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# THE FERNALD DOSIMETRY RECONSTRUCTION PROJECT

## Task 5. Review of Historic Data and Assessments for the FMPC

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

by

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### FINAL REPORT

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### Task 5

### **Review of Historic Data and Assessments for the FMPC**

Radiological Assessments Corporation Route 2, Box 122 Neeses, SC 29107

RAC Report No. CDC-4

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### FINAL REPORT

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### LIST OF ACRONYMS

Activity median aerodynamic diameter (AMAD) Air monitoring stations (AMS) Analytical Data Sheets (ADSs) Bioaccumulation factor (BF) Boundary air monitoring stations (BS) Centers for Disease Control and Prevention (CDC) Concentration Ratio (CR) Curies (Ci) Environmental Analysis Laboratories (EAL) Environmental Protection Agency (EPA) Feed Materials Production Center (FMPC) Fernald Environmental Management Project (FEMP) Geometric mean (GM) Geometric Standard Deviations (GSD) Industrial Hygiene and Radiation (IH&R) International Commission on Radiological Protection (ICRP) International Technology Corporation (IT) International units (SI) Mass median aerodynamic diameter (MMAD) Maximum allowable concentrations (MAC) Monthly dilution (MD) National Council on Radiation Protection and Measurements (NCRP) National Lead Company of Ohio (NLO) National Uranium Resource Evaluation (NURE) Northern Kentucky Environmental Services (NKES) Ohio Department of Health (ODH) Ohio State Plane (OSP) Old solid waste incinerator (OSWI) Parts per billion (ppb) Passive Environmental Radon Monitors (PERMs) Portsmouth Gaseous Diffusion Plant (PGDP) Predicted-to-observed (P/O) Ratio Quality assurance (QA) Radiological Assessments Corporation (RAC) Radon dispersion model (RNCHIQ4) Storm sewer outfall ditch (SSOD) Thermoluminescent dosimeter (TLD) U.S. Department of Energy (DOE) U.S. Geological Survey (USGS) United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR) Uranium (U) Westinghouse Materials Company of Ohio (WMCO)



### EXECUTIVE SUMMARY

The purpose of the Fernald Dosimetry Reconstruction Project is to estimate radiation doses to members of the public who lived near the Feed Materials Production Center (FMPC) from radioactive materials released to the environment during the operation of the facility (1951 to 1988). The goal of Task 5 of the project is to use available environmental measurement data from the FMPC area to verify and validate, to the greatest extent possible, the environmental transport methods developed in Task 4.

Overall, the Task 5 report: 1) provides a basis against which environmental data gathered around the FMPC can be compared (i.e. the radiation background); 2) documents long-term data sets of radionuclides in the environment around the FMPC for model validation (comparison of model predictions to independent field measurements); 3) performs quality assurance on basic particle-size information for airborne effluents; 4) compares the Radiological Assessments Corporation (RAC) environmental transport models to other models; 5) compares environmental radon and exposure rate measurements around the FMPC to the RAC model predictions; and 6) examines data for radionuclides other than uranium and radon, which may be important in the Project. The information in the Task 5 report is divided into this executive summary; a 27-page main text, which contains an introduction and overview of Task 5, as well as a summary of the Appendices and major findings; and the Appendices themselves, which present the review of historic data and detailed assessments.

One strength of the Fernald Dosimetry Reconstruction Project is the examination of historic records of many types, all of which contribute to our understanding of historic releases and their impacts. Since dose reconstruction involves putting together a complete picture of past operations, the environmental monitoring records, both past and present, provide important verification that our estimates of environmental releases and transport are reliable. Our research team has sought and compiled data from the most fundamental sources available (e.g. the original analytical data sheets, log books, first-level notes or memos) to do our analyses, and this is reflected in the Task 5 report as well as throughout the Fernald Dosimetry Reconstruction Project. Monitoring data generated by onsite contractors as well as offsite sources such as state agencies and universities were investigated.

The environmental data reviewed cover monitoring of uranium, radon, and other radionuclides in environmental samples, including:

- Gummed-film (a measure of deposition)
- Air
- Precipitation
- Soil
- Milk
- Vegetation
- Surface waters and cisterns
- Sediment and fish
- Groundwater

In addition, measurements of penetrating radiation around the K-65 Silos are compared to predicted levels using model calculations. For comparative purposes, estimates of radioactivity background concentrations for the FMPC area were derived. These background concentrations represent those which occur naturally in the environment, and provide perspective to measurements made in areas affected by FMPC releases.

Other sections of the Task 5 report discuss limited model verification exercises, in which the results of our models for environmental transport by atmospheric and liquid pathways are compared with results of other models. These comparisons generally indicate good agreement. In addition, measurements of radionuclides other than uranium in airborne and liquid effluents from the FMPC are reviewed. These radionuclides, including <sup>226</sup>Ra, thorium, <sup>99</sup>Tc, <sup>137</sup>Cs, <sup>90</sup>Sr, and <sup>239,240</sup>Pu, may not have contributed greatly to the radiation dose around the FMPC, but could be of interest to the reader. Finally, a quality assurance check was done on original particle size measurements from airborne effluents, and final particle size distributions used in the Project were derived.

Long-term model validations of uranium releases can not yet be presented in this report, because the final analysis of reconstructed source terms (quantity of material released from the site) for the FMPC is in progress. Shorter-term model validations were included in the Task 4 methodology report for the 1960–1962 period, for which a detailed reconstructed source term was available. The model validations for the longer time period will be included in the final Task 6 report of the Project.

In summary, the Task 5 report has provided an analysis of the types and quality of environmental data from the FMPC area. Although the environmental monitoring data are important to consider in developing methods for dose reconstruction, they are not complete enough, either temporally or spatially, to rely on exclusively for assessment of the exposure to surrounding populations from FMPC effluents. Rather, these data are used primarily to provide a quality check of the source term estimates and to validate the transport models.

The quality of data from more recent years is of higher quality than in the past. All the data have been extremely useful in providing the proof for our source term calculations, and model verification exercises. These data and assessments in Task 5 can be generally viewed as secondary sources of information for the dose reconstruction, as opposed to the primary sources which establish the amounts and characteristics of radionuclides released to the environment. However, they provide another important piece of information which lends support to the source term quantities and modeling methods.

### TASK 5

### **REVIEW OF HISTORIC DATA AND ASSESSMENTS FOR THE FMPC**

#### INTRODUCTION AND OVERVIEW

The Feed Materials Production Center (FMPC) near Fernald, Ohio, is a facility whose purpose was to convert uranium ore concentrates and materials recycled from other stages of nuclear weapons production to either uranium oxides or ingots of uranium metal that could be machined and extruded for production reactor fuel cores and target elements. Since operations began in 1951, uranium, uranium decay products, and other radionuclides have been released to the environment as part of routine operations and during unplanned, accidental occurrences. The location of the FMPC with respect to the surrounding area is shown in Figure 1.

The purposes of the Fernald Dosimetry Reconstruction Project for the Centers for Disease Control and Prevention (CDC) are to provide an independent analysis of the types and amounts of radioactive materials released to the environment from the FMPC and to establish a methodology that can be used to estimate doses to persons living near the facility. This methodology will also be suitable for use in an epidemiological study, if such a study is undertaken. The project has been divided into seven tasks. Task 1 dealt with the identification of release points at the FMPC site (RAC 1991). The goals of Tasks 2 and 3 are to determine the radionuclide source terms (that is, the amounts of radionuclides released to the environment) and the uncertainties associated with these historic releases. An interim Task 2/3 report for the operating years 1960 through 1962 was released in December 1991 (Voillequé et al. 1991). That report outlines the methods which would be used for reconstructing source terms for the entire operating history of the FMPC. A comprehensive source term report was issued in November 1993 (Voillequé et al. 1993); that report is currently being revised based on reviewer comments. The goal of Task 4 was to develop methods to describe the environmental transport of the released materials and how people may have been exposed to those materials (Killough et al. 1993).

The original goal of Task 5 was to VERIFY and VALIDATE, to the greatest extent possible, the environmental transport methods developed in Task 4 using available environmental measurement data from the FMPC site. VERIFICATION is the process of showing that a computer-implemented mathematical model is an appropriate description of the conceptual model of the transport process. Model verification can involve exercises such as (a) independently reviewing the model structure and basic equations of the model, (b) assuring that the mathematical equations employed in the computer code are correct and that the code properly implements those equations in the calculational procedure, and (c) checking the results of the computer calculations against "hand calculations" or the predictions of other models for a standard problem.

VALIDATION is the process of checking the predictions of the model(s) against the real world, in this case the FMPC environment. Validation will help assure that the transport models employed in the dose reconstruction adequately represent the physical processes involved in the transport of radionuclides at Fernald and will give added confidence in the results obtained from the computer codes.



Figure 1. Location of the FMPC. The assessment domain is the region around the site with which this study is principally concerned. Doses will be calculated (Task 6) for people within the assessment domain.

The scope of Task 5 has been refined based on the completed Task 4 report (Killough et al. 1993) and on plans for the remaining reports of the project. In order to develop the transport and dosimetry methodology of Task 4, some model validation exercises, mainly for the 1960–1962 period, were performed and were included in the Task 4 report. Many elements of the verification process were also incorporated, in their entirety, into the Task 4 report. In addition, the long-term model validations must be deferred to the final report for the project (Task 6), when the source term estimates and model predictions for the entire FMPC operating history are finalized.

This Task 5 report is now primarily intended to present much of the monitoring data obtained, that will be used for model validation in Task 6 and for source term reconstruction in Tasks 2/3 of the project. Some indirectly related analyses have also been performed,

which will support other aspects of this project; these analyses are logically reported here. The scope thus now includes (1) reporting the majority of the environmental monitoring data obtained, including that for background and regional concentrations of radioactivity in various environmental media, which will be used for validation exercises, source term development, and other purposes, (2) providing the results of a few additional validation exercises performed, (3) reporting the results of two verification activities, which were comparisons of Radiological Assessments Corporation (RAC) models with other transport models, and (4) giving results of other related evaluations, supporting other aspects of the project, including analysis of particle size data for airborne releases, and analysis of data on releases of radionuclides other than uranium.

Table 1 provides a summary of the historic environmental monitoring data and how they are used in the Fernald Dosimetry Reconstruction Project. Some data are useful for some purposes but not for others. For example, only air, surface water, and gummed-film monitoring data are used for verification or identification of possible episodic releases; and only groundwater monitoring data are used directly to compute doses for the affected members of the public. The scope of the historic data summarized in Table 1 includes ambient environmental monitoring data. Effluent monitoring data (e.g. uranium in liquid effluent or in stack samples) are used for source term reconstruction (Task 2/3), but are generally not included in this Task 5 report. Exceptions are the analyses of particle size distributions and other radionuclides in airborne effluents, which are discussed here.

The Task 5 report is divided into this summary and the following Appendices:

- Appendix A Radioactivity Background Around the Feed Materials Production Center
- Appendix B Regional Environmental Monitoring
  - Part 1 Deposition Measurements Using Gummed-Film
  - Part 2 Air Monitoring Data
  - Part 3 Wet Deposition
  - Part 4 Additional Soil Monitoring Data
  - Part 5 Milk Vegetation
  - Part 6 River Sediment Fish
  - Part 7 Groundwater, Cisterns, Ponds, and Pools

Appendix C Particle Size of Airborne Effluents

- Part 1 NKES Study Methodology QA
  - Part 2 Final Particle-Size Distributions
- Appendix D Comparison of the RAC Models with Other Models
  - Part 1 Comparison of Models for Airborne Uranium and Radon
  - Part 2 Surface Water Pathways
- Appendix E Monitoring Data for Radon in Air and Exposure Rate: With Comparisons to Predictions
- Appendix F Other Radionuclides in Airborne and Liquid Effluents

The diversity of Task 5 and the depth to which the data are presented are strengths of this dose reconstruction project. The remainder of this summary provides the reader with a guide to the various Appendices and a summary of the major findings.

Type of Monitoring Data	Dates of Data Examined	Source Term Reconstruction	Identification/ Verification of Episodic Releases	Source Term/ Model Validation <sup>a</sup>	Development of Model Structure and/or Parameters	Direct Dose Assessment
U in Soil <sup>b</sup>	1984–1988	X			x	
U on Gummed-film	19 <b>54–1964</b>		X	X	X	
U in Air <sup>e</sup>	1953-1984		x	<b>X</b> -	Х	
Rn in Air	1978–1 <b>991</b>	X		X		
Radiation from K-65 Silos	1957 and 1976–1990	X		x		
U in Wet Deposition	1961-1967			X	X	
U in Vegetation/Milk	1959–1991 <sup>d</sup>			X	· X	
U in Surface Water <sup>b</sup>	19551988		X	X		
U in Fish	1984–1991				X	
U in Sediment <sup>b</sup>	197 <b>4</b> –1991			X		
U in Groundwater	1981-1990					X

Table 1. Summary of Use of	Primary Types of Historic Environmental
Monitoring Data in the Fe	ernald Dosimetry Reconstruction Project

<sup>a</sup> Implicitly includes validation of source term quantities.

<sup>b</sup> Other radionuclides besides uranium were also examined.

<sup>c</sup> Taken at the FMPC perimeter and beyond.

<sup>d</sup> Lapse in vegetation data 1968–1984.

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## SUMMARY OF APPENDIX A --- RADIOACTIVITY BACKGROUND AROUND THE FEED MATERIALS PRODUCTION CENTER

Appendix A of this report presents information on background concentrations of uranium in air, soil, surface water, rain, and human diets, and of radon in air. Uranium and its decay products are radionuclides which occur naturally in the environment. The concentrations which are normally observed in the environment, without enrichment by man's activities, are referred to as BACKGROUND concentrations. An understanding of the background conditions is important to the dose reconstruction effort for several reasons. First, background concentrations must be known in order to assess the influence of emissions from the FMPC. Secondly, these concentrations can provide some perspective, in terms of risk, to the magnitude of concentrations observed in the environment around the FMPC.

Table 2 summarizes the background concentrations of uranium and radon in environmental media that apply to this project. Natural background concentrations vary globally; therefore, whenever possible, an estimate of background which is appropriate for southern Ohio or the FMPC area was obtained. Also, background concentrations of radon in outdoor air are dependent on season as well as time of day. Details and additional information can be found in Appendix A.

Medium	Contaminant	Concentration <sup>a</sup>	Method for determination
soil	uranium	1–3 pCi g <sup>-1</sup>	Measurements around the FMPC.
soil	uranium	1.5–4 pCi g <sup>-1</sup>	Regional measurements in Ohio.
air	uranium	70–100 aCi m <sup>-3</sup>	Site-specific mass loading calculation, assuming background in soil of 2–3 pCi $g^{-1}$ .
air	uranium	50–140 aCi m <sup>-3</sup>	Environmental Protection Agency (EPA) measurements in Columbus, Ohio.
air	uranium	40–120 aCi m <sup>-3</sup>	FMPC offsite measurements.
rain	uranium	0.03-0.2 pCi L <sup>-1</sup>	EPA measurements in Columbus, Ohio.
surface water	uranium	1–2 pCi L <sup>-1</sup>	Recent FMPC measurements in Great Miami River and Paddy's Run, upstream from FMPC.
drinking water	uranium	0.06 pCi L <sup>-1</sup>	EPA measurements in Cincinnati, Ohio.
groundwater	uranium	0.09–1.3 pCi L <sup>-1</sup>	Measurements in uncontaminated private wells in FMPC area.
air	<sup>222</sup> Rn	0.3 pCi L <sup>-1</sup>	Public Health Service measurements in Cincinnati, Ohio.
air	<sup>222</sup> Rn	0.5–0.7 pCi L <sup>-1</sup>	FMPC and Mound facility measurements in FMPC area.

Table 2. Summary of Background Concentrations of Uranium and Radon
in Environmental Media, Applicable to the FMPC

### SUMMARY OF APPENDIX B - REGIONAL ENVIRONMENTAL MONITORING

### Part 1 — Deposition Measurements Using Gummed-Film

Gummed-film was used to measure uranium deposition at locations on the FMPC plant site during the years 1953-1965. These measurements were relatively continuous during the periods of highest releases from the facility. They also indicate the trend of contamination as a function of distance from the center of the production area, one of the few sets of environmental measurements that provides such perspective.

The monitoring locations around the FMPC are listed in Table B1-1 of the Appendix. Those locations nearest the facility are shown in Figure 2. Not all of the locations were used throughout the thirteen years when gummed-film monitoring was performed. Like most of the environmental measurement programs, the gummed-film monitoring effort was expanded as the years passed. However, the program was discontinued in the mid-1960s.



Figure 2. Gummed-film deposition measurement locations near the FMPC. Locations are based on a map found in the FMPC archives and are approximate.

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when they were exposed for extended periods. This has been evaluated using measurements of collection efficiency made by others and data from a 2-year period when the effect of exposure duration was being assessed at the FMPC. The results of the evaluation are given in Table B1-2; details of the evaluation were reported in Appendix M of the Task 4 report (Killough et al. 1993).

All of the gummed-film data that were found in the FMPC files were compiled, corrected for collection efficiency, and have been tabulated at the end of Appendix B, Part 1. Initial use of the data for model validation comparisons was performed and reported in the Task 4 report. Appropriate portions of this data set will be used for further checks of calculated uranium deposition when the complete source term for the relevant years is available.

Uranium deposition estimates are provided for the sampling locations with the longest periods of record in Figures B1-3 and B1-4 in Appendix B. These plots show the time trends of deposition rate at four principal locations near the center of the production area. Estimates of annual depositions at eight onsite locations are shown in Figure 3. Uranium deposition in the FMPC area was highest in 1955. The dependence of uranium deposition density on distance from the facility center is shown in Figures B1-6 and B1-7 of the Appendix.

### Part 2 — Air Monitoring Data

Examination of historic air monitoring data around the Fernald site is important to the verification of release estimates and model predictions for the dose reconstruction project. The air monitoring data can provide measurements to compare with environmental transport model predictions, can assist in choosing appropriate models, and can provide one way of investigating possible episodic releases which may have been unmonitored or undetected at the release points.

Although the environmental monitoring data are important to consider in developing methods for dose reconstruction, they are not complete enough, either temporally or spatially, to rely on exclusively for assessment of the exposure to surrounding populations from FMPC effluents. Rather, these data are used primarily to provide a quality check of the source term estimates and to calibrate or validate the transport models.

Appendix L of Killough et al. (1993) focused on air monitoring data from the early 1960s, in support of the model simulations performed for this time period. The complete set of air monitoring data is included in Appendix B Part 2 of this Task 5 report. Summary tables and figures are included in the main body of the text; detailed data tables are included as an annex.

From the earliest years of operation, ambient air around the FMPC was sampled and analyzed for uranium. The amount and quality of data available has improved over the years. An evaluation of the quality of the air monitoring data was included in Appendix L of the Task 4 report of this project (Killough et al. 1993). Samples were routinely obtained at the FMPC perimeter from 1958 through 1971, at which time boundary stations were established (Figure 4). Prior to 1958 and at offsite locations, samples were taken infrequently for shorter periods of time. In the 1980s, permanent air monitoring stations at offsite locations were established. The monthly average concentrations of uranium in air at

the FMPC perimeter and boundary stations were computed from the weekly measurements and are presented in the Annex to Appendix B, Part 2. The monthly average concentrations are plotted to illustrate long-term trends of uranium in air around the FMPC.



Figure 3. Estimated annual uranium depositions at eight onsite gummedfilm monitoring locations between 1954 and 1964. In some cases the data are less complete, and the estimates are more uncertain. The largest data gaps occur in 1955 and 1959.



### Figure 4. Ambient air sampling locations around the FMPC.

A primary use of the air monitoring data will be model validation, which consists of comparison of model predictions to available measurements at different places and times. A model validation for the three-year period 1960–1962 was included in Killough et al. (1993),

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as part of that methodology development effort. Validations for other time periods will be included in the final Task 6 report. The measurements of uranium in air beyond the FMPC perimeter support the model predictions that concentrations decrease with distance from the site.

All long-term data sets of uranium in air around the FMPC show a decrease with time, consistent with declining production activities and increasing effluent control. Figure 5 summarizes the uranium in air in a NE direction from the site. More detailed plots of monthly average concentrations are included in Appendix B Part 2.



Figure 5. Summary of uranium in air at the perimeter and boundary stations NE of the FMPC from 1958–1991. The boundary station is about 800 m farther from the production area than the NE perimeter station.

In addition to providing data for model/source term validation, another use of the air monitoring data has been the identification of episodic releases. For the purposes of this dose reconstruction project, an episodic release is defined as one which increases the composite uranium release rate by a factor of at least 10 for a period of less than 10 days. All releases are included in the source term, but episodic releases warrant special dose assessment procedures. Plots of monthly average concentrations of uranium in air over time were examined for peaks, which were further investigated by reviewing weekly measurements. In addition, all individual measurements which were  $\geq$  ten times the annual average at that location were tabulated and investigated. There were 20 measurements representing 14 sampling periods which met this criterion. Some previously identified episodic releases were confirmed, and at least one other episodic release (February 1979) was newly-identified by examination of the air monitoring data record. It must be

emphasized that examination of the environmental monitoring data is only one method for identifying potential episodic releases. A complete review of episodic releases, including other types of historic records, will be included in the final source term report of the dose reconstruction project (Tasks 2 and 8).

### Part 3 — Wet Deposition

"Wet deposition" refers to the removal of uranium-bearing particulates from the air onto ground surfaces by the actions of rain, snow, or mixtures. Theoretical aspects of the wet deposition process were addressed in Appendix H of the Task 4 methodology report (Killough et al. 1993). Uranium measurements in wet deposition and air were used to compute a site-specific washout ratio (Table H-1, Killough et al. 1993), a parameter used in the environmental transport model. Additional data presented in Appendix B Part 3 of this Task 5 report are intended to assist in validation of the environmental transport models.

The main sources of information for this analysis were the original analytical data sheets from National Lead Company of Ohio and the monthly/weekly reports from the Industrial Hygiene and Radiation (IH&R) Department. There were only a limited number of measurements of wet deposition in the 1950s. The earliest records located of radioactivity in wet deposition were from the fourth quarter of 1953. These samples were collected in open "fallout trays," which collected both rain and snow as well as dry deposition. The samples were not specifically analyzed for uranium. The measured deposition rates range from 6 to 4700 dpm alpha m<sup>-2</sup> d<sup>-1</sup>, with large differences observed between the alpha activity collected at the various locations. [One picocurie equals 2.22 disintegrations per minute (dpm)]. The concentrations in precipitation ranged from 0.02 to 1.50 dpm alpha mL<sup>-1</sup>. It appears that this fallout tray sampling method was discontinued, as no other records of this type were found.

For the 1960s, a fairly complete data set of specific uranium measurements in precipitation was located. Rain and snow were collected and composited monthly from two locations, the east side of the Security Building at the FMPC and the Abbe Observatory in Cincinnati. The Security Building is located on the southern perimeter of the FMPC complex just west of D Street. The Abbe Observatory is a National Weather Service station located about 15 miles (24 km) south of the FMPC. Samples from the Abbe Observatory were analyzed for uranium concentration by the FMPC analytical department along with samples from the FMPC.

Figure 6 illustrates the data for uranium concentration in precipitation collected from the FMPC and the Abbe Observatory in the 1960s. This data set represents 81 measurements at the FMPC and 53 from Cincinnati. The concentrations at the FMPC are generally 1-2 orders of magnitude higher than those from Cincinnati.

For model validation, the predicted concentrations of uranium in rain at the location of the Abbe Observatory, using reconstructed source terms and the transport model, will be compared with the measured values shown in Figure 6. This comparison will be included with other model validations in the final Task 6 report.

The deposition rate (uranium deposited per unit area per unit time) is also computed and discussed in Appendix B Part 3. Higher wet deposition rates occur in the winter and spring. The total deposition rates are lower than those measured by the gummed-film

(Appendix B Part 1). The median deposition rate measured by gummed-film at the SE perimeter station (closest to the rainfall collection point) during 1961–1964 was 7.0 mg m<sup>-2</sup> d<sup>-1</sup>, whereas the median deposition rate measured in precipitation over the same period was 0.3 mg m<sup>-2</sup> d<sup>-1</sup>. It is not entirely clear why the two measurement results are not in closer agreement, given that they both measure dry and wet deposition, to some extent. Perhaps the open rainfall collector was not particularly efficient for intercepting and retaining dry deposition. Regardless, it does appear that dry deposition processes were more important than wet deposition processes for the particle sizes found in the vicinity of the FMPC perimeter (Killough et al. 1993).



Figure 6. Concentration of uranium in precipitation from the FMPC Security Building and the Cincinnati Abbe Observatory in the 1960s.

### Part 4 — Additional Soil Monitoring Data

Uranium. Appendix N of the Task 4 report (Killough et al. 1993) and Appendix A of this report both present data on uranium in soil. The purpose of Appendix N was to estimate the range of the uranium source term by a method other than those addressed in the Task 2/3 report (Voillequé et al. 1991). Hence the soil data may serve as an independent check of the final atmospheric source term developed by this dose reconstruction study.

Appendix B, Part 4 evaluates uranium levels not reported elsewhere in the FMPC dosimetry reconstruction task reports; illustrates the soil concentrations of uranium with depth; and discusses the occurrence of other radionuclides in the soil around the FMPC. The

other radionuclides include <sup>226</sup>Ra and thorium among the naturally occurring isotopes, and <sup>99</sup>Tc, <sup>137</sup>Cs, <sup>90</sup>Sr and <sup>239</sup>Pu among the man-made radionuclides.

A number of databases of radionuclide measurements in soil were reviewed in the Task 4 report (Killough et al. 1993). Additional databases were made available to the Fernald Dosimetry Reconstruction Project and were reviewed in the Annex to Shleien (1991). The general geographic distribution patterns of uranium in soil measured at different times and distances from the FMPC, are similar (RIFSSOIL 1990). All of the Figures in Appendix B, Part 4 (B4-1 through B4-3) highlight some areas of this contamination onsite. These areas could be due to spills of uranium-bearing materials or waste, or from airborne deposition (also see Figure N-3, Killough et al. 1993). The results show concentrations which are clearly elevated above background in the NE quadrant out to distances of about 8 km. The highest concentrations are found within 1 km of the emissions center. Since winds to the NE are about twice as frequent as those to other quadrants, it can be concluded that these elevated levels represent the deposition of uranium released to the air from FMPC activities. The area immediately to the east is characterized by the presence of the old solid waste incinerator (OSWI) which is definitely a source of localized deposition from airborne uranium. The NNW contamination is associated with the Plant 1 onsite storage area. A metal scrap area, the tank farm, and an unidentified source NE of Plant 9 all show high levels of contamination. The elevated uranium soil levels to the SW may represent uranium distribution by runoff and production activities. The results suggest that many of the areas with high concentrations of uranium within about 1 km of the site center represent contamination by industrial activity, such as localized spills.

During 1985–1989, the FMPC staff conducted various sampling programs that included uranium in soil at various depths as well as grass, vegetation, and produce. The soil depth data in Appendix B, Part 4 generally show the effect of environmental leaching of uranium. Those samples that do not follow the general pattern of decreasing concentration with depth may represent areas of soil mixing or, less likely, an underground source of uranium.

Thorium and <sup>226</sup>Ra. Tables B4–1 and B4–2 present surface soil values for <sup>226</sup>Ra and total thorium respectively. Although many locations lack specific data, no geographic patterns with distance or direction can be discerned. Other thorium isotopes, namely <sup>233</sup>Th, <sup>234</sup>Th and <sup>231</sup>Th may contribute to the total thorium levels reported in Table B4–2. The data in Table B4–1 and B4–2 are within the range of values reported in the scientific literature.

Given the lack of differences in the geographic distribution of <sup>226</sup>Ra and total thorium with distance from the site center, and the fact that levels are within the range of the natural occurrence of these isotopes, it cannot be concluded that their source is other than from natural sources.

Man-made Radionuclides. In order to assess the releases of any other radionuclides, the results for <sup>137</sup>Cs, <sup>90</sup>Sr, <sup>99</sup>Tc, and <sup>239,240</sup>Pu in the data file RIFSSOIL (RIFSSOIL-1988) have been examined. In many cases the results reported for these radionuclides are "less-than" (<) values indicating the actual level was below the minimum sensitivity of the measurement procedure. Sixteen sector averages were calculated for each of these radionuclides, excluding samples with a "less-than" designation. Not including those samples noted as "<" tends to raise the average concentrations for these radionuclides, but does not change the general conclusion regarding distribution or source.

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The results suggest that:

- Sampling and analysis for <sup>99</sup>Tc has been sparse, but some soil contamination with this isotope is present.
- <sup>137</sup>Cs and <sup>90</sup>Sr in soil at the FMPC are most likely from atmospheric weapons testing.
- Plutonium contamination of soil is present onsite, and the source (a Paducah UO<sub>3</sub> 1980 shipment) seems to have been identified.

### Parts 5 and 6 - Milk, Vegetation, River Water, Sediments, and Fish

Parts 5 and 6 of Appendix B present the analytical results of uranium concentrations in milk, vegetation, water, sediment, and fish sampled from the vicinity of the FMPC. The purpose of compiling these data is to observe general trends in concentrations in various components of the air and water pathways, and to provide information on the importance of various pathways for human exposure due to radionuclide releases from the FMPC. In addition, these data can be used to calculate site-specific parameters for use in our model calculations. One example of such a parameter is the concentration ratio (CR) for uranium from soil to grass.

Regular sampling programs for these environmental media began at quite different times during operations at the FMPC. Water samples have been collected regularly from upstream and downstream locations in the Great Miami River and in Paddy's Run Creek to the west of the site since the early fifties, while a fish sampling program was not initiated until 1984. Analysis of forage grass samples began in 1958, but the analysis of food crops did not begin until 1983. Milk samples from the Knollman Farm adjacent to the FMPC were analyzed as early as 1958, with a regular program underway by 1980. Sediment samples have been analyzed from onsite and offsite locations since 1974.

Appendix B, Part 5 summarizes the measurement data of uranium in milk and vegetation samples in the vicinity of the FMPC. The monthly milk samples were analyzed for total uranium using a fluorometric method. Since 1980, additional samples have been analyzed for <sup>90</sup>Sr, <sup>99</sup>Tc, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>228</sup>Th, <sup>230</sup>Th, <sup>232</sup>Th, <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U. Except for a few cases that have been traced to analytical or contamination errors, uranium concentrations in milk have been at or below the limit of detection. The higher than expected values occurred in control as well as local samples, however. Overall, the results indicate no increase in uranium in local milk compared to control samples.

Food crops around the FMPC were not monitored routinely until 1983 when potatoes from the vicinity of the FMPC and from control locations in Indiana were analyzed for uranium. Beginning in 1986, more extensive monitoring of leafy vegetables (cabbage and collards) and root vegetables (potatoes, carrots, and onions) was done. Generally, no clear differences between local and control concentrations have been observed.

Forage material, or grass, was monitored more extensively than food crops. We compiled results from analytical data sheets for the period 1958 to 1968, and for 1984 onward. The data indicate that the levels of uranium in forage grasses decrease with distance from the center of the FMPC. (See Figure B5-1, Appendix B). Furthermore, the annual average uranium concentrations in grass from offsite and onsite locations reflect the general trend of atmospheric releases of uranium from the FMPC. Figure 7 illustrates that the uranium concentrations in grasses decreased significantly from the late fifties when atmospheric releases were high to the late eighties when releases to air were much lower.

The uranium concentration in grasses is related to that in soil by the concentration ratio (CR), which is defined as a ratio of the radionuclide levels in plant material to the radionuclide levels in soil. Plant uptake of radionuclides from soils is affected by many factors, and consequently the CR can vary considerably. We determined a site-specific plant-to-soil CR from parallel measurements of uranium made in grass and soil at offsite FMPC locations during the sixties and the eighties. These data are presented in Appendix B, Part 6. Site-specific values based on these data can be compared to a range of CR values of 0.017 to 0.0053 published in the literature (Peterson 1983). The CRs from the earlier time period are high, outside the range of these published literature values. The CR, based on only the more recent data, yields a much lower median value of 0.03. We suggest that conditions under which the ratios were determined for the earlier years may not have been in equilibrium which is implicit in the definition of the CR ratio. Consequently, the ratio determined from the more recent data may be a better site-specific value to use for pathway analysis modeling if the air-soil-forage-cow-milk pathway is determined to be a key pathway of exposure to the residents in the FMPC area.



Figure 7. Average annual uranium concentrations in grasses from onsite and offsite sampling locations from 1958 to 1991. Production operations were suspended at the FMPC in 1989. Uranium emissions to air are estimates from the FMPC; they do not represent final estimates from the dose reconstruction project.

Appendix B, Part 6 examines the concentrations of uranium (and other radionuclides when available) in surface water, sediment, and fish from the vicinity of the FMPC during various years of operations. The site has conducted an extensive water sampling and uranium analysis program of the Great Miami River and Paddy's Run Creek since 1955

(Figure 8). The purpose of compiling the results of surface water uranium analysis is to observe general trends in uranium concentration in the surface water near the FMPC over time, and to compare these measurements with model-calculated concentrations based on our final source term estimates.



Figure 8. Diagram of the FMPC showing the main water sampling locations in the early years of operation.

Measurements from the original analytical data sheets for the Miami River and Paddy's Run Creek are compiled in tables in the Annex of Appendix B, Part 6. Figure B6-2 in the Appendix shows the monthly average uranium concentrations measured in the river at the

New Baltimore Bridge, approximately 2 km downstream from the site. The data show higher concentrations measured downstream in the river prior to mid-1957, when the monthly average uranium concentrations exhibited extreme fluctuations in concentrations. This change appears to be related to the installation of the storm sewer lift station in mid-1957. Prior to that time, all runoff from the storm sewer system went directly to the river. The concentrations of uranium measured in the Great Miami River have been much lower during all years than those measured in Paddy's Run Creek.

Figure B6–3 in the Appendix shows a gradual decrease in uranium concentrations in Paddy's Run Creek since the late fifties, both onsite above the confluence of the SSOD with Paddy's Run, and just below the site at the Willey Road Bridge. Uranium concentrations measured at the Willey Road Bridge have consistently been above background levels as well as being a source of groundwater contamination. Some of these uranium concentration data will be used in Task 6 to compare with our model-calculated concentrations.

Beginning in 1974, sediment in the Great Miami River was sampled at two locations upstream (at 1 and 2 km), and five sites at increasing distances downstream of the effluent outfall to the river. The 1974 through 1985 uranium concentration data for sediment in the river have been compiled in Appendix R of our Task 4 report (Killough et al. 1993), and are listed in Table B6-3 in Appendix B, Part 6. The average concentrations in sediment taken below the confluence of Paddy's Run with the river are slightly higher than upstream measurements for some years (1977, 1978, 1983), but the data indicate no consistent difference between uranium in sediment measured upstream, just downstream of the effluent discharge point, or further downstream below the point where Paddy's Run Creek flows into the Great Miami River. In addition, sediments collected from the Great Miami River upstream and downstream of the FMPC effluent discharge line, were analyzed for <sup>99</sup>Tc, <sup>235</sup>U, <sup>238</sup>U, <sup>236</sup>U, <sup>232</sup>Th, <sup>228</sup>Th, <sup>230</sup>Th, <sup>223</sup>Ra, <sup>224</sup>Ra, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>238</sup>Pu, and <sup>239,240</sup>Pu. No significant differences in average concentrations of these radionuclides have been observed.

Sediment from onsite locations in Paddy's Run Creek and the Storm Sewer Outfall Ditch have been sampled and analyzed for uranium also since 1974. However, offsite sediment sampling was done for the first time in Paddy's Run Creek south of Willey Road in 1985, while offsite sediment samples north of the site in Paddy's Run were not obtained until 1991. Figure B6-5 in the Appendix shows that the annual average uranium concentration in sediments from Paddy's Run Creek below the confluence of the storm sewer outfall ditch (SSOD) varies directly with the uranium concentration in water from the same location. In 1987, the concentration in both water and sediment from below the SSOD decreased markedly when the storm water retention basin became operational and began receiving runoff that had previously gone directly to Paddy's Run Creek.

Routine sampling of fish from the Great Miami River near the FMPC began only in 1984. Approximately 25 fish fillets have been analyzed each year from each of three locations on the river: 2.5 km upstream, at the main effluent outfall location, and downstream where Paddy's Run Creek drains into the river (Figure 8). Figure B6-6 in Appendix B, Part 6 shows the analytical results measured at these three locations from 1984 through 1990. Except for 1988, there appears to be a downward trend in concentration from 1984 to 1990. However, for each year, the uranium concentrations are not different among the three locations.

### Part 7 - Groundwater, Cisterns, Ponds, and Pools

Historical environmental monitoring data for uranium in groundwater, cistern water, and other water sources are discussed in Appendix B, Part 7. The historic monitoring data for uranium in private (groundwater) wells are important to the dose reconstruction work, because they will be used directly for exposure assessments for years when data are available, and will also be used to help estimate concentrations for years when no data are present.

The significant offsite uranium contamination in groundwater is south of the site, and is 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 at this time. Since this dose reconstruction project is concerned with past doses to people around the site, the groundwater contamination to be considered in this project is limited to the South Plume. Figure 9 shows the estimated areal extent of the South Plume uranium contamination as of the end of 1991, as well as the locations of the private wells monitored by the FMPC. The area of the South Plume has been estimated by the FMPC.

In the Task 4 report (Killough et al. 1993), 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. For years when groundwater uranium monitoring data are available, the measured concentrations in private wells around the FMPC will be used directly to calculate radiation doses to affected individuals.

For years when groundwater monitoring data are not available, the source term work of Tasks 2 and 3 of this project (in progress) will attempt to develop estimates of the uranium concentrations in wells in the South Plume, as a function of time. That work will use two major types of information: measured uranium concentrations in the private wells in the South Plume, and information about releases to the storm sewer outfall ditch and to Paddy's Run Creek (the source of the contamination). Estimates of the concentrations of uranium in water released to the storm sewer outfall ditch and to Paddy's Run Creek will be developed in the Tasks 2 and 3 work. Trends in the estimated discharges will be examined and compared to trends in the uranium concentrations in the South Plume, to help determine estimated concentrations in the plume for other time periods.

The FMPC routine groundwater well monitoring program is the most comprehensive for private wells in the area. Many wells are monitored monthly, and routine monitoring has been performed since 1982. Annual average concentrations of uranium in private wells around the FMPC have been compiled for 1983–1990 in Appendix B Part 7. These data show that uranium concentrations are significantly elevated above background in three wells, 12, 15, and 17, which are located within the South Plume area. For these three wells, additional monthly monitoring results have been compiled for November 1981 through February 1985. Concentrations in wells 12 and 17 show no significant trends, but concentrations in well 15 gradually increased in 1982 and then gradually decreased in 1983 and the first half of 1984. Table 3 compares the long-term average uranium concentrations in the three contaminated wells to the background concentration (see Appendix B, Part 7). Detailed monitoring results are available and will be discussed in the final Task 2/3 report.

### Review of Historic Data and Assessments for the FMPC

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Figure 9. Approximate area of uranium contamination in the South Plume, as of the end of 1991, and locations of the private wells around the FMPC sampled in the FMPC routine monitoring program. Although well 26 is within the area of groundwater contamination, the uranium concentrations from this well are at background levels, because it was installed in the mid-eighties at a greater depth than the others.

Uranium Concentrations in Contaminated Private Wells and Background Wells			
Well	Period	Concentration (pCi L <sup>-1</sup> )	
12	1982-1990	160	
15	1982-1990	220	
17	1982-1990	35	
Background	1983-1990	0.09–1.3	

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Monitoring of private wells around the FMPC for uranium has also been performed by other organizations. Though these data are much less comprehensive, the results corroborate the findings based on the FMPC routine monitoring. Results from duplicate analyses of water samples split between the FMPC and the Ohio Department of Health (ODH) have been summarized. These data show generally good agreement between FMPC and ODH results.

### SUMMARY OF APPENDIX C - PARTICLE SIZE OF AIRBORNE EFFLUENTS

#### Part 1 — NKES Study - Methodology QA

The particle-size distributions of uranium stack emissions are needed in order to calculate both the gravitational settling of uranium-containing particulates in airborne plumes and radiation exposures via the inhalation pathway. In addition, a knowledge of the particle-size distributions is necessary if corrections of uranium stack releases need to be made to account for losses through particle deposition in sampling lines. The only measurements of the particle sizes of stack emissions from the FMPC were conducted by Northern Kentucky Environmental Services (NKES) during 1985. An unpublished report is available on this work (Reed 1985). In the NKES study, measurements were made for both the inlet ducts and the outlet ducts of 15 major uranium-emitting stacks with dust collectors. The particle-size distributions determined in the study are listed in an FMPC report, FMPC-2082 (Boback et al. 1987).

Earlier in the project, distributions of the uranium species for both the inlet and outlet ducts of each of the 15 dust collectors were plotted using a procedure developed for interpolating and extrapolating the values from the FMPC-2082 report. The plots and procedure are reported in Appendix F of the RAC Task 2 and 3 interim report (Voillequé et al. 1991).

Appendix D of the Task 4 report contains the final particle size distributions as used in this study (Killough et al. 1993). Particle sizes for the outlet ducts (or emission stacks) are representative of emissions from stacks with intact bag filters in the dust collectors. The values for the inlet ducts, however, may be assumed to represent emissions from the same stacks during those periods in which the bag filters had failed in a manner that allowed unfiltered inlet air to escape to the atmosphere.

In Appendix C Part 1 of this Task 5 report, we evaluate the methodology employed by the NKES. The methodology is compared to that recommended in the operating manual for the Andersen Mark III stack sampler (Andersen 1984). To investigate the raw data and calculations from the NKES study, raw data from about 10 percent of randomly selected sampling runs were analyzed and compared with the reported results. The conclusions gleaned from these recalculations are presented in Appendix C, Part 1.

Additionally, other information of importance to the Fernald dose reconstruction project . present in the NKES report is noted, and comment is made on further particle size work required for environmental modeling.

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The sampling methods employed in the NKES study appear to follow the directions in the Andersen operating manual. The techniques employed should not have added to the existing uncertainties inherent in the sampling methodology.

The selected hand-written analytical laboratory sheets were inspected in detail to evaluate the raw input data and calculations. Except in a single case, unit conversions were rounded off and performed correctly, however, no explanation was given for the diversity of units employed. An error was noted in copying total dust loading in one case. There was a single instance where the volume of air sampled was off by a factor of two. For the most part, the errors led to erroneous emissions concentrations, and did not impact directly on the assessment of particle size determinations.

Inspection of the resultant particle size determination show discrepancies in five of the sixteen sets of runs (Table C1-1). Outlet (emissions) particle size is greater than the inlet (prior to the dust collection) particle size for these runs. Two of the five runs appear to be associated with anomalies in recording the data or in the analyses themselves. Extreme care needs to be exercised prior to using information on particle size without first checking the original data sources.

### Part 2 — Final Particle Size Distributions

Appendix C, Part 2 of this report contains a detailed analysis of final particle-size distributions of uranium-containing particulates emitted from FMPC stacks. This information is needed in order to estimate gravitational settling, radiation exposures via inhalation, and deposition losses in sampling lines. Particle-size measurements were made by Northern Kentucky Environmental Services (NKES) in 1985 for emissions from both inlet and outlet ducts of dust collectors serving 15 stacks. These stacks emitted either  $UF_4$  made by the hydrofluorination process or  $U_3O_8$  produced by air oxidation of uranium metal surfaces during foundry operations.

Particle-size distributions for all  $UF_4$ -emitting stacks and also for all  $U_3O_8$ -emitting stacks were averaged. The median values for  $UF_4$  inlet and outlet ducts were 9.5 and 8.1 mm aerodynamic diameter, respectively; corresponding values for  $U_3O_8$  were 8.3 and 6.0 mm, respectively. Neither the hydrofluorination process for producing  $UF_4$  nor the foundry operations producing  $U_3O_8$  particulates have changed significantly over the years of FMPC operation. Accordingly, the average particle-size distributions measured for these species can be applied to all emissions over the years of operation in which the same species is released from similar plant operations.

Some stacks at the FMPC served uranium metal machining operations. Average particle sizes of  $U_3O_8$  emitted from machining operations in other facilities may be applied to similar FMPC stack emissions. The average median value for airborne particulates produced by uranium machining at Los Alamos and at the United Kingdom was about 6.8 mm. This value may be assumed to apply to inlet ducts of dust collectors at the FMPC. An aerodynamic diameter of about 5.1 mm was assumed to apply to outlet ducts, which represents a typical reduction of 25% observed at the FMPC for filtration by dust collectors.

