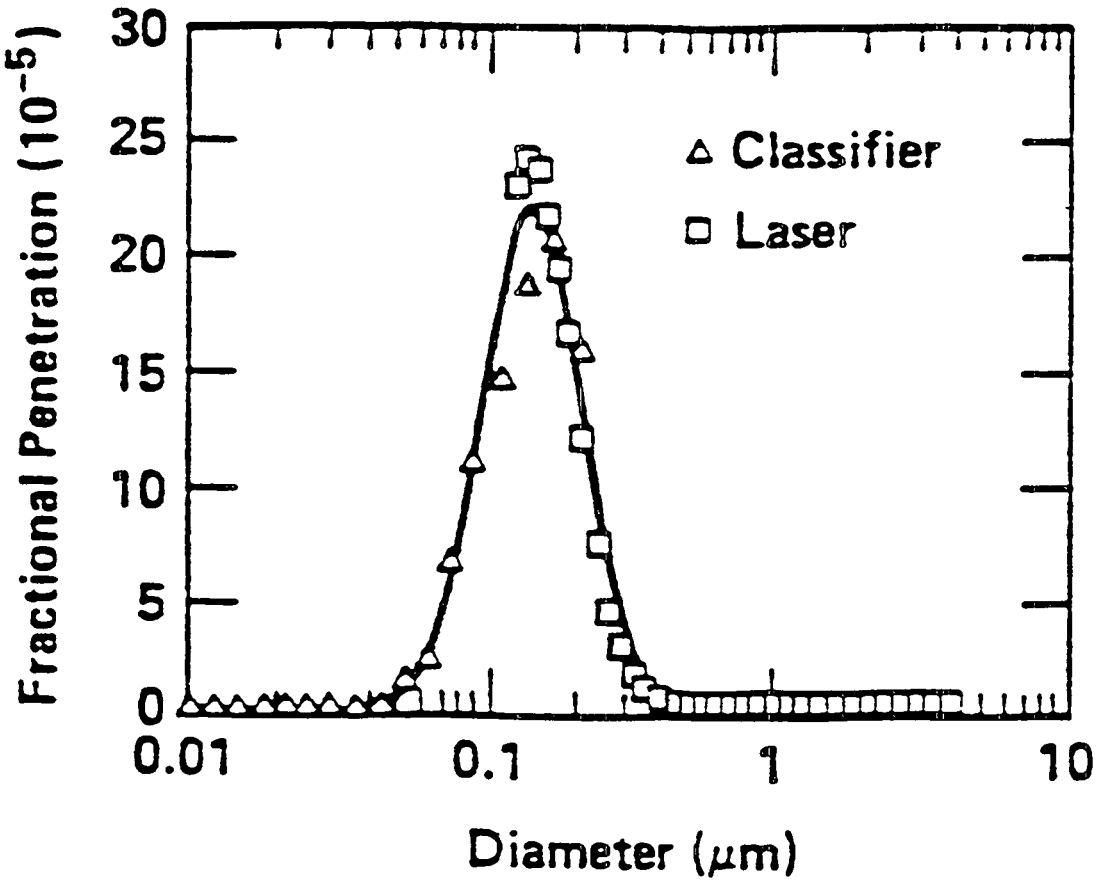
Elsewhere, Hayden (1972) noted that virtually all of the particles collected in Building 776 air were less than 0.3 micrometers in size. Figure A-2 (5.2 from Attachment 1 to EG&G Report 93-RF-2657, "Determination of Particle Size Distribution and Composition of the Effluent Air Emissions from Building 559"; Nininger and Osborne, 1992) also shows a narrow, lognormal physical particle size distribution for particles passing through HEPA filters, with a median value of about 0.1 micrometer and a maximum size of about 0.3 micrometer.

Other studies were also identified that characterized the particle size of effluents at various points before they reached the final filtration step and were released to the environment. While this information is of limited interest in addressing environmental release and transport, it provides a more complete picture of the effluents generated by Rocky Flats processes prior to final stages of filtration. One of these studies involved particle size analyses on filters from the Building 707 continuous air monitoring (CAM) system after a plutonium oxide spill (Hayden *et al.*, 1976). These analyses were conducted using fission track, alpha track, and optical and electron microscopy techniques. Although larger particles were present, most of the particles observed were less than 0.2 μm and were lognormally distributed.

Another study, conducted by Los Alamos researchers (Moss *et al.*, 1961), looked at the particle size distributions of *unfiltered* plutonium aerosols resulting from various chemical, metal preparation, and fabrication processes. Results of this study indicated that

- Particle mass median diameters were quite small (on the order of 0.14 to 0.65 μm).
- Standard deviations of the observed distributions were very low (on the order of 1.3 to 1.9).
- Size distribution characteristics varied little from one operation to another.

Particle size analysis was also performed on samples of magnesium nitrate salts collected from between the fifth and sixth stages of the HEPA filtration system in Building 771 (Hayden, 1978). This study showed a count median diameter of 0.075 μm. For plutonium oxide, this is equal to a 0.25 μm aerodynamic equivalent diameter and is again consistent with the particle size distribution expected downstream of HEPA filters.



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Reference: Biermann and Bergman, 1986.

FIGURE A-2
PENETRATION MEASUREMENT
FOR A STANDARD HEPA
FILTER WITH NO LEAKAGE

Another Los Alamos study looked at particle sizes of effluents from plutonium recovery activities in Building 771 and fabrication activities in Building 707 prior to final stages of HEPA filtration (Elder *et al.*, 1974). The study found that particles from the fabrication operations (Building 707) were predominantly collected with activity median aerodynamic diameters in the range of 3.3 to 4.7 μm, which are larger than those from the previously discussed studies. However, significant quantities of much smaller particles were also present. The majority of the particles present in effluent from recovery activities in Building 771 had activity median aerodynamic diameters of less than 1.0 μm. The study made the observation that the recovery operations produced the highest activity and smallest aerosol size, presenting the most difficult air cleaning problem for a number of different facilities that were characterized in the study.

A 1992 particle sizing study examined particles in effluents from Building 559 (Nininger and Osborne, 1992). A laser-based airborne particle counter was used to count particles in a series of size ranges, the smallest being 0.3 to 0.5 μm. Results showed the particle size distribution to be strongly biased toward very small particles, particles smaller than the 0.3 μm size that could be detected with the laser particle counter. It is reported that well below 2 percent of the particles estimated to have been present would have had aerodynamic diameters larger than 5 μm. Alpha track analyses of filtered effluent revealed no significant alpha-emitting radioactivity, but did suggest the possible presence of some very small alpha-emitting particles. Isotopic analyses showed no quantifiable radioactivity present.

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771-MAI - the main exhaust from Building 771 through the 145-foot tall stack;

444-MAI - the main exhaust from Building 444;

444-DOS - the Building 444 beryllium shop exhaust;

447-MAI - the main exhaust from Building 447;

883-AAA - BBB and CCC - the Building 883 exhausts.

Each of the DARs reviewed says that "Effluent sample is extracted through each sample probe at a rate of 2 dry standard cubic feet per minute (dscfm)." Reynolds numbers in all of these ducts were in excess of 400,000, corresponding to turbulent flow, and the DARs claim this results in a homogeneous distribution of particles in the effluent stream. The DARs reviewed indicate that there is one sampling nozzle per sampling probe except in the 771-MAI duct. Duct flow rates and velocities given in the DARs are not the same as those given in Repository Document RE-1029, but this is to be expected because flow rates in the exhaust systems differed at different times in the history of Rocky Flats operations.

The largest flow rate listed in Repository Document RE-1029 is for exhaust system 771-MAI (the main exhaust through the Building 771 stack), with a flow rate of 203,344 cubic feet per minute. The total exhaust volume from all vents listed in Repository Document RE-1029 is about 1,466,000 cubic feet per minute, excluding the exhaust volume from Building 371 (lines 51 and 52) because that building never became operational. Therefore, the 771 stack, which released about 90 percent of the plutonium from normal operations at Rocky Flats, was responsible for about 15 percent of the total exhaust air volume (excepting Building 371) tabulated in Document RE-1029.

The "pitot diameter" tabulated in Repository Document RE-1029 for each exhaust vent is believed to be the sampling tube inside diameter (i.d.), because the listed sample velocity for all exhaust vents corresponds to a sampling rate of 2 cubic feet per minute if the pitot diameter is used as the sampling tube inside diameter. The sample velocity of 1956 ft min^{-1} for the 771-MAI system is 1/3 of the sampling line velocity of 5867 ft min^{-1} calculated for the listed 0.25-inch sampling line for sampling at a rate of 2 cubic feet per minute. This is consistent if three 0.25-inch i.d. sampling nozzles were feeding each of the three sampling lines on the 771 MAI system, and the sampling lines also had an i.d. of 0.25 inch. Otherwise, if 2 cubic feet per minute were drawn through each sampling nozzle, the sample velocity in each nozzle would be 5867 ft min^{-1}. The DAR for the 771-MAI system reports sample nozzle inside diameters of 0.125 inch each for the three sample nozzles on each of the three sampling lines. Mr. W. Osborne of Rocky Flats (telephone conversation on 5/13/93) stated that the sampling rate is 2

cubic feet per minute in each of the three nozzles on the sampling lines, consistent with the isokinetic ratio of 12.17 reported in the 771-MAI DAR.

All pitot diameters in Repository Document RE-1029 are multiples of 0.125 inch. However, Mossoni and Kittinger reported that the sampling lines they studied in Buildings 707, 771, and 776 were between .305 and .430 inches inside diameter, and sampling line inside diameters in six of the seven DARs reviewed were not multiples of 0.125 inch. Discrepancies between the DARs reviewed and Repository Document RE-1029, for exhaust ducts other than 771-MAI, were as follows:

444-MAI: sample probe i.d. = 0.456 inch compared to 0.5 inch in RE-1029; duct cross-sectional area = 72 ft^2 as compared to 64 ft^2 in RE-1029;

444-DOS: sample probe i.d. = 0.4 inch, compared to 0.5 inch in RE-1029;

447-MAI: sample probe i.d. = 0.456 inch, compared to 0.625 inch in RE-1029;

883-AAA: sample probe i.d. = 0.402 inch, compared to 0.5 inch in RE-1029; duct cross-sectional area = 35.4 ft^2, compared to 48 ft^2 in RE-1029.

883-BBB: sample probe i.d. = 0.402 inch, compared to 0.5 inch in RE-1029; and

883-CCC: sample probe i.d. = 0.313 inch, compared to 0.5 inch in RE-1029; duct cross-sectional area where stack samples taken = 17.4 ft^2 and exhaust stack area = 12.6 ft^2, compared to duct cross-sectional area 12 ft^2 in RE-1029.

Mossoni and Kittinger (1973) reported sampling rates of 1.4 cubic feet per minute in the Building 771 lines tested, 1.5 cubic feet per minute in the Building 707 lines tested, and between 1.9 and 2.1 cubic feet per minute in the Building 776 lines tested.

Effects of anisokinetic sampling at Rocky Flats

Hayden (1976) reports that plutonium particles in Rocky Flats effluent air have a mean diameter of 0.09 μm and a very narrow size range (geometric standard deviation = 1.6). Taking 0.09 as the geometric mean particle size, and assuming a long-normal size distribution with geometric standard deviation 1.6, 99 percent of the particles are smaller than 0.27 μm. Elsewhere, Hayden (1972) noted that virtually all of the particles collected in Building 776 effluent air were less than 0.3 μm in size. Figure 5.2 in Attachment 1 to EG&7G Report 93RF-2657 "Determination of Particle Size Distribution and Composition of the Effluent Air Emissions from Building 559" (Nininger and Osborne, 1992) also shows a narrow log-normal particle size distribution for

particles passing through HEPA filters, with a median value of about 0.1 μm and a maximum size of about 0.3 μm.

Errors introduced by anisokinetic sampling were estimated using a particle density of 11.46 g cm^{-3}, corresponding to plutonium dioxide, and a computer program to perform the calculations described above. About 90 percent of the plutonium released from normal operations in buildings at Rocky Flats came from the Building 771 stack (effluent duct 771-MAI), and the possible effects of anisokinetic sampling in duct 771-MAI can be summarized as follows:

- With duct velocity 2542 ft min^{-1} and sampling line velocity 1956 ft min^{-1}, as reported in Repository Document RE-1029, the isokinetic ratio is 0.77. This would occur if three 0.25-inch i.d. sample nozzles fed each sampling line, the sampling line i.d. was also 0.25 inch and the sample was drawn at 2 cubic feet per minute through the sampling line. Under these conditions, the measured sample concentration is less than 1 percent higher than the duct concentration for the maximum particle diameter of 0.3 μm, but only 0.2 percent higher than the duct concentration for the median particle diameter of 0.1 μm. If the sampling rate were to drop to 1.4 cubic feet per minute, the isokinetic ratio would be 0.54. The measured sample concentration would then be 2 percent higher than the duct concentration for the maximum particle diameter of 0.3 μm, but only 0.4 percent higher than the duct concentration for the median particle diameter of 0.1 μm.

- If the duct velocity were 2542 ft min^{-1}, as reported in Repository Document RE-1029, and 2 cubic feet per minute were drawn through <u>each</u> sample nozzle, the sampling velocity would be 5867 ft min^{-1}. Under these conditions, the isokinetic ratio is 2.3 and the measured sample concentration is less than 2 percent lower than the duct concentration for the maximum particle diameter of 0.3 μm, and less than 0.5 percent lower than the duct concentration for the median particle diameter of 0.1 μm. If the sampling rate were to drop to 1.4 cubic feet per minute, the isokinetic ratio would be 1.6, and the measured sample concentration would again be less than 2 percent lower than the duct concentration for the maximum particle diameter of 0.3 μm, and less than 0.5 percent lower than the duct concentration for the median particle diameter of 0.1 μm.

- The 771-MAI DAR says the exhaust velocity was 17.1 ft sec^{-1} at a point in the 771 stack where the cross-sectional area is 150.3 ft^2. Using the hydrodynamic continuity equation $Q=AV$, where Q is the flow rate, A is the cross-sectional area of the conduit and V is the flow velocity, the exhaust velocity was 32.1 ft sec^{-1} (1926 ft min^{-1}) at the stack sampling locations where the cross-sectional area of the exhaust duct is 80 ft^2 The sampling nozzle inside diameters are 0.125 inch and each nozzle samples at a rate of 2 cubic feet per minute (Osborne, 1993), resulting in the isokinetic ratio of 12.2 reported in the DAR. Under these conditions, the measured sample concentration is 12 percent lower than the duct concentration for the maximum particle diameter of 0.3 μm, but only 3 percent lower than the duct concentration for the median particle diameter of 0.1 μm.

If the sampling rate were 1.4 cubic feet per minute, the isokinetic ratio is 8.5, the measured sample concentration is 9 percent lower than the duct concentration for the maximum particle diameter of 0.3 μm and 2 percent lower than the duct concentration for the median particle diameter of 0.1 μm.

- Historically, the maximum average flow in the main 771 exhaust duct was about 8.5 million cubic meters per day (3,500 cubic feet per second), corresponding to a duct velocity of 43.4 ft sec^{-1} (2604 ft min^{-1}) at the stack sampling location where the duct cross-sectional area is 80 ft^2. Assuming sample nozzle and sample line inside diameters of 0.125 inch and a sampling rate of 2 cubic feet per minute in each nozzle, the isokinetic ratio is 9. Under these conditions, the measured sample concentration is 12 percent lower than the duct concentration for the maximum particle diameter of 0.3 μm, and 3 percent lower than the duct concentration for the median particle diameters of 0.1 μm. If the sampling rate were 1.4 cubic feet per minute, the isokinetic ratio is 6.3, the measured sample concentration is 9 percent lower than the duct concentrations for the maximum particle diameter of 0.3 μm and 2 percent lower than the duct concentration for the median particle diameter of 0.1 μm.

Duct velocities and sampling probe inside diameters in the six DARs reviewed for buildings other than 771 indicated superisokinetic sampling with the sample concentration underestimating the stack concentration by less than 0.4 percent for 3 μm diameter particles, with a smaller error for 0.1 μm particles. The anisokinetic sampling errors for the duct velocities and sampling probe inside diameters for building other than Building 371 tabulated in Repository Document RE-1029 were 2 percent or less for 0.1 μm particles, the median particle size in Rocky Flats exhausts. For sampling rates between 1.4 and 2 cubic feet per minute, the maximum error for 0.3 μm particles, the approximate upper bound on the particles that pass through HEPA filters, was a measured sample concentration 8 percent higher than the duct concentration.

These results suggest that errors introduced by anisokinetic sampling in the estimation of radionuclide releases from Rocky Flats were of the order of 5 percent or less.

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APPENDIX D

ISOTOPIC COMPOSITION OF ALPHA-EMITTING EFFLUENTS

APPENDIX D

ISOTOPIC COMPOSITION OF ALPHA-EMITTING EFFLUENTS

This appendix discusses the composition of the primary radioactive metals that were used at the Rocky Flats Plant and methods that can be used to characterize the isotopic composition of historical radioactive effluents from the plant.

D.1 Rocky Flats Plutonium

The materials of concern identified in Task 2 of this project included individual isotopes of plutonium, that is, Pu-238, Pu-239, Pu-240, Pu-241, and Pu-242, as well as Am-241. Rocky Flats plutonium has contained the those radionuclides in the proportions reflected in Table D-1. The mass percentages in Table D-1 are averages over a two-year period from July 1976 to July 1978 (USDOE, 1980). The ranges of percentages by mass indicate the variability of isotopic content (Del Pizzo *et al.*, 1970). The isotopic fractions of alpha activity are presented for total long-lived alpha-emitting radioactivity and for Pu-239/240, which are both quantities that have been measured for airborne emissions. As a result, these fractions are useful for translating nonspecific or multiple isotope effluent measurements to releases of specific radionuclides.

Pu-241, which emits beta particles but no alpha particles, typically comprises over 80 percent of the total (alpha plus beta) radioactivity of Rocky Flats plutonium (USDOE, 1980). The multiples of total alpha activity and Pu-239/240 alpha activity that Pu-241 typically equals are useful in estimating quantities of the beta-emitter released based on measured activities.

On weight and alpha activity bases, Pu-239 and Pu-240 would be expected to make up nearly all of the plutonium in Rocky Flats airborne effluents. While alpha spectroscopy to specifically identify alpha emitters was practiced on environmental samples as early as the late 1950s or 1960 (Ray and Hammond, 1960), routine isotopic analyses of effluent sample filters did not start until around 1973. Specific measurement of Pu-239 and Pu-240 in Rocky Flats airborne effluents began in July 1973 (Dow, 1974). The alpha particles emitted from Pu-239 and Pu-240 are so similar in energy that the isotopes cannot be separately quantified by the alpha spectrometric method used to analyze plutonium effluent samples. Because of this, plutonium emissions since 1973 have been reported as "radiochemically determined Pu-239 plus Pu-240."

The contributions of Pu-238 to Rocky Flats plutonium emissions were first reported around 1986; they have been included in DOE's Effluent Information System since 1986 and were first included in Rocky Flats annual environmental reports in 1990.

TABLE D-1

TYPICAL ISOTOPIC COMPOSITION OF ROCKY FLATS PLUTONIUM

Isotope	Percentage by Mass[1]	Range of Mass Percentage[2]	Specific Activity in Rocky Flats Plutonium[1] (Ci g^{-1})	Percentage of Total Alpha Activity[3]	Percentage of Pu-239/240 Alpha Activity[3]
Pu-238	0.01	0.03 - 0.05	0.00171	1.79 - 2.07	2.39
Pu-239	93.79	92.84 - 93.84	0.05834	61.1 - 70.7	81.5
Pu-240	5.80	5.5 - 6.5	0.01322	13.8 - 16.0	18.5
Pu-241	0.36	Not Given	0.37260* (beta activity)	390 - 451* (beta activity)	521** (beta activity)
Pu-242	0.03	Not Given	0.00000118	0.00124 - 0.00143	0.00165
Am-241	Not Given	Not Given	0.00930 - 0.0222[4]	11.3 - 23.2	13 - 31[6]
Plutonium-239 plus Plutonium-240[7]			0.0716	75.0 - 86.7	100
Total of Alpha-Emitting Plutonium Isotopes[7]			0.0733	76.8 - 88.7	102
Total of All Alpha-Emitting Radionuclides[7]			0.0826 - 0.0955	100	115 - 133

* Pu-241 is a beta emitter. It does not emit alpha particles. In Rocky Flats plutonium it emits beta particles at 3.82 to 4.51 times the rate that alpha-emitting nuclides present emit alpha particles.

** Pu-241 present in Rocky Flats plutonium emits beta particles at 5.21 times the rate that the Pu-239 and Pu-240 present emit alpha particles.

REFERENCES/SOURCES:

1 Rockwell, 1989.
2 Del Pizzo *et al.*, 1970.
3 Calculated from the specific activities of the individual nuclides in Column 4.
4 Calculated based on the Column 6 range of Am-241 fraction of Pu-239/240 activity and the Pu-239 and Pu-240 specific activities in Column 4.
5 Calculated based on the Column 4 range of Am-241 specific activity and the sum of alpha-emitting plutonium isotope specific activities from Column 4.
6 Estimated based on Am-241 and Pu-239/240 measurements in Rocky Flats airborne effluents from 1985 through 1989.
7 Values in this row were calculated based on Column 4 specific activity values for the individual radionuclides.

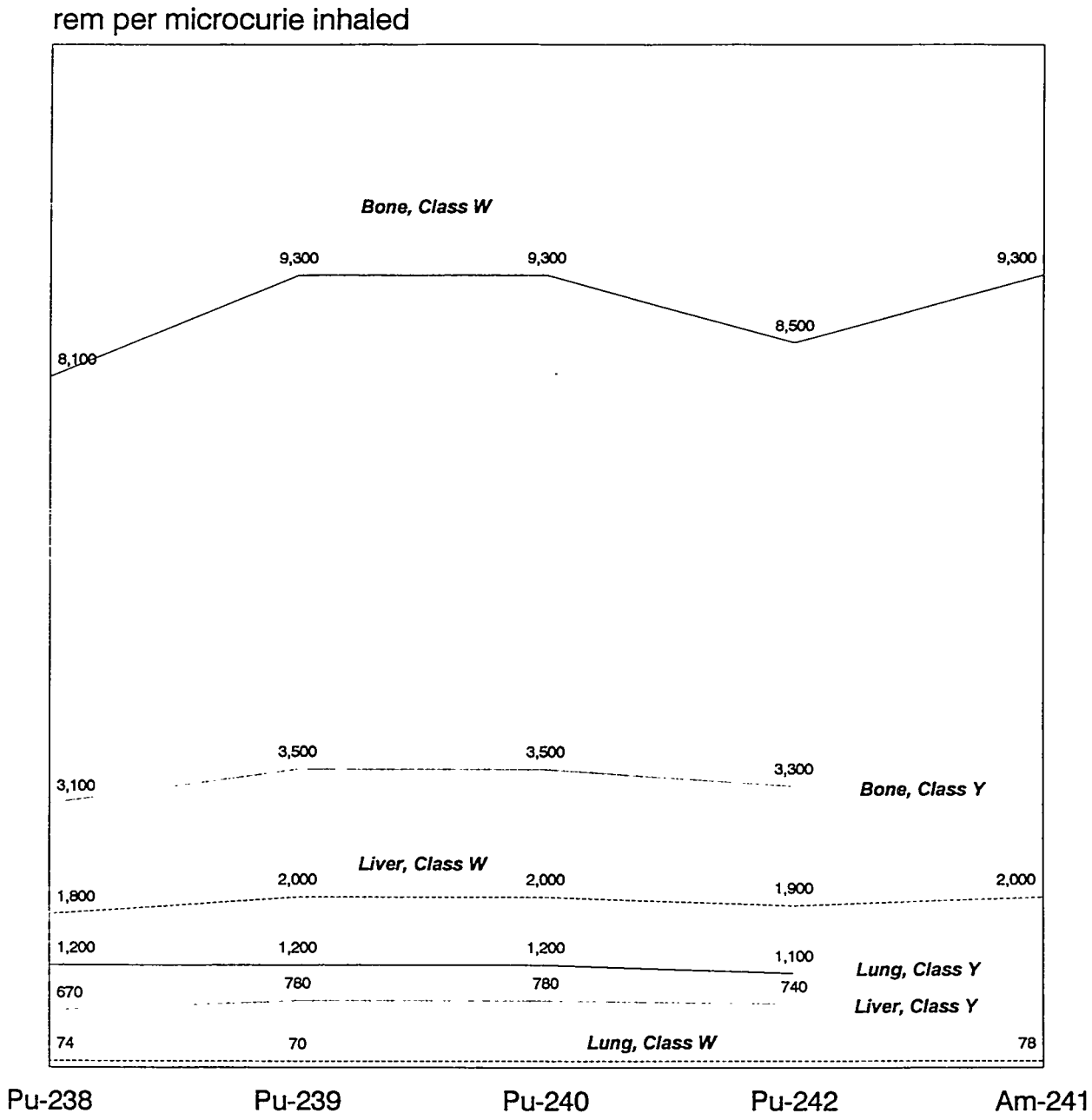
Am-241 is a decay product of Pu-241, and as such has been present in the plutonium handled at Rocky Flats since the early 1950s. The americium to plutonium activity ratio has reportedly ranged from 10 percent to 20 percent (USDOE, 1980). As an emitter of alpha and gamma radiations, americium would have contributed to the total long-lived alpha measurements made of particulate filters from plutonium production areas. Although the alpha spectroscopy procedure used for airborne samples since 1973 has separated plutonium, uranium, and americium content into separate specimens for analysis, Am-241 emissions were not reported until 1985 because of problems with performance of the laboratory method (Hornbacher, 1975-1982). Am-241 release totals were reported in USDOE's Effluent Information System starting in 1985 and in Rocky Flats annual environmental reports since 1988.

In order to translate measurements of total long-lived alpha radioactivity from plutonium facilities and more recent combined Pu-239/240 measurements to emission totals for the specific radionuclides listed in Table D-1, a number of relationships must be characterized:

- The fractions of total long-lived alpha radioactivity from plutonium facilities that were actually Pu-238, Pu-239, Pu-240, Pu-242, and Am-241 must be estimated.
- The ratio of relatively short-lived beta-emitting Pu-241 (13-year half-life) to measured total alpha or Pu-239/240 activities must be determined.
- The relative quantities of Pu-239 and Pu-240 historically present must be estimated to translate combined alpha spectral measurements to individual release totals.

Each of these relationships can be described to some extent based on available site-specific information and generic information characterizing compositions of special nuclear materials historically encountered within the U.S. nuclear weapons complex. For this project, source terms are provided for Pu-241, Am-241, and plutonium alpha activity. Given the relatively similar physical and toxicological properties of the alpha emitting plutonium isotopes, taking the source term analyses to the level of individual alpha-emitting plutonium isotopes would result in little benefit in terms of dose assessment, at the cost of introducing additional uncertainty into the process.

Figures D-1 and D-2 present inhalation and ingestion dose coefficients for the plutonium isotopes and for Am-241 based on ICRP Report 56 (ICRP, 1989) and ICRP Report 30 methodology as applied in DOE/EH-0071 (USDOE, 1988). ICRP Publication 56 presents age-dependent dose coefficients that reflect dose delivered to age 70 from an intake at various ages, while

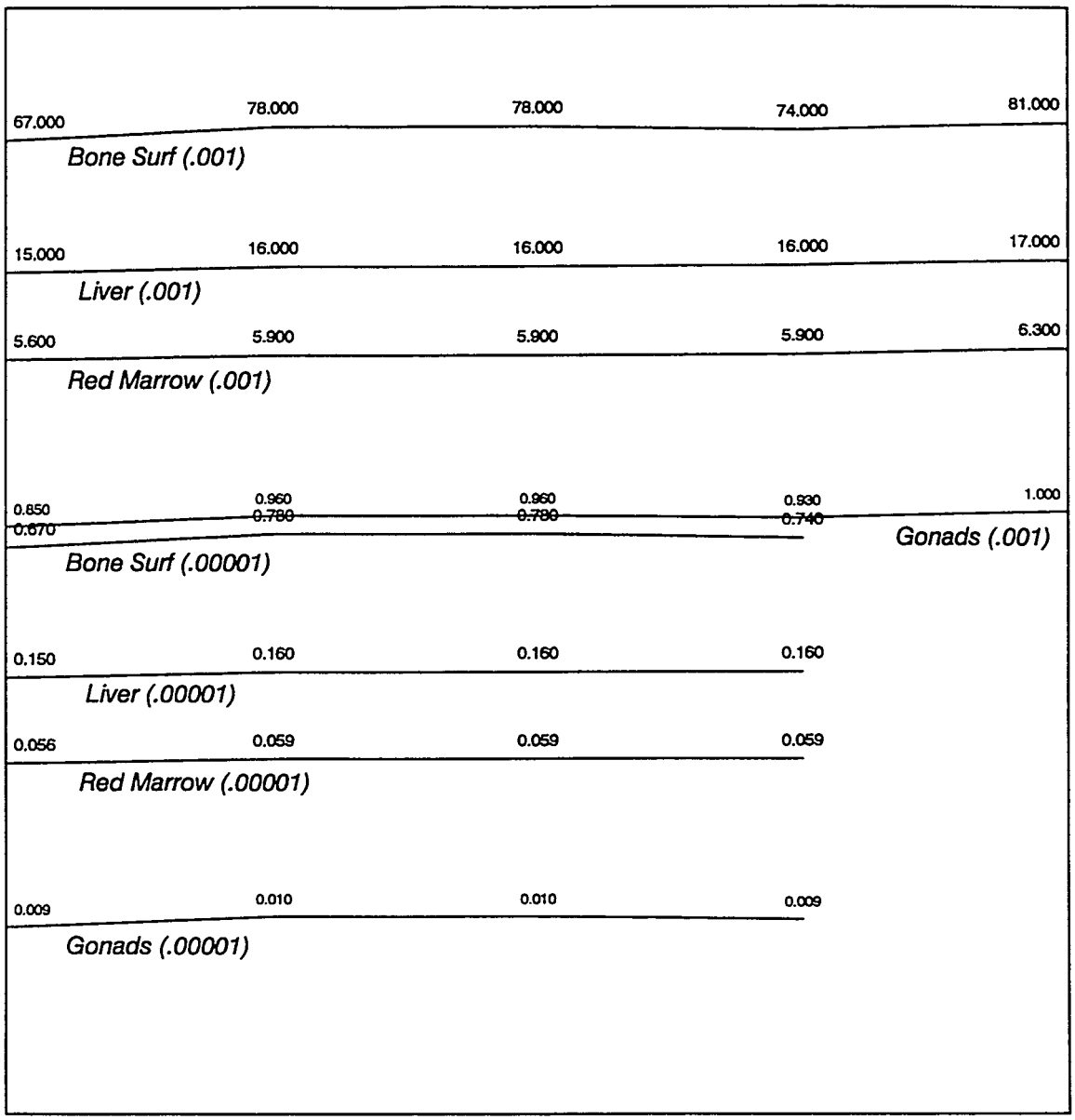


Classes reflect Lung Clearance Half-Times; W = weeks, Y= years

ChemRisk
A Division of McLaren/Hart

FIGURE D-1
INHALATION DOSE FACTORS
FOR PLUTONIUM
AND AMERICIUM

rem per microcurie ingested



Pu-238 Pu-239 Pu-240 Pu-242 Am-241

Numbers in parentheses are gastrointestinal absorption factors.

ChemRisk
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FIGURE D-2
INGESTION DOSE FACTORS
FOR PLUTONIUM
AND AMERICIUM

RKY294T5/D-2 REV 03-03-94

DOE/EH-0071 factors correspond to doses delivered over a 50-year period after intake by an adult. The doses that are calculated using these coefficients are called "committed" doses because they are estimated doses that a person will receive over a stated period (50 or 70 years) in the future after a one-time intake of a radionuclide. The quantity of radionuclide taken into the body is typically stated in units of becquerels (Bq) or curies (Ci), where 1 Bq is one disintegration per second, and 1 Ci equals 3.7×10^{10} disintegrations per second. The doses are also termed "effective" doses because they reflect the fact that a radionuclide taken into the body distributes to various organs, and weighting factors have been incorporated that reflect the relative importance of the impacted organs such that the result represents a total-body dose that represents equivalent risk. The values resulting from use of these coefficients are also called "dose equivalents" because they include application of quality factors that convert from estimates of the amount of energy absorbed in tissues (absorbed doses) to values that reflect the varying potential for damage associated with the different radiations (alpha particles, beta particles, gamma rays, etc.). Dose equivalents are stated in terms of sieverts (Sv) or rem, with one sievert equal to 100 rem.

The ICRP Task Group chose to continue to base their dose coefficients on the assumption that americium behaves like plutonium in adult humans. Figures D-1 and D-2 illustrate that there is relatively little variation in dose coefficients among the isotopes of plutonium and Am-241. While source terms used in the dose assessment phase of this project did not include specific release estimates for every radionuclide expected to be contained in Rocky Flats plutonium, the steps that can be taken to estimate historical emissions of the individual radionuclides listed in Table D-1 are described in this appendix.

D.2 Assignment of Plutonium Facility Emissions to Specific Radionuclides

An important step in the assignment of measured nonspecific radioactivity emissions (total long-lived alpha radioactivity) from plutonium facilities to the appropriate individual radionuclides is the characterization of Am-241 content. Am-241 is a decay product of plutonium and as such has been present at Rocky Flats since the early 1950s. It is an undesirable contaminant in weapons-grade plutonium and is present in increasing amounts as plutonium ages.

At Rocky Flats, americium was emitted in particulate form and was therefore collected by the exhaust sampling systems much like plutonium and uranium. The americium content of airborne effluents was not specifically measured for a large portion of the operational history of the Rocky Flats Plant. Am-241 release totals for airborne effluents were reported for calendar years 1985 through 1989 (EG&G, 1991).

Am-241 is formed when Pu-241 decays by emitting a beta particle. Plutonium is formed in a reactor when U-238 absorbs neutrons to form Pu-239, which in turn can absorb additional neutrons to form the heavier "contaminant" isotopes of plutonium, including Pu-241. The extent to which these contaminant nuclides build up is determined by the length of time the fuel remains in the reactor.

Chemical purification of reactor fuel that took place at the plutonium production sites (Hanford and Savannah River) separated the plutonium from americium present at that point in time, as did the peroxide precipitation step of the Rocky Flats plutonium recovery process and the molten salt extraction process used at Rocky Flats.

Weapons-grade plutonium like that used at Rocky Flats contains about 0.3 mass percent Pu-241 (Rockwell, 1985) and initially contains about 0.0001 percent Am-241 (Krey *et al.*, 1976). The Pu-241, however, decays relatively quickly (with a 13-year half-life) to form Am-241 as time passes after purification.

The Final Environmental Impact Statement for the Rocky Flats Plant states that the americium to plutonium activity ratio has ranged from 10 percent to 20 percent, but also states that this ratio can change during processing that separates americium from plutonium. In a referenced study, when americium was measured in a facility where it was handled in the chemically separated form, the americium to plutonium ratio in effluents did not exceed 0.42 (USDOE, 1980).

Based on reported Rocky Flats airborne release totals for calendar years 1985 and 1987 through 1989 (EG&G, 1991) and independently reconstructed emission totals for 1986, the ratios of Am-241 released to Pu-239/240 released were as shown in Table D-2. It is therefore estimated that airborne Am-241 emissions for each year from 1953 to 1984 were between 13 and 31 percent of the plutonium-239/240 release total for the same year.

