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RADIOLOGICAL ASSESSMENTS CORPORATION, THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT, TASKS 2&3, RADIONUCLIDE SOURCE TERMS AND UNCERTAINTIES - (USED AS A REFERENCE IN OU 5 RI REPORT)

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THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT

Tasks 2 and 3

Radionuclide Source Terms and Uncertainties

Submitted to the Centers for Disease Control and Prevention
in partial Fulfillment of Contract No. 200-90-0803

by

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RAC Report No. CDC-5

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October 28, 1994

From: ***Radiological Assessments Corporation***
To: **Individuals Interested in the Fernald Dosimetry Reconstruction Project**
Subject: **Status of the Project**

This note is to provide an update on the Fernald Dosimetry Reconstruction Project. As many of you know, the Centers for Disease Control and Prevention, the study's sponsor, requested that the National Academy of Sciences review key reports in the that project document our work. As a result, the Academy reviewed the Task 2/3 report on source terms and the Task 4 report describing our methodology. Although we received comments on the draft Task 2/3 report, and they have been considered for our final version, we still have not received the Academy's comments on the draft Task 4 report. Since Task 4 explains the methods to be used in estimating doses, we have not been able to proceed with dose calculations, and therefore the draft Task 6 report containing those results.

Although it is taking longer to complete our research and to provide the information for your comment than we had anticipated, we believe the Academy's review of our work is important. It is unfortunate that these reviews take considerable time to complete. They are, however, a crucial part of the scientific process.

We are currently finalizing our Task 2/3 report on source terms. Hopefully, we will soon be able to finalize our Task 4 report, taking into account the Academy's comments. Then we can proceed to estimate doses and document our results in Task 6.

We have no control over these events and, like you, we are disappointed that it is taking so long. However, we feel it is essential to take into account the Academy's peer review of our work. We appreciate the comments from many of you on the draft reports and we have given them careful consideration.

Thank you for your patience. Please do not hesitate to contact me if you have questions.

John E. Till
President

F-7340

THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT

Tasks 2 and 3

Radionuclide Source Terms and Uncertainties

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RAC Report CDC-6

January 1995

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CONTENTS

GLOSSARY OF TERMS AND ACRONYMS v

EXECUTIVE SUMMARY xi

INTRODUCTION AND OVERVIEW 1

PLANT PROCESSES AND WASTES 3

FMPC PRODUCTION INFORMATION 6

OTHER RADIONUCLIDE RELEASES 7

FMPC RELEASES TO THE ENVIRONMENT: FACTORS TO CONSIDER 8
- Period of Time Studied 9
- Characteristics of Radionuclide Releases 9
- Uncertainties in Estimating Releases 10
- Sources of Information 12

ATMOSPHERIC RELEASES FROM DUST COLLECTORS 14
- Dust Collector Operation 14
- Current Estimates of Release From FMPC Dust Collectors 15

DISCHARGES FROM PLANT 2/3 DENITRATION OPERATIONS 18

RELEASES FROM PLANT 8 SCRUBBERS 21

OTHER SOURCES AND EPISODIC RELEASES TO THE ATMOSPHERE 23
- Miscellaneous Unmonitored Emissions 25
- Accidental Releases 25

RADON AND DECAY PRODUCT RELEASES FROM K-65 SILOS AND MATERIALS 26
- History of K-65 Silos and K-65 Material at the FMPC 28
- Current Estimates of Radon Releases 28

DIRECT EXPOSURES FROM GAMMA RADIATION FROM THE SILOS 31

LIQUID WASTE DISCHARGES FROM FMPC 31
- Releases of Uranium in Liquid Effluents from the FMPC 32
- Other Radionuclides Released in Liquid Effluents 33

URANIUM CONTAMINATION IN GROUNDWATER OUTSIDE THE FMPC 35
- Potential Sources of Groundwater Contamination 36
- Estimated Uranium Concentrations in Private Wells 36

TASK 2 AND 3 SUMMARY AND CONCLUSIONS 39

REFERENCES 43

APPENDICES

A. Sources of Information
B. Plant Processes and Wastes
C. FMPC Production Information
D. Other Radionuclide Releases
E. Effluents from Dust Collector Exhausts
F. Fitting Particle Size Distributions for FMPC Dust Collectors
G. Estimates of Bias in Effluent Sampling for Particles
H. Discharges from Plant 2/3 Denitration Operations
I. Releases from Plant 8 Scrubber Systems
J. Emissions of Radon, Radon Daughters and Gamma Radiation from K-65 Silos
K. Other Sources and Episodic Releases to the Atmosphere
L. Surface Water Discharges
M. Groundwater Contamination Outside the FMPC

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GLOSSARY OF TERMS AND ACRONYMS

Each term or acronym is in italics the first time it appears in the text.

Activation products are radionuclides that result from the absorption of neutrons by uranium and other materials present in a nuclear reactor. An example is plutonium-239 produced following neutron absorption by uranium-238.

AMAD – Activity median aerodynamic diameter, a measure of particle size.

AMS – Air monitoring stations

Anisokinetic sampling –refers to a mismatch between the air or fluid velocity in the sampling probe and that in the stack releasing airborne effluents. It is a source of bias in effluent sampling. In contrast, *isokinetic sampling* results in an unbiased sample of the stack effluent.

Assessment Domain is the region surrounding a facility for which radiation doses to people are calculated; for this project, a circular region with a radius of 10 kilometers (km) (6.25 mi.) with its center in the FMPC production area..

Background Radioactivity – refers to radioactive elements in the natural environment including those in the crust of the earth (like radioactive potassium, uraniumand thorium isotopes) and those produced by cosmic rays.

Bias is a systematic distortion of measurements that makes the results inaccurate.

CDC – Centers for Disease Control and Prevention, who funded the Fernald study.

Chemical Symbols are abbreviations for different elements and compounds. Examples of elements include U for uranium, O for oxygen, N for nitrogen and F for fluorine. Examples of compounds include UF_4 for uranium tetrafluoride (green salt) and UO_3, or uranium trioxide (orange oxide).

Contamination refers to unwanted radioactive material, or to the deposition of radioactive material in the environment or in any place where it may make surfaces or equipment unsuitable for some specific use.

Decay (daughter) products refer to the isotopes or radionuclides that result from radioactive decay of isotopes, such as the uranium and thorium isotopes. In most of the feeds received by the FMPC, the uranium had previously been separated chemically from the other decay products. As a result, the facility's effluents consisted primarily of uranium, and decay product radionuclides were generally present in small quantities. In naturally-occurring uranium ores, the decay products include isotopes of uranium, protactinium, thorium, radium, radon and radon daughter products. *Radon daughter products* that are

derived from uranium are the short-lived decay products from radon-222, and include polonium-218, lead-214, bismuth-214 and polonium-214.

Denitration – a process in Plant 2/3 in which nitrates were driven off by heating uranyl nitrate hexahydrate (UNH) to produce uranium trioxide (UO_3, or orange oxide).

Derbies are masses of uranium metal fabricated in Plant 5. The derbies were then remelted and cast into ingots of metallic uranium.

Direct exposure – refers to one pathway of exposure of people to radiation from the FMPC. In this exposure pathway, penetrating radiation emitted from radioactive material is partially absorbed by individuals exposed to it. The amount of exposure decreases with distance from the source. An example is gamma radiation from the K-65 silos that resulted in low-level exposure of nearby residents.

DOE – U.S. Department of Energy

Dose is a general term denoting the quantity of radiation or energy that is absorbed by the body. There are technical terms with specific definitions, such as absorbed dose, dose equivalent, effective dose, etc.

Dust Collector is one type of filtration system for airborne effluents used at the FMPC to remove airborne particulate material before it was discharged through the stack to the outside. The filtering medium is similar to that used for large fiber vacuum cleaner bags.

Effluent is a gas or liquid containing contaminants that flows from a process, building or the site into the surrounding environment.

Empirical values are values which are measured (as opposed to theoretically determined or calculated values).

Enrichment of uranium – a process by which the relative abundances of the isotopes of uranium are altered, thereby producing a form of the element that has been enriched in one particular isotope and depleted in its other isotope. For example, natural or "normal" uranium contains 0.72% ^{235}U. Enriched uranium contains more than the natural concentration of ^{235}U, while depleted uranium contains significantly less than 0.72% ^{235}U.

Entrainment is a process in which the uranium-containing liquid droplets in a scrubber are carried by the exhaust air stream and are vented to the atmosphere with the exhaust gases.

Environmental exposure – exposure to radiation through environmental pathways.

Epidemiology – the study of diseases in human populations.

Fission products are radionuclides that result from the splitting of heavy elements like uranium in a nuclear reactor. Examples are strontium-90 (^{90}Sr), technetium-99 (^{99}Tc), ruthenium-106 (^{106}Ru) and cesium-137 (^{137}Cs).

FDRP – Fernald Dosimetry Reconstruction Project

FEMP – Fernald Environmental Management Project, the new name of the FMPC beginning in 1991.

FMPC – Feed Materials Production Center

GM – Geometric Mean, or median,the central point of a distribution. Half of the values are larger than the median value and half are smaller.

GSD – Geometric Standard Deviation, a measure of the spread of a distribution. A large GSD indicates a wide range of measured or calculated values.

Grab samples – samples, usually of relatively small volume, taken at random or at preselected frequencies. These samples define the concentration of a contaminant at the specific time when they are collected and differ from continuous or proportional samples which are intended to reflect the time averaged value.

Great Miami River is the major water flow near the Feed Materials Production Center (FMPC) that receives most of the liquid effluents from the FMPC. The river, located about a mile east and south of the FMPC, runs in a southerly direction and enters the Ohio River approximately 18 miles (29 km) downstream of Cincinnati. Upstream of the FMPC on the Great Miami River lie the communities of Fairfield, Hamilton, Middletown, and Dayton. The flow of the river at the Hamilton gauge averages 3300 cubic feet per second (cfs) (93.4 $m^3\ s^{-1}$) with a maximum of 352,000 cfs (9970 $m^3\ s^{-1}$) measured in March 1913 and a minimum of 100 cfs (2.8 $m^3\ s^{-1}$) measured in September 1941.

Green salt is the common name for uranium tetrafluoride (UF_4), the product from the Plant 4 operations that was sent to Plant 5 for conversion to derbies.

Gulping operations referes to a process in Plant 2/3 in which orange oxide (uranium trioxide, or UO_3) from the denitration pots was transferrred by a vacuum hose to a storage hopper. It appeared that the hose was "gulping" the orange oxide.

IH&R – Industrial Hygiene and Radiation Department at the FMPC

ICRP – International Commission on Radiological Protection

IT – International Technology Corporation

K-65 Silos – The *K-65 Storage Silos* are large concrete tank-like structures that store residues from the extraction of uranium from ores that were processed during the early years of FMPC operations.

kilo – a prefix that multiplies a basic unit by 1000. For example, 1 kilogram = 1000 grams.

Lognormal distribution – If the logarithms of a set of values are distributed according to a normal ("bell-shpaed") distribution the values are said to have a lognormal distribution, or be distributed "lognormally".

MTU – abbreviation for metric ton of uranium; one MTU equals 1,000 kg or 2,200 pounds

NCRP – National Council on Radiation Protection and Measurements

NKES – Northern Kentucky Environmental Services

NLO – National Lead Company of Ohio, the contractor for the FMPC through the end of 1985.

NO_x – nitrogen oxides, such as NO_2 and NO_3.

ODH – Ohio Department of Health

Orange oxide – abbreviation for uranium trioxide (UO_3), the product from the Plant 2/3 refinery that was sent to Plant 4 for further processing.

OSTI – the Office of Scientific and Technical Information, located in Oak Ridge, Tennessee, is the national center for worldwide literature on scientific and technical energy-related matters. It was one of the sources of information that *RAC* used for completion of the project.

Paddy's Run – a small intermittent stream lying along the west boundary of the site that joins the Great Miami River approximately 3 kilometers south of the FMPC. The flow in Paddy's Run, which generally exists only during January to May, averaged 2 to 4 cfs (0.065 to 0.1 $m^3 s^{-1}$). Since flow in Paddy's Run is dependent upon rainfall, discharges from the site to Paddy's Run generally occurred during periods of heavy rain and runoff when the storm sewer outfall overflowed, or when runoff from the west side the of site flowed into the Paddy's Run.

pico – a prefix that multiplies a basic unit by 1/1,000,000,000,000 or 1×10^{-12}. For example, one picocurie (pCi) equals 1×10^{-12} curie (Ci).

RAC – *Radiological Assessments Corporation* was the group chosen by CDC to do the Fernald Dosimetry Reconstruction Project.

Recycled uranium is uranium that had been irradiated in nuclear reactors, where finished uranium products were used. As a result, when the uranium was recovered and returned to the FMPC, small amounts of fission and activation products were introduced into the process stream.

Reentrainment is a process whereby the exhasut airflow creates new droplets from liquid that had been previously collected by a screen type filter.

Scrubber – a type of treatment system for airborne effluents that uses liquid droplets to remove particulate matter and reactive gases from airborne waste streams before they were discharged through the stack to the outside. At the FMPC, scrubbers were used in Plant 2/3 (refinery) and in Plant 8 (scrap recovery).

Scrub Liquor – the *scrub liquor* is the liquid in a scrubber that cleans or scrubs the exhaust air from certain plant operations. The liquid removes reactive gases and particles in the airstream before the airstream is discharged to the atmosphere.

South Plume –refers to the groundwater that has been contaminated by uranium from the FMPC. It extends southward from a point south of the waste pits and reflects the movement of contaminated groundwater.

Source Term – refers to the quantity, and chemical and physical form of radioactive materials released to the environment from various locations onsite.

SSOD – The Storm Sewer Outfall Ditch is a drainage ditch that runs south from the FMPC production area near the storm sewer lift station to Paddy's Run.

TLD – A thermoluminescent dosimeter is a device used at the FMPC to measure the amount of external radiation in the environment. These devices measure both radiation from naturally-occurring radioactivity in the soil and from the K-65 silos.

TRU – transuranic nuclides refer to isotopes heavier than uranium that are created by neutron capture by heavy elements.

Uncertainty –term used to describe probable bounds on, or how much evidence we have to support, our key findings. Uncertainty can result from two process: the first is due to random variations in sampling, measurement, and operational procedures. The second type of uncertainty occurs because of a lack of information about particular processes. This may occur because the right measurements were not done during part or most of the period of facility operation.

UF_4 – uranium tetrafluoride, or green salt was the product from Plant 4 that was sent on to Plant 5 for conversion to derbies.

UNH – uranyl nitrate hexahydrate was an intermediate step in the denitration process in Plant 2/3; nitrates were removed from UNH to produce uranium trioxide (UO_3, or orange oxide).

UO_3 – uranium trioxide, often called orange oxide, was produced in the Plant 2/3 refinery and was sent to Plant 4 for further processing.

$UO_2(NO_3)_2$ – uranyl nitrate was a product of the digestion phase in the Plant 2/3 refinery.

USGS – United States Geological Survey

Validation is the comparison of available measurements of the radionuclides in the local environment during the period of study with corresponding predictions from mathematical models.

WMCO – Westinghouse Materials Company of Ohio, the FMPC site contractor from 1986 through 1992.

EXECUTIVE SUMMARY

The purpose of the Fernald Dose Reconstruction Project (*FDRP*) is to estimate radiation doses to people who lived near the Fernald (Ohio) Feed Materials Production Center (*FMPC*) during its years of operation from 1951 to 1988. Exposures resulted from both planned and unplanned releases of radionuclides to the environment. The study was conducted for the Centers for Disease Control and Prevention.

The project was divided into seven tasks. The goal of Task 2 was to determine the radionuclide source term for the facility; that is, to determine both the amounts of radioactive material released to the environment and the variability of release rates. The Task 3 objective was to determine the uncertainties associated with those past releases.

This final report describes our estimates for source terms for the period 1951–1988. In finalizing this report, *RAC* has considered comments and suggestions received from a number of sources on our draft report (Voillequé et al. 1993). Initially we examined a three-year period in the early sixties to develop the methods that would be applicable to all years (Voillequé et al. 1991).

Our calculations are based on a thorough search of records documenting operations and effluent and environmental monitoring at the FMPC. In some cases, effluent measurement data from which estimates could be derived directly were not available. These situations were handled using statistical methods that simulate a possible range of values that could have existed. Source terms were divided into three categories of release: emissions to air, emissions to surface water, and *contamination* of groundwater.

The principal activity at the FMPC was processing uranium (U), with some thorium processing occurring at various times. In the early years, uranium ore was processed, and the waste materials were stored in drums and silos onsite. These waste materials are a source of radon and its decay products. Consequently, this report focuses primarily on emissions of uranium, and radon and its decay products. Some uranium was recycled, which is uranium that had been returned to the FMPC from other weapons material processing facilities. As a result, other radionuclides were also released at the site. Thus, release estimates are given for thorium, and selected activation products (plutonium-238, plutonium-239,240, neptunium-237), fission products (strontium-90, technetium-99, ruthenium-106, cesium-137), and decay products of uranium (radium-226) and thorium (radium-228). Table ES–1 summarizes the most important (uranium and radon) source term estimates and their uncertainties for 1951–1988.

Airborne waste streams were typically treated prior to release to the environment using either dust collectors (filters) or scrubbers (treatment systems employing liquids to remove particulate matter from gaseous waste streams). The efficiency of both of these methods varied greatly with the state of the technology at the time, maintenance of the system, and plant throughput. For dust collectors, our estimates accounted for anisokinetic sampling and sample line losses. Anisokinetic sampling occurs when the sampling probe in the dust collector stack does not record the stack exhaust gas velocity accurately. Losses of particles in the sampling line before they are detected at the sampler can significantly affect estimates of releases from stacks at the plant. These factors were not considered in previous studies.

Table ES-1. Summary of Median Uranium and Radon Releases Estimates From the FMPC for 1951-1988 With Uncertainty Bounds[a]

Release Point	Median Release Estimate	5th–95th Percentile Range
Uranium to atmosphere		
Dust collectors	140,000	120,000–170,000
Plant 2/3 scrubbers	66,000	56,000–78,000
Plant 8 scrubbers	81,000	56,000–130,000
Miscellaneous Sources[b]	16,000	9300–28,000
Total: airborne sources	310,000	270,000–360,000
Uranium to surface water		
Manhole 175	82,000	71,000–94,000
Paddy's Run	17,000	14,000–20,000
Total: surface water	99,000	85,000–120,000
Radon to Atmosphere		
K-65 Silos		
Radon	170,000 Ci	110,000–230,000 Ci
Radon decay products[c]	130,000 Ci	87,000–190,000 Ci

[a] Values are in kg of uranium, except for releases from the K-65 Silos which are reported in units of activity, called curies, Ci.
[b] Unmonitored and accidental releases.
[c] The release quantities for radon and its decay products are given in units of activity, curies (Ci); quantities of each of the short-lived decay products, polonium-218, lead-218, bismuth 214, and polonium-214.