Uranium ores and various uranium feedstock were handled in Plant 1 and Plant 2/3 of the FMPC. Particle sizes measured for airborne dust from mining and milling operations in

the Elliot Lake Area of Canada averaged about 7 mm. This value may be applied to the  $U_3O_8$  dust produced in Plant 1 and Plant 2/3 for similar ore-handling processes.

Assignment of particle sizes for uranium releases for all stacks over all years of operation requires identification of both the major released species and its generating process for each stack for each year. Particle-size values at midpoints of uncertainty ranges may be assigned for cases in which specific information is not available.

## SUMMARY OF APPENDIX D -- COMPARISON OF THE RAC MODELS WITH OTHER MODELS

### Part 1 -- Comparison of Models for Airborne Uranium and Radon

The modeling methodology of Task 4 (Killough et al. 1993) identifies two air transport models to be applied to releases of particulate uranium and to radon and radon daughters released from the FMPC site. The well-known GAUSSIAN PLUME model (Hanna et al. 1982) is used for releases of uranium from the old solid waste incinerator on the east boundary of the site, and from the oil burner, which was located in the production area during the period 1960–1962. For this three-year period, these sources accounted for less than 1% of the uranium released to the atmosphere from the site. For rooftop releases of uranium from the production plants, we used a variant of the Gaussian plume, called the TIME-DEPENDENT model (Ramsdell 1990), designed to account for building wake effects. We have also applied a specially coded version of this model to releases of radon and radon daughters from the K-65 silos west of the production area, on the assumption that wake effects from the silos should be considered.

Implementations of these models for specific purposes involve complexities that have been discussed elsewhere (Killough et al. 1993). This discussion is confined to tests to verify our interpretation of the basic form of each code — by comparisons to an independent code. For the Gaussian plume model, we have compared results calculated by our program with similar numbers computed by MICROAIRDOS<sup>TM</sup> (Moore et al. 1989). In the case of the timedependent model, we have used a graph from Ramsdell (1990) as our standard.

Initially, calculations were made for sector NE, the sector at Fernald where one would expect the highest air concentrations and ground depositions. We calculated results at 500 m, and then at 1000-m intervals out to 8000 m (Tables D1-1 and D1-2). Following this initial comparison, results were compared for various wind directions to ensure that this variable did not skew the results. Only <sup>238</sup>U and radon were compared in the latter case since no variations with uranium isotopes were observed (Table D1-3). However, for ground concentrations, predicted concentrations of <sup>234</sup>Th using the MICROAIRDOS<sup>TM</sup> were about one-tenth those predicted by the RAC model. This is because MICROAIRDOS<sup>TM</sup> assumes that the radionuclides are released over a year and decay on the ground for a year after deposition. The RAC model employs instantaneous release depositions. For long-lived radionuclides such as <sup>238</sup>U, <sup>234</sup>U, and <sup>235</sup>U with half-lives of  $4.468 \times 10^9$ ,  $2.445 \times 10^5$ , and 7.038  $\times 10^8$  years (Shleien 1992), the discrepancy would be unnoticed because radionuclide decay

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over a year is minimal. However for <sup>234</sup>Th, with a half-life of 24.1 days (Shleien 1992) the discrepancy due to decay is considerable.

The results for radionuclides other than  $^{234}$ Th show reasonable agreement. The ratio between MICROAIRDOS<sup>TM</sup> and the RAC program has a range of 1.26 to 1.52 in both comparisons of air concentrations and of ground depositions for  $^{238}$ U,  $^{234}$ U, and  $^{235}$ U (Table D1-1). For radon air concentrations the variation is somewhat less, from 1.07 to 1.21 (Table D1-2).

For the time-dependent model, we employ output from a study by Ramsdell (1990). In his paper, Ramsdell (1990) used a graph to summarize a model comparison involving the time-dependent model, and we have digitized the appropriate curve from that graph to serve as our standard for comparison.

This verification exercise discussed here is very narrow in scope. It tests our interpretation of the published algorithm and our method of coding the algorithm. It cannot test directly our more elaborate implementations of the model. However, this test of the algorithm and coding method for the time-dependent model showed our method of implementation is correct.

#### Part 2 — Surface Water Pathways

Part 2 of Appendix D compares our surface water modeling methodology for the transport and dispersion of radioactive materials from the FMPC with an independent, surface water dispersion model, GENII (Napier et al. 1988). Our methodology is based on a simple monthly dilution (MD) model for calculating concentrations of radionuclides in surface waters near the FMPC, which is described in Task 4 (Killough et al. 1993). We will ultimately use this model to calculate radiation doses from releases of radioactive materials from the FMPC. We presented the results of this comparison, based on our monthly source term estimates for 1960 to 1962 (Voillequé et al. 1991), in Task 4. In Appendix D, Part 2 of this report, the details of this comparison of uranium concentrations in the river, summarized in Table D2-2, and for Paddy's Run Creek in Table D2-3, are described. The results indicate good agreement between the models. This agreement suggests that the methods we have developed to determine surface water concentrations of uranium and other radionuclides based on our monthly source term data are reasonably congruent with other models developed for similar purposes.

In Task 4, we also compared our calculated uranium concentrations with actual environmental sampling measurements that were done in the Great Miami River and in Paddy's Run Creek (Killough et al. 1993). This procedure indicated quite close agreement between the measured uranium concentrations in the river and those calculated with our model, providing a measure of proof that our model of calculating environmental concentrations is reasonable.

### SUMMARY OF APPENDIX E — MONITORING DATA FOR RADON IN AIR AND EXPOSURE RATE: WITH COMPARISON TO PREDICTIONS

In addition to the particulate releases from the FMPC stacks, there are two types of releases from the waste storage silos, located in the waste disposal area west of the FMPC

production area, that were evaluated. Figure 10 shows the location of the waste storage silos. First, there is the release of <sup>222</sup>Rn (generally called "radon") and its short-lived daughters from the K-65 Silos, Silos 1 and 2. This release was described in our previous source term report (Voillequé et al. 1991). Second, there is gamma radiation that is emitted from the K-65 Silos and the Metal Oxide Silo, Silo 3. This gamma radiation represents a potential source of direct radiation exposure to people living near the Silos. Calculations of direct exposures from radiation emitted from the Silos are described in the Task 4 report (Killough et al. 1993) and final Tasks 2 and 3 report (Voillequé et al. 1993) of this project. In our preliminary source term assessment (Voillequé et al. 1991), we determined that the Metal Oxide Silo is not an important source of radon releases. However, because it contains high concentrations of radioactive materials, it does represent a potentially significant source of direct radiation exposure.



Figure 10. Location of the waste storage silos on the west side of the FMPC site.

Historic environmental monitoring data for radon in air and exposure rates from penetrating radiation around the FMPC are compiled and, in some cases, compared to predictions of our radon dispersion and direct radiation exposures models, in Appendix E. These data and comparisons are important for the dose reconstruction work, because the results can be used to evaluate the performance of our models. In addition, some of the data compiled here have not been published previously, and it is important to make these data available.

A number of environmental radon data sets were evaluated:

- Early FMPC monitoring performed prior to July 1980.
- Routine FMPC monitoring for 1981-1990, primarily along the site boundary.
- FMPC monitoring on the K-65 Area fence line, close to the K-65 Silos.
- Monitoring by the Ohio Department of Health for 1985–1989, on the site boundary.

In our previous source term work (Voillequé et al. 1991), we estimated radon release rates from the K-65 Silos for periods before and after the sealing of the K-65 Silos. Around the end of June 1979, the gooseneck vent pipes on the Silos were removed and the openings were sealed, and the metal covers for the manholes and fill pipes on the Silo domes were gasketed and bolted shut. This sealing of the K-65 Silos caused a decrease in the radon release rate from the Silos, but caused an increase in the radon concentration in the air inside the Silo head spaces. The estimated radon release rate from the K-65 Silos for the period 1959 to mid-1979 is about seven times higher than the estimated release rate for the period after the Silos were sealed (mid-1979 to 1987). Because the estimated release rate for this earlier period is much higher than later periods, it is especially important to have corroborating environmental data.

The early radon monitoring data from 1978–1980, which were previously unpublished, appear to be the only environmental radon monitoring performed before the K-65 Silos were sealed in mid-1979. From our analysis of the integrated radon measurements from April, May, and June 1979, the radon concentrations in air at the boundary station BS-6 prior to the sealing of the Silos agree well with our predicted concentrations. The data also show a significant decrease in radon concentration after the sealing.

For the period mid-1979 to 1987, we have made comparisons of predicted radon concentrations in air to measured concentrations for two data sets: (1) the monitoring performed by the Mound facility in 1985 and 1986 (Killough et al. 1993), and (2) the FMPC routine monitoring at boundary air monitoring stations for 1981-1990 (Appendix E of this report). In both of these comparisons, the predicted and measured concentrations agree relatively well, considering the significant uncertainties in the radon release rates, air dispersion model, and in the measurements. The comparisons did show some under-bias in our predicted concentrations.

Data for radon concentrations measured on the fence line around the K-65 Area in the FMPC monitoring program, from 1987 through 1991, are also compiled. Because these measurements bracket the end of 1987, when the foam layer was applied to the K-65 Silo domes, they may be useful for our development of the radon release rate for 1988 in the final report of Tasks 2 and 3.

In relation to direct exposures from gamma radiation emitted from materials in the K-65 and Metal Oxide Silos, we have compared predicted and measured exposure rates for three major studies of exposure rate measurements: (1) surveys along Paddy's Run Road in 1987 (in Task 4 of this project, Killough et al. 1993), (2) a 1957 survey relatively close to the K-65 Silos, and (3) the FMPC routine exposure rate monitoring at the site boundary air monitoring stations. For the Paddy's Run Road surveys in 1987, the predicted exposure rates were about one-half the measured values. For the 1957 survey, the geometric mean of predicted to observed ratios (P/O) was 3.0, although P/O values were generally less than 2.5

for greater distances from the Silos. For the FMPC routine monitoring, P/O ratios were about 1 both prior to and after the sealing of the Silos. These comparisons indicate reasonably good agreement between our predictions and the environmental measurements. These results will be used later in this project for final determinations about the performance of our direct radiation exposure model.

## SUMMARY OF APPENDIX F — OTHER RADIONUCLIDES IN AIRBORNE AND LIQUID EFFLUENTS

Appendix F of this report presents a critique of reported analytical data on radionuclides other than uranium and thorium which had been released to the atmosphere from the FMPC. These other radionuclides include daughters of <sup>238</sup>U in natural uranium and daughters of <sup>232</sup>Th in natural thorium produced through radioactive decay. Small amounts of fission and activation products were introduced to the FMPC in recycled uranium. Trace quantities of transuranic elements were also present as contaminants.

The only measurements of the other radionuclides in airborne releases at the FMPC were made in 1985. These measurements were made from bulk dust samples from dust collectors serving Plants 1, 4, 5, 8, 9, and the Pilot Plant and the Plant 8 scrubbers. Several discrepancies were noted in the reported results of the measurements. Accordingly, the data were subjected to careful verification and investigative procedures in attempts to resolve these questions.

The major discrepancies involved <sup>234</sup>Th and its daughter <sup>234</sup>mPa, which should have been in secular equilibrium with the <sup>238</sup>U in natural uranium. The 1985 analytical results for <sup>234</sup>Th were about 40% higher than expected for secular equilibrium for all of the plants except for Plant 5 and Plant 9. For these plants, the <sup>234</sup>Th was high by a factor of 10 or more.

The reported <sup>234</sup>Th concentrations were corrected to the actual times that the samples were taken in order to compare them directly with the <sup>234m</sup>Pa concentrations. The <sup>234</sup>Th values were still somewhat higher than expected for secular equilibrium for all plants except for Plant 5 and Plant 9. Interferences in the analytical procedures by other thorium nuclides are believed to account for these higher values.

The extremely high concentrations of  $^{234}$ Th for Plant 5 and Plant 9 dust can be explained by the fact that these plants processed liquid uranium. Thorium daughter impurities as oxides in liquid uranium are reported to migrate to the surfaces of the uranium during solidification. This migration would have resulted in higher than expected thorium concentrations in the U<sub>3</sub>O<sub>8</sub> solids accumulated on uranium metal surfaces.

Measurements of other radionuclides in liquid effluents were made since the midseventies. Concentrations of plutonium isotopes and <sup>237</sup>Np relative to that of uranium were measured in FMPC wastewater discharges over the period 1976 through 1984, and are listed in the Task 2/3 report (Voillequé et al. 1991). The mean value for <sup>99</sup>Tc is higher than the other values by factors ranging from 500 to 4800. These high levels are explained by the fact that technetium, unlike other fission products and transuranics, is very soluble and mobile in soils. Most of the wastewater discharged from the FMPC came from runoff over ground surfaces where it was in contact with soils (Voillequé et al. 1991).

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### APPENDIX A

### RADIOACTIVITY BACKGROUND AROUND THE FEED MATERIALS PRODUCTION CENTER

#### INTRODUCTION

Uranium and its decay products are radionuclides which occur naturally in the environment. The concentrations which are normally observed in the environment, without enrichment by man's activities, are often referred to as "background" concentrations. An understanding of the background conditions is important to the dose reconstruction effort for several reasons. First, background concentrations must be known in order to assess the influence of emissions from the FMPC. Secondly, these concentrations can provide some perspective, in terms of risk, to the magnitude of concentrations observed in the environment around the FMPC.

This Appendix presents information on background concentrations of uranium in air, soil, surface water, rain, and human diets, and of radon and daughter products in air. Because soil is the primary environmental reservoir for uranium, it will be discussed first. Natural background concentrations vary globally; therefore, whenever possible, an estimate of background which is appropriate for southern Ohio was obtained.

### URANIUM IN SOIL

Appendix B, Part 4 of this report discusses historic measurements of uranium in soil around the FMPC. Figures B4-1, B4-2, and B4-3 in that section present deposition patterns for total uranium in surface soil around the facility. The values in the figures are averages of samples taken in a certain distance and direction interval with respect to the site. Examination of these total uranium data is one method to estimate a "background" level for samples outside of the apparent deposition area. Visual inspection of unshaded areas in these figures indicates that the total uranium background level ranges from 0.8 to 2.9 pCi U g<sup>-1</sup>. The values for earlier soil samplings (1984-1986, Figures B4-1 and B4-2) are somewhat lower than those in Figure B4-4 (1986-1989) and may be due to analytical bias.

Other scientific groups have made measurements of background uranium in Ohio soils. Researchers at the University of Cincinnati determined concentrations of natural uranium in soil (0-5 cm depth) at locations distant from the FMPC (15 and 20 km away). Concentrations were determined by gamma spectrometry. The total uranium concentration ranged from 1.08 to 1.91 pCi g<sup>-1</sup> with an average value of 1.56 (Eckart 1992).

Myrick et al. (1983) present data on background radionuclide concentrations in soil at 356 locations in 33 states across the U.S. The samples were collected to a depth of 6 cm, dried, pulverized, and passed through a 35 mesh screen (<500  $\mu$ m particle size). Analysis for <sup>238</sup>U was by neutron absorption, which results in a sensitivity for <sup>238</sup>U of about 0.02 pCi g<sup>-1</sup>.
The nationwide average concentration of  $^{238}$ U in surface soil was determined to be 1.0 pCi g<sup>-1</sup>. The natural background concentrations of radionuclides in Ohio place it among the highest third of the states, which tend to be interior as opposed to coastal states. The data for 12 sampling locations in Ohio are presented in Table A-1; total uranium in soil would be about twice the listed  $^{238}$ U concentrations.

Tab	le A-1. Back	ground Co	oncentrati	ons (pCi g-	<sup>1</sup> ) of <sup>238</sup> U in 12
	Surface Soi	l Samples	from Ohio	) (Myrick e	t al. 1983)
-	072.)			) 0	

Range of Values	Arithmetic Mean and Standard Deviation <sup>a</sup>	Geometric Mean and Standard Deviation <sup>b</sup>
0.76-2.2	1.4 ± 0.79	1.3 : 1.4

<sup>a</sup>Standard deviation of arithmetic mean is the 2σ value. <sup>b</sup>The geometric standard deviation is a multiplicative parameter to the geometric mean containing 68% (1σ) of the frequency values.

#### URANIUM IN AIR

Airborne uranium is associated with particles of soil which are suspended in the air. A global average concentration of 32 aCi <sup>238</sup>U m<sup>-3</sup> air is given by UNSCEAR (1982), by assuming a particulate loading of 50  $\mu$ g m<sup>-3</sup> in surface air of populated areas, and an average of 0.68 pCi <sup>238</sup>U per gram of surface soil. [An attocurie (aCi) is equal to  $1 \times 10^{-18}$  Ci, or  $1 \times 10^{-6}$  pCi.] This corresponds to about 64 aCi total uranium per cubic meter air. Using this same mass loading approach, a site-specific estimate of background uranium in air would be 70–100 aCi m<sup>-3</sup>, based on a particulate loading in air of 35  $\mu$ g m<sup>-3</sup> (Killough et al. 1993, Appendix O), and a background concentration of total uranium in soil of 2–3 pCi g<sup>-1</sup> of soil (previous section, this report).

Direct measurements of background uranium concentrations in air have been published in the *Environmental Radiation Data* report series (EPA 1981–1988). The Environmental Protection Agency measures the concentrations of uranium isotopes by the analysis of semiannually composited samples (air filters) collected from continuously operating airborne particulate samplers at a number of stations throughout the U.S. Concentrations of the specific isotopes of <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U are determined by alpha spectroscopy following chemical separation. The closest air monitoring station to the FMPC is in Columbus, Ohio; data for 1980 through 1987 are presented in Table A-2. The average value for <sup>238</sup>U is 40 aCi m<sup>-3</sup> (range 24-68) and for total uranium it is 87 aCi m<sup>-3</sup> (range 50-140).

Monitoring of uranium in air around the FMPC has been conducted routinely by the site's operating contractors since the early 1960s through the present time. These data are presented and reviewed in Appendix B, Part 2. The annual average concentrations of uranium in air at seven permanent offsite air monitoring stations ranged between 40-60 aCi m<sup>-3</sup> in 1990 and 60-120 aCi m<sup>-3</sup> in 1989. These measurements are in good agreement with the background values reviewed here. However, only since production at the FMPC ceased have the boundary air monitoring stations registered concentrations of uranium in air which are representative of background (Appendix B, Part 2). A typical annual average

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concentration of uranium in air at the perimeter of the FMPC in the 1960s was about 2000 times greater than background.

					Corumbu	, ошо	
Time period	<sup>234</sup> U	2 σ <sup>b</sup>	<sup>235</sup> U	2σ <sup>b</sup>	<sup>238</sup> U	2σ <sup>b</sup>	Total U <sup>c</sup>
July-Dec 1987	37.0	6.9	2.1	1.5	24.4	5.1	63.5
Jan-June 1987	28.1	4.5	1.2	0.8	25.0	4.1	54.3
July-Dec 1986	23.0	3.8	1.2	0.8	26.1	4.1	50.3
Jan-June 1986	27.5	4.7	2.3	1.2	27.6	4.7	57.4
July-Dec 1985	27.3	3.7	1.1	0.6	28.4	3.8	56.8
Jan-Jun 1985	38.5	4.7	2.3	0.8	33.3	4.2	74.1
July-Sept 1984	45.1	6.2	2.0	0.8	40.2	5.6	87.3
April–June 1984	53.7	6.3	3.9	1.2	52.3	6.2	109.9
Jan-Mar 1984	50.3	6.7	1.1	0.7	46.8	6.4	98.2
Oct-Dec 1983	39.0	9.9	1.1	1.0	38.7	10.4	78.8
July-Sept 1983	49.3	6.7	2.1	0. <del>9</del>	49.3	6.7	100.7
April-June 1983	39.2	6.8	2.0	1.1	34.9	6.2	<b>76</b> .1
Jan-Mar 1983	27.3	6.6	1.0	0.9	28.3	6.7	56.6
Oct-Dec 1982	31.5	4.5	1.5	0.6	27.5	4.1	<del>6</del> 0.5
July-Sept 1982	51.4	7.7	2.7	1.1	39.3	6.2	93.4
April-Jun 1982	40.8	6.4	1.7	0.7	38.9	6.1	81.4
Jan-Mar 1982	44.6	6.7	2.1	0.8	38.6	5. <del>9</del>	85.3
Oct-Dec 1981	34. <b>9</b>	4.9	1.2	0.6	38.4	5.3	74.5
July-Sept 1981	76.1	9.2	4.3	1.2	67.6	8.3	148
Jan-Mar 1981	72.3	8.7	4.4	1.4	56.5	7.2	133.2
Apr-Jun 1981	67.8	10.1	6.9	2.5	64.8	9.7	139.5
Oct-Dec 1980	69.3	8.6	9.6	2.3	46.9	6.4	125.8
Average	44		2.6		40		87
Range	23-76		1.0-9.6		24-68		50140

Table A-2 Uranium in Air (aCi m<sup>-3</sup>) from Columbus Obio<sup>4</sup>

<sup>b</sup> Counting error at the 2σ (95%) confidence level.

<sup>c</sup> Determined by summation of the three individual isotopic measurements.

In summary, three methods were used to bracket the likely range of background uranium in ambient air in the Fernald area. These methods and the range of estimated background concentrations are listed in Table A-3.

Table A-3. Summar	y of Estimates of	Background for	• Total U in Ai	ir at the FMPC

Method of Estimation	Background Estimate (aCi U m <sup>-3</sup> air)
Mass loading calculation	70–100
Measurements in Columbus, OH	
19801987	50-140
Measurements at offsite monitoring	
stations around FMPC, 1989–1990	40-120

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#### URANIUM IN SURFACE WATER AND PRECIPITATION

Natural background levels of uranium in water depend upon whether the water comes from surface waters or ground water. In a large study by the National Uranium Resource Evaluation (NURE) program, plus data from the literature prepared from the US EPA, over 90,000 drinking water samples from around the U.S. were evaluated (Drury et al. 1981). The total data included about 35,000 surface water samples which averaged 1.1 pCi L<sup>-1</sup> and 55,000 ground water samples which averaged 3.2 pCi L<sup>-1</sup>. The 28,000 samples considered to be domestic drinking water supplies averaged 1.7 pCi L<sup>-1</sup> and a population-weighted mean value for finished waters (as opposed to raw, untreated water), based on 100 measurements, was 0.8 pCi L<sup>-1</sup>. In 1988, the Ohio Department of Health (ODH) found that typical background levels of radioactivity in surface water from ponds and small creeks in the Fernald area ranged from 1-2 pCi L<sup>-1</sup> (Steva 1988). In a study of Ohio rivers and streams in the sixties, uranium concentrations in 75 Ohio surface waters ranged from 0.07 to 1.2 pCi L<sup>-1</sup> and averaged 0.5 pCi L<sup>-1</sup> (Durfor and Becker 1964; Scott and Barker 1962).

Upstream samples collected north of the FMPC in both the Great Miami River and in Paddy's Run Creek provide information of background concentrations of uranium in surface water. Weekly surface water samples have been collected from the Great Miami River upstream of the FMPC at the Venice Bridge at Ross since the late fifties by NLO, Inc. (FMPC 1960-1985), and more recently by Westinghouse Materials Company of Ohio (FMPC 1986-1991, FEMP 1992). Figure A-1 shows the annual average uranium concentrations from 1959 to 1991. The average concentration from 1959 through 1970 was  $7.7 \pm 2.6$  pCi L<sup>-1</sup>, and from 1971 to 1991 was  $1.6 \pm 0.7$  pCi L<sup>-1</sup>.



Figure A-1. Annual average uranium concentration measured in the Great Miami River upstream and downstream of the FMPC from 1959 through 1991.

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Paddy's Run Creek samples collected from 1979 to 1991 north of the plant had an average uranium concentration of  $1.2 \pm 0.3$  pCi L<sup>-1</sup>, similar to the upstream concentration in the river from 1970 onward. In the sixties, however, the upstream concentration in Paddy's Run Creek averaged about 10 pCi L<sup>-1</sup>.

Beginning in 1963 a river sampling survey program was begun at the FMPC to obtain background information on water quality at 15 locations on both the Great Miami and Ohio Rivers (Klein 1963). The surveys were to be made twice per year, in spring and in the fall, and all samples were to be taken on the same day. Locations from approximately 100 km upstream to 30 km downstream of the FMPC were sampled. Water was collected from the bridges at the center of the river from the bottom, middle and top of the stream. A composite of the three samples was analyzed. The procedure directed that "the bottom sample was taken by lowering the sampler until it touched the bottom of the stream. The sampler is closed by dropping the weight. The sampler is then raised, shaken to assure that large particles will not settle out, and transferred to the sample bottle" (Klein 1963). Clearly, some sediment was included in the water sample with this procedure. The bioassay department at the FMPC analyzed the samples for nitrates, fluorides, chlorides, uranium, total alpha and total beta activity. The uranium concentration was reported in units of mg U per L<sup>-1</sup>.

Uranium measurements taken from analytical data sheets from this survey program for 1963 and 1964, and for the spring of 1965 and 1967 are listed in Table A-4. A description of the sampling locations and the average uranium concentrations from the available data sheets are shown in Table A-5. Figure A-2 shows the average and maximum uranium concentrations measured at these locations upstream and downstream of the FMPC for 1963 to 1967. The average values range from 2.2 pCi L<sup>-1</sup> in the Ohio River (sampling location 15) to 12.1 pCi L<sup>-1</sup> measured approximately 25 km north of the FMPC (sampling point 9). The average concentration at all locations over this time is 7.0  $\pm$  7.8 pCi L<sup>-1</sup>. This value agrees well with the average concentration (7.7  $\pm$  2.6 pCi L<sup>-1</sup>) measured at the routine sampling location, upstream of the FMPC at the Venice Bridge in Ross, for 1959 to 1970.

If upstream measurements were truly background, we would not expect to observe this decrease with time that is evident in the data for both Paddy's Run and the Great Miami River (Figure A-1). The higher upstream measurements prior to 1970 (versus after 1970) may be due to different analytical procedures. For example, water samples collected for uranium analysis at the FMPC were not filtered prior to acidification with nitric acid (Berger et al. 1985). Depending upon the chemical form of the uranium in the suspended particulates and the length of time between acidifying and analysis, this method could result in overestimating the concentration of dissolved uranium in water. Various amounts of sediments were certainly included in the water samples taken during the river survey program in the sixties (Klein 1963), and there is no indication that the sampling procedure was different for taking routine river samples. Furthermore, sampling bottles were re-used from sampling to sampling, and this may have resulted in contamination of samples (Berger et al. 1985). However, it appears that the sampling protocol or the analytical procedures did not change significantly until the mid-eighties. Furthermore, uranium concentration measurements made by the USGS in the sixties are in the background range seen upstream at the FMPC after 1970. In summary, the data indicate that the background uranium concentration in surface water in the Fernald area ranges from 1 to 2 pCi L<sup>-1</sup>.



Figure A-2. Average and maximum uranium concentrations measured at locations upstream and downstream of the FMPC in the Great Miami and Ohio Rivers from 1963 to 1967. The FMPC is located between sampling points 11 and 12. Data were converted from mass concentration units (Table A-4) using the conversion factor  $6.8 \times 10^{-7}$  Ci g<sup>-1</sup> for natural uranium.

Table A-4. Uranium Concentration Measurements in the Great Miami and	l Ohio
Rivers Upstream and Downstream of the FMPC $^{a}$	

Sample	Distance		Uran	ium Conce	ntration (m	g L <sup>-1</sup> )	
Number	from FMPC	20-Mar-63	17-Sep-63	14-Apr-64	23-Sep-64	8-Apr-65	20-Apr-67
1	100 km N	0.009	0.016	0.008	0.003	0.018	0.001
2	95 km N	0.016	0.03	0.018	0.004	0.007	0.003
3	85 km N	0.024	0.009	0.007	0.004	0.02	nd <sup>b</sup>
4	75 km N	0.009	0.008	0.006	0.002	0.006	0.001
5	60 km N	0.031	0.01	0.003	0.007	0.002	0.002
6	55 km N	0.028	0.003	0.003	0.003	0.005	0.003
7	50 km N	0.011	0.004	0.006	0.006	0.015	0.003
8	40 km N	0.011	0.023	0.011	0.014	0.018	0.002
9	25 km N	0.038	0.002	0.057	0.005	0.004	0.004
10	15 km N	0.01	0.007	0.033	0.008	0.007	0.001
11	2 km N	0.003	0.003	0.049	0.006	0.003	0.003
12	8 km S	0.02	0.008	0.041	0.007	0.006	0.001
13	15 km S	0.021	0.01	0.051	0.004	0.005	0.001
14	30 km S	0.013	0.003	0.006	0.001	0.002	0.004
15	30 km S	no sample	no sample	0.002	0.001	0.001	0.009

<sup>a</sup> Data given in mass concentration units as presented on analytical data sheets, NLO (1963 - 1967).

<sup>b</sup> Data sheet marked "nd" for this sample. This probably indicates "not detectable," or <0.001 mg  $L^{-1}$ .

<b>a</b> ,		Approximate	Uranium C	oncentration
Sample		Distance From	(pC)	(L <sup>-1</sup> )
Number	Location Description	the FMPC	Average	Std. Dev
1	Bridge Rte. 66 & Riverside St. Piqua, Ohio	100 km N	6.1	4.5
2	Bridge County Rd 61 and Peterson Rd below Piqua	95 km N	8.6	6. <del>9</del>
3	Tipp City - Rte. 71 bridge west of Rte. 202	85 km N	8.4	5.7
4	North Ridge Bridge above Dayton on Needmore Blvd. off Rte.202	75 km N	3.5	2.1
5	Upper River Bend, Miami Rd Rte. 25	60 km N	6.5	7.1
6	Bridge in Miamisburg on Rte. 725 west of Rte. 25	55 km N	4.9	6.7
7	Bridge on Chatauqua Rd. west of Rte. 25	50 km N	5.0	3.0
8	Bridge on Germantown Rd (Rte. 4) off Rte. 25	40 km N	8.7	4.7
9	Bridge on Rte. 127 below New Miami, Ohio	25 km N	12.1	15.4
10	Columbia Bridge below Hamilton, Ohio	15 km N	7.3	7.4
11	Bridge at Ross, Ohio	2 km N	7.4	12.3
12	Bridge at Miamitown, Ohio	8 km S	9.1	9.7
13	Bridge at Elizabethtown	15 km S	10.1	12.4
14	Ohio River (Aurora at ferry) upstream of Great Miami River	30 km S	3.2	2.9
15	Ohio River (Anderson Ferry) downstream of Great Miami River	30 km S	2.2	2.6

# Table A-5. Average Uranium Concentrations Measured From 1963 to 1967 in the Great Miami and Ohio River Sampling Survey <sup>a</sup>

Background concentrations of uranium in precipitation have been measured by the Environmental Protection Agency and reported in their *Environmental Radiation Data* reports (EPA 1981–1988). The most appropriate data for use in the Fernald dose reconstruction were collected from Columbus, Ohio. The three isotopes of uranium ( $^{234}$ U,  $^{235}$ U, and  $^{238}$ U) are reported separately in the EPA reports and were summed for our purposes. The median total uranium concentration measured in the 1980s is 0.07 µg L<sup>-1</sup>, with a 95% confidence range of 0.04 to 0.3 µg L<sup>-1</sup> (0.03–0.2 pCi L<sup>-1</sup>). Similar concentrations were reported for precipitation samples taken at 21 air monitoring sites throughout the U.S. in the 1973–1976 period (EPA 1977).

#### URANIUM IN DIET

One source of uranium in the diet is drinking water. Between 1954 and 1957, da: collected on total radium and uranium in water from wells and springs across the United States showed that over 40% of the samples had uranium concentrations greater than 3.5 pCi L<sup>-1</sup> (Scott and Barker 1962). For the East North Central region which includes Ohio, 30% of the wells had uranium concentrations above this level. Background levels in the U.S. for total uranium in groundwater range from 0.068 to 6.8 pCi L<sup>-1</sup> (Hem 1970), while local background levels range from 0.068 to about 2.2 pCi L<sup>-1</sup> (Varchol 1990). For wells in the FMPC area that have not been contaminated from FMPC uranium releases, long-term average concentrations range from 0.09 to 1.3 pCi U L<sup>-1</sup> (see Appendix B, Part 7).

The Office of Radiation Programs of the U.S. Environmental Protection Agency has analyzed some selected drinking water samples for uranium (Cothern and Lappenbush 1983). The concentrations in composite samples (July-December 1977) from 19 cities, including Cincinnati, Ohio, were usually less than 1 pCi L<sup>-1</sup>. The concentration in Cincinnati drinking water was  $0.028 \pm 0.009$  pCi <sup>238</sup>U L<sup>-1</sup> and  $0.035 \pm 0.011$  pCi <sup>234</sup>U L<sup>-1</sup>, for a total uranium concentration of 0.06 pCi L<sup>-1</sup> (0.09 µg L<sup>-1</sup>).

At typical concentrations, drinking water is not the primary contributor to total dietary intake. UNSCEAR (1982) reports a typical annual dietary intake of about 5 Bq (140 pCi) of  $^{238}$ U by people living in areas of "normal" natural radioactivity, which is equivalent to 0.77 pCi (1.1 µg) total uranium per day. Additional estimates of total dietary uranium intake for specific locations are given in Table A-6. No specific estimates of dietary intakes of uranium for the Cincinnati area have been located.

Table A-6. Total Dietary Uranium Intake						
Location Intake (µg U per day)						
New York City, U.S.	1.3ª					
Salt Lake City, U.S.	2.06 <sup>b</sup>					
United Kingdom	1¢					
<sup>a</sup> Fisenne et al. 1987.						
<sup>b</sup> Singh et al. 1990.						
<sup>c</sup> Hamilton 1972.						

#### RADON IN AIR

Background concentrations of radon (we discuss only <sup>222</sup>Rn here) in air are important in determining net radon concentrations due to releases from the K-65 Silos on the FMPC site. Net radon concentrations are used, in Appendix E of this report, for comparisons with predicted radon concentrations due to the releases. In this section we review some of the available information on background concentrations of radon in air around the U.S. and around the FMPC site.

#### Average Concentrations of Radon in Outdoor Air in the United States

The National Council on Radiation Protection and Measurements (NCRP) has produced a number of reports regarding exposures to radon and radon daughters. Although these reports were focused primarily on indoor radon, they also included some information on outdoor concentrations. The average concentrations of <sup>222</sup>Rn were reported to be 0.1 pCi L<sup>-1</sup> over the continents and 0.15 pCi L<sup>-1</sup> over land areas of the northern hemisphere (NCRP 1984a). For its estimates of exposures to the general population in the U.S., the NCRP assumed an average outdoor concentration of 0.18 pCi L<sup>-1</sup>, although this value was the average from a single study at 21 residences in New Jersey and New York (NCRP 1984b).

Gesell (1983) reviewed the available literature for background radon concentrations outdoors and indoors. Of the studies reviewed, only a small number contained year-round data from which an annual average concentration could be obtained. Table A-7 shows the annual average radon concentrations for a number of locations in the U.S., from the yearround studies included in Gesell's review. In some of these studies, measurements had been made only in the morning or only in the afternoon. For these, Gesell adjusted the reported average concentrations to estimate around-the-clock averages, based on mean ratios of average-to-morning and average-to-afternoon concentrations determined from other studies. Some of the studies were based on direct measurements (radon was collected by the sampling method), and some were based on indirect measurements (radon daughters were collected, with radon inferred from an equilibrium ratio). The concentration for Grand Junction, Colorado, was based on samples taken a significant distance from the uranium mill tailings pile. The relatively high concentration is likely due to the natural uranium mineralization in the area.

Gesell (1983) also reviewed average radon concentrations from some studies where measurements were made around the clock, but not for a full year. The data from those studies supported the data from the more complete studies shown in Table A-7. Gesell concluded that "The average outdoor radon level for the contiguous United States probably lies in the range of 100-400 pCi m<sup>-3</sup> [0.1-0.4 pCi L<sup>-1</sup>] and is probably about 250 pCi m<sup>-3</sup> [0.25 pCi L<sup>-1</sup>]."

Gesell (1983) also evaluated seasonal and diurnal variations in outdoor radon concentrations. From the data reviewed, he concluded that seasonal variations generally showed ratios of the maximum to minimum monthly concentrations of between 2 and 4. Data that show the seasonal variations for Cincinnati, Ohio, are presented later in this section. The diurnal variations generally yielded ratios of maximum to minimum concentrations in the range of 2-5.

Location	Adjusted average value (pCi L <sup>-1) b</sup>
Chester, New Jersey	0.22
Socorro, New Mexico	0.24
Cincinnati, Ohio (three-studies)	0.304 0.267 0.263
Washington, D.C.	0.234
Wales, Alaska	0.033
Kodiak, Alas	0.016
Grand Junctic Colorado	0.75

# Table A-7. Annual Average Radon Concentrations in Outdoor Air in the United States <sup>a</sup>

<sup>a</sup> Compiled by Gesell (1983) from other sources.

<sup>b</sup> Averages based on only morning or only afternoon measurements were adjusted by Gesell to estimate the average for continuous measurement.

#### **Regional Measurements of Background Concentrations of Radon in Outdoor Air**

In this section we present data on background radon concentrations in air around the FMPC site, from two sources: (1) the FMPC routine monitoring program, and (2) monitoring performed by the Mound facility, which is a Department of Energy facility in Miamisburg, Ohio. These data are thought to be the most useful for comparisons in other parts of this study. Radon monitoring conducted by the Ohio Department of Health (ODH) also included measurements at control (background) locations. For reasons discussed in Appendix E of this report, these ODH monitoring data are considered more uncertain and less useful than data from the FMPC routine monitoring program. Thus the ODH background data are not discussed here (see Appendix E for more information).

Because the FMPC monitoring program has been operational for the longest time at the greatest number of locations, compared with other monitoring data sets, the values from that monitoring program provide the best picture of average concentrations and their spatial and long-term temporal variability. Routine monitoring of radon in air around the FMPC site began in 1980 (Boback and Ross 1981), but background locations were not incorporated into the monitoring network until 1981 (Fleming et al. 1982). Table A-8 presents the average concentrations measured by the FMPC at the background stations, from 1981 through 1990 (Fleming et al. 1982, Fleming and Ross 1983, Fleming and Ross 1984, Facemire et al. 1985, Aas et al. 1986, WMCO 1988, WMCO 1989, Dugan et al. 1990, and Byrne et al. 1991). These measurements were made using alpha-track, integrating detectors which were exposed for three months (quarterly measurements). As seen in Table A-8, the difference between the maximum and minimum concentration is a factor of 3. When annual average concentrations for single locations (as opposed to the average over all

locations) are considered, the mean radon concentration is 0.58 pCi  $L^{-1}$ , with a standard deviation of 0.17 pCi  $L^{-1}$ .

 Table A-8. Average Background Concentrations of Radon in Air, from FMPC Annual

 Environmental Monitoring Reports

		A	verage n	neasured	background	l concentrat	ion (pCi L	-1) at locati	on <sup>a</sup> :						
Year	8 mi ENE	5 mi WNW	OS 1 5 km	OS 2 3 km	AMSBK1 10.5 km <sup>b</sup>	AMSBK2 6.4 km <sup>b</sup>	BKGD 1 34 km <sup>c</sup>	BKGD 2 25 km <sup>c</sup>	AMS 15 24.8 km	AMS 16 9.9 km	mean				
1981	0.67 <sup>d</sup>	0.36 <sup>e</sup>									0.59 <sup>f</sup>				
1982	0.56	0.66									0.61				
1983	0.77	0.61									0.69				
1984	0.836 <sup>g</sup>	0.357 <sup>g</sup>									0.596				
1985			0.59	0.37							0.48				
1986					0.60	0.57					0.58				
1987					0.66	0.80	0.43	0.76			0.66				
1988							0.3	0.9			0.6				
1989							0.4	0.6			0.5				
1 <b>990</b>							0.4	0.4	0.6	0.6	0.5				
mean							_				0.58				

<sup>a</sup> Data from annual environmental monitoring reports. Distances are from the FMPC to the monitoring location.

<sup>b</sup> In 1986, these locations were called OS 1 and OS 2, but distances are the same as AMSBK1 and AMSBK2.

<sup>c</sup> For BKGD 1 and BKGD 2, it appears that the locations were unchanged for these four years, although naming changed. Distances for BKGD 1 and BKGD 2 were given as 25 and 30 km for 1987, 25 and 34 km for 1988, and 34 and 25 km for 1989 and 1990, respectively.

d This value was based on three quarterly measurements.

<sup>e</sup> This value was based on only a single quarterly measurement.

<sup>f</sup> Weighted to account for one average based on three quarters and one based on one quarter.

<sup>g</sup> Averages reported for 1984 were apparently geometric means. We use them as if they were arithmetic means.

In the report of Task 4 of this Project (Killough et al. 1993), we discussed the radon monitoring performed on the FMPC site by the Mound facility in 1984 through 1986. The measurements were made using Passive Environmental Radon Monitors (PERMs) exposed for one- to two-week periods at many locations within the site boundary (Hagee et al. 1985, Jenkins 1986, and Berven and Cottrell 1987). The locations of this monitoring are shown in Figure A-3. Detailed results for the period July 2, 1985, through October 3, 1986, were given in Killough et al. (1993). The radon concentrations at the Mound locations 14, 17, and 18, all on the eastern boundary of the site, were assumed to be reasonably representative of the background radon concentration around the site. The predicted radon concentrations at these locations due to radon releases from the K-65 Silos were determined to be roughly 10% of the measured concentrations, which provides some support for the use of these locations as estimates of background.



Figure A-3. Monitoring locations of the Mound radon monitoring program on the FMPC site from September 1984 to October 1986 (from Hagee et al. 1985). Locations 14, 17, and 18 are used as estimates of background.

What is particularly useful about the Mound results is that the continuous monitoring using relatively short exposure periods (compared to the quarterly exposures used by the FMPC program) provides information about seasonal patterns in background concentrations. Individual results from the Mound monitoring are given in Killough et al. (1993), Table PS-1. Table A-9 shows the monthly and annual average concentrations, and the ratios of the monthly to the annual average concentration for locations 14, 17, and 18. The monthly averages for each location are plotted in Figure A-4, along with average monthly concentrations for Cincinnati, Ohio, from Gesell's (1983) review. The Cincinnati data include results from two studies, one of which included morning and afternoon measurements. For Study A (arbitrary name applied to differentiate the two), the values are averages of eight years of data. For Study B, the values are averages of four years of data.

		<sup>222</sup> R1				
Nominal period	- Monito <del>r</del> ing dates	14	17	18	Average	Ratio
Annual average	07/02/85-07/02/86	0.36	0.47	0.56	0.46	
July 1985	07/02/85-08/02/85	0.31	0.49	0.56	0.45	0.97
August 1985	08/02/85-08/29/85	0.37	0.53	0.61	0.50	1.08
September 1985	08/29/85-10/01/85	0.78	0.81	1.36	0.98	2.11
October 1985	10/01/85-11/06/85	0.52	0.61	0.78	0.63	1.37
November 1985	11/06/85-12/04/85	0.23	0.31	0.28	0.27	0.59
December 1985	12/04/85-01/02/86	0.36	0.46	0.35	0.39	0.83
January 1986	01/02/86-01/29/86	0.33	0.33	0.34	0.33	0.71
February 1986	01/29/86-02/27/86	0.23	0.26	0.27	0.25	0.55
March 1986	02/27/86-04/02/86	0.22	0.30	0.29	0.27	0.58
April 1986	04/02/86-04/29/86	0.28	0.51	0.55	0.45	0.96
May 1986	04/29/86-05/28/86	0.30	0.46	0.46	0.41	0.87
June 1986	05/28/86-07/02/86	0.35	0.52	0.83	0.57	1.22
July 1986	07/02/86-07/30/86	0.27	0.56	0.71	0.51	1.10
August 1986	07/30/86-09/03/86	0.44	0.87	0. <b>99</b>	0.77	1.66
September 1986	09/03/86-10/03/86	0.40	0.54	0.92	0.62	1.33

Table A-9. Monthly Average Radon Concentrations and Ratio to Annual Average Concentrations, for Locations 14, 17, and 18 of Mound Monitoring<sup>a</sup>

As shown by Figure A-4, radon concentrations at the "background" locations around the FMPC follow the same general trends as do the Cincinnati data reviewed by Gesell (1983). Although the data only cover 15 months, the ratios of the monthly average to annual average should be useful for estimating average background concentrations for periods shorter than a full year.