TABLE D-2

ACTIVITY RATIOS OF AM-241 TO PU-239/240 IN MONITORED ROCKY FLATS EFFLUENTS

Calendar Year	Ratio of Airborne Am-241 Released to Pu-239/240 Released
1985	22%
1986	31%
1987	21%
1988	13%
1989	24%
Average	22%

References: Independently reconstructed emissions for 1986, USDOE Effluent Information System (EG&G, 1991) for other years.

Long-lived alpha activity can be attributed to the various isotopes according to typical activity fractions such as those contained in Table D-1. The following steps can be taken to estimate the releases for the radionuclides listed in Table D-1:

For the period from 1953 through 1973 (based on nonspecific alpha measurements):

Emissions of the following radionuclides are estimated by multiplying the annual total long-lived alpha release total times the following values of Percentage of Total Alpha Activity from Table D-1. Because the americium content of total alpha-emitting radioactivity is specified as a range, all of the values derived from total alpha activity are also ranges.

Pu-238	0.0179 to 0.0207
Pu-239	0.611 to 0.707
Pu-240	0.138 to 0.160
Pu-241	3.90 to 4.51
Pu-242	0.0000124 to 0.0000143
Am-241	0.113 to 0.232

For the period from 1974 through 1989, except Am-241 1985-89 and Pu-238 1986-89 (based on Pu-239/240 measurements):

Emissions of the following radionuclides are estimated by multiplying the annual total long-lived alpha release total times the following values of Percentage of Pu-239/240 Alpha Activity from Table D-1. The americium content of Rocky Flats plutonium has been specified as a range of percentages of Pu-239/240 content. As a result, two multipliers are specified which define ranges of annual Am-241 emissions based on measured Pu-239/240 releases.

Pu-238	0.0239
Pu-239	0.815
Pu-240	0.185
Pu-241	5.21
Pu-242	0.0000165
Am-241	0.13 to 0.31

For the period 1985 through 1989 (based on specific Am-241 data):

Am-241 emissions are taken to be equal to the reported results of alpha spectrometric analyses of americium radiochemically separated from airborne particle samples.

For the period from 1986 through 1989 (based on specific Pu-238 data):

Pu-238 emissions are taken to be equal to the reported results of alpha spectrometric analyses of plutonium radiochemically separated from airborne particle samples.

The results of this process of assignment of activity released from plutonium facilities to specific radionuclides of concern are presented in Table D-3.

D.3 Rocky Flats Uranium

The materials of concern identified in Task 2 of this project included individual isotopes of uranium, that is, U-233, U-234, U-235, and U-238. Uranium has historically been processed and handled at Rocky Flats in two forms: enriched and depleted. Tables D-4 and D-5 show the reported activity fractions of the different radionuclides present in Rocky Flats enriched uranium and depleted uranium (USDOE, 1980).

TABLE D-3: ASSIGNMENT OF PLUTONIUM ACTIVITY TO SPECIFIC RADIONUCLIDES

Year	Plutonium-238 (μCi)	Plutonium-239 (μCi)	Plutonium-240 (μCi)	Plutonium-241 (μCi)	Plutonium-242 (μCi)	Americium-241 (μCi)
1953	2.9e-02 to 3.3e-02*	9.8e-01 to 1.1e+00	2.2e-01 to 2.6e-01	6.2e+00 to 7.2e+00	2.0e-05 to 2.3e-05	1.8e-01 to 3.7e-01
1954	9.5e-01 to 1.1e+00	3.2e+01 to 3.7e+01	7.3e+00 to 8.5e+00	2.1e+02 to 2.4e+02	6.6e-04 to 7.6e-04	6.0e+00 to 1.2e+01
1955	1.1e+00 to 1.2e+00	3.6e+01 to 4.2e+01	8.1e+00 to 9.4e+00	2.3e+02 to 2.7e+02	7.3e-04 to 8.4e-04	6.7e+00 to 1.4e+01
1956	3.4e+00 to 3.9e+00	1.2e+02 to 1.3e+02	2.6e+01 to 3.0e+01	7.4e+02 to 8.6e+02	2.4e-03 to 2.7e-03	2.1e+01 to 4.4e+01
1957	2.1e+02 to 2.5e+02	7.3e+03 to 8.5e+03	1.7e+03 to 1.9e+03	4.7e+04 to 5.4e+04	1.5e-01 to 1.7e-01	1.4e+03 to 2.8e+03
1958	4.5e+01 to 5.2e+01	1.5e+03 to 1.8e+03	3.5e+02 to 4.0e+02	9.8e+03 to 1.1e+04	3.1e-02 to 3.6e-02	2.8e+02 to 5.8e+02
1959	2.0e+01 to 2.3e+01	6.7e+02 to 7.8e+02	1.5e+02 to 1.8e+02	4.3e+03 to 5.0e+03	1.4e-02 to 1.6e-02	1.2e+02 to 2.6e+02
1960	2.0e+01 to 2.3e+01	6.7e+02 to 7.8e+02	1.5e+02 to 1.8e+02	4.3e+03 to 5.0e+03	1.4e-02 to 1.6e-02	1.2e+02 to 2.6e+02
1961	2.1e+01 to 2.5e+01	7.3e+02 to 8.5e+02	1.7e+02 to 1.9e+02	4.7e+03 to 5.4e+03	1.5e-02 to 1.7e-02	1.4e+02 to 2.8e+02
1962	4.5e+01 to 5.2e+01	1.5e+03 to 1.8e+03	3.5e+02 to 4.0e+02	9.8e+03 to 1.1e+04	3.1e-02 to 3.6e-02	2.8e+02 to 5.8e+02
1963	5.4e+01 to 6.2e+01	1.8e+03 to 2.1e+03	4.1e+02 to 4.8e+02	1.2e+04 to 1.4e+04	3.7e-02 to 4.3e-02	3.4e+02 to 7.0e+02
1964	4.1e+01 to 4.8e+01	1.4e+03 to 1.6e+03	3.2e+02 to 3.7e+02	9.0e+03 to 1.0e+04	2.9e-02 to 3.3e-02	2.6e+02 to 5.3e+02
1965	9.5e+01 to 1.1e+02	3.2e+03 to 3.7e+03	7.3e+02 to 8.5e+02	2.1e+04 to 2.4e+04	6.6e-02 to 7.6e-02	6.0e+02 to 1.2e+03
1966	4.7e+00 to 5.4e+00	1.6e+02 to 1.8e+02	3.6e+01 to 4.2e+01	1.0e+03 to 1.2e+03	3.2e-03 to 3.7e-03	2.9e+01 to 6.0e+01
1967	5.9e+00 to 6.8e+00	2.0e+02 to 2.3e+02	4.6e+01 to 5.3e+01	1.3e+03 to 1.5e+03	4.1e-03 to 4.7e-03	3.7e+01 to 7.7e+01
1968	7.2e+00 to 8.3e+00	2.4e+02 to 2.8e+02	5.5e+01 to 6.4e+01	1.6e+03 to 1.8e+03	5.0e-03 to 5.7e-03	4.5e+01 to 9.3e+01
1969	2.0e+01 to 2.3e+01	6.7e+02 to 7.8e+02	1.5e+02 to 1.8e+02	4.3e+03 to 5.0e+03	1.4e-02 to 1.6e-02	1.2e+02 to 2.6e+02
1970	5.5e+00 to 6.4e+00	1.9e+02 to 2.2e+02	4.3e+01 to 5.0e+01	1.2e+03 to 1.4e+03	3.8e-03 to 4.4e-03	3.5e+01 to 7.2e+01
1971	1.1e+00 to 1.3e+00	3.7e+01 to 4.3e+01	8.4e+00 to 9.8e+00	2.4e+02 to 2.8e+02	7.6e-04 to 8.7e-04	6.9e+00 to 1.4e+01
1972	9.0e-01 to 1.0e+00	3.1e+01 to 3.5e+01	6.9e+00 to 8.0e+00	2.0e+02 to 2.3e+02	6.2e-04 to 7.2e-04	5.7e+00 to 1.2e+01
1973	9.1e-01 to 1.1e+00	3.1e+01 to 3.6e+01	7.0e+00 to 8.2e+00	2.0e+02 to 2.3e+02	6.3e-04 to 7.3e-04	5.8e+00 to 1.2e+01
1974	2.3e+01	7.8e+02	1.8e+02	5.0e+03	1.6e-02	1.2e+02 to 3.0e+02
1975	2.4e-01	8.2e+00	1.9e+00	5.2e+01	1.7e-04	1.3e+00 to 3.1e+00
1976	9.6e-02	3.3e+00	7.4e-01	2.1e+01	6.6e-05	5.2e-01 to 1.2e+00
1977	9.6e-02	3.3e+00	7.4e-01	2.1e+01	6.6e-05	5.2e-01 to 1.2e+00
1978	6.7e-02	2.3e+00	5.2e-01	1.5e+01	4.6e-05	3.6e-01 to 8.7e-01
1979	1.3e-01	4.5e+00	1.0e+00	2.9e+01	9.1e-05	7.2e-01 to 1.7e+00
1980	2.9e-01	9.8e+00	2.2e+00	6.3e+01	2.0e-04	1.6e+00 to 3.7e+00
1981	2.0e-01	6.7e+00	1.5e+00	4.3e+01	1.4e-04	1.1e+00 to 2.5e+00
1982	4.8e-01	1.6e+01	3.7e+00	1.0e+02	3.3e-04	2.6e+00 to 6.2e+00
1983	1.9e+00	6.4e+01	1.4e+01	4.1e+02	1.3e-03	1.0e+01 to 2.4e+01
1984	1.9e+00	6.4e+01	1.4e+01	4.1e+02	1.3e-03	1.0e+01 to 2.4e+01
1985	2.2e-01	7.5e+00	1.7e+00	4.8e+01	1.5e-04	2.0e+00
1986	9.2e-01	2.4e+01	5.4e+00	1.5e+02	4.8e-04	4.8e+00
1987	5.6e-01	1.2e+01	2.8e+00	7.8e+01	2.5e-04	3.2e+00
1988	3.9e-01	1.2e+01	2.8e+00	7.8e+01	2.5e-04	2.0e+00
1989	2.0e-01	3.7e+00	8.3e-01	2.3e+01	7.4e-05	1.1e+00

* Note on Scientific Notation: 3.3e-02 equal 3.3×10^{-2} or 0.033.

TABLE D-4

ACTIVITY FRACTIONS OF ROCKY FLATS ENRICHED URANIUM

Nuclide	Percentage by Weight	Percentage of Alpha Activity	Percentage of Beta Activity
Th-231		-	99.1
Th-234		-	0.892
U-234		96.5	-
U-235	over 93	3.11	-
U-236		0.389	-
U-238		0.0280	-

TABLE D-5

ACTIVITY FRACTIONS OF ROCKY FLATS DEPLETED URANIUM

Nuclide	Percentage by Weight	Percentage of Alpha Activity	Percentage of Beta Activity
Th-231		-	1.42
Th-234		-	98.6
U-234		9.69	-
U-235	less than 0.7	1.28	-
U-238		89.0	-

The specific activity (curies per gram) of enriched uranium is approximately 6000 times lower than that of plutonium. Although enriched uranium is more than 93 percent U-235 by weight, U-234 constitutes over 96 percent of its alpha radioactivity. U-238 constitutes almost 90 percent of the alpha radioactivity of depleted uranium. Th-231 and Th-234 are beta-emitting decay products of U-235 and U-238, respectively. These thorium isotopes decay rather rapidly, with half-lives of 26 hours and 24 days, respectively.

On an alpha activity basis, U-234 and U-235 would be expected to make up nearly all of the uranium alpha activity in Rocky Flats airborne effluents from enriched uranium facilities. Likewise, U-238 and U-234 would be expected to comprise most of the uranium in effluents from depleted uranium facilities.

Routine isotopic analyses of effluent sample filters did not start until around 1973. However, reporting of total long-lived alpha radioactivity continued for uranium facilities until approximately 1978. Emissions from uranium facilities were "radiochemically determined as U-233, U-234, and U-238" for the first time in the 1978 Rocky Flats Plant annual environmental report (Rockwell, 1979). By calendar year 1979, there was reporting of both enriched and depleted uranium from essentially all uranium facilities (EG&G, 1991).

The analytical procedure used for chemical separation of uranium from other alpha emitters has historically involved addition of nonindigenous U-232 as an internal standard to gauge the yield of the separation process. Because U-232 emits alpha particles that interfere with the specific identification of U-235, the indistinguishable U-233/234 pair have been used to indicate the magnitude of enriched uranium releases in the same manner as U-238 has been used as an indicator of depleted uranium emissions. Although U-233 was indistinguishable from U-234 in measurement of uranium effluents, it was not a measurable constituent of the depleted or enriched uranium processed at Rocky Flats. All indications from review of historical operations at Rocky Flats are that no significant amounts of U-233 were processed or handled. As a result, no portion of measured emissions from the plant are attributed to U-233 in this analysis.

To translate measurements of total long-lived alpha radioactivity from uranium facilities and more recent U-233/234 and U-238 measurements to emission totals for the specific alpha-emitting radionuclides listed in Tables D-4 and D-5, a number of relationships must be characterized:

- The fractions of total long-lived alpha radioactivity from uranium facilities that were actually U-234, U-235, U-236, and U-238 must be estimated. The fractions might vary between periods of significant enriched uranium (oralloy line) recovery and machining compared to periods of large depleted uranium projects (e.g., M1A1 tank armor manufacturing in Building 883).

- The ratios of indicator isotope quantities (i.e., U-233/234 and U-238) to the quantities of other isotopes in enriched and depleted uranium (e.g., U-235 and U-236) must be characterized.

Each of these relationships can be described to some extent based on available site-specific information and generic information characterizing compositions of special nuclear materials historically encountered within the U.S. nuclear weapons complex. To support the dose assessment portions of this project, source terms were provided for enriched and depleted uranium. As with plutonium, given the relative similarities of the physical and toxicological properties of the uranium isotopes in question, taking the source term analyses to the level of individual radionuclides would result in little benefit in terms of dose assessment and would introduce additional uncertainty into the process. Figures D-3 and D-4 present inhalation and ingestion dose coefficients for the uranium isotopes of interest to this project from DOE/EH-0071 (USDOE, 1988). DOE/EH-0071 factors correspond to doses delivered over a 50-year period after intake by an adult. ICRP 56, which presents age-dependent dose coefficients, does not include uranium isotopes.

Figures D-3 and D-4 illustrate that there is relatively little variation in dose coefficients among the uranium isotopes. While the source terms used in the dose assessment phase of this project did not include specific release estimates for every radionuclide expected to be contained in Rocky Flats uranium, the steps that can be taken to estimate historical emissions of the individual radionuclides listed in Table D-4 and D-5 are described in this appendix.

D.4 Assignment of Uranium Facility Emissions to Specific Radionuclides

Assignment of alpha-emitting radioactivity released from facilities that processed and handled enriched and depleted uranium to specific radionuclides requires a number of approaches for different time periods because of the different forms of emission data that are available. The isotopic composition of the estimated release totals are based on observed measurements and the activity fractions presented in Tables D-4 and D-5.

Depleted Uranium from 1953-1977 (based on nonspecific alpha measurements):

Uranium isotopic releases are calculated by multiplying the measured total long-lived alpha activity released from depleted uranium facilities by the following, based on the fraction of depleted uranium alpha activity:

1.	U-234	0.0969
2.	U-235	0.0128
3.	U-238	0.890

rem per microcurie inhaled

	U-233	U-234	U-235	U-238
Lung, Class Y	1,100	1,100	1,000	1,000
Lung, Class W	59	59	56	52
Bone Surf, Class D	41	41	37	36
Kidneys, Class D	17	17	16	15
Red Marrow, Class D	2.6	2.6	2.4	2.4
Lung, Class D	1.2	1.2	1.1	1

"Classes" reflect Lung Clearance Half-Times; D = days, W = weeks, Y= years

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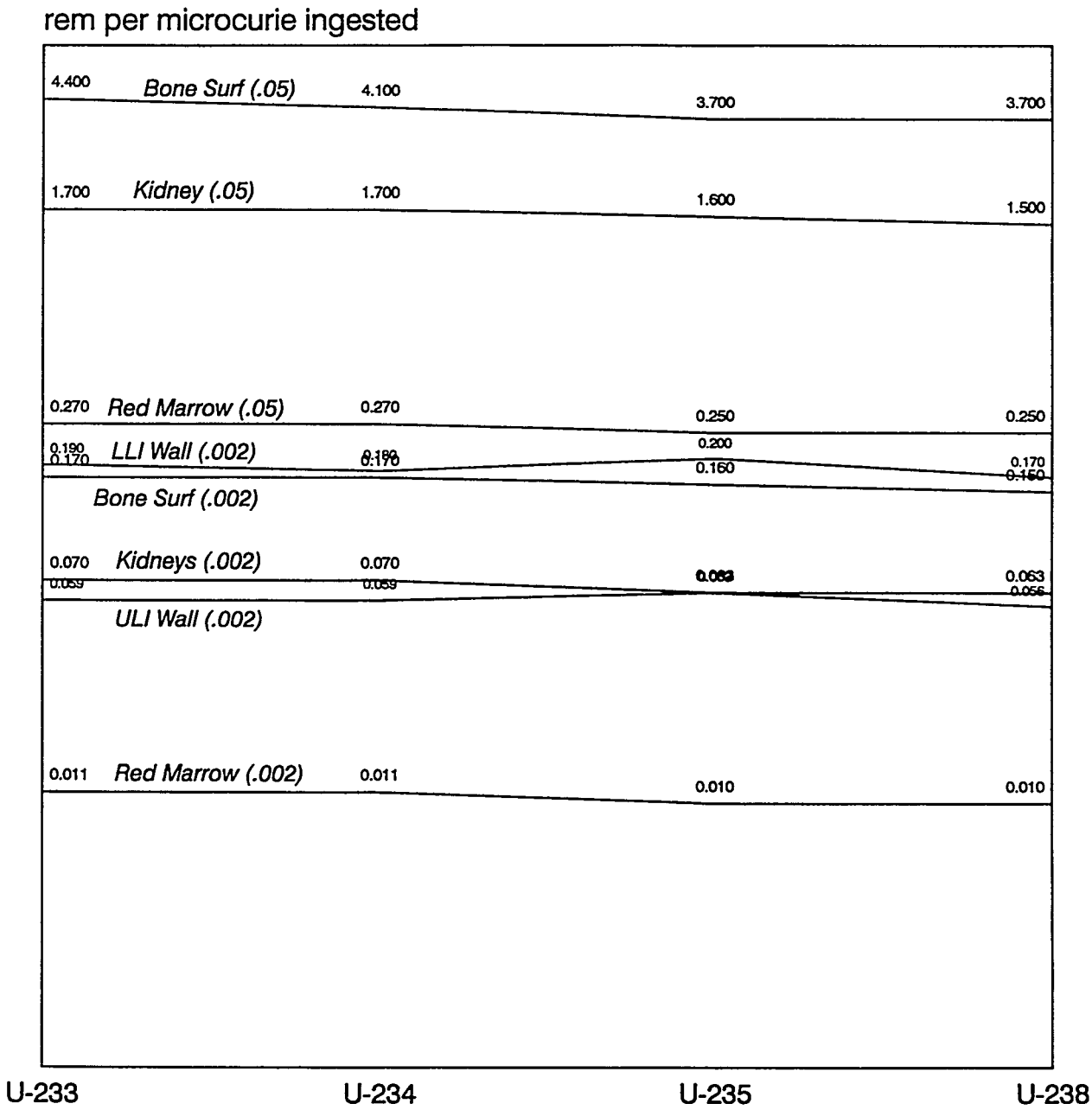
FIGURE D-3
INHALATION DOSE FACTORS
FOR URANIUM

RKY294T5/D-3 REV 03-03-94



Numbers in parentheses are gastrointestinal absorption factors. ULI and LLI are upper and lower large intestines.

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FIGURE D-4
INGESTION DOSE FACTORS
FOR URANIUM

RKY294T5/D-4 REV: 03-03-94

Enriched Uranium from 1953-1977 (based on nonspecific alpha measurements):

Uranium isotopic releases are calculated by multiplying the measured total long-lived alpha activity released from enriched uranium facilities by the following, based on the fraction of enriched uranium alpha activity:

1.	U-234	0.965
2.	U-235	0.0311
3.	U-236	0.00389
4.	U-238	0.000280

Depleted Uranium Emissions for 1978-1980 and for 1985-1989 (based on alpha spectrometric measurements of U-238):

1. U-234 releases are calculated by multiplying the reported U-238 site emission total by 0.109, the ratio of U-234 alpha activity to U-238 alpha activity in Rocky Flats depleted uranium (see Table D-5).

2. U-235 releases are calculated by multiplying the reported U-238 site emission total by 0.014, the ratio of U-235 alpha activity to U-238 alpha activity in Rocky Flats depleted uranium (see Table D-5).

3. U-238 releases are taken to be equal to those calculated based on U-238 measurements.

Enriched Uranium Emissions for 1978-1980 and for 1985-1989 (based on alpha spectrometric measurements of U-233/234):

1. U-234 releases are taken to be equal to the measured U-233/234 emissions.

2. U-235, U-236, and U-238 releases are calculated by multiplying the reported U-233/234 site emission total by the following, based on ratios of their activities to U-234 alpha activity in Rocky Flats enriched uranium:

a.	U-235	0.0322
b.	U-236	0.00403
c.	U-238	0.000290

Uranium Emissions for 1981-1984
(based on alpha spectrometric measurements of U-233, U-234, U-238):

For calendar years 1981 through 1984, Rocky Flats uranium emissions were reported only as total uranium emissions, based on alpha spectral measurements of U-233, U-234, and U-238. Separate depleted (or U-238) and enriched (or U-233/234) results were not reported. To estimate releases of specific uranium isotopes for these years, the following calculations were performed:

1. Reported enriched and depleted uranium emission totals for the period from 1978 to 1980, reconstructed uranium releases from 1984 and 1986, and reported emissions for 1985 and 1987 through 1989 were used to calculate average fractions of Rocky Flats uranium emissions that were in the enriched and depleted forms. The historical fractions of airborne depleted and enriched uranium emissions are depicted in Figure D-17. These calculations yielded 0.6 as the average depleted fraction and 0.4 as the average enriched fraction of total uranium alpha activity.

2. Reported combined uranium emission totals based on U-233/234 and U-238 measurements were multiplied by the enriched and depleted fractions to estimate enriched and depleted uranium release totals for the years in question. Results are as follows:

Year	Reported Total Uranium Release (μCi)	Calculated Enriched Uranium Release (μCi)	Calculated Depleted Uranium Release (μCi)
1981	29.9	12	18
1982	30.9	12	19
1983	51.0	20	31
1984	34.5	14	21

Depleted Uranium Facilities:

3. U-234, U-235, and U-238 release estimates were calculated by multiplying the estimated depleted uranium component of the reported U-233/234 plus U-238 site emission total (from Step 2 above) by the following, based on the ratios of their alpha activities to U-234 plus U-238 alpha activity in Rocky Flats depleted uranium:

1.	U-234	0.0982
2.	U-235	0.0130
3.	U-238	0.902

Enriched Uranium Facilities:

4. U-234, U-235, U-236, and U-238 release estimates were calculated by multiplying the estimated enriched uranium component of the reported U-233/234 plus U-238 site emission total (from Step 2 above) by the following, based on the ratio of their alpha activities to U-234 plus U-238 alpha activity in Rocky Flats enriched uranium:

1.	U-234	0.9997
2.	U-235	0.0322
3.	U-236	0.00403
4.	U-238	0.000290

The results of this process for assignment of activity released from uranium facilities to specific radionuclides of concern are presented in Table D-6.

TABLE D-6: ASSIGNMENT OF URANIUM ACTIVITY TO SPECIFIC RADIONUCLIDES

Year	Depleted Uranium (μCi)			Enriched Uranium (μCi)				Total Uranium (Depleted plus Enriched; μCi)			
	U-234	U-235	U-238	U-234	U-235	U-236	U-238	U-234	U-235	U-236	U-238
1953	1.2e+02 *	1.5e+01	1.1e+03	8.8e+00	2.8e-01	3.5e-02	2.5e-03	1.3e+02	1.6e+01	3.5e-02	1.1e+03
1954	1.2e+02	1.5e+01	1.1e+03	8.8e+00	2.8e-01	3.5e-02	2.5e-03	1.3e+02	1.6e+01	3.5e-02	1.1e+03
1955	2.0e+02	2.7e+01	1.9e+03	7.1e+01	2.3e+00	2.9e-01	2.1e-02	2.7e+02	2.9e+01	2.9e-01	1.9e+03
1956	9.7e+01	1.3e+01	8.9e+02	1.1e+03	3.4e+01	4.3e+00	3.1e-01	1.2e+03	4.7e+01	4.3e+00	8.9e+02
1957	7.1e+01	9.3e+00	6.5e+02	3.5e+02	1.1e+01	1.4e+00	1.0e-01	4.2e+02	2.0e+01	1.4e+00	6.5e+02
1958	1.6e+02	2.0e+01	1.4e+03	3.0e+02	9.6e+00	1.2e+00	8.7e-02	4.5e+02	3.0e+01	1.2e+00	1.4e+03
1959	2.6e+01	3.5e+00	2.4e+02	5.2e+02	1.7e+01	2.1e+00	1.5e-01	5.5e+02	2.0e+01	2.1e+00	2.4e+02
1960	3.4e+01	4.5e+00	3.1e+02	8.3e+02	2.7e+01	3.3e+00	2.4e-01	8.6e+02	3.1e+01	3.3e+00	3.1e+02
1961	5.0e+01	6.7e+00	4.6e+02	4.6e+02	1.5e+01	1.9e+00	1.3e-01	5.1e+02	2.2e+01	1.9e+00	4.6e+02
1962	3.6e+01	4.7e+00	3.3e+02	2.4e+02	7.8e+00	9.7e-01	7.0e-02	2.8e+02	1.3e+01	9.7e-01	3.3e+02
1963	4.7e+01	6.3e+00	4.4e+02	3.2e+02	1.0e+01	1.3e+00	9.2e-02	3.7e+02	1.7e+01	1.3e+00	4.4e+02
1964	2.3e+01	3.1e+00	2.1e+02	1.8e+02	5.9e+00	7.4e-01	5.3e-02	2.1e+02	9.0e+00	7.4e-01	2.1e+02
1965	2.7e+01	3.6e+00	2.5e+02	1.8e+02	5.9e+00	7.4e-01	5.3e-02	2.1e+02	9.5e+00	7.4e-01	2.5e+02
1966	1.4e+01	1.8e+00	1.2e+02	2.2e+02	7.2e+00	8.9e-01	6.4e-02	2.4e+02	8.9e+00	8.9e-01	1.2e+02
1967	1.4e+01	1.8e+00	1.2e+02	1.1e+02	3.4e+00	4.3e-01	3.1e-02	1.2e+02	5.2e+00	4.3e-01	1.2e+02
1968	1.4e+01	1.8e+00	1.2e+02	1.5e+02	5.0e+00	6.2e-01	4.5e-02	1.7e+02	6.8e+00	6.2e-01	1.2e+02
1969	1.6e+01	2.0e+00	1.4e+02	4.8e+01	1.6e+00	1.9e-01	1.4e-02	6.4e+01	3.6e+00	1.9e-01	1.4e+02
1970	1.8e+01	2.4e+00	1.7e+02	6.2e+01	2.0e+00	2.5e-01	1.8e-02	8.0e+01	4.4e+00	2.5e-01	1.7e+02
1971	5.6e+00	7.4e-01	5.2e+01	4.0e+01	1.3e+00	1.6e-01	1.1e-02	4.5e+01	2.0e+00	1.6e-01	5.2e+01
1972	4.1e+00	5.4e-01	3.7e+01	3.9e+00	1.2e-01	1.6e-02	1.1e-03	7.9e+00	6.6e-01	1.6e-02	3.7e+01
1973	5.2e+00	6.9e-01	4.8e+01	1.2e+01	3.7e-01	4.7e-02	3.4e-03	1.7e+01	1.1e+00	4.7e-02	4.8e+01
1974	8.7e-01	1.2e-01	8.0e+00	2.6e+01	8.4e-01	1.1e-01	7.6e-03	2.7e+01	9.5e-01	1.1e-01	8.0e+00
1975	2.7e+00	3.6e-01	2.5e+01	2.7e+01	8.7e-01	1.1e-01	7.8e-03	3.0e+01	1.2e+00	1.1e-01	2.5e+01
1976	1.2e+00	1.5e-01	1.1e+01	1.5e+01	5.0e-01	6.2e-02	4.5e-03	1.7e+01	6.5e-01	6.2e-02	1.1e+01
1977	1.8e+00	2.4e-01	1.7e+01	2.0e+01	6.5e-01	8.2e-02	5.9e-03	2.2e+01	9.0e-01	8.2e-02	1.7e+01
1978	3.6e+00	4.6e-01	3.3e+01	2.1e+01	6.8e-01	8.5e-02	6.1e-03	2.5e+01	1.1e+00	8.5e-02	3.3e+01
1979	2.8e+00	3.6e-01	2.6e+01	9.2e+00	3.0e-01	3.7e-02	2.7e-03	1.2e+01	6.6e-01	3.7e-02	2.6e+01
1980	1.6e+00	2.1e-01	1.5e+01	1.5e+01	4.8e-01	6.0e-02	4.4e-03	1.7e+01	6.9e-01	6.0e-02	1.5e+01
1981	1.8e+00	2.3e-01	1.6e+01	1.2e+01	3.9e-01	4.8e-02	3.5e-03	1.4e+01	6.2e-01	4.8e-02	1.6e+01
1982	1.8e+00	2.4e-01	1.7e+01	1.2e+01	4.0e-01	5.0e-02	3.6e-03	1.4e+01	6.4e-01	5.0e-02	1.7e+01
1983	3.0e+00	4.0e-01	2.8e+01	2.0e+01	6.6e-01	8.2e-02	5.9e-03	2.3e+01	1.1e+00	8.2e-02	2.8e+01
1984	2.0e+00	2.7e-01	1.9e+01	1.4e+01	4.4e-01	5.6e-02	4.0e-03	1.6e+01	7.1e-01	5.6e-02	1.9e+01
1985	4.3e+00	5.5e-01	3.9e+01	7.9e+00	2.5e-01	3.2e-02	2.3e-03	1.2e+01	8.0e-01	3.2e-02	3.9e+01
1986	3.2e-01	4.1e-02	2.9e+00	1.1e+01	3.5e-01	4.4e-02	3.2e-03	1.1e+01	3.9e-01	4.4e-02	2.9e+00
1987	1.3e+00	1.7e-01	1.2e+01	4.6e+00	1.5e-01	1.9e-02	1.3e-03	5.9e+00	3.2e-01	1.9e-02	1.2e+01
1988	1.0e+00	1.3e-01	9.3e+00	2.6e+00	8.4e-02	1.0e-02	7.5e-04	3.6e+00	2.1e-01	1.0e-02	9.3e+00
1989	2.7e-01	3.5e-02	2.5e+00	5.2e+00	1.7e-01	2.1e-02	1.5e-03	5.5e+00	2.0e-01	2.1e-02	2.5e+00

* Note on Scientific Notation: 1.2e+02 equals 1.2×10^{2} or 120.

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APPENDIX E

DETAILED DOCUMENT REVIEW OF ORGANIC SOLVENT EMISSIONS

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DETAILED DOCUMENT REVIEW OF ORGANIC SOLVENT EMISSIONS

The material in this appendix describes the detailed review performed on documentation relating to the emission of organic solvents of concern. The review included the thorough evaluation of the Air Pollution Emission Notice (APEN) documents including various calculation checks and the review of other documentation relevant to estimating emissions from Rocky Flats. Each of the organic solvents of concern is discussed in the following sections.

E.1 Carbon Tetrachloride Emission Evaluation

Information sources relevant to development of source terms for carbon tetrachloride emissions includes four APEN reports and seven other technical reports pertaining to Rocky Flats emissions of the chemical. These information sources are discussed and evaluated.

E.1.1 APEN Resources

Four APEN reports were identified that document carbon tetrachloride use and emissions. These reports correspond to Buildings 707, 776/777, 460, and 881. Associated buildings that APEN reports did not identify as carbon tetrachloride users were not considered further unless some other resource(s) indicated differently. The APEN report for Building 774 was added to the list for evaluation of carbon tetrachloride emissions when non-APEN resources indicated emissions from Building 774. Building 774 handles carbon tetrachloride contained in wastes received from Buildings 707 and 776/777.

APEN reports with identified carbon tetrachloride uses were reviewed in detail for any significant flaws in their emission estimate determinations. In particular, the estimate bases were evaluated and calculations checked. A significant flaw was considered any flaw that may have resulted in an emission estimate error equal to one percent or more of the estimated site total emission for that material. No significant flaws were identified. Several minor calculation discrepancies were identified and recalculated, and revised estimates evaluated. The errors were found to have had insignificant overall effects on emission estimates ($\leq$ 0.09 ton/yr), and the errors usually resulted in emission overestimates.

The emission estimate basis for each APEN report was usually a user estimate derived from inventory information or a derived evaporation rate. Mass balance information was usually not available. There was no chemical tracking system to document chemical use and movement. In several cases, estimates were based on standard emission factors published by USEPA in a document known as AP-42 (USEPA, 1985). However, in many cases, the AP-42 emission factors did not apply to Rocky Flats Plant processes.

The Building 774 APEN report stated that no carbon tetrachloride emissions need be accounted for from Building 774, because the APEN estimates for Building 707 and 776/777 assume

complete volatilization of used carbon tetrachloride. Review of the APEN reports for Building 707 and 776/777 confirmed uniform application of this conservative assumption and accounting. Therefore, monitoring data from Building 774 will be included in the process of comparing Building 707 and 776/777 emission estimates with available measurements.

The contribution of each building to the site carbon tetrachloride emission total was calculated, with the results shown in Table E-1.

TABLE E-1: CARBON TETRACHLORIDE EMISSIONS FROM RFP BUILDINGS BASED ON APENS

Building	Carbon Tetrachloride Emission Estimate (tons/yr)	Percent of Site Total Emission Estimate
707*	32.3	80
776/777*	8.1	20
460	0.00266	<1
881	0.000892	<1
Site Total	**40.4**	**100%**

* Building 774 emissions are included in emissions for these buildings.

All building emission estimates totaling less than one percent of the site total were not subject to a detailed review of the supporting APEN information. Detailed review of the supporting APEN information for minor chemical users is not expected to yield any significant difference in the final site-wide emission estimate even if the estimates are off by a factor of ten.

APEN reports for buildings contributing more than one percent to site-wide emissions were thoroughly evaluated. Emission estimate evaluations included but were not necessarily limited to:

- Checking calculations,
- Evaluating estimating methods,
- Checking assumptions,
- Looking for confirmatory information,
- Looking for overlooked emissions, and
- Addressing uncertainties or sources of error.

All calculations and mathematical extrapolations were checked. This usually just required converting the user quantity (gallons/year) to an emission estimate in tons/year. This calculation was generally found to be correct.

Each APEN accounted for building vents in detail to demonstrate inclusion of all emissions. However, none of the APENs for carbon tetrachloride users based the estimates on emission monitoring. The following is a summary of the major identified carbon tetrachloride users based on the APENs.

Building 707 Processes

Building 707 contained foundry and casting operations, and products assembly. Carbon tetrachloride was used as a cleaning agent. No emission controls for carbon tetrachloride were present in Building 707.

Module A - Casting Operations

Carbon tetrachloride was used to clean interior glove-box walls and furnaces in plutonium ingot casting furnace areas. A user estimate of 252 gallons per year was determined based upon inventory records and known routine cleaning practices. All of the carbon tetrachloride was assumed to evaporate. The calculated emission estimate is 1.68 tons per year.