Estimates of releases from the denitration processes scrubbers in Plant 2/3 (refinery) and from the scrubbers in Plant 8 (scrap recovery) were made considering uncertainty and variability in parameters that affect scrubber performance. Relevant site-specific data were used as much as possible. Monte Carlo techniques allowed us to sample the parameter uncertainty distributions to make the release estimates. The distributions represent uncertainties associated with these individual parameters and can be combined to form a distribution that characterizes the overall range of potential scrubber releases, in contrast to the point estimates of previous studies. Our estimates of releases from Plant 8 scrubbers relied heavily on data reporting monthly amounts of uranium found in the scrubber liquid residue (called scrub liquor) and measurements of scrubber penetration of uranium. The Plant 8 scrubbers dominated the uranium releases in the 1960s, with approximately 47,000 kg U released in that decade, compared to 21,000 and 19,000 kg U for the dust collectors and Plant 2/3 scrubbers, respectively. In the 1970s, the Plant 2/3 scrubbers were relatively more important. In the 1950s and 1980s, the dust collectors contributed most to the total uranium releases, although the magnitude of all releases in the 1980s was significantly less than in the 1950s.

A thorough evaluation of atmospheric releases of uranium from unmonitored sources (incinerators, building ventilation, lab hoods, unmonitored process emissions and waste pits) and accidental releases (fires, spills and episodic releases) indicates that these were relatively minor compared to the three primary sources of atmospheric emissions (dust collectors, Plant 8 and Plant 2/3 scrubbers). However, the detailed assessments of these sources provide thorough documentation of their magnitude with uncertainties. These release estimates are included as part of the total atmospheric source term.

Radon releases were calculated for the K-65 silos, located near the west side of the site, and for drummed K-65 material temporarily stored on the Plant 1 Pad in the early 1950s. The silos contained K-65 material, a waste from the extraction processing of uranium ore. This material contains high concentrations of radium-226, and thus, acts as a continuous source of radon-222, a highly mobile radioactive inert gas. Release estimates were complicated by a lack of data describing characteristics of the material in the silos, and by structural changes that occurred over the years. Our estimates of radon and radon decay product releases were derived from measurements found in the historical records and from previous studies. The rate of radon release from the K-65 Silos for 1959–1979 is greater than for other periods, and significantly greater than for later periods. Radon releases from the Plant 1 Pad drums were insignificant contributors to the total radon releases for the period 1951–1988, but were important contributors for 1951 and 1952.

Radioactive material left the site in liquid effluents at two key points: through Manhole 175 (MH 175), a final junction point for major effluent streams onsite to the Great Miami River, and, periodically, through the storm sewer outfall to Paddy's Run. Effluent concentrations and volumes were measured regularly at both locations, and records were used to reconstruct these source terms. More uncertainty is encountered with the release estimates to Paddy's Run because the frequency of sampling was less than at MH 175, and there were discharges to the stream that were not monitored. Nevertheless, estimates of releases in liquid discharges are relatively well known, and uncertainties are generally smaller than with releases to air.

An evaluation of the groundwater plumes underlying the FMPC indicated that, at the present time, three offsite wells are contaminated, and only a small number of people would have potentially received radiation doses from contaminated groundwater. Consequently, a simple model is used to estimate concentrations of uranium in the contaminated plume, based on recent measurements in the three offsite wells and on quantities of uranium released to the storm sewer outfall ditch and to Paddy's Run since the 1950s. Based on this simple model, it is likely that uranium contamination in the groundwater would not have reached the offsite wells prior to 1968.

There have been several previous assessments of uranium releases from the FMPC. Previous estimates of uranium discharged in liquid effluent fall within the uncertainty range of our estimates. Source terms from previous studies of airborne uranium releases have all fallen outside our uncertainty range except for one study. Exhaustive comparisons have not been made; however, reasons for our higher estimates include:

- the time to conduct a comprehensive review of historical documents, in particular original records, related to the FMPC operations;
- the use of a distribution of scrubber efficiencies for Plant 8 scrubbers;

- accounting for uranium losses from miscellaneous unmonitored sources and accidents;
- accounting for biases from sample line losses and o her sampling deviations in the calculation of dust collector losses.

Our results report not only best estimates of releases (as a median value) but also associated uncertainties that were calculated as an integral part of the estimates. This approach represents a significant improvement in the state-of-the-art of source term analysis. This depth of analysis was not undertaken in earlier estimates of releases. These source term estimates will be used in Task 6 to calculate radiation doses to people who live near the FMPC.

TASKS 2 AND 3

RADIONUCLIDE SOURCE TERMS AND ASSOCIATED UNCERTAINTIES FOR 1951–1988

INTRODUCTION AND OVERVIEW

The purpose of the Fernald Dose Reconstruction Project (*FDRP*) is to estimate radiation doses to people who lived near the Fernald (Ohio) Feed Materials Production Center (*FMPC*) during its years of operation from 1951 to 1988 (Figure 1). Exposures resulted from both planned and unplanned releases of radionuclides to the environment. The study was conducted for the Centers for Disease Control and Prevention.

The project was divided into seven tasks. The goal of Task 2 is to determine the radionuclide source term for the facility; that is, to determine both the amounts of radioactive material released to the environment and the variability of release rates. This information is fundamental to the assessment of radiation doses to persons in the vicinity of the site. The Task 3 objective was to determine the uncertainties associated with those past releases.

An interim Task 2/3 report (Voillequé et al. 1991) initially determined the source term for the years 1960, 1961 and 1962. This shorter time period was selected because environmental samples and records were available and there was a relatively consistent level of emissions. The pilot study tested and presented our methods for estimating the amounts of radioactive materials released and for assessing the uncertainties associated with those estimates. Based on the methods described in the interim Task 2/3 report, we estimated the amounts of radioactive materials released to air, surface water and in groundwater throughout the history of the Fernald plant's operation. Those results were presented in a draft report (Voillequé 1993). The draft report was reviewed, and comments were received from a number of people and organizations, including the CDC, members of the public, current employees at the FEMP, and former employees of NLO. All comments were considered in finalizing this current report, which reflects those changes and represents the final Task 2/3 report for this project. In addition to minor editorial changes, the main revisions to this report from the draft version include:

- Annexes listing the types of documents found in Central Files at the FMPC and of the boxes of contaminated documents that were examined in the Plant 4 storage area (Appendix A)
- Revised screening calculations using updated NCRP screening factors (Appendix D)
- Re-evaluation of the attachment fraction of particles in the calculation of sampling line losses for dust collector releases (Appendix G and E).
- Recalculation of discharges from the Plant 2/3 denitration operations using additional scrub liquor concentration data; determination of effect of alternative calculation of the outage fraction on Plant 2/3 scrubber releases (Appendix H)
- Two alternative calculations of releases from the Plant 8 scrubbers to test the effect of different modeling choices on the results. (Appendix I, page I–37)

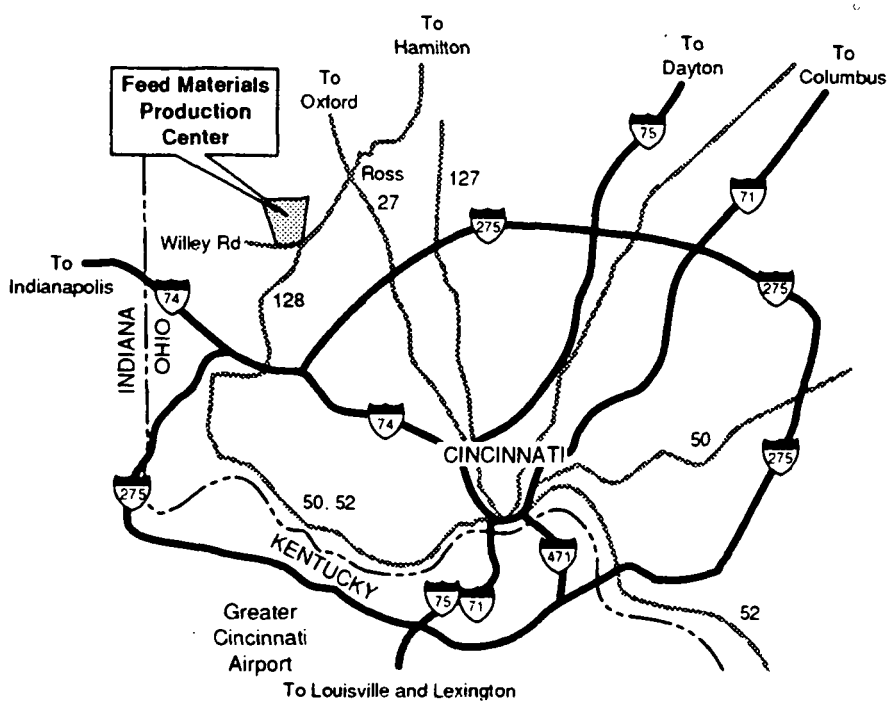

Figure 1. Location of the Fernald Feed Materials Production Center.

- An alternative calculation of radon and radon daughter product releases from the K-65 silos using a conventional methodology of radon releases from bulk quantities of ^{226}Ra-bearing materials (Appendix J, page J–73).
- Revision of fugitive emissions calculations for the waste pits using an improved model (Appendix K).
- Use of an empirical model to estimate uranium concentrations in offsite contaminated wells for years when no measurements were made; the model uses available uranium measurements in well water and considers the uranium released to Paddy's Run and the storm sewer outfall ditch (Appendix M).

This report is divided into this summary and 13 appendices. Each appendix is in bold type when it first appears in the discussion of that appendix. The appendices are:

Appendix A	Sources of Information
Appendix B	Plant Processes and Wastes
Appendix C	FMPC Production Information
Appendix D	Other Radionuclide Releases
Appendix E	Effluents from Dust Collector Exhausts
Appendix F	Fitting Particle Size Distributions for FMPC Dust Collectors
Appendix G	Estimates of Bias in Effluent Sampling for Particles
Appendix H	Discharges from Plant 2/3 Denitration Operations
Appendix I	Releases from Plant 8 Scrubber Systems
Appendix J	Releases of Radon, Radon Decay Products and Gamma Radiation from the K-65 Silos
Appendix K	Other Sources and Episodic Releases to the Atmosphere

Appendix L Surface Water Discharges
Appendix M Groundwater Contamination Outside the FMPC

The goal of this report is to provide the reader with a clear picture of the FMPC operations from 1951 through 1988. It explains the generation of effluents from those operations, and estimates effluent releases using relevant measurements and related information.

PLANT PROCESSES AND WASTES

The FMPC is a government-owned, contractor-operated facility whose primary purpose was to convert uranium (U) feed stocks to uranium metal ingots for machining or for extrusion into tubular form. Production reactor fuel cores and target elements were fabricated. Figure 2 gives an overview of the main features of the FMPC area. An aerial photograph shows the environs of the FMPC in 1965 (Figure 3).

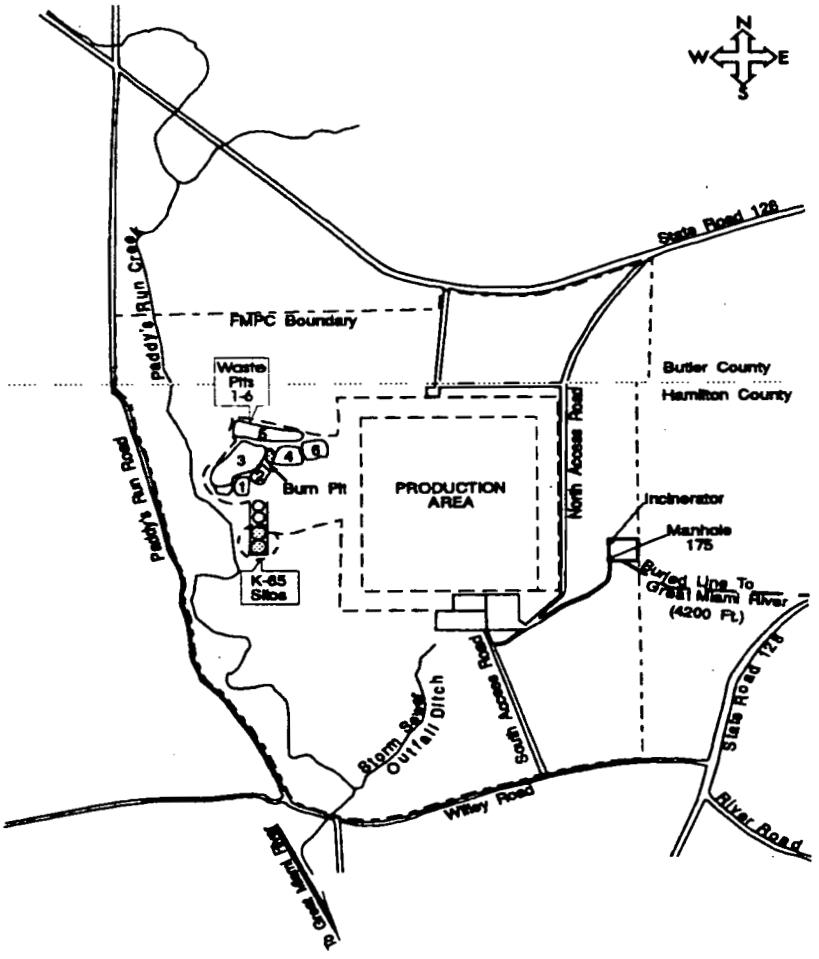

Figure 2. Overall view of the FMPC facility. The width of the production area is about 700 meters from east to west (inner fence).

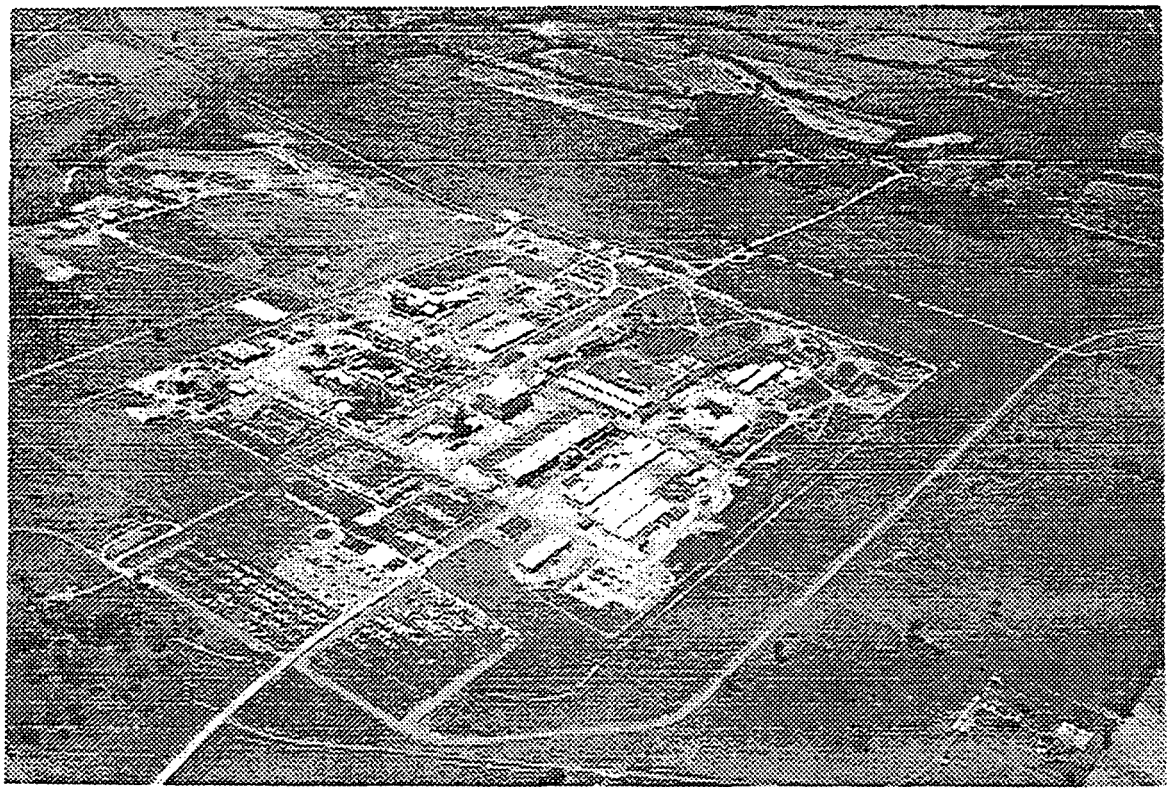

Figure 3. Aerial photograph taken from the southeast of the Feed Materials Production Center in 1965, showing the production area and general land features (digitized from DOE 1965). The area within 5 miles (8 km) from the center of the FMPC is populated with farm houses, small communities, and the small town of Ross, Ohio, with land use being primarily grazing and farming.

Appendix B describes the plant functions in some detail by following the flow of uranium through the various facilities as it was converted from raw material to finished products. Although uranium was the primary product at the FMPC, lesser amounts of thorium were produced intermittently during the mid-1950s, and from 1964 through 1980. In addition, the FMPC began processing materials recycled from other stages of the nuclear fuel cycle in 1962.

Figure 4 is a material flow diagram which shows the movement of incoming raw and recycled material (called feed materials) into the FMPC at Plant 1, the Sampling Plant, and their passage through various chemical and physical processes before leaving the site as finished products. Historic records and discussions with plant staff revealed that the same basic processing scheme was employed throughout all years of operation.

From Plant 1, the materials passed to Plant 2/3, the Refinery, where the uranium in the various feed materials was converted to uranium trioxide (UO_3, called orange oxide because of its color). The UO_3 was converted to uranium tetrafluoride (UF_4, called green salt) in Plant 4, and then sent to Plant 5, Metals Production. There the UF_4 was converted to uranium metal derbies or ingots. From Plant 5 the ingots were shipped offsite, or were sent to either Plant 6 (where the metal was fabricated into finished products) or to Plant 9 (where special products were machined).