#### Conclusions

Based on the FMPC routine radon monitoring program and the Mound monitoring program, the average background concentration of radon in outdoor air around the FMPC site appears somewhat higher than averages reported for the Cincinnati, Ohio. The FMPC monitoring data show significant changes in concentrations for different background locations and for different years. Data for Cincinnati and the FMPC also show similar, significant seasonal variations in outdoor radon concentrations. For these reasons, when the results from a particular radon study are evaluated, it is important to use background concentrations measured as part of the same study (with the same instruments and time of monitoring), or those from conditions as similar as possible.

If background concentrations are required for periods shorter than a year, the seasonal variations should be accounted for. Ratios of monthly average to annual average concentrations, obtained from the Mound monitoring at pseudo-background locations, can be used to estimate background concentrations for such shorter periods.

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Figure A-4. Seasonal variability of background radon concentrations in air for Cincinnati and the FMPC. The Cincinnati data are monthly averages based on a number of years of monitoring. The Mound monitoring data, for the FMPC, are monthly averages for July 1985 through September 1986. The three locations shown are considered to be reasonable substitutes for background locations (see text).

#### SUMMARY OF BACKGROUND CONCENTRATIONS

This appendix has presented concentrations of uranium and radon which can be considered background levels in the environment around the FMPC. As such, they are useful for comparison to dosimetric levels which may be associated with the FMPC, and they provide concentrations which need to be subtracted from monitored concentrations to assess contributions from the site.

Estimates of the annual average background concentrations of total uranium in the regional environment are 40–140 aCi m<sup>-3</sup> air, 2–3 pCi g<sup>-1</sup> soil, 0.03–0.2 pCi L<sup>-1</sup> rain, and 1–2 pCi L<sup>-1</sup> surface water. The mean concentration of total U in Cincinnati drinking water in 1977 was 0.06 pCi L<sup>-1</sup>. For wells in the FMPC area not contaminated from FMPC releases, long-term average concentrations range from 0.09 to 1.3 pCi uranium L<sup>-1</sup>. Although results are presented for uranium in the human diet, they are not specific to the FMPC area and are given for informational purposes only.

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Background concentrations of radon in outdoor air are dependent on season as well as time of day. An annual average concentration of radon in outdoor air around the FMPC is in the range of 0.5-0.7 pCi L<sup>-1</sup>.

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#### APPENDIX B – REGIONAL ENVIRONMENTAL MONITORING

#### PART 1 -- DEPOSITION MEASUREMENTS USING GUMMED-FILM

#### INTRODUCTION

Gummed-film was used to measure fallout deposition at locations throughout the United States during the 1950s and 1960s (Eisenbud and Harley 1953, 1955, 1956, 1958; Harley et al. 1960). Deposition was measured daily at 40 to 95 locations during major periods of nuclear weapons testing at the Nevada Test Site (NTS) (Beck et al. 1990). Most of these data have been retrieved and compiled for use in an assessment of radioiodine doses to thyroids of persons in the continental United States from weapons testing at the NTS (Wachholz 1990).

Beginning in 1953, measurements of deposition of particles at locations within the FMPC plant boundaries were performed using gummed-film (Barry 1953). Preliminary measurements were reported for two onsite locations in late 1953 (Barry 1954a, 1954b). Routine data collection at eight locations began in 1954. The number of measurement locations was increased with time to as many as thirty-one (Klein, 1965). The gummed-film monitoring program was discontinued at the end of 1965 (Noyes 1965).

Although the FMPC measurements were not part of the nationwide fallout monitoring effort, evidence indicates that the materials used were the same as in that program (Yoder 1954). Gummed-films, with an exposed area of  $0.093 \text{ m}^2$  (1 ft<sup>2</sup>), were mounted on pedestals that were about 0.9 m above ground. Figure B1-1, a photo taken at the Health and Safety Laboratory in New York, shows a worker preparing to place a square of gummed film on the pedestal. Most of the samples around the FMPC were exchanged monthly, although biweekly collections were more common during the earlier years of the program.

The samples were dry ashed, digested in nitric and hydrofluoric acids, and made up to volume. Aliquots were analyzed using the same fluorometric technique that was employed for many other measurements of uranium (Boback 1960). Gross beta and gross alpha analyses were also performed on most of the samples. The gross counting data are inherently of little interest, but were used when necessary to estimate the uranium depositions for some samples. Gross beta measurements reflected primarily the deposition of fallout from nuclear weapons testing that was underway at the NTS and subsequently in the South Pacific and the Soviet Union. Analytical results for the period between 1 October 1954 and 11 January 1955 were reported only in terms of total alpha activity (disintegrations per minute (dpm) per sample). These results were converted to uranium mass using the average ratio of uranium quantity to total alpha activity computed from many other paired measurements. The mean and sample standard deviation of the ratio were estimated to be 0.72 and 0.47  $\mu g U \, dpm^{-1}$ , respectively. This mean and its standard deviation of 0.66.

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Figure B1-1. Worker preparing to place gummed-film square on pedestal; photo from the files of the Environmental Measurements Laboratory courtesy of Harold Beck.

Examination of the gummed-film monitoring data is important to the Fernald Dosimetry Reconstruction Project for several reasons. First, these measurements were relatively continuous during years of highest airborne releases from the FMPC. Secondly, unlike most other environmental measurements in the 1950s and 1960s, the data provide a picture of the relationship between uranium in the environment as a function of distance from the site. Until 1972, the routine air monitoring data were collected at only four stations at the FMPC perimeter (Appendix B Part 2) whereas the gummed-film program obtained samples

at distances as great as 19 km from the center of the site. Only the rainwater data (Appendix B Part 3), recent air monitoring data (Appendix B Part 2), and the soil monitoring data (Appendix B Part 4) can provide similar insights into the spatial distribution of uranium in the environment.

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This appendix provides data that can be used for comparisons of model predictions with measurements. Comparisons for the 1960–1962 period were included in the Task 4 methodology report (Killough et al. 1993). Other comparisons over the longer period of record (1954–1964) will be included in the final Task 6 report for the project.

Another important use of the gummed film monitoring data has been verification of episodic releases; that is, releases that because of their magnitude and duration warrant special dose assessment procedures. For this project, episodic releases are defined as those that increase the composite uranium release rate by at least a factor of ten and have durations of less than ten days (Voillequé et al. 1991, Appendix K). During March 1960–March 1962, daily and weekly gummed-film samples were collected at a location north of the Health and Safety Building. These data were used to confirm an episodic release from the Pilot Plant during November 1960 (Killough et al. 1993, Appendix V). When longer sampling periods were used, the utility of the data for this purpose is limited.

The data presented in this appendix have been obtained from files of analytical data sheets that contain the results of the laboratory analyses of the gummed films (NLCO 1954–1964). The monitoring locations around the FMPC are discussed in the following section. An important aspect of the evaluation of the data was estimation of the gummed-film collection efficiency; that issue is discussed in the third section. Revised estimates of deposition are presented in the last section.

#### MONITORING LOCATIONS

Initially, deposition samples were collected at only eight locations near the perimeter of the FMPC. The program was gradually expanded until routine monthly deposition samples were obtained at about 30 locations within and around the FMPC production area. The locations are listed in Table B1-1, together with the approximate directions and distances from the center of the production area. The latter were determined from a hand-drawn map, found in the archives, that showed the 25 nearby locations and from descriptions of the locations of more distant stations. The angles and distances given in Table B1-1 are best estimates from measurements using the map and its accompanying scale. These estimates have uncertainties in position that are estimated to be about  $\pm 2-3$  degrees and  $\pm 10-20$  m. The positions of the five stations that were more than 2 km away from the plant center could not be shown on the small scale map and were estimated using another map of the Fernald area. Positional uncertainties for these locations are estimated to be  $\pm 5$  degrees and  $\pm 500$  m.

The table shows that although the locations were numbered as though they were along the compass lines, this was not exactly the case. When considered in terms of the 16-point compass sectors usually employed for meteorological dispersion calculations, a few of the locations are in sectors adjacent to those indicated by the station designation. Note that location SW-4 was actually located southeast of the facility.



Near the FMFC During 1934-1904								
Station	Distance (m)	Angle (deg)	Station	Distance (m)	Angle (deg)			
designation	from center	from north	designation	from center	from north			
N-1 <sup>c</sup>	340	6	A	220	201			
N-2	720	2	$S-1^{c}$	250	178			
N3	1010	0	S2	870	191			
N-4	12000	351	S3	1430	183			
NE-1 <sup>c</sup>	510	38	S4	7100	197			
NE-2	990	36	$SW-1^{c}$	520	219			
NE-3	1200	26	SW-3	1220	224			
$NE-4^a$	4200	72	$SW-4^{b}$	19000	155			
E-1c	320	89	$W-1^{c}$	330	262			
E-2	730	89	W-2	620	260			
E-4	15000	114	W3	1090	264			
$\mathbf{B}^{c}$	10	135	W4	12000	250			
SE-1 <sup>c</sup>	510	142	Cc	110	292			
SE-2	980	136	$NW-1^{c}$	510	319			
SE-3	1610	147	<b>NW_3</b>	1620	304			
SE-4	Unknown	Unknown						

#### Table B1-1. Approximate Locations of Deposition Measurements Near the FMPC During 1954–1964

<sup>a</sup> During 1960, this sampling station was located about 12500 m away at an angle of 73°.

<sup>b</sup> Actually located southeast of the facility center.

<sup>c</sup> Sampling location during 1954–1956.

The locations of the initial measurements were in the eight primary compass directions. Those locations (N-1, NE-1, E-1, SE-1, -1, SW-1, W-1, and NW-1 in Table B1-1) and two points within the production area were used during 1954–1956. Sampling locations at greater distances were added in 1957. The most distant locations were not added until late 1959. Figure B1-2 shows the approximate locations of the onsite and distant gummed film monitoring locations.

#### **GUMMED-FILM COLLECTION EFFICIENCY**

The collection efficiency for gummed-film is defined to be the ratio of the fallout activity collected by the gummed-film to the total amount deposited on a comparable ground surface. At the time of the measurements, the gummed-film collection efficiency was estimated to be about 60% (Harley et al. 1960). However, more recent evaluations (Beck 1984; Beck et al. 1990) indicate that the efficiency varies with precipitation amount and is substantially lower than originally thought. Comparisons of gummed-film data against integrated deposition results from soil samples in relatively arid locations near the NTS yielded an estimated efficiency of 20% for daily collections under dry conditions.



Figure B1-2. Gummed-film deposition measurement locations near (top section) and distant (lower section) from the FMPC. Locations are based upon a map and descriptions found in the FMPC archives and are approximate.



Contemporary measurements of total deposition and deposition on gummed-film, including Chernobyl fallout and field experimental data, led Beck et al. (1990) to develop revised estimates of collection efficiencies for gummed-film as a function of daily precipitation amount. Beck et al. (1990) estimated that the 1-sigma fractional uncertainty for each estimate was about  $\pm 25\%$ .

At the FMPC, gummed-film samples were not exchanged daily. The exposure period varied from about one week to about one month in the later years of the sampling program. Direct application of the collection efficiencies for one-day sampling periods appeared inappropriate. Washoff of deposited material by subsequent precipitation or blowoff by wind would be expected to further reduce the amount retained.

During a two-year period between March 1960 and March 1962, daily, weekly, biweekly, and monthly measurements of uranium deposition on gummed-film were obtained for a location on the FMPC site, just north of the Health and Safety Building. This special study was undertaken by the Industrial Hygiene and Radiation Department at the FMPC to determine the collection efficiency of the gummed-film for various exposure times and weather conditions (Starkey 1960). No report describing the results of the study has been found; however, the FMPC data files contained the measurement results. These data were analyzed to determine the gummed-film collection efficiencies for exposure periods longer than one day. The procedures used in the analysis and the results are described below.

National Weather Service daily precipitation measurements were obtained for both the Cincinnati airport, near Covington, Kentucky, and a downtown Cincinnati location (4th and Main Streets). Monthly precipitation totals at these two locations were compared with totals measured at the FMPC. From that comparison it appeared that the city location was more representative of the FMPC than the location at the airport.

The daily precipitation measurements from downtown Cincinnati were used with the daily efficiencies of Beck et al. (1990) to determine collection efficiencies for the daily gummed-film measurements and to estimate the true daily depositions at the Health and Safety Building. Those estimates were then summed for weekly, biweekly, and monthly periods for comparison with the total depositions measured for those periods. Weekly collections were compared with the sum of seven daily collections during the exposure period when the set of daily samples was complete. If only one daily value was missing, a comparison was also made. However, if two or more daily depositions were unavailable, the weekly collection was not compared with the sum of daily values. The same approach to missing data was used for the longer collection periods. Comparisons of biweekly deposition results were made with the sums of 12-14 daily values. For monthly comparisons, a maximum of four missing daily measurements was tolerated.

The results of the comparisons of longer term deposition results with sums of the estimated true daily depositions are shown in Table B1-2. Distributions of observed ratios are presented in Appendix M of Killough et al. (1993). Apparent mean collection efficiencies for the three longer exposure periods are comparable. The deposition-weighted average daily collection efficiency for the approximately two years of measurements was 0.16. This suggests that most losses occur during the day of deposition or that subsequent small losses on later days are counterbalanced by gains due to local effects or are masked.

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Gun	Gummed-Film Exposed for Longer Periods										
	Apparent										
Exposure	Number of _	collection	efficiency								
duration	comparisons	Mean	Std. dev.								
1 Week	58	0.15	0.06								
2 Weeks	31	0.16	0.06								
1 Month	15	0.14	0.03								

Table B1-2. Apparent Collection Efficiencies of

The collection efficiencies of greatest interest are those for biweekly and monthly exposure periods. Those sampling frequencies were used most often in the gummed-film monitoring program during 1954–1964. As might be expected, the variability of the ratios is smallest for the longest averaging time.

If there are losses due to weathering of material deposited on the gummed film, these results suggest that there are approximately compensating depositions, presumably due to resuspension of material from the ground surface. An alternative explanation, which can not be excluded based on these results, is that the collection efficiency of the gummed-film depends primarily on the conditions at the time of deposition and that there is little removal of material fixed at that time.

#### **REVISED URANIUM DEPOSITION ESTIMATES**

Deployment of gummed film collectors began with a limited number of sampling stations close to the facility boundary. Sampling locations at greater distances were added as the program developed. Recovery of data from the gummed-film monitoring program was generally good. Some samples, primarily at the distant locations, were not obtained routinely. These were in populated areas, so there are several possible reasons for lost samples; however, particular reasons were not given on the analysis sheets. A few samples were lost during analysis.

Revised estimates of uranium deposition on gummed-film were derived from the reported values (on analytical data sheets) and the apparent collection efficiencies given in Table B1-2. Most of the fluorometric analyses for uranium were originally reported in units of  $\mu g$  ft<sup>-2</sup>. The revised deposition density estimates are presented in metric units of mg  $m^{-2}$  and corrected for losses during the exposure period. Average deposition rates have been computed for the sampling periods on the assumption that sample changes were at approximately the same time of day (only the placement and removal dates are available). Uncertainties in the revised uranium depositions are estimated to be in the range of 20 to 30 percent of the tabulated values. Most of the uncertainty is associated with the estimates of the long-term collection efficiencies.

The following figures provide a general picture of the changes in uranium deposition rates with time during plant operation, as determined from the gummed-film measurements. The results for locations NE-1 and E-1 are plotted in Figure B1-3 while

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those for locations S-1 and W-1 are plotted in Figure B1-4. These four locations are among those with the longest period of record, more than ten years, although it can be seen that there are gaps in the record.



Location NE-1

Figure B1-3. Results of uranium deposition measurements using gummedfilm at locations NE-1 and E-1 throughout the period of monitoring. Values have been plotted as average deposition rates during the exposure period: however, not all the gummed-film exposure times are equal (see Table B1-3 below). The month of sampling is shown for every tenth sample. Blank spaces indicate that no data were collected during that period.

U deposition rate (mg per square meter per day)

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F

**Jan 63** Nov 63 Sep 64

**Mar 62** 



Location S-1



Figure B1-4. Results of uranium deposition measurements using gummedfilm at locations S-1 and W-1 throughout the period of monitoring. Values have been plotted as average deposition rates during the exposure period; however, not all the gummed-film exposure times are equal (see Table B1-3 below). The month of sampling is shown for every tenth sample. Blank spaces indicate that no data were collected during that period.

Figures B1-5 shows the estimated annual depositions at the nearest stations in the eight cardinal directions. The four primary directions are shown in the top portion of the figure and the intermediate directions in the bottom section. The plots illustrate the very high estimated depositions in 1955. It should be recognized that these estimates are based on incomplete measurment data in some years, as shown in Figures B1-3 and -4.

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Figure B1-5. Estimated annual uranium depositions at eight primary gummed-film monitoring locations between 1954 and 1964. In some cases the data are less complete and the estimates are more uncertain. The largest gaps occur in 1955 and 1959.

Figures B1-6 and -7 illustrate the observed dependence of the cumulative deposition on gummed-film with distance from the center of the Production Area. The first figure shows cumulative depositions during about seven years for locations at three distances along four directions from the plant. Measurements at greater distances were conducted for a shorter time period. Figure B1-7 contains cumulative depositions at four distances in three directions. The most distant locations in the plot are from 7 to 12 km from the facility center. The period of integration for the results in Figure B1-7 is about five years. The decrease in the cumulative deposition is not as rapid for distances beyond 2 km and the

cumulative depositions are not as low as would be expected at the more distant locations. An investigation of the possible reasons for the elevated depositions there is underway. Conclusions will be presented in the final project report.



Figure B1-6. Cumulative uranium depositions estimated from gummed-film data at three distances in four directions (north, southeast, south, and west) of the FMPC for the period August 1957—December 1964. Distances and directions of the sampling locations are shown in Table B1-1 above.

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Distance (m) from plant center

**Figure B1-7.** Cumulative uranium depositions estimated from gummed-film data at four distances in three directions (north, south, and west) of the FMPC for the period December 1959—December 1964. Distances and directions of the sampling locations are shown in Table B1-1 above.

#### SUMMARY

Data on deposition of uranium on gummed-film at locations on and around the FMPC plant site have been retrieved from the archives of analytical data sheets. Results of a special study conducted by plant staff have been used to obtain apparent collection efficiencies for gummed film for exposure periods longer than one day. The results from the analytical data sheets have been corrected for incomplete retention of uranium by the gummed-film. Revised deposition estimates have been compiled for use in model validation studies. Plots of the gummed film deposition results have been presented in this appendix to illustrate the scope of the data and general trends.

COMPILATION OF REVISED URANIUM DEPOSITION ESTIMATES

Table B1-3, in multiple parts, contains the revised uranium deposition densities for sampling locations and times for which data were found. The gummed-film data span the years from 1954 to 1964, although not all locations were sampled throughout that period. To minimize the space required for presentation, the tabulations are limited to locations for which data were found. All values in Table B1-3 have been rounded to a maximum of two significant figures.

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		<u>(Part</u>	<u>1: 5-4-54 thro</u>	ugh 8-20-54)		<u> </u>				
_	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>									
	5-4-54	5-18-54	6-7-54	6-21-54	7-9-54	8-6-54				
Station	5-18-54	6-7-54	6-21-54	7-9-54	8-6-54	8-20-54				
N-1	25	12	8	25	19	15				
NE-1	22	29	15	47	31	13				
E-1	28	480	25	10	160	33				
SE-1	57	35	8	4	41	53				
S-1	46	120	10	150	180	270				
SW-1	21	41	11	68	58	55				
W-1	22	24	25	3	41	32				
NW-1	10	8	13	9						

#### Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-1964 (Part 1: 5-4-54 through 8-20-54)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964 (Part 2: 8-20-54 through 1-25-55)

		(1 64 0		Jug 11 1 20 00)						
-	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period									
	8-20-54	9-3-54	10-1-54	12-14-54	12-28-54	1-11-55				
Station	9-3-54	10-1-54	12-14-54	12-28-54	1-11-55	1-25-55				
N-1	9	66	<b>98</b>	129	110	140				
NE-1	190	92	60	109	310	218				
E-1	68	120	180	260	930	24				
SE-1	16	74	59	489	120	500				
S-1	<b>7</b> 3	210	390	129	1300	460				
SW-1	55	54	76	23	240	120				
W-1	23	43	125	40	87	<u>63</u>				

-	Revis	ea uranium a	eposition den	sity (mg m ~)	or indicated p	erioa~
	1-25-55	2-8-55	2-22-55	3-8-55	8-24-55	9-19-55
Station	2-8-55	2-22-55	3-8-55	8-24-55	9-19-55	10-4-55
N-1	130	140	130		7900	1300
NE-1	120	270	250		2900	1500
E-1	3200	78	330		3700	6500
SE-1	180	3	220		3700	3700
S–1	790	220	1000		41000	1730
SW-1	340	260	180		4900	5600
W-1	170	110	79		2100	1500
В					3700	6900
С					3300	6000

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During
1954–1964
(Part 3: 1-25-55 through 10-4-55)

 $^{a}$  Blank spaces in the table indicate that no data were available.

		(Part 4	10-4-55 thr	ough 1-13-56	)					
_	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup><math>a</math></sup>									
	10-4-55 10-20-55 11-2-55 11-15-55 11-3-55 12-16-55									
Station	10-20-55	11-2-55	11-15-55	11-30-55	12-16-55	1-13-56				
N-1	770		88	61	120	150				
NE-1	1200	180	110	96	51	200				
E-1	860	8	210	220	260	950				
SE-1	770	20	43	43	130	410				
S1	560	180	120	150	340	1700				
SW-1	770	9	32	47	49	220				
W-1	810	52	35	130	190	270				
NW-1						150				
В	2000	2000	920		37	3500				
С	2500	180	190	79	540	1300				

# Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During1954-1964

<sup>a</sup> Blank spaces in the table indicate that no data were available.

		<u> </u>	b: 1-13-56 thro	ough 4-27-56)						
_	Revise	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period								
	1-13-56	1-30-56	2-21-56	3-6-56	3-22-56	4-10-5 <del>6</del>				
Station	1-30-56	2-21-56	3-6-56	3-22-56	4-10-56	4-27-56				
N-1	78	300	65	55	200	51				
NE-1	100	130	130	94	550	61				
E-1	160	91	150	460	510	1300				
SE-1	150	130	41	69	200	340				
S-1	1300	1300	200	380	1100	1100				
SW-1	260	380	57	<del>99</del>	240	120				
W-1	170	130	94	75	220	120				
NW-1	ଷ	45	41	80	82	32				
В	1600	2200	510	1200	1100	3200				
<u>C</u>	340	230	210	300	890	240				

#### Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964 (Part 5: 1-13-56 through 4-27-56)

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-1964 (Part 6: 4-27-56 through 8-15-56)

			arco21	vo un ough	01000/				
	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period								
	4-27-56	5-10-56	5-23-56	6-7-56	6-19-56	7-9-56	7-24-56		
Station	5-10-56	5-23-56	6-7-56	6- <u>19-56</u>	7-9-56	7-24-56	8-15-56		
N-1	49	95	37	76	76	100	160		
NE-1	110	300	57	76	190	330	160 ·		
E-1	67	280	120	240	620	1500	500		
SE-1	73	230	95	65	76	160	140		
S-1	320	1700	300	400	590	380	470		
SW-1	21	150	86	95	160	46	100		
W-1	58	100	76	110	95	100	86		
NW-1	25	34	150	62	44	29	52		
В	750	1100	680	480	1800	1200	1400		
C	240	280	210	320	360	420	350		

			. 0 10 00 440	APIL IN BO OO					
-	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>								
	8-15-56	8-29-56	9-17-56	9-25-56	10-25-56	11-27-56			
Station	8-29-56	9-17-56	9-25-56	10-25-56	11-27-56	12-26-56			
N-1	74	73	110	150	180	94			
NE-1	97	130	67	350	260	170			
E-1	97	150	82	290	220	360			
SE-1	78	90	250	270	600	67			
S-1	480	530	200	1800	1000	580			
SW-1	51	220	50	600	170	170			
W-1	84	110	74	360	170	56			
NW-1-	38	510	43	256	<del>59</del>				
В	480	590	780	3100	750	540			
С	540	500	420	7900	660	430			

#### Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-1964 (Part 7: 8-15-56 through 12-26-56)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

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		(Part 8	<u>s 12-26-56 thr</u>	ougn 0-28-57)					
	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>								
-	12-26-56	1-29-57	2-22-57	3-26-57	4-25-57	5-29-57			
_Station	1-29-57	2-22-57	3-26-57	4-25-57	5-29-57	6-28-57			
N-1	94	66	5 <del>9</del>	160					
N-2		-				42			
N-3						65			
NE-1	94	100	260	<del>99</del>		170			
NE-2						18			
NE-3					•	72			
E-1	150	950	430	160		120			
E-2						100			
SE-1	150	150	1300	120		78			
SE-2						18			
SE-3						16			
S-1	800	1500	110	65		170			
S–3						14			
SW-1	190		1700	180		160			
SW-3						30			
W-1	590	74	76	190		9			
W-2	•					120			
W-3						18			
NW-1	37	39	150	78		39			
NW-3						9			
Α						37			
В	890	1100	470	1800		630			
С	450	430	1700	690		310			

# Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964(Part 8: 12-26-56 through 6-28-57)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>							
	6-28-57	8-1-57	8-13-57	8-23-57	8-30-57	9-6-57		
	8-1-57	8-13-57	8-23-57	8-30-57	9-6-57	9-13-57		
N-1	110	30	10	28	4	32		
N-2		15	5	15	4	9		
N-3		11	5	10	2	8		
NE-1		37	11	33	9	24		
NE-2	65	55	_ 2	44				
NE-3	31	20	7	20				
E-1	190	65	17	70	28	31		
E-2		33	15	25	12	2		
SE-1	130	9	37	33		9		
SE-2	39	31	13	16		11		
SE–3	220	6	8	3	8	4		
S-1	280	54	5	90		55		
S3		17	13	4	12	13		
SW-1	120	46	30	54	37	62		
SW-3	110	24	22	18	5	22		
W-1	140	33	82	35	<i>7</i> 5	62		
W-2	160	24	41	35	35	64		
W-3	21	9	· 4	12		20		
NW-1	44	15	6	18	5	16		
NW-3	21	8	8	24	2	11		
Α	480	180	160	310	210	120		
В	1600	13	240	320	75	305		
С	170	13	140	220	640	180		

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-1	<b>964</b>
(Part 9: 6-28-57 through 9-13-57)	

<sup>a</sup> Blank spaces in the table indicate that no data were available.

		(Farti	N. 2-19-91 HI	Uugii 2-24-00)		
-	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>					
	9-13-57	8-30-57	9-30-57	10-31-57	11-29-57	1-15-58
Station	9-30-57	9-30-57	10-31-57	11-29-57	1-15-58	2-24-58
N-1	75	130	260		490	73
N-2	16	62	œ	45	76	32
N-3	13	26	52	27	83	12
NE-1	90	95	31	180	490	160
NE-2	46	64	48	80	150	61
NE-3	24	24	41	18	76	41
E-1	61	110		320	240	420
E-2	39	76	140	80	130	160
SE-1	39	98	93	80	180	160
SE-2	22	10	48	13	87	69
SE–3	10	31	14	4	40	20
S-1	86	130	220	60		1300
S–3	22	38	10	4	29	21
SW-1	135	400	240	80	76	100
SW-3	67	110	74	65	170	18
W-1	<del>64</del>	270	110	160	260	
W-2	55	150	220	22	1100	150
W-3	9	31	5	98	94	12
NW-1	20	48	24	45	43	49
NW-3	10	10	10	80	65	6
Α	120	230	1300	320	2300	1100
В	190	340	2700	950	1000	2000
С	360	150	850	3500	1700	1100

Table B1–3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964
(Part 10: 9-13-57 through 2-24-58)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

	Revised uranium deposition density (mg $m^{-2}$ ) for indicated period <sup>a</sup>					
-	2-24-58	3-21-58	4-22-58	5-21-58	6-6-58	6-19-58
Station	3-21-58	4-22-58	5-21-58	6-19-58	6-19-58	7-21-58
N-1	23	76	54	64		250
N-2	3	28	23	41		81
N-3	7		18	17		71
NE-1	<b>59</b>	76	150	100		340
NE-2		22	77			179
NE-3	11	28	69	34		109
E-1	78	240	100	170		560
E-2	16	76	220	80		220
SE-1	120	340	110	100		200
SE-2	28	30	160	40		73
SE3	14	11	9	15		<b>4</b> 0
S-1	1200	790	1200	540		650
S-3	12	43	39		9	32
SW-1	980	150	9	64		89
SW-3	98	76	170	18	8	12
W-1	<b>59</b>	170	140	54		73
W-2	120	48	120	40		89
W-3	10	19	14			24
NW-1	15	45	92	26		30
NW-3	5	35	33	22	10	14
Α	590		540	720		620
В	160		1300	400		880
С	3300		1200	<b>280</b>		1000

Table B1–3. Revised Estimates of Uraniu	m Deposition Near the FMPC During 1954–1964
(Part 11: 2-24	1-58 through 7-21-58)

<sup>a</sup> Blank spaces in the table indicate that no data were available.
	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>						
	7-21-58	8-25-58	9-24-58	10-23-58	11-19-58	12-16-58	
Station	8-25-58	9-24-58	10-23-58	11-19-58	12-16-58	1-5-59	
N-1	180	97	160	54	76	56	
N-2	100	41	39	49	19	21	
N-3	80	46	25	36	14	26	
NE-1	240	140	140	150	46	210	
NE-2	100	30	34	56	37	21	
NE-3	60	41	<del>99</del>	34	16	50	
E-1	460	290	340	410	450	150	
E-2	200	140	<del>99</del>	190	78	ស	
SE-1	160	210	140	56	12	450	
SE-2	57	45	<del>99</del>	22	23	50	
SE3	28	16	19	8	51	. 38	
S-1	1000	330		370	46	110	
S-2				14	40	75	
S-3	41	13	29	5	120	56	
SW-1	180	97	300	23	59	56	
SW-3	60	78	67	5	116	39	
W-1	280	120	240	37	79	24	
W-2	140	96	200	26	56	32	
W-3	47	260	11	8	16	13	
NW-1	61	51	<del>59</del>	31	20	30	
NW-3	12	11	13	280	9	15	
Α	2400	720		9	41	150	
В	1400	1320	2600	1100	54	410	
С	1200	41	1200	540	160	280	

Fable B1–3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964
(Part 12: 7-21-58 through 1-5-59)

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<sup>a</sup> Blank spaces in the table indicate that no data were available.

	Revised u	ranium deposit	ion density (m	g m <sup>-2</sup> ) for indica	ted period <sup>a</sup>
	1-5-59	2-2-59	3-2-59	3-24-59	4-7-59
Station	2-2-59	3-2-59	4-7-59	4-22-59	5-6-59
N-1	56	74	150		92
N-2	32	28	48		43
N-3	45	17	48		130
N-4				5	
NE-1	75	50	170		92
NE-2	58	53	68		73
NE-3	26	44	44		37
E-1	189	260	210		160
E-2	37	94	75		92
SE-1	15	74	130		71
SE-2	32	64	32		110
SE-3	75	27	13		24
S-1	470	580	300		380
S-2	41	53	34		22
S-3	19	8	27		18
S-4				12	
SW-1	94	31	62		200
SW-3	32	31	41		55
W-1	94	110	110		15
W-2	56	33	60		75
W-3	8	24	52		33
W-4				9	
NW-1	49	29	41		36
NW-3	15	69	21		20
Α	870	430	450		810
В	550	500	1200		750
С	960	820	1400		2000

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-1964
(Part 13: 1-5-59 through 5-6-59)

·····	Revised u	ranium deposit	ion density (mg	(m <sup>-2</sup> ) for indica	ted period <sup>a</sup>
-	4-22-59	5-6-59	5-26-59	6-9-59	6-29-59
Station	5-26-59	6-9-59	6-29-59	7-9-59	7-10-59
N-1		76		71	
N-2		57		24	
N-3		52		19	
N-4	3		3		8
NE-1		57		76	
NE-2		34		37	
NE-3		24		95	
E-1		76		380	
E-2		76		110	
E-4	5		1		4
SE-1		35		7	
SE-2		58		90	
SE-3		26		130	
S-1		630		11	
S-2		26		11	
S–3		34		1500	
S-4	8				3
SW-1		110		57	
SW-3		47		31	
W-1		93		150	
W-2		120		75	
W-3		28		17	
W-4	4				2
NW-1		65		55	
NW-3		17		22	
Α		400		1400	
В		610		1400	
C		95		656	

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-19	964
(Part 14: 4-22-59 through 7-10-59)	

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		(Fart 15: 7-9-5	5 through 12-14-	-9 C · 1			
Revised uranium deposition density (mg m 2) for indicated period							
	7-9-59	8-11-59	9-11-59	10-12-59	11-13-59		
Station	8-11-59	9-11-59	10-12-59	11-13-59	12-14-59		
N-1	71	75	220	270	89		
N-2	38	37	73	48	25		
N-3	37	22	73	36	34		
NE-1	190	190	290	110	80		
NE-2	95	110	24	94	54		
NE-3	80	57	73	58	31		
E-1	530	6600	440	890	230		
E-2	120	<i>7</i> 5	180	280	260		
SE-1	140	55	260	220	66		
SE-2	40	55	91	58	48		
SE-3	24	58	22	24	17		
S-1	720	270		830	480		
S-2	57	44	73	33	20		
S-3	31	42	22	30	17		
SW-1	75	150	310	150	34		
SW-3	43	37	51	55	17		
W-1	280	220	380	280	28		
W-2	74	91	550	160	51		
W-3	30	49	40	33	16		
NW-1	57	22	49	82	28		
NW-3	18	860	13	21	15		
Α	1000	70	860	1600	840		
В	1100	2000	1300	1800	840		
С	1300	730	2000	1400	1100		

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964
(Part 15: 7-9-59 through 12-14-59)

-	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>						
	12-14-59	1-14-60	2-15-60	3-17-60	4-1-60	5-2-60	
Station	1-14-60	2-15-60	3-17-60	4-1-60	5-2-60	6-1-60	
N-1	78	110	22	200	280	170	
N-2	30	33	17	53	74	68	
N-3	18	22	11	51	49	44	
N-4	6	15	8	10	13	15	
NE-1	120	78	35	49	150	76	
NE-2	52	37	20	32	76	61	
NE-3	20	29	20	21	44	34	
NE-4			12	2	2	1	
E-1	390	1200	1200	200	1000	400	
E-2	120	150	95	70	190	130	
E-4	3	2	1	1	3	2	
SE-1	98	110	60	<b>51</b> ·	550	440	
SE–2	22	37	17	32	28	ങ	
SE-3	24	21	32	21	13	36	
S-1	730	880	9	340	740	380	
S-2	24	100	610	170	38	21	
S–3	38	36	20	7	12	13	
S-4	5	17	6	12	9	14	
SW-1	84	140	130	17	190	72	
SW-3	-54	32	28	7	95	27	
SW-4		21	53	15	12	15	
W-1	98	160	130	78	250	190	
W-2	22	100	140		83	150	
W-3	8	30	26	15	19	47	
W-4	4	20	7	8	11	24	
NW-1	42	30	11	28	47	320	
NW-3	15	17	15	7	17	23	
Α	900	1200	1100	740	1000	1300	
В	1500	1400	830	910	1400	850	
С	900	500	640	4900	2000	1400	

## Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964 (Part 16: 12-14-59 through 6-1-60)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

	Revise	Revised uranium deposition density (mg $m^{-2}$ ) for indicated period <sup>a</sup>						
-	6-1-60	7-1-60	8-2-60	9-1-60	9-30-60	11-1-60		
Station	7-1-60	8-2-60	9-1-60	9-30-60	11-1-60	12-1-60		
N-1	170	320	170	110	130	700		
N-2	66	40	59	44	53	170		
N-3	55	44	<del>59</del>	36	36	190		
N-4	21		14	19	17	29		
NE-1	110	76	130	49	320	340		
NE-2	66	57	95	44	170	150		
NE-3	40	61	61	49	76	110		
NE-4		3						
E-1	930	510	570	360	490	2700		
E-2	110	130	95	110	210	360		
E-4	3	2	1			. 4		
SE-1	76	320	170	170	360	780		
SE–2	25	190	81	76	130	320		
SE–3	21	40	42	42	40	170		
S-1	590	1400	820	1000	400	1500		
S-2	53	91	95	130	95	210		
S3	28	47	40	55	460	61		
S-4	27	14	25	14	17	14		
SW-1	280	250	510	320	130	490		
SW-3	72	70	170	78	280	57		
SW-4		23	14	22	23	15		
W-1	260	620	260	590	110	930		
W-2	340	800	190	170	170	210		
W-3	95	70	59	40	28	30		
W-4	23	20	25	14	17	41		
NW-1	<b>59</b>	170	95	72	76	230		
NW-3	21	25	21	27	19	ស		
A	850	2000	1200	1400	3000	3300		
В	850	3000	1100	890	660	2000		
С	1300	2300	1600	700	2600	3500		

## Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964 (Part 17: 6-1-60 through 12-1-60)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

	Revise	Revised uranium deposition density (mg $m^{-2}$ ) for indicated period <sup>a</sup>					
-	12-1-60	1-5-61	2-14-61	3-15-61	4-18-61	5-19-61	
Station	1-5-61	2-14-61	3-15-61	4-18-61	5-19-61	6-1-61	
N-1	210	95	320	81	130	170	
N-2	ഒ	51	70	30	23	21	
N-3	68	44	45	32	25	28	
N-4	19	11	14	9	25	11	
NE-1	210	110	ങ	55	81	36	
NE-2	170	42	55	44	190	55	
NE-3	76	21	30	30	19	36	
NE-4				53	20	13	
E-1	760	1500	1100	680	760	17	
E-2	190	120	78	130	72	61	
E-4	3		3		4		
SE-1	120	260	110	280	190	95	
SE–2	160	100	45	133	55	34	
SE-3	130	51	23	40	25	21	
S-1	1000	1500	700	1300	800	360	
S-2	<b>7</b> 8	63	78	57	<del>59</del>	36	
S-3	70	32	21	23	21	47	
S-4	45	17	14	9	20	16	
SW-1	280	380	150	210	130	74	
SW-3	72	95	28	55	32	32	
SW-4	35	17	13	13	13	16	
W-1				210	210	70	
W-2	150	190	110	170	150	120	
W-3	55	53	28	47	27	21	
W-4	28	13	16	11	10	18	
NW-1	95	36	34	34	64	47	
NW-3	32	10	23	34	23	32	
Α	1800	2100	980	1800	1100	210	
В	970	1100	660	1000	570	250	
C	2300	1200	1800	610	620	230	

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-196	4
(Part 18: 12-1-60 through 6-1-61)	

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	Revised uranium deposition density (mg m <sup>-2</sup> ) for indicated period <sup>a</sup>						
	6-1-61	6-30-61	7-31-61	9-1-61	10-1-61	11-1-61	
Station	6-30-61	7-31-61	9-1-61	10-1-61	<u>11-</u> 1-61	12-1-61	
N-1	250	170	170	32	170	120	
N-2	55	130	45	140	36	44	
N-3	45	63	45	36	36	30	
N-4	17	53	<b>68</b>	40	27	11	
NE-1		152	85	63	110	89	
NE-2	260	89	5 <del>9</del>	55	55	40	
NE-3	74	66	44	34	36	28	
NE-4	30	68	14	11	31	11	
E-1	1800	590	700	530	610	470	
E-2	170	91	110	95	95	130	
SE-1	0	76	360	30	91	150	
SE–2	ଷ	36	66	63	36	47	
SE–3	55	34	32	380	19	19	
S-1	470	400	780	13	360	1000	
S-2	40	85	ស	25	30	55	
S3	36	34	42	17	13	25	
S-4	19	11	16	25	22	21	
SW-1		89	190	70	440	140	
SW-3	32	49	53	52	30	51	
SW-4	15	8	11	17	9	7	
W-1	95	130	210	120	85	170	
W-2	66	53	30	57	17	93	
W-3	36	70	78	17	5	28	
W-4	13	19	28	11	200	10	
NW-1		51	110	19	51	74	
NW-3	32	32	38	63	25	170	
Α	210	400	890	530	320	1200	
В	250	700	980	550	470	660	
C	230	1200	740	830	700		

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 195	1-1964
(Part 19: 6-1-61 through 12-1-61)	

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	Revised uranium denosition density (mg $m^{-2}$ ) for indicated period <sup>a</sup>							
-	12-1-61	12-29-61	2-5-62	3-1-62	4-4-62	5-2-62		
Station	12-29-61	2-5-62	3-1-62	4-4-62	5-2-62	6-1-62		
N-1	61	87	57	25	470	320		
N-2	8	21	21	110	80	64		
N-3	25	19	21	83	47	57		
N-4	17	0	14	34	10	13		
NE-1	30	130	32	21	230	170		
NE-2	28	49	21	28	89	63		
NE-3	25	32	89	21	53	44		
NE-4	10	16	12	13	37	8		
E-1	170	460	170	260	460	470		
E-2	78	97	47	120	230	150		
SE-1	66	5 <del>9</del>	110	230	260	210		
SE-2	28	23	44	44	110	32		
SE-3	17	21	17	53	57	42		
S-1	460	420	470	890	970	720		
S-2	21	47	30	49	28	44		
S-3	38	74	21	21	21	19		
S-4	11	0	11	10	10	17		
SW-1	47	<del>99</del>	120	510	190	280		
SW-3	64	25	34	27	68	49		
SW-4	7	0	8	14	8	10		
W-1	78	80	320	150	170	280		
W-2	61	72	66	80	280	190		
W-3	27	19	17	27	76	36		
W-4	5	0	9	45	8	25		
NW-1	30	36	28	36	100	89		
NW-3	7	93	17	6	25	47		
Α	660	550	760	1800	1300	3200		
B	420	780	510	970	850	1600		
<u> </u>	360	620	230	1100	1100	1300		

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 195	54-1964
(Part 20: 12-1-61 through 6-1-62)	

<sup>a</sup> Blank spaces in the table indicate that no data were available.