Module J - Casting Operations

Plutonium ingots were cast in this area. Carbon tetrachloride was used to clean the glove boxes. The same basis and calculation as used for Module A were used here, yielding an emission estimate of 1.68 tons per year.

Module K - Casting Operations and Stacker Retriever

This operation stored and retrieved plutonium for distribution to other processes. Plutonium was weighed, melted in a furnace, and formed into ingots. Carbon tetrachloride was used to clean the glove boxes. The same basis and calculation as used for Module A were used here, yielding an emission estimate of 1.68 tons per year.

Module B - Rolling and Forming

This process involved the forming and thermal treatment of plutonium ingots. Carbon tetrachloride was used to clean the rollers. A user estimate of 3 gallons per day performed 220 days per year was the basis of an emission estimate of 4.4 tons per year.

Module C - Briquetting

Metal trimmings from Module C machining and Module B scrap cutters were placed into metal baskets and dipped into a series of five carbon tetrachloride open surface baths. Each bath was a steel tank containing approximately 4 gallons of carbon tetrachloride. Emissions were calculated based on the cold cleaner factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating procedures (such as keeping the lids closed when not in use), and the lowest allowable reduction of 28 percent was conservatively applied. Assumptions were found to be reasonable. The calculated emission estimate was found to be in error by a factor of 10 due to a math error, resulting in an overestimation of the emission estimate as 0.10 ton per year (rather than 0.01 ton/yr).

Module C - Machining Operations

Plutonium parts were machined, weighed, and then cleaned with carbon tetrachloride. A user estimate of 3,400 gallons per year was the basis of an emission estimate of 22.61 ton per year.

Modules C and D - Inspection

Carbon tetrachloride was periodically used to clean parts prior to inspection. User estimates based on inventories of 60 liters for Module C and 24 liters for Module D were used to calculate an emission estimate of 0.147 ton per year.

The Building 707 APEN reported carbon tetrachloride feed and waste transportation lines within the building, but no emissions were specifically identified from these lines. Consideration was given as to whether losses from these lines should have been accounted for. Separate transportation line emission accounting was not found to be necessary or appropriate because the

APEN emission estimates were based on the assumptions that all used carbon tetrachloride evaporates and is accounted for as an emission.

All waste carbon tetrachloride was pumped to the "C-pit," a partial basement under Module C. The pit consisted of two storage tanks and 16 "pencil" tanks for the storage of carbon tetrachloride. The waste carbon tetrachloride was pumped to Building 774 after being sampled. All potential emissions from the C-pit area were considered accounted for by assuming all of the carbon tetrachloride evaporated from each module source. This assumption was found to be reasonable with the exception of the Module C briquetting operation in which it is not clear if the waste materials had been assumed to evaporate.

The total Building 707 carbon tetrachloride emission estimate was 32.3 tons per year.

Building 776/777 Processes

Building 776 was originally a manufacturing building until operations were transferred to Building 707 in 1972. Building 776 served as a waste storage and waste reduction building after that time. Building 777 was an assembly building. No emission controls for carbon tetrachloride were present in either building. Building 776 and 777 share a common wall and ventilation system.

Building 776 - Baler

The baler was used to reduce the volume of low-level combustible waste. Carbon tetrachloride was a solvent found present in a maximum concentration of 750 pounds carbon tetrachloride per million pounds of waste. Assuming all of the carbon tetrachloride evaporated, the maximum concentration was used to determine an emission estimate of 2.32 tons per year (assuming 6,193,290 pounds of waste per year).

Building 777 - Briquetting

Machine turnings were placed in metal baskets and dipped into a series of four open surface carbon tetrachloride baths prior to placing in a hydraulic press for puck production. Emissions were calculated based on the cold cleaning factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating procedures (such as keeping the lids closed when not in use), and the lowest allowable reduction of 28 percent was conservatively applied. Assumptions were found to be reasonable and an emissions estimate of 7.9×10^{-2} ton per year was calculated.

Building 777 - Machining

Carbon tetrachloride was used as a cleaning agent for parts prior to machining. A user estimate of 850 gallons per year was used to quantify emissions assuming all used carbon tetrachloride evaporated. The resulting calculated emission estimate is 5.65 tons per year.

Building 777 - Inspection

Parts were cleaned with carbon tetrachloride prior to inspection. A user estimated of 31.5 liters per year was used to estimate emissions assuming all used carbon tetrachloride evaporated. The resulting calculated emission estimate is 5.5×10^{-2} ton per year.

Building 776/777 waste carbon tetrachloride was collected in a series of five "pencil" tanks. The waste carbon tetrachloride was pumped to Building 774 after being sampled. Potential emissions from the waste and fuel tanks as well as all associated transportation lines were considered to be addressed by the assumption that all used carbon tetrachloride evaporated. However, it is not clear if the waste materials from the briquetting operation were assumed to evaporate.

The total Building 776/777 carbon tetrachloride emission estimate was 8.1 tons per year.

E.1.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain carbon tetrachloride emission information to support a chemical emission source term.

Building 707

A Rockwell International report (Rockwell, 1988) estimated Building 707 carbon tetrachloride use of 10,000 gallons per year based upon an eight-week solvent use study. Assuming all of the carbon tetrachloride evaporated, this represents potential Building 707 carbon tetrachloride emissions of 66 tons per year.

The volatile organic emission report (Hamilton and Moser, 1990) prepared by Martin Marietta was found to contain detailed emission estimate information based on an air monitoring program. The methodology was studied to evaluate the reliability of reported results. General confidence in the reported methodology and associated results is high for reasons including:

- there were six sampling points,

- simulated normal operations were conducted during sampling, and worst-case scenarios were often assumed for calculation purposes,
- carbon tetrachloride was often the target chemical,
- three different sampling collection media were used (Tedlar bags, Tenax tubes, and Tenax/charcoal tubes),
- the highest concentration obtained with either bag or solid sorbent collection method was used to calculate the emission estimate, and
- air flow rates were determined based upon a multi-point transverse monitoring method rather than a single-point method.

Some uncertainty in sample result accuracy is present due to reported little net sampling time and small volumes. The overall sampling times were reasonable, but the actual net times were short and volumes were very small (8-20 minutes, 2-5 liters). An environmental team audit conducted in June 1989 (USDOE, 1989) also questioned the accuracy of Rocky Flats Plant air monitoring results.

The Hamilton and Moser (1990) emission estimates for 1989 were reported in pounds of carbon tetrachloride per hour. Extrapolation to estimated annual emissions was accomplished by assuming:

- 480 hours of operation per year for bimonthly cleaning (six 5-day inventory periods, two shifts per day, eight hours per shift).
- 3,520 hours of operation per year for normal operations (44 weeks per year with six one-week inventory periods and a two-week Christmas shutdown, two shifts per day, eight hours per shift).

These assumptions were derived from, and are consistent with, APEN assumptions. The total estimate extrapolated from the Hamilton and Moser (1990) report is 53 tons per year.

An internal Rocky Flats Plant report (Fruehauf and Richter, 1974) provided an estimated Building 707 carbon tetrachloride usage of approximately 1,000 gallons per month based on warehouse dispensing records. Assuming all of the dispensed carbon tetrachloride evaporated, a Building 707 carbon tetrachloride emission estimate of 80 tons per year is calculated.

An internal Rocky Flats Plant report (Hobbs, 1982) provided Building 707 carbon tetrachloride emission estimate information based on both monitoring and material balance bases. Monitoring data from 1974 and 1975 indicated an average and maximum Building 707 carbon tetrachloride emission estimate of 46 tons per year and 120 tons per year, respectively, with wide concentration fluctuations. An attached material balance summary indicated a Building 707 carbon tetrachloride emission estimate of 20 tons per year. This report also provided some quantifiable associated uncertainty. Examples include:

- ± 10 percent error in the Building 707 maximum discharge rate due to observed rapid changes in carbon tetrachloride concentrations.

- Up to 12 percent error in the Building 707 monitoring data due to questionable air flow rates during monitoring.

A Rockwell International report (Rockwell, 1989) reported updated halogenated solvent usage in Building 707 for the period of July 1988 - July 1989. The monthly usage varied significantly due to inventory and production activities. Assuming all of the used carbon tetrachloride evaporated, an average and maximum Building 707 carbon tetrachloride emission estimate is 48 tons per year and 76 tons per year, respectively. This report also confirmed that the Rocky Flats Plant carbon tetrachloride uses as of 1988 were "exclusively in Buildings 707 and 776/777."

A Rockwell International report (Ferrera, 1988) provided weekly and monthly carbon tetrachloride usage rates for Building 707 during the period of June - November 1988. This report stated that the Rocky Flats Plant use of carbon tetrachloride was "almost exclusively in Building 707" and that use of halogenated solvents was close to the Rocky Flats Plant solvent use reduction goals, in part, because production rates had been down. Assuming all the used carbon tetrachloride evaporated, usage results indicated an average and maximum Building 707 carbon tetrachloride emission estimate of 50 tons per year and 80 tons per year, respectively. This report also referred to a baseline monthly carbon tetrachloride usage rate of 1,167 gallons based on 1987-1988 purchase records. Assuming all the purchased carbon tetrachloride evaporated, this baseline usage indicated a Building 707 carbon tetrachloride emission estimate of 93 tons per year.

Table E-2 summarizes the Building 707 carbon tetrachloride emission estimates including APEN and non-APEN resources.

TABLE E-2

ROCKY FLATS PLANT BUILDING 707
CARBON TETRACHLORIDE EMISSION ESTIMATES

Resource	Estimate Basis	Estimate Year	Carbon Tetrachloride Emission Estimate (tons/yr)
EG&G, 1990a (APEN)	Use	1989	32.3
Rockwell, 1988a	Use	1988	66
Hamilton and Moser, 1990	Monitoring	1989	53
Fruehauf and Richter, 1974	Use	1973	80
Hobbs, 1982	Monitoring	1974-75	46 Average
	Monitoring	1974-75	120 Maximum
	Material balance	1981	20
Rockwell, 1989	Use	1988-89	48 Average
	Use	1988-89	76 Maximum
Ferrera, 1989	Use	1988	50 Average
	Use	1988	80 Maximum
	Use	1987-88	93

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Building 776/777

A Dow Chemical report (Dow Chemical, 1974) provided summary air monitoring data for Building 776/777 carbon tetrachloride emissions over a period of 46 days in 1973. Samples were collected every 15 minutes and carbon tetrachloride concentrations at exhaust points were found in the range of 0 to 270 pounds per day (24 hours). The detailed data table was missing, but average and maximum Building 776/777 carbon tetrachloride estimated emissions were calculated to be 10 and 34 tons per year, respectively.

The volatile organic emission report (Hamilton and Moser, 1990) provided detailed emission estimates information for Building 776/777 in 1989. The same assumptions were applied to the Hamilton and Moser emissions as were applied to Building 707 (described previously) to determine annual emission estimates. The total Building 776/777 carbon tetrachloride emission estimate extrapolated from the Hamilton and Moser (1990) report is 33 tons per year.

An environmental team audit conducted in June 1989 (USDOE, 1989) provided a Building 776/777 carbon tetrachloride eleven-month usage rate of 6,125 gallons. Assuming all used carbon tetrachloride evaporated, an emission estimate for Building 776/777 carbon tetrachloride is 44 tons per year.

An internal Rocky Flats Plant report (Hobbs, 1982) provided a material balance based carbon tetrachloride emission estimate for Building 776/777 of 15 tons per year. The report also provided a summary of air monitoring data for Building 776/777, but the data was found to be the same as that discussed above in the Dow Chemical 1974 report. Table E-3 summarizes the Building 776/777 carbon tetrachloride emission estimates including APEN and non-APEN resources.

Rocky Flats Plant - Total

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) reported an estimated Rocky Flats Plant total carbon tetrachloride usage in 1977 of 5,334 gallons. Assuming all the carbon tetrachloride evaporated, a Rocky Flats Plant emission estimate is 36 tons per year. The same report listed a Rocky Flats Plant carbon tetrachloride effluent discharge rate of 4.73 grams per second for 1975. Applying the same operating assumptions as used in the APENs and as applied to the Hamilton and Moser (1990) report, an annual emission estimate of 66 tons per year was calculated.

TABLE E-3

ROCKY FLATS PLANT BUILDING 776/777
CARBON TETRACHLORIDE EMISSION ESTIMATES

Resource	Estimate Basis	Estimate Year	Carbon Tetrachloride Emission Estimate (tons/yr)
EG&G, 1990b (APEN)	Use	1989	8.10
Dow Chemical, 1974	Monitoring	1973	10 Average
	Monitoring	1973	34 Maximum
Hamilton and Moser, 1990	Monitoring	1989	33
U.S. Dept. of Energy, 1989	Use	1989	44
Hobbs, 1982	Material balance	1974-75	15

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An internal Rocky Flats Plant report (Fruehauf and Richter, 1974) provided an emission estimate for the Rocky Flats Plant for 1973 of 81 tons per year. This is based on the assumption that all used carbon tetrachloride evaporated and that the report authors considered all 700 area carbon tetrachloride users.

The volatile organics emission report for 1989 (Hamilton and Moser, 1990) provided detailed carbon tetrachloride emission estimates for Buildings 460, 707, 774, and 776/777. As discussed in the previous sections for Buildings 707 and 776/777, the Hamilton and Moser (1990) report provided emission monitoring results in pounds of carbon tetrachloride per hour that may be extrapolated to determine annual emission estimates. Applying the same assumptions, a total Rocky Flats Plant carbon tetrachloride emission estimate is 89 tons per year.

The environmental team audit conducted in June 1989 (USDOE, 1989) reported that the major use of carbon tetrachloride occurred in the 700 complex where it was estimated that 99 percent of the carbon tetrachloride was used. This supports the APEN evaluation results that indicated almost all (>99 percent) of the carbon tetrachloride use and emissions involved the 700 complex. The audit report contains good descriptions of identified carbon tetrachloride applications throughout the Rocky Flats Plant.

Agreement with APEN information was generally good with no major exceptions. The audit report did identify carbon tetrachloride users other than those identified in the APENs. These reported users were found to represent insignificant-quantity users.

The environmental team audit report also provided an estimated Rocky Flats Plant carbon tetrachloride usage of 186,816 pounds for 1988. No detail is available to defend the basis of this estimated usage. Assuming all of the used carbon tetrachloride evaporated, a Rocky Flats Plant carbon tetrachloride emission estimate is 93 tons per year.

An EG&G Rocky Flats air stack release tabulation (EG&G, circa 1990) indicated 1988 and 1989 air stack emissions from the Rocky Flats Plant of 130,000 and 48,212 pounds, respectively. Converting to tons per year, this corresponds to 65 tons per year (1988) and 24 tons per year (1989).

An internal Rocky Flats Plant report (Hobbs, 1982) provided carbon tetrachloride emission estimates, based on both monitoring and material balance approaches, that could be summed to estimate Rocky Flats Plant total carbon tetrachloride emissions. The monitoring summary yielded an average Rocky Flats Plant carbon tetrachloride emission estimate of 56 tons per year and a maximum Rocky Flats Plant carbon tetrachloride emission estimate of 153 tons per year. The material balance summary provided two different Rocky Flats Plant carbon tetrachloride emission

estimates of 34 tons per year and 106 tons per year. No explanation for this difference was available.

An internal Rocky Flats Plant report (Rockwell, 1989) provided updated solvent use status from July 1988 to July 1989. Assuming all the carbon tetrachloride used evaporated, an average Rocky Flats Plant carbon tetrachloride emission estimate was 48 tons per year.

Table E-4 summarizes these Rocky Flats Plant total carbon tetrachloride emission estimates, including APEN and non-APEN resources.

E.2.2 Chloroform Emission Evaluation

Information sources relevant to chloroform emissions from Rocky Flats and the evaluation process used to develop source terms are described in this section. Information sources relevant to chloroform emissions at Rocky Flats include four APEN reports and six other resources, ranging from personnel interviews to hazardous material inventories and technical reports. The key information sources are described and evaluated in this section.

E.2.1 APEN Resources

Four APENs were initially identified documenting chloroform use and emissions. These APENs correspond to Buildings 559/561, 881, 374, and 460. Selected APENs with identified chloroform users were reviewed in detail for any significant flaws in the emission estimate determinations. No significant flaws were identified. The emission estimate basis for each APEN was a user estimate derived from inventory information. Mass balance information was not available, there was no chemical tracking system to document chemical use and movement, and the USEPA AP-42 emission factors did not apply to the Rocky Flats Plant chloroform uses.

Each building's percent emission relative to the total was calculated with the results shown in Table E-5.

The building emission estimates for Buildings 374 and 460 totaled less than one percent of the total Rocky Flats Plant emission estimate, and a detailed review of the supporting APEN information was not conducted.

The building emission estimates for Buildings 559/561 and 881 were thoroughly evaluated. Each APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, none of the APENs for chloroform users based the estimates on emission monitoring. The following is a summary of the major identified chloroform users based on the APENs.

TABLE E-4

ROCKY FLATS PLANT — TOTAL
CARBON TETRACHLORIDE EMISSION ESTIMATES

Resource	Estimate Basis	Estimate Year	Carbon Tetrachloride Emission Estimate (tons/yr)
EG&G APENs	Use	1989	40
U.S. Dept. of Energy, 1980	Use	1977	36
	Monitoring	1975	66
Fruehauf and Richter, 1974	Use	1973	81
Hamilton and Moser, 1990	Monitoring	1989	89
U.S. Dept. of Energy, 1989	Use	1988	93
EG&G, circa 1990	Use	1988	65
	Use	1989	24
Hobbs, 1982	Monitoring	1974-75	56 Average
	Monitoring	1974-75	153 Maximum
	Material Balance	1974-75	34
	Material Balance	1974-75	106
Ferrera, 1989	Use	1988-89	48

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TABLE E-5: CHLOROFORM EMISSIONS FROM ROCKY FLATS BUILDINGS

Building	Chloroform Emission Estimate (ton/yr)	Percent of Site Total Emission Estimate
559/561	0.74	88
881	0.0968	11
374	0.0014	<1
460	0.00625	<1
Site Total	**0.84**	**100%**

Building 559/561 Processes

Building 559/561 contained laboratory facilities for conducting spectrochemical, chemical, and mass spectrometric analyses. No emission controls for chloroform were present in Building 559/561.

Gallium Determination

Chloroform was used to extract gallium oxide from plutonium metal samples. A user estimate of 60 milliliters of chloroform per sample was determined based on inventory records and process knowledge. All of the chloroform was assumed to evaporate even though some chloroform waste was known to have been disposed of and collected in Building 374. The calculated emission estimate is 0.74 ton per year.

Building 881 Processes

Building 881 contained laboratory, maintenance, and plant support facilities. No emission controls for chloroform were present in Building 881.

No explanation of Building 881 chloroform use was provided in the APEN. The source was identified in interviews to be joining of plastics by carpenters (see below). A user estimate of 15.5 gallons per year was the basis of an emission estimate of 9.68×10^{-2} ton per year.

E.2.2 Information Sources Other Than APEN Documents

A number of resources were identified, evaluated, and found to contain chloroform use or emission information to support a chemical emission source term.

Two Rocky Flats Plant personnel interviews indicated that chloroform had been used by carpenters (Building 881) to join plastics but is no longer used in this way (ChemRisk, 1991-1992).

A 1988-1989 inventory (EG&G, 1990d) identified two Building 881 chloroform operations for dissolving plastics and photoresists. This reference and the one above appear to identify the Building 881 use application, which was not provided in the APEN. The inventory also indicated a total Rocky Flats Plant chloroform inventory of 0.55 ton.

The Waste Stream and Residue Identification and Characterization report for Building 881 (Wastren Inc., 1991c) noted the use of chloroform to clean machines and metal parts in Process 881/18, "Special Assembly, Microshaping." There was no laboratory usage identified (possibly due to a discontinued operation).

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant chloroform inventory of 9 tons. The significant inventory difference compared to the 1988-1989 inventory (EG&G, 1990d) is not currently explainable. The inventory change may be an indication of significant decreased use of chloroform from 1974 to 1988-1989.

An interim Rocky Flats Plant industrial hygiene department printout (Rocky Flats Plant, 1990) identified Buildings 559/561 and 881 as the only chloroform control areas, confirming the APEN evaluation of these two buildings as the major chloroform users.

The Waste Stream and Residue Identification and Characterization report for Building 559/561 (Wastren Inc., 1991d) confirmed the use of chloroform for the extraction of gallium oxide.

No comparable material balance or monitoring basis resources were identified.

E.3 Methylene Chloride Emission Evaluation

Information sources relevant to methylene chloride emissions from Rocky Flats and the evaluation process being used to develop associated source terms are described in this section. Information sources relevant to methylene chloride emissions at Rocky Flats include seven APEN reports and five other resources, ranging from personnel interviews to hazardous material inventories and technical reports. The key information sources are described and evaluated in this section.

E.3.1 APEN Resources

Seven APENs were initially identified documenting methylene chloride use and emissions. These APENs correspond to Buildings 776/777, 771, 881, 551, 460, 374, and 228 A/B. Selected APENs with identified methylene chloride uses were reviewed in detail for any significant flaws in the emission estimate determinations. No significant flaws were identified. The emission estimate basis for most of the APENs was a user estimate derived from inventory information or engineering data. One of the APENs (Building 228 A/B) used sludge monitoring data as the emission estimate basis. Mass balance information was not available, there was no chemical tracking system to document chemical use and movement, and the USEPA AP-42 emission factors did not apply to the Rocky Flats Plant methylene chloride uses.

Each building's percent emission relative to the total was calculated, with the results as indicated in Table E-6.

The emission estimates for Buildings 374, 460, and 551 totaled less than one percent of the total Rocky Flats Plant emission estimate, and a detailed review of the supporting APEN information was not conducted.

The building emission estimate for Buildings 776/777, 771, and 881 were thoroughly evaluated. Each APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, none of the APENs for methylene chloride were based on emission monitoring.

The following is a summary of the major identified methylene chloride users based on the APENs.

Building 776/777 Processes

Building 776 was originally a manufacturing building until operations were transferred to Building 707 in 1972. Building 776 served as a waste storage and waste reduction building after that time. Building 777 was an assembly building. Buildings 776 and 777 share a common wall and ventilation system. No emission controls for methylene chloride were present.

TABLE E-6: METHYLENE CHLORIDE EMISSIONS FROM ROCKY FLATS PLANT BUILDINGS BASED ON APENS

Building	Methylene Chloride Emission Estimate (tons/yr)	Percent of Site Total Emission Estimate
776/777	2.32	70
771	0.70	21
881	0.28	8
374	0.025	<1
460	0.0014	<1
551	0.00053	<1
228 A/B	0.00012	<1
Site Total	**3.33**	**100%**

Baler

A baler was used to reduce the volume of low-level combustible waste. Methylene chloride was present in the wet, low-level waste. Methylene chloride was present in a maximum concentration of 750 pounds methylene chloride per million pounds of waste. Assuming all the methylene chloride evaporated, the maximum concentration was used to determine an emission estimate of 2.32 tons per year (assuming 6,193,290 pounds of waste per year).

Building 771 Processes

The principal operation in Building 771 was the recovery of plutonium from plutonium-bearing residues.

Maintenance

Maintenance personnel used chemical paint strippers consisting of approximately 85 percent methylene chloride (KS-3 paint remover). A user estimate of 165 gallons of paint stripper per year was used to determine an emission estimate of 0.70 ton per year. All of the methylene chloride was assumed to evaporate.

Building 881 Processes

Building 881 contained laboratories, maintenance shops, and plant support facilities.

Semivolatile Organics Analysis Laboratory

Methylene chloride was used to extract semivolatile organics from wastes using a Soxhlet extraction apparatus, or equivalent. Oil samples were first dissolved in methylene chloride, then prepared for gas chromatography/mass spectrometry (GC/MS) analysis. Laboratory personnel estimated that approximately 50 percent of the methylene chloride evaporated into the laboratory hood exhaust. The remaining methylene chloride was collected as waste in the satellite collection area. A user estimate of 100 gallons per year was used to determine an emission estimate of 0.28 ton per year.

E.3.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain methylene chloride use or emission information to support a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant methylene chloride inventory of 396.8 gallons (1,502 liters).

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) did not list methylene chloride as one of the major chemicals used in 1977. This report did identify methylene chloride as having been used in "various" buildings for paint stripping, confirming the APEN reports of such use.

A 1989 inventory printout of Rocky Flats Plant methylene chloride use (Grocki, 1989a) indicated a total inventory of approximately 73.8 gallons.

A memorandum regarding hazardous wastes potentially generated at the Rocky Flats Plant (Roy F. Weston, 1985) reported an estimated 60.8 gallons of methylene chloride as waste in 1977.

The Waste Stream and Residue Identification and Characterization report for Building 881 (Wastren, 1991c) cited the generation of methylene chloride waste from the Building 881 laboratory extraction process, confirming the APEN reported use. One hundred gallons of waste methylene chloride per year were reported. This may indicate that more than 50 percent of the APEN reported methylene chloride used (100 gallons per year) was recovered as waste, or it may indicate that the APEN estimated usage is low.

E.4 Tetrachloroethylene Emission Evaluation

Tetrachloroethylene is commonly known as percloroethylene (PCE). Information sources relevant to PCE emissions from Rocky Flats and the evaluation process used to develop an associated source term are described in this section.

Information sources relevant to PCE emissions at Rocky Flats include one APEN report and seven other comparable resources, ranging from personal interviews to hazardous material inventories and technical reports. The key information sources are described and evaluated in this section.

E.4.1 APEN Resources

One APEN was initially identified documenting PCE use and emissions. This APEN is for Building 374 and was reviewed in detail for any significant flaws in the emission estimate determinations.

No significant flaws were identified. The emission estimate basis was a user estimate derived from waste stream tests and waste generation records. Mass balance information was not available, there was no chemical use and movement tracking, and the USEPA AP-42 emission factors did not apply to the APEN-referenced emissions.

The Building 374 APEN was thoroughly evaluated. The APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, the APEN did not base the emission estimate on monitoring. The following is a summary of the identified PCE use based on the APEN.

Building 374 Processes

Building 374 was activated in 1978 as the process waste treatment facility for many of the production buildings.

Process Waste Treatment

PCE was detected in a 1986 waste stream test for Building 889 waste. A 1987 user estimate (waste generation rate) was the basis of an emission estimate of 7.14×10^{-5} ton per year. All PCE was assumed to evaporate.

The Building 374 APEN did not identify how the PCE was used in Building 889. There is also no Building 889 APEN available to further explain the Building 889 PCE use.

E.4.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain PCE use or emission information to support development of a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant PCE inventory of 1,179 gallons (4,462.75 liters).

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) did not list PCE as one of the major chemicals used in 1977.

The environmental team audit conducted in 1989 (USDOE, 1989) did not list PCE as a major or minor solvent in use.

Three ChemRisk investigation interviews (ChemRisk, 1991-1992, Interview Nos. 39, 48, 53) indicated significantly different historical PCE uses. PCE was reported to have been used in Building 881 at the rate of approximately 25 drums per month. Distillation was reported to have recovered approximately 10 percent of the used PCE in Building 881 during this period resulting in an estimated release of 100 tons per year. PCE was reported to have been used in Building 881 to clean scraps prior to briquetting and "lots of PCE" was reported to have been used in Buildings 444, 883, 771, 776, and 374. PCE was also reported to have been used in Building 886 during the approximate period of 1965-1975.

Industrial hygiene memorandums (Dow Chemical, 1965-1974) described air sampling efforts to evaluate potential occupational exposures in Buildings 776 (May 1966) and 444 (February 1974). No PCE use description or quantity was provided for the Building 776 application. The Building 444 application used PCE as a machine coolant.

Several resources described a four-month trial effort in 1966 to substitute PCE for carbon tetrachloride in "cold washing" applications. The PCE substitution was discontinued due to residue buildup on plutonium surfaces and inspection devices from inhibitors associated with the PCE.

A report describing potentially generated Rocky Flats Plant wastes (Roy F. Weston, 1985) reported PCE waste generation of 73.4 gallons (378 liters) in 1982, 109.6 gallons (415 liters) in 1983, and 121.5 gallons (460 liters) in 1984.

Table E-7 summarizes the Rocky Flats Plant PCE emission estimates including APEN and non-APEN resources.

TABLE E-7
ROCKY FLATS PLANT — TOTAL TETRACHLOROETHYLENE EMISSION ESTIMATES

Resource	Estimate Basis	Estimate Year	Tetrachloroethylene Emission Estimate (tons/yr)
Air Pollution Emission Notices, EG&G (1991d)	Use	1986-87	7.14×10^{-5}
ChemRisk (1991-1992; Interview No. 39)	Use	1958-62	100

E.5 1,1,1-Trichloroethane Emission Evaluation

Information sources relevant to 1,1,1-trichloroethane (TCA) emissions from Rocky Flats and the evaluation process used to develop associated source terms are described in this section.

Information sources relevant to TCA emissions at Rocky Flats include six APEN reports and twelve other resources, ranging from personnel interviews to hazardous material inventories and technical reports by Rocky Flats Plant personnel and outside groups. The key information sources are described and evaluated in this section.

E.5.1 APEN Resources

Six APENs were initially identified documenting TCA use and emissions. These APENs correspond to Buildings 776/777, 707, 774, 460, 374, and 881. In addition, trichloroethylene (TCE) use that was erroneously reported for Building 460 was determined to be TCA. The total TCA usage estimate for Building 460 reflects this correction. Associated buildings that APENs did not identify as TCA users were not considered further as TCA users unless some other resource indicated differently: The APEN for Building 444 was added to the list of APENs to be evaluated for TCA emissions when several non-APEN resources and monitored emissions from Building 444 indicated TCA use.

Selected APENs with identified TCA users were reviewed in detail for any significant flaws in the emission estimate determinations. No significant flaws were identified. The emission estimate basis for most of the APENs was a user estimate derived from inventory information or engineering data. Several Building 707 and 776/777 emission estimates were based on analogy to similar processes described in the USEPA AP-42 emission factors reference (USEPA, 1985). Mass balance information was not available, and there was no chemical tracking system to document chemical use and movement.

Each building's percent emission relative to the total was calculated, with the results as indicated in Table E-8.

TABLE E-8: 1,1,1-TRICHLOROETHANE EMISSIONS FROM ROCKY FLATS BUILDINGS BASED ON APEN REPORTS

Building	Estimate Basis	1,1,1-Trichloroethane Emission Estimate (tons/yr)	Percent of Site Total Emission Estimates
776/777	Use/Analogy	8.36	41
774	Use	8.3	40
707	Use/Analogy	3.8	18
881	Use	0.061	<1
460	Use	0.15*	<1
374	Use	0.0022	<1
Site Total		**20.7**	

* Corrected total.

The building emission estimates for Buildings 881, 460, and 374 totaled less than one percent of the total Rocky Flats Plant emission estimate, and a detailed review of the supporting APEN information was not conducted.

The emission estimates for Buildings 776/777, 774, and 707 were thoroughly evaluated. Each APEN accounted for building vents in detail to demonstrate complete emission inclusion. However, none of the APENs for TCA were based on emission monitoring. The following is a summary of the major identified TCA users based on the APENs.

Building 776/777 Processes

Building 776 was originally a manufacturing building until operations were transferred to Building 707 in 1972. Building 776 served as a waste storage and waste reduction building after that time. Building 777 was an assembly building. No emissions controls for TCA were present in either building. Building 776 and 777 share a common wall and ventilation system. All waste TCA was pumped to the waste TCA collection system.

Building 776 - Baler

The baler was used to reduce the volume of low-level combustible waste. TCA was a solvent present at a maximum concentration of 2,000 pounds of TCA per million pounds of waste. Assuming all of the TCA evaporated, the maximum concentration was used to determine an emission estimate of 6.19 tons per year (assuming 6,193,290 pounds of waste per year).

Building 777 - Foundry Coatings

Substrates to be coated with uranium or plutonium were cleaned with a heated combination vapor and ultrasonic degreaser containing TCA to remove oils. The TCA was changed out when dirty and at bimonthly inventories. Emissions were calculated based on the heated cleaner factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating practices (such as keeping the lids closed when not in use), and the lowest allowable reduction of 30 percent was conservatively applied. Assumptions were found to be reasonable. The calculated emission estimate was 0.20 ton per year.

Building 777 - Disassembly (Room 430)

Disassembled plutonium parts were cleaned with small quantities of TCA. A user estimate of one gallon per year was the basis of an emission estimate of 5.6×10^{-3} ton per year.

Building 777 - Assembly Superdry (Room 430)

Parts from inspection were cleaned in a 10-gallon TCA bath to remove oil, grime, and dirt prior to ultrasonic cleaning. The TCA was changed out when dirty or contaminated with water and during bimonthly inventories. Emissions were calculated based on the cold cleaner factors from AP-42, Section 4.6-1, "Solvent Degreasing" (USEPA, 1985). A common reduction factor was applied based on good operating practices, and the lowest allowable reduction of 28 percent was conservatively applied. Assumptions were found to be reasonable. The calculated emission estimate was 0.18 ton per year.

Building 777 - Ultrasonic Cleaner (Room 430)

A heated ultrasonic vapor degreaser containing TCA was used to clean parts. The emission estimate based on AP-42 heated cleaner emission factors previously described was 0.69 ton per year.

Building 777 - Ultrasonic Cleaner (Room 440)

A heated ultrasonic vapor degreaser containing TCA was used to clean metal filters. The emission estimate based on AP-42 heated cleaner factors previously described was 0.46 ton per year.

Building 777 - Downdraft Room 430, 432B, 433, and 440 Assembly and Cleaning

TCA was used to clean assembled parts using wetted lint-free wipes. A user estimate of 2 gallons per year was used. All used TCA was assumed to evaporate and the resulting calculated emission estimate was 1.1×10^{-2} ton per year.

Building 777 - Radiography

TCA was used for general cleaning. A user estimate of 5 liters per year was provided. Assuming all used TCA evaporated, the resulting calculated emission estimate was 7.4×10^{-3} ton per year.

Building 777 - Weighing

Parts were cleaned with TCA prior to weighing. A user estimate of 2 liters per year was used to estimate emissions. All TCA reportably evaporated into a glove box. The resulting calculated emission estimate was 3.0×10^{-3} ton per year.

Building 777 - Plutonium Metalography Laboratory

TCA was used as a cutting agent for grinding with a carbide tip. A user estimate of 24 gallons per year was provided. All TCA was assumed to evaporate, and the resulting calculated emission estimate was 0.13 ton per year.

Building 777 - Special Weapons Projects

TCA was used for general cleaning and in ultrasonic cleaners. A user estimate of 15 gallons per year for general cleaning was used, all TCA was assumed to evaporate, and the resultant emission estimate was 8.4×10^{-2} ton per year. Emission estimates for the ultrasonic cleaners based on AP-42 heated and cold cleaners (one of each assumed from the APEN calculations) emission

factors previously described were 0.14 ton per year (cold cleaner) and 0.25 ton per year (heated cleaner).

Building 777 - TCA Collection and Filtration System

The TCA collection and filtration system collected TCA from buildings 707 and 776/777. Emission estimates for two tanks (T-1 and T-2) based on the AP-42, Section 4.3.1, "Storage of Organic Liquids" equation 2 were 4.94×10^{-3} ton per year (T-1) and 4.94×10^{-3} ton per year (T-2).

The total Building 776/777 TCA emission estimate was 8.36 tons per year.

Building 774 Processes

Building 774 handled TCA wastes received from Buildings 707 and 776/777. No emission controls for TCA were present in the building.