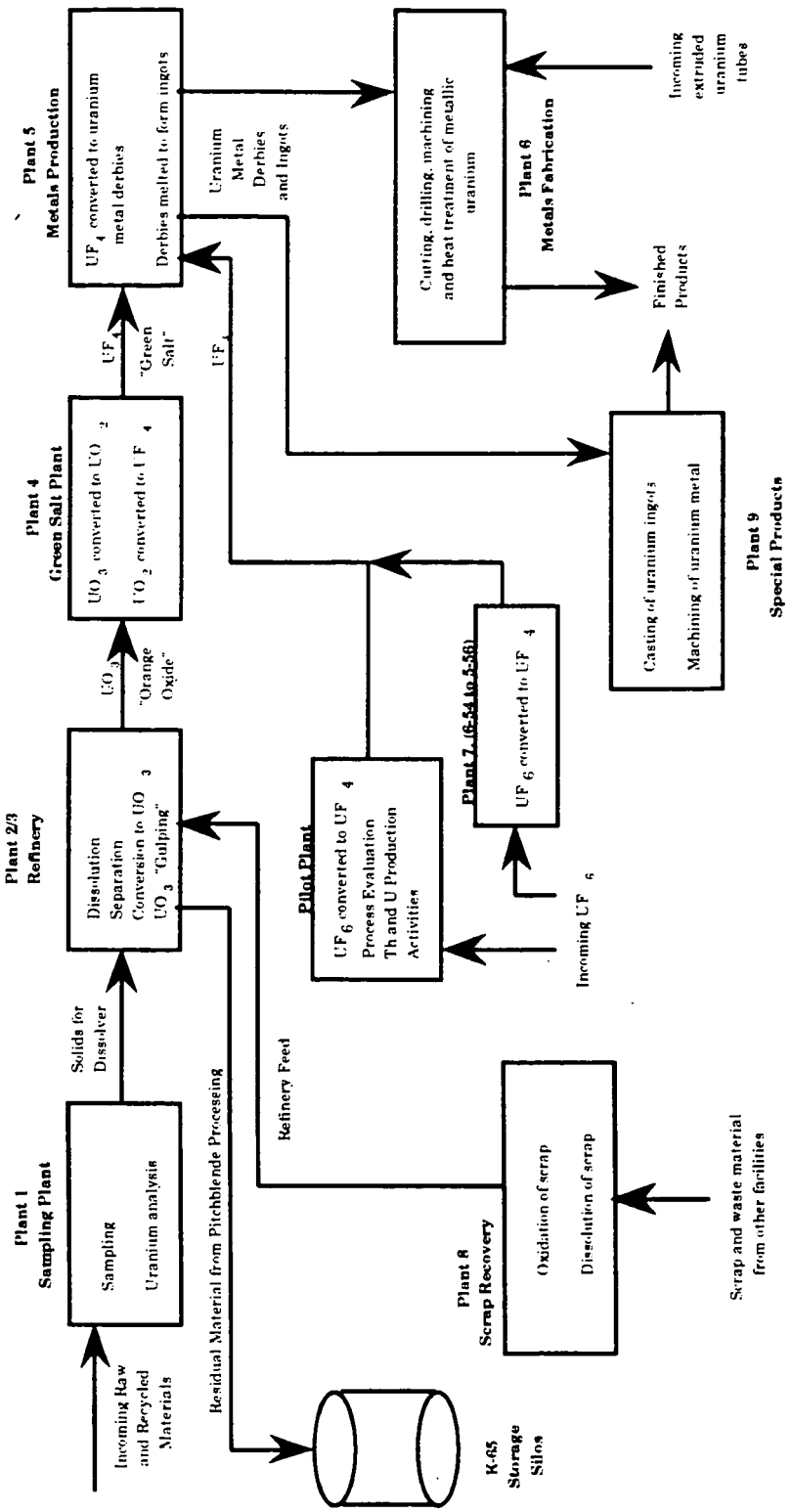

Figure 4. Schematic Diagram of Flows of Uranium Between Facilities at FMPC. Flows of recycled material were more complex than can be illustrated simply. For example, recycled uranium received as uranium trioxide would not be processed in Plant 2/3, but would be feed for Plant 4.

In Plant 8, the Scrap Recovery Plant, waste materials and metal scraps from the production processes were heated to remove impurities before being sent back through the Refinery (Plant 2/3) and the production process. The Pilot Plant was used for the direct conversion of incoming enriched UF_6 (uranium hexafluoride) to UF_4 (green salt).

Much of the thorium production activity at the FMPC took place in the Pilot Plant, beginning in 1954. Plant 7 operated only from 1954–1956 in converting UF_6 to UF_4.
Waste materials from these processes were treated in various ways at the FMPC depending upon their physical form. The K-65 Storage Silos, large concrete tank-like structures, store residues from the extraction of uranium from ores that were processed during the early years of FMPC operations. Liquid effluents were collected and treated at the general sump before being discarded to the waste disposal pits. Liquids from the clearwell portion of the waste pit, along with the storm sewer runoff and sewage treatment plant effluent were piped to the Great Miami River from Manhole 175 on the eastern boundary of the site. Solid waste materials were sent directly to the waste pits, or they were burned in the incinerator located near the eastern edge of the facility or in the burn pit near the waste pits. The FMPC also operated a graphite burner from 1965 to 1984, an oil burner from 1962 until 1979, and an incinerator for liquid organic wastes that was installed in 1983. Releases from these latter facilities are described in Appendix K.

FMPC PRODUCTION INFORMATION

Production information provides a guide to the magnitude of FMPC activities over the years. In the absence of other data, it can be used to help estimate releases from the facility to the environment. **Appendix C** contains details of the receipts and shipments of uranium at FMPC along with specific production data for each plant for the time period 1951–1988.

These records of shipment and receipts, and plant production provide several key pieces of information. First, they specify the level of *"enrichment"* of processed uranium, which relates to the concentration of uranium-235 (^{235}U) relative to uranium-238 (^{238}U).

- "Natural" uranium contains 0.72% ^{235}U.
- "Depleted" uranium contains less ^{235}U; typically 0.14–0.20% at FMPC.
- "Enriched" uranium contains more ^{235}U; typically, 0.95–1.25% at FMPC.

While most of the enriched uranium was in the above range, some processing of 2% enriched uranium occurred in the 1960s. The capability to digest 5% enriched uranium was added to Plant 1 in 1970.

Second, records of receipts of material by FMPC and shipments from FMPC provide a rough indication of production rates. Comparisons of the data on receipts and shipments indicate that material was received, processing occurred, and products were shipped on a fairly regular schedule during much of the time. During fiscal year (FY) 1952 through 1980, the FMPC received about 362,000 metric tons (MT; 1 MT = 1,000 kg = 2,200 pounds) of uranium and shipped about 358,000 MT to offsite locations (Audia 1977; FMPC 1988). Approximately 54% of the receipts and shipments were natural uranium, about 20% were enriched uranium, and some 26% were depleted uranium. Uranium shipments tended to follow the pattern of receipts during most of the years of operation.

Third, plant-specific production rates are useful for estimating releases of radioactive materials from specific facilities. Processing rates in each plant were increased or reduced because of changes in the demand for intermediate materials and finished metal products. Figure 5 summarizes the total production quantities in metric tons of uranium (MTU) for 1951 to 1988. In some plants, there was no production during certain years. For example, there was no production of UF_4 from UF_6 in the Pilot Plant from 1968 to 1984. Data on the enrichment categories of products are presented in Appendix C.

Thorium production at the FMPC was estimated to have been only about 0.4% of the uranium production. Processing was limited to a few facilities and to specific time periods. Some of the uranium received at the FMPC was recycled, that is, it had other radionuclides as contaminants in the uranium.

OTHER RADIONUCLIDE RELEASES

Radioactive decay of uranium and thorium isotopes produces series of other radionuclides that are collectively referred to as *decay or daughter products*. In most of the feeds received by the FMPC, the uranium had previously been separated chemically from the other decay products. As a result, the facility's effluents consisted primarily of uranium. Other radionuclides were generally present in small quantities. Early processing campaigns treated ores that contained nearly equilibrium amounts of the decay products. The wastes from that early processing were placed in the K-65 Storage Silos. Releases from the silos are discussed in Appendix J.

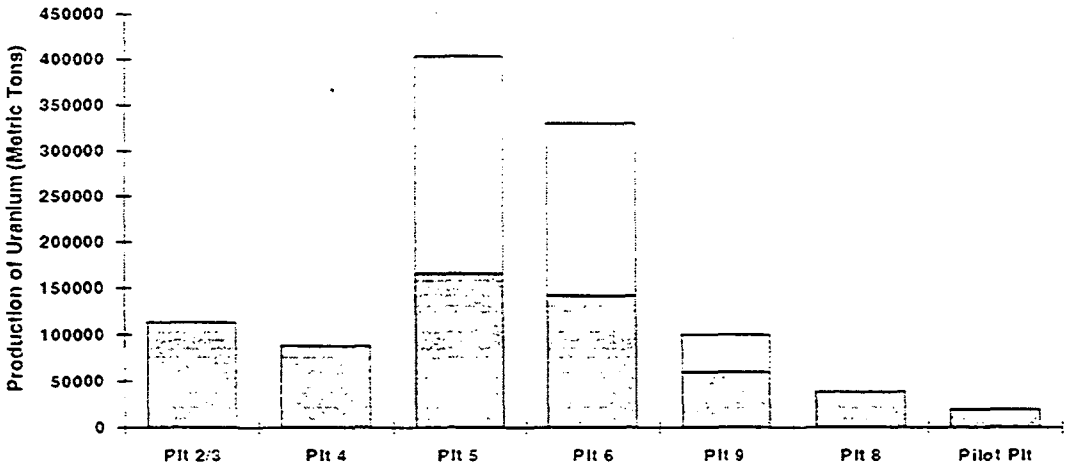

Figure 5. FMPC plant production for 1952 through 1988. Each plant produced a different product: uranium trioxide in Plant 2/3, uranium tetrafluoride in Plant 4, metal derbies (dark shading) and ingots (light shading) in Plant 5, machined (dark) or rolled products (light) in Plant 6, uranium ingots (light) and machined products(dark) in Plant 9, uranium recovered from scrap materials in Plant 8, and uranium tetrafluoride in the Pilot Plant.

Because *recycled feed materials* were sometimes used, small amounts of other radionuclides called *fission and activation products* were also introduced into the process stream and later released. Recycled uranium was not processed at the FMPC prior to October 1962, so releases of fission and activation products did not occur prior to that time. Measurements of the amounts of these radionuclides, relative to uranium, were not performed until years later. These products were measured from airborne effluents (in scrub liquor or dust collectors) at only one time in 1985.

Appendix D provides the measurement data of fission and activation products in particulate materials done at that time. The concentration of fission products— strontium-90, technetium-99 and cesium-137—were highly variable. The transuranic nuclides - neptunium-237, plutonium-238, and plutonium-239,240 were detected in all of the samples analyzed, but the observed concentrations varied over a wide range within the plants and from plant to plant. Only the short-lived decay products of uranium-238 were found in consistent concentrations. The concentrations of thorium isotopes and their radium decay products were found to be consistent in samples from some plants but not from others. In some plants, the concentrations of transuranic nuclides (*TRU*) were clearly affected by the processing of material containing unusually high concentrations of TRU between 1980 and 1985.

Measurements of radionuclides other than uranium in liquid effluents are available for a longer time period than for airborne effluents. There was no processing of thorium during the time periods 1952–1953, 1958–1963, or since 1980. Relative concentrations of thorium with respect to uranium were measured in the mid-1950s, and again beginning in 1967. Beginning in 1976, the concentrations of plutonium, neptunium, radium and the fission products, cesium-137, ruthenium-rhodium-106, technetium-99, and strontium-90, were measured relative to uranium. The concentrations of these other radionuclides in liquid effluents are shown in Appendix D. Estimates of the amounts discharged in liquid effluents are presented in **Appendix L.**

The relative importance of various radionuclides as potential contributors to offsite radiation doses was assessed using a methodology developed by the *NCRP* (National Council on Radiation Protection and Measurements) (NCRP 1989). These calculations show that releases of uranium are by far the most important contributors to the potential doses from releases to the atmosphere at the FMPC. For liquid releases, the radium isotopes were found to be of primary importance, depending upon the pathway considered.

FMPC RELEASES TO THE ENVIRONMENT: FACTORS TO CONSIDER

Several factors regarding source term estimates were considered at the outset of the project. These included the initial period of study, characteristics of radionuclide releases, the uncertainties involved in making source term estimates, and the sources of information that would be used for this process. To apply resources most efficiently, it was necessary to assign priorities to the three source terms — airborne effluents, liquid waste discharges, and inputs to the groundwater — according to their importance. The greatest emphasis was given to those releases that had the largest potential impact on the population residing in the vicinity of the FMPC. All the evidence, which will be documented throughout the report, indicates that airborne releases deserve the greatest attention. That conclusion influenced the level of detail of the investigations and the corresponding reports in this series.

Period of Time Studied

Although radionuclide source terms are reported here for the entire operating history of the FMPC (1951–1988), our initial effort focused on a shorter time period (Voillequé et al. 1991). Originally, we considered examining 1955, the year of the highest reported releases to the atmosphere (Boback et al. 1987). During a September 1990 site visit, it was confirmed that the installation of effluent sampling equipment was not complete during 1955. The quality and variability of results from an operational effluent sampling system are needed to estimate source term uncertainty, needed for Task 3.

Other factors indicated that a time period in the early 1960s was the best focal point for the initial work on source terms and their uncertainties. These included the availability of environmental samples and records along with a level of emissions which make uncertainty analysis workable. We were also able to locate other documentation that was needed to derive source term estimates.

Analysis of data from a period of relatively consistent operation (1960, 1961 and 1962) has provided a basis for estimating source terms for other periods when fewer measurements were made and when there were more unmonitored release points. The interim draft Task 2/3 report addressed releases to the atmosphere, to surface water and to groundwater by the FMPC for the period 1960–1962. In the current report, we use the same methods of investigation to derive annual source term estimates for uranium and other radionuclides released in air, surface water and ground water from the FMPC for the entire period 1951–1988.

Characteristics of Radionuclide Releases

Initially, it is important to identify specific attributes of the radionuclide release, or source term, to be documented. The most important parameters that are common to all releases include:

- nature of release: Was it routine or episodic?
- magnitude or size of the release
- radionuclides released

For the surface water source term, the discharged radionuclides in waste water were either in solution or in suspension as finely divided particles. In either case, the radioactivity was carried from the FMPC site via a pipeline to the Great Miami River or in the storm sewer overflow via Paddy's Run, a small stream at the west boundary of the site. Paddy's Run joins the Great Miami River approximately 3 kilometers south of the FMPC (Figure 2).

Radioactivity reached the groundwater by infiltration in a form similar to that in liquid discharges. The radiation doses from consumption of water from either source depend on the amounts released and upon the dilution in the river or the aquifer before withdrawal for human use.

For radionuclide releases to the atmosphere, there are two other factors, besides release rate and dispersion, that are important determinants of the radiation doses to members of the public. These are:

- the chemical form of the discharge
- its physical characteristics, primarily the size distribution of the released particles

Human metabolism of radionuclides that have been inhaled is dependent upon the chemical form of the radionuclides. Soluble compounds are readily taken up into the blood stream and are rapidly distributed throughout the body. Chemical forms that are insoluble in body fluids tend to be retained in the lung for a longer time and are only gradually transported to other tissues. The chemical form of the discharges are presented in the appendices describing atmospheric releases.

The particle-size distribution is important for calculating the amounts of radioactive material that were deposited on the ground following release. Particle size is also important for estimating the radiation dose from inhalation of the particles.

Uncertainties in Estimating Releases

Results of scientific investigations are, by their nature, uncertain, and it is a common practice for investigators to provide some estimate of uncertainties that affect their estimates. Estimating the uncertainties associated with the source term estimates (Task 3) is, therefore, an important part of this work. The absence of uncertainty estimates is a weakness in the previous source term information.

Knowledge of several parameters, or numbers, is required to define a radionuclide release. None of them is known exactly, and most are contributors to the overall uncertainty associated with the release estimate. Two types of parameter uncertainty affect the overall source term uncertainty (Hofer and Hoffman 1987). The first is due to random variations in sampling, measurement, and operational procedures. For example, estimates of uranium releases to the atmosphere are based upon analytical measurements of the sample mass, the percent of the collected mass that is uranium, the flow rate through the sampler, the flow rate through the stack, etc. The physical dimensions of the sampling probe and the exhaust duct are also factors. Although the latter two quantities are fixed and relatively well known, each of the other measurements is rather more uncertain, for various reasons. This uncertainty contributes to the overall uncertainty of a particular release estimate.

A second type of uncertainty occurs because of a lack of knowledge about particular parameters. This may occur because the parameters were not measured during part or, in some cases, most of the period of facility operation. Examples of this type are periods when the stack sampler flow rate was not measured, and periods when the stack flow rate was not measured. In these cases, estimates of the values of those parameters during the periods between measurements will be necessary. In the absence of definitive information, subjective judgment of experts can be used to estimate the range and distribution of values for the unknown parameters during such periods.

The technique of using a computer to draw many random samples from the parameter distributions and combining these sample releases to obtain information about the

distribution of the releases is an example of what is called a Monte Carlo procedure. Figure 6 illustrates this process.