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	Revised uranium deposition density (mg $m^{-2}$ ) for indicated period							
_	6-1-62	6-27-62	8-1-62	9-4-62	10-1-62	11-1-62	12-3-62	
Station _	6-27-62	8-1-62	9-4-62	10-1-62	11-1-62	12-3-62	12-31-62	
N-1	340	87	470	330	260	160	240	
N-2	63	18	90	62	52	52	71	
N-3	78	20	76	66	43	57	52	
N-4	28	13	9	10	8	10	4	
NE-1	170	64	220	160	210	160	190	
NE-2	74	42	38	120	100	71	62	
NE-3	44	30	76	76	90	47	62	
NE-4	35	13	28	14	71	7	11	
E-1	380	220	380	570	950	220	900	
E-2	320	110	180	220	210	95	200	
SE-1	280	<b>99</b>	240	380	140	220	340	
SE-2	110	38	81	120	62	81	110	
SE3	130	19	25	81	28	43	43	
S-1	1300	460	1100	2200	900	1000	1500	
S-2	36	17	71	170	52	76	95	
S–3	66	8	24	52	28	23	430	
S-4	31	11	9	12	8	5	4	
SW-1	640	49	420	910	140	950	45	
SW-3	110	30	76	200	95	120	140	
SW-4	18	9	5	18	18	10	5	
W-1	230	120	13000	660	160	230	290	
W-2	100	64	3900	430	81	85	190	
W-3	36	9	1600	71	28	25	71	
W-4	110	14	12	6	9	9	4	
NW-1	85	95	210	110	95	49	130	
NW-3	34	17	300	22	66	12	17	
Α	3300	85	1700	3800	1200	2100	2000	
В	1100	570	1100	2300	1400	4400	4500	
C	3800	26	1900	2300	1100	900	1000	

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 19	54-1964
(Part 21: 6-1-62 through 12-31-62)	

	Revised uranium deposition density (mg $m^{-2}$ ) for indicated period <sup>a</sup>						
	12-31-62	1-31-63	2-28-63	4-1-63	5-1-63	5-31-63	
Station	1-31-63	2-28-63	4-1-63	5-1-63	5-31-63	6-28-63	
N-1	96	140	340	480	1700	1500	
N-2	240	53	53	67	96	180	
N-3	58	53	38	48	57	110	
N-4	10	25	12		6	14	
NE-1	350	160	250	140	380	580	
NE-2	200	58	190	370	240	67	
NE-3	110	34	72	86	17	110	
NE-4	20	53	14	13	25	20	
E-1	1100	274	380	480	270	620	
E-2	380	960	100	140	160	430	
E-4		16	10		2	12	
SE-1	260	180	260	380	180	460	
SE-2	100	48	82	82	72	150	
SE-3	58	25	28	67	53	82	
S-1	2200	160	960	1300	1100	1300	
S-2	280	1800	53	19	82	82	
S–3	21	29	19	58	19	29	
S-4	13	17	220	20	21	10	
SW-1	690	65	58	770	290	540	
SW-3	220	36	53	180	21	140	
SW-4	9	13	6	27	7	15	
W-1	380	160	72	320	430	670	
W-2	190		140	150	210	430	
W-3	72	24	48	67	63	82	
W-4	7	5	14		12	17	
NW-1	110	50	220	770	170	380	
NW-3	36	25	23	17	86	35	
Α	2400	1300	2200	2500	2600	1800	
В	13000	2100	1700	1900	2000	2300	
C	1100	770	1800	1300	1700	1800	

Table B1-3. Revised Estim	nates of Uranium Deposition Near the FMPC During 195	4-1964
	(Part 22: 12-31-62 through 6-28-63)	

	Revised	uranium de	position dens	ity (mg m <sup>-2</sup> ) f	for indicated	perioda
	6-28-63	8-5-63	8-30-63	9-27-63	10-30-63	11-29-63
Station	8-5-63	8-30-63	9-27-63	10-30-63	11-29-63	12-30-63
N-1	240	820	380	480	620	100
N-2	85	180	43	69	220	58
N-3	72	85	38	62	160	46
N-4	15	17	15	· 46	5	4
NE-1	150	350	270	220	680	200
NE-2	120	150	100	110	220	110
NE-3	72	62	31	31	120	46
NE-4	11	12	15	31	26	15
E-1	620	620	770	1300	1300	580
E-2	210	200	280	200	480	340
E-4	6		15	. 8	2	4
SE-1	85	230	430	540	580	250
SE-2	85	120	120	250	190	46
SE-3	28	54	62	54	48	12
S-1	720	960	2200	1700	1800	920
S-2	62	85	110	180	54	12
S-3	58	26	31	23	14	7
S-4	8	48	15	7	5	6
SW-1	210	230	720	690	100	180
SW-3	67	85	180	62	58	38
SW-4	14	10	8	7	4	12
W-1	200	370	46	620	240	120
W-2	120	92	220	220	85	73
W-3	25	220	23	62	31	19
W-4	11	18	15	15	7	8
NW-1	110	280	150	170	160	46
NW-3	27	23	7	31	17	38
А	1100	2600	5800	7000	3800	1600
В	1300	1100	1500	2700	2000	1200
С	1400	1100	1300	3400	1900	720

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964
(Part 23: 6-28-63 through 12-30-63)

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, <del>_ ,</del>	Revised	uranium de	position dens	ity (mg m <sup>-2</sup> ) f	for indicated	perioda
	12-30-63	1-31-64	2-29-64	3-30-64	4-29-64	5-27-64
Station	1-31-64	2-29-64	3-30-64	4-29-64	5-27-64	7-1-64
N-1	430	410	480	320	580	580
N-2	170	480	110	100	140	100
N-3	100	100	100	72	110	77
N-4	7	3	6	9	5	13
NE-1	460	320	240	180	380	360
NE-2	210	120	110	120	170	170
NE-3	120	68	58	62	62	130
NE-4	23	14	11	11	15	15
E-1	920		480	<b>58</b> 0 <sup>′</sup>	1300	1000
E-2	480	620	170	210	170	280
E-4	3	5	3	3	6	9
SE-1	240	230	210	65	170	480
SE-2	<b>69</b> .	300	85	85	58	120
SE-3	23	85	14	14	18	25
S-1	1300	3400	1100	720	920	1300
S-2	69	180	18	19	58	380
S-3	38	92	22	12	17	19
S-4	12	6	8	10	4	4
SW-1	250	880	160	240	180	720
SW-3	31	260	62	62	77	140
SW-4	62	4	15	7	5	11
W-1	430	480	480	400	430	2200
W-2	120	120	180	92	85	300
W-3	69	150	62	48	23	85
W-4	17	8	8	5	7	72
NW-1	150	230	120	140	420	220
NW-3	31	33	17	18	25	. 21
Α	1500	9200	2700	2500	1500	3400
В	2000	3500	1300	1100	2800	3100
С	2700	2200	2300	3700	3700	770

### Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954–1964 (Part 24: 12-30-63 through 7-1-64)

<sup>a</sup> Blank spaces in the table indicate that no data were available.

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	Revised uranium deposition density (mg $m^{-2}$ ) for indicated period <sup>a</sup>						
	7-1-64	8-3-64	9-1-64	9-30-64	10-30-64	12-2-64	
Station	8-3-64	9-1-64	9-30-64	10-30-64	12-2-64	12-30-64	
N-1	140	350	480	190	250	180	
N-2	14	58	120	53	72	53	
N-3	30	34	85	43	62	15	
N-4	3	4	4	4	7	4	
NE-1	130	280	250	150	180	120	
NE-2	53	72	140	92	85	85	
NE-3	23	53	62	43	53	28	
NE-4	14	8	250	8	16	5	
E-1	620	1100	770	480	1000	280	
E-2	160	180	250	220	250	110	
E-4	25	1	6	1	1	1	
SE-1	130	220	200	310	180	58	
SE-2	62	85	77	68	92	15	
SE-3	23	48	16	8	30	14	
S-1	680	1500	430	.1800	580	770	
S-2	100	92	100	120	170	22	
S3	26	30	26	43	43	10	
S-4	7	6	15	6	10	4	
SW-1	460	380	580	1300	810	120	
SW-3	180	110	110	580		14	
SW-4	3	6	16	6	8	3	
W-1	370	280	1200	180	250	150	
W-2	140	100	200	160	190	48	
W-3	39	23	53	. 30	23	15	
W-4	3	8	10	4	10	5	
NW-1	50	73	110	54	85	50	
NW-3	11	5	33	10 .	13	13	
Α	14	2400	530	3500	1300	620	
В	1200	2200	2500	1100	920	530	
C	1200	2500	3900	1800	1500	1200	

Table B1-3. Revised Estimates of Uranium Deposition Near the FMPC During 1954-1964
(Part 25: 7-1-64 through 12-30-64)

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### APPENDIX B - REGIONAL ENVIRONMENTAL MONITORING

## PART 2 - AIR MONITORING DATA

#### INTRODUCTION

Examination of historic air monitoring data around the Fernald site is important to the verification of release estimates and model predictions for the dose reconstruction project. The air monitoring data can provide measurements to compare with environmental transport model predictions, and can assist in choosing appropriate models (e.g. of building wake effects, Appendix J, Killough et al. 1993). The environmental monitoring data can provide one way of investigating possible episodic releases which may have been unmonitored or undetected at the release points.

Although the environmental monitoring data are important to consider in developing methods for dose reconstruction, they are not complete enough, either temporally or spatially, to rely on exclusively for assessment of the exposure to surrounding populations from FMPC effluents. Rather, these data are used primarily to provide a quality check of the source term estimates and to calibrate or validate the transport models.

Appendix L of Killough et al. (1993) focused on air monitoring data from the early 1960s, in support of the model simulations performed for this time period. The complete set of air monitoring data is included in this Task 5 report. Summary tables and figures are included in the main body of the text; detailed data tables are included as an annex. Following a description of the air monitoring program and data summaries, factors affecting the quality of the measurements are discussed.

## DESCRIPTION OF MONITORING PROCEDURES AND SUMMARY PRESENTATIONS OF DATA

From the earliest years of operation, ambient air around the FMPC was sampled and analyzed for uranium. The amount and quality of data available have improved over the years. Air samples were obtained at the FMPC perimeter through 1971, at which time boundary stations were established (Figure B2-1). The objectives of the early perimeter air sampling program, as gathered from examination of historical memos and monthly reports, were twofold: 1) to determine the amount of uranium dust leaving the plant and 2) to compare the uranium concentration in air with the maximum permissible concentration in the National Bureau of Standards Handbook 69 (NBS 1959). The latter objective was met in quarterly and annual reports of the monitoring data. Assessments of the amount of uranium leaving the plant were made generally in a qualitative way and were normally stated to be "small".

The basic air monitoring technique was to draw a known volume of air through a filter and to measure the amount of uranium collected by the filter. For the dose reconstruction project, the measurements of uranium in air around the FMPC have been transcribed into



Figure B2-1. Ambient air sampling locations around the FMPC.

computer spreadsheet format directly from the original Analytical Data Sheets (NLCO 1953-1984). These data sheets record:

- location of the air sample
- sampling flow rate (m<sup>3</sup> min<sup>-1</sup>)
- sampling duration (min)
- total volume of air sampled (m<sup>3</sup>)
- total U on the filter (µg)
- uranium concentration in air ( $\mu$ Ci mL<sup>-1</sup>)
- alpha activity concentration in air (µCi mL<sup>-1</sup>)
- beta activity concentration in air ( $\mu$ Ci mL<sup>-1</sup>)
- uncertainty on the alpha concentration at the 95% confidence level

The U mass on the air filter was determined by the FMPC Analytical Department using the fluorimetric analytical method. Uranyl salts will absorb energy from ultraviolet light and release the absorbed energy as a yellow-green fluorescence. Numerous methods have been used for uranium analysis (Minczewski 1963), but until relatively recently, fluorimetry has been most frequently adopted for routine analysis of samples containing very small quantities of uranium (usually 0.001 to 10  $\mu$ g U). The procedure for analysis of uranium by the fluorimetric method at the FMPC laboratory is described in Boback (1960) and Dugan (1971). The uranium measurements are much more useful for our purposes than the gross alpha or beta analyses, which are strongly influenced by global fallout contributions.

The analytical measurement of  $\mu g$  U per filter was converted to  $\mu$ Ci U mL<sup>-1</sup> by multiplying by a Ci g<sup>-1</sup> ratio and dividing by the volume of air sampled. There were different activity-to-mass ratios used during the FMPC operating history, depending on whether the activity of the short-lived decay products are included with the <sup>238</sup>U activity. Until 1972 and after 1983, the ratio used on the analytical data sheets is  $6.8 \times 10^{-7}$ . Between 1972 and 1983, the ratio used was  $3.3 \times 10^{-7}$  Ci g<sup>-1</sup>, which would only represent the <sup>238</sup>U activity. In the detailed data tables in the Annex of this report, we have transcribed the activity concentrations directly from the analytical data sheets. On some of the summary tables and figures, however, we have standardized the results to a common basis. In all cases the activity-to-mass ratio is noted, to avoid misinterpretation.

Uranium concentrations in this part of Appendix B are expressed in femtocuries per cubic meter of air. A femtocurie (fCi) is  $1 \times 10^{-15}$  Ci, which is equivalent to 0.001 picocurie (pCi) or 1000 attocurie (aCi). The attocurie (aCi) is the unit used to express background concentrations of uranium in air in Appendix A.

#### Perimeter Air Monitoring (1953–1971)

There were a very limited number of samples of "out-plant air" during 1953-1957. Those at the FMPC perimeter were taken in the open air at up to seven guard towers and the south gate house. Because of the importance of this period to the total dose reconstruction, we have examined these data in some detail; however, their utility is probably limited, for the reasons summarized below:

- Prior to August 1957, all air monitoring at FMPC perimeter guard towers was done with low volume samplers (0.02 m<sup>3</sup> min<sup>-1</sup>) for a very short period of time (15-60 min), resulting in a small total volume of air sampled. However, there was replication (2-5 replicates per location/time).
- With the exception of two samples of >24 hours each in March 1954, all perimeter air samples taken before August 1957 were analyzed for gross alpha, not specifically for uranium.
- Because of the low total volume of air sampled (generally less than 1 m<sup>3</sup>), the alpha count rate was very low (0-10 dpm per sample).
- Conversion from measured gross alpha concentrations to estimated uranium concentrations is subject to large uncertainty. The regression between gross alpha and µg U for air samples having less than 100 dpm alpha per sample shows much scatter compared with samples with higher activity levels (Figures B2-2 and B2-3).
- The percentage of the year sampled was very low in the early years, and the sampling intervals throughout the year were irregular (Table B2-1).

	Different lime Periods	
Time Period (location)	Percentage of Year Encompassed by Air Monitoring	Months per Year When No Sampling Occurred
1953–1957 (perimeter)	<1%	6–11
1958–1960 (perimeter)	15-19%	4–5
1961–1971 (perimeter)	28-48%	0
1972–present (boundary)	continuous	0

Table B2-1. Extent of Air Monitoring at the FMPC Perimeter or Boundary During Different Time Periods

Table B2-2 summarizes the results of the very limited perimeter sampling for the years 1953 through 1957. The estimated uranium concentrations were mainly determined from the regression equation shown on Figure B2-3, and are subject to such large uncertainty that quantitative use of these data is discouraged.

Beginning in 1958, perimeter air samples were routinely analyzed specifically for uranium; however, the sampling frequency was still less than 20% of the year until 1961 (Table B2-1). Dodd (1958a) indicated that protective covers were being constructed for the perimeter samplers and that "more frequent sampling and more reliable data will be available as soon as these are put in use." The perimeter guard towers were removed in late 1959, and four permanent, wooden louvered-sided instrument shelters for the air samplers were completed by 22 April 1960 (Quigley 1960, 1961). Appendix B — Part 2 Air Monitoring Data



Figure B2-2. Uranium versus gross alpha measurements for 149 out-plant air filters in 1957–1959.



Figure B2-3. Uranium versus gross alpha measurements for 43 out-plant air filters from 1957–1959 which counted <100 dpm alpha. One extreme outlier was deleted. Regression equation is Y ( $\mu$ g U) = 0.80 X (dpm alpha) – 2.9, with R<sup>2</sup> = 0.45.

	Number of	-	
Year	Days Sampled	Estimated Uranium Co	ncentration (fCi $m^{-3}$ )
		Arithmetic Average of All	
		Measurements at Perimeter Guard Towers <sup>b</sup>	Maximum Individual Measurement
1953	12	<200	2900
1954	2	1600	3200
1955	6	1400	13,000 <sup>c</sup>
1956	6	<200	1000
1957	6	1600	25,000 <sup>d</sup>

# Table B2-2. Estimates of Uranium Concentrations in Air at the FMPC Perimeter for 1953–1957<sup>a</sup>

<sup>a</sup>Determined by conversion from gross alpha measurements, except for 1954 and August-October 1957, which were specific U analyses.

<sup>b</sup>For determination of averages, a measurement of <200 was set equal to 100. <sup>c</sup>July 20, 1955.

<sup>d</sup>Specific uranium measurement (colorimetric method) at NE guard tower on August 30, 1957. Measurement on same day at the east guard tower was 17,000 fCi m<sup>-3</sup>.

Beginning in May 1960, perimeter air samples were taken generally at a frequency of one week. However, the air was not continuously sampled. A typical sampling period was 3360 min (56 hours), or 33% of the week. One primary reason for the discontinuous sampling was that at these relatively high flow rates, the filters (approximately 4 inches in diameter) would load up with dust after several days, resulting in frequent pump failures. For two weeks in October 1960, a continuous air sampler (manufacturer: Unico, model 300) was tried along with the Staplex sampler at the SE perimeter station. This test sampler had a flow rate of 15 cfm ( $0.47 \text{ m}^3 \text{ min}^{-1}$ ), about 1/3 the flow rate of the Staplex high volume air samplers. After these two weeks, the new sampler was pulled in for maintenance, and there is no indication that the Staplex samplers were replaced. No routine continuous air monitors were employed at the FMPC until the *boundary* air monitoring stations were established in 1972 (Figure B2-1, Table B2-1).

To summarize, from these written sources as well as personal communications with site personnel (Dugan 1992), we have deduced that the typical air sampler used at the FMPC perimeter during the 1960s was a Staplex high-volume air sampler inside a louvered weather shelter, drawing air at  $1.5 \text{ m}^3 \text{ min}^{-1}$  through a 4-inch diameter Mine Safety Appliances Company (MSA) Type S pleated filter. The filter face was oriented perpendicular to the ground surface (Dugan 1992). The average inlet velocity through the sampler filter would have been  $3.2 \text{ m s}^{-1}$ .

Monthly average concentrations of uranium in air for each perimeter station were computed from the individual weekly measurements. A weekly sample was included in a given monthly average if the midpoint of the sampling period fell within that month. A tabular presentation of the monthly average concentrations of uranium in air for 1958-1971 is included in Table B2S-1 ("S" for "Special") in the annex following the main body of this Part. Some individual measurements were invalidated due to conditions such as the

Appendix B — Part 2 Air Monitoring Data

following, which were noted by the sampling or analytical technicians on the analytical data sheets:

- defective pump
- uncertain sample volume or time
- partial loss of sample
- filters came loose.

A general picture of the differences between the stations and the long-term trends is illustrated in Figure B2-4, which shows the annual average concentrations at each station on the same plot. A set of summary figures of the monthly average concentrations follows in Figures B2-5 through B2-10. The monthly data are useful for model validation and for identification of possible episodic releases. Two plots are presented for the NE and SE stations, encompassing first 1958-1971 and then 1961-1971. The second time period provides a better view of some of the peaks in that period of measurements.

The uranium concentration at the NW station is consistently the lowest, with an average over the entire time period 1958–1971 of 80 fCi U m<sup>-3</sup> (Figure B2-4). The SE station is next lowest, with a long-term average of 120 fCi U m<sup>-3</sup>. The SW and NE stations are similar, showing long-term averages of 160 and 150 fCi U m<sup>-3</sup>, respectively. Although the NE station is in the prevailing wind direction, the SW station is closest to the major production area release points. It also shows the most erratic concentration patterns (Figure B2-7).



Figure B2-4. Annual average concentrations of uranium in air at four perimeter stations between 1958 and 1971.



Figure B2-5. Monthly average concentration of uranium in air at NE perimeter (1958-1971).



Figure B2-6. Monthly average concentration of uranium in air at NE perimeter (1961-1971).



Figure B2-7. Monthly average concentration of uranium in air at SW perimeter.



Figure B2-8. Monthly average concentration of uranium in air at NW perimeter.

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Figure B2-10. Monthly average concentration of uranium in air at SE perimeter (1961-1971).

#### **Boundary Air Monitoring**

In 1971, the four air monitors at the perimeter of the production area were replaced by six samplers at what are called "boundary stations" (Figure B2-1). The seventh station (BS-7) was operated infrequently until 1981. Ross and Boback (1971) summarized the air sampling program being used to measure airborne contaminants at the boundary of the FMPC at that time. Continuous air samples were collected by pulling air through an  $8 \times 10$  inch fiberglass or paper filter at a rate of about 1 cubic meter per minute. The permanent equipment at the boundary stations consisted of a high volume air sampler housed in an aluminum enclosure (Figure B2-11). A flow switch, activated by the vacuum pump exhaust, controlled a running time meter and also shut off the power if the pump stopped. The switch and timer provided a record of pump operation. The authors state, "In previous sampling stations which did not have timers, occasional pump failures resulted in discarded filters because there was no record of the sample collection period."



Figure B2-11. Diagram of shed-roof air sampler used for boundary air sampling at the FMPC after 1971 (modified from Ross and Boback 1971).

The monthly average concentrations of uranium in air at the boundary stations were determined from the weekly measurements on the analytical data sheets and are tabulated through 1984 in Table B2S-2 in the annex to Part 2 of this report. The FMPC used an activity-to-mass ratio of  $3.3 \times 10^{-7}$  Ci U g<sup>-1</sup> on the analytical data sheets during this time period, based on the Atomic Energy Commission's definition of a "special curie" of natural uranium which was in effect at that time. The monthly averages in Table B2S-2 in the annex retain the original activity basis as recorded on the data sheets. However, for comparison with the previous and later periods, the data in Figures B2-12 through B2-16 have been converted to an activity-to-mass ratio of  $6.8 \times 10^{-7}$  Ci U g<sup>-1</sup> by multiplying the data by the ratio of 6.77 to 3.33, or 2.03. Thus, all the summary plots in the main part of this appendix are on a comparable basis.

Similar to the presentation of perimeter air data, the annual average measurements at all stations are shown first in Figures B2-12 and B2-13, in order to illustrate the long-term trends ar d differences between stations. For 1985 through 1991, the annual averages were obtained from the FMPC annual environmental reports. The stations track each other from year to year and exhibit a marked decline in the late 1980s as FMPC production activities declined. The concentrations are considerably lower than those measured at the closer perimeter stations in the previous decade. The long-term average concentrations for 1972 through 1984 range from 5 fCi m<sup>-3</sup> for BS-4 to 20 fCi m<sup>-3</sup> for BS-3. These concentrations can be compared with long-term averages ranging from 80 to 160 fCi m<sup>-3</sup> at the perimeter stations during the period 1958-1971 (Figure B2-4).

BS-3 is the boundary station showing the highest uranium concentrations in air, primarily due to it being the closest to the production area (Figure B2-1). It is also near the old solid waste incinerator (at the sewage treatment plant area). However, operations were discontinued at the incinerator at the end of 1979, and BS-3 continued to show the highest uranium concentrations of the boundary stations (Figure B2-12). This suggests that either proximity to the production area or resuspension of contaminated soil around the incinerator are likely to be more significant contributors to airborne uranium at BS-3 than incinerator operations. The SW boundary station (BS-5) has the second lowest long-term average uranium concentration in air, whereas the SW perimeter station was relatively high. This is consistent with the belief that the SW perimeter station was affected by its proximity to the major release points in that part of the production area. However, prevailing winds probably tended to carry those releases towards the NE rather than towards the more distant SW boundary station.

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Figure B2-12. Annual average concentrations of uranium in air at boundary stations 1, 2 and 3 between 1972 and 1991.



Figure B2-13. Annual average concentrations of uranium in air at boundary stations 4, 5, 6, and 7 between 1972 and 1991.

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Figure B2-14. Monthly average concentrations of uranium in air at boundary stations 1 and 2.







Figure B2-16. Monthly average concentrations of uranium in air at boundary stations 5, 6 and 7.

#### Offsite Air Monitoring — Noncontinuous Sampling Through 1970

A limited amount of monitoring of uranium in air at locations beyond the FMPC boundary was performed as early as the 1950s. Our objectives for examining these data were:

- to investigate model validation opportunities, particularly locating data which could elucidate the relationship between distance and direction from the FMPC and groundlevel concentrations of uranium in air;
- to communicate to the interested residents of surrounding communities what the actual measurements of uranium in air indicated during the early years of FMPC operation.

We examined the original analytical data sheets as well as the monthly reports from the Industrial Hygiene and Radiation (IH&R) department. Until the mid-1980s, offsite air samples were not taken with any regular frequency or at established locations. Rather, the location (e.g. "intersection of Paddy's Run Road and New Haven Road") was written on the data sheet along with a general indication of the weather conditions on the day of sampling. Typical sampling times were 45 or 60 minutes per sample at flow rates ranging from 0.5 to  $1.5 \text{ m}^3 \text{ min}^{-1}$ . Under these conditions, the lower limit of detection was about 10 fCi U m<sup>-3</sup>.

From the mid-60s onward, it was common to obtain two separate field replicates at the same time and place, and blank filters were also analyzed. A data set of these field replicates

was used to evaluate the precision of the air sampling method (Killough et al. 1993, Figure L-3).

Sampling notes on the data sheets suggest that the offsite samples were, more often than not, purposefully taken in a direction which was believed to be downwind of the FMPC on that day. Thus, quantitative interpretation of these data should be limited because of this known positive bias. Because samples were not continuous, were infrequent, and were taken with no regular frequency, they can not be used to quantify the amount of airborne uranium to which people were exposed over all time periods. However, they do indicate "snapshots" in time which can be compared with our estimates of offsite concentrations using source term reconstruction and modeling techniques.

There were seven locations outside of the FMPC property boundary which were sampled frequently enough to examine the time history of the uranium concentration in air. These locations are shown in Figure B2-17. Some summary statistics are shown in Table B2-3. It should be emphasized that these measurements are hourly grab samples which do not represent a large coverage of the time period. For example, Ross was sampled 153 times, but over a 159-month period, this constitutes only about 0.1% of the total time.

		Uranium Concentration (fCi U m <sup>-3</sup> air)				
Location	Number of samples	Minimum	Maximum	Mean	Median	
Ross/Venice	153	<10	532	70	40	
Shandon .	48	<10	512	70 <sup>·</sup>	30	
New Haven	24	10	270	70	50	
Fernald	24	<10	440	100	30	
Miami Whitewater Forest/Golf Course	68	<10	389	40	20	
New Baltimore	98	<10	651	70	40	
Roadside Park on Route 128 to Hamilton	79	<10	750	50	30	

# Table B2–3. Uranium in Air (Hourly Grab Samples) at Locations Outside the FMPC Property Boundary through 1970

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**Roadside** Par **forgan** Ross Road oure Shandon Route 126 Ross Ohio Paddy's Run R Route Butler County Hamilton County River FMPC SITE New Haven-Fernald X Baughman Road Grea Mami 1.25 Miami Whitewater New Baltimore Forest Measurements of uranium in air were compiled for these general offsite areas through 1970 .,

Figure B2-17. Offsite air sampling locations.

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The monitoring data for Ross/Venice are plotted in Figure B2-18. A downward trend over time is suggested by the data. The arithmetic average of measurements up through 1965 is 104 fCi m<sup>-3</sup> compared with 53 fCi m<sup>-3</sup> for 1965 through 1970 (medians are 50 and 40 fCi m<sup>-3</sup>, respectively). A similar downward trend with time is exhibited for the three stations which are generally west of the FMPC (Figure B2-19).



Figure B2-18. Hourly grab samples of uranium in air at Ross/Venice (NE of the FMPC) through 1970.



Figure B2-19. Hourly grab samples of uranium in air at three offsite locations through 1970. These locations are generally W of the FMPC site (Figure B2-17).

In addition to these seven locations outside the FMPC boundary, data were compiled for several locations inside the FMPC boundary: the North Access Road Area, the South Access Road Area (includes Willey Rd), and the Coan House Area. The access road locations are shown in Figure B2-1. The exact location of the Coan house has not been verified, but from the analytical data sheets, we deduced that it was several hundred yards from the NE corner of the FMPC perimeter fence. A long-time resident of the area (Clausen 1993) has confirmed that the Coan family owned an old farmhouse on the NE corner of the site, and that they moved when the town of Fernald was established.

Summary statistics for measurements of uranium at these three locations are shown in Table B2-4. The medians of these hourly measurements in the downwind directions (80 fCi  $m^{-3}$  at the N Access Rd area and 260 fCi  $m^{-3}$  at the Coan House area) are 2 to 6 times higher than the median of the hourly measurements made at Ross (40 fCi U  $m^{-3}$ ) during this time period. The maximum concentrations observed at these three areas within the FMPC boundary (Table B2-4) are roughly four times the maximum concentrations observed at locations outside the FMPC boundary (Table B2-3).

In addition to the analytical data sheets, another source (Dodd 1958b) described a set of measurements of uranium in air outside of the production area. Sixteen out-plant air dust samples were collected on October 10, 1958 in pairs from the roof of a truck cab using Staplex high volume air samplers and Whatman #41 filter discs. The sample locations were between 2500 feet and 6200 feet from the production area in seven directions. Concentrations ranged from 370 fCi m<sup>-3</sup> at 6200 feet from the FMPC to 1060 fCi m<sup>-3</sup> at 2500 feet from the site. Taken together, these measurements of uranium beyond the FMPC perimeter lend qualitative support to the model predictions that ground-level concentrations of uranium in air decrease with distance from the site. They are not adequate, however, to give quantitative estimates of the rate of decrease.

	Coan House Area	N Access Road Area	S Access Road Area
Number of Samples	22	132	93
Minimum	<10	<10	<10
Maximum	2070	1380	2800
Mean	430	160	205
Median	260	80	50

Table B2-4. Hourly Grab Samples of Uranium in Air (fCi m <sup>¬</sup>	) at Three General
Areas Between the FMPC Perimeter and the FMPC Bound	ry through 1970

#### Offsite Air Monitoring — Continuous Sampling in the 1980s

Continuous air samplers were established at several offsite locations in the late 1980s. Although this time period is relatively unimportant for the dose reconstruction, we examined these data for insight into the patterns of ground-level concentrations of uranium in air with distance and direction from the site. We did not locate original data sheets for these measurements; rather, information was obtained from the annual environmental monitoring reports (Table B2-5). The information may be used in final model validation exercises in Task 6.

Station	Nearby Community				
Code	and Bearing <sup>b</sup>	1987	1988	1989	1990
AMS 10	Fernald	0.71	0.25	0.13	0.06
	(2.2  km S)				
AMS 11	New Haven	0.60	0.2 <b>9</b>	0.07	0.04
	(3.7  km SW)				
AMS 12	Shandon	0.29	0.14	0.06	0.04
	(5.7  km NW)				
AMS 13	Ross	1.26	0.63	0.09	0.06
	(4.1 km NE)				
AMS14	Ross	NA	0.18	0.07	0.04
	(4.3 km NE)				
AMS15	Cincinnati	NA	NA	0.08	0.05
	(24.8  km SE)				
AMS16	Miamitown	NA	NA	0.12	0.06
(9.9 km SSW)					
<sup>a</sup> Monitoring data for offsite stations are not reported in 1991					
Environm	ental Report (WEMCO 19	9Z).			
°Dista	nce and direction from	the cent	ter of the	FMPC at	t Plant 4
<u>estimated</u>	from Figure 21 of WEMC	<u>O (1992).</u>			

Table B2-5. Average Concentrations of Uranium in Air (fCi m<sup>-3</sup>) at Permanent Offsite Air Monitoring Stations (AMS)<sup>a</sup>

### IDENTIFICATION OF POSSIBLE EPISODIC RELEASES

For the dose reconstruction process, examining the long-term trends in air monitoring data has provided an opportunity to pinpoint possible episodic releases which might not have been detected adequately by effluent monitoring. For the purposes of this dose reconstruction project, an episodic release is defined as one which increases the composite uranium release rate by a factor of at least 10 for a period of less than 10 days. All releases are included in the source term, but episodic releases warrant special dose assessment procedures.

Possible episodic releases were selected using the following methodology. First, plots of air monitoring data were visually examined for obvious peaks. These peaks are marked with arrows in Figures B2-5 through B2-10 and in Figure B2-14 through B2-16. The individual weekly air monitoring measurements were reviewed to better define the timing of the potential release. IH&R Department Monthly Reports and miscellaneous incident reports were then reviewed to help confirm that potential episodic releases suggested by the air monitoring data occurred. If the timing of the peak coincided with the occurrence of a documented unplanned release, it was assumed that the measured peak represents contamination from that release. Gummed-film data sets were also used to help verify possible episodic releases. A few of the gummed-film results encompass time periods short enough that peaks due to episodic events could be discerned.
In addition to a visual examination of plots and subsequent review of supporting documents and gummed-film data, a more quantitative method was used to determine whether certain times could contain episodic releases which warrant special dose assessment procedures. Individual measurements for each year were tabulated, along with the annual average concentration at that monitoring station for that year, and the ratio of the individual to annual average concentration was determined (Table B2-6). A ratio of greater than 10 suggests that an episodic release occurred. There were 20 measurements representing 14 sampling periods which met this criterion (Table B2-6).

Several possible episodic releases were identified using the screening methods described above and are discussed below. It must be emphasized that examination of the environmental monitoring data is only one method for identifying potential episodic releases. A complete review of episodic releases, including other types of historic records, will be included in the final source term report for this dose reconstruction project (Tasks 2 and 3).

	101 1 Hat Station, 1800-1804								
	Sample	Annual	Individual						
	Ending Date	Average	Measurement	Station	Ratio <sup>b</sup>				
	3/19/66	130	1310	SW	10				
	10/28/67	40	430	NW	11				
	9/21/68	96	1000	NW	10				
	3/13/70	90	940	SW	10				
	5/26/72	19	240	BS-6	13				
_	3/30/78	17	179	BS-3 ·	11				
-	9/28/78	7	94	BS6	13				
	2/8/79	13	167	BS-1	13				
	2/8/79	7.7	81	BS-2	11				
	2/8/79	1 <b>9</b>	463	BS3	24				
	2/8/79	8.1	15 <b>2</b> ·	BS-4	19				
	2/8/79	10.8	252	BS-5	23				
	2/8/79	9.2	164	BS-6	18				
	10/30/80	3.5	42	BS-4	12				
	11/25/80	4.6	47	BS-3	10				
	7/23/81	3.6	58	BS-4	16				
	9/3/81	.8.8	124	BS-2	14				
	4/26/83	25	246	BS-3	10				
	4/26/83	8.6	88	BS-4	10				
	9/20/83	9.9	100	BS-5	10				

#### Table B2-6. Uranium Concentrations (fCi m<sup>-3</sup>) in Air Samples from Perimeter and Boundary Stations Which Were ≥Ten Times the Annual Average Concentration for That Station, 1958-1984<sup>a</sup>

<sup>a</sup> Data from analytical data sheets (NLCO 1953-1984). All concentrations standardized to an activity-to-mass ratio of 6.8  $\times 10^{-7}$  Ci g<sup>-1</sup> for natural uranium.

<sup>b</sup> Ratio of individual measurement concentration to annual average concentration for that station.

#### November 1960

There was a uranium release from the pilot plant in November 1960 (around November 12 to 20) which met our criteria for an episodic release. This event was reviewed in the Task 4 methodology report (Appendix V, Killough et al. 1993). The air monitoring stations at the perimeter were not operating during the period of suspected highest releases. The last records of air monitoring in 1960 were from the NE and SE perimeter stations during November 6–8. The concentrations of uranium in air for those samples were in fact the highest observed for the entire year (Figure B2-5), suggesting that some deterioration of the bag filter may have occurred earlier in the month. In addition, gummed film monitoring clearly confirmed an episodic release (Killough et al. 1993, Figures V–1 and V–2).

#### March 1966

The uranium concentration measured in the weekly air sample collected at the SW perimeter station on March 19, 1966 (1300 fCi m<sup>-3</sup>) was ten times greater than the annual average concentration at the SW station in 1966. There was nothing in the literature reviewed to suggest that an episodic release occurred during this period. In addition, elevated concentrations were not observed at the other perimeter locations during this time period.

#### October 1967

The uranium concentration measured in the air sample collected on October 28, 1967 at the NW perimeter station (430 fCi m<sup>-3</sup>) was eleven times higher than the annual average measured during that year. Although a slight increase in concentration was noted in the NE perimeter station (about 2.5 times the annual average concentration), no references to an incident occurring at this time could be found. A potential source is the waste pit area, located just W and SW of the air sampler.

#### September 1968

The uranium concentration measured in the air sample collected at the NW perimeter station on September 21, 1968 (1000 fCi m<sup>-3</sup>) was ten times greater than the annual average for that station in 1968. Increases in air concentrations were not observed at the other perimeter stations during this week, nor was any documentation of an episodic release occurring during this time period found.

#### March 1970

On March 13, 1970, the uranium concentration measured in the air sample collected at the SW perimeter station (940 fCi m<sup>-3</sup>) was ten times higher than the annual average measured during that year. Similar increases were not evident at the other perimeter locations. No references to an incident occurring during this time period were found in the documents reviewed.

#### May 1972

An elevated uranium concentration was observed in air samples collected at BS-6 during May 1972 (Figure B2-16). Similar increases were not noted at other boundary stations. The highest concentration (240 fCi m<sup>-3</sup>) was recorded for the week ending May 26, 1972 and was 13 times greater than the annual average. Adams (1985) reported a dust loss of 184 lbs of uranium from dust collector G1-856 during the month of May, although the exact dates of the release are not recorded. Another potential source is the waste pit area, which is located near the air sampling station in the NE direction.

#### March 1978

A potential episodic release is indicated during the week ending March 30, 1978 by the elevated uranium concentrations measured in air sampled at BS-1, BS-2, and BS-3. The uranium concentration measured in the air sample collected at BS-3 (179 fCi m<sup>-3</sup>) is approximately eleven times greater than the annual average. The concentrations measured at BS-1 and BS-2 are approximately eight times greater than the average annual concentration calculated for each of those stations. Adams (1978) reports that "a significant dust loss occurred in the Plant 9 dust collector servicing the NPR furnace and the crucible burnout area" during the period from 3/15/78 to 6/14/78. The total dust loss was 256 lbs, and the total uranium loss was 153 lbs. The loss resulted from the mechanical failure of the collector blow ring. The loss was not reported until June 14, 1958, so it is difficult to pinpoint when the release occurred. However, it was estimated that the loss would have required 20-35 operating days. Thus, this release does not fit our definition of an episodic release (i.e., total release must occur in less than 10 days).

#### September 1978

On September 28, 1978, peaks in uranium concentrations were observed in weekly air samples collected at BS-2, BS-3, BS-4, BS-5, and BS-6. The highest increase (ten times the annual average) was measured at BS-6 (94 fCi m<sup>-3</sup>). Concentrations measured at the other stations measured from three to nine times the annual averages for those stations. Although an episodic release is implicated by the air data, no information related to such a release could yet be found in the available references.

#### February 1979

This month appeared to contain an episodic release according to several of the air monitoring station data sets (Figures B2-14, B2-15, and B2-16). In order to investigate the timing more carefully, the weekly data were examined (Figure B2-20). These data show the elevated concentrations were limited to the week ending February 8, 1979. The peak concentrations at all stations were over an order of magnitude higher than the annual averages for the year (Table B2-6), indicating that the release meets our definition of an episodic release. The maximum value of 230 fCi m<sup>-3</sup> (470 fCi m<sup>-3</sup> using a specific activity of  $6.8 \times 10^{-7}$  Ci g<sup>-1</sup>) was confirmed in the annual environmental monitoring report for that

year (Table 1 of Boback and Ross 1980); although no explanation was given there. However, the IH&R Department Monthly Report for February, 1979 (Boback 1979) did include the following discussion:

"An unexplained increase in uranium concentration occurred recently in Boundary Station air samples. Generally the average concentration is about  $0.4 \times 10^{-14} \mu \text{Ci/mL}$ , but during the period February 1 to February 8 the average was  $10.49 \times 10^{-14} \mu \text{Ci/mL}$ . All six Boundary Stations showed high uranium, alpha, and beta. ... Investigations were made, but the cause of the high uranium and activity was not discovered. No stack losses occurred and no large spills were reported. Material burned at the incinerator near BS-3 was the normal noncontaminated paper and scrap. No dumping of material to the pit was reported or observed.

The highest uranium concentration was only 11.4% of the NCG [sic] but the high U concentration found at all six locations would indicate a rather large source leak, continuous for two or more days. The wind during this period was mostly from the west but there was wind during this period from all around the compass."



Figure B2-20. Uranium in air at boundary stations in 1979, illustrating episodic release in February.

#### October 1980

An elevated uranium concentration was observed in a weekly air sample collected at BS-4 on October 30, 1980. This result (42 fCi m<sup>-3</sup>) was approximately twelve times higher than the average for 1980. Concomitant increases were not noted in other boundary stations

during this week, nor was any documentation of an episodic release found in the references reviewed.

#### November 1980

On November 25, 1980 a uranium concentration of 47 fCi m<sup>-3</sup> was measured in the weekly sample collected at BS-3. The result was an order of magnitude greater than the annual average. The results obtained from other boundary stations were smaller than the annual average. Reference to an incident occurring during this period could not be found. Active burning at the solid waste incinerator had halted by this time, but resuspension of ground contamination in that area could have been responsible for the elevated airborne contamination.

#### July 1981

The uranium concentration measured in the weekly air sample collected at BS-4 on July 23, 1981 (58 fCi m<sup>-3</sup>) was 16 times greater than the average for the year 1981. There was nothing in the literature reviewed to suggest that an episodic release occurred during this period. In addition, elevated concentrations were not observed at the other boundary stations during this time period.

#### September 1981

During the week ending September 3, 1981, increases in uranium concentrations were observed in weekly air samples collected at BS-1 and BS-2. The concentrations were approximately three and fourteen times, respectively, greater than the annual average concentration measured at those locations. Nutter (1981) reports a 263 kg loss of green salt from Plant 4 dust collector G4-2 sometime during the period from August 29 to through September 8, 1981. The dust collector was operated intermittently during the period from August 31 through September 3. The Plant was not in operation September 5-8, but dust collector G4-2 and others were turned on September 5 and 8. After the collector was shut down on September 8, a torn bag was found. The magnitude of the loss was not noted until September 9. The report notes that "the nearly constant differential pressure during seven days of operation prior to the discovery of the torn bag on September 8 indicated that the dust collector was not functioning properly." It thus appears that the air monitoring results could reflect this dust loss.

#### **April 1983**

Uranium concentrations in weekly air samples collected at BS-3 and BS-4 on April 26, 1983 were an order of magnitude greater than the respective annual averages. Reference to an incident occurring during this period could not be found.

#### September 1983

Increased concentrations of uranium were reported for air samples collected at BS-1, BS-2, BS-3, and BS-4 during the month of September 1983. The result at BS-5 was approximately ten times greater than the annual average calculated for 1983. No documentation of an incident occurring during this period of time could be found.

#### **Summary of Episodic Releases**

Based on the criteria used to establish an episodic release, 14 possible episodic releases were identified from the air monitoring data collected during the period from 1958 through 1984. Of these, only three appear to be supported by documentation and, in one case, by gummed-film results. These releases occurred during November 1960, February 1979, and September 1981. The remaining potential releases lacked documentation or other confirming information. However, they are listed here and will be combined with other methods for investigating episodic releases in the final source term report (Tasks 2 and 3).

#### EVALUATION OF QUALITY OF ENVIRONMENTAL MONITORING DATA FOR URANIUM IN AIR

For model validation purposes, it is critical to assess the quality of the environmental data before comparing the observed measurements with predicted concentrations. An assessment of the precision and bias of the perimeter air sampling results was presented in Appendix L of the Task 4 report (Killough et al. 1993), and the reader is referred to that source for detailed information. An important conclusion was that the high-volume air samplers were only about 50% efficient for the particle sizes present at the FMPC perimeter in the 1960–1962 period. In a qualitative sense, the efficiency of the samplers at the FMPC boundary should be higher, because the larger particles would have been selectively deposited on the ground between the perimeter and the boundary stations. However, depending on which data are used for model validations in the final Task 6 report, a similar assessment of sources, particle sizes, and sampler efficiency will be performed for the measurements of uranium in air at the boundary and offsite stations.

#### SUMMARY

The air monitoring data collected from the environs of the Fernald site have been thoroughly examined for usefulness in supporting the methodology and conclusions of the dose reconstruction project. A primary use of the data will be model validation, which consists of comparison of model predictions to available measurements at different places and times. A model validation for the three-year period 1960-1962 was included in Killough et al. (1993), as part of that methodology development effort. Validations for other time periods will be included in the final Task 6 report. The measurements of uranium in air beyond the FMPC perimeter support the model predictions that concentrations decrease with distance from the site.



Monitoring results for perimeter and boundary stations have been presented in separate sections of this part. To permit a longer-term, summary view, the results of monitoring in the NE direction are plotted together in Figure B2-21, below. After 1971, air monitoring in the NE direction was performed at boundary station BS-2, which is about 800 m further from the production area than the NE perimeter station. There is a clear decrease in uranium concentration over time. For perspective, the current DOE concentration guide for uranium in air is 100 fCi m<sup>-3</sup>, which corresponds to a committed effective dose equivalent of 100 mrem for the most insoluble class of uranium compounds.



Figure B2-21. Summary of uranium in air at the perimeter and boundary stations NE of the FMPC from 1958–1991. The boundary station is about 800 m further from the production area than the NE perimeter station. The current DOE standard for uranium in air is 100 fCi m<sup>-3</sup>.

In addition to providing data for model/source term validation, another use of the air monitoring data has been the identification of episodic releases. Plots of monthly average concentrations of uranium in air over time were examined for peaks, which were further investigated by reviewing weekly measurements. In addition, all individual measurements which were  $\geq$  ten times the annual average at that location were tabulated and investigated. Some previously identified episodic releases were confirmed in this manner. At least one other episodic release (February 1979) was newly identified by this examination of the air monitoring data record.