Organic and Sludge Immobilization System (OASIS)

OASIS received TCA and other organics from Buildings 707 and 776/777 and treated them by solidifying with gypsum cement. The treatment was performed in a glove box.

The OASIS emission estimate addressed any TCA not assumed to have evaporated from originating APENs (Buildings 707 and 776/777). Assuming all of the 1,478.4 gallons of TCA per year received evaporates, the emission estimate was 8.3 tons per year.

The total Building 774 TCA emission estimate was 8.3 tons per year.

Building 707 Processes

Building 707 contained foundry and casting operations and products assembly. No emission controls for TCA were present in Building 707. All waste TCA was gravity drained to tanks in the basement.

Assembly Operations - Modules D, E, and G

TCA was used for cleaning assembled parts in five ultrasonic cleaners (degreasers). There were four heated degreasers and one cold degreaser. Emission estimates for the degreasers based on AP-42 heated and cold cleaner emission factors (USEPA, 1985), as previously described, totaled 3.1 tons per year for all five degreasers. The degreaser emission estimates were considered conservative since the degreasers were used no more than 4 hours per day and the emission estimates assumed 16 hours of operation per day.

Assembly - Superdry - Module F

Parts were cleaned with TCA-wetted Kimwipes® prior to assembly. A user estimate of one gallon per month was provided. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 6.7×10^{-2} ton per year.

Assembly - Electron Bombardment Brazing/Scanning - Module G

TCA-wetted Kimwipes® were used to clean brazing operation bell jars. A user estimate of 24 gallons per year was provided. All used TCA was assumed to evaporate and the resulting calculated emission estimate was 0.13 ton per year.

Assembly Testing - Module H

Components were cleaned after quality control testing with TCA-wetted cheesecloth (infrequent use). A user estimate of less than a half gallon per year was provided. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 2.8×10^{-3} ton per year.

Radiography (Room 173)

TCA was used to clean parts prior to radiography. A user estimate of 5 liters per year was provided to estimate emissions. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 7.4×10^{-3} ton per year.

Weighing - Module D

TCA was used for general cleaning. A user estimate of 6 liters per year was provided. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 8.9×10^{-3} ton per year.

Eddy Current Testing - Module E

The same TCA use and quantity as Weighing — Module D were applied here with an emission estimate of 8.9×10^{-3} ton per year.

Weld Scanners and Fluorescent Penetrant Operations

TCA was used to clean dye from parts. A user estimate of 150 liters per year was used to estimate emissions. All used TCA was assumed to evaporate, and the resulting calculated emission estimate was 0.22 ton per year.

Production Control Operations - Module D

A 4-gallon capacity TCA dip tank was used to clean parts after grit blasting. A user estimate of 44 gallons per year based on estimated tank replenishment rates was used to estimate emissions. All replenished TCA was assumed to evaporate, and the resulting calculated emission estimate was 0.24 ton per year. This emission estimate did not account for bimonthly inventory refills or refills due to dirty TCA. Assuming 6 bimonthly inventory refills and 6 refills due to dirty TCA, a total emission estimate is 0.51 ton (versus the 0.24 ton per year APEN estimate). The larger emission estimate is more consistent with APEN assumptions, is more conservative, and will be used as an adjusted APEN emission estimate.

Calibration Laboratory - Modules D and G

TCA was used to clean gauges prior to performing precision measurements. A user estimate of 16 ounces per year was considered insignificant.

TCA Waste System

Waste TCA from ultrasonic cleaners (degreasers) was collected in a sump tank (V-100). The waste TCA was then pumped to Building 777 for subsequent pumping to Building 774 for treatment. Breathing losses were not calculated for the three feed tanks (V-36 A, B, and C) and the waste tank (V-100) due to the very small temperature change for indoor tanks and the fact that there were no day tanks involved. Working losses were calculated using AP-42, Section 4.3.1, "Storage of Organic Liquids" (USEPA, 1985) equation 2. The resulting calculated emission estimate for working losses from the three feed tanks was 7.6×10^{-3} ton per year based on a total 1989 TCA purchase for Buildings 707 and 777 of 2,450 gallons. The resulting calculated emission estimate for working losses from the waste tank was 9.0×10^{-4} ton per year based on an assumed waste tank throughput of 723 gallons per year.

The total Building 707 TCA emission estimate was 4.1 tons per year. This includes an adjustment for the higher emission estimate associated with the Production Control Operations — Module D TCA dip tanks.

E.5.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain TCA use or emission information to support a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant TCA inventory of 6,013 gallons (22,763 liters).

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) listed a Rocky Flats Plant 1977 TCA consumption of 26 tons per year (4,675 gallons).

The volatile organics emissions report (Hamilton and Moser, 1990) provided detailed TCA emission estimates for Buildings 444, 460, 707, 774, and 776/777. The Hamilton and Moser report provides emission monitoring results in pounds per hour, which can be extrapolated using standard APEN operating assumptions to determine annual emission estimates. The resulting calculated emission estimates are summarized in Table E-9. All emission sampling was performed in July 1989. The Hamilton and Moser report was the first identified resource indicating significant TCA use and emissions from Building 444. Three other non-APEN resources were identified to confirm the use of TCA in Building 444 (USDOE, 1989; Ferrera, 1988; and Rockwell, 1976). The APEN for Building 444 (EG&G, 1991e) was reviewed, and although it did not identify any TCA emissions, it did list three TCA tanks as being out of service "due to plant program to minimize use of criteria, hazardous and toxic chemicals. . . ." Both the APEN report and the Hamilton and Moser report used 1989 as the basis year so the discrepancy cannot currently be explained. It is possible that the building TCA use was discontinued after the Hamilton and Moser monitoring and before the APEN evaluation. The documented use and emissions from Building 444 have been included in the source term evaluation.

The environmental team audit conducted in June 1989 (USDOE, 1989) identified "major" TCA users that generally supported the APEN information. Some APEN reported users were not identified by the environmental team audit report. The environmental team audit also reported a 1988 TCA usage of 47,630 pounds. Assuming all used TCA evaporated, a Rocky Flats Plant total emission estimate is 24 tons per year.

TABLE E-9: 1,1,1-TRICHLOROETHANE EMISSION ESTIMATES FROM MONITORING AND APENS

Building	Based on Monitoring* (tons/yr)	Based on APENs (tons/yr)	Operation/Comments
444	2.1	0	Intermittent part immersion in a TCA vapor degreaser vented only 40 minutes per week.
460	5.3	.0072	Submerging parts in Cee Bee cleaner followed by a TCA vapor degreaser bath followed by a TCA ultrasonic bath.
707	21.7	4.1**	Ultrasonic degreasers and general TCA use as a cleaning agent.
774	15.7	8.3	Organic waste solidification (OASIS).
776/777	1.2	8.36	Normal unspecified operations.
Total	**46**	**20.8**	

*Hamilton and Moser, 1990
**Corrected total.

An internal Rockwell International chlorinated solvent usage report (Rockwell, 1988) confirmed APEN reported TCA uses for Building 707. The projected TCA usage in Building 707 was 3,500 gallons for 1988. Assuming all the used TCA evaporated, the resulting Building 707 TCA emission estimate is 20 tons per year.

A monthly status report on halogenated solvent use (Ferrara, 1988) documented planned efforts to reduce TCA use including reducing the Building 444 TCA use by 2,700 gallons per year. A "baseline" Rocky Flats Plant TCA use of 650 gallons per month was referenced based on 1987 and 1988 purchase records. Assuming all the used baseline TCA evaporated, a Rocky Flats Plant emission estimate is 44 tons per year. The Rocky Flats Plant solvent goal was to reduce the monthly TCA use from 650 gallons per month to 325 gallons per month.

An internal Rockwell International halogenated solvent usage update report (Rockwell, 1989) indicated an average TCA usage of 250 gallons per month for 1988-1989. Assuming all the used TCA evaporated, a Rocky Flats Plant emission estimate is 17 tons per year.

An EG&G Rocky Flats air stack release tabulation (EG&G, circa 1990) indicated 1988 and 1989 air stack emissions from the Rocky Flats Plant of 47,000 and 45,600 pounds, respectively. Converting to tons per year, this corresponds to 24 tons per year (1988) and 23 tons per year (1989).

A Rockwell International monitoring report (Rockwell, 1976) provided Building 444 TCA exhaust duct monitoring results in parts per million (ppm). An extrapolated emission estimate based on available exhaust duct flow rate and operating data (Hamilton and Moser, 1990) was calculated to be 5.0×10^{-5} ton per year. This result appears to contradict the Hamilton and Moser (1990) emission estimate of 2.1 tons per year; however, the discrepancy can likely be explained by the difference in time periods and monitoring methodologies.

The Waste Stream and Residue Identification and Characterization report for Building 776 (Wastren Inc., 1991e) did not cite any TCA waste generation. This may support the APEN assumption that all TCA associated with the Building 776 baler evaporated.

The Waste Stream and Residue Identification and Characterization report for Building 777 (Wastren Inc., 1991b) cited the generation of 2,802 gallons of TCA waste per year. The documented TCA waste sources generally supported the APEN reported users.

The Waste Stream and Residue Identification and Characterization report for Building 707 (Wastren Inc., 1991a) cited the generation of 3,854.5 gallons of TCA waste per year. The documented TCA waste sources generally supported the APEN reported users; however, the waste generation rate estimate is very high compared to the waste tank throughput reported in the Building 707 APEN.

Table E-10 summarizes the Rocky Flats Plant total TCA emission estimates including APEN and non-APEN resources.

E.6 Trichloroethylene Emission Evaluation

Information sources relevant to trichloroethylene (TCE) emissions from Rocky Flats and the evaluation process used to develop associated source terms are described in this section.

Information Sources relevant to TCE emissions at Rocky Flats include two APEN reports and twelve other comparable resources, ranging from personal interviews to hazardous material inventories and technical reports by Rocky Flats Plant and outside groups. The key information sources are described and evaluated in this section.

E.6.1 APEN Resources

Two APENs were initially identified documenting TCE use and emissions. These APENs are for Buildings 460 and 374. Other buildings that APENs did not identify as TCE users were not considered further unless some other resource indicated differently. The APEN for Building 444 was added to the list of APENs to be evaluated for TCE emissions when the Building 374 APEN (EG&G, 1991d) reported TCE in waste from Building 444.

Selected APENs with identified TCE users were reviewed in detail for any significant flaws in the emission estimate determinations. Given the plant's elimination of TCE usage for activities other than those related to research and analytic functions starting in 1975, the use of TCE in Building 460 reported in the APEN was questioned. A plant review, initiated at ChemRisk's request, revealed that TCE use reported for Building 460 was in fact TCA (Costain, 1992). TCE emission estimates for Building 460 have been reviewed and treated as TCA emissions.

The TCE emission estimate basis for the Building 374 APEN was a user estimate. Mass balance information was not available, there was no chemical tracking system to document chemical use and movement, and the USEPA AP-42 emission factors did not apply to the Rocky Flats Plant TCE uses.

The emission estimates for Building 374 were thoroughly evaluated. The APEN accounted for building vents in detail to demonstrate complete emission inclusion. The following is a summary of the major identified TCE users based on the APEN.

TABLE E-10

ROCKY FLATS PLANT — TOTAL 1,1,1-TRICHLOROETHANE EMISSION ESTIMATES

Resources	Estimate Basis	Estimate Year	Trichloroethane Emission Estimate (tons/year)
EG&G APENs	Use/Analogy	1989	20.8*
U.S. Department of Energy, 1980	Use	1977	26
Hamilton and Moser, 1990	Monitoring	1989	46
U.S. Department of Energy, 1989	Use	1988	24
Weis, 1988	Use	1988	20
Ferrera, 1988	Use	1987-89	44
Church, 1989	Use	1988-89	17
EG&G, circa 1990	Use	1988	24
	Use	1989	23

* Adjusted to include a higher emission estimate for Building 707 Production Control Operations — Module D — TCA dip tanks.

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Building 374 Processes

Building 374 was activated in 1978 as the process waste treatment facility for many of the production buildings. Emission controls for TCE were not present.

Process Waste Treatment

TCE was detected in 1986 waste stream tests for Building 444, 779, and 889. A 1987 user estimate (waste generation rate) was the basis of an emission estimate of 1.5×10^{-3} ton per year. All TCE was assumed to evaporate. This emission estimate did not include the Building 444 emission estimate (4.95×10^{-5} ton per year), with the reasoning that solvent use in Building 444 had been eliminated since the 1986 data had been collected.

ChemRisk attempted to evaluate APENs for other buildings identified as waste sources of TCE in the Building 374 APEN. The Building 444 APEN was reviewed and no reference to TCE use or emission was identified. APENs are not available for Buildings 779 and 889.

E.6.2 Information Sources Other Than APEN Documents

The following resources were identified, evaluated, and found to contain information to support a chemical emission source term.

A 1974 inventory (Barrick, 1974) indicated a total Rocky Flats Plant TCE inventory of 4,4041.6 gallons (15,300 liters).

A 1989 inventory printout (Grocki, 1989b) indicated a total Rocky Flats Plant TCE inventory of 13 milliliters.

Stack emission monitoring results for Building 776/777 (Johnson, 1973b) indicated TCE emissions from Building 776/777 in 1973 as 5.0 tons per year (average) and 17.9 tons per year (maximum). A separate resource (Dow Chemical, 1974) described the Johnson, 1973b, sampling methodology and discussed the results. Sampling and analysis was performed with a portable gas chromatograph and a flame ionization detector. Samples were collected every 15 minutes from Booster No. 1 exhaust. The TCE source was ultrasonic cleaners and cleaning baths.
A Dow Chemical report on the annual use of TCE (Dow Chemical, 1972-1974) indicated a total Rocky Flats Plant TCE 1973 usage of 60 tons per year (9,790 gallons) (Table E-11).

TABLE E-11

INFORMATION CONCERNING USE OF TRICHLOROETHYLENE DURING 1973

Building	User	Annual Use (gallons)
444	Chem Tech R&D	55
447/881	Metallurgical Operations	495
551	Stores	55
705	Ceramic R&D	55
771	Plutonium Fab R&D	55
771/707	Assembly Operations	5,720
774	Waste Treatment	165
777	Special Assembly	55
865	General Fab R&D	550
881	Assembly Operations	1,815
881	Fabrication	440
883	Metallurgical Operations	55
Site Wide	Chem Tech R&D Activities	55
Site Wide	Waste Management Activities	55
Miscellaneous	Unknown	165
TOTAL		9,790

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An undated Rocky Flats Plant report (Musgrave, circa 1973) discussed possible replacements for TCE. TCE was described as the "workhorse solvent for metal degreasing over the past 20 years." Prior to 1963, Rocky Flats Plant personnel cleaned metal parts with acetone, isopropanol, and other solvents. Beginning in 1963, ultrasonic vapor degreasing with stabilized trichloroethylene was the prime degreasing process. TCA was considered the best choice replacement for TCE for degreasing beryllium, plutonium, and uranium. Fiscal year 1973 Rocky Flats Plant TCE use was reported to be 10,000 gallons. Assuming all used TCE evaporated, an emission estimate is 62 tons per year.

The Final Environmental Impact Statement for Rocky Flats (USDOE, 1980) listed a Rocky Flats Plant 1977 TCE consumption of 2 tons per year (330 gallons).

A Rocky Flats Plant memorandum to the USEPA (Rocky Flats Plant, 1975) described the Rocky Flats Plant schedule for compliance with USEPA regulations requiring the use reduction and/or control of TCE for degreasing operations. This memorandum stated that TCE was no longer used at the Rocky Flats Plant as of December 1, 1974 except in one plutonium operation, which used up all TCE inventory by February 10, 1975. Since then, the only TCE use was reported to have been for research and analytical purposes using "insignificant small quantities." TCE was replaced with detergent washes in non-plutonium areas and TCA in plutonium areas. Other resources confirming the historical (pre-1975) large-scale use of TCE for degreasing were identified including, Hamilton and Moser (1990), ChemRisk (1991-1992; Interview No. 35); Dow Chemical (1974), and Dow Chemical (1965-1974) industrial hygiene memoranda.

The Waste Stream and Residue Identification and Characterization reports for Buildings 460 and 364 (Wastren Inc., 1991f and 1991g) referenced TCE waste streams.

Table E-12 summarizes the Rocky Flats Plant total TCE emission estimates including APEN and non-APEN resources.

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TABLE E-12

ROCKY FLATS PLANT — TOTAL TRICHLOROETHYLENE EMISSION ESTIMATES

Resource	Estimate Basis	Estimate Year	Trichloroethylene Emission Estimate (tons/yr)
EG&G APENs	Use	1986, 1987, 1988, 1989	1.5×10^{-3}
Dow Chemical (1972-1974)	Use	1973	60
Musgrave (circa 1973)	Use	1973	62
U.S. Department of Energy (1980)	Use	1977	2

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APPENDIX F

UNCERTAINTIES ASSOCIATED WITH ESTIMATING SPECIFIC ISOTOPE RELEASES BASED ON NONSPECIFIC MONITORING DATA

APPENDIX F

UNCERTAINTIES ASSOCIATED WITH ESTIMATING SPECIFIC ISOTOPE RELEASES BASED ON NONSPECIFIC MONITORING DATA

As described in Section 2.4.1, from 1953 to 1973 only long-lived gross alpha activity was routinely monitored in Rocky Flats airborne effluents. Release estimates of specific isotopes for this time period were calculated from the reported long-lived gross alpha activity. Even after 1973, some isotopes such as Am-241 and Pu-241, were not routinely monitored and their annual release quantities have to be derived from the release estimates of Pu-239/240. Because of the limited information available, uncertainty and error might have been introduced into the estimation of source terms of these radionuclides. This appendix evaluates and quantifies this source of uncertainty.

Because different information and sources of uncertainty were involved in the calculation of release estimates of plutonium and uranium isotopes, they are discussed separately in the following sections.

F.1 Uncertainties Associated with Estimating Pu-239/240, Am-241 and Pu-241 Releases Based on Nonspecific Monitoring Data

1953-1973

Before 1973, only long-lived gross alpha activity was routinely monitored in Rocky Flats airborne effluents. As a result, it is necessary to assume that long-lived gross alpha activity sampled in effluents from buildings in which plutonium was handled consisted solely of those alpha-emitting radionuclides associated with weapons grade plutonium. Since gross alpha analysis is not specific to any radionuclide, the plant made attempts during the 1970s to determine the accuracy of this practice. As discussed in Section 2.2.2.1, results of studies carried out in the mid and late 1970s indicate the correlation between total long-lived alpha and plutonium measured was poor. However, if samples with activity below 0.002 pCi m^{-3} were excluded from the study, the correlation was very good. For the purpose of this evaluation, it was estimated that the uncertainty associated with this practice is ± 20 percent. In other words, in order to compensate for this source of uncertainty, an uncertainty factor with an uniform distribution that ranges from 0.8 to 1.2 was applied to the annual release estimates developed in Section 2.8.

As discussed in Section 2.4.1.1, in the development of source terms for Pu-239/240 and Am-241 between 1953 and 1973, long-lived gross alpha activities measured from plutonium buildings were partitioned into Pu-239/240 and Am-241 release estimates. This method is based on the assumptions that there is a constant ratio between the two radionuclides and that the ratio established in 1980s can be applied to long-lived alpha monitored in the earlier years. In order to account for the uncertainty associated with the development of this ratio, other sources of information were consulted. According to Table 2-35, annual airborne Am-241 emissions for each year from 1985 to 1989 were between 13 percent and 31 percent of the plutonium alpha

activity release total for the same year. Theoretical calculations based on the initial purity and average age of plutonium handled at Rocky Flats indicate that the americium to plutonium activity ratio ranged from 10 percent to 20 percent. For the purpose of this evaluation, uncertainty factors of source terms of Pu-239/240 and Am-241 were developed using 0.31 and 0.1 as the upper and lower bounds of the americium to plutonium activity ratio:

When $F_{Am} / F_{Pu} = 0.10$ and $F_{Am} + F_{Pu} = 1$

$F_{Am} = 0.09$ and $F_{Pu} = 0.91$

And when $F_{Am} / F_{Pu} = 0.31$ and $F_{Am} + F_{Pu} = 1$

$F_{Am} = 0.24$ and $F_{Pu} = 0.76$

Based on the calculation shown above, the upper and lower bounds of the uncertainty factor of Pu-239/240 release estimate were calculated to be 1.11 (0.91/0.82) and 0.93 (0.76/0.82), respectively. Similarly, the upper and lower bounds of the uncertainty factor of Am-241 release estimate were determined to be 1.33 (0.24/0.18) and 0.50 (0.09/0.18), respectively. It was assumed that these uncertainty factors are triangularly distributed with best estimates equal to 1.0.

The uncertainty factor of the source term of Pu-241 was calculated by combining the uncertainties associated with the release estimates of total plutonium alpha activity and the ratio of Pu-241 to total plutonium alpha activity. As shown above, the uncertainty factor associated with the source terms of Pu-239/240 was represented by a triangular distribution with an upper bound of 1.11, a best estimate of 1 and a lower bound of 0.93. The range of beta to alpha activity percentage was not available. Since Pu-241 decays into Am-241, it is reasonable to assume that the uncertainty associated with the ratio of Pu-241 to Pu-239/240 is the same as the uncertainty associated with the ratio of Am-241 to Pu-239/240. As described earlier, release ratio of Am-241 to Pu-239/240 ranged from 0.1 to 0.31, with a most likely value of 0.22. Therefore, the uncertainty factor of the ratio was assumed to have a triangular distribution with a best estimate of 1 and upper and lower bounds of 1.4 (0.31/0.22) and 0.45 (0.1/0.22), respectively. The uncertainty associated with the calculation of Pu-241 release estimates based on the long-lived gross alpha data reported between 1953 and 1973 was obtained by combining the two distributions by Monte Carlo simulation. This resulted in an uncertainty factor with a normal distribution with a mean of 0.96 and a standard deviation of 0.2.

1974-1984

During this period, Pu-239/240 was monitored directly by alpha spectral analysis. However, Am-241 and Pu-241 were not routinely measured and release estimates of these two isotopes have to be calculated from the source terms of Pu-239/240. As described in Section 2.4.1.2, this calculation was based on the ratios of Am-241 to Pu-239/240 and Pu-241 to Pu-239/240. The uncertainty factor of the ratio of Am-241 to Pu-239/240 was developed in the section above, and was represented by a triangular distribution with a best estimate of 1 and upper and lower bounds of 1.4 (0.31/0.22) and 0.45 (0.1/0.22), respectively.

Information regarding the range of ratios of Pu-241 to Pu-239/240 is not available. Since Pu-241 decays into Am-241, it was assumed that the uncertainty associated with the Pu-241 to Pu-239/240 ratio is the same as the uncertainty associated with the Am-241 to Pu-239/240 ratio. Therefore, the uncertainty factor associated with the Pu-241 to Pu-239/240 ratio was also represented by a triangular distribution, with a best estimate of 1.0 and upper and lower bounds of 1.4 and 0.45, respectively.

1985-1989

During this period, both Pu-239/240 and Am-241 were monitored by alpha spectral analysis. Only Pu-241 release estimates were derived from the source terms of Pu-239/240. As explained above, this calculation was based on the ratio of Pu-241 to Pu-239/240. The uncertainty factor of this ratio was developed in the section above, and was represented by a triangular distribution, with a best estimate of 1.0 and upper and lower bounds of 1.4 and 0.45, respectively.

F.2 Uncertainties Associated with Estimating Enriched and Depleted Uranium Releases Based on Nonspecific Monitoring Data

1953-1977

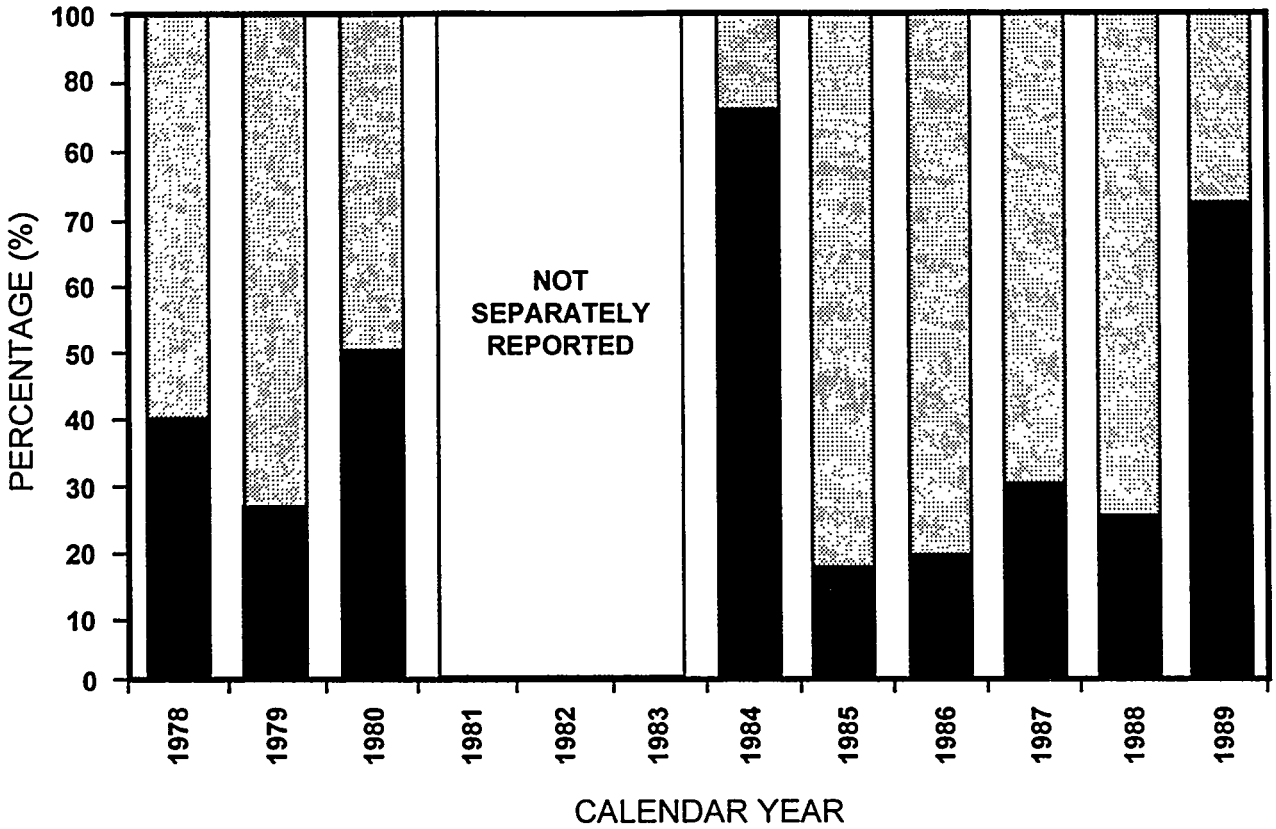
Routine isotopic analysis of effluent sample filters did not start until around 1973. However, reporting of long-lived gross alpha activity continued for uranium facilities until approximately 1978. Emission from uranium facilities were "radiochemically determined as U-233, U-234 and U-238" for the first time in the 1978 Rocky Flats Plant annual environmental report (Rockwell, 1979). Before that time, long-lived gross alpha activity sampled in effluents from buildings in which enriched or depleted uranium were handled was assumed to be 100 percent enriched or depleted uranium, respectively. As discussed above, an uncertainty factor represented by a uniform distribution with an upper bound of 1.2 and a lower bound of 0.8 was developed to compensate for the uncertainty introduced by this practice.

1978-1980 and 1984-1989

From 1978 through 1989, with the exception of 1981-1983, reported airborne uranium emissions were based on alpha spectral measurements of uranium isotopes. Due to the specific nature of these analysis, it was assumed that there was no uncertainty associated with the identity of analytes measured during this period of time.

1981-1983

Uranium emissions for calendar years 1981, 1982, and 1983 were reported only as total uranium emissions—separate depleted and enriched results were not reported. Based on the historical fractions of airborne depleted and enriched uranium emissions reported in 1978-1980 and 1984-1989, release estimates of enriched and depleted uranium were calculated from the total uranium emission. However, because of the fluctuation of the relative importance of enriched and depleted uranium in the total uranium emission, uncertainty was introduced in this translation. Based on the values shown in Figure F-1, the correction factor for enriched uranium was assumed to have a triangular distribution with a best estimate of 1 and upper and lower bounds of 1.95 and 0.42, respectively. Similarly, the correction factor for depleted uranium was assumed to have a triangular distribution with a best estimate of 1 and upper and lower bounds of 1.38 and 0.37, respectively.



LEGEND

ENRICHED URANIUM

DEPLETED URANIUM

ChemRisk
A Division of McLaren/Hart

FIGURE F-1
ENRICHED AND DEPLETED
FRACTIONS
OF RFP URANIUM EMISSIONS

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APPENDIX G

OVERALL UNCERTAINTIES ASSOCIATED WITH RELEASE OF ESTIMATES OF CONTAMINANTS OF CONCERN

APPENDIX G

OVERALL UNCERTAINTIES ASSOCIATED WITH RELEASE OF ESTIMATES OF CONTAMINANTS OF CONCERN

As discussed in Section 4, there are many sources of uncertainty that may have an impact on the release estimates of contaminants of concern. Uncertainty factors that can be used to represent uncertainties associated with monitoring programs and the identity of contaminants measured were developed in Section 4 and Appendix F, respectively. In this appendix, the identified uncertainty factors of a particular contaminant are combined where they overlap in time by a statistical technique called Monte Carlo simulation to produce an overall uncertainty factor.

Annual emission estimates were calculated by multiplying the source terms developed in Sections 2.8 and 3.1 by the appropriate overall uncertainty factor distribution. This produced a probability distribution for the annual emissions. It is believed that the use of these emission probability distributions will bound the actual emissions in a year.

In the following section, the calculation of overall uncertainty factors for plutonium and americium isotopes, uranium isotopes, tritium and beryllium are discussed.

G.1 Overall Uncertainties of Release Estimates of Pu-239/240, Am-241 and Pu-241

As discussed in Sections 2.2 and 4, five potential sources of uncertainty were identified in the development of release estimates of plutonium and americium isotopes:

(1) Sampling flow rate,
(2) Effluent flow rate,
(3) Analytical procedure,
(4) Identity of long-lived alpha emitters, and
(5) Apportioning of plutonium alpha into Pu-239/240 and Am-241.

Because monitoring data available for the calculation of release estimates of plutonium and americium isotopes from 1953 to 1973, from 1974 to 1984, and from 1985 to 1989 are different, different uncertainty factors were developed to account for various sources of uncertainty. They are listed in Tables G-1 through G-3. The overall uncertainty factor of each isotope and time period was calculated by combining all relevant uncertainty factors by Monte Carlo simulation. The results of the simulations are also provided in Tables G-1 through G-3. Since overall uncertainty factors calculated for a specific isotope do not change significantly over the three time periods, it was assumed that they can be represented by a single factor. The overall uncertainty factors of Pu-239/240, Am-241, and Pu-241 that were used to characterize plant emission from 1953 to 1989 are summarized below:

Overall Uncertainty Factor

Pu-239/240	Lognormal Distribution (GM=1.3, GSD=1.6)
Am-241	Lognormal Distribution (GM=1.4, GSD=1.6)
Pu-241	Lognormal Distribution (GM=1.2, GSD=1.6)

TABLE G-1

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF PU-239/240, AM-241 AND PU-241, 1953-1973

Nuclides	Source of Uncertainty					
	Partitioning of Total Alpha into Specific Isotopes		Sampling Flow Rate	Effluent Flow Rate	Analytical Procedure	Overall Method
Pu-239/240	Uniform 0.8 - 1.2	Triangular 0.93 - 1 - 1.11	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.1 SD = 0.3	Lognormal GM = 1.3 GSD = 1.6
Am-241	Uniform 0.8 - 1.2	Triangular 0.5 - 1 - 1.33	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.1 SD = 0.3	Lognormal GM = 1.2 GSD = 1.6
Pu-241	Uniform 0.8 - 1.2	Normal Mean = 0.96 SD = 0.2	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.1 SD = 0.3	Lognormal GM = 1.2 GSD = 1.6

Note:
SD = Standard deviation
GM = Geometric mean
GSD = Geometric standard deviation

TABLE G-2

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF PU-239/240, AM-241 AND PU-241, 1974-1984

Nuclides	Source of Uncertainty				
	Partitioning of Total Alpha into Specific Isotopes	Sampling Flow Rate	Effluent Flow Rate	Analytical Procedure	Overall Method
Pu-239/240	NA	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.07 SD = 0.14	Lognormal GM = 1.3 GSD = 1.4
Am-241	Triangular 0.45 - 1 - 1.4	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.07 SD = 0.14	Lognormal GM = 1.2 GSD = 1.5
Pu-241	Triangular 0.45 - 1 - 1.4	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.07 SD = 0.14	Lognormal GM = 1.2 GSD = 1.5

TABLE G-3

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF PU-239/240, AM-241 AND PU-241, 1985-1989

Nuclides	Source of Uncertainty				
	Partitioning of Total Alpha into Specific Isotopes	Sampling Flow Rate	Effluent Flow Rate	Analytical Procedure	Overall Method
Pu-239/240	NA	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.07 SD = 0.14	Lognormal GM = 1.3 GSD = 1.4
Am-241	NA	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.07 SD = 0.14	Lognormal GM = 1.4 GSD = 1.5
Pu-241	Triangular 0.45 - 1 - 1.4	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.07 SD = 0.14	Lognormal GM = 1.2 GSD = 1.5

Note:
SD = Standard deviation
GM = Geometric mean
GSD = Geometric standard deviation
NA = Not Applicable

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G.2 Overall Uncertainties Associated with Release Estimates of Enriched and Depleted Uranium

As discussed in Sections 2.2 and 4, four potential sources of uncertainty were identified in the source term development of uranium isotopes:

(1) Sampling flow rate,
(2) Effluent flow rate,
(3) Analytical procedure, and
(4) Identity of the long-lived alpha emitter.

To establish the overall uncertainty in the emission estimates of enriched and depleted uranium, the uncertainties described above were combined where they overlap in time. The uncertainty factors used in Monte Carlo modeling and the results obtained are listed in Tables G-4 through G-6. Since overall uncertainty factors calculated for a specific isotope do not change significantly over time, they can be represented by a single factor. The overall uncertainty factors of enriched and depleted uranium over 1953-1989 are as follows:

	Overall Uncertainty Factor
Enriched uranium	Lognormal Distribution (GM=1.3, GSD=1.6)
Depleted uranium	Lognormal Distribution (GM=1.3, GSD=1.6)

G.3 Overall Uncertainties Associated with Release Estimates of Tritium

1953-1973

As described in Section 2.2.1.1, sampling for tritium in airborne effluents was not routinely conducted until 1974. Therefore, emission estimates of tritium prior to 1974 are not based on measurements. According to Section 2.5.5, annual tritium emissions for this period will be treated as a uniform distribution with the identified lower and upper bounds of 140 and 390 for the period of 1968 through 1973, and 1 and 800 for the period of 1953 through 1967.

1974-1989

The approach used to evaluate uncertainties associated with the monitoring of airborne plutonium and uranium can also be applied to the monitoring of airborne tritium. As discussed in Section 2.2, four potential sources of uncertainty were identified in the development of release estimates for tritium:

(1) Sampling flow rate,
(2) Effluent flow rate,
(3) Analytical procedure, and
(4) Collection efficiency.