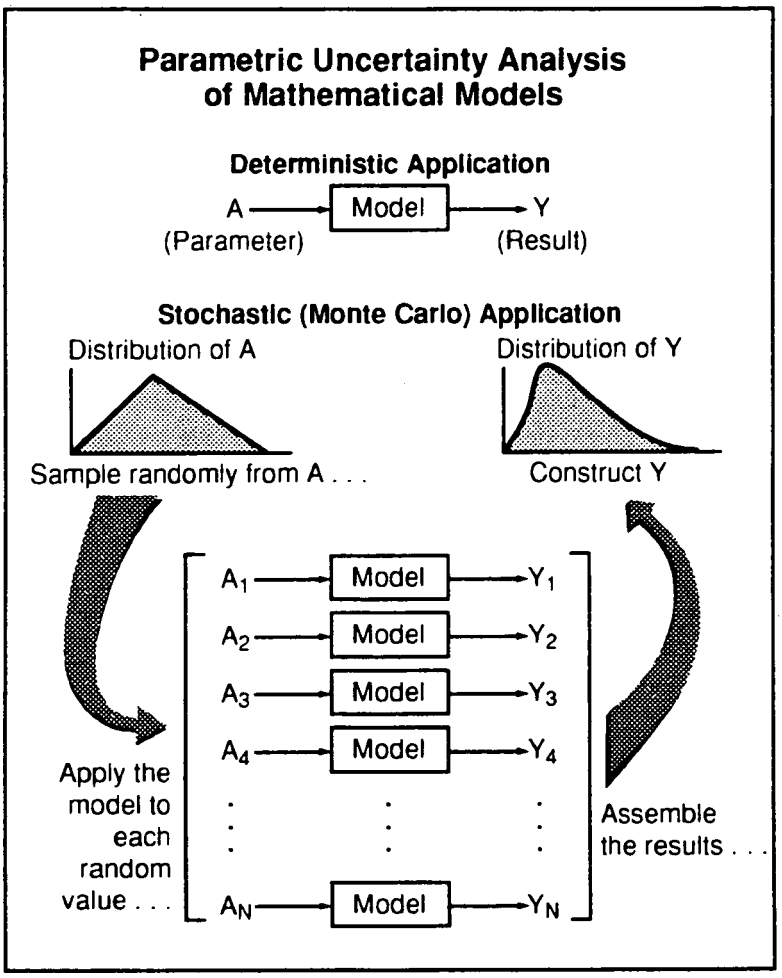

Figure 6. Schematic presentation of Monte Carlo methods for propagating a parametric uncertainty distribution through a model to its results. In this simplified illustration, A is an input parameter to the model, and Y is the result, or output, corresponding to A. For each specific value of A, the model produces a unique output Y. Such an application of the model is deterministic, because A determines Y. But A may not be known with certainty. If uncertainty about A is represented by a distribution, such as the triangular one in the figure, repeatedly sampling the distribution at random ad applying the model to each of the sample input values A_1, A_2... gives a set of outputs Y_1, Y_2,..., which can be arranged into a distribution for Y. The distribution of Y is then our estimate of the uncertainty in Y that is attributable to uncertainty in A. This is a stochastic, or Monte Carlo application of the model.

Our use of a Monte Carlo procedure to estimate releases explicitly recognizes that those estimates are uncertain because of variability or lack of knowledge of the parameters upon which the estimates depend. This procedure applies our best estimates of the distributions of parameter values to produce a distribution of results. Our approach contrasts with one in

which a calculation is based upon point estimates of the various parameters and yields a single result. The Monte Carlo calculation carries the underlying uncertainty in the parameters forward and displays it in the breadth of the distribution of results.

This process was illustrated in the interim Task 2 and 3 report (Voillequé et al. 1991) by examining the distributions involved in the calculation of releases from the Plant 8 scrubbers for May 1961, and for that entire year. The estimated release from those scrubbers depends upon two parameters: the amount of uranium collected in the scrub liquor and the penetration of uranium through the scrubbers. The Monte Carlo procedure for estimating the Plant 8 scrubber releases involves independent selection of values of the two parameters and the use of the selected values to compute an estimate of the release. This procedure was performed repetitively (5000 times in the current example) and yielded a distribution of results.

Just as these source term estimates reflect the underlying variability and lack of knowledge about individual parameters, the radiation dose calculations, performed in a subsequent task (Task 6), will consider the range of source term values for a given year. They will also incorporate uncertainties about meteorological dispersion, particle deposition, and other parameters to produce distributions of estimated doses to people residing near the FMPC.

Sources of Information

A major effort in the Fernald Dosimetry Reconstruction Project has been searching for, and reviewing, hundreds of documents related to the operation of the Feed Materials Production Center since operations began in 1951. It has been our practice to trace the information back to original sources whenever possible. In the Task 1 report, issued in January 1991 (RAC 1991), we outlined the general approaches that we have taken to obtain this information. These five methods, which have formed the foundation for the project in providing the technical data for this study, are:

- site visits to the FMPC facility;
- investigation of records and scientific literature pertaining to the FMPC;
- retrieval and review of documents from NLO, Inc. using their computer database of document titles;
- examination of engineering diagrams, site blueprints, historic photographs and maps; and
- discussions with current and former longtime employees.

Because we realized the importance of retrieving documents from a wide range of sources, considerable time has been spent identifying types and locations of reports and records pertinent to the completion of this project. We visited a number of locations around the country to review documents that might provide background information on FMPC operations (Figure 7). Generally, this documentation of FMPC operations and releases comes from two broad areas: (a) from National Lead Company of Ohio, Inc. (NLO), the former operator of the site, the Westinghouse Materials Company of Ohio (WMCO), the site operator from January 1, 1986 through 1992, and the Department of Energy (DOE); and (b) from FMPC-independent sources. **Appendix A** provides a detailed look at the sources and locations of documents used for the project.

While not all the original records are still available, many original documents remain in the files at the FMPC facility, in the library of the NLO offices, and in storage facilities utilized by WMCO. Many hours have been spent examining original plant documents, particularly those related to effluent discharge measurements and procedures. The information sources can be categorized as follows:

- processes descriptions for the various facilities
- plant operating procedures
- effluent sampling procedures
- daily and monthly reports of liquid effluent discharges
- monthly reports of airborne effluent discharges
- original analytical data sheets recording sample concentrations
- plant operating process logbooks
- nuclear materials control reports
- daily sump discharge logbooks
- topical reports related to effluent characteristics
- reports of ventilation system tests and evaluations
- incident reports
- investigation reports
- letter reports of operational problems
- production records for specific processes

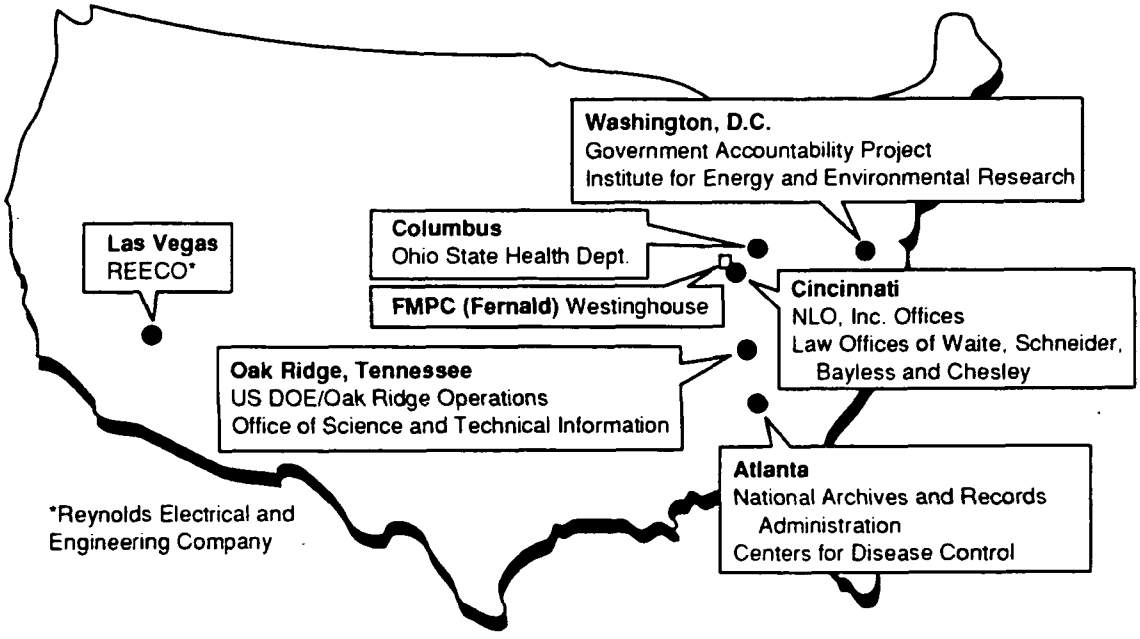

Figure 7. Locations visited in obtaining FMPC-related documentation and information.

Discussions with long-time employees and retirees from the FMPC provided another source of information for the project (RAC 1991). Their recollections on processes and procedures that routinely occurred since facility start-up served to identify sources and

locations of documentation. Documents used in the construction of the source terms are referenced in the appropriate section of the text, with the references listed at the end of the appendix or section. In addition, we have maintained a collection of all documentation that we have reviewed since the Fernald Dosimetry Reconstruction project began in 1990. Appendix A lists all documents that have been added to the RAC Document Repository up to this time.

In general, data from original records used in this study are reported in the same units that appeared in the source documents. For example, the uranium concentrations in liquid effluents and volume measurements, compiled in Appendix L, are reported in mg L^{-1} and gallons, respectively. In contrast to some of the original sources of information, our final release estimates and results of other calculations are reported to only two significant figures.

ATMOSPHERIC RELEASES FROM DUST COLLECTORS

Atmospheric releases from FMPC operations came from buildings where uranium processing took place and from outside areas such as the waste pits and incinerators. Appendix K reviews estimates of emissions of uranium from miscellaneous unmonitored processes, non-routine events, and episodic releases. Some airborne effluents were treated with one of the two treatment systems used at the FMPC: scrubbers or dust collectors. These treatment systems are discussed in **Appendix B**. Dust collectors employed bag filters to remove airborne particulates from an exhaust stream. Information on effluents from dust collectors is presented in **Appendix E**. The key points of the dust collector operation and our estimates are presented here.

Dust Collector Operation

Process area ventilation air was ducted to dust collectors where airborne particulate material was removed before discharge through the stack to the outside. The dust collectors recovered valuable uranium that would otherwise be lost and reduced worker exposure in the process area. When operating as designed, the dust collector systems could be quite efficient (Drinker and Hatch 1956, Ross and Boback 1971).

The sampling systems installed in the dust collector stacks were simple in concept. A schematic diagram of the sampling system is drawn in Figure 8. Air was drawn from the exhaust stack through a sampling line to a pleated cellulose filter for collection of particulate material in the sample of discharged air. The filters were periodically changed and submitted for analysis. Details of the design and operation of these systems and of the sample analysis and data reporting are given in Appendix E.

Distribution to all the plants of an initial stack sampling procedure seems to have occurred in February 1956 (Starkey 1956). Later that year a formalized procedure was developed (Boone 1956). Initial sampling frequencies were weekly, biweekly, or monthly depending on the magnitude of the previous effluent measurements. Monthly reports of releases were made to plant management by the Industrial Hygiene and Radiation (*IH&R*) group.

The sequence of reports itself documents the onset and growth of the dust collector effluent sampling program. Periodic sampling of some stacks was performed as early as

1953; however, the continuous sampling program did not begin until April 1955. Initiated in seven stacks in Plant 4 and 5, the sampling program grew fairly rapidly to encompass thirty stacks six months later. There were increases in the 1950s to a maximum of 50 sampling systems in May 1958.

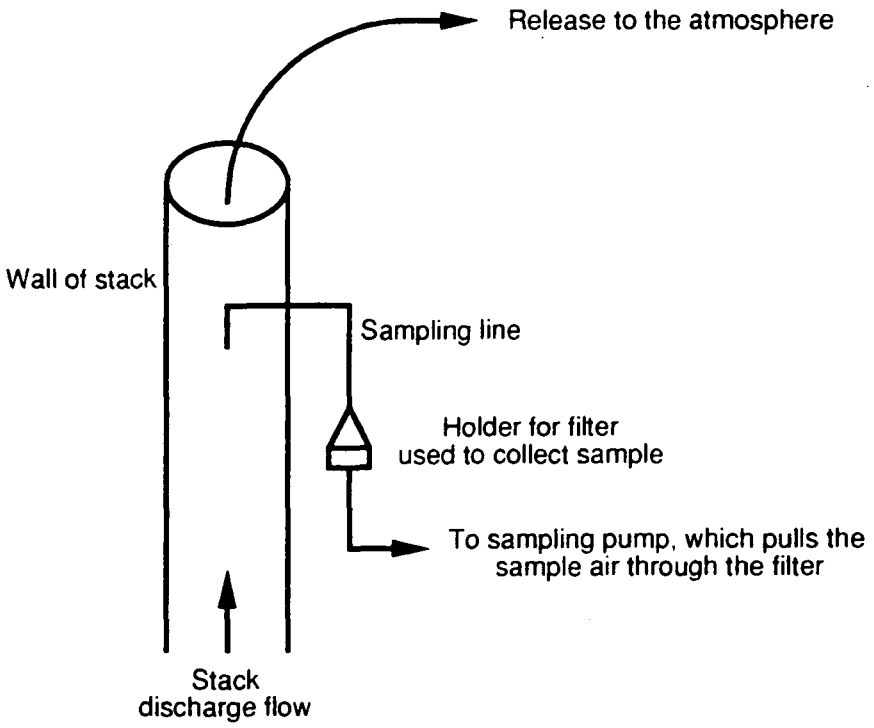

Figure 8. A schematic diagram of the dust collector stack sampling system

At the start of 1960, there was a decline to 44 samplers for dust collector exhaust due to shutdown of systems in Plant 1 and in the Pilot Plant. At that time, the most common sampling interval was one month, although a few stacks were sampled more frequently. In the 1960s, sampling intervals were occasionally as long as six weeks for discharge points that were minor contributors to plant uranium releases. In later years, both plant production and staff were reduced. Intervals between sample analyses were greater and routine reports contained less detail. Filters were no longer changed and analyzed regularly. Filter changes and analysis occurred primarily when the filter had collected a visually detectable amount of particulate material.

Current Estimates of Release From FMPC Dust Collectors

Estimates of releases from individual dust collectors at the FMPC were tabulated from original records, which were usually monthly reports of the measurements. Review of the reported results revealed periods when samplers were not in operation and other times when the releases were too low to be detected. Estimates were made for these periods based on other sampling results and information about the sampling and analysis procedures.

Estimates were also made for years before monitoring was established as a routine procedure. These estimates were based either upon normalized release rates soon after routine monitoring was established or representative measurements during the mid- to late-1950s. In some cases, evaluations of unmonitored effluents led to significant increases over previous release estimates.

There are two major deficiencies in the tabulations of reported releases in the monthly reports. The first is that the release estimates were incomplete. Release estimates were not provided for sampling periods when samplers were not installed or were not operational for the entire period. The second deficiency in the tabulations is the failure to properly account for undetected releases. If no material was detected on the filter from a dust collector exhaust sample, the reported release was shown as zero. There were entire months when either no samples were collected or no releases were detected in the dust collector exhausts because the total reported releases from some of the plants were zero. To develop a better estimate of the releases for this report, it was necessary to estimate the unmonitored and undetected releases by determining the maximum release that could have occurred when none was detected. The details of this method are given in **Appendix E**.

In addition to correcting for unmonitored and undetected releases, the initial releases estimates are subject to further revision to account for biases in the effluent measurements themselves. While the design of the sampling systems was generally well conceived, three types of deviations from ideal sampling conditions may have biased the dust collector discharge estimates.

- Nonrepresentative sampling may have occurred when particles were not uniformly mixed in the exhaust at the location of the sampler. This is more likely to happen when the exhaust ducts are greater than 15 cm in diameter. The ANSI (1969) guide recommends multiple sample withdrawal points for ducts greater than 15 cm in diameter. The reason for multiple probes is to provide assurance that the samples will not be biased because of a nonuniform distribution of the contaminant in the stack. The sample extracted from the center of a dust collector exhaust stack would be representative if the particles were uniformly mixed in the exhaust or if the concentration on the centerline happened to be equal to the average concentration in the stack. When this is not the case, the sample is not representative of the material being discharged. The bias introduced may be positive or negative. A qualitative assessment of nonrepresentative sampling in presented in **Appendix G**.

- Anisokinetic sampling may have occurred. This occurs when there is a mismatch between the fluid velocity in the probe and that in the stack. If the velocities are not the same, over- or under-sampling of particles of various sizes could occur. The possible effects of anisokinetic sampling conditions were calculated using the methods described in Appendix G. That appendix contains example calculations and the basis for parameters used in Monte Carlo calculations of bias due to anisokinetic sampling.

- Losses of particles in the sampling line can occur when particles are deposited on the walls of the line, or when they are impacted due to the presence of bends in the lines between the probe and the collection filter. Neither topic has been addressed in

previous analyses of the uranium release data. It should be emphasized that sample line losses lead only to underestimates of the effluent releases. The magnitudes of such losses depend upon particle size and density (**Appendix F**), the configuration of the sampling line, and the operating conditions for the line. These relationships are described in Appendix G.

A Monte Carlo procedure was used to estimate the sampling biases and their uncertainties. The calculations considered the three sources of bias identified above to obtain a measure of overall sampling bias. Major contributors to the uncertainty were the velocity of air in the sampling probe and in the duct, the bias due to nonrepresentative sampling, and a parameter used in computation of the attachment fractions. There is no simple way to reduce the largest uncertainties, which principally reflect the absence of information about conditions of past operations and sampling. Corrections for these biases are applied in estimating the dust collector uranium losses in **Appendix E**.

Once released from the stack, the physical and chemical characteristics of the uranium are important in the transport and deposition of released uranium and in the estimation of the radiation dose due to uranium inhalation.

- Particle size distributions were measured for some of the effluent streams in 1985. Those data and information about other uranium processing facilities have been used to estimate particle size distributions for the dust collector exhausts in this report (See Appendix F and Appendix E). Particle-size distributions for the stack emissions measured in 1985 are included as a part of the source-term characterization for stacks for all years because the plant processes served by the stacks have not changed significantly since the start of FMPC operations. Appendix F contains information on the reported measurements done in 1985. The distributions cover wide ranges of particle sizes and are not truly lognormal. The ranges of particle sizes have been subdivided into intervals and representative sizes are used in the calculations. Average particle-size distributions for both the inlet and the outlet ducts for stacks emitting UF_4 and U_3O_8 were derived from the data in Appendix F. The average distributions and distributions obtained from similar facilities are used for FMPC exhausts for which particle size measurements were not made. In spite of some substantial variations from stack to stack, the particles were relatively large.

- The chemical form of the materials discharged from the dust collectors affects the particle density, the transport and deposition of released uranium, and the estimation of the radiation dose due to uranium inhalation. The predominant uranium species emitted from each stack was identified from FMPC reports and engineering drawings of process equipment. About three-fourths of the releases from the dust collectors were in the form of uranium oxides.