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#### ANNEX TO APPENDIX B PART 2

### DETAILED TABLES OF URANIUM IN AIR AT PERIMETER AND BOUNDARY STATIONS

# Table B2S-1. Monthly Average Concentrations of Uranium (fCi m<sup>-3</sup>) in Air at the FMPC Perimeter From 1958 through 1971<sup>a</sup>

	Perimeter Station						
<u>Month/year</u>	SW	NW	NE	SE			
Feb-58	160	400	1255	260			
Mar-58	13	5	6	1090			
Jun-58	66	182	600	303			
Jul-58	NA <sup>b</sup>	NA	245	NA			
Aug-58	81	54	153	211			
Oct-58	43	25	58	57			
Nov-58	91	65	221	84			
Dec-58	136	25	67	65			
Jan-59	110	87	NA	87			
Apr-59	NA	68	111	73			
May-59	NA	150	133	59			
Jun-59	176	56	104	59			
Jul-59	106	NA	121	73			
Aug-59	120	NA	49	37			
Sep-59	145	NA	97	NA			
May-60	NA	311	190	NA			
Jun-60	NA	65	101	NA			
Jul-60	357	199	125	123			
Aug-60	253	96	146	132			
Sep-60	311	169	94	120			
Oct-60	510	104	173	220			
Nov-60	NA	NA	906	410			
Jan-61	NA	99	151	62			
Feb-61	NA	38	101	115			
Mar-61	161	75	148	172			
Арт-61	111	118	127	246			
Мау-61	350	76	104	177			
Jun-61	96	96	96	96			
Jul-61	84	317	130	52			
Aug-61	70	109	117	188			
Sep-61	230	92	144	28			
Oct-61	81	79	191	173			
Nov-61	176	130	142	152			
Dec-61	68	68	68	68			
Jan-62	81	41	232	57			

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Table B2S-1. Monthly Average Concentrations of Uranium (fCi m <sup>-3</sup> ) in Air at th	he
FMPC Perimeter From 1958 through 1971 <sup>a</sup> (cont.)	

	Perimeter Station					
Month/year	SW_	NW	NE	SE		
Feb-62	243	33	85	186		
Mar-62	527	40	84	247		
Apr-62	234	66	135	316		
May-62	161	187	179	148		
Jun-62	408	48	133	106		
Jul-62	59	48	107	113		
Aug-62	250	88	132	193		
Sep-62	211	119	131	198		
Oct-62	101	50	207	113		
Nov-62	375	45	115	143		
Dec-62	217	56	214	86		
Jan-63	334	54	310	150		
Feb-63	155	80	115	224		
Mar-63	248	51	319	283		
Apr-63	396	108	636	310		
May-63	282	212	364	236		
Jun-63	245	123	208	260		
Jul-63	505	153	305	163		
Aug-63	172	104	126	108		
Sep-63	458	83	170	143		
Oct-63	160	140	352	224		
Nov-63	68	158	410	268		
Dec-63	118	90	173	• 153		
Jan-64	235	122	258	95		
Feb-64	258	93	255	450		
Mar-64	108	150	230	190		
Apr-64	270	103	243	198		
May-64	242	125 .	283	110		
Jun-64	305	83	355	157		
Jul-64	150	63	123	90		
Aug-64	292	54	70	136		
Sep-64	207	93	133	123		
Oct-64	196	48	166	140		
Nov-64	298	98	200	188		
Dec-64	93	47	210	47		
Jan-65	138	58	174	114		
Feb-65	100	90	153	85		
Mar-65	95	25	55	108		
Apr-65	128	23	100	65		

(continued next page)

	Perimeter Station					
<u>Month/year</u>	SW	NW	NE	SE		
May-65	100	38	196	84		
Jun-65	153	29	135	40		
Jul-65	86	20	126	65		
Aug-65	83	30	148	60		
Sep-65	45	48	95	73		
Oct-65	110	73	68	48		
Nov-65	48	26	82	42		
Dec-65	12	16	116	38		
Jan-66	138	23	40	55		
eb-66	92	30	60	40		
Mar-66	358	68	105	43		
\pr-66	174	42	104	72		
flay-66	93	30	63	50		
un-66	38	33	128	150		
ul-66	144	24	60	40		
Aug-66	35	27	33	50		
ep-66	83	17	28	60		
)ct-66	220	71	98	38		
lov-66	53	43	345	123		
)ec-66	158	52 <sup>.</sup>	252	196		
an-67	45	28	283	85		
'eb-67	65	10	105	38		
lar-67	1 <b>66</b>	28	122	90		
Apr-67	203	33	130	90		
lay-67	90	43	80	148		
un-67	150	53	118	65		
ul-67	30	23	33	73		
ug-67	215	25	85	70		
ер-67	244	35	58	42		
kt-67	14	128	163	203		
lov-67	93	63	185	145		
<b>)ec-67</b>	198	52	96	118		
an-68	288	43	· 78	83		
`eb-68	175	40	125	160		
far-68	316	42	134	96		
pr-68	98	83	73	115		
fay-68	158	70	88	104		
un-68	85	73	145	. 88		
ul-68	10	93	183	148		
Aug-68	NA	38	174	222		
Sep-68	NA	473	188	255		

Table B2S-1. Monthly Average Concentrations of Uranium (fCi m<sup>-3</sup>) in Air at the FMPC Perimeter From 1958 through 1971<sup>a</sup> (cont.)

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Perimeter Station									
<u>Month/year</u>	SW	NW	NE	SE					
Oct-68	NA	135	145	240					
Nov-68	NA	44	140	80					
Dec-68	45	80	88	118					
Jan-69	154	112	70	110					
Feb-69	488	18	43	63					
Mar-69	218	87	104	173					
Apr-69	123	40	112	153					
May-69	136	45	88	76					
Jun-69	395	90	183	53					
Jul-69	53	53	260	207					
Aug-69	148	33	25	100					
Sep-69	150	33	33	29					
Oct-69	36	24	50	50					
Nov-69	23	30	85	50					
Dec-69	60	13	45	65					
Jan-70	40	45	48	35					
Feb-70	108	30	123	55					
Mar-70	315	45	110	63					
Apr-70	208	34	40	42					
May-70	60	43	144	24					
Jun-70	50	85	83	35					
Jul-70	40	30	60	28					
Aug-70	35	23	20	28					
Sep-70	18	28	38	24					
Oct-70	40	35	28	20					
Nov-70	31	23	23	13					
Dec-70	54	28	58	54					
Jan-71	25	38	23	35					
Feb-71	21	35	35	25					
Mar-71	70	30	40	48					
Apr-71	86	44	38	62					
May-71	54	44	70	26					
Jun-71	53	58	48	55					
Jul-71	23	20	38	23					
Aug-71	58	23	25	15					

#### Table B2S-1. Mo at the

<sup>a</sup> An activity to mass ratio of  $6.8 \times 10^{-7}$  Ci g<sup>-1</sup> was used during this time period.

48

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 $^{b}$  NA = No data available.

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Sep-71

Oct-71

Nov-71

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Boundary Monitoring Stations, 1971–1984									
Month/yr	<b>BS-1</b>	BS-2	BS-3	BS-4	BS-5	<b>BS-6</b>	BS-7		
Jan-72	6.68	6.40	9.75	4.58	2.90	10.63	NA		
Feb-72	10.60	3.80	15.53	2.68	4.43	4.10	NA		
Mar-72	2.80	2.15	5.53	1.77	2.03	4.13	NA		
Apr-72	8.58	7.47	13.30	1.58	1.60	5.15	NA		
May-72	6.80	2.93	5.18	2.46	7.25	34.50	NA		
Jun-72	NA	8.23	NA	4.03	5.20	5.43	NA		
Jul-72	7.27	6.90	9.93	2.00	4.77	4.17	NA		
Aug-72	7.67	22.47	7.10	3.73	3.37	4.97	NA		
Sep-72	5.46	5.58	6.84	2.14	2.96	5.62	NA		
Oct-72	4.70	5.73	7.77	5.83	4.40	6.97	NA		
Nov-72	1.80	2.00	8.20	2.25	2.48	9.60	NA		
Dec-72	5.22	2.08	9.47	1.72	1.36	7.13	NA		
Jan-73	13.10	16. <b>9</b> 8	22.05	1.65	3.48	8.33	NA		
Feb-73	4.03	3.35	7.68	3.40	1.45	5.73	NA		
Mar-73	4.46	2.7 <b>4</b>	4.03	1.04	1.90	6.58	NA		
Apr-73	7.20	3.80	11.63	1.85	6.50	7.30	NA		
May-73	9.43	5.53	11.68	1.53	1.00	3.18	NA		
Jun-73	8.66	9.00	9.18	1.32	1.86	3.44	11.90		
Jul-73	9.33	7.40	7.65	2.38	2.78	6.17	8.05		
Aug-73	30.20	14.00	10.30	2.90	5.53	8.40	25.43		
Sep-73	10.88	14.30	20.36	6.88	6.38	21.16	7.80		
Oct-73	11.78	9.98	15.40	8.55	4.33	10.90	12.70		
Nov-73	14.70	14.08	16.44	3.38	4.34	16.04	NA		
Dec-73	2.15	3.25	4.30	2.50	2.13	4.80	7.25		
Jan-74	4.85	4.65	4.25	1.83	2.00	3.88	6.20		
Feb-74	5.90	6.35	7.73	1.95	4.78	6.43	NA		
Mar-74	7.78	5.88	8.26	1.20	3.30	6.64	NA		
Apr-74	19.65	8.98	8.20	3.80	2.00	3.50	NA		
May-74	10.88	9.06	9.30	2.32	3.70	5.68	NA		
Jun-74	13.93	10.50	8.25	1.20	<b>3.86</b>	4.48	NA		
Jul-74	8.25	8.10	6.23	2.18	4.85	4.03	NA		
Aug-74	14.18	13. <b>36</b>	10.80	2.76	3.70	7.52	NA		
Sep-74	8.65	6.65	4.90	1.43	4.45	6.13	NA		
Oct-74	16.00	10.08	14.35	4.90	4.08	6.30	NA		
Nov-74	11.70	7.38	10.86	-3.24	3.64	7.60	NA		
Dec-74	7.70	10.30	5.73	1.63	1.45	7.70	NA		
Jan-75	14.18	13.30	20.18	2.08	2.55	10.53	NA		
Feb-75	3.68	4.10	12.58	1.43	2.65	14.00	NA		
Mar-75	6.58	3.66	7.48	2.24	2.08	6.46	NA		
Apr-75	9.28	7.93	6.43	3.98	4.30	10.23	NA		
May-75	16.78	21.80	25.95	10.83	5.70	18.55	NA		

Table B2S-2.	Monthly Average	<b>Concentrations</b> or	f Uranium	(fCi U m <sup>-</sup>	<sup>3</sup> ) in Air at
	Boundary M	onitoring Stations	s. 1971–198	4a ·	

(continued next page)

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_	<u> </u>	undary M	onitoring	Stations,	1971-1984	" (cont.)	
<u>Month/yr</u>	BS-1	BS-2	BS-3	BS-4	<u>BS-5</u>	BS-6	BS-7
Jun-75	20.62	18.22	15.56	3.04	5.62	7.28	NA
Jul-75	10.03	12.99	13.23	5.65	4.68	7.38	NA
Aug-75	7.18	11.98	9.83	2.43	4.50	4.88	NA
Sep-75	6.50	5.30	8.80	2.60	4.56	6.32	NA
Oct-75	26.58	12.33	24.65	6.65	10.70	11.53	NA
Nov-75	19.53	16.58	16.08	4.35	5.43	6.13	NA
Dec-75	8.33	4.73	4.22	1.20	1.76	3.30	NA
Jan-76	23.30	6.78	9.43	1.53	0.70	1.83	NA
Feb-76	9.68	9.85	7.55	1.98	1.10	2.55	NA
Mar-76	11.46	5.84	10.84	1.42	1.20	3.34	0.20
Apr-76	9.88	4.10	9.13	3.50	4.55	4.66	6.85
May-76	6.34	4.62	5.54	1.86	3.40	4.94	5.74
Jun-76	9.65	8.58	13.20	1.83	1.73	3.90	4.63
Jul-76	7.03	7.65	11.48	4.27	2.03	2.23	2.73
Aug-76	9.90	4.18	4.76	2.88	5.88	9.64	10.50
Sep-76	7.85	6.45	10.80	5.95	5.20	12.03	6.20
Oct-76	8.20	2.30	3.90	2.80	6.70	10.80	6.40
Nov-76	NA	NA	NA	NA	NA	NA	NA
Dec-76	NA	NA	NA	NA	NA	NA	NA
Jan-77	3.76	3.24	8.70	2.72	3.90	2.94	NA
Feb-77	7.05	11.90	10.85	4.93	1.83	3.55	NA
Mar-77	18.98	11.15	12.50	4.45	6.00	4.05	NA
Apr-77	15.35	5.63	12.08	2.58	3.50	3.28	NA
May-77	7.86	7.58	9.10	2.78	6.40	4.54	NA
Jun-77	3.10	2.55	3.83	1.65	1.85	3.95	NA
Jul-77	2.73	1.08	3.08	0.40	0.48	0.57	NA
Aug-77	4.56	2.06	5.78	1.32	1.46	2.80	NA
Sep-77	1.05	0.85	1.88	0.68	0.63	0.60	NA
Oct-77	2.52	0.68	2.76	1.26	1.34	1.10	NA
Nov-77	1.55	1.63	3.20	0.80	1.93	0.68	NA
Dec-77	1.40	1.20	2.55	0.78	0.98	1.08	NA
Jan-78	1.58	0.96	5.40	2.28	3.86	1.26	NA
Feb-78	2.83	3.30	3.78	6.13	13.60	4.48	NA
Mar-78	9.15	17.88	30.53	7.23	16.90	5.80	NA
Apr-78	12.53	14.20	23.63	3.65	6.08	7.55	NA
May-78	5.80	1.78	6.34	0.90	1.42	2.06	NA
Jun-78	1.43	1.53	1.60	0.93	1.13	1.18	NA
Jul-78	0.85	0.80	1.80	0.60	0.48	0.63	NA
Aug-78	1.64	1.48	1.48	0.64	1.04	1.06	NA
Sep-78	2.33	4.55	12.45	3.98	12.28	12.20	NA
Oct-78	4.48	3.32	7.64	2.44	5.14	3.16	NA

(continued next page)

Boundary Monitoring Stations, 1971–1984 <sup>a</sup> (cont.)									
Month/yr	BS-1	BS-2	BS-3	<b>BS</b> -4	BS-5	BS-6	<b>BS-7</b>		
Nov-78	4.28	1.28	3.18	1.90	1.95	2.18	NA		
Dec-78	1.57	1.70	2.80	0.50	0.67	0.83	NA		
Jan-79	2.20	1.12	5.46	1.08	4.10	1.54	NA		
Feb-79	24.68	14.38	66.08	23.80	39.15	27.15	NA		
Mar-79	2.55	0.93	3.28	3.80	2.40	1.08	NA		
Apr-79	1.56	1.44	. 3.30	0.84	1.06	1.84	NA		
May-79	12.53	1.63	3.53	2.03	1.43	2.37	NA		
Jun-79	2.68	1.90	4.03	2.53	1.13	1.47	NA		
Jul-79	1.83	0.83	1.37	0.50	0.43	0.83	NA		
Aug-79	1.03	1.53	1.10	0.63	0.50	0.75	NA		
Sep-79	4.85	2.08	4.45	4.50	5.75	5.28	NA		
Oct-79	13.46	4.22	13.40	4.62	4.22	6.70	NA		
Nov-79	6.13	9.65	5.75	3.05	3.15	3.05	NA		
Dec-79	3.08	5.18	2.24	1.04	0.70	0.74	NA		
Jan-80	1.80	0.95	2.30	2.15	4.60	4.90	NA		
Feb-80	2.23	3.43	2.43	2.20	1.13	1.70	NA		
Mar-80	2.34	1.78	1.76	0.78	1.80	1.94	NA		
Apr-80	0.80	1.03	1.15	1.08	0.80	1.43	NA		
May-80	1.40	1.10	1.30	1.68	1.20	2.03	NA		
Jun-80	1.76	2.06	1.08	2.01	1.37	1.92	NA		
Jul-80	0.69	0.57	0.79	0.58	1.21	0.55	NA		
Aug-80	2.12	1.34	1.57	1.00	1.29	1.34	NA		
Sep-80	2.06	1.30	2.60	1.07	1.20	2.49	NA		
Oct-80	3.94	2.21	3.76	5.68	0.88	2.73 .	NA		
Nov-80	4.22	4.00	7.45	2.32	1.64	2.36	NA		
Dec-80	1.31	1.47	1.18	0.74	0.97	0.84	NA		
Jan-81	2.15	1.63	2.20	0.92	0.69	0.54	1.08		
Feb-81	1.26	1.13	2.83	0.52	1.10	1.96	1.50		
Mar-81	2.12	1.39	2.28	0.63	0.76	1.53	0.31		
Apr-81	5.22	3.48	2.86	0.65	1.20	1.33	0.62		
May-81	9.57	3.51	3.58	0.98	5.91	1.49	0.63		
Jun-81	8.52	6.82	10.33	1.37	1.38	1.59	0.97		
Jul-81	2.65	2.48	5.04	7.97	5.56	7.42	1.99		
Aug-81	4.44	15.33	5. <del>64</del>	0.65	2.75	3.47	2.88		
Sep-81	3.65	5. <b>92</b>	9.93	3.09	3.87	2.27	0.98		
Oct-81	3.86	4.65	10.57	1.98	4.55	3.26	2.22		
Nov-81	2.57	1.91	3.98	0.84	3.89	3.55	1.58		
Dec-81	1.97	1.23	4.76	2:44	0.78	0.94	0.62		
Jan-82	2.51	2.47	4.87	1.72	1.62	1.55	1.06		
Feb-82	3.78	3.70	5.10	1.67	7.45	1.63	0.77		
Mar-82	6.83	3.34	7.11	2.65	5.67	5.07	1.35		

Table B2S-2. Monthly Average Concentrations of Uranium (fCi U m<sup>-3</sup>) in Air at Boundary Monitoring Stations, 1971-1984<sup>a</sup> (cont.)

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<u>Month/yr</u>	BS-1	BS-2	BS-3	BS-4	BS-5	<b>BS-6</b>	BS-7
Apr-82	9.37	4.00	10.34	2.50	5.72	5.13	1.03
May-82	11.81	5.62	12.48	2.56	5.21	6.05	4.97
Jun-82	15.33	8.05	10.17	4.31	4.53	4.38	2.34
Jul-82	6.00	3.51	4.27	0.71	1.41	1.54	1.83
Aug-82	2.27	1.98	4.25	1.25	2.60	9.62	1.85
Sep-82	4.42	2.14	4.05	1.10	0.73	0.84	0.99
Oct-82	7.93	5.88	5.79	2.37	2.72	4.55	2.47
Nov-82	10.44	6.18	7.78	3.40	3.08	2.81	2.18
Dec-82	14.00	5.00	9.85	1.06	2.66	4.20	2.70
Jan-83	2.84	2.86	4.75	1.84	2.12	3.61	1.33
Feb-83	3.35	4.68	8.66	3.54	6.75	8.68	2.91
Mar-83	9.62	9.08	31.48	6.88	6.93	4.95	3.06
Apr-83	27.81	21.51	30.32	14.85	7.68	8.85	1.72
May-83	9.42	4.55	8.91	1.90	2.58	5.00	2.03
Jun-83	13.44	9.09	9.14	2.61	3.52	7.77	4.77
Jul-83	5.47	3.68	4.28	2.88	2.30	4.11	0.93
Aug-83	6.32	5.29	9.53	3.19	3.30	4.20	1.29
Sep-83	32.10	11.77	24.84	4.64	15.86	9.01	6.27
Oct-83	8.08	5.51	4.87	2.87	4.15	5.83	1.97
Nov-83	6.28	4.80	5.94	3.74	3.29	2.86	2.32
Dec-83	1.61	1.83	4.22	0.53	0.91	1.92	0.88
Jan-84	3.53	3.63	5.42	1.44	1.36	1.41	0.66
Feb-84	7.34	5.26	9.38	2.18	1.67	4.04	2.32
Mar-84	2.56	1.90	5.55	1.79	1.67	6.53	1.34
Apr-84	4.23	3.38	22.24	1.14	2.20	4.90	1.50
May-84	10.39	6.61	9.69	1.65	3.12	6.02	1.80
Jun-84	13.03	11.24	9.18	2.81	2.35	2.57	1.31
Jul-84	2.29	3.83	2.98	1.24	0.93	1.82	0. <b>99</b>
Aug-84	12.15	10.74	15.19	4.42	4.01	7.25	2.24
Sep-84	21.80	8.89	12.21	2.80	4.33	3.61	2.73
Oct-84	4.06	3.76	5.06	3.23	6.67	9.60	4.14
Nov-84	13.60	10.23	18.18	3.17	8.71	8.79	5.62
Dec-84	5.60	6.34	14.02	2.56	2.14	2.62	2.20

Table B2S-2. Monthly Average Concentrations of Uranium (fCi U m<sup>-3</sup>) in Air at

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 $3.3 \times 10^{-7}$  Ci g<sup>-1</sup>.

#### APPENDIX B -- REGIONAL ENVIRONMENTAL MONITORING

#### PART 3 - WET DEPOSITION

#### INTRODUCTION

"Wet deposition" refers to the removal of uranium-bearing particulates from the air onto ground surfaces by the actions of rain, snow, or mixtures. Theoretical aspects of the wet deposition process were addressed in Appendix H of the Task 4 methodology report (Killough et al. 1993). Uranium measurements in wet deposition and air were used to compute a site-specific washout ratio (Table H-1, Task 4). Additional data presented in this Task 5 report are intended to assist in validation of the environmental transport models.

#### SOURCES OF INFORMATION, METHODS, AND DATA FOR THE 1950s

The main sources of information for this analysis were the original analytical data sheets from National Lead Company of Ohio (NLCO 1953-1967) and the monthly/weekly reports from the Industrial Hygiene and Radiation (IH&R) Department. The analytical data sheets provided measurements of the concentrations of uranium, gross alpha, and gross beta in precipitation as well as precipitation amounts. The IH&R reports briefly discussed monitoring objectives and included some data for which analytical data sheets were not found. In addition, Klein and Ross (1966) indicate that the FMPC and the Division of Air Pollution Control of the city of Cincinnati had agreed to exchange information on uranium in rainwater.

There were only a limited number of measurements of wet deposition in the 1950s. The earliest records located of radioactivity in wet deposition were from the fourth quarter of 1953. The samples were collected in open "fallout trays," having a surface area of 9  $ft^2$  (0.836  $m^2$ ) which collected both rain and snow as well as dry deposition. These fallout trays should not be confused with the gummed-film fallout trays which were used well into the 1960s. Only three records of samples from five locations over the period 11/19/53 through 12/8/53 were located; these samples were analyzed for gross alpha, not uranium (Table B3-1). The measured deposition rates range from 6 to 4700 dpm alpha  $m^{-2} d^{-1}$ , with large differences observed between the alpha activity collected at the various locations. The concentrations ranged from 0.02 to 1.50 dpm alpha mL<sup>-1</sup>. It appears that this fallout tray sampling method was discontinued, as no other records of this type were found. However, a few other records of analyses of "clean snow" were located from the late 1950s, several associated with a metal oxide spill on January 26, 1956 (Table B3-2). This spill will be evaluated along with other episodic events in the final report of Tasks 2 and 3 of the dose reconstruction project. These data in Tables B3-1 and B3-2 are included here because the 1950s are very important to the dose reconstruction at Fernald. However, there are so few measurements, they may not be useful for validation purposes.

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			Water	•.	
Dates of		Brick House	Treatment		
Sampling	NE Area	Area <sup>b</sup>	Area (E)	SW Area	NW Area
Trays set out	$0.08 \text{ dpm mL}^{-1}$	0.03 dpm mL <sup>-1</sup>	0.02 dpm mL <sup>-1</sup>	0.04 dpm mL <sup>-1</sup>	1.50 dpm mL <sup>-1</sup>
11/19/53. Rained	860 dpm m <sup>-2</sup>	330 dpm m <sup>-2</sup>	$270  dpm  m^{-2}$	550 dpm m <sup>-2</sup>	19,000 dpm m <sup>-2</sup>
1 1/22/53. Collected	210 dpm m <sup>-2</sup> d <sup>-1</sup>	82 dpm m $^{-2}$ d $^{-1}$	$68 \text{ dpm m}^{-2} \text{ d}^{-1}$	140 dpm m <sup>-2</sup> d <sup>-1</sup>	4700 dpm m $^{-2}$ d $^{-1}$
11/23/53.					
Trays set out 11/23/53. Both snow and rain over the 10-day period were collected 12/3/53.	0.61 dpm mL <sup>-1</sup> 690 dpm m <sup>-2</sup> 69 dpm m <sup>-2</sup> d <sup>-1</sup>	0.54 dpm mL <sup>-1</sup> 610 dpm m <sup>-2</sup> 61 dpm m <sup>-2</sup> d <sup>-1</sup>	1.47 dpm mL <sup>-1</sup> 830 dpm m <sup>-2</sup> 83 dpm m <sup>-2</sup> d <sup>-1</sup>	0.11 dpm mL <sup>-1</sup> 63 dpm m <sup>-2</sup> 6 dpm m <sup>-2</sup> d <sup>-1</sup>	0.40 dpm mL <sup>-1</sup> 230 dpm m <sup>-2</sup> 23 dpm m <sup>-2</sup> d <sup>-1</sup>
Trays set out 12/3/53. Rained 12/5/53. Collected 12/8/53.	0.20 dpm mL <sup>-1</sup> 1000 dpm m <sup>-2</sup> 210 dpm m <sup>-2</sup> d <sup>-1</sup>	0.22 dpm mL <sup>-1</sup> 980 dpm m <sup>-2</sup> 200 dpm m <sup>-2</sup> d <sup>-1</sup>	1.0 dpm mL <sup>-1</sup> 4700 dpm m <sup>-2</sup> 950 dpm m <sup>-2</sup> d <sup>-1</sup>	0.12 dpm mL <sup>-1</sup> 540 dpm m <sup>-2</sup> 110 dpm m <sup>-2</sup> d <sup>-1</sup>	0.11 dpm mL <sup>-1</sup> 490 dpm m <sup>-2</sup> 98 dpm m <sup>-2</sup> d <sup>-1</sup>

### Table B3-1. Gross Alpha Content of Wet Deposition Samples Collected in Open Fallout Trays around the FMPC in 1953<sup>a</sup>

<sup>a</sup> Source: Analytical Data Sheets.

<sup>b</sup>Brick house is believed to be the old Coan farmhouse, located several hundred yards from the NE corner of the FMPC perimeter fence in a NE to E direction from the center of the FMPC. (See also Appendix B, Part 2 for air sampling results at this location.)

# Table B3-2. Uranium in Snow Following Metal Oxide Spill on January 26, 1956and in Hamilton Ohio in 1961

Date Collected	Location	Surface Area Collected	Uranium content (mg L <sup>-1</sup> )	Beta content (dpm mL <sup>-1</sup> )	Alpha content (dpm mL <sup>-1</sup> )	Reference
V2 <b>6/56</b>	Snow directly underneath metal oxide silos	25 in <sup>2</sup>	242	181,526	4198.8	IH&R weekly report dated 2/13/56
1/26/56	Snow 200 yds SW of metal oxide silos	25 in <sup>2</sup>	387	3104	390.0	IH&R weekly report dated 2/13/56
1/26/56	200 mL clean snow 100 yds W of drum baler on NE side of Project	25 in <sup>2</sup>	0.968	59.6	8.76	IH&R weekly report dated 2/13/56
1/26/56	200 mL clean snow between Plant 5 and 7 near gumpaper fallout tray	25 in <sup>2</sup>	2.033	92.9	22.99	IH&R weekly report dated 2/13/56
12/9/61	Hamilton, OH First heavy snow of 1961.	Not available.	0.005	0.68	0.05	Analytical Data Sheet





#### URANIUM CONCENTRATIONS IN PRECIPITATION IN THE 1960s

For the 1960s, a fairly complete set of original data sheets for uranium in wet deposition was located. The IH&R monthly report for October 1960 (Quigley 1960) refers to the new study whose objective was:

"...to determine the activity of rainwater. This will provide additional data since rainwater scavenges air particulates from the air; thus, determination of radioactivity in rainwater samples will provide a measure of ... increases or decreases in the atmospheric radioactivity level. It would be helpful if we could obtain an off-site collection of rainwater for background data. This is planned for the near future."

Rain and snow were collected and composited monthly from two locations, the East side of the Security Building at the FMPC and the Abbe Observatory in Cincinnati. The Security Building is located on the southern perimeter of the FMPC complex just west of D Street. The Abbe Observatory is a National Weather Service station located about 15 miles (24 km) south of the FMPC. Samples from the Abbe Observatory were analyzed for uranium concentration by the FMPC analytical department along with samples from the FMPC.

Precipitation samples were analyzed for total uranium concentration (mg  $L^{-1}$ ), as well as gross alpha and beta activity. The minimum detectable concentration for the fluorimetric method appears to be about 0.001 mg U  $L^{-1}$  (Dugan 1971). None of the rainwater concentrations are reported as "less than detectable," although several from the Abbe Observatory were reported as 0.001 mg U  $L^{-1}$ . It is not known whether or not the samples were pretreated or filtered before analysis.

In most cases, the volume of water collected was noted on the analytical sheet. At the FMPC only, the inches of water which fell during the month is also noted. These data are included in Table B3-3, following the main body of this part. The complete precipitation record for the FMPC is included in Table B3-4. Approximately 800 mL of water were collected per inch of rain (Table B3-3), which would represent a collection area of 315 cm<sup>2</sup>. The standard rain gauge has a diameter of 8 inches, or a 324 cm<sup>2</sup> opening. For the purposes of calculating deposition per unit area (see next section), a deposition area of 320 cm<sup>2</sup> was used.

Figure B3-1 illustrates the data for uranium concentration in precipitation collected from the FMPC and the Abbe Observatory (Cincinnati) in the 1960s. This data set represents 81 measurements at the FMPC and 53 from Cincinnati. The concentrations at the FMPC, ranging from 0.012 to 3.8 mg L<sup>-1</sup>, are generally 1-2 orders of magnitude higher than those from Cincinnati, which range from 0.001 to 0.023 mg L<sup>-1</sup>. The median concentration for the entire time period is 0.1 mg L<sup>-1</sup> at the FMPC and 0.003 mg L<sup>-1</sup> in Cincinnati. As noted above, the FMPC measurement capability was not able to detect concentrations lower than 0.001 mg L<sup>-1</sup>, as illustrated by the truncation of the data at that level (Figure B3-1).

The FMPC data set was used to determine the washout ratio to be used in the model for assessing transport of releases from the FMPC (Killough et al. 1993). However, the Cincinnati data are independent of the FMPC data and can be used as a model validation data set. For model validation, the predicted concentrations of uranium in rain at the location of the Abbe Observatory, using reconstructed source terms and the transport model, will be compared with the measured values shown in Figure B3-1. This comparison will be included with other model validations in the final Task 6 report.



Figure B3-1. Concentration of uranium in precipitation from the FMPC Security Building and the Cincinnati Abbe Observatory in the 1960s.

Background concentrations of uranium in rain have been measured by the Environmental Protection Agency and reported in their Environmental Radiation Data reports (EPA 1981–1988). The most appropriate data for comparison in this study were collected from Columbus, Ohio, and are illustrated in Figure B3–2. The data are the sum of three isotopes of uranium ( $^{234}$ U,  $^{235}$ U, and  $^{238}$ U), which are reported separately in the EPA reports. The median of these measurements, made in the 1980s, is 0.00007 mg uranium L<sup>-1</sup>, with a 95% confidence range of 0.00004 to 0.0003. Similar concentrations were reported for 21 air sampling sites throughout the U.S. in the 1973–1976 period (EPA 1977). The FMPC monitoring procedure was unlikely to be able to monitor at these low levels, but they do give perspective as to normal background levels of uranium in wet deposition.

#### WET DEPOSITION RATE OF URANIUM

The wet deposition of uranium to the ground depends not only on the concentration in precipitation but also on the amount of precipitation that falls during a particular time 0001239

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interval. The deposition rate (uranium deposited per unit area per unit time) is computed by multiplying the uranium concentration in rain by the total volume of precipitation collected and dividing by the surface area of the collector and the sampling time. Figures B3-3 and B3-4 illustrate the monthly wet deposition rate at the FMPC and at the Abbe Observatory in Cincinnati. It was assumed that the Cincinnati collector was also a standard rain collector with an 8-in diameter. The measured concentration of uranium in air at the closest perimeter station at Fernald (about 200 m from the precipiation collector) is superimposed on the wet deposition bar chart, to illustrate the relationship between the uranium in air and the wet deposition rate (Figure B3-3).



Figure B3-2. Background concentrations of total uranium in rain in Columbus, Ohio in the 1980s.

Some seasonal trends are apparent in Figures B3-3 and B3-4 — higher depositions tend to occur in the winter and spring. Months with high rainfall can result in relatively high deposition rates even though the concentration in rain that month is moderate to low (e.g. March 1963, March 1964). During the 1961-1967 period, all of the monthly depositions >15 mg m<sup>-2</sup> mo<sup>-1</sup> at the FMPC occurred between November and April, and all of those >10 mg m<sup>-2</sup> mo<sup>-1</sup> occurred between November and May.

These seasonal differences can not be accounted for solely by precipitation quantities. Typically, March does have the highest monthly precipitation. However, July has the second highest monthly precipitation, yet deposition rates during that month are relatively low. A statistical analysis of the entire data set showed that the wet deposition rate at the FMPC is not strongly correlated with monthly rainfall amount. Other factors must play important roles in the wet deposition process, such as precipitation rate and timing, precipitation type, wind speed and direction, atmospheric stability, and source term quantity and characteristics.

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Figure B3-3. Wet deposition rate of uranium at the FMPC in the 1960s — comparison to air monitoring results at the SE perimeter station.

In addition to the seasonal dynamics, a longer term temporal trend is also suggested by the data in Figure B3-3. In 1965, 1966, and 1967, only two months (January 1965 and November 1967) had a monthly deposition of greater than 10 mg m<sup>-2</sup> mo<sup>-1</sup>, whereas that rate was exceeded during 14 months of the previous four years (1961-1964). Further investigations of the relationship between the estimated source term and the environmental monitoring data, including this wet deposition data set, are continuing.

The uranium in air measurements from the SE perimeter station do show some similarities to the wet deposition trends (Figure B3-3). The combination of the relatively high air concentration and the high rainfall in April 1961 probably contributed to this being one of the highest months for wet deposition. In March 1964, the precipitation rate was the highest for this entire time period (over 11 inches in that month), and the air concentration was also relatively high, resulting in the second highest wet deposition rate of 48 mg U m<sup>-2</sup> mo<sup>-1</sup>. Months with the lowest air concentrations almost always show low wet deposition rates, yet some months with relatively high air concentrations also show low deposition (e.g. August 1961). Again, there are a number of contributing factors to the wet deposition rate which can not be easily separated and quantified.



Figure B3-4. Wet deposition rate of uranium at the Cincinnati Abbe Observatory in the 1960s.

#### SUMMARY

The historic records of measurements of uranium in precipitation at the FMPC and at the Abbe Observatory, 24 km S of the FMPC, have provided useful information for the dose reconstruction project. The FMPC data were used in conjunction with air monitoring results to quanitify the washout ratio, a parameter which is used in the environmental transport model (Killough et al. 1993). An independent data set of measurements at the Abbe Observatory will provide an opportunity for validation of the model predictions in the final Task 6 report.

The concentrations of uranium in precipitation at the FMPC, ranging from 0.012 to 3.8 mg  $L^{-1}$ , are generally 1-2 orders of magnitude higher than those from the Abbe Observatory in Cincinnati, which range from 0.001 to 0.023 mg  $L^{-1}$ . The median uranium concentrations for December 1960 through December 1967 are 0.1 mg  $L^{-1}$  at the FMPC and 0.003 mg  $L^{-1}$  in Cincinnati. The difference in uranium concentrations in air at these two locations is clearly due to FMPC releases. Background concentrations of uranium in rain are on the order of 0.00007 mg uranium L<sup>-1</sup>, based on EPA measurements in the 1980s from Columbus, Ohio.

Higher wet deposition rates occur in the winter and spring. The total deposition rates, ranging up to a maximum of 69 mg U m<sup>-2</sup> mo<sup>-1</sup> (2.2 mg U m<sup>-2</sup> d<sup>-1</sup>) in December 1960, are lower than those measured by the gummed-film (Appendix B Part 1). The median deposition rate measured by gummed-film at the SE perimeter station (closest to the rainfall collection 000132

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point) during 1961-1964 was 7.0 mg m<sup>-2</sup> d<sup>-1</sup>, whereas the median deposition rate measured in precipitation over the same period was 0.3 mg m<sup>-2</sup> d<sup>-1</sup>. It is not entirely clear why the two measurement results are not in closer agreement, given that they both measure dry and wet deposition, to some extent. Perhaps the open rainfall collector was not particularly efficient for intercepting and retaining dry deposition. Perhaps particulate material was filtered from the rainfall before analysis. Another possibility is that the collection efficiency of gummedfilm for particulates is higher than we thought (see Appendix B, Part 1). Regardless, it does appear that dry deposition processes were more important than wet deposition processes, for the particle sizes found in the vicinity of the FMPC perimeter (Killough et al. 1993).

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	Uranium in Precipitation							Deposition Rate	
	(mg l	U L <sup>-1</sup> )		FMPC	FMPC	mL Precip.	Abbe	(mg m <sup>-</sup>	$2 mo^{-1}$
Month	FMPC	Abbe		Vol. (mL)	Inches	per Inch	Vol. (mL)	FMPC	Abbe
Dec-60	1.6	NA <sup>b</sup>	a	1384	1.73	NA	NA	69.2	NA
Jan-61	0.4	NA	a	1056	1.32	NA	NA	13.2	NA
Feb-61	0.12	NA	a	2864	3.58	NA	NA	10.7	NA
Mar-61	0.13	NA		4190	5.17	810	NA	17.0	NA
Apr-61	0.4	NA		3035	3.63	836	NA	37.9	NA
May-61	0.062	NA		4750	6.16	771	NA	9.2	NA
Jun-61	0.028	0.009	a	2960	3.7	NA	NA	2.6	NA
Jul-61	0.03	0.001	a	7000	8.75	NA	NA	6.6	NA
Aug-61	0.02	0.007	a	1616	2.02	NA ·	NA	1.0	NA
Sep-61	0.023	0.023		2420	3.05	793	335	1.7	0.24
Oct-61	0.044	0.001		1400	1.71	819	NA	1.9	NA
Nov-61	0.11	0.003	a	2816	3.52	NA	NA	9.7	NA
Dec-61	0.065	0.008		2300	3.08	747	NA	4.7	NA
Jan-62	0.04	0.0015	a	2776	3.47	NA	NA	3.5	NA
Feb-62	0.029	0.001	a	3680	4.6	NA	NA	3.3	NA
Mar-62	0.13	0.002	a	2648	3.31	NA	NA	10.8	NA
Apr-62	0.58	0.022	a	456	0.57	NA	NA	8.3	NA
May-62	0.18	0.003	a	<b>3344</b>	4.18	NA	NA	18.8	NA
Jun-62	0.12	0.005	a	832	1.04	NA	NA	3.1	NA
Jul-62	0.018	0.003	a	5304	6.63	800	NA	3.0	NA
Aug-62	0.12	0.002	a	1776	2 <b>.22</b>	NA	NA	6.7	NA
Sep-62	0.27	0.008	a	1024	1.28	NA	NA	8.6	NA
Oct-62	0.11	0.002	a	2408	3.01	NA	NA	8.3	NA
Nov-62	NL	0.02	a	1528	1 <b>.91</b>	NA	NA	NA	NA
Dec-62	0.65	0.02	a	984	1.23	NA	NA	20.0	,NA
Jan-63	0.27	0.02	a	1240	1 <b>.55</b>	NA	NA	10.5	NA
Feb-63	1	0.012	a	536	0.67	NA	NA	16.8	NA
Mar-63	0.08	0.02		6090	9.78	623	4195	15.2	2.62
Apr-63	0.2	0.01		2250	2.8	804	1000	14.1	0.31
May-63	0.13	0.01	a	3024	3.78	NA	1000	12.3	0.31
Jun-63	0.1 <b>6</b>	0.01		1055	1.34	787	1570	5.3	0.49
Jul <b>-63</b>	0.07	0.023	a	2704	3.38	NA	NA	5.9	NA
Aug-63	0.05	0.002	a	3064	3.83	NA	1800	4.8	0.11
Sep-63	NL	0.005		375	0.45	833	406	NA	0.06
Oct-63	3.8	0.003		34	0.04	850	72	4.0	0.01
Nov-63	0.44	0.005		660	0.75	880	575	9.1	0.09
Dec-63	0.44	0.005		710	0.86	826	755	9.76	0.12
Jan-64	0.22	0.003		1170	1.96	597	2230	8.0	0.21
Feb-64	1.15	0.008		990	1.21	818	1150	35. <del>6</del>	0.29
Mar-64	0.2	0.01		7690	11.15	690	8640	48.1	2.70
Apr-64	0.215	0.002		6100	7.46	818	5180	41.0	0.32
May-64	0.145	0.02		640	0.8	800	860	2 <b>.9</b>	0.54
Jun-64	0.07	0.001		4410	5.91	746	NA	9.6	NA
Jul-64	0.11	NLC		2050	2.53	810	1500	7.0	NA
Aug-64	0.095	NL		1210	1.47	823	1530	3.6	NA
Sep-64	NL	0.007		1500	2.2	682	1900	NA	0.42

Table B3-3. Uranium in Precipitation from the FMPC and the Abbe Observatory

(continued next page)

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					cont.)					
-	Uranium in Wet Deposition (mg U L <sup>-1</sup> )			FMPC	FMPC	mL Precip.	Abbe	Deposit	Deposition Rate	
								$(mg m^{-2} mo^{-1})$		
Month	FMPC	Abbe	_	Vol. (mL)	Inches	per Inch	Vol. (mL)	FMPC	Abbe	
Oct-64	0.25	0.007		550	0.67	821	550	4.3	0.12	
Nov-64	0.4	0.003		1700	2.09	813	2080	21.2	0.20	
Dec-64	0.17	0.001		3170	4.07	· 779	4160	16.8	0.13	
Jan-65	0.21 d	0.001	a	2752	3.44	NA	3150	18.1	0.10	
Feb-65	0.084	0.001		4330	4.23	1024	4000	11.4	0.13	
Mar-65	0.095	0.011		2175	2.65	821	2390	6.5	0.82	
Apr-65	0.04 <sup>d</sup>	0.001		NA	5.86	NA	4600	5.9	0.14	
May-65	0.115	0.001		465	0.82	567	465	1.7	0.01	
Jun-65	0.15	0.001		1880	2.38	790	1240	8.8	0.04	
Jul-65	0.03	0.022		2540	3.24	784	2750	2.4	1.89	
Aug-65	0.05	0.002 <sup>d</sup>		2710	3.3 <b>9</b>	799	2210 <sup>d</sup>	4.2	0.14	
Sep-65	0.04	0.001		5320	6. <b>56</b>	811	5560	6.6	0.17	
Oct-65	0.025	0.001		2705	3.31	817	1900	2.1	0.06	
Nov-65	0.22	0.001		1060	1.36	779	1125	7.3	0.04	
Dec-65	0.205	0.001		1140	1. <b>39</b>	820	118	7.3	0.00	
Jan-66	0.085	NA		2450	3	817	NA	6.5	NA	
Feb-66	0.075	NA		2550	3.05	836	NA	6.0	NA	
Mar-66	0.23	NA		743	0.93	7 <b>99</b>	NA	5. <b>3</b>	NA	
Apr-66	0.034	NA		3945	4.83	817	NA	4.2	NA .	
May-66	0.075	NA		2550	2.87	889	NA	6.0	NA	
Jun-66	0.045	NA		2350	3.16	744	NA	3.3	NA	
Jul-66	0.016	NA		2100	2.56	820	NA	1.0	NA	
Aug-66	0.023	NA		2440	2. <b>99</b>	816	NA	1.8	NA	
Sep-66	0.013	NA		3580	4.42	810	NA	1.4	NA	
Oct-66	NA	NA	a	624	0.78	800	NA	NA	NA	
Nov-66	0.022	NA		3100	4.19	740	NA	2.1	NA	
Dec-66	0.012	NA		2500	3.11	804	NA	0.9	NA	
Jan-67	0.36	NA		375	0.44	852	NA	4.2	NA	
Feb-67	0.1	NA		1400	1.75	800	NA	4.4	NA	
Mar-67	0.05	NA		2700	3.37	801	NA	4.2	NA	
Apr-67	0.017	NA		3430	4.24	809	NA	1.8	NA	
May-67	0.018	NA		4800	5.86	819	NA	2.7	NA	
Jun-67	0.07	NA		1750	<b>2.1</b> ·	833	NA	3.8	NA	
Jul-67	0.034	NA		2875	3.58	803	NA	3.0	NA	
Aug-67	0.043	NA		465	0.58	802	NA	0.6	NA	
Sep-67	0.028	NA		1078	1. <b>36</b>	793	NA	0.9	NA	
Oct-67	0.022	NA		2050	2.51	817	NA	1.4	NA	
Nov-67	0.13	NA		3052	3.74	816	NA	12.4	NA	
Dec-67	0.095	NA		2780	3.42	813	NA	8.2	NA	
Average	0.22	7.0E-03				797				
Std. Dev.	0.48	7.2E-03				65				
Number	81	53				60				
Median	9.5 E-02	3.0E-03				810				

# Table B3-3. Uranium in Precipitation from the FMPC and the Abbe Observatory

<sup>a</sup> FMPC volume estimated from inches recorded  $\times$  800 mL per inch.

b NA = Not available.