TABLE G-4

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF ENRICHED AND DEPLETED URANIUM, 1953-1977

Nuclides	Source of Uncertainty				
	Partitioning of Total Alpha into Specific Isotopes	Sampling Flow Rate	Effluent Flow Rate	Analytical Procedure	Overall Method
Enriched Uranium	Uniform 0.8 - 1.2	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.1 SD = 0.3	Lognormal GM = 1.3 GSD = 1.6
Depleted Uranium	Uniform 0.8 - 1.2	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.1 SD = 0.3	Lognormal GM = 1.3 GSD = 1.6

TABLE G-5

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF ENRICHED AND DEPLETED URANIUM, 1978-1980 and 1984-1989

Nuclides	Source of Uncertainty				
	Partitioning of Total Alpha into Specific Isotopes	**Sampling Flow Rate**	**Effluent Flow Rate**	**Analytical Procedure**	**Overall Method**
Enriched Uranium	NA	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.06 SD = 0.2	Lognormal GM = 1.3 GSD = 1.4
Depleted Uranium	NA	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.06 SD = 0.2	Lognormal GM = 1.3 GSD = 1.4

TABLE G-6

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF ENRICHED AND DEPLETED URANIUM, 1981-1983

Nuclides	Source of Uncertainty				
	Partitioning of Total Alpha into Specific Isotopes	**Sampling Flow Rate**	**Effluent Flow Rate**	**Analytical Procedure**	**Overall Method**
Enriched Uranium	Triangular 0.42 - 1 - 1.95	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.06 SD = 0.2	Lognormal GM = 1.4 GSD = 1.6
Depleted Uranium	Triangular 0.37 - 1 - 1.38	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Normal Mean = 1.06 SD = 0.2	Lognormal GM = 1.1 GSD = 1.6

Note:
SD = Standard deviation
GM = Geometric mean
GSD = Geometric standard deviation
NA = Not Applicable

As described in Section 2.2.1.2, sampling flow rates have historically been set at approximately 50 cm^3 min^{-1} for tritium samplers. However, the actual average sampling flow rate is likely to be larger than this value. It is because water was used as the trapping medium for tritium; as sampled air was bubbled through the medium, a portion of water was lost to evaporation. This resulted in a drop in resistance to air flow and an increase of sampling flow rate. Since data to characterize the variability of the actual sample flow rates were not located, it is assumed that the correction factor of tritium sampling has a triangular distribution with a best estimate of 1.2 and upper and lower bounds of 1.5 and 0.9, respectively.

The uncertainty associated with the estimation of effluent flow rate has been discussed and quantified in Section 4. It was used in the determination of overall uncertainty associated with the tritium monitoring data.

As discussed before, beginning in 1974, the plant began reporting the average relative error associated with tritium analysis in the annual environmental reports. The reported annual errors are summarized in Table 4-1. Investigators were unable to clearly establish whether the plant corrected the reported release estimates for these errors; however, it is believed that they did not. Therefore, an uncertainty factor is developed to compensate for this potential source of error. It is assumed to have a normal distribution with a mean of 0.95 and a standard deviation of 0.08 as indicated in Table G-7.

TABLE G-7

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF TRITIUM, 1974-1989

Source of Uncertainty				
Sampling Flow Rate	**Collection Efficiency**	**Effluent Flow Rate**	**Analytical Procedure**	**Overall Methods**
Triangular 0.9 - 1.2 - 1.5	Triangular 1.3 - 3.0 - 4.8	Triangular 0.5 - 1 - 2	Normal Mean = 0.95 SD = 0.08	Lognormal GM = 2.6 GSD = 1.5

Another source of uncertainty that is unique to tritium is the collection efficiency of the analyte. Based on the result of a special tritium study performed by the plant in 1978 (Section 2.2.1.1), it is estimated that the collection efficiency of tritium is 48 $\pm$ 27 percent, meaning actual emission to the air ranged from 1.3 to 4.8 times those reported. Using this information, the

correction factor for collection efficiency of tritium was assumed to have a triangular distribution with a best estimate of 3.0 (average of 1.3 and 4.8) and an upper and lower bounds of 1.3 and 4.8, respectively.

Monte Carlo simulation was used to combine the four uncertainty factors described above to estimate the overall uncertainty in the tritium emission data after 1973. The uncertainty factors used in Monte Carlo modeling and the results obtained are listed in Table G-7. The overall uncertainty associated with the determined release estimates between 1974 and 1989 can be represented by a lognormal distribution with a GM of 2.6 and a GSD of 1.5.

G.4 Overall Uncertainties Associated with Release Estimates of Beryllium

As described in Section 3.1.2, a relatively complete record of the annual beryllium emissions was compiled from sample data logbooks for 1960 through 1970 and annual beryllium releases reported in the Annual Environmental Monitoring Reports for 1971 through 1989. No sampling data were located for the period prior to 1960. Based on document review and personnel interviews, it is believed that beryllium was not used in the manufacturing process until 1958 and in the absence of any data, it is assumed that the emissions from 1958 and 1959 were approximately the same as those reported in 1960.

Upon reviewing the beryllium monitoring program used at Rocky Flats, three potential sources of uncertainty were identified in the development of release estimates of beryllium:

(1) Sampling flow rate,
(2) Effluent flow rate, and
(3) Analytical procedure.

The sources of uncertainty related to the collection of samples and the quantification of effluent flow rates discussed for plutonium and uranium measurements also apply to beryllium measurements. Therefore, uncertainty factors associated with sample flow rate and effluent flow rate developed earlier can also be used in this section.

A brief description of different analytical methods used at Rocky Flats to measure beryllium throughout the history of the plant is provided in Section 3.1.1. Listed chronologically, they are: emission spectroscopy with photographic plates as detector, emission spectroscopy with photomultiplier as detector, flame atomic absorption spectroscopy and nonflame atomic absorption spectroscopy. It is believed that detection limit and precision of a new method is generally better than the method it replaced. Annual average relative errors of the latest method, nonflame atomic absorption spectroscopy, were reported from 1974 through 1989 and are reproduced in Table 4-1. Based on the information provided in this table, it is estimated that

for the period from 1971 through 1989, the potential error associated with beryllium analysis can be represented by an uncertainty factor with a normal distribution which has a mean of 1.21 and a standard deviation of 0.49. However, this would lead to an upper bound of approximately 2.7 (mean + 3 standard deviations) and a lower bound of approximately -0.3 (mean - 3 standard deviations). As it is physically meaningless to have a correction factor with a negative value, a surrogate uncertainty factor was created to represent this source of uncertainty. The new uncertainty factor is assumed to have a triangular distribution with a best estimate of 1.2 and an upper and lower bounds of 2.7 and 0, respectively.

Although information about the accuracy of the methods used in the earlier years is not available, it is reasonable to assume that they are larger than those reported after 1973. For the purpose of this evaluation, it is assumed that variability of relative error of earlier methods is about twice as large as those reported for nonflame atomic absorption spectroscopy. In other words, the correction factor would have a normal distribution which has a mean of 1.21 and a standard deviation of 1.0. However, this would give an upper bound of approximately 4.2 (mean + 3 standard deviations) and a lower bound of approximately -1.8 (mean - 3 standard deviations). For the same reason given above, a surrogate uncertainty factor was created to represent this source of uncertainty. The new uncertainty factor is assumed to have a triangular distribution with a best estimate of 1.2 and an upper and lower bounds of 4.2 and 0, respectively.

Like before, Monte Carlo simulation was used to combine the uncertainty factors developed to estimate the overall uncertainties in the beryllium emission data from 1960 to 1970 and from 1971 to 1989. The uncertainty factors used in Monte Carlo modeling and the results obtained are listed in Table G-8. The overall uncertainty factors developed for beryllium emissions are as follows:

	Overall Uncertainty Factor
1960-1970	Lognormal Distribution (GM=1.9, GSD=2)
1971-1989	Lognormal Distribution (GM=1.4, GSD=1.9)

TABLE G-8

INDIVIDUAL AND OVERALL UNCERTAINTY FACTORS ASSOCIATED WITH RELEASE ESTIMATES OF BERYLLIUM

Period	Source of Uncertainty			
	Sampling Flow Rate	**Effluent Flow Rate**	**Analytical Procedure**	**Overall Method**
1960-1970	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Triangular 0 - 1.2 - 4.2	Lognormal GM = 1.9 GSD = 2.0
1971-1989	Normal Mean = 0.92 SD = 0.11	Triangular 0.5 - 1 - 2	Triangular 0 - 1.2 - 2.7	Lognormal GM = 1.4 GSD = 1.9

Note:
SD = Standard deviation
GM = Geometric mean
GSD = Geometric standard deviation

APPENDIX H

CALCULATION OF OVERALL UNCERTAINTY FACTORS BY MONTE CARLO SIMULATION

APPENDIX H

CALCULATION OF OVERALL UNCERTAINTY FACTORS BY MONTE CARLO SIMULATION

Potential systematic errors and degrees of biases in the estimation of annual release rates of radionuclides and beryllium are identified in Section 4.0 of this report. Because of the uncertainties associated with the identified parameters, they are not defined in terms of a single, discrete number, but instead in terms of a probability distribution of values that we are confident includes the true but unknown value of a particular parameter. When using inputs that are described in terms of probability distributions to perform calculations, there is more than one possible answer, and an equation must be solved many times using discrete input values that are sampled from the probability distributions defined for each of the inputs. The answer that this calculation process produces is itself a probability distribution. One method that is commonly used to perform this type of calculation is known as Monte Carlo simulation. It permits the propagation of the errors identified throughout the analytic process so that they are accurately reflected in the result. The purpose of this appendix is to describe Monte Carlo simulation and its application in calculating the overall uncertainty associated with release estimates of a contaminant.

Monte Carlo Simulation

A simple equation like the one shown below can be used to illustrate how Monte Carlo simulation may be used to propagate uncertainties in the input parameters of an equation.

$$A = B * C$$

When the exact values of parameters B and C are known, A can be calculated by simply multiplying B by C. This is also known as a point estimate or deterministic calculation, because it produces a single value of A. However, when there are uncertainties associated with parameters B and C, A cannot be determined by multiplying B by C. Figure H-1 shows how Monte Carlo simulation can be used to propagate the uncertainties in B and C through the equation and produce a probability distribution of A. The process can be divided into three steps. First, many values of each parameter are selected according to the probability distribution of the parameter. Second, the selected values of parameter B are randomly paired with the selected values of parameter C. Lastly, the paired values are multiplied together consistent with the equation to produce an estimate of A. For example, if 500 pairs of parameter B and C are selected, Monte Carlo simulation would produce 500 estimates of A. These estimates can be arranged numerically to provide a probability distribution of A as shown in Figure H-1.

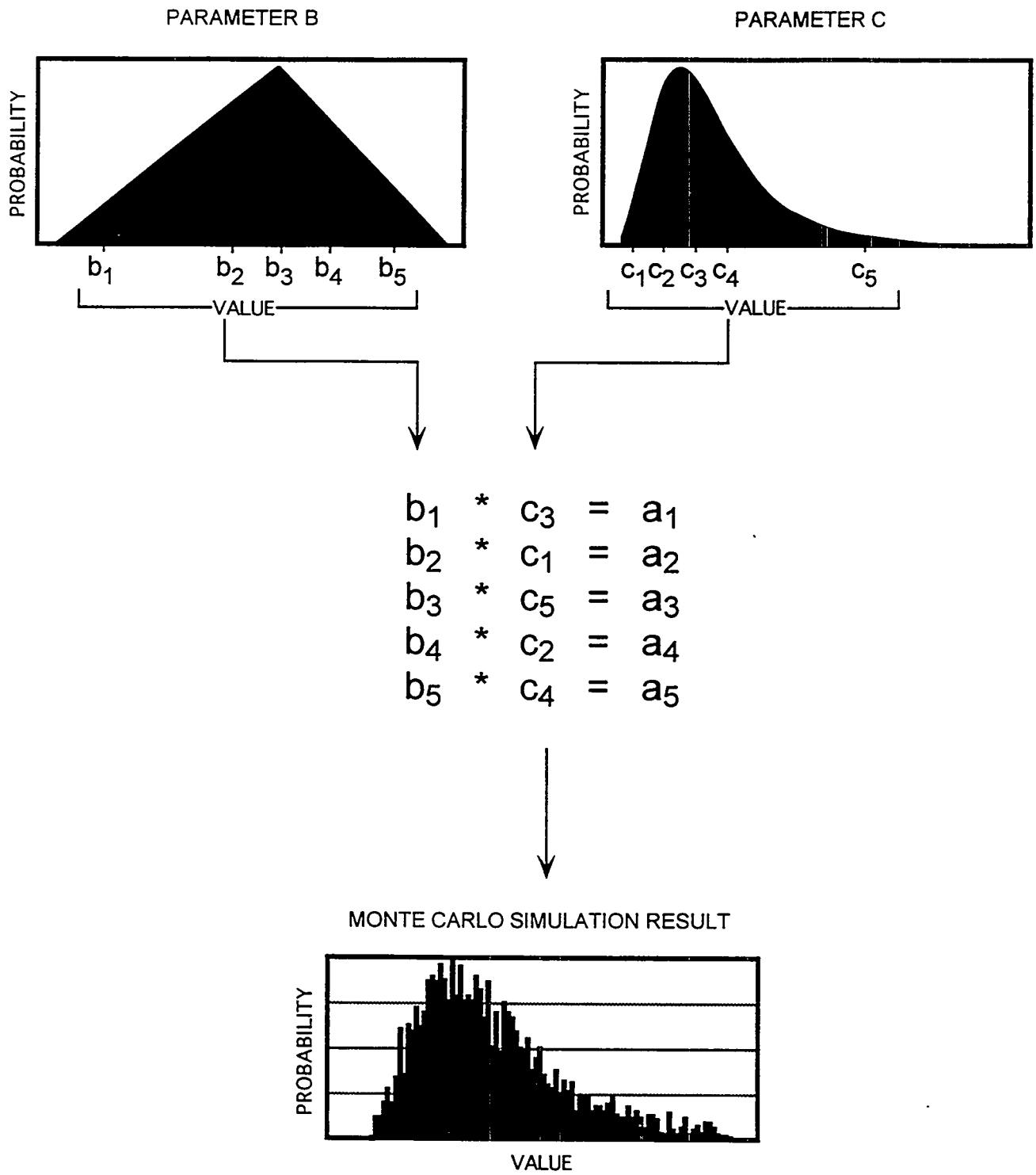

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FIGURE H-1
SCHEMATIC REPRESENTATION
OF MONTE CARLO SIMULATION

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Calculation of Overall Uncertainty Factors Associated with Contaminant Release Estimates

As described in Section 4 of this task report, potential systematic errors associated with the sampling and analytical processes, estimation of effluent flow rates and lack of information about the identity of the analyte are identified in the estimation of annual release rates of radionuclides and beryllium. The overall uncertainty factors of radionuclides and beryllium were determined by combining the appropriate uncertainty factors with Monte Carlo simulation (Appendix G). Calculation of the overall uncertainty factor associated with release estimates of Pu-239/240 between 1953 and 1973 is used in this appendix to illustrate this process.

As described in Appendix G (Table G-1), there are five sources of uncertainty in the determination of release estimates of Pu-239/240 between 1953 and 1973:

- Assignment of total long-lived alpha to plutonium and americium isotopes, U_a;
- Partition of plutonium and americium isotopes into specific isotopes, U_{Pu};
- Measurement of sampling flow rate, U_s;
- Estimation of effluent flow rate, U_e; and
- Measurement of total long-lived alpha particles, U_m.

If each of these five sources of uncertainty can be represented by an uncertainty factor, the overall uncertainty of the Pu-239/240 release estimates can be determined by the following equation:

$$\textit{Overall Uncertainty Factor} = U_a * U_{Pu} * U_e * U_m / U_s$$

A commercial software package called Crystal Ball® (Decisioneering, 1993) was used to perform the simulation. Probability distributions of the five uncertainty factors defined in Table G-1 of Appendix G were used as inputs to the equation and are presented in Figure H-2. In this example, 3000 runs were performed by the program to generate a probability distribution of the overall uncertainty factor (Figure H-3). Since the distribution appears to be lognormally distributed, it is best characterized by its geometric mean (GM) and geometric standard deviation (GSD).

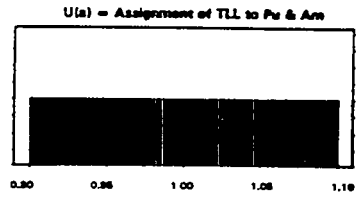
In order to facilitate the determination of GM and GSD of the overall uncertainty factor, the probability distribution of the overall uncertainty factor in logarithmic scale was also generated (Figure H-4).

Assumption: U(a) = Assignment of TLL to Pu & Am

Uniform distribution with parameters:

Minimum	0.90
Maximum	1.10

Mean value in simulation was 1.00

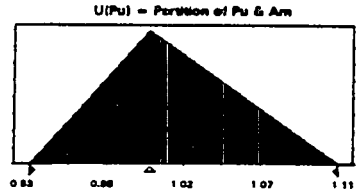


Assumption: U(Pu) = Partition of Pu & Am

Triangular distribution with parameters:

Minimum	0.93
Likeliest	1.00
Maximum	1.11

Selected range is from 0.93 to 1.11
Mean value in simulation was 1.01



Assumption: U(s) = Sampling Flow Rate

Normal distribution with parameters:

Mean	0.92
Standard Dev.	0.11

Selected range is from -Infinity to +Infinity
Mean value in simulation was 0.92

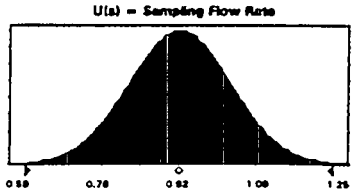


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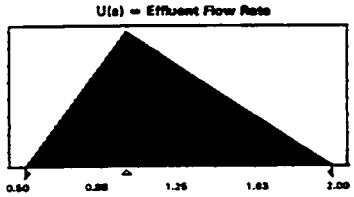
FIGURE H-2
PROBABILITY DISTRIBUTIONS OF UNCERTAINTY FACTORS USED IN THE DETERMINATION OF THE OVERALL UNCERTAINTY FACTOR OF Pu-239/240 RELEASE ESTIMATES (1953-1973)

Assumption: U(e) = Effluent Flow Rate

Triangular distribution with parameters:

Minimum	0.50
Likeliest	1.00
Maximum	2.00

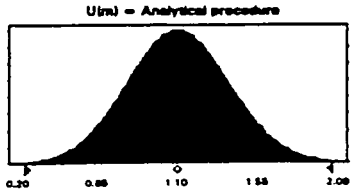


Selected range is from 0.50 to 2.00
Mean value in simulation was 1.16

Assumption: U(m) = Analytical procedure

Normal distribution with parameters:

Mean	1.10
Standard Dev.	0.30



Selected range is from -Infinity to +Infinity
Mean value in simulation was 1.11

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FIGURE H-2 Continued
PROBABILITY DISTRIBUTIONS OF UNCERTAINTY FACTORS USED IN THE DETERMINATION OF THE OVERALL UNCERTAINTY FACTOR OF Pu-239/240 RELEASE ESTIMATES (1953-1973)

Forecast: Overall uncertainty factor

Summary:

Display Range is from -0.50 to 3.50
Entire Range is from -0.28 to 4.42
After 3,000 Trials, the Std. Error of the Mean is 0.01

Statistics:	Value
Trials	3000
Mean	1.43
Median (approx.)	1.35
Mode (approx.)	1.25
Standard Deviation	0.60
Variance	0.36
Skewness	0.90
Kurtosis	4.33
Coeff. of Variability	0.42
Range Minimum	-0.28
Range Maximum	4.42
Range Width	4.70
Mean Std. Error	0.01

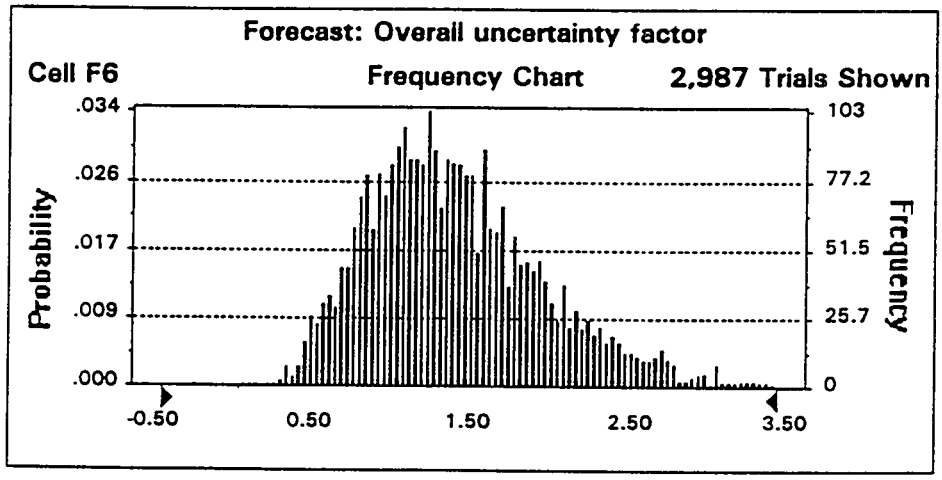

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FIGURE H-3
PROBABILITY DISTRIBUTION
OF THE OVERALL UNCERTAINTY FACTOR
OF Pu-239/240 RELEASE ESTIMATES
(1953-1973)

Forecast: ln(overall uncertainty factor)

Summary:

Display Range is from -1.00 to 1.50
Entire Range is from -3.25 to 1.49
After 2,999 Trials, the Std. Error of the Mean is 0.01

Statistics:	Value
Trials	2999
Mean	0.27
Median (approx.)	0.30
Mode (approx.)	0.23
Standard Deviation	0.44
Variance	0.20
Skewness	-0.63
Kurtosis	5.16
Coeff. of Variability	1.64
Range Minimum	-3.25
Range Maximum	1.49
Range Width	4.73
Mean Std. Error	0.01

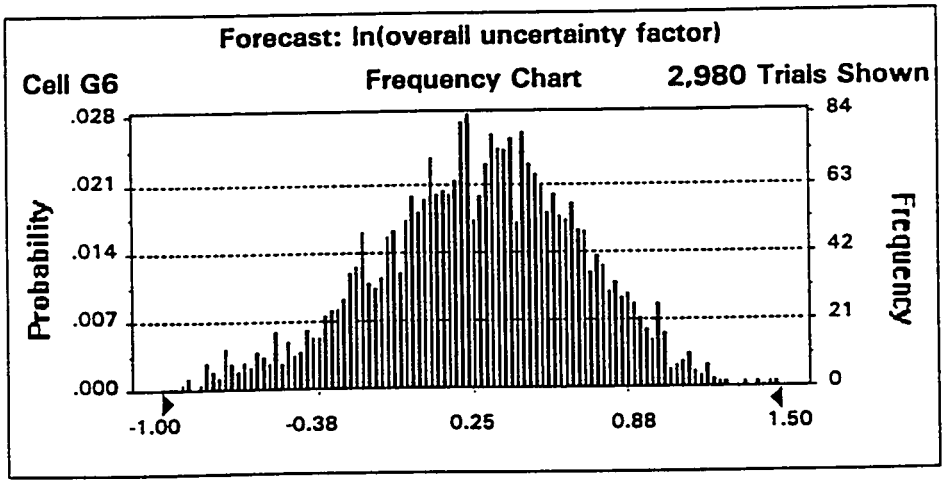

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FIGURE H-4
PROBABILITY DISTRIBUTION OF THE OVERALL UNCERTAINTY FACTOR OF Pu-239/240 RELEASE ESTIMATES IN LOGARITHMIC SCALE (1953-1973)

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GM and GSD of the overall uncertainty factor are related to the mean and standard deviation of the overall uncertainty factor in logarithmic scale by the following two equations:

$$GM = \exp[mean(L)]$$

$$GSD = \exp[SD(L)]$$

Where:

GM = Geometric mean of the probability distribution of the overall uncertainty factor,

GSD = Geometric standard deviation of the probability distribution of the overall uncertainty factor,

mean(L) = Mean of the probability distribution of the overall uncertainty factor in logarithmic scale,

SD(L) = Standard deviation of the probability distribution of the overall uncertainty factor in logarithmic scale.

GM and GSD of the overall uncertainty factor associated with release estimates of Pu-239/240 between 1953 and 1973 are calculated based on the Monte Carlo simulation output and the equations shown above:

$$GM = \exp[0.27] = 1.3$$

$$GSD = \exp[0.44] = 1.6$$

GMs and GSDs of overall uncertainty factors of other contaminants of concern were determined in a similar manner.

REFERENCE

Decisioneering (1993). Crystal Ball, version 3.0. User Manual. Decisioneering, Inc., Denver, CO.

APPENDIX I

COLORADO DEPARTMENT OF HEALTH DRINKING WATER DATA SUMMARY AND STATISTICAL TESTING

APPENDIX I

COLORADO DEPARTMENT OF HEALTH DRINKING WATER DATA SUMMARY AND STATISTICAL TESTING

During the period of 1970 to 1989, the Colorado Department of Health (CDH) sampled drinking waters from the cities of Broomfield and Westminster. These cities draw their water from Great Western Reservoir and Standley Lake, which are potentially impacted by waterborne effluents from the Rocky Flats plant. In addition, CHD also sampled drinking waters from the cities of Arvada, Boulder, and Golden which derive their waters from reservoirs at a considerable distance from Rocky Flats and that do not receive run-off or effluents from Rocky Flats. The drinking water samples from the cities deriving their water from distant water supplies relative to Rocky Flats provide reference points for comparing the levels of radioactivity found in waters possibly impacted by the plant. These comparisons are presented in the main body of the text of this report in Section 5.5. This appendix presents details about the CDH data set in terms of the frequency of detection and the maximum values in Tables I-1 through I-4 and the annual average radionuclide concentrations in Table I-5 through I-8. As indicated in Table I-1 through I-4, a large number of samples were below detection limits. In calculating the annual average values, one-half the applicable detection limit was used whenever non-detect results were reported. As described in the main text of the report, the average is not necessarily the best statistical descriptor to use to characterize the data since the data are not normally (or even log-normally) distributed because of the large number of non-detects. The average, in this case, would tend to provide an over-estimate of the central tendency of the data set. For this reason, other methods of examining the data were explored to determine if they might yield additional information.

Statistical Testing

The objective of the analysis is to construct a statistical test that can be used to show if the radionuclide concentrations found in the drinking water from Broomfield and Westminster are the same as those from other cities (Arvada, Boulder, and Golden) which do not derive their water from reservoirs that were likely impacted by the Rocky Flats Plant.

The available water sample data cover an approximately 10-year period, with over 50 percent of the sample results below detection limits. The detection limits varied over time.

There are no known "direct" statistical tests that can be used when so many non-detects are involved. However, for large sample sizes, the generalized likelihood-ratio test (Mood *et al.*, 1963; pages 440-442) can be used to give an approximate solution to this problem. Like all likelihood tests, both "point" and interval data can be directly incorporated into the analyses without resorting to using "mid-value" or any other artificial value for data that are below a level of detection. Besides being restricted to large sample sizes, the only other constraint is that the underlying distribution of the data must be assumed. However, the test can be repeated by first assuming a log-normal distribution, then a normal, Weibull, etc. A more detailed description

of the underlying theory is given by Sverdrup (1967; pages 133-140), Wald (1943) and Wilks (1938). Measurements of contaminant concentrations, as in this application, are typically well-approximated by a lognormal distribution.

The generalized likelihood-ratio test is performed by defining a null hypothesis and its alternative, computing the parameters of the assumed distribution using the maximum likelihood algorithm, and then taking the ratio, λ, of the two likelihood functions that have different assumptions concerning the equality of the parameters:

$$\lambda = sup\ L(\mu_n\ ,\ \sigma_{1n}\ ,\ \sigma_{2n})\ /\ sup\ L(\mu_{1d}\ ,\ \mu_{2d}\ ,\ \sigma_{1d}\ ,\ \sigma_{2d})$$

where sup means to find the largest value; L(.) is the likelihood function for the joint log-normal distributions of two data samples containing both point and interval data; μ is the true mean of the assumed normal distribution of the logarithms of the original measurements (i.e, μ = Mean[y] and y = Ln(Conc)); σ is the true standard deviation of the same (i.e., σ = Std Dev[y] and y = Ln(Conc)). The subscripts 1 and 2 refer to the two data sets or cities from which the chemical concentrations are being compared and the subscripts "n" and "d" refer to the fact that the parameters for the numerator and may be different. In the numerator likelihood function, the mean of the two data distributions are assumed to be equal and the variances to be unequal. In the denominator likelihood function, both the mean and the standard deviations of the two data distributions are assumed to be unequal. The λ ratio will always be less than one because the numerator term has one degree of freedom less than that of the denominator term (the numerator term will always be more restrictive than the denominator, and therefore also less likely).

The exact distribution of the λ ratio is intractable (Mood *et al.*, 1963; page 440), but an approximate size-σ test can be obtained. The asymptotic distribution of the generalized likelihood-ratio reduces to that of the chi-square distribution with 1 degree of freedom for large sample sizes. If the null hypothesis is defined such that the means of the data distributions are assumed to be equal, then the null hypothesis should be rejected when the following condition holds:

$$-2Ln(\lambda) > \chi^2(1-\alpha,1)$$

when Ln(.) is the natural logarithm and χ^2(.) is the chi-square function evaluated at 1 degree of freedom and at the quantile 1-α.

The above test was performed with the radionuclide concentration data from Broomfield and Westminster compared to that of three surrounding cities (Arvada, Boulder, and Golden). A computer code was written to solve the maximum likelihood function for the joint distribution parameters, i.e., the set (μ_n, σ_{1n}, σ_{2n}) and (μ_{1d}, μ_{2d}, σ_{1d}, σ_{2d}) and then perform the above chi-square test. Solving the maximum likelihood problem for joint distributions can be very difficult. A brute force method of systematically guessing the parameters over various ranges was first used to get a rough estimate of the maximum likelihood parameters and then a Newton-Raphson iteration scheme was used to refine the solution to a high degree of accuracy.

The p-value or "size" of the test was found by setting $-2\mathrm{Ln}(\lambda) = \chi^2(1-\alpha,1)$ and solving for the resultant α that just satisfied this condition. Summaries of the results are given in Tables I-9 and I-10. The analysis shows that over the ten year period, the mean values of the Broomfield and Westminster water concentrations of plutonium, tritium, and uranium were statistically no different than those of the other cities. The only exceptions were those for uranium concentrations compared for Boulder and Broomfield and Westminster and Arvada.

Discussion

The likelihood-ratio test comparing means is reasonably reliable, provided the sample sized are not too small. Some of the sample sizes under consideration are rather small, particularly those for plutonium-238, and this problem is exacerbated by the fact that the non-detects contain less information than the detected concentrations. A more serious problem for the tests involving the smaller sample sized is lack of power to detect (that is, declare statistically significant) a difference between means when the difference between the true means is large enough to be considered important. Given that there are no better alternatives to the likelihood-ratio test for these data, the best that can be done is to exercise caution in interpreting the results of this analysis and emphasize that these analyses are not definitive given the limitations of the data.

Caution should also be exercised in the interpretation of the two p-values that are marginally less than 0.05 (i.e., uranium for Broomfield—Boulder and Westminster—Arvada). The probability of getting at least one p-value less than 0.05 when performing 12 independent tests, if there are no true differences between the 12 pairs of means, is 0.46. These tests are not completely independent, but clearly the chance of one "false positive" in the set is substantial. In addition, any imprecision in the tests due to small sample sizes and deviations from the distributional assumption is likely to result in p-values that are too small rather than too large. Therefore, the isolated significant differences provide, at most, weakly suggestive evidence of difference between the true means.

The bottom-line is that, as was concluded from the qualitative evaluation of the data in the main text of the report, there is little or no evidence based on the drinking water sampling conducted by CDH that waters from Broomfield and Westminster were any different with respect to the long-term (1970-1989) concentrations of the radionuclides sampled from those of Arvada, Boulder, or Golden.

TABLE I-1

DETECTION OF FREQUENCY OF PU-238 IN DRINKING WATER, 1970-1989

Location	Number of Samples	Frequency of Detection	Percent Detects	Maximum Concentration (pCi L^{-1})	Date of Maximum
Broomfield	253	33/253	13	0.16	3/19/74
Westminster	100	10/100	10	0.62	10/25/73
Arvada	28	5/28	18	0.58	2/14/75
Golden	30	3/30	10	0.19	2/14/75
Boulder	22	4/22	18	0.21	4/7/75

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-2

DETECTION FREQUENCY OF PU-239/240 IN DRINKING WATER, 1970-1989

Location	Number of Samples	Frequency of Detection	Percent Detects	Maximum Concentration (pCi L^{-1})	Date of Maximum
Broomfield	343	84/343	24	4.52	4/26/73
Westminster	150	47/150	31	0.75	7/5/72
Arvada	47	8/47	17	1.15	3/1/75
Golden	51	10/51	20	0.09	2/14/75
Boulder	49	6/49	12	0.48	4/7/75

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-3

DETECTION FREQUENCY OF NATURAL URANIUM IN DRINKING WATER, 1970-1989

Location	Number of Samples	Frequency of Detection	Percent Detects	Maximum Concentration (pCi L^{-1})	Date of Maximum
Broomfield	356	188/356	53	346.4	12/13/76
Westminster	280	94/280	34	29.15	7/10/74
Arvada	91	31/91	34	35	2/1/80
Golden	45	14/45	31	13.89	3/29/74
Boulder	42	8/42	19	15.46	4/16/73

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-4

DETECTION FREQUENCY OF TRITIUM IN DRINKING WATER, 1970-1989

Location	Number of Samples	Frequency of Detection	Percent Detects	Maximum Concentration (pCi L^{-1})	Date of Maximum
Broomfield	854	345/854	41	23293	6/1/73
Westminster	280	131/280	47	3450	6/2/75
Arvada	124	29/124	23	1291	11/13/91
Golden	59	16/59	27	1776	10/6/71
Boulder	65	12/65	19	1101	9/26/72

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-5

ANNUAL AVERAGE PU-238 CONCENTRATIONS IN DRINKING WATER (pCi L^{-1}), 1970-1989

Year	Arvada	Boulder	Golden	Broomfield	Westminster
1970	0.015	no samples	0.015	0.015	0.015
1971	0.015	no samples	no samples	0.015	0.015
1972	0.015	0.015	0.015	0.0155	0.015
1973	0.015	0.053	0.015	0.023	0.051
1974	0.015	0.015	0.0125	0.039	0.021
1975	0.32	0.1125	0.0775	0.017	0.017
1976	no samples	no samples	no samples	0.013	0.015
1977	no samples	no samples	no samples	0.014	no samples
1978	0.015	0.015	0.015	0.015	0.015
1979	0.015	no samples	0.015	0.018	0.015
1980	0.015	0.01375	0.015	0.015	0.015
1981	0.015	0.01375	0.01375	0.015	0.015
1982	0.015	0.015	0.015	0.018	0.02
1983	0.015	no samples	0.015	0.015	0.015
1984-1989	No Analyses for Pu-238				

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-6

ANNUAL AVERAGE PU-239/240 CONCENTRATIONS IN DRINKING WATER (pCi L^{-1}), 1970-1989

Year	Arvada	Boulder	Golden	Broomfield	Westminster
1970	0.015	no samples	0.02	0.027	0.11
1971	0.01	no samples	no samples	0.022	0.014
1972	0.01	0.025	0.027	0.073	0.07
1973	0.013	0.01	0.013	0.174	0.055
1974	0.01	0.01	0.01	0.063	0.019
1975	0.43	0.245	0.045	0.016	0.018
1976	no samples	no samples	no samples	0.015	0.01
1977	no samples	no samples	no samples	0.018	no samples
1978	0.03	0.01	0.03	0.01	0.01
1979	0.01	no samples	0.01	0.084	0.015
1980	0.01	0.01	0.01	0.01	0.01
1981	0.01	0.01	0.011	0.01	0.011
1982	0.01	0.0175	0.015	0.011	0.17
1983	0.0275	0.041	0.01	0.021	0.029
1984	0.46	0.256	0.0275	0.0275	0.073
1985	0.35	0.03	0.02	0.017	0.02
1986	0.0075	0.014	0.005	0.0055	0.006
1987	0.004	0.004	0.004	0.006	0.006
1988	0.002	0.0013	0.003	0.0023	0.0063
1989	no samples	no samples	no samples	0.0004	0.0015

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-7

ANNUAL AVERAGE NATURAL URANIUM CONCENTRATIONS IN DRINKING WATER (pCi L^{-1}), 1970-1989

Year	Arvada	Boulder	Golden	Broomfield	Westminster
1970	no samples	no samples	no samples	no samples	no samples
1971	no samples	no samples	no samples	3.52	3.78
1972	6.41	15	1.69	3.29	4.53
1973	5.32	8.48	5.64	5.48	5.84
1974	3.86	6.53	11.68	5.19	6.15
1975	1.5	1.5	1.5	4.52	3.6
1976	no samples	no samples	no samples	16.74	0.852
1977	no samples	no samples	no samples	1	1.5
1978	3.52	1.5	1.21	1.29	0.99
1979	0.69	0.35	0.69	0.85	0.69
1980	14.5	1.5	1.5	2.21	1.75
1981	4.64	1.5	1.5	2.86	1.72
1982	2.53	2.375	2.25	2.925	2.01
1983	1.52	1.8	1.375	1.125	1.27
1984	1.27	1	1	1.23	1.18
1985	1.12	1	1.3	1	1
1986	1	1	1	1.18	1.05
1987	1	1	1.6	1.75	1
1988	1	1	2.1	1	1
1989	no samples	no samples	no samples	1	1

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-8

ANNUAL AVERAGE TRITIUM CONCENTRATIONS IN DRINKING WATER (pCi L^{-1}), 1970-1989

Year	Arvada	Boulder	Golden	Broomfield	Westminster
1970	no samples	no samples	no samples	1051.8	1151
1971	982.5	735	1776	836	698
1972	no samples	533.7	650	995.4	892.75
1973	454.67	250	741.25	8555.2	678.3
1974	381.33	469	250	5432.1	648.2
1975	250	443	405.5	1899.6	724.1
1976	317	329.5	351	754	540.8
1977	495.5	250	330	446	370.04
1978	670.5	524	307.5	362.1	352.1
1979	280.25	212.5	212.5	302.8	281
1980	258	175	329.7	282.5	252.6
1981	412.3	262	332.3	439	275.4
1982	329	355.25	463.25	295.8	330.48
1983	218.2	231.6	175	265.3	284.4
1984	175	175	175	223.5	235.6
1985	175	175	175	208.9	209.9
1986	189.6	175	235.5	188.9	190.2
1987	233.9	175	175	194	202.24
1988	175	175	175	187	187.13
1989	no samples	no samples	no samples	102.1	100

Source: Calculated from data reported by the Colorado Department of Health in Monthly Environmental Surveillance Reports.