The process of developing revised estimates of releases from the FMPC dust collectors is complex. Reported releases were incomplete because sampling was not initiated when production began. The reported releases do not include estimates of releases that were

undetected by the analytical procedure or because a sampling system was temporarily out of service. The three sources of possible bias in the reported results, discussed above, have been estimated as part of this effort.

The first step in the approach adopted was to return whenever possible to the original release reports that were prepared routinely by the IH&R department. In the early years of full operation of the effluent sampling program, these reports contained a great deal of information about sample collection and about operational problems in all the plants. These detailed reports made it possible to estimate the magnitudes of undetected releases. Later reports of results, when production rates and releases were lower, were not as detailed and were much less helpful in this regard. In general, inclusion of undetected releases does not have a large effect on the estimates for early years when releases were large. In plants whose releases were relatively small (tens of kilograms of uranium per year) the relative contribution of estimates of releases that had gone undetected was greater.

Overall, corrections for unmeasured releases and for sampling bias led to revised release estimates that were about 50% higher than previous estimates of dust collector releases. Table 1 shows that the median estimate of total releases from the FMPC dust collectors from 1951 to 1988 was about 140,000 kg uranium. Most releases occurred during the 1950s. Principal contributors to the releases during that decade were Plants 4, 7, and 5. Plant 8 also contributed significantly to the total, but most of those releases occurred over a longer period of time. Although releases from the other facilities were not small, those releases were not major fractions of the total release. However, some of the releases from plants that were lesser contributors to the total were important in individual years.

Table 1. Summary Release Estimates for FMPC Dust Collectors

Period	Best estimate of release (kg U)	Other percentiles in distribution of release estimates (kg U)			
		5th percentile	25th percentile	75th percentile	95th percentile
1950s	120,000	96,000	110,000	130,000	150,000
1960s	21,000	18,000	19,000	22,000	24,000
1970s	3,100	2,500	2,800	3,400	3,800
1980s	2,100	1,700	1,900	2,400	2,700
1951–1988	140,000	120,000	130,000	160,000	170,000

DISCHARGES FROM PLANT 2/3 DENITRATION OPERATIONS

The air emitted from release points not equipped with dust collectors was cleaned through scrubbers. Scrubbers used either acid or caustic solutions to scavenge particles from the air stream being discharged to the atmosphere. Most of the particles are scavenged by mist droplets, which, for the most part, are collected by mist-eliminating devices and recycled to the liquid reservoir. This liquid (*scrub liquor*) is changed periodically. The uranium-containing droplets accumulate on the mist-eliminators, and some of the liquid is agglomerated into larger droplets and escapes back into the exhaust gas stream in a process called *reentrainment*. Figure 9 illustrates these processes. In this manner, the scrubbers of Plant 2/3 and Plant 8 emitted liquid droplets of reentrained scrub liquor of varying uranium concentration.

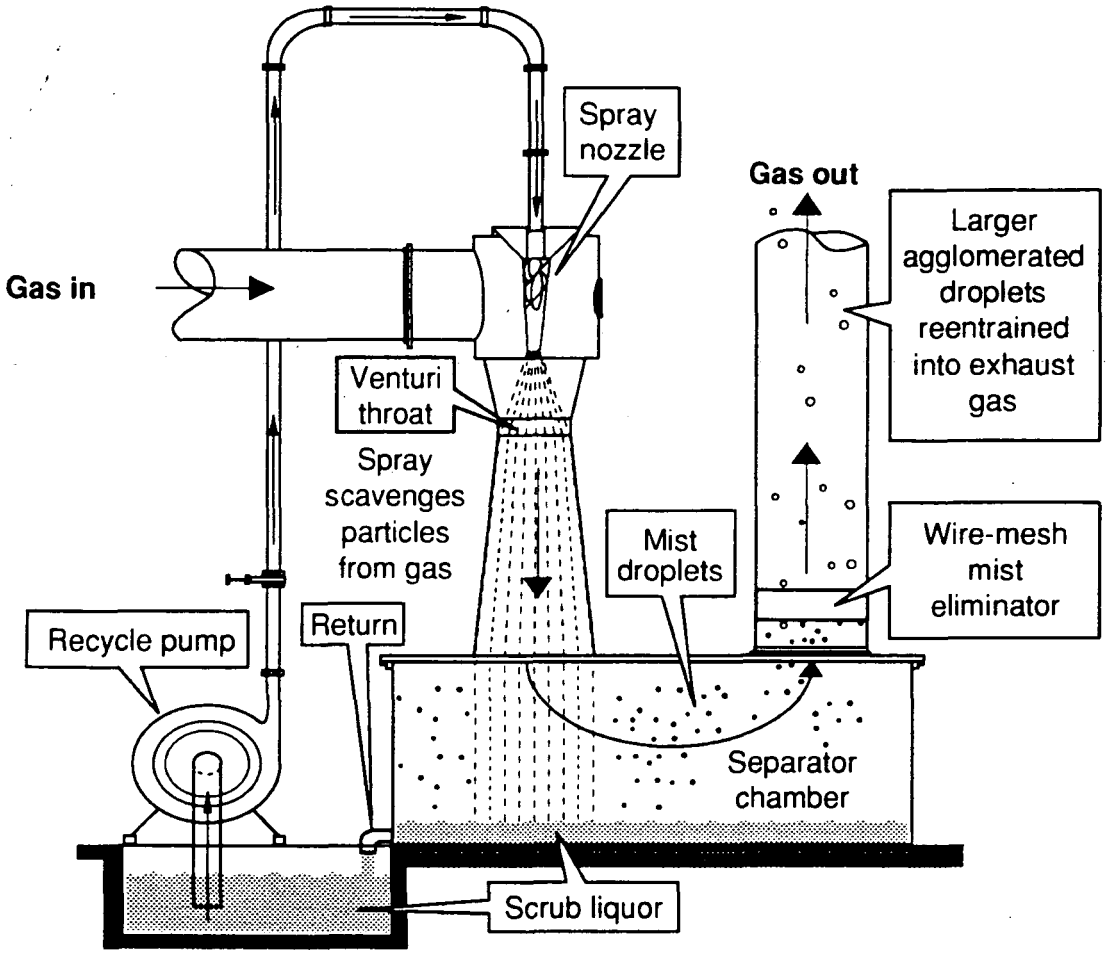

Figure 9. Scrubber schematic. Exhaust gas entering the scrubber is forced through a liquid spray into a Venturi tube. The gas then passes through a separator chamber and into the outlet duct. The spray entrains most particles into liquid droplets. Most of the liquid (or scrub liquor) is collected in the separator chamber and returns to a reservoir from which it is recycled. The scrub liquor of the Plant 2/3 and Plant 8 scrubbers was changed periodically and uranium was recovered from it. To inhibit the escape of the uranium-containing droplets various mist-eliminating systems were used. The figure indicates a wire mesh mist eliminator in the outlet duct (as in Plant 2/3), which would trap most droplets. But some of the trapped liquid was reentrained into the gas stream as large agglomerates and escaped to the atmosphere. Evaporation of the liquid produced relatively large solid particles.

After 1956, exhausts from the *denitration* process in Plant 2/3 were treated by a wet scrubber prior to discharge to the atmosphere. In the denitration process, nitrates were removed from uranyl nitrate hexahydrate (UNH) to produce uranium trioxide (UO_3, or orange oxide). Fumes of oxides of nitrogen that were produced during denitration were routed to the scrubber system. In a second process, orange oxide from the denitration pots was transferred by vacuum or "*gulping*" to a storage hopper. The releases of uranium from the scrubber exhausts were not sampled, even periodically, until recently. In June 1988, an investigation of higher than expected environmental radioactivity measurements led to the

conclusion that releases from Plant 2/3 processing activities were the source of the observed higher offsite air concentrations (Investigation Board 1988). **Appendix H** provides details of the scrubber exhaust system, our current approach to estimating releases from the Plant 2/3 scrubbers, and previous release estimates. Because information is lacking on early operations with dust collectors, releases for those years are estimated using the same model used for years when the scrubbers were in operation.

Current release estimates are based upon a review of the following:

- previous release estimates (Semones and Sverdrup 1988);
- plant operating data from 1969, 1970 and 1973;
- the Shift Foremen's Logs for 1956–1962 and 1967; and
- uranium trioxide production data.

The log sheets and logbooks contained information on parameters important for the calculation of releases due to gulping operations. Uranium released from the Plant 2/3 scrubbers is composed of releases due to scrub liquor *entrainment* and to particles of UO_3 in the air stream that pass through the scrubber. Independent estimates of releases from the Plant 2/3 scrubber system were performed using models of scrubber penetration by particles and mist reentrainment that were based upon the recent effluent measurements. Monte Carlo techniques were then used to sample the parameter distributions and the randomly selected parameter values were used to make the release estimates. The parameters considered in calculating the releases estimates are:

- scrubber outage fraction
- scrub liquor concentration
- entrainment release factor
- amount of UO_3 in a pot
- gulping time
- gulping release factor.

Estimates of Plant 2/3 scrubber releases obtained from the Monte Carlo calculations are shown in Table 2 by decade. Median estimates of releases during three of the four decades of operation are comparable, about 20,000 kg, while the value for the 1980s was much lower. The median release estimate for the entire period of operation was 66,000 kg uranium. This estimate was bounded by 5th and 95th percentile values of 56,000 and 78,000 kg uranium, respectively. The highest annual releases were estimated for the period 1957–1961.

Table 2. Summary Release Estimates for Plant 2/3 Scrubbers

Period	Best Estimate of Release (kg U)	Other percentiles in distribution of release estimate (kg U)			
		5th	25th	75th	95th
1950s	24,000	18,000	21,000	26,000	32,000
1960s	19,000	14,000	17,000	21,000	25,000
1970s	22,000	17,000	20,000	25,000	29,000
1980s	980	730	850	1,100	1,600
1953–1988	66,000	56,000	62,000	71,000	78,000

About 40% of the release is estimated to have been small particles of UO_3 that penetrated through the scrubber. The larger fraction (~60%) would have been uranyl nitrate hexahydrate (UNH). The estimated size range for these particles is 19–100 μm. An alternative calculation of releases from the Plant 2/3 denitration operations, based on a change in the outage fraction, is described in **Appendix H**.

RELEASES FROM PLANT 8 SCRUBBERS

Descriptions of Plant 8 operations, scrubber efficiency measurements, and the basis for both previous and current release estimates are given in **Appendix I** of this report and in the Task 4 report (Killough et al. 1993). Ten air scrubbing systems in Plant 8 cleansed, or scrubbed, the exhaust air by contact with droplets of caustic liquid. Six of the scrubbers—the rotary kiln, oxidation #1, the caustic or primary calciner, uranium ammonium phosphate (UAP) furnace, the oxidation #2 or NPR, and the green salt reverter—handled hot exhaust gases from the kiln and furnaces. The other four scrubbers—old digester, new digester, the ammonium diuranate (ADU), and the leach tank—treated ventilation air collected above the digestion and other process tanks. Some of the key findings that affect the current release estimates are:

- The exhausts from these systems were not sampled on a regular basis. Periodic measurements of discharge concentrations and of scrubber efficiencies were performed by the Industrial Hygiene and Radiation Department. A number of their measurements for the caustic, kiln, UAP, and NPR scrubbers were made during the early 1960s, a period of substantial concern about releases of uranium from these systems. In the early 1980s, when Plant 8 production was lower, measurements were made to determine emission factors for the Plant 8 scrubber discharges.

- There were no reported measurements of the sizes of the particles or liquid droplets released to the atmosphere from the Plant 8 scrubbers. A theoretical analysis of Plant 8 scrubber operations was conducted to estimate these particle size distributions [see Appendix D of the Task 4 report (Killough et al. 1993)]. About 30% of the total uranium emitted from the Plant 8 scrubbers included solid particles of U_3O_8 of less than 10 micrometers in diameter. The remainder of the released uranium from the scrubbers escaped as large droplets (80 to 180 μm in diameter) of reentrained scrub liquor. Evaporation of the liquid produced relatively large solid particles.

Previous estimates of releases from the Plant 8 scrubber systems were reviewed. An important difficulty with previous estimates of the Plant 8 scrubber releases was the assumption of a constant scrubber efficiency. Just as with these previous estimates, current estimates require knowledge of scrubber efficiencies and uranium concentrations in the scrubber liquor. Plant records were found in storage that provided data on the amounts of uranium scrubbed from the airborne effluents during periods ranging from one month to one year. Plant 8 production (uranium recovery) data were compiled to indicate the changing scale of plant operations. Memoranda and analytical data sheets were located that

described measurements of scrubber efficiencies performed in Plant 8, primarily during 1961–1965. These data were compiled for each scrubber for use in calculations of releases from 1953 through 1981. Data collected in the 1980s on short-term measurements of release rates from the various stacks were also compiled and used for calculations for this later period.

For the years 1953–1981, annual uranium releases from the Plant 8 scrubbers and the uncertainties associated with them were estimated by applying a simple model to each scrubber. The calculations used the following plant-specific data:

- Plant 8 production (uranium recovery) data;
- amounts of uranium found in scrub liquor;
- the amount of uranium in scrub liquor per unit production;
- the use and performance of the scrubbers serving the calciner, rotary kiln, UAP furnace and the two oxidation furnaces.

For the latter years of FMPC operation (1982–1988), release estimates were based upon the operating times for the various scrubbers and measurements that had been made of release rates during scrubber operation. For both time periods, simple models of releases were applied to individual scrubbers. When information on scrub liquor collections was not available, the 6- to 12-month average ratio of plant production to the amount of uranium collected in scrub liquor was found to be a reasonable link between production data and scrubber operations.

Monte Carlo calculations were performed to estimate uranium releases from the Plant 8 scrubbers. The ranges of all of the parameters used in calculations were relatively broad, owing both to variability and to limited historic data. Table 3 contains summary release estimates by decade and for the entire period from 1953 through 1988. The table illustrates the importance of the releases during the 1960s when plant production was highest. The median estimate for the 1950s was second highest, about 60% of that for the following full decade of operation. Alternative calculations of releases from the Plant 8 scrubbers, performed to test the effect of different modeling choices on the results, are described in Appendix I. The first involved the use of correlations between scrubber penetration and the accumulation of uranium in the scrub liquor. The second alternative approach was based on release to production ratios for the early 1960s, when the scrubbers were studied most intensively, that were applied to the entire period of operation.

Table 3. Summary Release Estimates for Plant 8 Scrubbers

Period	Best Estimate of Release (kg U)	Other percentiles in distribution of release estimate (kg U)			
		5th	25th	75th	95th
1950s	29,000	17,000	23,000	37,000	53,000
1960s[a]	47,000	30,000	39,000	57,000	78,000
1970s	1,700	1,000	1,400	2,100	2,700
1980s	1,400	980	1,200	1,600	2,000
1953–1988	81,000	56,000	69,000	95,000	130,000

[a] In making these estimates it was assumed that the bypass for the UAP scrubber operated 10 per cent of the time between September 1963 and April 1966.

To put these atmospheric releases into perspective, Figure 10 compares the uranium released annually from the dust collectors, the Plant 8 scrubbers, and the Plant 2/3 denitration processes. The dust collectors dominated the releases in the 1950s with 120,000 kg of uranium released, with a maximum of 54,000 kg of uranium released from them in 1955 alone. In the 1960s, the Plant 8 scrubbers dominated the releases, with approximately 47,000 kg uranium released during that decade, compared to 21,000 and 19,000 kg U for the dust collectors and Plant 2/3 scrubbers, respectively. In the 1970s, the Plant 2/3 scrubbers were relatively more important, discharging 22,000 kg U, compared to 3,100 and 1,700 kg U, respectively for the dust collectors and Plant 8 scrubbers. Again in the 1980s, the dust collectors contributed most to the total uranium releases, although the magnitude of all releases in the 1980s was significantly less than at any other time.

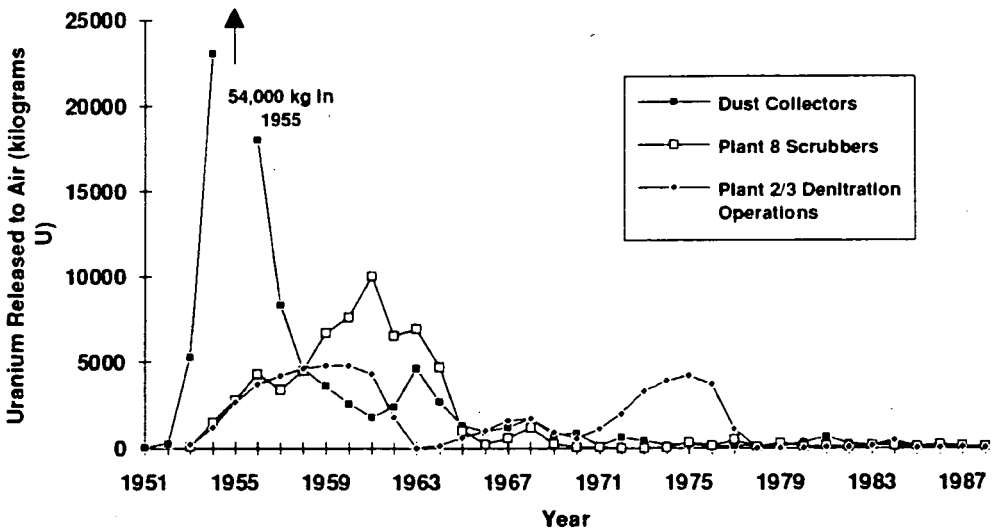

Figure 10. The best estimates of annual releases of uranium to the atmosphere from the Plant 8 scrubbers (square), the dust collectors (triangle) and the Plant 2/3 scrubbers (circle). The relative importance of each of these sources to the total atmospheric uranium release changes with each decade.

OTHER SOURCES AND EPISODIC RELEASES TO THE ATMOSPHERE

Appendix K addresses other miscellaneous unmonitored sources and accidental releases to the atmosphere. The unmonitored sources include emissions from:

- five waste incinerators,
- building exhaust and lab hood ventilation,
- miscellaneous unmonitored process emissions, and
- the waste pits.

Accidental releases include:

- non-routine events, and
- episodic releases.