<sup>c</sup> NL = Data sheet not legible.

 $^{d}$  Analytical data sheet not located. Data presented in Ross and Klein (1966).



#### Table B3-4. Precipitation Amounts (inches) Recorded at the Feed Materials Production Conter 1960 1991

			Г	rouucu	on ter	iver, 19	00 - 13	71			
Month	1960	1961	1962	1 <b>963</b>	1 <b>964</b>	1965	1966	1967	1968	1969	1970
January		1.32	3.47	1.55	1.96	3.44	3.00	0.44	1.36	4.19	0.98
February		3.58	4.60	0.67	1.21	4.23	3.05	1.75	0.30	1.14	1.51
March		5.17	3.31	9.78	11.15	2.65	0.93	3.37	3.38	0.91	3.74
April	1.24	3.63	0.57	2.80	7.46	5.86	4.83	4.24	2.02	2.97	5.20
May	3.92	6.16	4.18	3.78	0.80	0.82	2.87	5.86	10. <b>36</b>	2.20	1.96
June	6.04	3.70	1.04	1.34	5.91	2.38	3.16	2.10	2.71	3.17	3.11
July	4.60	8.75	6.63	3.38	2.53	3.24	2.56	3.58	5.22	3.58	4.18
August	1. <b>9</b> 8	2.02	2.22	3.83	1.47	3.39	2.99	0.60	2.61	2.95	1.83
September	0. <b>9</b> 1	3.05	1.28	0.45	2.20	6.56	4.42	1.36	3.51	5. <b>26</b>	3.74
October	2.07	1.71	3.01	0.04	0.67	3.31	0.78	2.51	1.24	1.53	3.11
November	2.06	3.52	1. <b>9</b> 1	0.75	2.09	1. <b>36</b>	4.19	3.74	3.37	3.31	2.08
December	1.73	3.08	1.23	0.86	4.07	1. <b>39</b>	3.11	3.42	3.49	2.46	2.87
Total	24.55	45.69	33.45	29.23	41.52	38.63	35.89	32.97	39.57	33.67	34.31
Month	1 <b>97</b> 1	1972	1 <b>973</b>	1974	1975	1976	1 <b>977</b>	1978	1 <b>979</b>	1980	1981
January	1.87	1.70	1.79	2.65	3.07	3.09	1.60	3.73	3.38	2.04	0.17
February	4.06	1.08	1.07	2.06	3.94	1.85	1.18	0.19	3.17	1.24	3.28
March	1. <b>95</b>	2.84	5.31	2.90	6.19	2.01	4.15	2.21	1.10	3. <b>29</b>	1.42
April	1.11	5.49	4.49	5.56	2.69	0.94	2.73	2.58	4.22	1. <b>50</b>	4.55
Мау	3.22	4.87	4.75	4.92	2.47	1.33	3.27	4.02	2. <del>94</del>	3.98	4.14
June	3.84	1. <b>99</b>	6.48	4.21	3.47	4.87	3.57	5.80	4.47	3.04	3.85
July	3.28	1.88	7.61	1.17	1.50	1.89	1.65	4.58	4.05	8.16	3.85
August	3.05	1. <b>98</b>	3.24	7.09	4.72	5.59	5.1 <b>6</b>	4 <i>.</i> 99	6.09	4.54	3.25
September	4.35	4.27	1.40	5.24	4.22	3.71	1.33	0.54	7.37	0.88	2.43
October	1.70	3.12	4.62	1.03	4.40	3.23	5.70	3.23	1. <b>48</b>	3.26	2.08
November	1.33	5. <b>46</b>	4.80	3.71	1.71	0.61	3.37	2.22	4.72	2.09	2.91
December	3.10	4.00	2.16	1.89	3.05	0.41	4.47	5.10	2.50	0.87	2.27
Total	32.86	38.68	47.72	42.43	41.43	29.53	38.18	39.19	45.44	34.89	34.20
											1 <b>960-199</b> 1
-	1982	1983	1984	1985	1986	1987	1988	1989	1990	1991	Average
January	6.05	1.38	0.66	0.74	0.90	0.82	2.32	1.82	3.27	2.37	2.16
February	1.20	0 <b>.62</b>	2.36	1.78	3.01	1.08	3.65	4.88	4.80	3.40	2.32
March	4.18	2.37	2.88	5.34	3.09	2.85	2.99	5.18	2.59	4.34	3.66
April	1.43	4.20	3.96	1.30	1.61	2.33	1 <b>.97</b>	6.86	3.11	4.48	3.37
May	4.79	7. <b>9</b> 5	3.56	4.85	2.06	2.14	0.40	5.28	9.81	2.61	3.95
June	3.51	1.56	1.49	2.77	3.44	3.00	0.16	2.74	4.02	0.17	3.22
July	2.11	2.33	3.29	3.76	3.01	5.47	3.20	4.21	3.65	2.58	3.80
August	1.98	1.20	1.71	4.38	2.82	1.11	2.44	4.57	3.40	3.43	3.21
September	0.81	0.55	3.17	0.54	5.87	1.09	1. <b>66</b>	1. <b>50</b>	3.30	2.13	2.78
October	0.62	7.34	2.81	3.82	2.64	1.05	2.92	2.41	6.74	1.14	2.67
November	4.23	3. <b>69</b>	4.25	8.98	3.67	1.54	3.89	2.86	2.03	1.07	3.05
December	3.62	2.47	3.84	2.41	2.72	2.41	2.69	1. <b>59</b>	7.01	3.19	2.80
Total	34.53	35.66	33.98	40.67	34.84	24.89	28.29	43.90	53.73	<b>30.9</b> 1	36.73

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#### APPENDIX B -- REGIONAL ENVIRONMENTAL MONITORING

#### PART 4 — ADDITIONAL SOIL MONITORING DATA

#### INTRODUCTION

Appendix N of the Task 4 report (Killough et al. 1993) and Appendix A of this report both present data on uranium in soil. The purpose of the Task 4 report Appendix N was to estimate the range of the uranium source term by a method other than those addressed in the Task 2/3 report (Voillequé et al. 1991). Hence it may serve as an independent check of the final atmospheric source term developed by this dose reconstruction study.

Appendix A of this report had as its goal the determination of the background levels of uranium around the FMPC. It remains for this Appendix to report upon uranium levels not reported elsewhere in the FMPC dosimetry reconstruction task reports; to illustrate the soil concentrations of uranium with depth; and, to discuss the occurrence of other radionuclides in the soil around the FMPC. The other radionuclides include  $^{226}$ Ra and thorium among the naturally occurring isotopes, and  $^{99}$ Tc,  $^{137}$ Cs,  $^{90}$ Sr and  $^{239}$ Pu among the man-made radionuclides.

#### URANIUM

Uranium concentrations in soil as a function of depth may be observed from three sets of soil data collected around the FMPC. These data sets are: the EG&G measurements taken in 1985 (Shipman 1985), the SOIL-13-86 data set (IT Corp. 1986), and the RIFS-1988 (RIFSSOIL 1988) data set, which is the most complete of the three.

#### Uranium Geographic Distribution Data in Soil

Studies of uranium in surface soil have been conducted around the FMPC prior to this dose reconstruction project. Appendix N of the Task 4 Report of the Dosimetry Reconstruction Project (Killough et al. 1993) used soil data for locations near the FMPC to make an independent estimate of uranium depositions around the FMPC. Some of the material found in that appendix is repeated here to provide a full picture of uranium in soil around the FMPC, and specifically to estimate the natural uranium background in soil in the vicinity of Fernald, Ohio. Makhijani (1988, 1989) used soil data to estimate uranium releases from the FMPC. As part of this dose reconstruction project, data from previous analyses have been reviewed (Shleien 1991; and Appendix G, this report).

Data not previously discussed in Task 4, Appendix N (Killough et al. 1993) appeared in a database (computer disk) from the IT study (IT Corp., undated). This database has been referred to as "SOIL-13" and consists of several separate groups of data entries. The database was analyzed in our initial review of historic soil measurements relevant to the FMPC (Shleien 1991). Those discussed here are the SOIL-13 file, which contains data from the 1984 survey by FMPC reported in the FMPC 1984 annual report (below), and the 1986 survey contained in the IT Report. The 1986 study was undertaken as an independent assessment of the 1984 FMPC soil sampling data. Shleien (1991) concluded that the 1984 (and 1986) data can be used with confidence to describe the regional distribution of uranium in soil.

The data reported here as "SOIL-13-84" contains results for 138 samples analyzed for total uranium. The "SOIL-13-86" data contains results for uranium and uranium nuclides as well as thorium, <sup>232</sup>Th, <sup>226</sup>Ra, <sup>99</sup>Tc, <sup>90</sup>Sr, <sup>137</sup>Cs, and <sup>239,240</sup>Pu. The IT report notes that as of April 12, 1986, 939 samples had been collected at 311 locations (and vegetation had been sampled at 235 of these locations). The computer disk file contained results for 426 soil samples for uranium.

There is not a detailed description of the sampling procedures used in the IT Report or the FMPC annual reports. The SOIL-13-84 data is reported in the IT report as having been collected at 2-15 cm, but is given on the disk as 1-6 inches (value used here). The SOIL-13 1986 includes samples taken at a depth of 0-5 cm, except for some sampling by 2.5-cm increments to a depth of 15 cm at a location near the incinerator and in a background area. These data form the bases for Figures B4-1 and B4-2, which show the geographic distributions of total uranium in surface soil for two of the databases yielding the most complete geographic distributions (SOIL-13-86 and SOIL-13-84). The average concentration of total uranium in soil is listed as a function of distance and direction from the air emissions center of the FMPC (see Killough et al. 1993, Appendix N for geographic coordinates).

The general geographic distribution patterns observed from the two databases discussed here are similar. The results show concentrations which are clearly elevated above background, in the NE quadrant out to distances of about 8 km. The highest concentrations are found within 1 km of the emissions center. Since winds to the northeast are about twice as frequent as those to other quadrants, it can be concluded that these elevated levels represent the deposition of uranium released to the air from FMPC activities. With regard to levels to the southwest, these may represent distribution by run-off and production activities. It further appears that many of the high samples within about 1 km represent contamination by industrial activity, such as localized spills.

	Distance from FMPC Air Emission Center (km)													
Direction	0-1	1-2	2-3	3-4	4-5	·5-6	6-7	7-8	8-9	9-10				
N		7.7		3.5	2.1	1.2	0. <del>9</del>	0. <b>9</b>	1.2					
NNE		7.3		4.4	1.8	2.3	2.1	1.3	1.1					
NE			2.3	1.7	1.1	1.3	2.2	1.3	0.8					
ENE			2.5	1.7	1.8	1.2	1.3	1.6						
Е	59.1	8.7	1.7	2.5	2.0	1.8	1.2	1.1						
ESE	30.6	2.5	1.5	1.1	1.5	1.0	0.7	0.9	1.2					
SE		3.7		1.6	1.0	1.1	0.7	0.9	1.0					
SSE	25.1	3.5	1.5	1.1	1.0	0.9	1.2	1.1						
S		20.7	3.6	1.8	1.2	0.7	1.1	0.8	0.8					
SSW		4.2	0.8	1.5	0.9	1.4	1.3							
SW		6.5	5.0	1.3		1.0	1.0							
WSW			1.6	2.7	2.8	2.2	1.4	1.3	1.3					
w	6.3	8.5	0.6	2.0		0.8	1.1							
WNW														
NW		15.7		1.3	0.9	1.1	1.2	0.8	0.8	0.9				
NNW	35.6	2.6	3.8	2.2	1.0	0.9	1.0	1.3	1.3					

Figure B4-1. Average total uranium concentrations in soil (pCi g-1; depth 0-5 cm) by sector and 1-km distance increment. Data are from the "SOIL-13-86" database. Concentration ranges are indicated by shading: 0-2 (none), 2-4 (light shaded area) and  $\geq$  4 (darker shaded area). A blank space means no data are available for that sector.



Figure B4-2. Average total uranium concentrations in soil (pCi  $g^{-1}$ ; depth 1-6 in.) by sector and 1-km distance increment. Data are from the "SOIL-13-84" database. Concentration ranges are indicated by shading: 0-2 (none), 2-4 (light shaded area) and  $\geq$  4 (darker shaded area). A blank space means no data are available for that sector.

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At a later time, additional databases were made available to the Fernald Dosimetry Reconstruction Project and were reviewed in the Annex to Shleien (1991). During 1985– 1989, the FMPC staff conducted various sampling programs that included uranium in soil as well as grass, vegetation, and produce. The soil data from those studies, including the routine soil samples were analyzed (Shleien 1991) and include:

- Parallel Sampling Locations (Grass and Soil) 1985
- Farm/Garden Produce (Soil and Fertilizer) 1986, 1987
- Parallel Soil and Vegetation (0-5 and 5-10 cm cm depth) 1987, 1988
- Soil and Grass (0-5 and 5-10 cm depth) 1989
- Soil and Produce (0-5 cm depth) 1988
- Routine Soil (0–5 and 5–10 cm depth) 1986, 1987

These data have been used for composition of Figure B4-3. The geographic distribution is similar to Figures B4-1 and B4-2, but areas of higher uranium concentrations are evident NW of the plant at distances greater than 5 km.



Figure B4-3. Distribution of uranium in soil (pCi g<sup>-1</sup>; depth 0-5 cm) based on the "FMPC 1986-1989" database (Shleien 1991). Darker shading represents average concentrations of > 5 pCi g<sup>-1</sup>; lighter shading represents concentrations > 3 but < 5 pCi g<sup>-1</sup>. No shading represents < 3 pCi g<sup>-1</sup>. A blank space means no data are available for that sector.

Figures B4-1 through B4-3 highlight some areas of high contamination on-site. These areas could be due to spills of uranium-bearing materials or waste, or from airborne deposition (also see Figure N-3, Killough et al. 1993). The area immediately to the east is characterized by the presence of the old solid waste incinerator (OSWI) which is definitely a source of localized deposition from airborne uranium. Extension of the ground

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contamination pattern in a NE direction lends credence to this assumption. Contamination in the ESE and SSE are not in the direction of the prevailing winds. These areas are likely due to waste materials, not from airborne deposition. The NNW contamination is associated with the Plant 1 on-site storage area. A metal scrap area, the tank farm, and an unidentified source NE of Plant 9 all show high levels of contamination. Except for the incinerator area, it appears reasonable to attribute levels of uranium in soil above about 100 pCi g<sup>-1</sup> to causes other than airborne deposition.

#### Distribution of Uranium with Depth in Soil

Distribution of  $^{238}$ U and total uranium with depth is illustrated in Figure B4-4 and B4-5 respectively. The samples are in approximately the same location. The sample collected later in time (B4-5) shows a lower concentration and a steeper decrease in concentration with depth. Whether this is due to further weathering or because of the small difference in location is unknown.

The RIFS-1988 data contains samples for the 0-6 inch soil layer (divided into three, 2-inch strata), and the 0-18 inch soil layer (divided into three, 6-inch strata). Figure B4-6 shows the ratio of  $^{238}$ U to the 0-6 inch layer as a function of depth for relatively uncontaminated samples. A similar figure (B4-7) is given for highly contaminated samples. The locations are different, and not a great deal may be discerned from the comparison of the figures, except a decrease in concentration with increasing depth. In Figure B4-8, the 2-4 inch strata show a somewhat higher uranium level for several samples than do the 0-2 and 4-6 inch strata.



Figure B4-4. Concentration of <sup>238</sup>U versus depth in soil. Data (from Shipman, 1985) are for samples collected at 97 degrees and 0.70 km from the FMPC air emissions center.

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Figure B4-5. Concentration of total uranium versus depth in soil. Data (from Soil 13, 1986) are for samples collected at 85 degrees and 0.73 km from the FMPC air emissions center.

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**Figure B4-6.** Relative concentration of  $^{238}$ U in two deeper soil layers as compared to concentration in surface (0-6 inch) layer. Data (from RIFS 1988) are for samples in which the  $^{238}$ U in surface layer was < 50 pCi g<sup>-1</sup>.


**Figure B4-7.** Relative concentration of  $^{238}$ U in two deeper soil layers as compared to concentration in surface (0-6 inch) layer. Data (from RIFS 1988) are for samples in which the  $^{238}$ U in surface layer was > 1000 pCi g<sup>-1</sup>.

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**Figure B4-8**. Relative concentration of  $^{238}$ U in two deeper soil layers as compared to concentration in surface (0-2 inch) layer. Data (from RIFS 1988) are for samples in which the  $^{238}$ U in surface layer was >100 pCi g<sup>-1</sup>.

The depth data generally show the effect of environmental leaching of uranium. Those samples that do not follow the general pattern of decreasing concentration with depth may represent areas of soil mixing or, less likely, an underground source of uranium.

#### **GEOGRAPHIC DISTRIBUTION OF RADIUM-226 AND THORIUM**

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Tables B4-1 and B4-2 present surface soil values for <sup>226</sup>Ra and total thorium respectively. Although many locations lack specific data, no geographic patterns with distance or direction can be discerned. Data on <sup>226</sup>Ra and <sup>232</sup>Th collected in twelve samples of Ohio surface soil samples had arithmetic mean concentrations of  $1.5 \pm 0.93$  and  $1.0 \pm 0.50$  pCi g<sup>-1</sup> (uncertainties at  $2\sigma$  level) for <sup>226</sup>Ra and <sup>232</sup>Th, respectively (Myrick et al. 1993). The mean <sup>226</sup>Ra concentration in limestone is 0.42 pCi g<sup>-1</sup> and 1.3 pCi g<sup>-1</sup> in igneous rock (Eisenbud 1987). The average concentration of <sup>232</sup>Th in the upper crust of continental soils is 1.2 pCi g<sup>-1</sup>, although granitic and salic igneous rock may be about twice this level (NCRP 1987). Other thorium isotopes, namely <sup>233</sup>Th, <sup>234</sup>Th and <sup>231</sup>Th may contribute to the total thorium levels reported in Table B4-2. The data in Table B4-1 and B4-2 are within the range of values reported in the scientific literature.

Given the lack of differences in the geographic distribution of <sup>226</sup>Ra and total thorium with distance from the site center. and the fact that levels are within the range of the natural occurrence of these isotopes. it cannot be concluded that their source is other than from natural sources.

#### IAN-MADE RADIONUCLIDES

In order to assess the releases of any other radionuclides, the results for  $^{137}$ Cs,  $^{90}$ Sr.  $^{99}$ Tc, and  $^{239.240}$ Pu in file RIFSSOIL (RIFSSOIL–1988) have been examined. In many cases the results reported for these radionuclides are less than (<) values indicating the actual level was below the minimum sensitivity of the measurement procedure. Such "less than" values were not used in the analysis since the actual level is unknown. Sixteen sector averages were calculated for each of these radionuclides, excluding samples with a "less-than" designation. Not including those samples noted as "<" would tend to raise the average results for these radionuclides, but does not change our conclusion regarding distribution or source.

#### **TECHNICIUM-99**

The results for  $^{99}$ Tc, where few samples were available (none off-site) are shown in Table B4-3. The table is cut off at 2 km because no data were available beyond this distance. The eastern and southwestern sectors showed high concentrations of this radionuclide. (These sectors also have high values of 228 pCi g<sup>-1</sup> and 320 pCi g<sup>-1</sup> respectively.) Not a great deal can be said about the occurrence of these levels of  $^{99}$ Tc other than to note their presence and indicate that local soil contamination is their most likely origin. It is noted later in this task report that  $^{99}$ Tc in waste water at the FMPC may have originated by run-off of  $^{99}$ Tc from soil leaching and contamination into the waste water (see Appendix F this report).

							(nchu	v-2 m)								
		0 <u>_1 km</u>		<u>n</u>	<u>1–2 km</u>		2	<u>2–3 kr</u>	<u>n</u>		<u>3–4 kr</u>	<u>n</u>	4	4– <u>5 k</u>	m	
	Sectors	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS
N	(>348.75,<=11.25	) 0.68	13	0.22	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
NNE	(>11.25,<=33.75)	NA	0	NA	NA	0	NA	0.75	3	0.21	0.63	2	0.43	0.46	1	NA
NE	(>33.75,<=56.25)	NA	0	NA	NA	0	NA	NA	0	NA	0.79	1	NA	NA	0	NA
ENE	(>56.25,<=78.75)	NA	0	NA	0.53	2	0.18	NA	0	NA	NA	0	NA	NA	0	NA
Е	(>78.75,<=101.25	) NA	0	NA	0.57	2	0.08	NA	0	NA	NA	0	NA	0.76	2	0.02
ESE	(>101.25,<=123.7	5) NA	0	NA	NA	0	NA	0.61	1	NA	0.85	4	0.05	0.70	3	b.12
SE	(>123.75,<=146.2	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
SSE	(>146.25,<=168.7	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
S	(>168.75,<=191.2	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
SSW	(>191.25,<=213.7	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	0.78	2	0.15
SW	(>213.75,<=236.2	5) NA	0	NA	NA	2	0.46	0.77	1	NA	0.52	12	0.10	NA	0	NA
wsw	(>236.25,<=258.7	5) NA	0	NA	NA	0	NA	0.49	1	NA	NA	0	NA	0.61	3	0.05
W	(>258.75,<=281.2	5) NA	0	NA	NA	0	NA	0.98	1	NA	NA	0	NA	NA	0	NA
WNW	(>281.25,<=303.7	5) NA	0	NA	NA	0	NA	0.72	1	NA	NA	0	NA	NA	0	NA
NW	(>303.75,<=326.2	5) NA	0	NA	NA	1	NA	NA	0	NA	NA	0	NA	NA	0	NA
NNW	(>326.25,<=348.7	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA

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## Table B4-1. SOIL-13 1986 Ra-226 (pCi g<sup>-1</sup>) 16 Sector Average (Depth (-2 in)

			5_6 km6-		<u>–7 km7–8 km _</u>		n	8–9 km			9–10 km					
	Sectors	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS
N	(>348.75,<=11.25	) NA	0	NA	0.67	4	0.19	0.96	3	0.36	NA	0	NA	NA	0	NA
NNE	(>11.25,<=33.75)	0.72	4	0.19	0.86	7	0.20	0.93	2	0.33	NA	0	NA	NA	0	NA
NE	(>33.75,<=56.25)	0.66	2	0.11	0.61	8	0.22	0.76	4	0.23	0.65	2	0.40	NA	0	NA
ENE	(>56.25,<=78.75)	1.02	1	NA	0.76	3	0.15	0.84	4	0.11	0.92	4	0.06	NA	0	NA
E	(>78.75,<=101.25	<b>) 0.78</b>	4	0.12	NA	0	NA	0.52	3	0.24	0.59	2	0.21	NA	0	NA
ESE	(>101.25,<=123.7	/5)0.83	3	0.19	0.68	3	0.03	0.73	4	0.28	0.59	2	0.18	NA	0	NA
SE	(>123.75,<=146.2	25) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
SSE	(>146.25,<=168.7	'5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
S	(>168.75,<=191.2	25) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
SSW	(>191.25,<=213.7	/5)0.90	2	0.08	NA	0	NA	0.69	3	0.12	NA	0	NA	NA	0	NA
sw	(>213.75,<=236.2	25) NA	0	NA	0.61	1	NA	NA	0	NA	NA	0	NA	NA	0	NA
wsw	(>236.25,<=258.7	5)0.70	1	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA .
W	(>258.75,<=281.2	25) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
WNW	(>281.25,<=303.7	(5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
NW	(>303.75,<=326.2	25) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
NNW	(>326.25,<=348.7	75) NA	0	NA	0.65	1	NA	0.49	1	NA	NA	0	NA	NA	0	NA

## Table B4–2. SOIL–13 1986 Total Thorium (pCi g<sup>-1</sup>) 16 Sector Average (Depth 0–2 in)

		<u>0–1 km</u>		1	<u>1–2 km</u>		2	<u>–3 kn</u>	<u>n 3-</u>		<u>3-4 kn</u>	n	<u> </u>		<u>m</u>	
<u> </u>	Sectors	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS
N	(>348.75,<=11.25	) 2.84	13	1.57	1.46	3	0.15	NA	0	NA	2.13	2	0.33	2.72	4	1.06
NNE	(>11.25,<=33.75)	NA	0	NA	, NA	0	NA	2.99	4	0.19	1.52	2	0.29	2.10	3	0.65
NE	(>33.75,<=56.25)	NA	0	NA	NA	0	NA	2.65	7	0.29	2.56	1	NA	2.44	2	0.71
ENE	(>56.25,<=78.75)	NA	0	NA	3.99	2	0.77	NA	0	NA	NA	0	NA	NA	0	NA
Ε	(>78.75,<=101.25	) NA	0	NA	2.00	2	0.51	NA	0	NA	1.92	1	NA	2.86	6	1.48
ESE	(>101.25,<=123.7	5) NA	0	NA	NA	0	NA	1.86	1	NA	1.88	5	0.21	2.54	5	0.64
SE	(>123.75,<=146.2	5) NA	0	NA	NA	0	NA	NA	0	NA	2.36	1	NA	2.24	5	0.36
SSE	(>146.25,<=168.7	5) NA	0	NA	NA	0	NA	NA	0	NA	3.64	1	NA	2.63	4	0.34
S	(>168.75,<=191.2	5) NA	0	NA	NA	0	NA	NA	0	NA	1.39	3	0.23	NA	0	NA
SSW	(>191.25,<=213.7	5) NA	0	NA	NA	0	NA	NA	0	NA	2.26	2	0.45	1.77	2	0.84
SW	(>213.75,<=236.2	5) NA	0	NA	2.85	2	1.87	1.56	2	0.08	1.16	10	0.30	NA	0	NA
wsw	(>236.25,<=258.7	5) NA	0	NA	NA	0	NA	1.58	1	NA	NA	0	NA	2.53	4	0.09
W	(>258.75,<=281.2	5) NA	0	NA	NA	0	NA	1 42	1	NA	2.00	1	NA -	1.92	1	NA
WNW	(>281.25,<=303.7	5) NA	0	NA	NA	0	NA	NA	1	NA	4,33	1	NA	NA	2	0.02
NW	(>303.75,<=326.2	5) NA	0	NA	2.58	1	NA	NA	0	NA	2,17	1.	NA	3.91	5	1.57
NNW	(>326.25,<=348.7	5) NA	0	NA	NA	0	NA	NA	0	NA	2.20	2	0.08	1.92	3	0.00

			<u>56 kr</u>	<u>n</u>	6	<u>–7 kn</u>	<u>n</u>	7	<u>8 kn</u>	<u>1</u>		<u>8–9 kr</u>	<u>n</u>	<u>9–10 km</u>		m
	Sectors	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS	Value	No.	STDS
N	(>348.75,<=11.25	) 1.61	6	1.00	2.09	9	0.66	2.29	5	0.84	NA	0	NA	NA	0	NA
NNE	(>11.25,<=33.75)	1.72	6	0.16	2.27	7	0.48	2.09	2	0.63	NA	0	NA	NA	0	NA
NE	(>33.75,<=56.25)	2.90	3	0.17	1.75	8	0.89	1.43	4	0.55	1.40	2	0.25	NA	0	NA
ENE	(>56.25,<=78.75)	3.39	1	NA	3.55	3	0.29	2.31	4	0.90	2.70	2	1.61	NA	0	NA
Е	(>78.75,<=101.25	) 2.86	4	0.95	NA	0	NA	2.68	4	0.44	2.61	5	0.56	NA	0	NA
ESE	(>101.25,<=123.7	5)1.70	3	0.04	1.93	5	0.24	2.00	4	0.70	1.40	2	1.00	NA	0	'NA
SE	(>123.75,<=146.2	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
SSE	(>146.25,<=168.7	5) NA	0	NA	NA	0	ŇA	NA	0	NA	NA	0	NA	NA	0	NA
S	(>168.75,<=191.2	5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
SSW	(>191.25,<=213.7	5)1.39	2	0.63	NA	0	NA	2.24	4	1.14	2.01	2	0.61	NA	0	NA
SW	(>213.75,<=236.2	5) NA	0	NA	1.11	1	NA	NA	0	NA	NA	0	NA	NA	0	NA
wsw	(>236.25,<=258.7	5)1.25	1	NA	2.22	1	NA	NA	0	NA	NA	0	NA	NA	0	NA
W	(>258.75,<=281.2	5)2.07	2	0.41	2.02	2	0.88	3.07	4	0.84	NA	0	NA	NA	0	NA
WNW	(>281.25,<=303.7	'5) NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
NW	(>303.75,<=326.2	(5)2.55	2	1.39	NA	0	NA	NA	0	NA	NA	0	NA	NA	0	NA
NNW	(>326.25,<=348.7	5)0.94	1	NA	1.53	1	NA	2.00	4	0.93	NA	0	NA	NA	0	NA

Table B4-2. SOIL-13 1986 Total Thorium (pCi g<sup>-1</sup>) 16 Sector Average (Continued) (Depth 0-2 in)

Appendix B - Part 4 Additional Soil Monitoring Data

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		0_	<u>1 km</u>		1-	<u>1–2 km</u>			
	Sectors	Value	No.	STDS	Value	No.	STDS		
Ν	(>348.75,<=11.25)	3.07	7	4.0	1.15	2	0.2		
NNE	(>11.25,<=33.75)	5.82	6	· 9.0	NA	0	NA		
NE	(>33.75.<=56.25)	1.10	1	NA	NA	0	NA		
ENE	(>56.25,<=78.75)	3.45	2	2.6	NA	0	NA		
E	(>78.75,<=101.25)	27.51	10	70.6	NA	0	NA		
ESE	(>101.25.<=123.75)	NA	0	NA	NA	0	NA		
SE	(>123.75.<=146.25)	1.00	1	NA	NA	0	NA		
SSE	(>146.25,<=168.75)	1.83	3	0.2	NA	0	NA		
S	(>168.75,<=191.25)	NA	0	NA	NA	0	NA		
SSW	(>191.25,<=213.75)	NA	0	NA	NA	0	NA		
SW	(>213.75,<=236.25)	44.13	9	104.0	NA	0	NA		
WSW	(>236.25,<=258.75)	2.40	1	NA	NA	0	NA		
W	(>258.75,<=281.25)	NA	0	NA	NA	0	NA		
WNW	(>281.25,<=303.75)	2.50	1	NA	NA	0	NA		
NW	(>303.75.<=326.25)	0.90	1	NA	NA	0	NA		
NNW	(>326.25.<=348.75)	1.25	2	0.5	NA	0	NA		

Fable	B4-3.	RIFSSOIL	<sup>99</sup> Tc (pCi	<b>g</b> <sup>-1</sup> )	16 Sector	Average
		(Deptl	h 0-2 and	0–6 i	n)	۰.

#### CESIUM-137 AND STRONTIUM-90

Table B4-4 shows the concentration of  $^{137}$ Cs and  $^{90}$ Sr in soil samples with depth (Shleien 1991). The ratio of  $^{137}$ Cs to  $^{90}$ Sr in 56 surface soil samples (0-2 in. layer) collected around the FMPC in 1988 was 0.82 with a standard deviation of 0.46. The soil concentrations of  $^{137}$ Cs decreased with depth relative to the  $^{90}$ Sr concentration.

Samples taken at the 0–6 inch strata have a  $^{137}Cs/^{90}Sr$  ratio of 0.54 with a standard deviation of 0.37. The other strata samples indicate a decreasing ratio with depth. The number of samples available in the lower strata is very limited. Data obtained from the open literature indicate that deposition of  $^{137}Cs$  in the northern mid-latitudes between 1965 to 1967 ranged from 60–100 mCi km<sup>-2</sup> whereas the deposition from  $^{90}Sr$  was 60–80 mCi km<sup>-2</sup> (Eisenbud 1987) in the same area and time frame. If the deposition of either  $^{137}Cs$  or  $^{90}Sr$  from fallout in the northern hemisphere is 80 mCi km<sup>-2</sup>, then the concentration in the top centimeter of soil would be:

$$(80 \text{ mCi km}^{-2})(1 \times 10^{-10} \text{ km}^2 \text{ cm}^{-2})(1 \times 10^9 \text{ pCi mCi}^{-1}) = 8 \text{ pCi cm}^{-3}$$

Assuming a typical soil density of 1.4 g cm<sup>-3</sup>, the concentration per unit mass would be 5.7 pCi g<sup>-1</sup>. Leaching would move some of the radionuclide into lower layers of soil. Thus, a ratio of near unity is to be expected, given the half-lives of the two radionuclides and the

possible introduction of small amounts of fresh fallout post 1967. The ratio of the two radionuclides found in surface soil samples around the FMPC indicates that atmospheric fallout is the most likely source.

The data on  ${}^{137}\text{Cs}/{}^{90}\text{Sr}$  ratio indicates that  ${}^{137}\text{Cs}$  is being more tightly bound to the upper strata of soil than is the  ${}^{90}\text{Sr}$ . This agrees with the general perception about the binding of cesium relative to strontium by soil. On the other hand, one would expect higher absolute concentrations of  ${}^{137}\text{Cs}$  than  ${}^{90}\text{Sr}$  (a  ${}^{137}\text{Cs}/{}^{90}\text{Sr}$  ratio of greater than 1) due to lesser leaching and runoff of the  ${}^{137}\text{Cs}$ . This is not illustrated by the data. We presently have no explanation of this phenomena.

		<sup>137</sup> Cs (p	Ci g <sup>-1</sup> )	9	<sup>0</sup> Sr (pCi g	g <sup>-1</sup> )	13	<sup>37</sup> Cs/ <sup>90</sup> Sr Ratio			
Soil Layer	Ave	n <sup>b</sup>	Std. Dev.	Ave.	n <sup>b</sup>	Std. Dev.	Ave.	n <sup>b</sup>	Std. Dev.		
0-2	0.72	123	0.28	1.31	66	1.17	0.82	56	0.46		
0-6	0.55	118	0.32	1.65	64	1.83	0.54	53	0.37		
2-4	0.60	10	0.22	1.36	12	0.77	0.73	4	0.46		
4-6	0.53	4	0.26	1.10	11	0.44	0.40	2	0.14		
6-12	0.45	9	0.30	1.26	24	0.94	0.37	2	0.08		
12-18	0.60	4	0.78	1.32	23	0.98	0.27	3	0.14		

## Table B4-4. RIFSSOIL-1988 Depth and Ratio Data for <sup>137</sup>Cs and <sup>90</sup>Sr<sup>a</sup>

<sup>o</sup>Two samples (one for  $^{137}$ Cs and one for  $^{90}$ Sr) having concentrations above 10 pCi g<sup>-1</sup> were excluded from averages.

<sup>b</sup> n = Number of samples.

#### PLUTONIUM

Transfer of <sup>239</sup>Pu from the stratosphere to the earth's surface is said to occur at the same rate as <sup>90</sup>Sr, resulting in a constant ratio between the two radionuclides since the cessation of large atmospheric atomic tests in 1963. The ratio of <sup>239</sup>Pu/<sup>90</sup>Sr is about 0.017 in the stratosphere (corrected for decay to 1987) (Eisenbud 1987). It may be assumed to be the same on the earth's surface given the similar transfer rates from the stratosphere. The average <sup>90</sup>Sr concentration in surface soil around the FMPC is 1.3 pCi g<sup>-1</sup> with a standard deviation of 1.2. The expected concentration in surface soil of <sup>239</sup>Pu is about 0.02 pCi g<sup>-1</sup> considering the ratio noted above in this paragraph. The plutonium concentrations are about 50 to 500 times that expected from the fallout ratio of <sup>239</sup>Pu/<sup>90</sup>Sr. (Analysis for plutonium usually is unable to separate <sup>239</sup>Pu from <sup>240</sup>Pu, but the expected level of <sup>240</sup>Pu is relatively low.) Plutonium contamination occurred around the incinerator. Similar contamination with uranium occurred around the incinerator, the pilot plant, south of plants 7 and 5, and north of the coal pile. This pattern is similar to some of the areas of uranium soil deposition (See Figure N–3 of Killough et al. 1993).

Thus far, no off-site soil samples have been located that have been analyzed for plutonium.

Distance	Degrees	Date	Depth (In.)	pCi g <sup>-1</sup>
0 643	94 76	05-Nov-87	0-2	1 1
0.287	229.23	12–Mav–88	0–2	3.5
0.666	99.42	23-Jun-88	0-6	1.5
0.152	149.89	22–Jun–88	0–6	1.1
0.278	218.74	23–Apr–88	0–6	5.3
0.279	218.88	30-Jun-88	0–6	11.4
0.278	219.80	30–Jun–88	0-6	12.9
0.278	220.9 <del>9</del>	30-Jun-88	0-6	1.8
0.237	229.23	22-Jun-88	06	7.4
0.048	358.41	29-Jun-88	2–4	2.9
0.648	308.41	29-Jun-88	4-6	1.9
0.279	218.88	30-Jun-88	6-12	1.2
0.278	219.80	30-Jun-88	6-12	4.3
0.278	219.80	30-Jun-88	12-14	0.7

## Table B4–5. Concentrations of <sup>239,240</sup>Pu in Soil Around the FMPC (from RIFSSOIL 1988)

#### Source of Plutonium in Soil

The first step in discerning the source of plutonium in the soil samples was to obtain, from the compilation of soil sampling data (Shleien 1991), information on the uranium (U) concentrations that were measured at the locations where Pu contamination was detected. In some cases,  $^{238}$ U was measured in the soil layer of interest; for other strata, data on total uranium activity were found. We looked for results for all samples (of any vertical stratification) that had been collected at the location.

The data on measured concentrations of both U and Pu are shown in Table B4–6. As can be seen from the direction and distance columns, many of the samples where plutonium was detected were in close proximity. These have been grouped and have been given common primary location numbers. In the last column of the table are the Pu/U ratios in parts per billion (ppb). When only <sup>238</sup>U was measured, it was assumed that the total uranium activity was twice that of <sup>238</sup>U. When both activities were measured, the value for total uranium was used in the calculation of the Pu/U ratio. It can be seen from samples for which both <sup>238</sup>U and total uranium were measured that the ratio of these two activities is rather variable.

The highest Pu/U ratios were found for Location 4. In that area, Pu/U ratios of up to 213 ppb were found in the top soil layer and all samples from the vicinity contained elevated



Pu/U ratios in at least the top soil stratum. Substantially elevated level was also seen in the lowest soil layer at Location 6, where the soil appears to have been disturbed.

#### Plutonium Concentrations in Recycled Materials Received at the FMPC

Historic data on plutonium content of recycled materials that were received by the FMPC, compiled by NLO in 1985 (Spenceley 1985). were reviewed. The plutonium concentrations in the various forms of recycled uranium compounds differed substantially. with Pu/U ratios ranging from about 0.3 ppb for receipts of offsite  $UO_2$  to more than 1100 ppb for  $UO_3$  received from Paducah in 1980. Except for the 1980 shipment, the Pu/U ratios of incoming materials, while variable, were less than 10 ppb. The ratios of Pu/U in 16 hoppers of  $UO_3$  from the Paducah shipment ranged from 67 ppb to more than 7700 ppb. Only four of the lots exhibited ratios of less than 200 ppb; in three lots the Pu/U ratios exceeded 1000 ppb.

Part of the material from Paducah was repackaged, from hoppers to drums, in Plant 4. It was later blended with sump cake in the rotary kiln in Plant 8 and converted to calcium uranate, which was subsequently used as feed for the refinery. Production of  $UO_3$  from this feed stock appears to have begun in May 1982 and 110 lots had been produced by May 1985. The Pu content of each lot was measured and Pu/U ratios ranging from 4 to 46 ppb were found (Spenceley 1985). The ratio generally increased with time, but not monotonically, as the feed with higher Pu content became incorporated into the refinery inventory. Samples of UNH from 14 tanks in the refinery were analyzed for Pu in April 1985. The measured Pu/U ratios in samples of UNH ranged from 6.5 to 81 ppb.

#### Plutonium Concentrations in Samples of Dust and Scrub Liquor in 1985

Concentrations of plutonium and other transuranic nuclides were measured in various samples of dusts and scrub liquors in 1985 (Boback et al. 1987). In Plant 8, where the Paducah material was processed, ratios of Pu/U in samples of scrub liquor averaged about 60 ppb. Dust from primary dust collector for that facility was found to have a Pu/U ratio of about 80 ppb. Samples of dusts collected in Plant 4, which presumably represent historically more typical Pu/U ratios, averaged about 5 ppb. Similar low concentration ratios were also found in the dusts collected from the Pilot Plant. A somewhat higher average Pu/U ratio was found in dusts from Plant 5, but the results appear to be highly dependent upon the specific process exhaust treated. The highest Pu/U ratio was found in a sample of dust from Plant 1; it was about 3600 ppb in dust from collector G2-64. This finding apparently reflects dust from grinding and homogenization of samples of the original Paducah  $UO_3$ .

#### **Possible Sources of Surface Contamination**

It seems clear that some spillage or release of  $UO_3$  received from Paducah in 1980 could account for the elevated Pu/U ratios that have been observed in soil samples. The distributions of plutonium and uranium with depth in the soil samples suggest that the plutonium contamination was relatively recent. This latter observation is consistent with the Paducah  $UO_3$  as the source.

	Distance	Direction	Sample	Depth	239/240Pu	238 <sub>U</sub>	U	(Pu/U)
Loc.	(m)	(° from N)	ID	<u>(in)</u>	(pCi <b>g</b> <sup>-1</sup> )	(pCi g <sup>-1</sup> )	(pCi g <sup>-1</sup> )	(ppb)
1	643	94.76	5092 5093 5094	0-2 2-4 4-6	1.1 < 0.6	25670	15571 3642	0.24 < 0.21
2	665	99.42	5477 5478 5479	06 612 1218	1.5 < 0.6	1477	1063 661	5.6 < 3.1
3	152	149.89	5455 5456 5457	0-6 6-12 12-18	1.1 < 0.6	695	2133 1124 272	5.7 < 5.9
4a	278	218.74	5412 5413 5414	0–6 6–12 12–18	5.3 < 0.6	186	234 51	157 < 28
4b	278	219.8	5483 5484 5485	06 612 1218	12.9 4.3 0.7	333 281 453	718 425 663	213 84 9
4c	279	218.88	5486 5487 5488	06 612 1218	11.4 1.2	1441 316	2620 588 137	44 21
4d	273	220.99	5480 5481 5482	0–6 6–12 12–18	1.8 < 0.6	7944 394 1929		1.25 < 8.4
58	237	229.23	5462 5463 5464	06 612 1218	7.4 < 0.6	2343	5044 724 384	17 < 9.1
5b	237	229.23	5851 5852 5853	0-2 2-4 4-6	3.5 < 0.6	2374 339 250	570 624	8.1 < 9.7
6	64 <b>8</b>	358.41	5671 5672 5673	0-2 2-4 4-6	< 0.6 2.9 1.9	295 80.9	792 565 144	< 8.3 54 129_

## Table B4-6. Plutonium and Uranium Concentrations at Environmental Soil Sampling Locations Where Plutonium Was Detected

## SUMMARY

Appendix B, Part 4 summarizes data on natural and man-made radionuclides which have not been discussed elsewhere in reports on the FMPC dose reconstruction study. From these data it appears that:

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- Uranium soil contamination is noted on-site at the FMPC (also see Killough et al. 1993. Appendix N.).
- Uranium soil contamination is noted out to 8 km in the NE direction, which is also the direction toward which the prevailing winds blow.
- Uranium is leached into the soil, its concentration decreasing with depth in most cases.
- Radium-226 and thorium present on- and offsite is of natural origin.
- Sampling and analysis for <sup>99</sup>Tc has been sparse, but some soil contamination with this isotope is present.
- <sup>137</sup>Cs and <sup>90</sup>Sr in soil at the FMPC are most likely from atmospheric weapons testing.
- Plutonium contamination of soil is present on-site. The source seems to have been identified.

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#### **APPENDIX B --- REGIONAL ENVIRONMENTAL MONITORING**

#### PART 5 — MILK-VEGETATION

#### INTRODUCTION

This part of Appendix B presents the analytical data of uranium concentrations measured in milk and vegetation samples gathered in the vicinity of the FMPC during various years of operations. The purpose of compiling these analytical results of uranium in milk and vegetation is to observe general trends in concentrations in various components of the air pathway. These data may provide information on the importance of the air-to-grassto-cow-to-milk pathway for human exposure due to radionuclide releases from the FMPC. These data are also useful for calculating a site-specific parameter called the Concentration Ratio (CR) for uranium in grass.