TABLE I-9

RESULTS OF PAIRED STATISTICAL COMPARISON TEST OF RADIOACTIVITY CONCENTRATIONS IN BROOMFIELD DRINKING WATER WITH DRINKING WATER FROM ARVADA, BOULDER, AND GOLDEN

City Paired with Broomfield	Resulting p-Value with [Data Set Size]			
	^{238}Pu	^{239}Pu	Tritium	Uranium
Broomfield Sample Size	[253]	[343]	[854]	[356]
Arvada	0.202 [28]	0.683 [47]	0.878 [124]	0.211 [91]
Boulder	0.541 [22]	0.156 [49]	0.393 [65]	0.023* [42]
Golden	0.139 [30]	0.402 [51]	0.849 [59]	0.522 [45]
Westminster	0.146 [100]	0.643 [197]	0.605 [418]	0.124 [279]

* You must reject the null hypotheses that the means of the log-normal distributions are the same. Test also rejected the null hypotheses for normal and Weibull distributions.

NOTE: Statistical test of the hypotheses that the concentrations of plutonium, tritium, and uranium in the water supply from Broomfield are the same as that from the cities of Arvada, Boulder, and Golden. The resultant p-value of the generalized likelihood-ratio test is given. The null hypothesis assumes that the statistical distribution of the data is log-normal and that the mean of the assumed normal distribution of the logarithms of the original measurements for radionuclide concentrations in Broomfield drinking water and that of any other city is the same but unknown and that the variance of the same are different and unknown. The alternative hypothesis assumes that the means of the distributions are different. The null hypotheses is accepted for any p-value greater than 0.05.

TABLE I-10

RESULTS OF PAIRED STATISTICAL COMPARISON TEST OF RADIOACTIVITY CONCENTRATIONS IN WESTMINSTER DRINKING WATER WITH DRINKING WATER FROM ARVADA, BOULDER, AND GOLDEN

City Paired with Westminster	Resulting p-value with [Data Set Size]			
	^{238}Pu	^{239}Pu	Tritium	Uranium
Westminster Sample Size	[100]	[197]	[418]	[279]
Arvada	0.700 [28]	0.853 [47]	0.569 [124]	0.036* [91]
Boulder	0.817 [22]	0.264 [49]	0.250 [65]	0.116 [42]
Golden	0.568 [30]	0.290 [51]	0.939 [59]	0.919 [45]

* You must reject the null hypotheses that the means of the log-normal distributions are the same. Test also rejected the null hypotheses for normal and Weibull distributions.

NOTE: Statistical test of the hypotheses that the concentrations of plutonium, tritium, and uranium in the water supply from Westminster are the same as that from the cities of Arvada, Boulder, and Golden. The resultant p-value of the generalized likelihood-ratio test is given. The null hypothesis assumes that the statistical distribution of the data is log-normal and that the mean of the assumed normal distribution of the logarithms of the original measurements for radionuclide concentrations in Westminster drinking water and that of any other city is the same but unknown and that the variance of the same are different and unknown. The alternative hypothesis assumes that the means of the distributions are different. The null hypotheses is accepted for any p-value greater than 0.05.

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APPENDIX J

RESOLUTION OF COMMENTS ON THE DRAFT TASK 5 REPORT

COMMENTS AND RESPONSES CONCERNING THE DRAFT PHASE I TASK 5 REPORT "ESTIMATING HISTORICAL EMISSIONS FROM ROCKY FLATS"

Comments numbered 1 through 150 were submitted by Radiological Assessments Corporation staff. Comments numbered 151 through 180 were submitted by Mr. Dick Fox of the Colorado Department of Health. Comments numbered 181 through 192 were submitted by Ms. Judy Brunch of the Colorado Department of Health, Water Quality Liaison. All responses were prepared by ChemRisk project staff.

Comment 1. Page 20. Appendix A indicates that the question of particle size for plutonium emissions is more complicated than the two-paragraph summary on page 20 suggests. The text indicates that there are studies that are not discussed in Appendix A. Summarize the important features of all the studies relied upon in drawing the conclusions in a table or tables in the appendix.

Response 1. *The discussion of airborne effluent particle sizes in Section 2.1 was revised as suggested. The available reports of particle sizing studies are described in Appendix A to the report. A table has been added that summarizes their methods and results. Discussion of a recently completed plant study was also added (Nininger & Osborne, 1992).*

Comment 2. Page 20. Present information on the particle size of other released materials, particularly uranium and beryllium, or state the assumptions that were made regarding particle size distributions for these materials.

Response 2. *No particle size information for uranium and beryllium were located. It was assumed that the results from the plutonium particle size studies described in Appendix A were representative of uranium and beryllium particle sizes from Rocky Flats HEPA-filtered effluents.*

Comment 3. Page 21. A more credible example of an organic form of tritium (T) would be CH_3T. What do the tritium species measurements imply about the chemical forms of tritium released during the life of the plant? The conclusion appears to require answers to the following questions: when was the incinerator installed? what fraction of the time did it operate? what were the average tritium release rates for the two operating modes?

Response 3. *The report has been revised to present CH_3T as an example of an organic form of tritium.*

The report has been revised to indicate HTO and HT emission rates during periods when oxide-containing materials were being processed and the plutonium recovery incinerator was in operation and during "nonoperating" periods based on the Hurley (1979) study.

The report has been revised to indicate that oxide-containing materials were being processed and the plutonium recovery incinerator was in operation about 27 percent of days during the period of the Hurley (1979) study (i.e., 25 days out of 92). Beyond this information, records of the operational history of the incinerator were not located during Phase I investigations.

Comment 4. Page 21. No references or other justifications are provided for the statements about the chemical forms of uranium and plutonium that were released. Plutonium and uranium processing involve several chemical forms. What are the reasons that the oxides are most likely to be released? Are there data that show most releases were from machining operations? Is there a problem with classified information here?

Response 4. *The "Plutonium and Uranium Compound Solubility" section of Section 2.1 has been expanded to describe likely forms of plutonium and uranium in Rocky Flats airborne effluents.*

Comment 5. Page 23. Under the section "Sampling of Airborne Particulate Matter" it is stated that sampling line lengths varied between 14 inches and 18 feet. This is a very large range and sample-line losses should be calculated.

Response 5. *The statement that the sample line lengths varied between 14 inches and 18 feet contained a typographical error. The statement has been corrected to read "Sample line lengths were reported to vary between about 14 inches and 10 feet." Compared to the sample lines found at many nuclear facilities, even the 10-foot lines are quite short.*

Section 2.2.1.3 now presents the results of sample line loss calculations for various sample line conditions representative of Rocky Flats systems. Calculational methods are described in Appendix B.

Comment 6. Page 23, last paragraph. (Marril, 1965) is misspelled and should be corrected to (Merril, 1965).

Response 6. *The identified typographical error has been corrected.*

Comment 7. Page 26. It is not clear how one arrives at tritium release underestimates of factors of 1.3 to 4.8 from a relative collection efficiency of 48±27 percent. It would seem that (as a minimum) the range of relative collection efficiencies needs to be specified; it would be better to include a figure that shows the distribution of relative collection efficiencies that were determined in the special study. Furthermore, because of the relatively large amount of information to be presented on the topic, the use of the text box for the special studies seems inappropriate.

Response 7. *The report has been revised to indicate how the factors of 1.3 and 4.8 are calculated (i.e., $1.3 = (0.48 + 0.27)^{-1}$ and $4.8 = (0.48 - 0.27)^{-1}$).*

Information describing the range of collection efficiencies typically experienced with tritium samplers similar to those used at Rocky Flats has been added. These revisions reflect data described in NCRP Report No. 47 and results of tritium sampling efficiency studies performed at Los Alamos (see Valentine, 1968). The data that are available at this time from the referenced special tritium sampling study do not support preparation of a distribution of relative tritium collection efficiencies; detailed data are given only for the Östlund method sampling.

As suggested, the information on special tritium sampling studies has been removed from text box formatting.

Comment 8. Pages 26,27. The term percent is replaced with the symbol %. A standard approach should be adopted.

Response 8. *The % symbol has been replaced with the word "percent" through the report.*

Comment 9. Page 28. 2nd paragraph. The units cfm are used for the first time, the rest of the text refers to ft^3/min.

Response 9. *As suggested, flow rate terminology in the report has been standardized.*

Comment 10. Page 28. The text gives the impression that there are more effluent sampler worksheets available for review, but only a limited number were reviewed because of time limitations. What fraction (approximately) of the available sheets were reviewed? The sampling flow rate bias is estimated to range from –50% to +25%. Based on the worksheet review, what was the distribution of biases within that range?

Response 10. *The discussion of the sampling flow rates has been revised to indicate that Phase I record searches yielded only a few air effluent calibration worksheets for 1991 from a plant employee's personal file. This documentation provides an extremely limited record of sampling flow rate variation.*

A sample flow rate correction factor distribution (observed values divided by assumed standard flow rate) is now discussed in Section 2.2.1.2. The distribution, which is assumed to be normal, is shown in Figure 2-2 of the revised report. The distribution has a mean of 0.92 and a standard deviation of 0.11.

Comment 11. Page 28. In the third line of the "Particle Sampling Velocity" subsection, the text should be revised to read " . . . standard ($\pm$20%) 2 ft^3 min^{-1} flow rate . . . ." and it would be better to place the reference at the end of the sentence if possible. This subsection deals with both sampling flows and probe placement.

Response 11. *The referenced sentence has been revised in response to the editorial comment.*

Comment 12. Problems with the sampling systems were raised as an issue at the December public meeting. It would be useful to examine in some detail the worst case situations for the expected aerosols. For example, (a) use the ANSI Guide or a calculation to evaluate the effect of the reported factor of 17 velocity mismatch on the measured effluent concentration and (b) make a similar estimate of effect for the most poorly placed sampling probe.

Response 12. *The results from an analysis of sampling errors associated with anisokinetic sampling at Rocky Flats was added to the text. The associated analysis is described in Appendix C. This analysis utilizes the results of the recently completed monitoring study conducted at Rocky Flats (Nininger & Osborne,*

1992) and approaches outlined in the draft radionuclide source terms and uncertainties report from the Fernald dosimetry reconstruction project (Voillequé et al., 1991).

Comment 13. Pages 29/30. Stating that there were significant deviations from isokinetic conditions and that none of the systems was in compliance does raise some large questions about the validity of the data. Unsupported statements that the resultant errors were inconsequential or not significant are not adequate to resolve these questions. All sampling systems are not of equal importance. List the release points in approximate order of contribution to the total releases and describe for each the likely impact of the identified sampling problems. This will identify the probable magnitude of the sampling errors.

Response 13. *The analyses performed in response to comments 5 and 12 address this comment.*

Comment 14. Page 31. The pressure drop for an impinger system is not as likely to increase with sampling time as it would for a particulate filter system. What are the reasons for expecting the sampling bias to be the same for the tritium samplers as for the particle samplers?

Response 14. *Sampling biases for tritium samplers are no longer assumed to be the same as for particle samplers. The "Tritium Sampling Flow Rates" portion of Section 2.2.1.2 now describes the likely increase of tritium sample flow rates with time due to evaporation of impinger fluid. This is in contrast to particle sampling by filtration, in which buildup on filters can cause flow rates to decrease with time. The range of uncertainty in tritium emission estimates due to lack of knowledge of sample flow rates is estimated to be between 0.9 and 1.5 times the estimated emissions.*

Comment 15. Page 32. Provide more information on the study of filter collection efficiency by Langer. The statement about the challenge aerosol indicates particles with a density of about 2.8 g cm^{-3} , which is not suggestive of any compounds of uranium or plutonium, although a radioactive aerosol is implied.

Response 15. *The confusion about challenge particle density in this comment is due to an apparent error in the report of the Langer (1984) study. Aerodynamic Equivalent Diameter (AED) is related to physical particle diameter (PHY) as follows:*

$$AED = PHY \sqrt{\frac{\rho_p}{\rho_0}}$$

Where ρ_p is the density of the particle and ρ_0 is the density of a unit density particle, i.e., 1. Langer (1984) states that the particle "size" averaged 0.3 micrometer, with plutonium density of 4.5 g cm^{-3}. But these data would indicate an average AED of 0.63 micrometer ($0.3 \sqrt{4.5}$, not the 0.5 micrometer stated in the Langer report. The text has been revised to indicate that standard calculation methods used with these data yield an AED of about 0.6 micrometer.

Comment 16. Page 32. In the comments on Appendix A, inclusion of a figure that shows filter efficiency versus particle size is recommended. Reference to such a figure would bolster the argument on page 32 as well.

Response 16. *A graph showing filter collection efficiency as a function of aerosol particle diameter has been added as Figure 2-3. A graph showing fractional penetration through a standard HEPA filter of particles from 0.01 to about 10 μm was also added as Figure A-2.*

Comment 17. Page 38. Plutonium-241 is not a "high-energy alpha emitter". The dose factor for 241Pu is about a factor of 50 lower than those of the other plutonium isotopes.

Response 17. *The Section 2.2.2 discussion of radionuclides found in weapons grade plutonium has been revised.*

Comment 18. Pages 40–43. The discussion of detection efficiency and self-absorption needs to be more carefully structured. It may be best to start with the equation (similar to that in Figure 2–4) that shows how the two factors are used and then exercise care when referring to them and to the loss of counts due to

self-absorption. The self-absorption factor given in Table 2–1 should be consistent with the value described in the equation and the text.

Response 18. *The discussion of detection efficiency and alpha particle self-absorption has been revised as suggested. Equations describing alpha counting efficiency and self-absorption factors were added to the report.*

Comment 19. Page 40. The reported "total efficiency" of 0.21 was not verified by daily measurements of NBS-traceable electroplated sources; only the counting efficiency can be assessed or set by such measurements (as indicated on page 41). The counting efficiency is stated to be 0.30±2% in Table 2-1; presumably what is meant is 30±2% or 0.30±0.02. However, if the calibrations source activity is only known to ±10% (footnote 3) then the efficiency can't be better than 30±3%.

Response 19. *Revisions to Table 2-1 and applicable portions of the report were made as suggested.*

Comment 20. Page 43. What is the basis for the conclusion that there is little uncertainty associated with the undocumented self-absorption factor? The implication is that all filters were approximately uniformly loaded with particles and that the depth of penetration is independent of the type of filter used. The text on page 42 is presently contradictory: "impossible to render any judgment regarding its technical validity" but that the correction factor of 0.7 "is consistent with values one would normally expect" and that the choice "should not be a source of significant bias or uncertainty." Relying so heavily on personal communications with plant personnel (Haynes, 1992; Tyree, 1992) for establishing these correction factors is not convincing. A more thorough investigation of the scientific literature should be done, if plant records indeed do not have any more information on these topics. In addition, an uncertainty analysis is necessary — these factors should not be viewed as point estimates, but as distributions.

Response 20. *Pages from the report referenced in this comment were revised to remove the contradiction. Unfortunately, additional review of plant records did not identify plant-specific supportive evidence or documentation of the 0.7 correction factor used to account for alpha particle absorption in filter paper.*

Comment 21. Page 44. Explain the factor 4.66 in the equation in Figure 2–4. It would be appropriate to use N_b and t_b to be consistent with S_b and to avoid confusion with the quantity t in Figure 2–5. Do not use computer notation (such as 2.22E6) in the figure or the equation beneath it.

Response 21. *The derivation of the factor of 4.66 found in the Lower Limit of Detection equation has been described, and the symbols used in the associated equations have been revised as suggested.*

Comment 22. Page 45. Provide a reference for the Koval method; the basis is not intuitively obvious from the text and Figure 2–5. The text needs to be revised so that it states that the goal is to correct for the contributions of the alpha-emitting daughters of ^{212}Pb; ^{212}Pb is not an alpha-emitter. It seems to have been assumed that the counting efficiency and self-absorption factor were both independent of alpha energy, which could be one source of the discrepancies that were observed when the specific analyses were compared with gross analyses.

Response 22. *References for the Koval methods have been provided. The suggested text revision regarding alpha-emitting daughters of lead-212 has been made, and the noted possible contributing factor in discrepancies between gross alpha analyses and specific measurements is acknowledged in the discussion of the detection efficiencies and self-absorption factors.*

Comment 23. Were any measurements made of the uranium or americium content of releases from the "plutonium buildings?" Were there comparisons of isotope-specific uranium concentrations and concentrations of long-lived alpha emitters (similar to those described for plutonium on page 47)?

Response 23. *All identified comparison efforts have been described. Measurements of the type needed to support evaluations of the type stated are generally only possible for the period after the mid-1970s, when isotope-specific monitoring was routinely conducted.*

The report was revised to include discussion of comparisons between independently calculated gross alpha measurements for the second half of 1973 and reported Pu-239 measurements for the same period (See page 53).

Comment 24. Page 47. Provide more detail on the comparison of plutonium concentrations with those of long-lived alpha emitters in the concentration range that was measured when only alpha counting was used. What was the range of the observed ratios? Include a figure that shows the distribution of ratios for the higher concentration range. It seems that the conclusion—that this source of uncertainty is unquantifiable—is not correct.

Response 24. *The available information does not support such an analysis.*

Comment 25. Page 49. The explanation of the problems with americium analyses is confusing. If the U, Pu, and Am are partitioned into separate fractions, how does the addition of ^{236}Pu tracer interfere with the estimate of Am recovery? If the Pu tracer is in the Am fraction, the method, not the tracer, is the problem. Is there a reason why it took 12 years to overcome the difficulty?

Response 25. *The use of Pu-236 as a nonindigenous tracer in radiochemical determination of alpha-emitting radionuclides in effluent samples caused problems because it has actinium daughters that are chemically similar to americium. These decay products were carried along with americium in chemical separation processes and interfered with efficient detection of americium-241.*

Comment 26. Page 49. Are the detection limits for the 1980s based on the same confidence interval as those for the earlier method? It would be useful to present both sets of values in the same units.

Response 26. *Detection limits presented are based on the same confidence level. Units have been standardized to facilitate comparison.*

Comment 27. Pages 50 and 51. The information on the internal QA program should be made into a separate section that contains, in addition to the description, information on how good the analytical data were/are. Particularly important are the results for samples analyzed in comparisons with the EPA laboratory.

Response 27. *Formal quality assurance programs did not begin at Rocky Flats until the 1970s. Very little of the information that was collected during the Phase I study describes the quality assurance program that was applied during the study period. Additional interviews and targeted records searches would be necessary to prepare a useful report section describing the Rocky Flats*

internal QA program, and such a section would likely add little information regarding the periods of most significant releases from Rocky Flats.

Comment 28. Page 56. The section titled "1957 Reconstruction" uses the release estimate presumably being questioned (from the 1980 FEIS) as the standard of truth and concludes that the raw data contained a calculation error and that the FEIS estimate is correct (apparently without knowing how it was obtained). The analysis does not adequately consider alternatives and, in our opinion, does not withstand critical examination.

The results from the October monthly report are inconsistent, considering the flow that would have been required. However, suppose the error was a misplaced decimal point in the flow rate and that it was 4.8 x 108 instead of 4.8 x 109 m3/month, about double that normally observed. The fact that during the fire the fans were turned up to a higher speed (prior to the explosion) lends some plausibility to such a flow rate, but we do not know whether it was possible to double the flow rate. If that were the case, then the estimated October release would be 23,000 μg. If the flow rate were increased to clear the contamination from the building during cleanup operations, the release during the last 11 days of September could likely be estimated to be about 97,000 μg. The total release for the year could have been more than 5 times greater than the DOE estimate of 26,000 μg (1595 μCi).

Look at it another way. Not considering September or October, the monthly reports give a 10-month total release of about 16,000 μg. That means that the DOE estimate for routine releases in September and October must be $\sim$10,000 μg ($\sim$610 μCi) if it is otherwise consistent with Ray (1957). The average concentration during the 53 days when the sampling system was operating was about 20 dpm/m3. The total air discharge during that period would have to be 6.7 x 107 m3, an average of 0.38 x 108 m3/month. Is there information that would support a factor-of-6 reduction in the exhaust flow rate during that 6-week period?

Response 28. *We have acknowledged and discussed in the text the potential for a number of different possible interpretations of the anomalies that we found in the data for estimating releases from Building 771 in October of 1957 following the fire. We have chosen to adopt the value reported in the monthly Health Physics report for October as the most plausible release since it represents the most basic level of data we have been able to locate to date. While we can*

postulate a number of reasons why this number is inconsistent with that reported in the Final Environmental Impact Statement we have no basis to establish any of these as fact.

Comment 29. Tables 2–5 and 2–6. If the notes are to be kept in the present format, the word "average" should be included with "dpm/m^3" in the equation used to calculate the total release. It is recommended that symbols be defined for each of the quantities and that the equation use the quantities (not units).

Response 29. *The tables have been revised as suggested.*

Comment 30. Table 2–5. Not that it affects the bottom line, but the average concentration for the Building 774 Stack should be a time-weighted mean rather than the simple arithmetic mean that it appears to be.

Response 30. *The time-weighted average (TWA) value has been inserted in Table 2-5 for the Building 774 stack. Throughout the remainder of the report, TWA values have been inserted for all stacks.*

Comment 31. Table 2–6. Is there any evidence to justify the assumptions that the flow rates through the three ducts that contributed to the Building 771 exhaust and that the flow rates through the four ducts that contributed to the Building 774 exhaust are equal? The reference to Building 559 should be deleted from the notes to the part of Table 2–6 that is on page 62. Give some explanation as to the nature of the ventilation systems and the functions of the "booster" and "water" ducts.

Response 31. *In the case of Building 771, the three measurements are averaged because they are from the same duct. The reference to Building 559 in the notes to Table 2-6 has been deleted. Notes that accompany tables have been improved to be more descriptive of methods used to calculate emission totals. Further information regarding distribution of flows between ventilation system components was not identified during the Phase I study.*

Comment 32. Page 59/64. In the section titled "1973 Reconstruction" it appears that the comparison amounts only to summations of previously compiled monthly releases for each plant. If that is the case, why should there be any difference between the current estimates and those in the FEIS? Presumably, the authors of that document had access to the same summaries. One would not expect a difference of 25% between two additions of roughly 100 numbers. How many spot checks of the compiled versus raw data were made and what was the distribution of ratios? Is there another explanation for the difference?

Response 32. *Raw data for calendar year 1973 were located for plutonium emissions but not for uranium emissions. Approximately 10 percent of the raw data associated with plutonium monitoring for this period were spot checked for accuracy. We do not have documentation of the specific data formats were used to generate FEIS summary values. In contrast to some earlier years, we do not have any reason to expect that independently calculated emission totals for calendar year 1973 would differ significantly from those reported in the FEIS. Evaluations performed as part of this study are based on independently calculated emission totals in all cases where they are available rather than on reported release totals.*

Comment 33. Table 2-7. Unless the November release from Building 771A was <0.054 μCi instead of <0.54 μCi, the total for the year is incorrect. Why is it that the November and December results are all less than detectable limits and those limits are much higher than previously measured release quantities?

Response 33. *The identified value has been corrected. An explanation for the exclusive appearance of values below limits of detection for November and December 1973 has not been identified. Because 1973 was a period of significant changes in analytical methods, from radiometric determination of gross alpha radioactivity to radiochemical determination of Pu-239/240, it is possible that the appearance of all less-than values in late 1973 was a result of the adoption of new reporting conventions associated with these changes in analytical methods.*

Comment 34. It would seem that the goal (page 55) of checking two additional years, including one from the 1980s is still valid. Is there not a year in the 1980s (other than 1984) for which a comparison of original records and reported releases would be feasible? This applies to both uranium and plutonium

releases. A year that includes americium release estimates as well might be preferred.

Response 34. *Release estimates for calendar years 1984 and 1986 were independently calculated from analytical data and are included in the Task 5 report. Table 2-27 presents 1984 results for Pu-239/240, Pu-238, U-233/234, and U-238. Table 2-28 presents 1986 results for the same radionuclides as well as for Am-241.*

Comment 35. Page 66. The sentence in the first full paragraph should read: "All other uranium discharge ducts appeared and were assumed to be . . . ." The footnote in Table 2–8, page 67, should read 141, not 153, days. See also comment 24 regarding the use of an equation employing quantities instead of units. To be correct, time-weighted averages should be used for Ducts A and B of Building 883 and Duct 5 of Building 881; the resulting average concentrations are 0.35, 0.42, and 0.16 dpm/m3, respectively.

Response 35. *The identified revisions have been made. The time-weighted averages given in the tables differ from those given in the comment because they are based on averages over the year rather than the period of sampling. This allows the same method of calculating totals to be used for all release points. We assumed no contribution before sampling started, 19 days of sampling in Building 883 ducts in August 1957, and 31 days of sampling in Building 881 duct 5 during July 1957. In the end, the results of the two methods are equivalent.*

Comment 36. Page 66. Which of the uranium processing buildings were considered in the DOE FEIS and how do those estimates compare with the appropriate part of Table 2–8?

Response 36. *The depleted uranium release totals reported in 5the FEIS for 1960 and earlier included Building 883 Duct A. They did not include Buildings 444 and 447. Building 444 sampling began in 1954 and Building 447 sampling began in 1956. Enriched uranium totals included Building 881 and (beginning in 1957) Building 883 Duct B. Results of comparison of reconstructed and reported values are presented in Section 2.3.2.2. See also Table 2-30, "Reported and Reconstructed Uranium Release Estimates."*

Comment 37. Table 2–9 should be expanded to include all of the monthly data on enriched uranium concentrations in exhaust ducts in Building 881. The need for presentation of all the data is emphasized by the first paragraph on page 70; a release of the magnitude indicated for a single day may require special treatment.

Response 37. *The table has been expanded as suggested. In addition, this single day release was subject of further evaluation in Task 6 from the standpoint of air transport and exposure.*

Comment 38. Page 66. Although the text states that uranium emissions were reconstructed for years prior to 1961, no data for the years 1958–1960 are presented and no explanation is given. Was Building 881 not operating during those years or does this imply that only releases from that building were included in the FEIS estimates?

Response 38. *Depleted uranium releases were independently reconstructed for every year from 1954 through 1960, 1963, 1969, and 1973. Enriched uranium releases were independently reconstructed for 1954 through 1957, 1963, 1969, and 1973. Independent reconstructions of depleted uranium emissions data for the 1958-1960 time period were performed because of the fact that early reporting neglected the contributions of several key buildings. Examination of enriched uranium data indicated that similar errors were not made in conjunction with enriched uranium releases, so enriched uranium emissions for these additional years were not reconstructed. This has been clarified in the text.*

Comment 39. Table 2-10. Using the data in Table 2–9 and the flow rates in the footnote to Table 2–2 leads to a release of about 210 μCi of enriched uranium during 1956. What is the source of the estimate of 1100 μCi in Table 2–10? Was the release on 24 January 1956 excluded from the averaging process? More explanation is needed.

Response 39. *Further explanation has been provided. See Table 2-12, "Summary of 1956 Airborne Enriched Uranium Emissions."*

Comment 40. Table 2–11 should be expanded to include all of the monthly data on depleted uranium concentrations in ducts in Buildings 444 and 447.

Response 40. *The tables have been expanded as suggested.*

Comment 41. Page 70. Which depleted uranium release points were considered in the FEIS estimates for 1957–1960 and how do estimates for those ducts compare with the comparable reconstructed release estimates?

Response 41. *The depleted uranium release totals reported in the FEIS for 1960 and earlier included Building 883 Duct A. They did not include Buildings 444 and 447. Building 444 sampling began in 1954 and Building 447 sampling began in 1956. Enriched uranium totals included Building 881 and (beginning in 1957) Building 883 Duct B. Results of comparison of reconstructed and reported values are presented in Section 2.3.2.2. See also Table 2-30, "Reported and Reconstructed Uranium Release Estimates."*

Comment 42. The FEIS apparently lists releases for enriched and depleted uranium separately (per Table 2–12). The depleted and enriched uranium release estimates from Table 2–13 should be compared with the corresponding DOE estimates. The comparison for total uranium given on page 72 is not particularly meaningful. The same comment applies to the text that compares the estimates for 1969 and 1973. Specify the discharge locations that contribute to the totals for enriched and depleted uranium.

Response 42. *The text and tables have been revised to include the requested comparisons and to identify the emission points associated with enriched and depleted uranium.*

Comment 43. Tables 2–17 and 2–18 give differing estimates of depleted uranium releases for 1957 (730 vs. 880 μCi). Given the comparisons in Table 2–17, it is not clear why the DOE uranium release estimates are considered to be valid prior to 1969, and have been selected for use.

Response 43. *Further explanation has been provided. Tables 2-30 and 2-32 are in agreement. For 1957 through 1960, depleted uranium emissions for Buildings 444 and 447 were independently reconstructed and were added to the reported depleted uranium emissions from other areas (Building 883). As shown in Table 2-32, release totals that reflect independent calculations are used instead of FEIS values in all cases where they are available.*

Comment 44. Page 90. What was the basis for the operating loss estimates for thorium? For alpha-emitters, whose concentrations in solid wastes are very difficult to quantify, it is not clear that either NOL or MUF would provide more than a very crude guide to releases to air or water.

Response 44. *The document that is the source of the thorium information does not describe the basis for the operating loss estimates for thorium, and this was not a focus area of our investigations. We agree that NOL and MUF provide only a crude estimate of potential release and have not attempted to develop a release estimate based on the limited information available and the relative unimportance of this material at Rocky Flats.*

Comment 45. Page 91. Delete the word "spontaneous" from the sixth line of the first paragraph of Section 2.7.

Response 45. *The requested editorial change has been incorporated.*

Comment 46. Page 97. In the first line of the normal text, replace "have" with "has."

Response 46. *The referenced subject matter has been relocated to Section 4, "Uncertainty in Airborne Emission Estimates." The requested editorial change is no longer needed.*

Comment 47. Page 97. The basis for the following important statement is not given or referenced: "Each of these have been discussed in some detail and ruled out as significant sources of systematic error in the effluent measurements, in many cases, because of correction factors used by the plant to compensate for these errors." As noted in an earlier comment, additional support is needed for the decision to dismiss these factors which are listed on pages 96 and 97. Sampling probes have been described as being behind support structures; why would such a sample be representative?

Response 47. *The referenced subject matter has been relocated to Section 4, "Uncertainty in Airborne Emission Estimates." Additional information regarding the likely magnitude of contributors to uncertainty has been added in earlier sections and in appendices. For example, see discussions of recent studies of sampling system effectiveness (Nininger & Osborne, 1992) and analyses of impacts of sample line deposition and anisokinetic sampling.*

Comment 48. Page 98. If you believe that there was a bias toward lower flow rates, why is a multiplicative correction factor of 1 considered the "most probable"?

Response 48. *A most probable correction factor of 1 is no longer applied. A sample flow rate correction factor distribution is now discussed in Section 2.2.1.2 and Section 4. It is a distribution of observed flow rates divided by 56.6 L min^{-1}, the flow rate assumed by plant staff when estimating particulate emissions. The distribution, which is assumed to be normal, is shown in Figure 2-2 of the revised report. The distribution has a mean of 0.92 and a standard deviation of 0.11.*

Comment 49. Page 98. How was it determined that a triangular distribution was most representative of the distribution in effluent flow rates? Is it based on data in Figures 2–2 and 2–3 and similar information? An explanation and plot would be useful in this section.

Response 49. *Very little information was identified during the Phase I study that supports quantification of variability of exhaust flow rates. Uncertainties were estimated after review of information such as that depicted in Figures 2-4 and 2-5, which reflect flow rates for Buildings 771 and 883A based on back-calculations, engineering estimates, and flow totalizer measurements. These data suggest that errors in the range of 50 percent, and on the high end no more than 100 percent, could plausibly be associated with the use of fixed effluent volume assumptions in the 1950s, 1960s, and early 1970s. The uncertainty in emission estimates due to the lack of documentation of exhaust flow volumes was represented by treating emission estimates as triangular distributions with lower and upper bounds at 0.5 and 2 times the reported value, with the most probable value equal to the reported value.*

Comment 50. Page 98. The recommendation is made that plant release estimates should be adjusted, annually, for measurement errors tabulated in Table 2-23. These relative errors are essentially quality assurance measurements, to ensure that analytical techniques fall within some acceptable level of accuracy. However, correcting the release estimates based on the bias in that year's QA samples may not be a good idea. First, it should be established definitively whether or not the plant already corrected the estimates (see page 98). Also it is important to assess the degree of confidence on the average annual relative errors in Table 2-23. You should look at the number of samples, variance,

and perhaps the temporal trend over a year before concluding that a correction is appropriate. Again, uncertainty analysis is called for.