Episodic releases are actual accidental releases which occurred in the past, and which were large enough to be given special treatment in terms of environmental transport and dose assessment. In addition to actual episodic releases, non-routine releases from other events, such as spills, fires and leaks of gaseous uranium hexafluoride and uranyl nitrate, were estimated in a generic way based on the frequency of occurrence of such events. Table 4 presents the total release estimates from the miscellaneous unmonitored sources. In addition, the table illustrates the difference between our reconstructed source terms and those previously developed by the FMPC contractor. In contrast to previous estimates, the reconstructed source terms all carry some estimate of uncertainty and are well documented.

Table 4. Summary of Total Estimated Releases of Uranium from Miscellaneous Unmonitored and Accidental Sources at the FMPC

		Total Release Estimate (kg U)		
Source	Inclusive Dates	Median	5th–95th Percentile Range	Previous Estimate[a]
Miscellaneous Unmonitored Releases				
Old Solid Waste Incinerator	1954–1979	2200	1600–2900	2471
Oil Burner	1962–1979	370	270–470	467
Graphite Burner	1965–1984	230	61–730	129
New Solid Waste Incinerator	1979–1986	8	0.6–90	14
Liquid Waste Incinerator	1983–1986	4	0.9–9	12[d]
Building Ventilation	1954–1987	4100	970–15,000	390
Unmonitored Process Emissions	1953–1988	[b]	110–970[c]	324
Lab Hoods	1953–1987	[b]	20–200[c]	66.5
Waste Pits	1953–1988	3000	900–12,000	1560
Accidental Releases				
Non-routine Releases[e]	1952–1988	1300	780–2900	2784
Episodic Releases[f]	1953, 1960, 1966, 1978, 1979, 1983	1700[f]	1300–2100[f]	Not defined previously

[a] From FMPC operating contractor. See individual sections of Appendix K for sources of information.
[b] Not reconstructed; estimate developed previously by the FMPC contractor.
[c] Subjective uncertainty of a factor of 3 applied to previous estimate.
[d] Based on maximum processing rate.
[e] Includes fires, spills, and leaks of uranium hexafluoride and uranyl nitrate.
[f] Does not include the November 1960 episodic release from the Pilot Plant dust collectors, which is included in the total dust collector source term. Does include two accidental releases of uranium hexafluoride and three releases (unknown sources) identified from ambient air monitoring.

Releases from these sources were more thoroughly examined than they had been in the interim source term report (Voillequé et al. 1991). There, only a few revised source term estimates were developed. Although releases from these sources were believed to be relatively minor compared with the dust collectors and scrubber emissions, the documentation to support that conclusion was lacking in most cases, and some of the previous methods used to estimate releases needed improvement. The detailed assessments in Appendix K provide thorough documentation of the magnitude of these sources, with uncertainties.

Miscellaneous Unmonitored Emissions

The agreement between past and revised release estimates is good for the incinerators. Of all incinerators at the FMPC, the old solid waste incinerator had the highest total release of uranium, with a median estimate of 2200 kg. The reconstructed median release estimate from building ventilation or exhausts (4100 kg U) is over ten times higher than the previous estimate, due to two main reasons:

(1) the use of lower dilution factor for building make-up air, and

(2) the use of higher in-plant airborne contamination levels, measured in the 1950s, to make a forward projection through 1970.

The median release estimate for non-routine releases (1300 kg U) is less than that previously calculated by Vaaler and Nuhfer (1988), although the 5th and 95th percentile range encompasses the previous estimate. The median estimate of releases from the waste pits (3000 kg U) was about twice as high as previous results, because we used a model (i.e., the resuspension algorithms found in MILDOS) that was highly sensitive to soil particle size which varied greatly among the pits.

Accidental Releases

Accidental releases are frequently characterized as increases in the effluent discharge rates due to unplanned and non-routine events. In previous historic reports, typical events included spills, fires, and cleanup system failures. However, when the frequency of the unusual events is high, one questions whether the adjective "accidental" is correct. Similarly, when a large release is the result of a conscious operational decision, it hardly qualifies as unplanned. Such situations complicate the definition of the term accidental releases; so the term "episodic releases" has been defined and used in the Fernald Dose Reconstruction Project. Criteria for an episodic release, discussed fully in **Appendix K**, that were used to determine whether special evaluation of a release from a particular event is warranted include:

- the event under consideration caused the composite release rate of the FMPC to increase by a factor of ten or more above the value that would otherwise have been observed, and
- the duration of the high release rate caused by the particular event was less than 10 days.

Six incidents involving releases of uranium were identified which met our criteria for special treatment as episodic releases. It should be emphasized that all known releases are included in the total source term estimates, but only a small number are truly episodic releases, by our definition. Three episodes, documented in incident reports, occurred on November 7, 1953, in November 1960, and on February 14, 1966. The remaining three episodes were identified by air monitoring data, although documentation could not be found to identify the sources. These events occurred sometime during the weeks ending on September 28, 1978, February 8, 1979, and September 20, 1983. In terms of total quantity of uranium released, the dust loss episode in November 1960 had the most impact, releasing about 2100 kg U over a 5 to 7 day period. However, the episode on February 14, 1966 had the largest release rate, releasing 750 kg U in one hour. A release of about 30 Ci of radon occurred on April 25, 1986, from unauthorized venting of the K-65 silos. This source term may also be treated separately as an episodic release.

Figure 11 compares the relative importance of the various unmonitored sources with releases from the dust collectors, the Plant 2/3 denitration operations and the Plant 8 scrubbers. It is clear that the magnitude of uranium releases from the miscellaneous unmonitored sources is minor relative to the three major sources of atmospheric emissions from the FMPC (Figure 10). When all of the miscellaneous sources investigated in Appendix K are combined, using appropriate statistical measures, the grand total of the releases is 16,000 kg (median estimate), with a 5th–95th percentile range of 9,300 to 28,000 kg. This total does not include the November 1960 dust loss from the Pilot Plant, which is included with the total dust collector source term.

RADON AND DECAY PRODUCT RELEASES FROM K-65 SILOS AND MATERIALS

The main source of radon-222 release from the FMPC is material stored in the K-65 silos, which contain residue, called K-65 material, from the extraction of uranium from pitchblende or other uranium ores. Originally, the waste residues from the processing, including the K-65 material, were to be returned to the supplier, the African Metals Corporation. On an "interim" basis, the wastes were stored at processing facilities, where they remain. The K-65 material contains very high concentrations of radium-226, and consequently, is a significant source of radon-222 emissions.

The K-65 material at the FMPC has primarily been stored in large concrete storage tanks, called the K-65 Silos, located in the waste storage area of the site. Figure 2 shows the location of the K-65 Silos, as well as two other waste storage silos. Silo 3, the Metal Oxide Silo, contains the metal oxide waste material, another waste residue from the extraction processing of uranium ores. The metal oxide material is also contaminated with radioactivity, but the concentration of radium-226 is much lower than in the K-65 material. Silo 4 has never been used, and contains only a small quantity of water with very low levels of radioactive and chemical contaminants. The Metal Oxide Silo and Silo 4 are not considered significant sources of radon-222 releases. Belgian Congo uranium ores were also processed at the Mallinckrodt Chemical Works (MCW) facility in St. Louis. Due to insufficient storage capacity at MCW, K-65 material from MCW was shipped to the FMPC, beginning in 1951, before construction of the K-65 Silos was complete. That K-65 material was stored in 55-gallon drums on the storage pad around Plant 1.

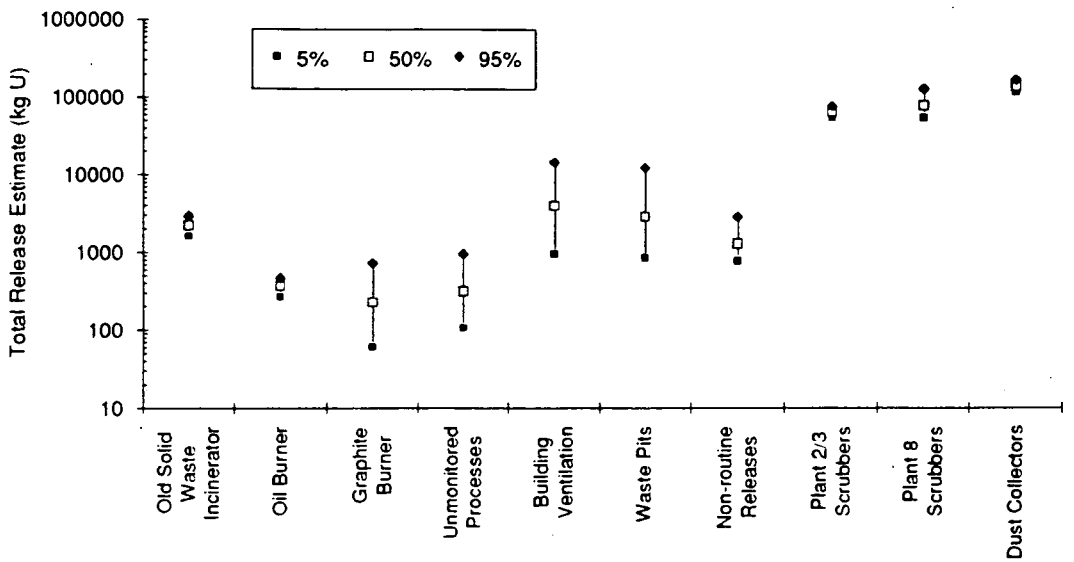

Figure 11a. *Note logarithmic scale.*

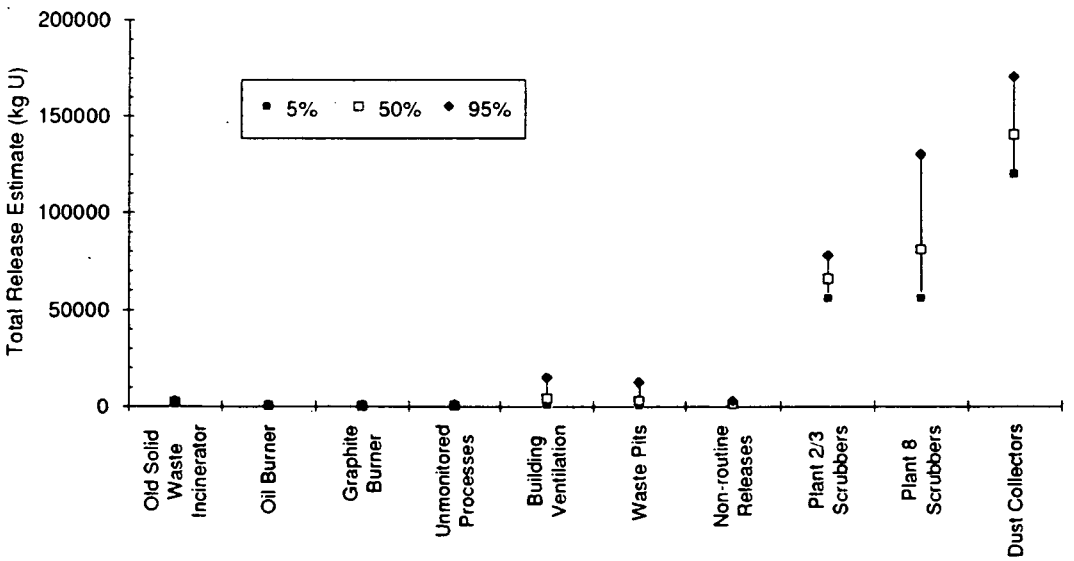

Figure 11b. *Note linear scale.*

Figure 11. Relative importance of miscellaneous unmonitored sources of atmospheric releases of uranium compared with releases through scrubbers and dust collectors. The 50% point represents the median (best estimate). The 5% and 95% points encompass a 90% probability range on the total estimates. Figure 11a is plotted on a logarithmic scale, so that the uncertainty distributions can be seen more clearly, while Figure 11b is plotted using a linear scale, which more accurately illustrates the true relative magnitude of these sources.

Appendix J contains the detailed descriptions of the radon-222 and radon daughter release estimates, including more information about the K-65 and metal oxide materials and storage silos; a summary of previous estimates of radon releases, by others; a discussion of potential radon sources at the FMPC; descriptions of our calculational strategies for current estimates of releases; models and calculated releases for the different time periods assessed; and a discussion of an alternative calculation, for comparison with current estimates. The following sections provide some information about the history of K-65 materials at the FMPC, and our estimates of radon-222 and radon decay releases from the site.

History of K-65 Silos and K-65 Material at the FMPC

The K-65 Silos were constructed in August 1951 through July 1952 for storage of K-65 materials. However, MCW began shipping K-65 material to the FMPC before construction of the FMPC silos was complete. By the end of July 1952, about 13,000 55-gallon drums of K-65 material (equal to about half the capacity of one Silo) had been received at the FMPC. Before disposal in the Silos began, the drummed K-65 material was stored on the concrete ore storage pad around Plant 1, the Sampling Plant, for the period September 1951–mid-June 1953. The K-65 material was added to the Silos from July 1952 through September 1958. We thus calculate radon-222 and radon decay product releases from:

- the K-65 Silos, and
- stored drums of K-65 material on the storage pad near Plant 1 for 1951–1953.

The K-65 Silos have had problems of deterioration, almost since the time of construction. Significant cracking in the walls and seepage of the contents was noted from the 1950s (Wunder 1954; Martin 1957). Because of these problems, repairs and improvements to the Silos occurred from the 1960s through the 1980s. Not all of the changes to the Silos would have had a significant effect on the releases of radon. The most important change, for radon emissions, was the sealing of penetrations of the Silo domes in 1979. This action would have significantly reduced the ventilation of the silo air spaces, and thus also reduced the radon releases from the Silos. The addition of an exterior foam layer on the silo domes in 1987 may have further reduced the emissions of radon. Earthen berms were built around the Silos in 1964. However, at that time the radon releases occurred primarily through openings in the silo domes, so the addition of the berms would not have altered the releases.

Based on these changes to the K-65 Silos and on the operational periods of them, we estimate radon and radon daughter releases from the silos separately for each of the following periods:

- mid-July 1952 to mid-June 1953 (operational period for Silo 1)
- mid-June 1953 to mid-September 1958 (operational period of Silo 2)
- mid-September 1958 to June 1979 (both silos inactive; prior to sealing penetrations),
- July 1979-to December 1987 (both silos inactive; after sealing penetrations), and
- 1988 (1988 is the last year of concern for this project).

Current Estimates of Radon Releases

For some other releases at the FMPC, extensive data sets of direct measurements of release quantities are available. However, for radon and radon decay product releases there are no direct measurements of release quantities. In addition, until the 1980s there were

very few measurements of parameters that can be used indirectly to calculate radon releases. Because of this limited availability of data, we use models to estimate radon release quantities.

The traditional model used to estimate radon releases from radium-226-bearing material, such as uranium mill tailings, involves calculations of the quantity of radon formed in the material, and the subsequent diffusion of the radon through the material to the outside air. For the K-65 materials, measurements have not been made of the radon diffusion coefficient and radon emanation fraction, which are two key parameters in this traditional calculation. Literature values can be obtained for these parameters, but without site-specific values, the uncertainty ranges are extremely large. To reduce the uncertainties in our results, we have used different models, which we believe make the best use of the limited data that are available. Appendix J describes the available, useful information; the information lacking, that would be useful to improve estimates; and the general approach to estimating radon releases. The methods used for 1980–1987 are generally similar to those used in previous release estimates (Borak 1985; IT 1989; Grumski 1987; Boback et al. 1987), though additional data have been obtained and used.

There are no direct data available for estimating releases of radon decay products. Thus, radon decay product releases are calculated to be equal to radon releases multiplied by two correction factors. The first correction factor accounts for the expected ratio of radon decay product concentrations in the silo air to the radon concentration (equilibrium fraction). The second is a fractional release factor, that accounts for deposition of radon decay products along the release path (such as cracks in the silo domes, or penetrations in the domes), which reduces the quantities of decay products released.

As for other releases, we use Monte Carlo methods to perform the calculations of radon and radon decay product releases, so that uncertainties are calculated along with best estimates. The estimated release rates from the K-65 Silos are plotted versus time in Figure 12. The cumulative quantity of radon released from the K-65 Silos for 1959–1979 is larger than for other periods, due to the length of this period and the higher release rate for the period. Releases for this period may also be important in terms of potential doses to offsite people. The predicted radon release rate from the K-65 Silos remained elevated through most of the 1970s, while uranium releases to air generally decreased through the 1970s compared to the 1960s (see Figure 10 and Table 11).

The predicted total quantities of radon released from the FMPC for 1951–1988, are summarized in Table 5. From this summary, it can be seen that radon releases from the drummed K-65 material stored on the Plant 1 pad are relatively insignificant contributors to the total radon releases for the period 1951–1988. However, the radon releases from the drummed K-65 material occurred when operations at the FMPC were just beginning and releases of uranium were relatively small. Consequently, radon releases from the drummed K-65 material may be significant contributors to site-wide releases of all radionuclides from 1951–1953.

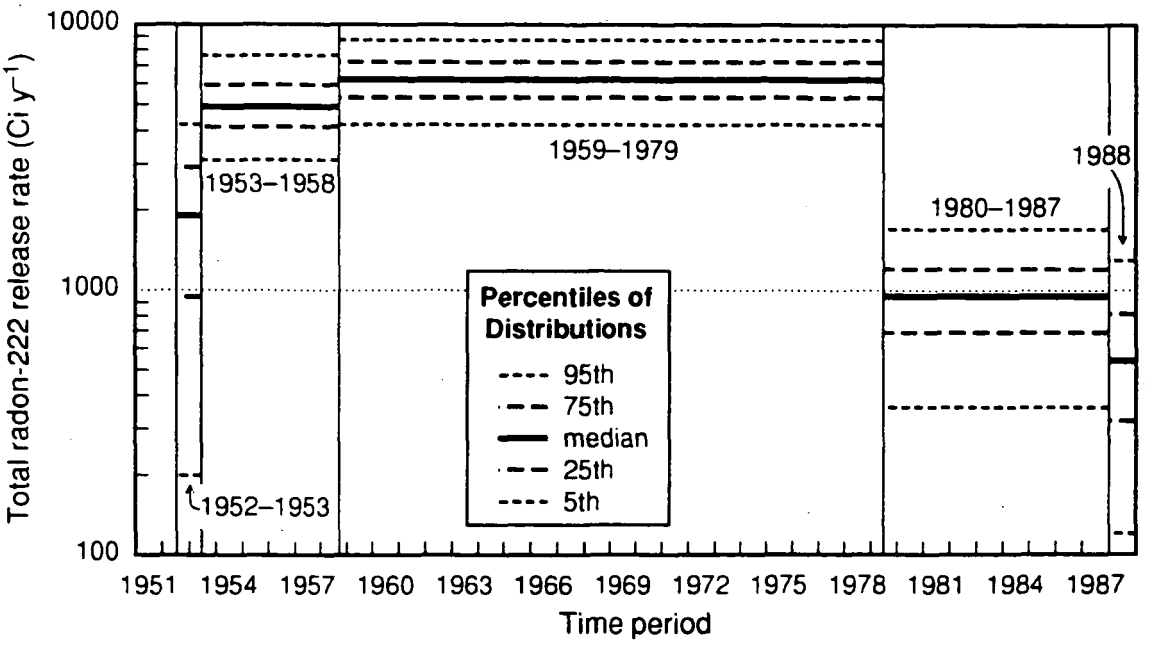

Figure 12. Estimated radon-222 release rates from the K-65 Silos as a function of time. The periods indicated are only the nominal periods; the more precise dates are given in Appendix J.