#### MILK SAMPLING

Initial milk sampling at the FMPC occurred in 1959 and 1960 when single local samples were collected from the Knollman Farms, a dairy adjacent to the FMPC. The cows grazed on leased government property bordering the FMPC. Samples were collected in five consecutive months in 1965, and five months in 1966. Milk was sampled once a month as a composite of all the milk in the cooler at the time sampled. The samples in 1959, 1960, 1965, and 1966 were analyzed for total uranium by the FMPC Analytical Department (NLCO 1959, 1965, 1966). Because of low levels of uranium in the milk samples in 1965 and 1966, the site reduced the frequency of sampling to every 6 months (Nelson 1966). The activity measured in milk samples during this time compared favorably with results obtained by the U.S. Public Health Service milk testing stations in the area (Ross 1965).

A regular milk sampling program at the FMPC began in January 1980 when milk samples were taken from Knollman Farms, Inc., which grazes its cows on a leased portion of the FMPC site. Control samples were taken from a dairy farm in Sunman, Indiana located about 20 miles west of the site until September 1982, when milk was no longer available (EAL 1980-1984). A new control location in Edgewood, Kentucky (Foltz Brothers Dairy) is about 18 miles (35 km) southeast of the site. Foltz dairy distributes milk collected from various farms located in several counties of northern Kentucky. The monthly samples were analyzed for total uranium using a fluorometric method after sample wet-ashing with nitric acid. An additional sample is analyzed annually for gross alpha, gross beta, <sup>90</sup>Sr, <sup>99</sup>Tc,<sup>226</sup>Ra, <sup>228</sup>Ra, <sup>228</sup>Th, <sup>230</sup>Th, <sup>232</sup>Th,<sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U.

Initially, samples were collected monthly in 1980, 1981 and 1982. In 1983, the frequency of milk sample collections was reduced to quarterly, but was switched back to monthly sampling in September 1986 when analytical problems with the milk sampling program arose. Since 1980, all samples have been sent to an offsite laboratory, Environmental Analysis Laboratories (EAL) Corporation in Richmond, California. They analyzed uranium in milk by a technique called kinetic phosphorescence. The uranium measurements are tabulated in Table B5S-1 in the annex to this appendix. A summary of the uranium measurements in milk from the Knollman Farm is shown in Table B5-1. Except for a few cases that have been traced to analytical or contamination errors, the uranium concentrations in milk samples have been at or below the limit of detection of 1  $\mu$ g per liter (0.68 pCi L<sup>-1</sup>). The results indicate no increase in uranium in local milk compared to control samples. However, problems of high reported values of uranium have been attributed to contamination of samples (EAL 1980-1984). These higher than expected values occurred in control as well as local samples (e.g. July 1980, February and March 1981, April 1990). In June 1991, the site began using a different offsite laboratory to perform uranium analysis of milk samples using alpha spectroscopy.

Year	Number of Samples	Results <sup>b</sup>									
1959	1	nd <sup>c</sup>									
1965	5	3 of 5 ≤ DL									
1966	4	$3 \text{ of } 4 \leq DL$									
1980	12	8 of 12 ≤ DL									
1981	12	10 of 12 ≤ DL									
1982	12	all < DL									
1983	4	$3 \text{ of } 4 \leq DL$									
1984	5	all < DL									
1985	3	all < DL									
1986	3	all < DL									
1987	12	all < DL									
1988	12	11 of 12 ≤ DL									
1989	12	$11 \text{ of } 12 \leq DL$									
1990	12	10 of 12 ≤ DL									

Table B5-1. Summary of Uranium Concentration Measurements
in Milk From Knollman Farm <sup>a</sup>

<sup>a</sup> Values taken from NLCO 1959, 1965-1966; Nelson 1966; EAL 1980-1984; WMCO 1987, WMCO 1988, WMCO 1989.

<sup>b</sup> The detection limit for the analytical method was 0.68 pCi L<sup>-1</sup>. For some years, control as well as farm samples had above the detection limits of uranium. <sup>c</sup> None detected.

None detected.

### VEGETATION SAMPLING

Radionuclide contamination of forage and food crops can be a principal component of several human exposure pathways that result in the intake of radioactive materials. External contamination of vegetation involves mainly physical processes such as wet and dry deposition of airborne effluents and resuspended materials (Appendices G and H, Killough et al. 1993). Internal contamination of plants occurs primarily from root uptake of radionuclides from the soil. Food crops around the FMPC were not monitored routinely until 1983 when potatoes from the vicinity of the FMPC and from control locations in Indiana



were analyzed for uranium. Beginning in 1986, more extensive monitoring of leafy vegetables (cabbage and collards) and root vegetables (potatoes, carrots, and onions) was done. Generally, no clear differences between local and control concentrations have been observed.

Forage material from around the FMPC was monitored more extensively than food crops. In this section, datasets of uranium concentration in grasses from two time periods are examined. Data taken from analytical data sheets from the period 1958 to 1968 comprise one set, and data from 1984 onward represent the second.

Common varieties of grasses were gathered from the FMPC area and analyzed for uranium, gross alpha, beta and fluoride beginning in 1958 and 1959, 1961 and from 1963 to 1968. The grass samples, identified visually as blue grass with long, thin blades (NLCO 1958–1968), were collected twice a year in the spring and late summer near the gummed film stations. [The approximate locations of the gummed-film stations have been described previously (Killough et al. 1993)]. Table B5S–2 in the annex of this appendix contains the results of uranium in grass monitoring taken from analytical data sheets for this early period (1958–1968). These samples were oven dried, analyzed fluorometrically for uranium, and reported in units of  $\mu$ g U g<sup>-1</sup> dry weight. The sample designations indicate the general compass direction and distance from the center of the FMPC for locations designated 1 (300–500 m), 2 (600 to 1000 m), 3 (1000 to 1600 meters) and 4 (7000 to 14,000 meters). An exception is SW-4 which was actually located southeast of the facility center. The sample designations 3 and 4 generally were located at the site boundary or beyond.

Table B5S-3 in the annex gives the annual averages for these years at the designated locations in units of pCi  $g^{-1}$ , to be consistent with measurement units used in later years. Figure B5-1 shows that the 8-year average uranium concentration in forage decreased with increasing distance from the center of the production area in all directions. The onsite concentrations ranged from approximately 9 pCi  $g^{-1}$  to over 100 pCi  $g^{-1}$  at the east and south, locations which were near the incinerator and the storm sewer outfall to Paddy's Run Creek, respectively. The offsite concentrations at 1 to 2 km from the site center ranged from 4 to 11 pCi  $g^{-1}$ , while those from 7 to 14 km (location 4) were less than 1.5 pCi  $g^{-1}$ .

Beginning in 1984, analytical results from the routine sampling of grasses from the vicinity of the FMPC were reported in the annual environmental monitoring reports. The plant material sampled was primarily brome grass (Bromus sp.), but other genera represented were Allium, Daucus, Hordeum, Medicago, Melilotus, Poa, Secale and Triticum (Facemire et al. 1985). Each vegetation sample was a composite of a number of subsamples up to about 500 g (wet weight) total. Each subsample consisted of all above-ground plant materials from a 0.5-m (1.5 ft) diameter circular quadrant. Five of these subsamples equaled  $1 \text{ m}^2$  (11 ft<sup>2</sup>) of ground cover (Facemire et al. 1985, Aas et al. 1986, WMCO 1989). After collection, the samples were air dried before analysis for uranium and fluoride. Samples were sent to an offsite laboratory for analysis. In 1987, there was a change in analytical laboratories used to analyze the vegetation samples.





**Figure B5-1.** Long-term average uranium concentration in grass for 1958 to 1968 from onsite (<0.5, 0.5-1.0 km) to increasing distances offsite (1-2, 7-14 km). The east (dotted line) and south (broken line) locations onsite (<0.5 km) are near the incinerator and storm sewer outfall at Paddy's Run Creek, respectively.

Table B5S-4 lists the uranium concentrations in grass from onsite and offsite locations for the eighties. The table lists the approximate distance and directions from the center of the site. Prior to 1988 the sampling locations and designations varied from year to year although the majority of samples were collected to the northeast of the site. The onsite concentrations were higher than the offsite average values:

Figure B5-2 shows the annual average concentrations for 1958 to 1968 and from 1984 to the present. The concentrations measured during the eighties are 10 to 100 times lower than those measured in the late fifties and sixties. Although our complete source term data, for these periods will not be reported until later, the FMPC has estimated uranium emissions to air (Semones and Sverdrup 1988, FEMP 1992). Figure B5-2 includes these estimates for comparison. The uranium concentration in grass reflects the general trend of atmospheric releases of uranium from the FMPC. On July 10, 1989, the FMPC suspended production operations.



Appendix B—Part 5 Milk-Vegetation Page B5-5



**Figure B5–2.** Average annual uranium concentrations in grasses from onsite and offsite sampling locations from 1958 to 1991. Production operations were suspended at the FMPC in 1989. Uranium emissions to air are estimates from the FMPC; they do not represent final estimates from the dose reconstruction project.

### PLANT-TO-SOIL CONCENTRATION RATIO

Radionuclide uptake by plants from soil has generally been described by an empirical concentration ratio, CR, which is defined as the radionuclide activity per unit mass of plant material to the radionuclide activity per unit mass of soil. The soil and plant concentrations are usually reported in units of dry weight. Plant uptake of radionuclides from soils is affected by many factors, and, consequently, the CR can vary considerably. Some of the factors which affect plant uptake are:

- soil characteristics,
- plant species.
- translocation within the plant,
- the physical and chemical form of the radionuclide, and
- the distribution of radionuclides within the soil.

Some measurements of concentration ratios (CR) for uranium in grasses have been made. Peterson (1983) gives a range of CR for uranium of 5 x 10<sup>-3</sup> to 1.7 x 10<sup>-2</sup>. The latter value represents gross plant-to-soil CRs and includes external contamination from deposited and resuspended material as well as root uptake. This situation more closely reflects the conditions of measurement of uranium in grasses made at the FMPC where deposition onto vegetation and resuspension were not evaluated separately from root uptake. At the FMPC, grass samples were not washed prior to analysis and, therefore, the grass concentrations represent uranium from soil uptake, and from atmospheric deposition.

A site-specific plant-to-soil *CR* was determined from measurements of uranium made in grass and soil at offsite FMPC locations at various times during site operations. The data were obtained from analytical data sheets from the National Lead Company of Ohio from 1963 to 1968 (NLCO 1958-1968), and from environmental monitoring reports (Facemire et al. 1985, Aas et al. 1986, WMCO 1987, WMCO 1988, WMCO 1989, Dugan et al. 1990, Byrne et al. 1991, FEMP 1992) for more recent years. Sample locations were described in Tables B5S-2 and B5S-3, and included both on- and offsite locations. For this analysis, we use sampling results from locations within 1 to 3 km of the center of the facility. Onsite sample pairs were not used because of uncertainty about exact sampling locations for the early years, and the possibility of spills contributing to uranium in soil. Table B5S-5 tabulates the uranium concentration measurements in parallel soil and grasses from 1963 to 1968 analytical data sheets. Uranium concentrations during this time were recorded in units of  $\mu g g^{-1} dry$  weight. The median *CR* based on these data is 0.50, with the 25<sup>th</sup> and 75<sup>th</sup> percentiles of 0.3 and 1.6, respectively.

The soil samples for the parallel soil and vegetation samples from the late eighties onward were collected as part of the routine soil sampling program. Each soil sample was a composite of ten cores 2 cm (1 inch) in diameter and 5 cm (2 inches) deep. The cores were taken at two depths, 0-5 cm and 5-10 cm, within the soil profile, and at the four corners and the center of two grids. For the *CR* analysis, the upper layer soil concentrations were used because this situation more closely duplicated the soil sampling procedure in the sixties. Table B5S-6 tabulates the uranium concentration measurements in parallel soil and grasses samples for 1985 to 1991. Uranium concentrations during this time were recorded in units of pCi U g<sup>-1</sup> dry weight.

Figure B5-3 represents a histogram of the plant-to-soil concentration ratios determined from uranium data collected in 1963 to 1968 and from 1985 to 1991. The median CR for these ratios is 0.25, with the 25<sup>th</sup> and 75<sup>th</sup> percentiles of 0.04 and 0.58, respectively. The ratio, based on only the more recent data, is much lower, 0.03, with the 25<sup>th</sup> and 75<sup>th</sup> percentiles of 0.0089 and 0.065, respectively. These values are compared to published values of 0.017 to 0.0053 (Peterson 1983) in Table B5-2. The CRs from the earlier time period are high, outside the range of published literature values.

Table B5-2. Comparison of Published and Empirical Plant-to	-Soil
Concentration Ratios	

5 <sup>th</sup> percentiles	
Current FMPC data only	Published values <sup>b</sup>
0.0089, 0.03, 0.065	0.017, 0.0053
	5 <sup>th</sup> percentiles Current FMPC data only 0.0089, 0.03, 0.065

<sup>n</sup> Historic data refer to the 1963 to 1968 series of soil/vegetation samples; the current data refer to the 1985 to 1991 parallel samples.
 <sup>h</sup> From Peterson 1983.

In the earlier years, the plants were intercepting a relatively concentrated aerosol of uranium-bearing particles which were deposited on plant as well as ground surfaces. The uranium concentrations on plants could be relatively high because they were not treated to remove external contamination. In contrast, the soil samples are diluted with deeper layers of soil which are probably lower in uranium content than the very surface layer, particularly for the earlier years when the cumulative uranium depositions on soil are less than in later years. These circumstances suggest that conditions under which the ratios were determined for the earlier years may not have been in equilibrium which is implicit in the definition of the CR ratio. This possibility is currently being studied. Meanwhile, we suggest that the ratio determined from the more recent data is a better site-specific value to use for pathway analysis modeling if the soil-forage-cow-milk pathway is determined to be a key pathway of exposure to the residents in the FMPC area.



Figure B5-3. Histogram of plant-to-soil concentration ratios for uranium determined from 145 paired samples collected near the FMPC in 1963-1968 and in 1985 to 1991.

#### SUMMARY

This appendix summarizes the measurement data of uranium in milk and vegetation samples in the vicinity of the FMPC. Except for a few cases that have been traced to analytical or contamination errors, the uranium concentrations in milk samples have been at or below the limit of detection of 1  $\mu$ g per liter (0.68 pCi L<sup>-1</sup>). The higher than expected values occurred in control as well as local samples (e.g. July 1980, February and March 1981, April 1990). The results indicate no increase in uranium in local milk compared to control samples. The milk data could be compared to model-calculated concentrations of

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uranium in milk, if this pathway (air to soil to vegetation to cow to milk) proves to be a key pathway of exposure to people in the assessment domain (Appendix B, Killough et al. 1993).

There were no clear differences observed between local and control concentrations of uranium in food crops, although extensive monitoring of garden vegetables began only in 1986. The level of uranium in forage grasses is correlated with distance from the center of the FMPC. Concentrations in grass onsite but outside the production area ranged from 4 to 11 pCi g<sup>-1</sup>, while those from 7 to 14 km from the site center were less than 1.5 pCi g<sup>-1</sup>. The annual average uranium concentrations in grass from offsite and onsite locations reflect the general trend of atmospheric releases of uranium from the FMPC.

The median concentration ratio (CR) for uranium in grass, based on parallel sampling of grass and soil in the sixties and eighties, is 0.25. The more recent grass/soil data, however, yield a much lower median value of 0.03. We suggest that conditions under which the ratios were determined for the earlier years may not have been in equilibrium which is implicit in the definition of the CR. Consequently, the ratio determined from the more recent data may be a better site-specific value to use for pathway analysis modeling if the air-soil-forage-cow-milk pathway is determined to be a key pathway of exposure to the residents in the FMPC area.

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## APPENDIX B – PART 5 ANNEX

Table B5S-1. Uranium Concentration (pCi L <sup>-1</sup> ) Measurements in Milk From
Knollman Farm and Control Locations <sup>a</sup>

Year	Sample	Knollman	Control <sup>h</sup>	Year	Sample	Knollman	Control b
	Date				Date		
1959	Jul	nd		1984	Jan	<0.68	<0.68
1965	Aug	$13.3^{\circ}$			Apr	<0.68	<0.68
	Sep	0.66			Jul	<0.68	<0.68
	Oct	0.66			Sep	<0.68	<0.68
	Nov	<0.66			Dec	<0.68	<0.68
	Dec	2.00		1985	Jun	<0.68	<0.68
1966	Feb	<0.66			Oct	<0.68	<0.68
	Jun	2.00			Dec	<0.68	<0.68
	Jul	nd		1986	Feb	<0.68	<0.68
	Aug	0.66			Jun	<0.68	<0.68
1980	Jan	3.89	<0.40		Sep	<0.68	<0.68
	Feb	<0.40	<0.46	1987	Ann Avg	<0.7	<0.7
	Mar	6.60	0.53	1988	Jan	<0.7	<0.7
	Apr	<0.68	<0.68		Feb	1.00-	<0.7
	May	<0.68	<0.68		Mar	<0.7	<0.7
	Jun	<0.68	<0.53		Apr	<0.7	<0.7
	July	0.68	29.7 <sup>./</sup>		May	<0.7	<0.7
	Aug	0.53	0.33		Jun	<0.7	<0.7
	Sep	6.6 c	1.32		Jul	<0.7	<0.7
	Oct	0.68	1.98		Aug	<0.7	<0.7
	Nov	6.6 <sup>r</sup>	1.98		Sep	<0.7	<0.7
	Dec	0.68	0.68		Oct	<0.7	<0.7
1981	Jan	0.68	0.68		Nov	<0.7	<0.7
	Feb	4.62 <sup>r</sup>	4.95 <sup>r</sup>		Dec	<0.7	<0.7
	Mar	<0.68	- 3.50 °	1989	Jan <sup>e</sup>	<0.7	<0.7
	Apr	1.98	<0.68		Feb	<0.7	<0.7
	May	<0.5	<0.68		Mar	<0.7	<0.7
	Jun	<0.68	1.98		Арг	< 0.7	<0.7
	July	<0.68	<0.68		May	<0.7	<0.7
	Aug	<0.68	<0.68		Jun	<0.7	<0.7
	Sep	<0.68	1.32		Jul	<0.7	<0.7
	Oct	<0.68	<0.68		Aug	<0.7	<0.7
	Nov	<0.68	<0.68		Sep	<0.7	<0.7
	Dec	<0.68	<0.68		Oct	12.80 //	1.90

(continued next page)



		Knollman Fa	arm and Co	ntrol Lo	cations " (	cont'd.)	
Year	Sample Date	Knollman	Control <sup>b</sup>	Year	Sample Date	Knollman	Control <sup>b</sup>
1982	Jan	<0.68	<0.68	1989	Nov	<0.7	<0.7
	Feb	<0.68	<0.68		Dec	<0.7	<0.7
	Mar	<0.68	<0.68	1990	Jan	<0.68	<0.68
	Apr	<0.68	<0.68		Feb	<0.68	<0.68
	May	<0.68	<0.68		Mar	0.00	0.00
	Jun	<0.68	<0.68		Apr	4.80 <sup>c</sup>	3.70 <sup>c</sup>
	July	<0.68	<0.68		May	11.00 <sup>d</sup>	2.70
	Aug	<0.68	<0.68		Jun	<0.68	2.20
	Sep	<0.68	<0.68		Jul	<0.68	<0.68
	Oct	<0.68	<0.68		Aug	<0.68	<0.68
	Nov	<0.68	<0.68		Sep	0.02	0.05
	Dec	<0.68	<0.68		Oct	0.10	0.09
1983	Jan	0.68	0.68		Nov	0.05	0.05
	Feb	0.68	0.68		Dec	0.06	0.02
	Apr	1.35	1.35	1991	Jan	0.048	0.065
	Sep	<0.68	<0.68		Apr	0.068	0.11

#### Table B5S-1. Uranium Concentration (pCi L<sup>-1</sup>) Measurements in Milk From Knollman Farm and Control Locations " (cont'd.)

 <sup>n</sup> Values taken from analytical data sheets (1959, 1965, 1966, 1980-1985), offsite laboratory reports (EAL 1980-1985) and site environmental monitoring reports (WMCO 1987, WMCO 1988, WMCO 1989, Dugan et al. 1990, Byrne et al. 1991, FEMP 1992).

<sup>b</sup> Control location changed from a farm in Sunman, Indiana in 1983 to a dairy in Edgewood, Kentucky, about 35 km from the FMPC.

<sup>c</sup> Analysis was repeated to verify result.

"Result confirmed by duplicate analysis; concluded that sample was contaminated before analysis (EAL 1980-1984).

" Samples analyzed by Argonne National Laboratory and the Radiological and Environmental Sciences Laboratory had less than detectable concentrations as well.

Sample	19	58		1959		19	)63	19	64
Location "	25-Aug	24-Sep	7-Apr	22-Apr	11-Sep	1-Apr	27-Sep	30-Apr	15-Sep
N1	Lost h	10.89	45.6	203		110	39	80	43
N2	16.1		19.6	Lost <sup>b</sup>	11.74	35	5	6	9
N3	12.22		8.7	6.5	1.96	30	6.7	6	4
N4						2.1	2	0.1	10
NE1	27.48					93	37	46	23
NE2	24.8		10.2	18.7	12.65	6.7	55	18	20
NE3	9.57		9.5	4.8	33.9	26	6.7	12	8
NE4						4.1	3.2	2	6
E1	56.64		73.2	20.8		300	160	370	164
E2	24.99		20.4	18.1	2.67	110	33	26	32
E4						2.1	1.6	0.2	1
SE1	25.66		12.7	48		130	39	84	70
SE2	11.45		8.1	17.5		22	27	8	25
SE3	3.99		76.7	5.5	1.72	9.5	5	1	5
S1		175.54	23.7	461		680	500	<b>49</b> 0	258
S2		9.93	5	9.8	4.1	75	23	10	16
S3	Lost <sup>h</sup>	15.12	0.5	1.4	2.44	15	3	12	11
S4						3.4	1.2	1	2
SW1	Lost <sup>h</sup>	51.25	37	98.7		12	130	80	42
SW2	Lost <sup>h</sup>								
SW3		3.14	6.4	5.1	3.12	34	43	4	21
SW4						1.6	1.8	1	1
W1		25.1	59.2	53.3			110	80	76
W2		9.37	77.7	33.7		160	47	30	16
<b>W</b> 3	10.24		7.3	1.4	0.9	21	11	10	7
W4						5.6	0.4	0. <del>9</del>	2
NW1	7.54	58.07	22.4	18.9		67	27	38	14
NW3	13.97		4.7	2.4	1.4	17	4.7	3	4

Table B5S-2. Uranium Concentrations ( $\mu$ g U g<sup>-1</sup>) in Grass Samples Near the FMPC During the Early Years

(continued next page)

	the FMPC (continued)							
Sample	19	65	19	66	19	67	19	68
Location "	27-Apr	1-Sep	4-May	17-Aug	14-Apr	9-Aug	16-Apr	16-Aug
- N-1	11	13	9	13	82	13	17	13
N-2	3	3	4	2.3	26	3.9	8.3	5.8
N-3	4	5	3	2	15	7	3.2	2.6
N-4	2	2 ·	2	1	2	2.3	0.6	1
NE-1	27	46	21	33	72	13	40	24
NE-2	5	9	17	7	5 <del>9</del>	5.3	9.3	11
NE-3	3	5	3	3	9	4	5.8	3.3
NE-4	2	2	1	1	3	2.3	1.2	3.8
E-1	45	158	8	124	607	116	58	211
E-2	35	12	8	5	132	23	17	7.3
E-4	0.7	0.4	1	4	1	1	0.5	1
SE-1	22	18	17	12	114	65	37	8.3
SE-2	14	7	6	3.6	24	8	14	3.3
SE-3	5	2	4	2	3	1.7	3.7	0.6
S-1	110	58	82	167	241	356	90	40
S-2	8	8	<u></u> 8	2	21	4.8	5	3.3
S-3	5	3	`4	2	5	1.8	2.2	1.7
S-4	0.5	0.5	0.4	1	2	0.6	0.6	0.3
SW-1	14	23	19	16	100	12	13	16
SW-2								
SW-3	10	4	4	6	22	2.3	3.1	2.7
SW-4	3	2	0.7	2	1	0.9	0.4	0.5
W-1	16	11	27	11	42	14	13	7.5
W-2	25	60	55	4	50	9	22	9.3
<b>W</b> -3	4	4	2	2	8	2.6	1.8	2
W-4	3	2	0.4	2	1	1.6	2.2	1
NW-1	9	12	4	4	22	8	13	12
NW-3	2	2	1	1	2	2.5	1.3	1
<sup><i>a</i></sup> Approxima <sup><i>b</i></sup> The analyt	te locatio ical data	ns are d sheets ir	escribed ndicate th	in Appen nat "samr	dix M in oles were	Killough lost in t	n et al. 19 reatment	993. "

# Table B5S-2. Uranium Concentrations (up U $g^{-1}$ ) in Grass Samples Near

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				ic anu	onane.	LUCALIU	41.5			
Sample				-						8-yr
Location "	1958	1959	<u>1961<sup>b</sup></u>	1963	1964	1965	1966	1967	1968	average
N1	7.2	82	44 <sup><i>h</i></sup>	49	41	7.9	7.3	31	9.9	29
N2	11	24	22 <sup>b</sup>	13	5.0	2.0	2.1	9.9	4.6	8.9
N3	8.1	3.8	7.9 <sup>k</sup>	12	3.3	3.0	1.6	7.3	1.9	5.1
N4			4.0 <sup><i>b</i></sup>	1.4	3.3	1.3	0.99	1.4	0.53	1.5
NE1	18			43	23	24	18	28	21	25
NE2	16	9.1		20	12	4.6	7.9	21	6.7	12
NE3	6.3	11		11	6.6	2.6	2.0	4.3	3.0	5.8
NE4				2.4	2.6	1.3	0.66	1.8	1.6	1.5
E1	37	31		150	180	67	44	240	89	100
E2	16	9.1		47	19	15	4.3	51	8.0	21
E4				1.2	0.40	0.4	1.6	0.66	0.50	0.80
SE1	17	20		56	51	13	9.6	59	15	30
SE2	7.6	8.5		16	11	6.9	3.2	11	5.7	8.7
SE3	2.6	18		4.8	2.0	2.3	2.0	1.5	1.4	4.4
<b>S</b> 1	120	160		390	250	55	82	200	43	160
S2	6.6	4.2		32	8.6	5.3	3.3	8.5	2.7	8.9
S3	10	0.95		5.9	7.6	2.6	2.0	2.2	1.3	4.1
S4				1.5	0.99	0.33	0.46	0.86	0.30	0.7
SW1	34	45		47	40	12	12	37	9.6	30
SW2	11									11
SW3	2.1	3.2		25	8.3	4.6	3.3	8.0	1.9	7.1
SW4		-		1.1	0.66	1.6	0.89	0.63	0.30	0.9
W1	17	37		73	52	<b>8.9</b>	12	18	6.8	28
W2	6.2	37		68	15	28.	19	20	10	25
<b>W</b> 3	6.8	2.1		11	5.6	2.6	1.3	3.5	1.2	4.2
W4				2.0	0.96	1.6	0. <b>79</b>	0.86	1.1	1.2
NW1	24	14		31	17	6.9	· 2.6	9.9	8.2	14
NW3	9.2	1.9		7.2	2.3	1.3	0.66	1.5	0.76	3.1
Average	17	26	19	42	28	11	9	29	9	21

### Table B5S-3. Annual Average Uranium Concentration in Grass (pCi g<sup>-1</sup>) for Onsite and Offsite Locations <sup>a</sup>

<sup>a</sup> Values taken from NLCO 1958-1968. Approximate locations are described in Appendix M of Killough et al. 1993. The onsite locations are outside of the production area.
 <sup>b</sup> From Klein 1963. This value represents an average of all locations designated 1, 2, 3 or 4.

	ass rivin (	JUSILE		ISITE LU	cation	s at the	FMFU		
Location/distance	Direction			•					
From Site (km) <sup>h</sup>	From Site	1984	1985	1986	1987	1988	1989	1990	1991
Onsite:									
AMS 9	NE		1.57	0.6	0.28	0.02	2.90	0.48	0.16
AMS 8	NE	5.06	2.34	2.29	0.45	0.54	0.25	0.04	0.17
AMS 1	N-Fence	4.59	0.88			5.60	0.26	0.01	0.095
AMS 3	E-Fence	7.09	1.63	4.29	0.32	0.68	1.00	0.28	0.041
AMS 4	SE-Fence	0.66	0.37	0.72	0.39	1.50	0.05	0.007	0.22
AMS 6	W-Fence	0.66	0.02	0.40	0.35	0.24	0.26	0.016	0.095
AMS 5	SW-Fence		0.31	0.39	0.20	0.28	0.16	0.016	0.28
AMS 2	NE-Fence	1.78	1.40	1.40	0.27	0.61	0.12	0.005	0.074
AMS 7	NW-Fence	4.33				0.14	0.01	0.02	0.15
Production Area		6.67	1.50	3.25	0.96				
Fenceline			0.67	0.49	0.32				
Onsite Average		4.17	1.07	1.54	0.39	1.07	0.56	0.10	0.14
Offsite:									
1.3	N						0.05	0.08	0.02
1.8	NE					0.09			
1.8	NE					0.002			
1.9	NE						0.07	0.01	0.034
1.9	E	0.90	0.48	0.2	0.66	0.04	0.03	0.23	0.068
1.7	SE	0.44							
1.7	S		0.26	0.13	0.27				
1.3	NW					0.14			
2.4	NE	1.12	0.54	0.31	0.22	0.74	0.04	0.01	0.14
2.7	NE		0.4	0.21		0.05	0.02	0.01	0.034
2.2	SE	0.32							
2.6	s	1.06				0.12	0.08	0.09	0.047
2.2	NW				0.14		0.04	0.08	0.088
3.8	SE							0.11	0.034
3.7	S					0.03	0.03	0.01	0.095
3.7	S					0.01	0.01	0.02	0.095
3.7	SW					0.08	0.06	0.06	0.027
3.9	NW			0.06	0.14	0.01	0.02	0.02	0.081
4.2	NE	0.48	0.25	0.24			0.04	0.01	0.68
4.3	NE					0.01	0.02	0.01	0.027
4.3	SE						0.01	0.00	
5	Ε				0.03	0.01		0.04	0.027
5.4	w				0.04	0.02		0.01	0.14
5.1	NW	0.26		0.14	0.03	0.02		0.08	0.041
6.2	NE	0.12		0.08					
8.8	NE	0.10	0.1	0.13	0.03	0.03		0.02	0.02
24	SE				0.03	0.01			0.1
40	NW	0.25	0		0.28	0.01		0.01	0.16
Offsite Average		0.51	0.29	0.17	0.17	0.08	0.04	0.04	0.10

#### Table B5S-4. Annual Average Uranium Concentrations (pCi g<sup>-1</sup>) Measured in Grass From Onsite and Offsite Locations at the FMPC <sup>a</sup>

<sup>*a*</sup> Values are taken from the annual environmental monitoring reports.

<sup>h</sup> Onsite samples were collected near the air monitoring stations (AMS). Offsite sampling locations are designated by the distance in km from the center of the FMPC production area.

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Sampling			CR	Sampling			CR
period	Grass	Soil	(unitless)	period	Grass	Soil	(unitless)
Apr-63	30	4.2	7.14	May-66	3	11	0.27
• •	15	3.8	3.95	-	4	9	0.44
	34	12	2.83		4	19	0.21
	17	2.3	7.39		1	6	0.17
	26	1.5	17.33		3	11	0.27
	10	5	1.90		4	13	0.31
	5	6	0.78		2	9	0.22
Sep-63	7	0.4	16.75	Aug-66	2	4	0.50
	3	0.5	6.00		2	6	0.33
	3	2.7	1.19		6	13	0.46
	2	5	0.36		1	3	0.33
	2	4.4	0.36		3	7	0.43
	2	4.5	0.44		2	15	0.13
Apr-64	10	16	0.63		8	13	0.62
	6	21	0.29	Apr-67	15	11	1.36
	12	4	3.00		5	6	0.83
	4	8	0.50		22	18	1.22
	3	8	0.38		2	5	0.40
	12	3	4.00		9	9	1.00
	1	10	0.10		3	4	0.75
	7	2	3.50		3	7	0.37
Sep-64	4	4	1.00	Aug-67	7	8	0.88
	11	5	2.20		1.8	5	0.36
	21	5	4.20		2.3	12	0.19
	4	1	4.00		2.5	. 4	0.63
	8	7	1.14		4	11	0.36
	5	3	1.67		1.7	8	0.21
	4	13	0.31		1.8	9	0.20
Apr-65	4	7	0.57	Apr-68	3.2	8	0.40
	5	1	3.85		2.2	7	0.31
	10	1	8.33		3.1	14	0.22
	2	16	0.13		1.3	4	0.33
	3	10	0.30		5.8	10	0.58
	5	3	1.67		3.7	8	0.46
Sep-65	4	10	0.40		2	12	0.17
	5	10	0.50	Aug-68	2.6	9	0.30
	3	4	0.75		1.7	9	0.20
	4	12	0.33		2.7	9	0.31
	2	3	0.67		1	4	0.26
	5	6	0.83		3.3	10	0.33
	2	6	0.33		0.6	10	0.06
	2	11	0.18	Mediar	n <i>CR</i> (1963 t	o 1968)	0.50

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## Table B5S-5. Uranium Concentrations (µg g<sup>-1</sup> dry wt) in Parallel Soil/Vegetation Samples Collected by FMPC in the Sixties

Sampling	inpies col	necteu r	$\frac{10m}{CR}$	Sampling		n 1305 tu	<u>1331</u> CR
period	Grass	Soil	(unitless)	period	Grass	Soil	(unitless)
1985	0.65	2.84	0.23	1989	0.05	15.00	0.003
	0.31	1.08	0.29		0.07	7.90	0.01
	1.63	31.14	0.05		0.03	2.20	0.01
	1.50	17.60	0.09		0.04	8.90	0.00
	0.37	5.75	0.06		0.04	7.20	0.01
	0.31	3.25	0.10		0.08	3.20	0.03
	0.26	4.27	0.06		0.02	7.40	0.00
	0.67	5.08	0.13		0.06	5.90	0.01
	0.18	3.11	0.06		0.03	6.80	0.004
	0.40	2.23	0.18		0.01	2.40	0.004
1987	0.33	3.20	0.10	1990	0.08	7.20	0.011
	0.08	6.10	0.01		0.01	7.20	0.001
	0.20	15.00	0.01		0.23	6.30	0.04
	0.39	23.80	0.02		0.08	2.50	0.03
	1.03	2.37	0.43		0.01	6.40	0.002
	0.22	4.30	0.05		0.09	4.80	0.02
	0.28	6.50	0.04		0.01	4.20	0.003
	0.27	4.50	0.06		0.01	3.40	0.002
	0.14	3.00	0.05		0.01	3.40	0.003
1988	0.61	9.40	0.06		0.02	1.60	0.01
	0.14	5.70	0.02	1991	0.02	14.00	0.001
	0.12	1.40	0.09		0.03	4.50	0.008
	0.08	2.60	0.03		0.07	3.90	0.017
	5.00	2.00	2.50		0.09	0.54	0.16
	0.04	1.60	0.03		0.14	4.10	0.03
	0.03	1.40	0.02		0.05	0.41	0.11
	0.01	2.70	0.00		0.03	1.30	0.026
	0.74	5.40	0.14		0.03	0.88	0.03
	0.05	3.40	0.01		0.10	1.50	0.063
	0.09	4.10	0.02		0.10	0.74	0.13
	0.00	5.40	0.00037	Median	CR (1985 t	o 1991)	0.03

Table B5S-6. Uranium Concentration (pCi g<sup>-1</sup> dry wt) in Parallel Soil/Vegetation Samples Collected From the Vicinity of the FMPC From 1985 to 1991

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#### APPENDIX B - REGIONAL ENVIRONMENTAL MONITORING

#### PART 6 - RIVER-SEDIMENT-FISH

#### INTRODUCTION

This part of Appendix B examines the concentrations of uranium (and other radionuclides when available) in surface water, sediment, and fish from the vicinity of the FMPC during various years of operations. The purpose of compiling the results of surface water uranium analysis is to observe general trends in uranium concentration in the surface water near the FMPC over time, and to compare these measurements with modelcalculated concentrations based on our final source term estimates. Our final source term estimates will be reported later in the final Task 2 and 3 report. In our Task 4 report (Killough et al. 1993), we used the water sampling data from 1960-1962 as a comparison with model-calculated uranium concentrations in the river and in Paddy's Run, and presented the sediment sampling data in the river to show that uranium build-up had not occurred in the sediments of the river from operations at the FMPC (Killough et al 1993). These measurements of uranium in water can also used in combination with sediment and fish sampling results to calculate site-specific parameters such as the bioaccumulation factor (BF) for uranium in fish, if needed for radiation dose calculations in Task 6. In this appendix we review the water sampling data for years other than 1960 to 1962, the sediment sampling data from 1974 onward, and the fish sampling results from 1984 onward.

#### SURFACE WATER SAMPLING

The FMPC sampled and analyzed surface water from the Great Miami River and from Paddy's Run beginning in the early years of operation. A water sampling program was planned but not yet initiated in September 1952 (Davis 1952). However, by the next month (October 1952), some samples from Paddy's Run and the river had been sent to the New York Operations Office of the Atomic Energy Commission for analysis (Blase 1952). By 1953, the site was analyzing water samples for gross alpha and gross beta on a somewhat limited schedule. From 1954 onward, uranium analysis was done routinely in surface water samples collected at locations upstream and downstream of the site in the Great Miami River and in Paddy's Run to the west of the facility. Figure B6-1 shows the liquid effluent release points and the main water sampling locations in the early years of operations.

For Paddy's Run, water samples were collected upstream and downstream, and analyzed routinely for total uranium (mg U  $L^{-1}$ ), gross alpha and beta activity, total suspended solids, some chemical constituents, and occasionally for radium. In the fifties, water samples were collected three times daily, and a composite analyzed every third day. Samples were collected by the NLO water department downstream in Paddy's Run at Willey Road, or from the New Haven Bridge if there was no water flow at the Willey Road bridge.

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Figure B6-1. Diagram of the FMPC showing the main water sampling locations in the early years of operation.

In the fifties, weekly samples were taken "above" the storm sewer outfall ditch (SSOD) onsite. and only occasionally, offsite at the bridge north of route 126, upstream of the FMPC. Routine offsite sampling upstream in Paddy's Run, north of Route 126 did not begin until April 1959 (NLCO 1959).



In the Great Miami River, water samples were taken upstream at the Venice Bridge in Ross, about a mile (1.6 km) north of the effluent discharge point, at the New Baltimore Bridge, about 2 miles (3.2 km) downstream, and at the Miamitown Bridge, approximately 5 km downstream of the FMPC, at 39°12'30" north latitude and 84°42'30" west longitude according to the Hamilton County Engineer's office (Fuchs 1977). The sampling frequency at these locations varied in the early years, but followed a more regular schedule later.

#### Sources of Data and Information to Evaluate Data Quality

We have located and compiled water sampling data for uranium (and for radium, when the analysis was done) from original analytical data sheets from the Analytical Department of the National Lead Company of Ohio onsite, from Industrial Hygiene and Radiation monthly reports, and from annual environmental monitoring reports. Data for the river and Paddy's Run for the 1960-1962 period were tabulated in an earlier RAC report (Killough et al. 1993). The source of the data are referenced as they are presented in the text or appropriate tables.

Radionuclides other than total uranium were analyzed in later years of operations. Water from Paddy's Run was analyzed for radium occasionally beginning in the sixties, and was done on a monthly or semimonthly basis along with thorium analysis from the seventies onward. No other specific radionuclide analysis was done on water samples until 1984, when semiannual samples were analyzed for <sup>90</sup>Sr, <sup>99</sup>Tc, <sup>234</sup>U, <sup>236</sup>U, <sup>235</sup>U, and <sup>238</sup>U. Cesium-137 was added in 1987.

The analytical data sheets did not indicate a minimum detectable concentration for uranium until the early seventies. Prior to that time, the minimum reported value in the data sheets was approximately 0.001 mg U  $L^{-1}$  (0.68 pCi  $L^{-1}$ ). In the seventies, the detection level was 0.33 pCi  $L^{-1}$  (NLCO 1975). A regular quality control program was not in place until the late seventies when interlaboratory quality assurance practices such as daily calibrations of instrumentation and routine analysis of blanks, standard solutions and spiked sample aliquots were documented and performed (NLCO 1978). Prior to that time, the water sampling program was focused on meeting state of Ohio or federal guidelines, or not exceeding maximum allowable concentrations (MAC), and only occasionally ran "blank" samples. Usually, these tests were performed when a contamination problem was suspected. For example, the uranium concentration measured in a "blank" (distilled water) in June 1956 was quite high at 0.01 mg U  $L^{-1}$  (10 pCi  $L^{-1}$ ) (NLCO 1956). The contamination problems seemed to be related to the reuse of sampling bottles. When a blank (distilled water) from a new bottle was analyzed, no uranium was detected on August 19, 1955 (NLCO 1955). However, a blank (distilled water) from a previously-used bottle yielded a uranium concentration of 0.019 mg  $L^{-1}$ . As discussed in Appendix A, some FMPC sample contamination is suggested when the upstream or "background" uranium measurements made by the site are compared to background measurements made upstream of the FMPC by other facilities.

By the late sixties, there were monthly quality control reports detailing laboratory analytical accuracy and precision; however, these relate more to onsite operations than to environmental monitoring. Nevertheless, information on the uncertainties surrounding the fluorometric analysis of uranium was provided (Brown 1967).

#### **Measurement Data**

Tables B6S-1 to B6S-15 ("S" for "Special") give the original measurements from the analytical data sheets for 1954, 1955, 1956, 1957, 1959, 1963, 1964 and 1965 for the Great Miami River and Paddy's Run. These tables are provided in the annex of Part 6 of this appendix. The original measurements were reported in mg L<sup>-1</sup>, as shown in these tables. For the summary tables and graphics, however, we have converted these units to pCi L<sup>-1</sup> using the conversion of  $6.8 \times 10^{-7}$  Ci g<sup>-1</sup> for natural uranium. Table B6-1 summarizes some of these data as annual averages for uranium in the river from 1955 to 1991. Figure B6-2 shows the monthly average uranium concentrations measured in the river at the New Baltimore Bridge, approximately 2 km downstream from the site. This trend analysis clearly shows the higher concentrations measured in the river in 1955 through 1957. This may be related to the installation of the storm sewer lift station in 1957. Prior to that time, all runoff from the storm sewer system went directly to the river. Other changes in the liquid effluent control system at the FMPC that may have affected the quantity of material discharged to the river will be described in more detail in the final Task 2 and 3 report.





0002.-

Measured in the	Great Miami River	<u>r from 1955 to 1991</u>
Year	Venice Bridge	New Baltimore
	(upstream)	(downstream)
1955	50	240
1956	54	67
1957	37	45
1958	13	47
1959	10	11
1960	12	18
1961	10	11
1962	10	14
1963	5.9	12
1964	6.2	8.0
1965	6.1	11
1966	9.3	11
1967	7.2	11
1968	5.9	7.1
1969	11	10
1970	5.0	8.0
1971	3.0	2.1
1972	1.0	4.0
1973	3.9	3.0
1974	2.0	2.0
1975	1.3	1.2
1976	1.7	2.7
1977	1.4	1.7
1978	1.7	2.0
1979	1.4	1.4
1980	1.4	1.4
1981	1.4	1.4
1982	1.4	1.4
1983	1.4	2
1984	1.6	1.6
1985	1.6	1.6
1986	1.2	1.4
1987	1.1	2.1
1988	1.0	1.5
1989	1.4	1.5
1990	1.2	1.4
1991	1.1	1.2

## Table B6-1. Annual Average U Concentrations (pCi $L^{-1}$ ) Measured in the Great Miami River from 1955 to 1991

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For Paddy's Run, Figure B6-3 shows the gradual decrease in uranium concentrations with time both above the confluence of the storm sewer outfall ditch (SSOD) with Paddy's Run, and just below the site at the Willey Road Bridge. Table B6-2 summarizes the measurement data from Paddy's Run as annual averages for 1955 to 1965 and from 1975 to 1990, years for which data were located.



Figure B6-3. Annual average uranium concentration in Paddy's Run from 1955 to 1965 and from 1975 to 1990. The data from 1965 to 1974 were not located.