Response 50. *Release estimates are no longer altered based on reported annual average relative analytical errors. Reported relative analytical errors are, however, reflected in the uncertainty analyses for each emission type as described in Appendix G.*

Comment 51. Page 98. No reference is given to support tritium sampling efficiency. No justification is provided for the distribution.

Response 51. *References to the Section 2.2.1.1 discussion of tritium sampling efficiency and to the special study documented by Hurley (1979) were added to Section 4.2.*

Comment 52. Table 2-23. What is the basis for the values given in Table 2–23? How many comparison measurements were used to estimate the biases shown? What was the distribution of observed biases? The same "new spectrographic technique" was surely not used for all the constituents of the effluent shown in the table. Were the estimates for 3H (not H3) and Be made in a different way?

Response 52. *The data contained in the former Table 2-23 are based on laboratory analyses of control samples, and were reproduced from Rocky Flats annual environmental reports for 1974 through 1989. The values are described as follows in the 1976 report: "The ratio of the standard deviation of the six-month differences to the average standard value in percent; i.e., observed value minus standard value, divided by average standard value, times 100 equals the ratio as expressed in percent."*

The "new spectrographic technique" applies only to the alpha spectral analyses of plutonium, uranium, and americium. Beryllium analyses were performed by atomic absorption analysis, and tritium was quantified by beta liquid scintillation.

The numbers of control analyses upon which the reported relative errors are based are identified in some of the annual reports. Numbers ranged between 25 and 240 control samples per analysis type per year for analytes in airborne effluents. Ranges of relative errors (in addition to annual averages) are presented for 1987 through 1989 only.

Comment 53. Page 98. Show the distribution of observed correction values for tritium collection efficiency to convince the reader that they can be represented by a triangular distribution.

Response 53. *Data available to us at this time do not support the suggested presentation.*

Comment 54. Page 100. If there were measurement biases of various magnitudes between 1974 and 1989, is it reasonable to assume that such biases did not contribute to the overall uncertainty in the release estimates for earlier years?

Response 54. *Uncertainties of measurements performed before 1974, including contributions from analytical procedures, are addressed in Appendix G.*

Comment 55. Figure 2–9 requires revision.

Response 55. *The plots of tritium emission estimates and uncertainty bounds have been improved.*

Comment 56. Table 2–24. The first 21 years in the "analytic error adjusted emission estimate" columns should be blank to be consistent with the way the estimates were made (see Table 2–25).

Response 56. *Due to restructuring of the uncertainty analyses and related tables, this comment no longer applies.*

Comment 57. Tables 2–24 and 2–25. The bounding values are not consistent within these two tables. In the early years, the complete distribution is represented, while the 95% confidence interval is used for later years. Consistent use of the 95% confidence interval could be accomplished by simple calculation.

Response 57. *Emission estimates and uncertainties for all radionuclides and beryllium are specified with geometric means and 95 percent lower and upper confidence limits. Methods for determination of overall uncertainties with these emission estimates are described in Appendix G. Plausible ranges of historical emissions of organic solvents are specified with lower and upper bounds. These values are provided to support screening to determine appropriate levels*

of concern. The estimated ranges of emissions derived from review of the available data are believed to encompass the actual emission rates.

Comment 58. Table 2-25. As indicated by previous comments, adopting some of the DOE Pu and U release estimates and then describing them as "most probable" is difficult to accept. The "most probable" estimates of ^3H releases for the years 1953–67 and 1968–73 are 400 Ci and 270 Ci, respectively, according to the ChemRisk assumptions of uniform distributions for those periods. However, given the way the upper bound of 800 Ci was obtained, it may be understandable that the column was left blank. The assumption of a uniform distribution should perhaps be reconsidered. Footnote 3 to Table 2–25 contains the wrong GM.

Response 58. *Methods have been revised as described in Section 4 and Appendix G. "Most probable" values for early tritium releases are not provided. The selection of a uniform distribution reflects the lack of specific information regarding releases during this period—releases within the described range are believed to be equally likely for these periods.*

Comment 59. Page 120. The spelling of (Bukowski,1968) is not consistent with its citation in the reference section.

Response 59. *The requested editorial change has been incorporated. The correct spelling is Bokowski.*

Comment 60. Page 120. The detection limit for the third analytic procedure for beryllium is reported in ng. It would be more helpful to express it in μg as for the other procedures.

Response 60. *The requested editorial change has been incorporated.*

Comment 61. Page 120. The efficiency of the direct flame is given as 2 μg per filter paper, this does not appear consistent with the detection limit given for it in the previous paragraph (0.003 μg). Is there a reason for this? Is one of the units incorrect or does the 2 μg refer only to Gelman Type E filters or is there an alternative explanation?

Response 61. *The detection limit of 0.003 μg is per milliliter in aqueous solution. This additional information has been added.*

Comment 62. Page 121. Explain why one-half the reported value was used to calculate the average beryllium concentrations in stack air, in cases where less-than values were reported What percentage of Be measurements were less-than-detectable? Use of " <x/2 " in computing averages could have introduced bias. It's better to do probability plotting to estimate the distribution shape and determine a central estimate and uncertainty.

Response 62. *Additional information concerning the practice of replacing less-than values with one-half the limit of detection is provided in Section 3.1.2. In order to evaluate the impact of this method on the release estimates of beryllium, a more detailed analysis was performed on the 1962 and 1968 beryllium monitoring data. Although about 30 percent of the air monitoring data reported in 1962 were below the applicable detection limit (0.0001 μg m^{-3} in most instances), their impact on the release estimate for the year was relatively small. This was demonstrated by calculating annual averages setting less-than values equal to the detection limit and then setting them to zero. The difference between beryllium release estimates for 1962 calculated by these two methods was less than 2 percent. In 1968, less than 1 percent of beryllium air monitoring results were below detection limits, and the difference between release estimates calculated by the two methods described above was less than 1 percent.*

Comment 63. Table 3-1. Annual average beryllium concentrations in stack air are provided. This includes buildings 886, 889, 865, and 774 for the year 1970 because a broader sampling program was initiated. How should these values be interpreted? Do they indicate beryllium activities occurred in these buildings or was it the case that all measurements were below detection limits,with half the detection value being taken for calculation purposes?

Response 63. *The information regarding a broader sampling program has been added to the text. However, in review of the data, it was determined that buildings 889 and 865 were not part of the beryllium air sampling program during that time period. Therefore, the data for these buildings have been removed from the table.*

Beryllium was not machined in Buildings 774 or 886, but may have been present in wastes handled in Building 774 and parts handled in Building 886. Results below limits of detection were rare in 1970.

Comment 64. Tables 3-1, 3-2. Simply stating ND for no data is misleading. A distinction should be made between years when no known beryllium activities took place in a building and therefore no beryllium releases could be assumed, and years where estimates for beryllium releases from a building are unavailable, but should be assumed.

Response 64. *The requested distinction has been made. "NA" is used to indicate periods during which no beryllium processing activity is believed to have occurred. "ND" is used to indicate periods when beryllium processing is believed to have occurred, but for which monitoring data were not located.*

Comment 65. Table 3–2 contains a release estimate for Building 447 in 1962, but there were no data for that year according to Table 3–1.

Response 65. *The appropriate correction has been made. No beryllium monitoring data were located for Building 447 in 1962.*

Comment 66. Page 124. Were any independent estimates of Be releases made and compared with the values in Table 3–3? If so, what were the results of those comparisons?

Response 66. *A independent reconstruction of beryllium emissions during 1984 was performed and is described in Section 3.1.2. Results are given in Table 3-4.*

Comment 67. Table 3.4. ** is used to indicate that the plant reported a less-than value, and that a value equal to the reported value was conservatively assumed. How can a "most probable value" be based on an assumption that is known to be conservative? The release estimates for the years 1972-80 are clearly conservative estimates; a best estimate procedure was not begun until 1981. It is not clear whether the estimates for the early years are biased; more information is needed regarding unmonitored releases during that period. Conservative assumptions can be used in the absence of data, to help determine the upper bound to the emission estimate. Given the paucity of

data it may be more useful to define an upper and lower bound to a most probable range rather than a single most probable value, thus giving a "top hat" distribution.

Response 67. *Methods for determination of release estimates have been revised as described in Sections 3 and 4 and Appendix G. "Most probable" values of beryllium emissions are no longer provided. Emission estimates and uncertainties for beryllium are specified with geometric means and 95 percent lower and upper confidence limits. Methods for determination of overall uncertainties with these emission estimates are described in Appendix G.*

Comment 68. Pages 130–131. It is not clear how the estimated range of carbon tetrachloride (CCl_4) emissions (40–120 tons per year), for the period 1958–1970, is selected based on the information presented in Appendix C. For example, in Table C-4 values ranging from 34–153 tons per year, are presented. Since the time period, 1958–1970, is reported to correspond to the period of maximum use for CCl_4, it does not follow that a value smaller than the one estimated in 1974–1975, at which time "attempts were being made to eliminate carbon tetrachloride from some production operations", should be selected. Are the quality of some data considered better than others? What criteria are used to discount data? More explanation is required here.

Response 68. *The upper bounds of plausible carbon tetrachloride have been revised to reflect available information. The upper bound of 200 tons per year is approximately 30 percent above the maximum release rate (153 tons per year) that corresponds to peak levels of carbon tetrachloride measured around 1974/1975 by Hobbs (1982). The upper bound is above the estimated 1974 peak emission rate to account for some decrease in carbon tetrachloride use beginning around 1972. The fact that carbon tetrachloride usage was in a downward trend was likely offset somewhat by the fact that the 153 tons per year estimate reflects peak measurements rather than average emissions over long periods of time. The elimination of uses of carbon tetrachloride at Rocky Flats was neither immediate nor comprehensive- significant uses remained in 1970. In fact, at the time of plant shutdown in 1989, procedures still called for use of carbon tetrachloride in plutonium facilities.*

Unlike radionuclides and beryllium, there were no efforts at Rocky Flats to control organic solvent emissions by use of exhaust system recovery devices, scrubbers, or traps. Because of this fact, significant fluctuations due to failure of control devices did not occur. Emissions were more directly related

to the quantities of each solvent that were historically used and the fractions of the quantities that were used that became airborne.

Comment 69. Figure 3-2. Is the factor-of-8 step increase in CCl_4 usage in 1958 justified or should a linear change over time be employed here (as indicated in the text on page 129)? A similar concern applies to Figure 3–6. Figure 3–4 is inconsistent with the text.

Response 69. Linear increases from the carbon tetrachloride emission bounds for 1953-1957 to those for 1961-1970 have been adopted. However, we caution that we have no specific knowledge regarding the rate of change in usage— ramp-up on the use of a solvent may have occurred over 6 months or 3 years.

Comment 70. Pages 130–131. It appears that the basic assumption is that all CCl_4 that was used volatilized and is therefore considered as an airborne emission. This approach seems justified, especially for determining a maximum emission rate. How is the CCl_4 that is known to have been stored in drums and subsequently disposed of at pad 903 taken into consideration? How much could have been released to water and could drinking water have been a significant exposure pathway? Estimates of the quantity of CCl_4 that was disposed of via these and other routes would allow these pathways to be quantified as well as providing a method for determining a lower bound to the airborne emissions.

Response 70. A discussion of potential carbon tetrachloride emissions from 903 Pad drums has been added to Section 3.2.1. If all of the carbon tetrachloride estimated to have been contained in the drums (about 260 tons) volatilized, emissions could have approached approximately 13 percent of the upper-bound annual release estimate for the period. As part of the surface water discussion we have provided appropriate references to support the assumption of nearly 100% evaporation of volatiles from surface waters.

Comment 71. Figures 3-2 through 3-6, Tables 3-5 through 3-10. The figures and tables showing inventories and emission rates for organics should contain two scales and two columns, respectively, that clearly separate these distinctly different quantities. Were there no estimates of uncertainty given in any of the referenced documents? Certainly the monitoring results indicated variability, but this information has been lost in the compilation of the tables (for

example, the last entry in Table 3-5 could surely show a range of emission rates).

Response 71. *The tables of inventory and emission estimates for the organic solvents have been revised to include separate columns for inventory data and emission estimates. It was not necessary to use multiple scales on the emission estimate graphs to include both inventory data and material use or emission estimates. While uncertainties of inventory, usage, and monitoring values are not well characterized in the referenced reports, some ranges are provided that indicate variability.*

Comment 72. Table 3-5. Reference is made to (Johnson, 1973) for the monitoring of CCl_4 emissions from booster 1. In Appendix C, however, the reference for this monitoring is given as Dow Chemicals, 1974a. A reference to Johnson, 1973a is also given in Appendix C, but it does not correspond exactly to Johnson, 1973 which appears in the main text.

Response 72. *The (Johnson, 1973) reference in Section 3 is the same document as (Johnson, 1973a) in Appendix E; it is repository document CH-850. Listings in the references sections have been standardized to eliminate the potential for confusion. The report cited as (Dow Chemical, 1974) in Appendix E is a different reference for the same series of measurements in Building 776/777; it is repository document RE-822. Both documents are based on the sampling of Building 776/777 Booster 1 during June and July 1973. A few of the data points given in RE-822 match data in CH-850. An average emission rate based on data in CH-850 is 12 tons per year, while RE-822 yields an estimate of 10 tons per year.*

Comment 73. Page 134. The document (Barrick, 1984) which is used as a basis for estimating emissions from 1952 to 1974 is not discussed in Appendix C.

Response 73. *We assume that the reviewer is referring to the (Barrick, 1974) citation rather that 1984 as stated in the comment. The Barrick document was discussed in Appendix C of the draft report, referred to as (Dow Chemical, 1974b). This 1974 hazardous material inventory is now (Barrick, 1974) in Appendix E.*

Comment 74. Page 134. The basis for estimating that chloroform emissions ranged between 1.5 and 15 tons a year, given that a single inventory value of 8.9 tons in 1974 is known, should be given and discussed.

Response 74. *It was estimated that chloroform supplies were replenished at a rate such that approximately two-times the indicated inventory quantity (or about 18 tons per year) were used and released each year. Based upon the reported nature and extent of chloroform uses, it was unlikely that the replenishment rate for the solvent was significantly greater than this or that annual emissions were less than approximately one-quarter of the quantity kept on hand.*

The 1989 value that defines the lower bound of plausible chloroform emissions for this period (.5 ton per year) is approximately 25 percent below the APEN chloroform emission estimate and slightly below the 1988/1989 inventory quantity for chloroform. In the later years of Rocky Flats operations, uses of chloroform were relatively minor. As a result, there was an increasing likelihood that minor operations contributing to the site emission total went unidentified. While the APEN assumption of complete volatilization likely tended to cause emissions to be overstated, it is also possible that unidentified minor chloroform emission sources existed and that inventories reported in 1988/1989 underwent some replenishment during each year.

Review of available information indicates that it is unlikely that emissions of chloroform in the late-1980s exceeded the 1988/1989 inventory quantity by a factor of ten. As a result, the 1989 chloroform emission rate that defines the upper bound of plausible chloroform emissions was estimated to be 5 tons per year.

Comment 75. Page 134. It is not clear how the upper values of 15 and 1.5 tons per year, in 1974 and 1989, respectively are established.

Response 75. *See the response for comment number 74.*

Comment 76. Table 3-6. Splitting the final column into two would allow emission rate estimates to be distinguished from inventory estimates. It would also be very helpful if a more detailed discussion were to be included about the uncertainties associated with using inventory data to estimate emission rates. Are values based on purchase records considered equally as useful?

Response 76. *The tables of inventory and emission estimates for the organic solvents have been revised to include separate columns for inventory data and emission estimates. Inventory reports and purchasing records identified during the Phase I study are not extensive or complete enough to support a quantitative uncertainty analysis of these information sources.*

Comment 77. Page 137. The document (Barrick, 1984) which is used as a basis for estimating emissions from 1952 to 1974 is not mentioned in Appendix C.

Response 77. *We assume that the reviewer is referring to the (Barrick, 1974) citation rather that 1984 as stated in the comment. The Barrick document was discussed in Appendix C of the draft report, referred to as (Dow Chemical, 1974b). This 1974 hazardous material inventory is now cited as (Barrick, 1974) in Appendix E.*

Comment 78. Page 137. The basis for estimating that methylene chloride emissions ranged between 5 and 15 tons a year, given that a single inventory value of 2.2 tons in 1974 is known, should be given and discussed.

Response 78. *Based upon the 1974 inventory quantity of 2.2 tons, purchasing record indications of replenishment during the year, and the indication in worker interviews that methylene chloride usage was significant before 1974, it is unlikely that emissions from 1953 to 1974 were less than 3 tons per year. The plausible upper bound of annual methylene chloride emissions is consistent with an ordering frequency of between five and seven times per year observed in warehouse purchasing records (EG&G, 1974-1988) and the 1974 inventory of 2.2 tons.*

Based upon the 1989 inventory of 0.31 tons of methylene chloride, documented replenishment rates during the 1980s, and the APEN emission estimate of 3.3 tons per year, it is unlikely that annual emissions of methylene chloride were less than 0.5 ton per year.

The 1989 point (5 tons per year) that defines the upper bound of methylene chloride emissions is based upon the APEN emission estimate of 3.3 tons per year and the possibility that; due to the continued widespread U.S. use of methylene chloride in paint strippers, pesticides, and certain aerosol products (ATSDR, 1991); unidentified minor methylene chloride emission sources existed. It is unlikely that actual emissions significantly exceeded the APEN

estimate, however, due to the conservative assumption in the APEN assessment that all methylene chloride that was used was lost to the atmosphere.

Comment 79. Figure 3-6. The upper and lower estimates for the plausible range of 1,1,1-trichloroethane emissions show an abrupt stepwise change at the end of years 1957 and 1974. Given the paucity of documented information would it not be more reasonable to assume more gradual increases in emission rates occurring over one or more years?

Response 79. *Linear increases have been adopted; see Section 3.2.5 and Figure 3-5. However, we caution that we have no specific knowledge regarding the rate of changes in usage.*

Comment 80. Page 146. It is stated that the estimated range for TCA emissions during the period 1974 to 1984 is conservative in the sense that it overestimates emissions. Can a non-conservative lower estimate be provided? Otherwise there is a danger that the real value could lie outside the plausible range given.

Response 80. *Bounds of plausible 1,1,1-TCA emission have been revised as discussed in Section 3.2.5 and Figure 3-5.*

Comment 81. Page 168. Baseline Reservoir as "background" will probably be hard to support in the public arena. Information should be collected from other regional waters and the complexities explained more thoroughly. Many people would perhaps assume airborne deposition at 6 miles would affect concentrations in the reservoir. Same comment applies to page 171 re: drinking water.

Response 81. *We contacted USGS, USEPA, CDH, and the Northern Colorado Water Conservancy in an unsuccessful attempt to locate a pre-1971 gross alpha or radionuclide data set for surface water bodies at a distance greater than Baseline Reservoir. It is possible that Baseline Reservoir has been impacted by routine airborne emissions from Rocky Flats. However, because Baseline is not in a predominant downwind direction from the plant, the magnitude of deposition is not expected to be large. To illustrate this point, Fugitive Dust Model predictions of dispersion of releases from the 903 Pad showed that only about 0.07 percent of emissions from the Pad deposited in the north sector*

between 10 and 20 kilometers from the plant. Baseline Reservoir is 6 miles or about 10 kilometers from the Rocky Flats Plant.

There have been some concerns voiced about possible notices of violation for high radioactivity in the water of Baseline Reservoir. Follow-up by Ann Lockhart of CDH and ChemRisk with the CDH Water Quality Control Division, the Radiation Control Division, and the Public Service Company led to the conclusion that the identified problem was with the Valmont Reservoir. Further investigation with Gail Biggs regarding these findings led to his further questioning whether Valmont Reservoir drained into Baseline Reservoir. Discussions with Judy Brunch of CDH and review of USGS maps suggest that Baseline is up-gradient of Valmont and that it is more likely that discharges flow from Baseline to Valmont rather than the other way around.

Comment 82. Page 172 et seq. It is not clear whether the solvents evaporated from surface waters are included in the atmospheric source term amounts. Is there sufficient evidence from the literature to support the assumption of 100% volatilization of the chemicals from surface waters? It would appear that the quantity of chlorinated hydrocarbons that were released to water needs to be checked.

Response 82. *References to support the assumption of 100 percent volatilization of solvents from surface waters have been provided at the beginning of Section 5.0.*

Comment 83. Page 172. The statement is made that "data were not collected on the waterborne effluents for any of the non radioactive chemicals of concern." What types of chemical data were collected? At the least, these data should be referenced, as they may be useful to understanding water chemistry. They also could even help to support that no other non radioactive contaminants of concern were present (other metals, e.g.).

Response 83. *The other types of chemical data that are available have been added to Section 5.2.2 of the report.*

Comment 84. Page 172. There should be some agricultural or research data for elements (including Be) in regional soils and perhaps sediments. A possible lead is the Soils Testing Laboratory at Colorado State University. If establishment of background levels of Be is important, then this avenue should be investigated further.

Response 84. *Discussion of measurements of beryllium in reservoir sediments has been added to Section 5.2.2 . "Typical" levels of beryllium in Colorado soils are also described.*

Comment 85. Page 173. Indicate where in the reservoirs the monitoring was done, or if a variety of locations were sampled. Depending on other sources of inflow/runoff, it could make a difference to the conclusions made in the subsequent pages (e.g. Table 4-3) if the monitoring was done near the inlet drainage vs. the opposite part of the water body, after dilution and mixing.

Response 85. *Locations of reservoir monitoring are not documented in the Rocky Flats Environmental Survey reports. We are unable to elaborate without further investigation.*

As described in Section 5.4.1, gross alpha concentration values presented in Table 5-4 were calculated based on reported Rocky Flats emissions and some conservative assumptions regarding dilution. These are not monitoring results.

Comment 86. Section 4.4.1. The referenced "nearly twofold" increase in the release was actually more than a factor of two. Compare the three years before and after the period 1959–1962. An appropriate comparison between the release and reservoir concentration data would reflect the year to year variations in the reservoir's volume. A time history of reservoir volume should be sought. Before concluding that the increase in reservoir concentration in 1966 is correlated with plant effluents, the question of reservoir volume must be addressed as well as the impact of the ^{238}Pu contribution from the SNAP-9A power supply.

Response 86. *The suggested editorial revision has been made. We agree that data on reservoir volume and reservoir inflow and outflow are critical to understanding the relationships between releases and reservoir concentrations. We are not aware of the availability of reservoir volume data for the period*

prior to 1971. As no data on the volume of Great Western Reservoir prior to 1971 were located, annual rainfall at Rocky Flats was investigated as a possible surrogate of reservoir volume. See Section 5.4.1 for this discussion.

A discussion of possible contributions of Pu-238 from the abortive reentry of the SNAP 9-A navigational satellite power supply was added to Section 5.4.1.

Comment 87. Page 174. It seems odd that the second time period, 1971–1989 has less information on plant effluents to surface water than the previous period. Usually the reverse is true.

Response 87. We agree that it is unusual that less information on releases to surface water is available for the second time period. The paucity of data could be tied to the adoption of a "zero-release" policy by the Rocky Flats Plant in the mid-1970s.

Comment 88. Page 174 and subsequent pages. It is unfortunate that you must rely on gross alpha for your comparison, since these are so heavily influenced by natural radionuclides. You should indicate in this presentation whether or not the water was filtered, as this could greatly affect the amount and variability of gross alpha associated with suspended particulate.

Response 88. We are unable to elaborate on the methods used to analyze the samples without additional investigation.

Comment 89. Page 176. Table 4-2 should indicate that releases listed are gross alpha.

Response 89. The suggested change has been incorporated in Table 5-2.

Comment 90. Page 177. Are there data for "water inflow and outflow rates" which influence contaminant dilution? Have they been located and examined?

Response 90. The Phase I investigation did not result in the identification of any data on inflow or outflow rates for the period prior to 1971.

Comment 91. Page 179. A correlation coefficient of 0.9 (between radioactivity levels in Standley Lake and GWR) would qualify as more than "fairly closely correlated".

Response 91. The suggested editorial change has been incorporated.

Comment 92. Page 179, subsequent figures, and Table 4-4. It would be good to state here whether the drinking water analyzed was "raw" or "finished". Analysis of only annual averages in drinking water could mask important differences in the tails of the distributions of concentrations. Could some other percentiles of the distribution (e.g. the 5%, 25%, 75%, and 95%) be derived and presented? If there are insufficient samples to do this for a given year, perhaps pooling a number of years would be worthwhile for this comparative purpose.

Response 92. The drinking waters that were analyzed were "finished." Drinking water data with finer resolution than annual averages have been added to the report as described elsewhere.

Comment 93. Page 180. Figure 4-9 is a good comparison of trends which helps to support the conclusions. As mentioned previously, it would be useful to include another regional water body which is even further away than Baseline reservoir. Also do data exist to extend this graph into the period in the 1970s when holding pond reconstruction resulted in resuspension of pond sediments into Great Western Reservoir (page 159)?

Response 93. As mentioned in 81, we have not located comparable data for other more distant reservoirs. Drinking water monitoring data for the cities of Broomfield and Westminster taken between 1971 and 1975 with finer resolution than the annual data were plotted. See Figure 5-20 and Section 5.5. Elevated levels of Pu-239/240 in Broomfield water between 1972 and 1973 are evident in this figure, and may be attributable to pond reconstruction activities.

Comment 94. Page 186. As mentioned above for the reservoir data, computing and analyzing only annual average concentrations of radionuclides in drinking water (CDH data) could mask some important differences in the distributions. Other percentiles should be computed and/or a finer time resolution (e.g. monthly) should be examined. However this may be difficult since so many

of the samples contained less-than-detectable concentrations. Examination of data on a finer time resolution is warranted during the period (1972–1974) of pond reconstruction.

Response 94. *As mentioned above, higher-resolution drinking water monitoring data for Broomfield and Westminster from 1971 through 1975 have been plotted. See Figure 5-20 and Section 5.5. Elevated levels of Pu-239/240 in Broomfield water between 1972 and 1973 are evident in this figure, and may be attributable to pond reconstruction activities.*

Comment 95. Page 187. The reported concentrations for specific radionuclides in drinking water require investigation. For example, there needs to be an explanation for the increase in Pu-238 in 1975 (Figure 4-12) and Pu-239/240 in 1975 and 1984 (Figure 4-14). It is unacceptable to leave the reader hanging about something so obvious. Is it known why there were no data for 1976 and 1977? It may be most appropriate to consider the data for a particular reservoir as a censored distribution and determine the form and central measure for that distribution.

Response 95. *We recognize the anomalies in the data but are unable to explain the high values in background. We have been informed that background drinking water data for Pu-238 and Pu-239/240 for 1976 and 1977 are unavailable because either the lab or measurement station was not operative during those years.*

Comment 96. Page 192. Another source of information for regional concentrations of radionuclides in drinking water (as well as surface water and air) is the U.S. EPA's Environmental Radiation Data series, published quarterly. They probably have several monitoring locations in the Denver area.

Response 96. *Some USEPA Environmental Radiation Data series information has been acquired and will be added to the project information repository.*

Comment 97. Page 193. Can the accidental tritium release be used as a "tracer" study to indicate the degree of contaminant transfer from Rocky Flats to drinking water? This would be an upper bound for release of contaminants which are particle-reactive. The hydrological conditions at the time would need to be

determined so that the conclusions would not be inappropriately applied to other hydrological conditions.

Response 97. *This is an interesting possibility. However, a good record of hydrological conditions would be important if this were pursued, and, to-date, such information has not been located.*

Comment 98. Page 198. Summary box. It is stated that "available data are not sufficient for reconstructing contaminant releases in surface water from the plant." However, no alternative recommendations are made. Also, it is not well established that the sediment data are not useful for reconstructing deposition patterns (spatial and temporal) if not amounts. For example, Hardy et al. (1980) used mass isotopic analysis of Pu isotopes to differentiate global fallout and Plant contributions in a sediment core collected from Standley Lake (downwind of the Plant). They observed a rather definite time-pattern of Pu deposition including a peak in the transuranic concentrations during late 1969, which corresponded with the peak release from Pad 903. Thus, we believe the sediment data are likely to be quite important in this regard.

Response 98. *If this issue is to be pursued further, the most likely alternative to be pursued is the evaluation of sediment data, particularly the information that will be generated as part of the on-going Superfund investigations, as well as the study you have cited.*

Comment 99. A variety of soil studies are noted in the report, which have been carried out at various times in the Rocky Flats area to estimate radionuclide releases from the facility. It would appear, however,that an independent review of these data is called for to see if further insight into the source term can be gained. While the organic source term would not be amenable to retrospective soil analysis, the longer lived radionuclides and Be would be.

Response 99. *The soil studies are discussed and relied on in the Task 6 report to support the reconstruction of the 903 Pad releases. Independent review of soil data could logically occur as part of Phase II efforts.*

Comment 100. Page 203. What evaluation has been done of the amount of Pu which could have been on the HEPA filters and thus possibly released during the fire? The alleged amounts of 10 to 250 kg (Church lawsuit) dwarf the total source term estimates in the 1 Ci (14 gram) range. Is there a published reference for the Church lawsuit arguments that suggest this release range for during the 1957 fire? The basis for these estimates must be discussed. Instead of presenting the various release estimates and letting them "hang there" without any evaluation, it may be more appropriate to defer all consideration of the accident source terms to the Task 6 report.

Response 100. *As requested, detailed considerations of accident source term issues has been deferred to the Task 6 report. In most cases the additional information requested in these comments is provided in the revised Task 6 report.*

Comment 101. Page 203. Are there suggestions of time scenarios other than the one given on page 203? The text implies that there are but none are discussed. Does Langell give any times for the installation of temporary filters?

Response 101. *See Response to comment number 100.*

Comment 102. Pages 204-5. There is a large variation in the quoted studies relevant to particle sizes released from the 1957 fire. Has ChemRisk done additional evaluations and developed recommendations for the dose reconstruction?

Response 102. *See Response to comment number 100.*

Comment 103. Page 205. The section titled "Emission temperature and release velocity." The purpose of the dose reconstruction is not to address a scenario which results "in greatest community exposure," but rather to develop the best estimate of what actually happened with associated uncertainties. The reference to personal communication in this section, while perhaps germane, is not enough. A calculational, engineering approach is necessary also.

Response 103. *See Response to comment number 100.*

Comment 104. Page 205. The section titled "Meteorological Data." Give a better indication of what kind of meteorological data are contained in the Dow letter. Where were the data taken? What was the frequency of sampling?

Response 104. See Response to comment number 100.

Comment 105. Page 205, last paragraph. Are the ambient air data referenced by Hammond (1971) available in a more contemporaneous source?

Response 105. See Response to comment number 100.

Comment 106. Page 206. Barrick refers only to offsite sampler measurements. Were there no onsite air monitors operating? What about the one referred to in the previous paragraph by Hammond? This section needs a map showing the locations of the fire and the samplers. Locating the original data sheets for these offsite and any onsite air monitoring stations during the 1957 fire is crucial to the quality of the dose reconstruction. Referencing these summary sources is not convincing.

Response 106. See Response to comment number 100.

Comment 107. Page 207-8. The section titled "Modeling Approach for the 1957 Fire." The approach outlined here may be adequate for a first cut, but we do not believe it is adequate for a dose reconstruction. Making use of the vegetation data is good, if uncertainty analysis is carried through the computations. However, other angles must be used as well (e.g. starting in-plant with inventories, release fractions, ventilation rates, etc..). All available methods of reconstructing this incident must be integrated to evaluate consistency, gaps, uncertainties.

Response 107. See Response to comment number 100.

Comment 108. Page 210. A citation for the Fugitive Dust Model should be given in the first paragraph of "Modeling Approach for the 903 Pad."

Response 108. See Response to comment number 100.

Comment 109. Page 211, first paragraph. Provide the current reference for this study (i.e., Webb 1992).

Response 109. See Response to comment number 100.

Comment 110. Page 211. The contamination from the 1965 glove-box drain fire was "released through the main exhaust plenum filters ... incorporated in the normal emission data and does not have to be modeled separately." Shouldn't the amount be separated from the routine releases for dose assessment purposes? That is, the meteorology for that particular day should be used to evaluate the dispersion pattern. Same comment applies to the 1969 fire (page 212).

Response 110. See Response to comment number 100.

Comment 111. Page 213, last paragraph. A defensible dose reconstruction would look at these original data and draw an independent conclusion, not stop at citing a "Rocky Flats analysis," which would have zero credibility with most of the public.

Response 111. The analysis of impacts of the 1969 fire went well beyond review of the Rocky Flats analysis. Further analysis and interpretation are provided in the Task 6 report.

Comment 112. Page 216, last sentence. The range of measurements cited could not support the Rocky Flats conclusion that "burning oils contaminated with depleted uranium did not pose a health hazard." Without additional interpretive information (e.g. a simple dose calculation), all that can be said is that the burning raised the alpha concentration in air by up to a factor of 10.

Response 112. Further analysis and interpretation are provided in the Task 6 report.

Comment 113. Page 217. The lack of a method for evaluating the source term from Oil Burn Pit 1 could perhaps be accepted in that this burn was a small amount (10 drums) compared with Oil Burn Pit 2 (>1354 drums). However, some estimate for Pit 2 burning is necessary, and should be possible, with uncertainties, from the air monitoring data and/or other information. The

radionuclide content of the waste was apparently not measured. What about the residue? Can the amount of U in the oil be bounded? The technical literature provides data on release fractions of U from burning contaminated oils, which could be used to complement the site-specific measurements.

Response 113. *Further analysis and interpretation are provided in the Task 6 report.*

Comment 114. page 221. Re the statement: "In addition, a source term for a potential release cannot be constructed due to the lack of any information on what was actually burned and in what quantities." This type of statement calls into question the completeness of the entire dose reconstruction unless it can be confirmed that these unquantified releases are much less than those that are quantified.

Response 114. *An estimate of the quantity of uranium released to the air from the 1965 burning of a pallet containing 60 kg of depleted uranium has been added to Section 6.2.2.*

Comment 115. A plot of a lognormal PuO_2 particle size distribution that shows the relationship between the count median and mass median diameters needs to be included to illustrate the definitions given on page A-1.

Response 115. *Figure A-1 has been added. It shows a log probability plot of particle sampling data and illustrates the relationships between the data, mass median diameter, geometric standard deviation, and count median diameter.*

Comment 116. A figure that shows the collection efficiency for HEPA filters needs to be included to bolster the statements on page A-2 regarding expected particle size.

Response 116. *A figure depicting the fractional penetration of particles from 0.01 to 10 micrometers in diameter through a standard HEPA filter with no leakage has been added to Appendix A. It is Figure A-2.*

Comment 117. Tabulations or figures that describe more completely the results of the particle size measurements discussed should be included. The tabulation should include all of the studies, not just a selected few (page 20 of the main text indicates that there are others, perhaps many more). Were the distributions

measured by Hayden lognormal? If so, what was the GSD? If not, what was the shape of the distribution?

Response 117. *A summary table of particle size methods and results has been added. It is Table A-1. Additional details from the cited studies have been added to the Appendix A discussion.*

Comment 118. Page A-3. The final study by Elder et al. raises questions that need to be resolved. If there were HEPA filters upstream of the sampling point (the text says "prior to final stages of HEPA filtration") it would be extremely surprising to find such large (3 to 5 μm) particles. Is "were predominantly collected" intended to imply that small particles were not collected efficiently using their method? More explanation and discussion of these results is needed.