Table 5. Summary of Predicted Total Radon and Radon Decay Product Release Quantities (Ci) from the FMPC for the Period 1951–1988

	Radon released			Decay products released[a]		
Source of releases	5th	median	95th	5th	median	95th
K-65 Silos	110,000	170,000	230,000	87,000	130,000	190,000
Drummed K-65 material stored on Plant 1 pad	54	720	3,400	4.5	130	880
Both sources	110,000	170,000	230,000	87,000	130,000	190,000

[a] The release quantities for radon-222 decay products are release quantities of each of the short-lived decay products, polonium-218, lead-214, bismuth-214, and polonium-214.

Table 6 presents a comparison of our results with previous estimates of the emissions of radon from the K-65 Silos. The other studies did not report uncertainties associated with the release rate estimates. However, results of the other studies generally fall within, or close to, our 90% probability interval (5th to 95th percentile) of release rates.

Table 6. Comparisons of Current Estimates of Radon Release Rates (Ci y^{-1}) from K-65 Silos to Release Rates from Other Studies

	Percentiles of our estimates			Results of other studies	
Period, release pathway	5th	median	95th	Value	Reference
1980–1987, diffusion	72	130	240	60 [a]	Borak 1985; IT 1989[b]
1980–1987, air exchange	230	810	1600	1023 [a]	IT 1989 [b]
1980–1987, total	360	950	1700	1083 [a]	IT 1989 [b]
1988, total	120	540	1300	1150 [b]	Hamilton et al. 1993

[a] These results were considered by IT (1989b) to apply to the complete period 1953–1984, but we believe that the conditions and parameters used to develop the estimates were only valid for the period July 1979–1987.

[b] This result was the average release rate calculated for 1989–1990. We compare it to our results for 1988 because we believe conditions of the Silos were unchanged for 1988–1991.

We did an alternative calculation of radon releases using more conventional methods. This method estimates radon releases that would exist if the Silo domes did not cover the K-65 material. The results of the alternative method are generally consistent with, but not as satisfactory as the current methodology because of very large uncertainties and the apparent underprediction of the radon releases.

DIRECT EXPOSURES FROM GAMMA RADIATION FROM THE SILOS

Radium-226 and other radionuclides in the materials stored in the K-65 and Metal Oxide Silos produce emissions of gamma radiation, which may have exposed people outside the FMPC. In our Task 4 Report (Killough et al. 1993), we described the methodology to be used to calculate exposures and doses due to this direct radiation. Exposure rates will be calculated using the MicroShield 4 computer software (Negin and Worku 1992). In Appendix J, we provide additional information, necessary to complete the exposure calculations that will be reported in the Task 6 report.

The two K-65 (Silos 1 and 2) and the Metal Oxide (Silo 3) Silos are the only significant sources of direct radiation exposures to people outside the FMPC boundary. This conclusion is based on the results of aerial radiation surveys of the FMPC site and surrounding area, and results of penetrating radiation monitoring performed by the FMPC along the site boundary. Additional information is used for direct exposure calculations, including:

- concentrations of radionuclides in the Silos 1,2 and 3,
- concentrations of radionuclides in the air space of the K-65 Silos,
- densities and moisture content of the materials stored in the Silos, and
- information about the time-history of filling of the K-65 Silos.

LIQUID WASTE DISCHARGES FROM FMPC

Liquid wastes that are generated at the FMPC come from three main sources: process water via the clearwell portion of the waste pit, sanitary sewage, and storm water. Figure 2

shows that liquid effluent streams from FMPC are released to the offsite environment at two locations. These are (1) the combined sewer outfall which discharges through Manhole 175 into the Great Miami River at a point almost directly east of the plant site, about three miles upstream from New Baltimore and (2) the storm sewer outfall which discharges into a branch of Paddy's Run onsite. **Appendix L** provides more detailed descriptions of the principal contributors to liquid discharges from the FMPC and the types of documentation used to tabulate the discharges.

Releases of Uranium in Liquid Effluents from the FMPC

To the Great Miami River. Manhole 175, located on the eastern side of the facility, is the discharge point for waste water leaving the site through the main effluent line to the Great Miami River. It is the final junction point of the major waste effluent streams from the facility. The discharge flow to the Miami River was continuously measured. A composite sample was collected and analyzed for uranium on a daily basis. These daily uranium measurements were found for most years in the 1950s and 1960s. Daily flow rate measurements were located for 1958–1964, and monthly totals were available for later years. When specific information was not located for a particular month, an average value, based on the other months in the same year, was used.

The quantity of uranium released to the river is the product of the uranium concentration multiplied by the flow volume. Sources of uncertainty for these estimates of uranium losses through Manhole 175 to the Great Miami River come primarily from the analytical errors in measuring effluent flow, and in sampling and measuring uranium concentrations in the water.

To Paddy's Run. Runoff water collected in the storm sewer system passed through the storm sewer lift station before release through Manhole 175 to the river. Since the storm sewer lift station was not connected to any process, all the uranium lost through it was assumed to be from leaks and spills (Ross, 1972). When the capacity of the storm sewer lift station was reached, water overflowed through the storm sewer outfall to Paddy's Run. The volume of storm water that overflowed the storm sewer lift station was related to rainfall amounts and patterns.

Estimates of uranium losses from the storm sewer outfall to Paddy's Run were based upon analytical data sheets and monthly reports which listed the individual outfall events occurring during that month. There are three major components of uncertainty associated with estimation of uranium losses to Paddy's Run:

- the analytical errors associated with determining uranium concentration and water flow before discharge to Paddy's Run.
- time periods when rainfall, and consequently runoff, were quite high and the capacity of the storm sewer lift station flow meter and v-notch weir at Paddy's Run was exceeded.
- unmeasured losses from the site above the point where the storm sewer outfall enters Paddy's Run (where the measured losses were recorded).

Figure 13 shows the annual uranium release estimates to the Great Miami River and to Paddy's Run for all years. The magnitude of the uranium releases to the river peaked in 1961 with 7300 ± 140 kg uranium. From 1974 onward, the annual releases were below 1000

kg. The uranium losses to Paddy's Run show much more month-to-month variation than do the uranium losses to Manhole 175 (MH 175). However, the average quantity of 500 kg uranium discharged through Manhole 175 to the Great Miami River each month during the early 1960s was roughly five times greater than the average quantity of 100 kg of uranium lost to Paddy's Run during that same time.

Other Radionuclides Released in Liquid Effluents

Release estimates for thorium, radium-226, radium-228, and fission and activation products are based on correlations between the total annual releases of uranium and those of the other radionuclides. These ratios of releases, computed for years when measurements were made, provide a basis for estimating the release of the other radionuclides for years when they were not measured. This methodology is described in **Appendix D** in the present report, and in Appendix C of Task 4 (Killough et al. 1993). Ratios of the annual average activity of a radionuclide (or quantity of thorium) to the annual uranium quantity were calculated for years when data were available. The measured concentrations at MH 175 reported in analytical data sheets were used to calculate the ratio for some years. Annual average concentrations of radium, thorium and the fission and activation products in liquid effluents were reported by the FMPC in historic release reports (Boback et al. 1987), and in annual environmental monitoring reports beginning in 1976. The variability of the release ratio from year to year was considered in deriving the uncertainty associated with the estimated releases of these other radionuclides. The release estimates and uncertainty analysis were computed using Monte Carlo techniques in the Crystal Ball™ program (Decisioneering 1993).

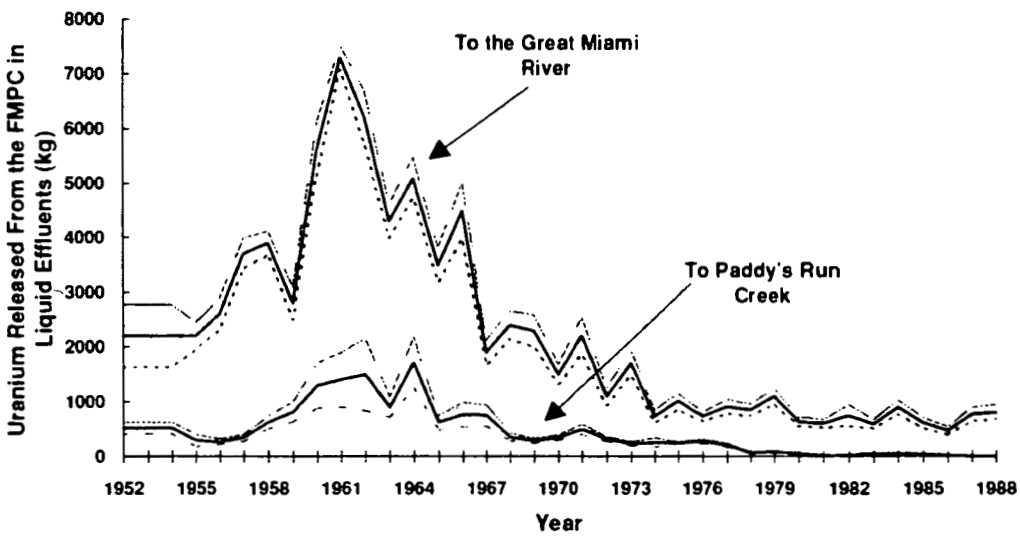

Figure 13. Uranium losses to the Great Miami River via Manhole 175 and to Paddy's Run from the FMPC from 1952–1988. The uncertainty of each estimate is described by the 95th percentile (top, broken line), and the 5th percentile (lower, dotted line).

Table 7 summarizes our estimates for releases of materials in liquid effluents from the FMPC for all years of operation. Our best estimate of uranium released to the Great Miami River for all years is 82,000 kg. The 5th to 95th percentile uncertainty range is 71,000 to 94,000 kg of uranium. Some estimates of uranium in liquid wastes have been made by others on an annual basis (Boback 1971), or in summary reports evaluating the past discharge history of the facility (Rathgens 1974, Boback et al., 1985). These estimates of uranium to surface water from 1951 through 1984 range from 74,000 to 77,000 kg (Boback et al. 1987, Galper 1988) and fall within the uncertainty range of our estimates. Revisions to historic discharge reports generally focused on amending estimates of uranium loss to airborne effluents, and did not include updated figures for liquid effluents (Boback et al. 1985, Boback et al. 1987).

The total release estimate for uranium to Paddy's Run via the storm sewer outfall ditch and runoff is 17,000 kg of uranium. The 5th to 95th percentile uncertainty range is 14,000 to 20,000 kg of uranium. Losses to Paddy's Run show much more month to month variation than do the uranium loss estimates to the Great Miami River. The highest annual releases of uranium occurred from 1960 to 1964, when the average quantity of uranium discharged through MH 175 to the river was approximately 500 kg each month, about 3 to 4 times greater than the average quantity of uranium lost to Paddy's Run each month.

Table 7. Summary of Total Estimates of Radioactive Materials Released From the FMPC in Liquid Effluents For All Years of Operation

Material Released to Great Miami River	Median Value	Uncertainty Range (5th %ile to 95th %ile)
	Quantity (kg)	Quantity (kg)
Uranium	82,000	71,000 to 94,000
Uranium (To Paddy's Run)	17,000	14,000 to 20,000
Thorium	5,800	3800 to 9400
	Activity (Ci)	Activity (Ci)
Radium-228	2.7	0.33 to 20
Radium226	18	15 to 22
Plutonium 239,240	0.0088	0.0019 to 0.033
Plutonium-238	0.00028	0.00016 to 0.0034
Neptunium-237	0.0044	0.0011 to 0.018
Cesium-137	0.54	0.14 to 1.9
Ruthenium-106	0.056	0.014 to 0.22
Technetium-99	300	110 to 800
Strontium-90	6.0	1.5 to 24

The other materials released at various times over the years include decay, fission and activation products of uranium, thorium and recycled uranium. Recycled uranium was not processed until late 1962, so releases of fission and activation products began at that time. Releases of thorium, and one of its decay products, radium-228, occurred when thorium was processed at the site in 1954–1957, and 1964–1988. Releases of radium-226 occurred

throughout the history of the site; and the total release is estimated at 18,000 mCi or 18 Ci, with an uncertainty range of 15 to 22 Ci. These values will be used to calculate radiation doses to the population in the vicinity of the FMPC in our final task report.

The chemical form of uranium in liquid effluents is not known with certainty, but several uranium species of both the +4 and +6 oxidation states may have been present in solution in liquid waste streams during this period. The ratios of these various ionic species in the process waste streams, in Paddy's Run, or in the main effluent pipeline to the river, would be a function of the pH of the water. The presence of suspended solids in the liquid wastes is considered in assessing the relative solubility of uranium in liquid releases. Daily measurements of total suspended solids (TSS) were made on 24-hour composite effluent samples at MH 175 beginning in 1956 (NLCO 1956). Among the suspended solids may have been very small particulates of the insoluble U_3O_8 and UO_2. Not all the suspended solids measured on a daily basis were uranium, but the average monthly values may provide an upper bound, or conservative estimate, for the amount of insoluble uranium that was released in liquid effluent. Furthermore, some uranium-containing suspended solids that were released into the waste streams might have dissolved during dilution downstream from the FMPC.

URANIUM CONTAMINATION IN GROUNDWATER OUTSIDE THE FMPC

Contamination of the groundwater could occur either by direct discharge of waste waters to it or by infiltration of contaminated water through the soil. No evidence of direct discharges to the groundwater from the facility has been found in review of historic documents. Concern about the infiltration pathway has been evident in FMPC documents since the late 1950s, and a variety of studies and analyses have been conducted from that time to the present day (Eye 1961, Dove and Norris 1951, Hartsock 1960, Spieker and Norris 1962). Recent reports describe the measured contamination levels in groundwater, primarily to the south and southwest of the FMPC that have resulted from infiltration of water bearing uranium to the aquifer (GeoTrans 1985, ASI-IT 1990). Uranium contamination of groundwater outside the FMPC has been known since late 1981, when the first samples of water from private wells were analyzed. The significant offsite uranium contamination in groundwater is south of the site, and is now called the "South Plume." There are additional known areas of groundwater contamination on the FMPC site, but only the South Plume area extends outside the site boundary. Since this dose reconstruction project is concerned with past doses to people around the site, the groundwater contamination under consideration here is limited to the South Plume. Figure 14 shows the estimated area of the South Plume contamination, as of 1991. Also shown are the locations of the private wells sampled by the FMPC monitoring program.

In our Task 4 report (Killough et al. 1993), we examined the potential importance of the groundwater contamination for doses to people around the FMPC. It was shown that only three of the private wells monitored, numbers 12, 15, and 17, have had measured uranium concentrations above the range of background. Although well 26 is within the area of groundwater contamination, it is installed deeper in the aquifer, and the uranium concentrations are at background levels. We concluded that because of the limited area of the South Plume, only a small number of people would have potentially received radiation

doses from contaminated groundwater. Toward the main objective of this project, the determination of the feasibility of an epidemiological study, doses to these people would be less significant to the collective population dose than doses through other pathways. For this reason, we further concluded that a detailed assessment of the groundwater transport of radionuclides, and detailed assessments of doses to individuals potentially exposed through groundwater pathways, are not warranted. For other project objectives, it is still important to estimate potential doses through the groundwater pathway, so instead we use simple methods to estimate concentrations of uranium in the three contaminated wells. **Appendix M** contains details of our groundwater assessments.

Potential Sources of Groundwater Contamination

The status of groundwater contamination in the vicinity of the FMPC has been investigated. Appendix M describes a special study that was conducted to determine the primary transport pathway for uranium deposited on soil around the FMPC. The study compared uranium migration due to infiltration, surface soil erosion, and surface water runoff. Results of the study show that uranium deposited on soils is primarily transported by infiltration and that soil erosion transports the least amount of uranium. There are two potential sources of groundwater contamination originating on the FMPC site (see Figure 14): (1) historical releases of uranium-contaminated water to Paddy's Run and to the Storm Sewer Outfall Ditch (SSOD), and (2) possible releases from the solid and liquid waste pits in the waste storage area.

Of these two potential sources, the principal source of uranium contamination in the South Plume has been determined to be the historical releases to Paddy's Run and the SSOD (DOE 1990). The bottom sediments of Paddy's Run and the SSOD are very permeable in the area north and west of the South Plume, so these areas are recharge areas for the regional aquifer. Thus, uranium contamination in Paddy's Run and the SSOD percolates downward through the permeable sediments to ultimately reach the groundwater.

Estimated Uranium Concentrations in Private Wells

A preliminary investigation of the movement of contaminated groundwater was performed, to determine the transport times required for uranium contamination to move from the source (waters in Paddy's Run and the SSOD) to offsite locations. The study is described more fully in Appendix M. Based on results of this preliminary assessment, we concluded that the South Plume would not have reached the offsite private wells in the South Plume area until after 1962. Thus, exposures of people using wells in the South Plume might have occurred from 1963 onward.

Monitoring of the three contaminated wells (wells 12, 15 and 17) was initiated in late 1981. Routine monitoring of these wells, as well as other private wells, has been performed by the FMPC since 1982. We obtained results of monthly measurements of uranium concentration in well water for the three contaminated wells for late 1981 through 1992. Annual average uranium concentrations are shown in Table 8. The annual average concentrations for 1982–1988 will be used as the basis of dosimetry calculations for these years.