Response 118. *Sampling locations for the referenced study were selected in "process or glovebox ventilation ducts making major contributions to the overall activity concentrations incident on exhaust HEPA filters." The article (Health Physics 27, pp. 45-53) goes on to state that "Since these ducts served more than one process and in some cases contained a stage of HEPA prefiltration at the glovebox, wide variations in size characteristics and activity concentration were expected." For the Rocky Flats Building 707 sampling point ("Location Designation E" from Elder et al. per Bill Osborne), high mean percent activities (>10%) on the backup filter indicated that significant quantities of small particles may also be present.*

Comment 119. The reports cited in the appendix should be included as references to the appendix. The citations of Moss and Elder should both include "et al."

Response 119. *The references have been specified within the appendix as suggested.*

Comment 120. It would be helpful to include the terms defined in Appendix A into the Glossary.

Response 120. *The definitions at the beginning of Appendix A have been added to the report glossary.*

Comment 121. Table B-1. The percentage by mass of ^{238}Pu falls outside the stated range of that quantity. The last entry in the row for ^{241}Pu should be 520.7 to be consistent with other information in the table. The second footnote also contains the wrong number.

Response 121. The noted values for mass percentages of Pu-238 are from different references, and are not in full agreement. The values for Pu-241's beta activity multiples of Pu-239/240 alpha activity have been corrected.

Comment 122. Figures B-1 and B-2. The chemical form of the plutonium and americium isotopes that is assumed in the presentation of dose factors should be clearly stated. Tissue by tissue (lung, liver, and bone tissues) comparisons of doses due to intakes of the isotopes would be more relevant to assessment of potential consequence for exposed individuals. Comparison of effective dose equivalents blurs any differences between Pu and Am.

Response 122. The chemical forms of plutonium, americium, and uranium (as indicated by gastrointestinal absorption factors and lung clearance classes) are now indicated in Figures D-1 through D-4. These figures have been revised to provide tissue-specific dose conversion factors for relevant isotopes of those elements.

Comment 123. In Table B-2, both the range and the average of the reported relative analytical errors should be provided. As noted elsewhere, the distributions of these ratios are also important and should be given.

Response 123. This table has been deleted from the appendix, but the data on which it is based are discussed in Sections 4 and in Appendix G. Ranges of relative errors are only provided for 1988 and beyond. Raw data upon which to base evaluations of distributions have not been located.

Comment 124. Table B-3. None of the measured ratios of Am to Pu is as small as the largest ratio (<12%) from results of the theoretical computation (plotted in Figure B-12). What is the relevance of the calculation to the Am/Pu ratio at Rocky Flats?

Response 124. Discussion of the theoretical computations has been deleted from the appendix.

Comment 125. Page B-13. The analysis effectively assumes a uniform distribution of the Am/Pu ratio between the bounds 10–35%. This assumption is not consistent with the data in Table (B-3) and exaggerates the uncertainty associated with all the computed releases of specific radionuclides.

Response 125. *The range of Am-241 to Pu-239/240 observed in Rocky Flats airborne effluents has been revised to between 13 and 31 percent. Methods for evaluation of uncertainties associated with estimating specific isotope releases based on nonspecific monitoring data are now described in Appendix F.*

Comment 126. Page B-16. In the third paragraph, the second sentence should read "U-238 and U-234." Alternatives to "essentially all" would be more appropriate in both sentences; perhaps "nearly all" in the first sentence and "most" in the second.

Response 126. *The suggested editorial changes have been incorporated in Appendix D.*

Comment 127. Page B-16. In the fourth paragraph, it should be pointed out that there was no significant amount of ^{233}U in the uranium processed at Rocky Flats. Recognition of this fact, shown in tables already identified, would simplify much of the subsequent discussion. Even though ^{233}U and ^{234}U may be indistinguishable, there is no need to maintain the fiction that the alpha spectrometric results are equivocal (for example, see the second bullet on page B-18).

Response 127. *The suggested editorial changes have been incorporated in Appendix D.*

Comment 128. Page B-18. State the chemical form of the uranium assumed in the presentations of dose conversion factors.

Response 128. *The chemical forms of plutonium, americium, and uranium (as indicated by gastrointestinal absorption factors and lung clearance classes) are now indicated in Figures D-1 through D-4. These figures have been revised to provide tissue-specific dose conversion factors for relevant isotopes of those elements.*

Comment 129. Page B-21. In the second inset isotopic distribution, the value for U-236 should be 0.0039, not 0.39.

Response 129. The referenced typographical error has been corrected.

Comment 130. Table C-1. Building 774 should be included in the table noting that the carbon tetrachloride emission estimates for it are implicitly accounted for in buildings 707 and 776/777.

Response 130. The requested addition has been made to Table E-1.

Comment 131. Page C-7. In the last bulleted item, the text should read "six 5-day inventory periods."

Response 131. The requested change has been made.

Comment 132. Page C-9. The citation (Dow Chemical, 1974) should read (Dow Chemical, 1974a).

Response 132. As there is now only one Dow Chemical reference for 1974 in Appendix E, the comment does not apply.

Comment 133. Tables C-2. The estimate year for the reference by Kreuhauf and Richter is 1973 not 1974.

Response 133. The estimate year for the Fruehauf and Richter data has been corrected to 1973. The Fruehauf and Richter report was issued in January 1974.

Comment 134. Tables C-2 and C-3. It would be more helpful to organize the information by year rather than information source, so that estimates for the same time periods can be compared more easily. The average emission estimates could be presented in a separate column from the maximum emission estimates.

Response 134. Data have been arranged chronologically in the Section 3 tables, with separate column for inventory data and emission estimates.

Comment 135. Page C-16, in the section titled "Building 881 processes", rather than speculating that the source was probably associated with the laboratories, the findings in the next section below should be noted.

Response 135. The suggested revision has been incorporated.

Comment 136. Page C-17. There is a reference to an APEN for Building 228 A/B. The corresponding release estimate is not given in Table C-6, nor is it mentioned as one of the contributors accounting for <1% of the total release of methylene chloride.

Response 136. The APEN volatile organic chemical emission estimate for Building 228A/B (sludge drying beds adjacent to Building 910), has been added to Table E-6. Methylene chloride was the only VOC detected in the sludge monitoring that formed the basis for that estimate.

Comment 137. There are two references listed for (Grocki, 1989) (page C-40). The one concerning methylene chloride (page C-20) should, perhaps, be designated 1989a and the other as 1989b. In the main text, however, these are listed as a single document (page 153/page 137). A consistent approach needs to be adopted.

Response 137. The two 1989 reports by Larry Grocki are now more clearly cited.

Comment 138. Page C-20. The volume of methylene chloride waste generated in Building 881 reported in Wastren (1991c) is not consistent with the estimate based on the APEN. Two reasons for this discrepancy were suggested. Is it not possible to explore these further and provide better estimates for the volume evaporated and the volume collected as waste and therefore released to a different media?

Response 138. We are unable to elaborate without further investigation.

Comment 139. Page C-26. At the bottom of the page it is stated that building 774 handled TCA wastes received from buildings 707 and 774. Should this read from buildings 707 and 776/777?

Response 139. *Yes. The noted typographical error has been corrected.*

Comment 140. It does not appear that a single density for TCA was used consistently throughout all calculations.

Response 140. *The calculations reflected in Appendix E are based on a 1,1,1-TCA density of 0.006 tons per gallon, or 11.2 pounds per gallon.*

Comment 141. Table C-9. According to the text on page C-29, the total TCA emissions from Building 707 was estimated as 4.1 tons per year from the APENs. A value of 3.8 tons per year is indicated in Table C-9 and Table C-8.

Response 141. *The value of 4.1 tons per year includes an adjusted emission total for the TCA dip tank used in Production Control Operations-Module D (See "Building 707 Processes"). The APEN estimate (0.24 tons per year) did not account for the bimonthly inventory refills or refills due to dirty TCA. APENs values are reported as such in Table E-8. After the reasoning for the correction is given, the revised values are included in Tables E-9 and E-10.*

Comment 142. Page C-31. The material cited in the fourth paragraph for Building 444 is not self-consistent.

Response 142. *No inconsistency is evident. At 11.2 pounds per gallon, 7800 gallons of TCA (650 gal mo^{-1} × 12 mo) corresponds to 87,360 pounds or 44 short tons. The site-wide TCA usage reduction goal was 3900 gallons per year (325 gal mo^{-1} × 12 mo). Part of this campaign was a goal of reducing TCA use by 2700 gallons per year by substituting detergent cleaners for TCA in Building 444.*

Comment 143. Page C-31. The reference cited as "EG&G, 1988" must have been dated 1989 or 1990 because it contains data for the year 1989.

Response 143. *The referenced document does not have an issuance date indicated. Its citation has been changed to EG&G, circa 1990.*

Comment 144. Page C-32. The estimated emission of TCA from building 707 was 4.1 tons per year in 1989, based on APENs (page C-29). However, based on a monitoring report (Martin Marietta, 1990) for the same year, an emission rate of 21.7 tons per year was determined for the same building (Table C-9). This larger value is supported by the internal Rockwell International chlorinated solvent usage report (Rockwell 1988a) from which an emission estimate of 20 tons per year is derived (page C-31). This value was based on a projected usage of 3500 gallons. On page C-32, the report by Wastren Inc. (1991a) is called into question, because the cited generation of 3854.5 gallons of TCA waste per year is considered very high compared to the waste tank throughput reported in the building 707 APEN. Is it possible that it is the emission estimate, based on the APEN (4.1 tons per year) which should be treated with some skepticism. If an emission value of 20 tons per year from building 707 were assumed, the total TCA emission estimate based on the APENs would be 36.7 tons per year. This value is more consistent with the emission estimate of 46 tons per year based on monitoring.

Response 144. The range of plausible 1,1,1-TCA emissions that was determined in the Phase I investigation was based on review of all of the indicated data, and is not inconsistent with any of the values given in Table 3-9. We are unable to elaborate about the apparent inconsistencies between the referenced reports without further investigation.

Comment 145. Page C-35, line 13. The volume of 9790 gallons disagrees with the value in Table C-11. The tabled value is consistent with the other numbers in the table.

Response 145. The Section E.6.2 text is correct. Table E-11 has been corrected.

Comment 146. The lack of correspondence between the text and the tables in the Appendix C and Section 3 is confusing. Some references in the appendix are listed differently from those (for the same work) in the main text.

Response 146. Text and table references have been revised so that they are consistent.

Comment 147. The calculations that were made using the CDH data should be described in an introductory section of text for this appendix.

Response 147. *A detailed description of the calculations has been added.*

Comment 148. Consideration of the data sets for the reservoirs as censored distributions and the display of those distributions would be beneficial to this appendix.

Response 148. *See the Appendix I discussion for analytical methods that were applied.*

Comment 149. Table D-6 is printed twice.

Response 149. *The noted reproduction error has been corrected.*

Comment 150. Text is required to explain the contents of this appendix.

Response 150. *Appendix H has been expanded to present an overview of the methods in which Monte Carlo simulation was used in this project.*

Comment 151. Page 23. Sampling of Airborne Particulate Matter - monitor locations in some of the stacks do not meet EPA siting criteria. There should be checked and noted.

Response 151. *As stated in the report, the effluent sampling systems in place in all 63 monitored dusts at Rocky Flats were out of compliance with 40 CFR 61.93 and ANSI N13.1-1969 requirements as of April 1992. Several relatively recent studies were performed at the plant to evaluate the particle sizes present in Rocky Flats exhausts, the performance of the flow rate monitors that have historically been in place, and the performance of existing sampling systems compared to isokinetic sampling systems. Results of these studies (reports authored by Nininger and Osborne) are referenced in the Task 5 report, and sources of sampling and analytical error and bias were included in the uncertainty analysis for monitored airborne effluents.*

Comment 152. Page 27. Particle Sampling Flow Rates - there are problems with most of the instack monitoring systems at the Plant as they cannot be calibrated in place, therefore, making the air flows somewhat questionable.

Response 152. *See response to comment 151. Sources of uncertainty and bias associated with flow rate measurements were included in the uncertainty analysis for monitored airborne effluents.*

Comment 153. Table 2-2. Average air flow for Building 881, Duct 1, 2, 3 should indicate that it is the total or the three stacks and not an average for each stack.

Response 153. *The table has been annotated as suggested.*

Comment 154. Table 2-4. Total for Total μ for Month should be 246,073 unless using rounding procedures. If so this could be noted.

Response 154. *As stated in the Task 5 report, the convention in the report was to round values to two significant figures.*

Comment 155. Table 2-5. Assume the values listed for the stacks are monthly averages, if so this should be noted.

Response 155. *Table values have been labeled as Average Daily Concentrations.*

Comment 156. Table 2-6. Total μCi Released for the Building 771 Ducts assumed to be an average of the three averages, if so should be noted. Also my calculations show 467, assume the 470 is a round off which should be noted.

Response 156. *A footnote has been added to Table 2-6 that states that the Building 771 release total given is based on the average concentration in the Main 1, 2, and 3 ducts. Per our convention, the value of 470 is rounded to 2 significant figures.*

Comment 157. Table 2-6. My calculation for the Total μCi vary as follows:

Duct	Mine	Table
Main	880	890
Booster 1	0.86	0.91
Booster 2	57.9	55
Booster 3	6.0	5.9

Response 157. *Our values agree very closely with your calculations. Table 2-6 release totals (microcuries) for Building 776 ducts have been verified to be 870 for Main, 0.84 for Booster 1, 58 for Booster 2, and 5.8 for Booster 3.*

Comment 158 Table 2-7. Total for Building 778 would be 0.706 not .27 unless there is a typo for October which should be 0.027 rather than 0.17 and for December which should be 0.021 rather than 0.21. One of the two need to be corrected. Also for Building 771A Total should be 0.965 not 0.48 unless there is a typo for November which should be 0.054 rather than 0.54. Correction should be made for either cases.

Response 158. *The release totals presented in the draft report are correct. Typographical errors were found in Table 2-7 for Building 778 in October and December and Building 771A for November. The data in the table are now consistent with the totals presented.*

Comment 159. Page 66. List results of 1100 μCi of uranium released, my calculation is 1094, if rounded off used it should be noted.

Response 159. *A total uranium release estimate is no longer presented in the referenced discussion. Release totals are, however, presented for enriched and depleted uranium. The values are 360 Ci and 730 Ci, respectively, which add to yield a total of 1100 Ci as was presented in the draft report.*

Comment 160. Table 2-8. Building 444 Duct 2 total shows 0.82 while my calculation total 1.521. if there a typo in the monthly average? Also the average would be 0.126 rather than 0.068 if there are no typos and the total μCi released would be 102 not 54.

Response 160. *The old Table 2-8 was split into separate tables for depleted and enriched uranium (now Tables 2-8 and 2-9). Time-weighted averaging was performed for Building 883 Ducts A and B and Building 881 Duct 5, of which sampling began in mid-year. All monthly values of average daily concentration were verified to be correct with the following exceptions: Building 444 Duct 2 for October and November, Building 881 Duct 1 for February, and Building 881 Duct 4 for June.*

Comment 161. Table 2-8 (Continued). Building 881 Duct 1 average shows 0.096 while my calculations show 0.17. If there are no typos the μCi released should be 79 not 81.

Response 161. See response for comment 160.

Comment 162. Table 2-9. May have math errors but without additional data a comment cannot be made. This comment based upon other tables with errors.

Response 162. Examination of associated data showed the given values to be accurate. However, the former Table 2-9 was deleted from the Task 5 report.

Comment 163. Table 2-10. Raw data reconstruction (μCi) for 1956 shows 1,100 while my calculation using Table 2-9 data is 210.9.

Response 163. See Tables 2-12 and 2-30. What was missing in your calculation was the contribution of the large release on January 24th, 1956. See new Tables 2-12 and 2-30.

Comment 164. Page 70. The statement that the 1956 release is approximately 20% of the 1953-1989 total release of enriched uranium is questionable using my calculations. Would like a clarification on the calculations.

Response 164. Per Table 2-45, the 1953-1989 enriched uranium release total is 5600 microcuries. 1100 ÷ 5600 = 20%.

Comment 165. Table 2-12 raw data reconstruction result (μCi) other than 1957 are close by my calculations except 1959 where my calculations show 257 vs 290 and 1960 where my calculations show 283 vs 240. Since there was no data for 1957 in Table 2-11 comparison calculations could not be made.

Response 165. The 1957 values are based on Table 2-4. For 1958, 1959, and 1960, depleted uranium emissions from Buildings 444 and 447 were independently reconstructed and added to reported emissions from Building 883. Table 2-20 has been revised to indicate the error in the FEIS data and the methods used to generate the given airborne depleted uranium emission totals.

Comment 166. Page 72. 1963 Reconstruction lists a release of 880 μCi while the total from Table 2-13 is 886.9.

Response 166. Table 2-13 has been split into two tables, and several typographical errors have been corrected. See Tables 2-21 and 2-22. The correct total is 820 microcuries.

Comment 167. Page 72. 1969 Reconstruction lists a release of 210 μCi while the total from Table 2-14 is 212.

Response 167. Your value would have been rounded to 210, as presented. See Tables 2-23 and 2-24. The correct total is 210 microcuries.

Comment 168. Table 2-13, Building 444 Duct A-1 #2 total shows 10 while my calculations show 0.0777 and μCi released shows 85 while my calculations show 62. Building 444 Duct A-1 #3 total shows 20 while my calculation shows 2.44 and μCi released shows 98 vs. 97. Building 447 μCi shows 250 vs. 253. Building 331 μCi shows 0.90 vs. 0.92. If there are no typos in the lists the numbers needed to be corrected.

Response 168. Several typographical errors were corrected in the tables of 1963 uranium monitoring data. The data now appear in Tables 2-21 and 2-22.

Comment 169. Table 2-13 (Continued). Building 881 Duct 1 μCi released lists 47 while my calculations show 44.2. Duct 2 and Duct 3 both lists μCi of 73 while my calculations show 69. Duct 5 μCi Released lists 14 while my calculation show 30.7. Duct 6 average shows 0.74 while my calculation show 0.92. The μCi of Duct 6 could not be calculated as there is no air flew for that duct listed in the document unless sit is actually 5A rather than 6 then the 73 is corrected.

Response 169. The heading for Duct 6 has been changed to Duct 5A. See response to comment 168.

Comment 170. Table 2-14. Building 444 #3 shows total of 100 while my calculations show 105.

Response 170. *Your results do not really differ from ours. Our calculations yielded a value of 104, which was rounded to 100.*

Comment 171. Table 2-15. Building 883 B total lists 5.0 while my calculation shows 5.5.

Response 171. *Subsequent recalculation also gave us a result of 5.5. Table 2-25 has been revised in accordance.*

Comment 172. Tables 2-16 through 2-21. Tables were not checked due to the lack of time, however, considering math errors in other tables they should be double checked to determine any errors or typos.

Response 172. *Tables in the Task 5 report have gone through multiple stages of checking and verification.*

Comment 173. Tables 3-1 and 3-2. Tables are in different orders which can lead to confusion.

Response 173. *Table 3-1 has been reformatted to correspond with Table 3-2; there is a column for each year and a row for each building.*

Comment 174. Table 3-2. Building 441, 865, and 889 calculations could not be made as there are no air flows listed for these buildings.

Response 174. *As stated in the text, documentation of an exhaust flow rate for Building 441 could not be located, therefore the flow rate for Building 331 was used in the calculations for Building 441, a building of similar size.*

A review of beryllium monitoring data indicated that, although sampling systems were placed in Buildings 865 and 889 in November or December of 1970, no data were located for these buildings. The beryllium concentrations originally calculated for these buildings were estimates based on the applicable limit of detection. It was by an oversight that these values were included in the draft Table 3-2. Buildings 865 and 889 were removed from Table 3-1 and 3-2.

Comment 175. Table 3-2. A value is shown for Building 447 in 1962 while Table 3-1 lists no data for that year.

Response 175. A correction was made to Table 3-2. A check of the raw data indicated that no values were reported for Building 447 in 1962.

Comment 176. Table 3-2. Several calculation differences were found of which the largest was for Building 886 in 1970 with 0.005 vs. 0.06 and the yearly total for 1966 with 34 vs. 32.9.

Response 176. The 1966 release total was changed from 34 grams to 33 grams. The Building 886 value in Table 3-2 for 1970 was corrected from 0.005 to 0.06.

Comment 177. Table 3-3. Table shows a -0.01 for 1983, it is not possible to have a negative value and should show <0.01.

Response 177. Your suggested change is not appropriate. It is possible to have negative discharge totals when beryllium background is subtracted from near-background analytical results. A total of 556 beryllium analyses contributed to the 1983 site beryllium emission total. Seven months of 1983 had negative discharge totals, and the total discharge reported for the year was -0.101 grams. The 1982 annual environmental monitoring report (RFP-ENV-83) states that "this value is indistinguishable from the background associated with the analyses." A value of zero was carried through to the dose assessment process.

Comment 178. Table 4-8. Table lists background average of 0.041 while my calculation shows 0.043. Table lists Westminster average of 0.034 while my calculations shows 0.026. The calculation for table results are inaccurate or there is a typo on the number.

Response 178. Some typographical errors were corrected in Table 5-8. The 1970-1983 averages are 0.029 pCi L^{-1} for "background," 0.019 pCi L^{-1} for Westminster, and 0.018 pCi L^{-1} for Broomfield. These values were inadvertently rounded to a single significant figure in Table 5-8.

Comment 179. Table 4-10. Table averages should be reviewed, background listed as 360 my calculation shows 364, Westminster listed as 420 my calculations shows 418 and Broomfield listed as 1200 my calculations shows 1155.

Response 179. Your calculations are not inconsistent with ours. As mentioned earlier, our values have been rounded to two significant figures.

Comment 180. General comment for the entire document is that the math needs to be closely reviewed as do rounding to two significant figures.

Response 180. The Task 5 report has been carefully reviewed.

Comment 181. It would be more useful if the Index and Glossary were moved to the front or the back (behind Appendices) of the document.

Response 181. The Task 5 report glossary and index have been moved to the end of the document, behind the appendices.

Comment 182. It would be helpful to have a Table of units and conversions at the front or back of the document.

Response 182. A table of units and conversions has been added near the end of the document, immediately following the glossary.

Comment 183. A map showing the different buildings on the plant site, as well as the location of various burn pits and sources of releases would be helpful. It would also be nice to have a description of the building uses included with the map, preferably in the front or back of the document for easy reference.

Response 183. The information described is contained in the report of Tasks 3 and 4. The reports are meant to complement each other. We do not feel it is advisable or practical to repeat the information in the Task 5 report.

Comment 184. Page 161. Although Pond C2 is off-channel and does not receive plant site runoff, after a holding period in this pond, the water was then released back into Woman Creek (until the present line to the Broomfield Diversion Ditch

was constructed). It may have been periodically diverted via Mower ditch to Mower reservoir, but it is not accurate to say, "Woman Creek remained isolated from Rocky Flats surface runoff, and Pond C2 remained isolated from Woman Creek".

Response 184. *The indication in Section 5.1.1.3 that Pond C-2 and Woman Creek remained isolated from Rocky Flats runoff has been removed. Section 5.1.1.3 now states that Pond C-2 water was released into Woman Creek until the Broomfield Diversion Ditch was constructed, and that Pond C-2 water may have also been diverted by Mower ditch to Mower Reservoir.*

Comment 185. Figure 4-3 (last item on time line). I believe Pond C1 is located in Woman Creek itself, therefore the statement "Pond C1 isolated from Woman Creek" is incorrect.

Response 185. *The time line entry on Figure 5-3 has been revised to state that "Pond C-1 was isolated from Rocky Flats surface runoff."*

Comment 186. Page 163-164. Have the uses of Mower Reservoir been confirmed by contact with the current reservoir owner?

Response 186. *Additional investigations of uses of Mower Reservoir were not conducted during revision of the Task 5 report.*

Comment 187. Figure 4-4. Is there a discreet conveyance from Mower Reservoir to Standley Lake?

Response 187. *Mower Reservoir flows into Church Ditch, which supplies water to Upper Church Lake and Great Western Reservoir (References: Rocky Flats FEIS, RFP-ENV-88 Fig. 6, and Drawing 304923-B9 from the OU-3 Final Past Remedy Report). Figure 5-4 was misleading in this respect and has been revised.*

Comment 188. Page 171. "Until the early 1980's, both the City of Arvada and Rocky Flats received a majority of their water from Ralston Reservoir," What has happened since then? Where do they receive their water now? This paragraph need clarification and the information should be confirmed with the Colorado Department of Health's Drinking Water Section (Jerry Biberstine).

Response 188. *We are unable to expand the discussion without additional investigation.*

Comment 189. Figure 4-14 through 4-17. It is interesting that the annual average concentration of Pu 239/240 and natural uranium for Golden and Westminster, both of whose primary sources are Clear Creek, appear to be consistently relatively low.

Response 189. *No response necessary.*

Comment 190. Section 4.0. What is the significance of "natural uranium" in evaluating releases from the plan? (Does "natural uranium" provide releases from the plant? (Does "natural uranium" provide useful information about what releases may be related to Rocky Flats plant?) How doe it relate to the "enriched" and "depleted" uranium, in terms of trying to quantify off-site releases?

Response 190. *"Natural uranium" is approximately 99.276 percent U-238, 0.7196 percent U-235, and 0.0057 percent U-234. It is not the most useful form of monitoring data for evaluation of impacts from Rocky Flats, however, measurements in this form are presented for possible utility in evaluating overall environmental concentrations of uranium.*

If large releases of depleted or enriched uranium were to have occurred, "finger printing" of sources by analysis of isotopic ratios could be possible. Unfortunately, because uranium is ubiquitous in the environment and concentrations found in nature fluctuate widely, analyses of this type are rarely very powerful. Attribution of uranium in surface waters near Rocky Flats to specific sources was not possible based on the data collected during the Phase I investigation.

Comment 191. Page 198. What does the contractor propose to do in future studies when they "cannot determine whether the plant was the source of fluctuations" in radioactive? How will this be handled?

Response 191. *Questions regarding plans for future health studies should be directed to the Department of Health and the Phase II contractor.*

Comment 192. Page 201 and 224. Page 201 second paragraph statement that the chromic acid incident "likely resulted in some degree of off-site contamination" appears to be contradictory to the last sentence on Page 224. Perhaps Page 224 should include more details of the conclusions on Page 201. The second paragraph of the DOE citation states that "daily water samples taken from the impoundment ponds after the ice thawed on the ponds ... indicated chromium levels below the Clean Water Act drinking water standard of 0.05 ppm." Were there any release from those impoundments at that time and what were the results of the specific daily samples? More detail here might be helpful.

Response 192. The report was revised to indicate that the chromic acid spill resulted in some degree of on-site contamination. No new information concerning the chromic acid incident that warrants addition to the report has been identified.

GLOSSARY

activity fraction	The fraction of a mixture of materials that a radionuclide constitutes in terms of its contribution to the total radioactivity of the mixture. This is in contrast to the mass fraction, which reflects a portion of the total mass of mixture.
aerodynamic equivalent diameter	The diameter of a unit density (1 g cm^{-3}) sphere with the same settling velocity as the particle in question
aerosol	A suspension of fine solid or liquid particles in gas.
alpha emitter	A radioactive material that releases energy in the form of alpha particles.
alpha pulse-height analysis	A method for identifying and quantifying alpha-emitting radionuclides by recording the frequency of emission of alpha particles of specific energies.
alpha track method	A method for measuring alpha radiation by recording the passage of alpha particles on photographic film.
back-calculation	A calculation technique used to work back from a known or given result of a calculation to determine an unknown input value to the calculation.
"background" radioactivity	Radioactivity normally present in the natural environment, or not attributable to a particular known source of interest.
beta emitter	A radioactive material that releases energy in the form of beta particles.
blowdown	Cooling tower water routinely released from the system to maintain water quality (total dissolved solids) at an acceptable level.
cfm	Cubic feet per minute — a volumetric rate measurement.
Church litigation	Lawsuits brought against the Rocky Flats Plant by neighboring landowners.

computer modeling	A series of mathematical calculations used to predict the effect of such things as physical, chemical or environmental processes that are performed with the aid of a computer.
Count median diameter	For a distribution of particles of various sizes, the count median diameter identifies the size for which half the total *number* of particles collected is contributed by smaller particles and half by larger particles.
critical mass experiments, or criticality tests	Tests performed with a sufficient quantity of fissile materials to produce a nuclear fission chain reaction.
criticality safety program	A plant program involving the use of critical mass experiments for the purposes of developing routine practices for avoiding spontaneous nuclear fission chain reactions.
curie (Ci)	The conventional unit of activity equal to 3.7×10^{10} nuclear disintegrations per second.
cutting oil	A liquid applied to a cutting tool to assist in the machining operation by washing away the chips and serving as a lubricant or coolant.
decay product	Nuclide formed from the disintegration of an unstable atom.
degrease	To remove grease from.
depleted uranium	Uranium in which the fraction of the U-235 isotope has been decreased below the 0.7 percent found in nature.
disintegrations per minute (dpm)	The rate of nuclear transformations exhibited by a radionuclide. Nuclear transformations are the events that lead to emission of radiations as the atom changes to a more stable form.
dose coefficient	A constant used to convert from a quantity of radionuclide to which a person is exposed to some measure of the resulting dose.
EG&G, Inc.	Current operator of the Rocky Flats Plant for the Department of Energy.
effluent	Any gas or liquid emerging from a pipe or similar outlet; waste products from industrial plants as stack gases or liquid mixtures.

emission point	Location of release.
enriched uranium	Uranium in which the fraction of the fissionable U-235 isotope has been increased above the 0.7 percent found in nature.
filter plenum	The portion of a building air exhaust or ventilation system that contains media to collect or separate out matter carried by the air.
fission	The splitting of a heavy atomic nucleus into approximately equal parts, accompanied by release of a large amount of energy.
fission product	Nuclides formed as a result of fission.
fission track method	A method for characterizing fissionable materials by placing samples in a neutron source and detecting tracks produced by resulting fission fragments in materials held in contact with the sample during irradiation.
gamma emitter	A radionuclide that emits photons which are identical in form to X rays.
glove box	A sealed, protectively lined compartment having holes to which are attached gloves for use in handling dangerous materials inside the compartment.
gram	A metric unit of mass and weight equal to one-thousandth of a kilogram and nearly equal to one cubic centimeter of water at its maximum density.
gross alpha activity	Radioactivity measured in terms of alpha particles emitted, with no determination of their energy or the identity of the specific radionuclides from which they were emitted.
groundwater	Water within the earth that supplies wells and springs.
half-life	The time required for an unstable element or nuclide to lose one-half of its radioactive intensity in the form of alpha, beta, and gamma radiation.
halogen	Any of the five elements fluorine, chlorine, bromine, iodine, and astatine that form part of the group VII A of the periodic table of elements.

HEPA filter	High efficiency particulate air filter.
isokinetic sampling	Refers to the removal of a sample from an air stream where the velocity of the air entering the sampling device is the same as the velocity of the air in the duct at the sampling point.
isotopes	Atoms having the same atomic number but different atomic weights; they have similar chemical properties but different physical properties.
kilogram	The basic metric unit of mass nearly equal to 1,000 cubic centimeters of water at the temperature of its maximum density.
liquid scintillation counter	An instrument which measures radioactivity by placement of a sample in a liquid "cocktail" that emits light that can be related to the quantity of radioactivity present.
lognormal	Refers to a statistical distribution of a data set relating to or being a normal distribution that is the distribution of the logarithm of a random variable.
long-lived alpha activity	Alpha-emitting radioactivity from which short-lived radionuclides have been allowed to decay away or have been subtracted using an algorithm designed to quantify only those nuclides with long half-lives.
mass median diameter	For a distribution of particles of various sizes, the mass median diameter identifies the size for which half the total *mass* of material collected is contributed by smaller particles and half by larger particles.
microcurie (μCi)	One-millionth of a curie.
noble gases	Any of a group of rare gases that include helium, neon, argon, krypton, xenon, and sometimes radon that exhibit great stability and extremely low reaction rates.
nuclide	An individual species of an atom characterized by its mass number (number of protons plus neutrons in its nucleus), atomic number (number of protons in its nucleus), and the energy state of its nucleus.
organic solvents	Non-polar carbon-containing substances (hydrocarbons) capable of dissolving another substance.

particle size distribution	Frequency of occurrence of particles by size as measured by diameter.
particulate	Of or relating to minute separate particles.
radioactive daughter	An atomic species that is the product of the radioactive decay of a radionuclide and is itself radioactive.
radionuclide	A radioactive form of an element distinguished by its atomic number, atomic weight, and energy state.
runoff	To drain off.
sampling train	All the components or devices used to collect a sample.
source term	Information relating the quantity and characteristics of a contaminant release.
specific activity	The total activity of a given radionuclide per gram of a compound, element, or radioactive nuclide.
spectrophotometry	An important technique of instrumental analysis involving measurement of the absorption of radiant energy by a substance as a function of the energy incident upon it.
spectroscopy	A branch of analytical chemistry devoted to identification of elements and elucidation of atomic and molecular structure by measurement of the radiant energy absorbed or emitted by a substance.
spontaneous fissioning	The property of certain materials which undergo splitting without the external application of neutrons.
surface water	Water present on the earth's surface, e.g. rivers, streams and lakes.
totalizer	An instrument that indicates or records a cumulative measure of a parameter of interest, for example the total volume of air which is exhausted through a stack in a given period.
tritiated water	Water in which one or more hydrogen atoms have been replaced with tritium, the radioactive form of hydrogen.

weapons grade plutonium	Plutonium that is approximately 94 percent Pu-239 by mass, with about 5.8 percent Pu-240 and small amounts of Pu-238, Pu-241, Am-241, and Pu-242.

METRIC FRACTIONS

Multiple	Decimal Equivalent	Prefix	Symbol
10^6	1,000,000	mega-	M
10^3	1,000	kilo-	k
10^2	100	hecto-	h
10	10	deka-	da
10^{-1}	0.1	deci-	d
10^{-2}	0.01	centi-	c
10^{-3}	0.001	milli-	m
10^{-6}	0.000001	micro-	μ
10^{-9}	0.000000001	nano-	n
10^{-12}	0.000000000001	pico-	p
10^{-15}	0.000000000000001	femto-	f
10^{-18}	0.000000000000000001	atto-	a

METRIC CONVERSION TABLE

Multiply	By	Equals	Multiply	By	Equals
in.	2.54	cm	cm	0.394	in.
ft	0.305	m	m	3.28	ft
ac	0.404	ha	ha	2.47	ac
mi	1.61	km	km	0.621	mi
lb	0.4536	kg	kg	2.205	lb
liq. qt.-U.S.	0.946	l	l	1.057	liq. qt.-U.S.
ft^2	0.093	m^2	m^2	10.764	ft^2
mi^2	2.59	km^2	km^2	0.386	mi^2
ft^3	0.028	m^3	m^3	35.31	ft^3
d/m	0.450	pCi	pCi	2.22	d/m
pCi/l(water)	10^{-9}	μCi/ml(water)	μCi/ml(water)	10^9	pCi/l(water)
pCi/m^3(air)	10^{-12}	μCi/cc(air)	μCi/cc(air)	10^{12}	pCi/m^3(air

TRADITIONAL AND INTERNATIONAL SYSTEMS OF RADIOLOGICAL UNITS

(Traditional units are in parentheses.)

Quantity	Name	Symbol	Expression in Terms of Other Units
absorbed dose	Gray	Gy	J/Kg^{-1}
	(rad)	rad	10^{-2} Gy
activity	Becquerel	Bq	1 dps
	(curie)	Ci	3.7×12^{10} Bq
dose equivalent	Sievert	Sv	J/Kg^{-1}
	(rem)	rem	10^{-2} Sv
exposure	Coulomb per kilogram		C/Kg^{-1}
	(roentgen)	R	2.58×10^{-4} C/Kg^{-1}

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