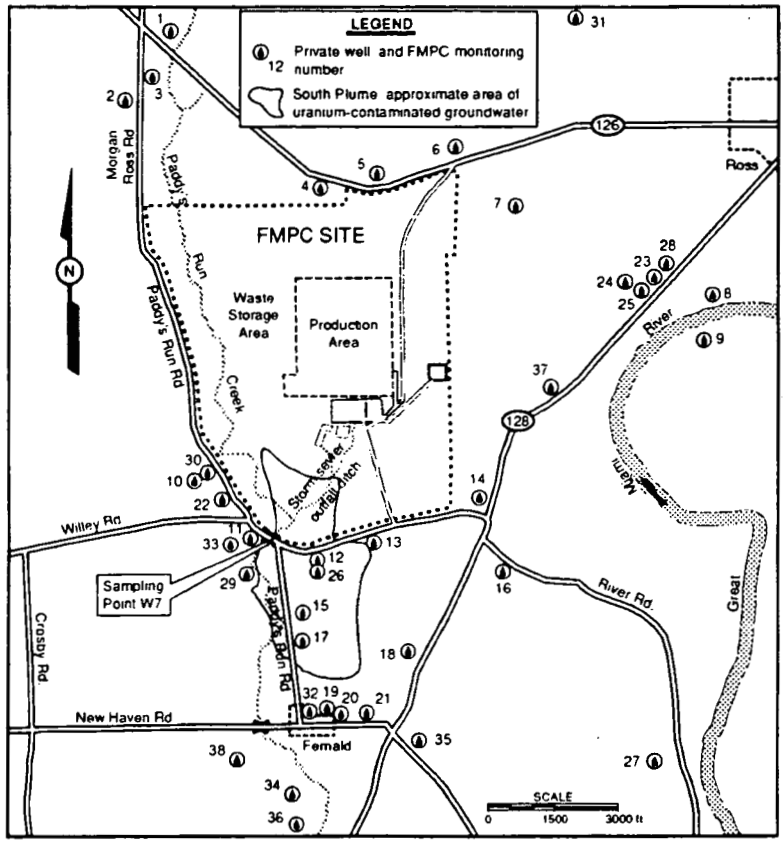

Figure 14. Approximate area of uranium contamination in the South Plume at the end of 1991, and locations of the private wells around the FMPC sampled in the FMPC routine monitoring program. Sampling point W7 is a location for sampling the surface water in Paddy's Run, at the Willey Road bridge.

For the period 1963–1981, for which well monitoring was not performed, we used models to estimate concentrations of uranium that might have existed in well water of the South Plume. We first developed an estimated upper bound on the annual average uranium concentration that could have existed in wells 12, 15, and 17. As mentioned above, the primary source of uranium contamination of the South Plume has been determined to be uranium-bearing waters released into Paddy's Run and the SSOD. Thus, uranium concentrations in the groundwater are expected to be at the most, equal to concentrations in Paddy's Run and the SSOD. Uranium concentration data for Paddy's Run and the SSOD were obtained and compiled in Appendices L and M. Uranium concentrations were higher in the SSOD than in Paddy's Run. In the SSOD, the maximum concentration of uranium was 8,300 pCi L^{-1}, for the year 1960. Thus, this value is used as the upper bound of the annual average uranium concentration that might have existed in the contaminated wells during 1963–1981.

We recognize that this upper bound is an extremely conservative estimate (that is, the estimated value is too high) of the uranium concentrations in the three contaminated wells

for 1963–1981. The conservatism results because: (1) the maximum annual average concentration was used to represent the concentrations for the complete period, (2) dilution of the uranium with water from Paddy's Run (with lower concentrations than that of the SSOD) was ignored, and (3) dilution in the groundwater (from other groundwater sources) was also ignored. For the dosimetry calculations, we believe the use of the upper bound uranium concentration of 8,300 pCi L^{-1}, to represent concentrations in private wells of the South Plume area for 1963–1981, is unrealistically conservative.

Table 8. Annual Average Concentrations of Uranium (pCi L^{-1}) in the Three Contaminated Wells[a]

Year	Well 12	Well 15	Well 17
1982	170	320	45
1983	180	290	39
1984	170	220	36
1985	140	200	31
1986	150	190	31
1987	200	200	40
1988	170	190	38
1989	170	190	27
1990	130	180	30
1991	100	170	27
1992	100	150	25

[a] The range of long-term average, background concentrations of total uranium in private well water around the FMPC is 0.09 to 1.3 pCi L^{-1} (Shleien et al. 1994).

Thus, we developed an empirical model to estimate uranium concentrations in the contaminated wells. An empirical model is one based primarily on measurement data, rather than on theory, to explain the particular conditions. In this case, the data we used are the annual average measured uranium concentrations in the contaminated wells for 1982–1992, and the calculated quantities of uranium released to Paddy's Run and the SSOD for 1952–1988 (these releases are discussed in Appendix L). Details of the model are described in Appendix M. We think that the use of this model provides more realistic, though still somewhat conservative, estimates of uranium concentrations that might have existed in the contaminated wells for 1963–1981.

Table 9 summarizes the uranium concentrations in well water from the South Plume, that will be used for the dosimetry calculations (Task 6). The values for 1963–1981 are based on the empirical model. Based on the empirical model calculations, it is likely that uranium contamination in the groundwater would not have reached the offsite wells prior to 1968 (estimated concentrations are zero prior to 1968). The values for 1982–1988 are the annual averages based on measurements for well 15. Concentrations from well 15 are used in this assessment because they are the highest concentrations of the three contaminated wells.

Table 9. Values of Uranium Concentration (pCi L^{-1}) Used to Represent Annual Average Concentrations in Contaminated Wells of the South Plume Area

Year	Concentration	Year	Concentration	Year	Concentration
1951–1967[a]	0	1975	490	1983	290
1968	180	1976	580	1984	220
1969	230	1977	620	1985	200
1970	230	1978	620	1986	190
1971	230	1979	570	1987	200
1972	240	1980	510	1988	190
1973	290	1981	460		
1974	370	1982	320		

[a] The concentration listed is applied to each year in this range.

TASK 2 AND 3 SUMMARY AND CONCLUSIONS

The purpose of the Fernald Dosimetry Reconstruction Project is to estimate doses to the public who lived near the Feed Materials Production Center near Fernald, Ohio from the radionuclides released to the environment during operation of the facility. This report describes our best estimates of releases to the atmosphere and to surface water from FMPC operations, and from the K-65 Silos, during the period 1951–1988. Table 10 provides a summary of our best estimates these results.

Figure 15 shows the relative contributions of uranium released from the major sources at the FMPC facilities during the period. These major sources are uranium released to the atmosphere, uranium released in liquid effluents, and releases of radon gas and its decay products. They are shown in three main sections separated by vertical lines. Numerical values of the best estimate of release are shown next to the heavy bars that represent them. The methods used to determine these release estimates are described carefully and fully in the accompanying appendices.

It should be noted that uncertainties associated with the parameters used to determine these values vary considerably. In some cases, detailed measurements had been made and were located. An example is the uranium discharged in liquid effluent to the Great Miami River. In other cases, however, measurements of uranium losses were not made, and current release estimates are based on other information (for example, the Plant 8 scrubber releases). The median release estimates do not stand alone. The statistical parameters reported with these values in the appendices are an integral part of the release estimates; they should always be reported with them. The table and figure include ranges of estimates as well as the *best* estimates to provide a general comparative overview of annual release estimates for these years.

For the operational period of the FMPC, the total releases from atmospheric sources (dust collectors, Plant 2/3 scrubbers, Plant 8 scrubbers and miscellaneous sources) are 310,000 kg uranium, with the 5th to 95th percentile range of 270,000 to 360,000 kg. The predicted total quantities of radon and radon decay products released from the FMPC through 1988 are 170,000 Ci (5th to 95th percentile range of 110,000 to 230,000 Ci), and

130,000 Ci (5th to 95th percentile range of 87,000 to 190,000 Ci). For releases of uranium in liquid effluents, the median release estimate to the Great Miami River during this time period, is 82,000 kg (5th to 95th percentile range of 71,000 to 94,000 kg), while that to Paddy's Run is 17,000 kg, with the 5th to 95th percentile range of 14,000 to 20,000 kg.

It is important to realize that median estimates of releases from various sources may *not* be directly added to obtain a corresponding median estimate of the annual total release for all sources. The reason is that the medians do not have the additive properties that are familiar to most people from dealing with (arithmetic) means. We have chosen to use median estimates because they represent the 50th percentile of their distributions. For nonsymmetric distributions such as those encountered in this work (principally lognormal or approximately so), the mean is larger than the median by an amount that increases with the weight of extremely large values. For this reason, the median is considered a more stable measure of the central tendency of the distribution, and it is generally used in this study to represent best estimates of uncertain quantities.

Table 10. Summary of Median Uranium and Radon Release Estimates From the FMPC for 1951–1988 With Uncertainty Bounds[a]

Source	Median release estimate	5th percentile	95th percentile
U to Atmosphere			
Dust Collectors	140,000	120,000	170,000
Plant 2/3 Scrubbers	66,000	56,000	78,000
Plant 8 Scrubbers	81,000	56,000	130,000
Miscellaneous Sources[b]	16,000	9,300	28,000
Total: airborne sources	310,000	270,000	360,000
U to Surface Water			
To the Great Miami River	82,000	71,000	94,000
To Paddy's Run	17,000	14,000	20,000
Total: surface water	99,000	85,000	120,000
Radon to Atmosphere			
K-65 Silos			
Radon-222	170,000 Ci	110,000 Ci	230,000 Ci
Radon-222 decay products[c]	130,000 Ci	87,000 Ci	190,000 Ci

[a] Values are in kg of uranium, except for releases from the K-65 silos which are reported in units of activity, called curie, Ci. Median estimates of releases from the various sources cannot be directly added to obtain a corresponding total median release estimate for all sources because medians do not have the additive properties that are associated with arithmetic means. See discussion on uncertainty in release estimates on page 12.

[b] These estimates do not include the November 1960 release from the Pilot Plant which is included in the dust collector releases.

[c] The release quantities for decay products are quantities of each of the short-lived decay products, polonium-218, lead-214, bismuth-214, and polonium-214.

Our work strongly supports the conclusion that atmospheric releases account for the greatest fraction of uranium released from the FMPC facility. Table 11 summarizes the grand medians and percentile values for the releases by decade for the three primary sources—the dust collectors, the Plant 8 scrubbers, and the Plant 2/3 scrubbers. The total

releases estimate for 1951–1084 is a summary for all release points, including the unmonitored and accidental releases. The unmonitored releases are relatively minor compared to the three major sources, contributing only 16,000 kg uranium over the 47-year time span (Figure 15). The uranium releases to the atmosphere were highest in the 1950s with 180,000 kg uranium released from the three primary sources, and declined to almost half that in the 1960s. Total release estimates for the 1970s and 1980s are significantly less at 30,000 and 4,400 kg, respectively.

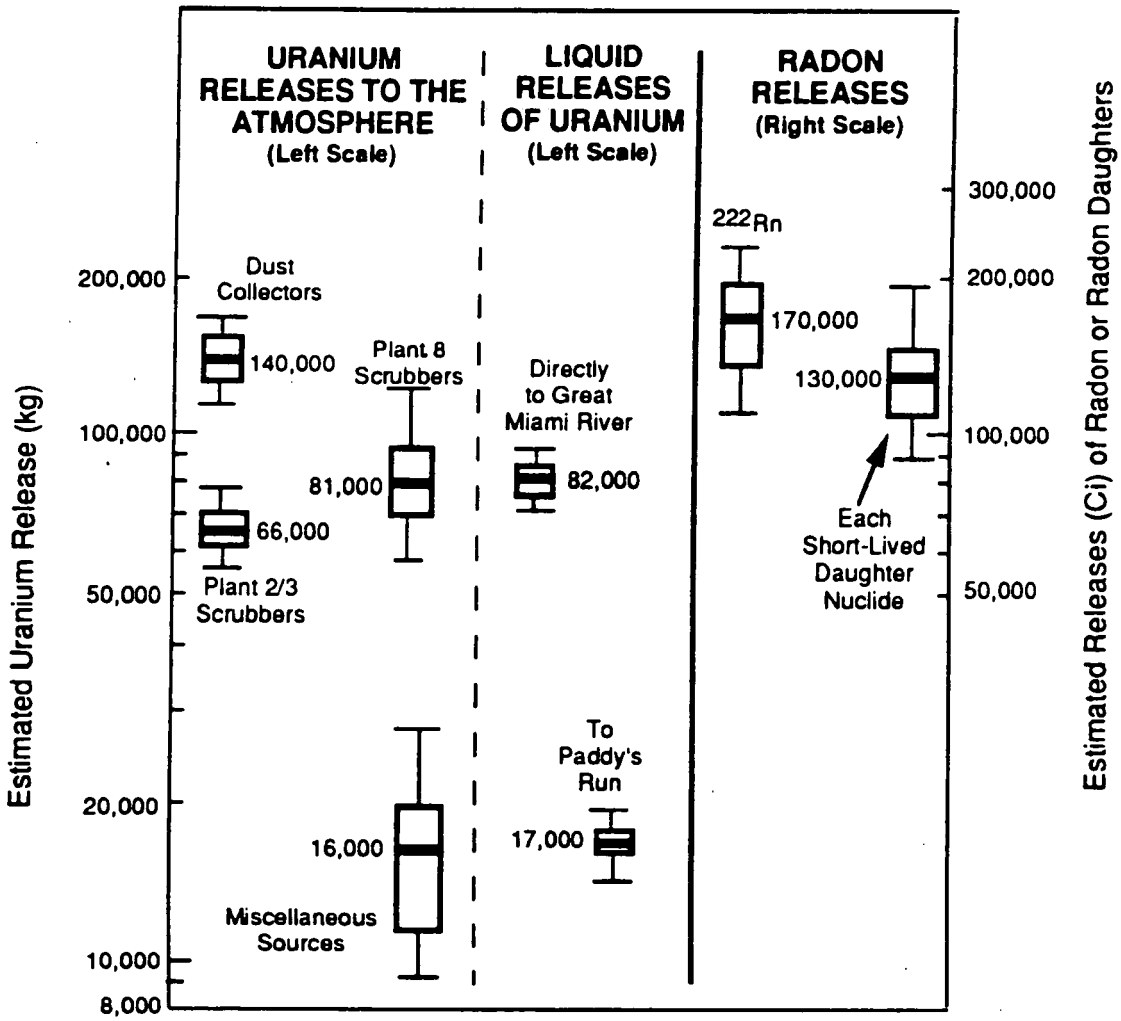

Figure 15. Summary of release estimates from the FMPC for the years 1951–1988. Releases are divided into three main sections which are separated by vertical lines. The center square represents the median or *best* estimate. The dark square on tope represents the 95th percentile value, while the lower diamond represents the 5th percentile value. Ninety percent of the estimates lie within the range defined by top and bottom values that surround the best estimate.

Table 11. Summary of Uranium Release Estimates for the Airborne Sources

Period	Best Estimate (kg U)[a]
1950s	175,000
1960s	90,000
1970s	30,000
1980s	4,400

Period	Best Estimate (kg U)[b]	Other percentiles in distribution of release estimate (kg U)			
		5th	25th	75th	95th
1951–1988	310,000	270,000	290,000	340,000	360,000

[a] Releases by decade are releases from the dust collectors, the Plant 8 scrubbers and the Plant 2/3 denitration processes.
[b] Total releases with best or 50th, 5th, 25th, 75th and 95th percentile values, represent releases from dust collectors, Plant 8 scrubbers, Plant 2/3 denitration processes and the miscellaneous sources.

There have been several previous attempts at determination of uranium releases from the FMPC. Estimates of uranium discharged in liquid effluent were have been made by others on an annual basis (Boback 1971), or in summary reports evaluating the past discharge history of the facility (Rathgens 1974, Boback et al., 1985). These estimates of uranium to surface water from 1951 through 1984 range from 74,000 to 77,000 kg (Boback et al. 1987, Galper 1988) and fall within the uncertainty range of our estimates. Revisions to historic discharge reports generally focused on amending estimates of uranium loss to airborne effluents, and did not include updated figures for liquid effluents (Boback et al. 1985, Boback et al. 1987).

Table 12. Summary of Previous Atmospheric Uranium Release Estimates

Years (inclusive)	Uranium (kg)	Reference
1953–1984	96,000	Data for EPA estimate[a]
1951–1985	135,000	FMPC-2082 report[b]
1951–1987	179,000	Addendum to FMPC-2082 Report; IT report[c]
1951–1985	390,000	Reports prepared by Institute for Energy and Environmental Research for litigation involving the US DOE[d]

[a] From Kennedy 1985 and Meyers, no date; no specific documentation for source term estimate is provided.
[b] From Boback et al. 1985; this report estimated airborne uranium releases from plant operations only.
[c] From Clark et al. 1989 and IT 1989; the addendum included other sources of uranium releases from Plant 2/3 scrubber operations, unmonitored releases and accidental releases. The IT report employed the source term from the Addendum to the 2082 report.
[d] From Makhijani and Franke 1989; this estimate from their "alternative #2" calculations included additional scrubber losses from Plant 8 based on 70% efficiency for scrubbers instead of 85%.

Previous reports of airborne uranium releases which have been used to estimate radiation doses in the offsite population around the FMPC have been reviewed for this project (Shleien 1991). Table 12 summarizes estimates of atmospheric releases of uranium which have been presented by others previously. These previous studies to determine the releases of radionuclides from the FMPC have yielded source terms which are less than our median or best estimates described in the present report. Our uncertainty ranges do not encompass these estimates except for that of the IEER. Exhaustive comparisons have not been made; however, reasons for our higher estimates include:

- the time to examine numerous documents, in particular original records, related to the FMPC operations;
- the use of a distribution of scrubber efficiencies for Plant 8 scrubbers;
- accounting for uranium losses from miscellaneous unmonitored sources and accidents;
- accounting for biases from sample line losses and other sampling deviations in the calculation of dust collector losses.

Our methodology represents a significant improvement in the state-of-the-art of source terms analysis over previously reported data. It involves estimating a median, or best estimate of the releases in addition to a formal uncertainty analysis of parameters associated with these estimates. The Monte Carlo procedure uses our best estimates of the distributions of parameter values to produce a distribution of results. This process has resulted in obtaining a distribution of release estimates, instead of determining a single point estimate of the various parameters, with a single result. As a result, the source term has been characterized by a distribution of uncertainty for each year's releases.

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