In summary, the site has conducted an extensive water sampling and uranium analysis program of the Great Miami River and Paddy's Run since 1955. Although the sampling protocols and analytical procedures were not clearly stated in the early years, the measurement data from these programs does provide important information for our dose reconstruction study. The concentrations of uranium measured in the Great Miami River have been much lower during all years than those measured in Paddy's Run. The concentrations measured in the river downstream of the effluent outfall were, to some extent, higher than the upstream measurements in the fifties and sixties, although the facility always emphasized that the reported concentrations were never greater than DOE guidelines in effect at the time.

The Willey Road Bridge data have consistently shown above background concentrations of uranium in Paddy's Run as well as being a source of groundwater contamination. Recent studies of the groundwater around the FMPC (Dames and Moore 1985, DOE 1990) have concluded that the primary source of the uranium contamination in the groundwater south of the site is uranium in waters released to the Storm Sewer Outfall Ditch (SSOD) and to Paddy's Run. Prior to 1957 when the storm sewer lift station was installed, much of the runoff from the site went directly into Paddy's Run. A storm sewer detention sump was

originally built in the vicinity of the main storm sewer outfall to catch initial runoff after rainfall (Starkey et al. 1962). However, no means of emptying this sump was provided, and it was very seldom used. In addition, the erratic and seasonal water flow in Paddy's Run has contributed to greater fluctuations in uranium measurements. Some of these uranium concentration data will be used in future reports to compare with our model-calculated concentrations.

Year	Upstream <sup>a</sup>	Willey Road Bridge
		(downstream)
1955	35	100
1956	55	240
1957	34	100
1958	26	480
1959	27	780
1960	14	1100
1961	20	470
1962	14	367
1963	7	690
1964	21	720
1965	19	580
1975	4.1	92
1976	2.7	160
1977	5.4	20
1978	5.4	63
1979	2.7	11
1980	2.7	19
1981	2.7	21
1982	2.7	5
1983	1.4	8
1984	1.4	9.5
1985	1.6	7.2
1986	. 1.1	9.5
1987	1.0	1.9
1988	0.8	2.1
1989	0.9	4.5
1990	0.8	4.5
1991	0.8	3.9

# Table B6-2. Annual Average Uranium Concentrations(pCi L<sup>-1</sup>) in Paddy's Run

<sup>a</sup> The upstream location changed from an onsite location above the storm sewer outfall ditch (SSOD) in the fifties and sixties to an offsite location just north of Route 126 in the later years.

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#### SEDIMENT SAMPLING

Annual sediment sampling of the Great Miami River and the Paddy's Run was begun in 1974 to determine "if material was accumulating below the site outfall" (NLCO 1975). Initially samples were collected from seven locations along the river bed by dragging a heavy metal container along the bottom, and from the river bank by scraping up the top one or two inches. Only the portion passing a 50-mesh screen was analyzed for uranium. Two locations were sampled upstream (at 1 and 2 km), and five sites at increasing distances downstream of the effluent outfall to the river. These distances ranged from immediately below the outfall, to 3 km at the confluence of Paddy's Run with the Great Miami River. In 1986, two more sampling locations were added above and below the effluent outfall (WMCO 1987). In the early eighties, semiannual sampling was done. The 1974 through 1985 uranium concentration data for sediment in the river have been compiled in Appendix R of our Task 4 report (Killough et al. 1993), and are listed in Table B6-3.

Figure B6-4 shows the 12-year average of uranium concentrations in river sediment at the four sampling locations. Although the average concentration in sediment taken near the effluent outfall is slightly higher, the data indicate no consistent difference between uranium in sediment measured upstream, just downstream of the effluent discharge point, or further downstream below the point where Paddy's Run flows into the Great Miami River. The results from 1974 onward indicate no build-up of uranium in the sediments where settling might be expected to occur.



**Figure B6-4.** Twelve-year averages, with standard deviations, of uranium concentrations in sediments from upstream of the FMPC, near the effluent outfall, below the outfall, and below the confluence of Paddy's Run with the Great Miami River. The dotted lines represent the background range of uranium concentrations in soils in Ohio.

	Total Uranium (pCi g <sup>-1</sup> dry wt)"				
	Upstream of		Downstream of	Below	
Year	the FMPC	At Effluent Line	Discharge Point	Confluence of Paddy's Run	
1974	1.30	1.8	0.80	1.10	
1975	1.80	3.3	0.60	0.90	
1976	0.70	1.6	0.40	0.70	
1977	0.85	1.0	0.50	1.20	
1978	0.90	1.8	0.50	2.10	
1979	0.80	1.4	0.60	0.90	
1980	0.84	. 0.7	0.47	0.68 ·	
1981	0.44	0.54	0.68	0.54	
1982	0.90	1.5	0.87	1.00	
1983	1.75	3.1	1.86	2.10	
1984	1.30	2.64	2.41	1.36	
1985	0.90	2.4	1.4	0.70	
1986	0.25 <sup><i>b</i></sup>	Ь	ь	Ь	
1987	$1.2^{b}$	Ь	Ь	Ь	
1988	1.4		1.4	2.0	
1989	2.0		2.0 °	2.0	
1990	1.6		1.8 "	0.79	
1991	1.2	1.1	1.1	0.98	
Average	1.02	1.76	0.88	1.14	
Stdev	0.39	0.61	0.53	0.60	

Table B6–3.	Uranium i	in Sediment fro	om the Great	Miami River

 $^{\alpha}$  Total uranium is reported for all years except 1987 to 1989 when  $^{238}$ U concentrations are given. The FMPC annual environmental reports stated that "the 95% CI was ± 25% for all samples.

<sup>b</sup> Only an average concentration for all river locations was given with statement that there was no significant difference among sampling locations.

In addition to uranium, other radionuclides were analyzed in sediments collected in the eighties. Sediments were analyzed for <sup>99</sup>Tc beginning in 1983 (Fleming and Ross 1984), and for <sup>235</sup>U, <sup>238</sup>U, <sup>236</sup>U, <sup>232</sup>Th, <sup>222</sup>Th, <sup>223</sup>Th, <sup>223</sup>Ra, <sup>224</sup>Ra, <sup>224</sup>Ra, <sup>226</sup>Ra, <sup>228</sup>Ra, <sup>238</sup>Pu, <sup>239</sup>. <sup>240</sup>Pu and <sup>99</sup>Tc beginning in 1986 (WMCO 1987). The data indicate no significant difference in average concentrations of these radionuclides in sediments collected from the Great Miami River upstream and downstream of the FMPC effluent discharge line.

Sediment from onsite locations in Paddy's Run and the Storm Sewer Outfall Ditch have been sampled and analyzed for uranium since 1974. Paddy's Run has been divided into three general areas for sediment sampling purposes: upstream from just north of the waste pits to the confluence with the storm sewer outfall ditch, along the outfall ditch, and downstream of the confluence with the outfall ditch to the site boundary. The sampling locations in these three areas have varied from just a few to over 70 in 1989 (WMCO 1990). The uranium concentrations in onsite samples from these locations generally varied spatially and temporally (Facemire et al. 1985). The temporal variation can most likely be related to the erratic and seasonal water flow in the creek. In 1985 the sediment collection process was standardized to decrease the variation in results from location to location by erecting permanent steel posts at all Paddy's Run and storm sewer outfall ditch sampling points (Aas et al. 1986). From 1986 through 1990, the FMPC focused on characterizing the sediments in Paddy's Run and in the storm sewer outfall ditch. During this time, more than 750 sediment samples from along Paddy's Run were analyzed by an offsite laboratory for eleven radionuclides. Table B6-4 displays the data for uranium, <sup>99</sup>Tc and <sup>226</sup>Ra concentrations in sediments from the Paddy's Run. The data indicate that higher concentrations of most radionuclides from onsite samples were associated with pools or areas in Paddy's Run where sediments tended to settle, or where infiltration may have occurred (NLCO 1955, NLCO 1956, NLCO 1959).

With the commencement of the sediment characterization program in 1985, offsite sediment sampling was done for the first time in Paddy's Run south of Willey Road. Offsite sediment samples north of the site in Paddy's Run were not obtained until 1991 (FEMP 1992). Figure B6-5 shows the annual average uranium concentration in sediments from Paddy's Run below the confluence of the storm sewer outfall ditch (SSOD) varies directly with the uranium concentration in water from the same location, suggesting that uranium is flushed regularly from sediments, preventing any long-term uranium accumulation. In 1987, the concentration in both water and sediment from below the SSOD decreased markedly when the storm water retention basin became operational and began receiving runoff that had previously gone directly to Paddy's Run.

	Uranium	$\frac{(\mathbf{n}\mathbf{Ci}\boldsymbol{\sigma}^{-1})^{n}}{(\mathbf{n}\mathbf{Ci}\boldsymbol{\sigma}^{-1})^{n}}$	99 Te (	nCi m <sup>1</sup> )	<sup>226</sup> Ra	$(nCi \sigma^1)$
Year	Onsite north of waste pits	Offsite below Willey Road	Onsite north of waste pits	Offsite below Willey Road	Onsite north òf waste pits	Offsite below Willey Road
1985	1.0	4.80 <sup>h</sup>	0.5	2.3	na <sup>d</sup>	na <sup>d</sup>
1986	1.5	10 <sup>b</sup>	<0.5	<0.5	0.86	0.83
1987	0.86	0.44	<1.1	<1.2	0.67	0.56
1988	1.2	1.5	<1.1	<0.90	0.65	0.69
1989	<2.4	<2.2	<(),9()	<(),9()	0.63	0.5
1990	2.8	1.6	< 0.71	<0.77	0.89	0.7
1991	1.4 °	1.2	na <sup>d</sup>	na	0.64	na <u>d</u>

Table B6-4.	Sediment	Sampling	<b>Results</b> in	Paddy's Rur

 $^n\,$  Measurement data for  $^{238}{\rm U}$  concentrations for 1986 to 1990 have been standardized to total uranium.

<sup>b</sup> These averages include onsite locations above Willey Road at the confluence of the storm sewer outfall ditch (SSOD) with Paddy's Run. Data were not separated by offsite and onsite locations.

' Includes sediment 'samples taken north of Route 126 (offsite), the first year that a background offsite sample was taken north of the site.

<sup>d</sup> Analysis not done.

Appendix B — Part 6 River-Sediment-Fish \* **\* \* 3** 3 9



**Figure B6-5.** Annual average uranium concentration in sediments from Paddy's Run above and below the confluence of the storm sewer outfall ditch, and in water from below the confluence of the storm sewer outfall ditch (SSOD). In 1987, the storm water retention basin became operational and received runoff that had gone directly to Paddy's Run.

#### FISH SAMPLING

Routine sampling of fish from the Great Miami River near the FMPC began only in 1984. We have not located other data regarding uranium measurements in fish from the river before this time. Therefore, we compiled these more recent data for use in validating our source term estimates for this time period and to, perhaps, calculate a site-specific Bioaccumulation Factor (BF) for uranium for dose calculations in Task 6.

Approximately 25 fish were analyzed each year from each of three locations on the river: 2.5 km upstream, at the main effluent outfall location, and downstream where Paddy's Run drains into the river (Figure B6-1). After collection, the fish are placed in plastic bags and packed in ice. Later they are scaled, and the heads and entrails removed. Fish are filleted if their total weight is greater than 800–900 grams (about 2 lb.). The fillets are frozen, packed in dry ice and shipped to an independent testing laboratory for uranium analysis. Figure B6 –6 shows average uranium concentrations measured in fish fillets taken upstream, at the main effluent outfall, and downstream of the FMPC from 1984 through 1990. Except for 1988, there appears to be a downward trend from 1984 to 1987. However, for each year, the uranium concentrations are not different among the three locations. The 1988 results were questioned in the annual Environmental Monitoring Report, but no reason was given for the high values. Table B6-5 lists the uranium concentration values used in Figure B6-6.



**Figure B6-6**. Average uranium concentrations measured in fish fillets taken upstream, at the main effluent outfall, and downstream of the FMPC. The uranium concentrations in water from the river near the New Baltimore Bridge ranged from 1.4 to  $2.1 \text{ pCi L}^{-1}$  during this time.

From	From the oreat mann tiver hear the FMFC					
Year	2.4 km Upstream	Near Effluent Outfall	Downstream at Confluence with Paddy's Run			
1984	0.24	0.30	0.22			
1985	0.11	0.16	0.09			
1986	0.07	0.06	0.07			
1987	0.01	0.01	0.01			
1988	0.11	0.13	0.30			
1989	0.01	0.01	0.02			
1990	0.06	0.01	0.02			

Table B6-5. Measured Uranium Concentration in Fish (pCi g<sup>-1</sup>) From the Great Miami River Near the FMPC

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#### ANNEX TO APPENDIX B PART 6

#### DETAILED DATA TABLES

#### Table B6S-1. Reported Uranium and Radium Concentrations in the Miami River and Paddy's Run in 1953 and 1954 <sup>a</sup>

Great Miami River						
	Ura	nium (mg I	<u></u>	Total rac	lium (mmg	mL <sup>-1</sup> ) <sup><i>b</i></sup>
Sample		New			New	
Collection	Venice	Baltimore	Miamitown	Venice	Baltimore	Miamitown
Date	Bridge	Bridge	Bridge	Bridge	Bridge	Bridge
	(upstream)			(upstream)		
17-Sep-53	0.02	0.055	0.021	0.011	0.009	0.006
29-Jan-54	0.005	0.046	0.021	0.0033	0.0013	0.002
	0.023	0.004	0.019	0.002	0.0001	0.0006
		0.005			0.0008	
8-Mar-54	0.006	0.011	0.003	0.0008	0.0013	0.0008
	0.005	0.007	0.009	0.0008	0.0004	0.0007
			Paddy's Run			
	Urani	um (mg L <sup>-</sup>	1)	Total ra	ndium (mm	$g m L^{-1} b$
	Willey Road			Willey F	load	
	Bridge	New	Haven	Bridg	e N	ew Haven
17-Sep-53	0.013			0.001	3	
29-Jan-54	0.141	(	).2	0.69		0.5
8-Mar-54	0.876	1	.08	0.018	3	0.045
29-Dec-54	0.112	0.	008			
" From Barry 1953 and NLCO 1954.						
The symbo	<u>οι, μμ. is an o</u>	utdated, pr	eviously-used	<u>notation, which</u>	<u>is equival</u>	e <u>nt to pico.</u>

		<u> </u>	mann nive	er water in Ia	999 -		
1955	Venice	New		1955	Venice	New	
Collection	Bridge	Baltimore	Miamitown	Collection	Bridge	Baltimore	Miamitown
Date	(upstream)	Bridge	_Bridge	Date	(upstream)	Bridge	<u>Bridge</u>
11-Jan	0.052	0.204	0.064	6-Jun	0.096	0.064	0.019
	0.1	0.064	0.072		0.148	0.056	0.02
	0.084	0.092		24-Jun	0.005	0.045	0.062
1-Feb	0.011	0.017	0.023		0.037	0.009	0.02
	0.018	0.018	0.028		0.065		
	0.024	0.014		30-Jun	0.011	0.055	0.033
14-Feb		0.028			0.012	0.023	0.06
		0.036		8-Jul	0.157	0.065	0.04
16-Feb	0.009	0.064	0.04		0.028	0.037	0.016
	0.008	1.02			0.018	0.051	
21-Feb	0.028	0.012		18-Jul	0.056	0.102	0.032
	0.018	0.024			0.102	0.102	0.084
7-Mar	0.022	0.062		28-Jul	0.015	0.111	0.12
	0.008	0.034			0.008	0.042	0.042
	0.054	0.038		19-Aug	0.009	0.064	0.016
14-Mar	0.025	0.204		_	0.02	0.072	0.064
	0.024	0.5		26-Aug		0.063	
	0.084	0.186		30-Aug	0.005	0.005	0.019
24-Mar	0.048	0.028			nd <sup>//</sup>	0.005	0.021
5-Apr	0.126	0.016	nd <sup>h</sup>	9-Sep	0.007	0.01	0.01
			nd <sup>b</sup>		0.008	0.022	0.043
21-Apr	0.169	0.096	0.076	22-Sep	0.02	0.028	0.013
	0.112	0.088	0.058		0.036	0.064	0.048
22-Apr	0.062	nd <sup>h</sup>	0.012	30-Sep	0.068	0.03	0.017
	0.032	0.012	0.014		0.016	0.078	0.068
25-Apr		0.014	0.008	19-Oct	0.036	0.03	0.056
		0.011		<b>.</b>	0.056	0.064	0.058
27-Apr	nd <sup>b</sup>	0.615	0.022	31-Oct	0.076	0.058	0.06
	0.004	0.053	0.026		0.046	0.07	0.052
5-May	0.56	0.082	0.022	10-Nov	0.038	0.062	0.038
•	0.005	0.58	0.004		0.068	0.026	0.018
		3.46		18-Nov	0.03	0.064	0.028
18-May	0.664	14.22	0.041		0.032	0.036	0.008
·	0.214	0.214	0.019	Avg (mg $L^{-1}$ )	0.08	0.36	0.04
26-Mav	0.568	0.03	0.148	Stdev (mg L <sup>-1</sup> )	0.13	1.8	0.03
•	0.041	0.011	0.014	Avg (pCi $L^{-1}$ )	51	240	27
				Stdev (pCi L <sup>-1</sup> )	88	1200	20
' From NL	.CO 1955.			<u> </u>	<u></u> ,		<u> </u>

Table B6S-2. Measured Uranium Concentration	(mg L <sup>-1</sup>	)
in Miami River Water in 1955 <sup>a</sup>		

<sup>h</sup> Not detectable.

11	n Miami River	r Water in 1956	<i>a</i>
1956	Venice	New	Miamitown
Collection	Bridge	Baltimore	Bridge
Date	(upriver)	(downstream)	(downstream)
5-Jan	0.084	0.092	0.062
	0.062	0.052	0.246
8-Feb	1.17	1.16	0.14
	0.158	0.176	0.3
6-Jun	0.816	0.018	0.154
	0.078	0.116	0.028
11-Jun	0.126	0.02	0.018
	0.044	0.036	0.032
13-Jun	0.484	0.056	
22-Jun	0.034	0.154	0.046
	0.068	0.096	0.03
26-Jun	0.04	0.096	0.096
	0.096	0.042	0.06
6-Jul	0.004	0.019	0.003
	0.01	0.019	0.015
12-Jul	0.003	0.008	0.026
	0.003	0.021	0.014
19-Jul		0.019	0.008
24-Jul	0.016	0.015	0.007
14-Aug	0.027	0.039	0.007
23-Aug	0.007	0.028	0.01
	0.007	0.016	0.02
27-Aug	0.175	0.037	0.248
29-Aug	0.036	0.04	0.029
10-Sep	0.101	0.23	0.147
13-Sep	0.023	0.031	0.064
19-Sep	0.007	0.023	0.012
25-Sep	0.068	0.043	0.039
	0.034	0.026	0.064
1-Oct	0.213	0.018	0.038
	0.017	0.028	0.032
5-Oct	0.014	0.024	0.02
	0.026	0.032	0.037
6-Oct	nd <sup><i>h</i></sup>	0.041	0.006
	0.068	0.179	0.013

#### Table B6S-3. Measured Uranium Concentration (mg L<sup>-1</sup>) in Miami River Water in 1956 <sup>a</sup>

(continued next page)

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in Miami River Water in 1956 <sup><i>a</i></sup> (continued)						
1956	Venice	New	Miamitown			
Collection	Bridge	Baltimore	Bridge			
Date	(upriver)	(downstream)	(downstream)			
8-Oct	0.025	0.022	0.038			
	0.038	0.016	0.068			
9-Oct	0.005	0.041	0.028			
	0.003	0.026	0.016			
10-Oct	0.008	0.044				
	0.009	0.03				
11-Oct	0.068	0.128				
	0.012	0.03				
12-Oct	0.005	0.02	0.034			
	0.012	0.012	0.026			
16-Oct	0.006	0.011	0.012			
	0.002	0.009	0.014			
17-Oct	0.057	0.06	0.055			
	0.003	0.031	0.018			
18-Oct	0.019	0.046				
	0.007	0.018				
22-Oct			0.016			
			0.032			
25-Oct	0.018	0.01	0.017			
	0.002	0.024				
8-Nov	0.026	0.058	0.035			
	0.026	0.086	0.067			
20-Dec	0.058	0.058	0.067			
	0.026	0.106	0.086			
$Avg (mg L^{-1})$	0.08	0.069	0.053			
Stdev (mg $L^{-1}$ )	0.20	0.15	0.064			
Avg (pCi $L^{-1}$ )	54	67	36			
Stdev (pCi L <sup>-1</sup> )	140	160	44			
" From NLCO 19	956.					
<sup>b</sup> None detected.						

Table B6S-3.	Measured	Uranium Cor	ncentration	$(\mathbf{mg} \mathbf{L}^{-1})$
in Miam	ni R <u>iver W</u> a	ter in 1956 a (	(continued)	

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In	Miami River	water in 1957	
1957 Collection	Venice Bridge	New Baltimore	Miamitown
Date	(upstream)	(downstream)	(downstream)
23-Jan	0.18	0.68	1.02
	0.3	0.52	0.34
18-Feb	0.034	0.074	0.012
	0.544	0.098	0.052
26-Mar	0.03	0.01	0.04
	0.02	0.01	0.015
8-Apr	0.009	0.042	0.011
	0.041	0.009	0.018
24-Apr	0.013	0.002	0.009
	0.005	0.011	0.005
14-May	0.001	0.001	0.01
	0.006	0.008	0.031
19-Jun	0.1	0.1	0.05
	0.02	0.02	0.06
19-Jul	0.004	0.01	0.025
	0.009	0.01	0.013
	0.007	0.002	0.014
	nd <sup><i>h</i></sup>	0.025	0.013
20-Aug	0.027		0.067
30-Sep	0.005	0.005	
	0.034	0.025	
3-Oct	0.006	0.009	•
	0.004	0.001	•
17-Oct	0.004	0.016	
26-Nov	0.005	0.005	
	0.002	0.007	
20-Dec	0.008	0.01	
$Avg(mg L^{-1})$	0.05	0.07 ·	0.10
Stdev (mg $L^{-1}$ )	0.12	0.16	0.24
Avg ( $pCiL^{-1}$ )	37	45	65
Stdev (pCi L <sup>-1</sup> )	81	110	160
a From NLCO a	nalytical data sh	eets for 1957	

Table B6S-4. Measured Uranium Concentration (mg L<sup>-1</sup>) in Miami River Water in 1957 <sup>a</sup>

h None detected.

<b></b> ¥	-				Page B6–21
2	17	Q	9	0	
		0	6.4		

1959	• • • • • • • • • • • • • • • • • • • •		1959	•.	
Collection	Venice Bridge	New Baltimore	Collection	Venice Bridge	New Baltimore
Date	(upstream)	(downstream)	Date	(upstream)	(downstream)
5-Jan	0.009	0.009	9-Sep	0.018	0.023
4-Feb	0.012	0.038	11-Sep	0.007	0.008
2-Mar	0.006	0.01	24-Sep	0.006	0.009
3-Mar	0.002	0.004		0.037	
19-Mar	0.006	0.008	28-Sep	0.008	0.008
30-Mar	0.005	0.017	1-Oct	0.006	0.007
28-Apr	0.014	0.027	12-Oct	0.008	0.026
6-May	0.009	0.004	16-Oct	0.02	0.002
20-May	0.006	0.009		0.013	
26-May	0.002	, 0.014	26-Oct	0.013	0.008
9-Jun	0.005	0.004	30-Oct		0.009
26-Jun	0.011	0.01	18-Nov	0.019	0:019
29-Jun	0.005	0.008	25-Nov	0.047	0.033
10-Jul	0.005	0.003	30-Nov	0.004	0.021
15-Jul	0.096	0.009	16-Dec	0.021	0.018
27-Jul	0.001	0.004	23-Dec	0.016	0.057
6-Aug	0.013	0.012	28-Dec	0.006	0.01
11-Aug	0.019	0.006	30-Dec	0.11	0.13
-		0.006	$Avg(mg L^{-1})$	0.02	0.02
		0.01	Stdev (mg $L^{-1}$ )	0.02	0.02
25-Aug	0.009	0.013	$Avg(pCiL^{-1})$	11	11
31-Aug	0.003	0.013	Stdev (pCi L <sup>-1</sup> )	16	15
<sup>o</sup> From NLCO	1959		· · · · · · · · · · · · · · · · · · ·		

# Table B6S-5. Measured Uranium Concentration (mg L<sup>-1</sup>) in Miami River Water in 1959 °

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	<u> </u>	liami River	water in 1963	3	
1963	Venice	New	1963	Venice	New
Collection	Bridge	Baltimore	Collection	Bridge	Baltimore
Date	(upstream)	<u>Bridge</u>	Date	(upstream)	Bridge
3-Jan	0.01	0.006	3-Sep	0.008	0.008
10-Jan	0.005	0.015	13-Sep	0.012	0.012
17-Jan	0.03	0.01	17-Sep	0.006	0.011
24-Jan	0.01	0.03	27-Sep	0.007	0.011
31-Jan	0.01	0.01	1-Oct	0.007	0.008
7-Feb	0.01	0.01	11-Oct	0.006	0.02
14-Feb	0.01	0.02	15-Oct	0.009	0.027
21-Feb	0.003	0.013	16-Oct		0.027
28-Feb	0.006	0.007	17-Oct		0.034
7-Mar	0.012	0.01	18-Oct		0.016
14-Mar	0.002	0.008	19-Oct		0.012
21-Mar	0.005	0.005	20-Oct		0.022
28-Mar	0.007	0.009	21-Oct		0.012
4-Apr	0.006	0.016	22-Oct		0.016
11-Apr	0.025	0.008	23-Oct		0.018
18-Apr	0.018	0.045	24-Oct		0.021
25-Apr	0.01	0.009	25-Oct	0.003	0.004
2-May	0.02	0.03	26-Oct		0.015
9-May	0.03	0.03	27-Oct		0.009
16-May	0.014	0.006	28-Oct		0.016
23-May	0.01	0.01	29-Oct	0.011	0.011
30-May	0.01	0.02	30-Oct		0.024
6-Jun	0.01	0.01	31-Oct		0.013
13-Jun	0.01	0.01	1-Nov		0.027
20-Jun	0.01	0.01	2-Nov		0.016
25-Jun	0.01	0.01	3-Nov		0.014
27-Jun	0.01	0.01	4-Nov		0.023
5-Jul	0.01	0.01	5-Nov		0.016
9-Jul	0.003	0.013	6-Nov		0.01
19-Jul	0.006	0.012	7-Nov		0.024
23-Jul	0.001	0.003	8-Nov	0.011	0.012
2-Aug	0.01	0.17	9-Nov		0.06
6-Aug	0.006	0.025	10-Nov		0.021
16-Aug	0.003	0.008	11-Nov		0.008
20-Aug	0.007	0.022	12-Nov	0.006	0.015
29-Aug		0.009	13-Nov		0.04
30-Aug	0.006	0.013	14-Nov		0.016

Table B6S-6. Measured Uranium Concentration (mg  $L^{-1}$ ) in Miami River Water in 1963 "

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1963	Vonice	Now	1063 (COI	Vanico	Now
Collection	Bridge	Baltimore	Collection	Bridge	Baltimore
Date	(upstream)	Bridge	Date	(upstream)	Bridge
15-Nov		0.026	28-Nov	· · · ·	0.006
16-Nov	0.007	0.012			0.009
17-Nov		0.018	29-Nov		0.017
18-Nov		0.011	30-Nov		0.011
19-Nov		0.015	1-Dec		0.012
20-Nov		0.014	2-Dec		0.009
21-Nov		0.019	3-Dec		0.011
22-Nov	0.013	0.014	6-Dec	0.006	0.024
23-Nov		0.05	10-Dec	0.005	0.054
24-Nov		0.018	20-Dec	0.018	0.01
25-Nov		0.012	24-Dec	0.005	0.045
26-Nov	0.008	0.028	$Avg(mg L^{-1})$	0.01	0.02
	0.012	0.017	Stdev (mg L <sup>-1</sup> )	0.01	0.02
27-Nov		0.026	Avg (pCi L <sup>-1</sup> )	6.5	12
		0.005	Stdev $(pCiL^{-1})$	4.1	13
From NLCO	1963.			· · · · · · · · · · · · · · · · · · ·	

Table B6S-6. Measured Uranium Concentration (mg L <sup>-1</sup> )	)
in Miami River Water in 1963 <sup>a</sup> (continued)	

		mann nive	water in 150	)4	
1964	Venice	New	1964	Venice	New
Collection	Bridge	Baltimore	Collection	Bridge	Baltimore
<u>Date</u>	(upstream)	Bridge	Date	(upstream)	Bridge
3-Jan	0.033	0.017	17-Aug		0.022
7-Jan	0.019	0.038	18-Aug	0.011	0.02
17-Jan	0.006	0.009	19-Aug		0.009
21-Jan	0.012	0.017	20-Aug		0.008
31-Jan	0.015	0.023	21-Aug		0.013
4-Feb	0.014	0.05	24-Aug		0.009
14-Feb	0.006	0.013	25-Aug		0.016
18-Feb	0.007	0.007	26-Aug		0.018
28-Feb	0.011	0.021	29-Aug		0.007
3-Mar	0.005	0.031	30-Aug		0.006
13-Mar	0.01	0.003	31-Aug	-	0.005
14-Mar		0.007	1-Sep	0.009	0.006
17-Mar	0.009	0.006	2-Sep		0.007
28-Mar	0.008	0.018	3-Sep		0.014
31-Mar	0.005	0.008	4-Sep		0.011
- 10-Apr	0.007	0.006	7-Sep		0.008
14-Apr	0.012	0.011	8-Sep		0.08
24-Apr	0.006	0.005	9-Sep		0.013
28-Apr	0.003	0.003	10-Sep		0.008
8-May	0.045	0.03 <del>9</del>	11-Sep	0.004	0.006
12-May	0.003	0.007	12-Sep		0.007
22-May	0.008	0.012	13-Sep		0.003
28-May	0.017	0.008	14-Sep		0.007
4-Jun	0.009	0.009	15-Sep	0.007	0.006
9-Jun	0.006	0.006	16-Sep		0.009
19-Jun	0.009	0.012	17-Sep		0.012
23-Jun	0.022	0.013	18-Sep		0.01
3-Jul	0.012	0.017	19-Sep		0.012
7-Jul	0.013	0.014	20-Sep		0.022
17-Jul	0.007	0.014	21-Sep		0.008
21-Jul	0.005	0.001	22-Sep		0.022
4-Aug	0.008	0.013	23-Sep		0.027
12-Aug		0.01	24-Sep		0.017
13-Aug		0.011	25-Sep	0.017	0.014
14-Aug	0.012	0.006	26-Sep		0.012
15-Aug		0.027	27-Sep		0.065
16-Aug		0.06	28-Sep		0.017

#### Table B6S-7. Measured Uranium Concentration (mg L<sup>-1</sup>) in Miami River Water in 1964 °

(continued next page)

	III MIAIIII	Rivel water	111904 (COI	itinueu)	
1964	Venice	New	1964	Venice	New
Collection	Bridge	Baltimore	Collection	Bridge	Baltimore
Date	(upstream)	Bridge	Date	(upstream)	Bridge
29-Sep	0.004	0.02	6-Nov	0.006	0.003
30-Sep		0.009	7-Nov		0.006
1-Oct		0.03	8-Nov		0.007
2-Oct		0.032	9-Nov		0.005
3-Oct		0.042	10-Nov	0.006	0.007
4-Oct		0.107	11-Nov		0.006
5-Oct		0.016	12-Nov		0.011
6-Oct		0.005	" 13-Nov		0.007
7-Oct		0.009	15-Nov		0.008
8-Oct		0.002	16-Nov		0.012
9-Oct	0.024	0.2	17-Nov		0.014
10-Oct		0.003	18-Nov		0.011
11-Oct		0.007	19-Nov		0.02
13-Oct	0.003	0.055	20-Nov	0.008	0.019
14-Oct		0.006	21-Nov		0.013
15-Oct		0.003	22-Nov		0.008
16-Oct		0.005	23-Nov		0.01
17-Oct		0.006	24-Nov	0.003	0.005
18-Oct		0.01	25-Nov		0.008
19-Oct		0.018	26-Nov		0.008
20-Oct		0.018	27-Nov		0.01
21-Oct		0.013	28-Nov		0.016
22-Oct		0.019	29-Nov		0.019
23-Oct	0.003	0.01	30-Nov		0.005
24-Oct		0.011	1-Dec		0.005
25-Oct		0.008	2-Dec		0.005
26-Oct		0.018	3-Dec		0.005
27-Oct	0.005	0.007	4-Dec	0.005	0.006
28-Oct		0.008	8-Dec	0.012	0.011
29-Oct		0.021	18-Dec	0.007	0.008
30-Oct		0.01	31-Dec	0.008	0.023
1-Nov		0.015	$Avg(mgL^{-1})$	0.01	0.02
3-Nov		0.075	Stdev (mg $L^{-1}$ )	0.01	0.02
4-Nov		0.009	Avg (pCi $L^{-1}$ )	6.9	11
5-Nov		0.006	Stdev (pCi L <sup>-1</sup> )	5.3	15
" From NLCO	O 1964.				

#### Table B6S-7. Measured Uranium Concentration (mg L<sup>-1</sup>) in Miami River Water in 1964 <sup>a</sup> (continued)

Radiological Assessments Corporation "Setting the standard in environmental health"

+=:	in Miami River Water in 1965							
1965	Venice	New	1965	Venice	New			
Collection	Bridge	Baltimore	Collection	Bridge	Baltimore			
Date	(upstream)	(downstream)	Date	(upstream)	(downstream)			
5-Jan	0.004	0.01	9-Jul	0.006	0.038			
15-Jan	0.007	0.019	13-Jul	0.007	0.006			
19-Jan	0.002	0.006	21-Jul	0.006	0.006			
29-Jan	0.005	0.007	27-Jul	0.006	0.001			
2-Feb	0.004	0.006	6-Aug	0.003	0.007			
12-Feb	0.01	0.014	20-Aug	0.012	0.01			
16-Feb	0.006	0.006	24-Aug	0.006	0.009			
26-Feb	0.009	0.015	3-Sep	0.003	0.008			
2-Mar	0.006	0.004	7-Sep	0.006	0.006			
12-Mar	0.005	0.006	17-Sep	0.008	0.006			
16-Mar	0.404	0.007	21-Sep	0.006	0.006			
19-Mar	0.0 <b>2</b>	0.017	1-Oct	0.005	0.014			
26-Mar	0.05	0.01	15-Oct	0.02	0.018			
30-Mar	0.006	0.006	19-Oct	0.006	0.01			
9-Apr	0.001	0.025	29-Oct	0.01	0.01			
13-Apr	0.022	0.01	2-Nov	0.006	0.01			
23-Apr	0.014	0.023	3-Nov	0.19	0.03			
27-Apr	0.009	0.01	12-Nov	0.003	0.016			
6-May	0.006	0.014	16-Nov	0.01	0.011			
11-May	0.005	0.013	26-Nov	0.009	0.009			
21-May	0.009	0.022	30-Nov	0.009	0.01			
25-May	0-Jan	0.026	10-Dec	0.002	0.008			
4-Jun	0.008	0.012	14-Dec	0.006	0.013			
8-Jun	0.013	0.009	22-Dec	0.003	0.007			
18-Jun	0.011	0.01	28-Dec	0.005	0.007			
22-Jun	0.006	0.008	Avg (mg $L^{-1}$ )	0.01	0.01			
1-Jul	0.006	0.012	Stdev(mg L <sup>-1</sup> )	0.03	0.01			
4-Jul	0.008	0.014	Avg (pCi L <sup>-1</sup> )	7.6	7.9			
5-Jul	0.005	0.008	Stdev(pCi L <sup>-1</sup> )	17	4.7			
<sup>n</sup> From NLCO	1965.							

Table B6S-8.	Measured	Uranium	Concentration	$(mg L^{-1})$
	in Miami H	River Wate	er in 1965 °	

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	in Paddy's Run Water in 1955 "						
1955		Willey	New	1955		Willey	New
Collection	"Above"	Road-	Haven	Collection	"Above"	Road	Haven
Date	SSOD h	Bridge	Road	Date.	SSOD *	Bridge	Road
6-Jan	0.139	0.514		8-Apr	0.006	0.213	
9-Jan	0.046	0.055		11-Apr	0.029	0.63	
11-Jan		0.088	0.097	14-Apr	0.074	0.408	
12-Jan	0.065	0.046		17-Apr	0.051	0.093	
15-Jan	0.126	0.025		20-Apr	0.012	0.084	
18-Jan	0.012	0.13	0.079	21-Apr		0.032	0.106
21-Jan	0.079	0.195	0.361	22-Apr		0.013	0.044
24-Jan	0.093	0.076	0.167	23-Apr	0.106	0.058	
27-Jan	0.213	0.056	0.301	25-Apr	0.082	0.021	
30-Jan	0.046	0.06	0.148	28-Apr	0.025	0.148	
1-Feb		0.241	0.148	2-May	0.077	0.091	
2-Feb	0.098	0.38		5-May	0.018	0.284	0.148
5-Feb	0.013	0.324		8-May	0.011	0.193	
8-Feb	0.023	0.03		11-May	0.011	0.051	
11-Feb	0.04	0.026		14-May	0.02	0.14	
14-Feb				17-May	0.014	0.8	
15-Feb	0.062			18-May		0.132	0.009
16-Feb				20-May	0.064	0.14	
18-Feb	0.022	0.065		23-May	0.041	0.332	
21-Feb	0.126	0.122		26-May	0.025	0.076	0.045
24-Feb	0.006	0.06		29-May	0.286	0.11	
27-Feb	0.038	0.074		1-Jun	0.295`	0.028	
2-Mar	0.008	0.05		4-Jun	0.02	0.051	
7-Mar		0.06	0.056	6-Jun		0.069	0.166
8-Mar	0.013	0.052		7-Jun	0.099	0.5	
11-Mar	0.074	0.773		9-Jun	0.058	0.175	-
14-Mar	0.06	0.079		12-Jun	0.042	0.069	
17-Mar	0.056	0.038		15-Jun	0.001	0.102	
21-Mar	0.074	0.148		16-Jun	0.102	0.12	
24-Mar	0.039	0.144	0.06	19-Jun	0.023	0.148	
27-Mar	0.052	0.023		22-Jun	0.065	0.152	
30-Mar	0.02	0.056		24-Jun		0.134	0.074
2-Apr	0.028	0.079		25-Jun	0.138	0.166	
5-Apr	0.026	0.139		28-Jun	0.019	0.129	

Table B6S-9. Measured Uranium Concentrations (mg  $L^{-1}$ )

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(continued next page)

Radiological Assessments Corporation "Setting the standard in environmental health"

1955WilleyNew1955WilleyNCollection"Above"RoadHavenCollection"Above"RoadHaDateSSOD hBridgeBridgeDateSSOD hBridgeBridgeBridge30-Jun0.2220.23512-Oct0.0480.029	ew iven idge
Collection"Above"RoadHavenCollection"Above"RoadHaDateSSOD hBridgeBridgeDateSSOD hBridgeBridgeBridge30-Jun0.2220.23512-Oct0.0480.029	iven idge
DateSSOD hBridgeBridgeDateSSOD hBridgeBri30-Jun0.2220.23512-Oct0.0480.029	idge
30-Jun 0.222 0.235 12-Oct 0.048 0.029	
1-Jul 0.037 0.185 15-Oct 0.02 0.068	
4-Jul 0.024 0.217 18-Oct 0.029 0.15	
7-Jul 0.029 0.721 19-Oct 0.03 0.0	038
8-Jul 0.39 0.488 21-Oct 0.009 0.169	
0.244 24-Oct 0.007 0.67	
10-Jul 0.164 0.155 27-Oct 0.009 0.008	
13-Jul 0.034 0.258 30-Oct 0.007 0.014	
16-Jul 0.028 0.126 31-Oct no flow 0.0	034
18-Jul 0.111 0.156 2-Nov 0.02 0.566	
19-Jul 0.097 1.03 5-Nov 0.017 0.082	
22-Jul 0.03 0.029 9-Nov 0.058 0.073	
25-Jul 0.121 0.005 10-Nov 0.027 0.0	068
28-Jul 0.087 0.175 0.074 12-Nov 0.003 0.028	
0.242 15-Nov 0.009 0.396	
1-Aug 0.034 0.189 18-Nov 0.141 0.021 0.0	039
4-Aug 0.03 0.223 0.075	
8-Aug 0.309 21-Nov 0.017 0.019	
11-Aug 0.121 24-Nov 0.037 0.047	
19-Aug 0.126 0.028 30-Nov 0.013 0.042	
20-Aug 0.193 3-Dec 0.033 0.028	
26-Aug 6-Dec 0.024 0.047	
27-Aug 0.069 9-Dec 0.015 0.047	
30-Aug no flow 0.058 12-Dec 0.018 0.027	
15-Dec 0.015 0.075	
9-Sep 18-Dec 0.017 0.035	
21-Sep 0.019 0.073 21-Dec 0.015 0.015	
22-Sep 0.082 0.116 24-Dec 0.035 0.025	
24-Sep 0.222 0.063 28-Dec 0.02 0.019	
27-Sep $0.058$ Avg (mg L <sup>-1</sup> ) 12 43	27
30-Sep 0.038 0.048 0.058 Stdev (mg L <sup>-1</sup> ) 25 94	33
0.068 Aver (nCi L <sup>-1</sup> ) 35 100	83
3-Oct 0.009 0.013 Stdev (pCi L <sup>-1</sup> ) 38 120	75
6-Oct 0.029 0.102	
9-Oct 0.015 0.037	

Table B6S-9. Measured Uranium Concentrations (mg L <sup>-</sup>	1)
in Paddy's Run Water in 1955 <sup>a</sup> (continued)	

 <sup>a</sup> From NLCO analytical data sheets for 1955.
 <sup>b</sup> These samples were collected onsite above the confluence of the storm sewer outfall ditch (SSOD) with Paddy's Run.

in Paddy's Run water in 1956 "								
1956		Willey	New	1956		Willey	New	
Collection	"Above"	Road	Haven	Collection	"Above"	Road	Haven	
Date	SSOD *	Bridge	Bridge	Date	SSOD h	Bridge	Bridge	
1-Jan	0.034	0.044		15-Jul	0.031	0.068		
4-Jan	0.012	0.035	0.03	17-Jul		0.102	·	
7-Jan	0.026	0.031		22-Jul	0.003	0.205		
10-Jan	0.008	0.031		29-Jul	0.053	1.36		
13-Jan	0.032	0.011		5-Aug	0.004	0.425		
16-Jan	0.024	0.023		12-Aug	0.015	0.63		
22-Jan	0.036	0.019		14-Aug			0.107	
28-Jan	0.009	0.044		19-Aug	0.012			
1-Feb	0.026	0.057		21-Aug		0.792		
4-Feb	0.044	0.021		23-Aug		1.29	0.046	
8-Feb		0.088	0.068	26-Aug	0.064			
15-Apr	0.008	0.217		2-Sep	0.019	no flow		
18-Apr	0.015	0.039		9-Sep	0.347	0.162		
27-Apr		0.029		23-Sep	0.043			
30-Apr	0.048	0.049		30-Sep	0.005		0.034	
6-May	0.027	0.044		7-Oct	0.022	2.29		
14-May	0.008	0.035		14-Oct	0.153			
20-May	0.052	0.029		21-Öct	0.672			
27-May	0.022	0.078		28-Oct	0.071	1.92		
28-May		0.052		4-Nov	0.038			
28-May		0.306		18-Nov	0.058			
31-May	0.009	0.087		21-Nov	0.027	2.09		
3-Jun	0.035	0.106		3-Dec	0.053	3.99		
6-Jun		0.077	0.145	7-Dec		1.14		
		0.436		8-Dec		1.71		
9-Jun	0.007	0.306		9-Dec	0.19	0.25		
11-Jun		0.319	0.15	10-Dec		0.086		
17-Jun	0.494	0.028		13-Dec		0.317		
21-Jun		0.29		16-Dec	0.12	0.134		
24-Jun	0.036	0.194		19-Dec	0.23	0.144		
26-Jun		0.044	0.097	26-Dec		0.106		
4-Jul	0.116	0.156		31-Dec	0.058	0.086		
7-Jul	0.156	0.058		$Avg(mgL^{-1})$	0.08	0.37	0.08	
10-Jul		0.054		Stdev (mg $L^{-1}$ )	0.13	0.70	0.05	
12-Jul		0.027	0.022	Avg (pCi $L^{-1}$ )	55	240	53	
13-Jul		0.449		Stdev (pCi L <sup>-1</sup> )	91	468	33	

Table B6S-10. Measured Uranium Concentrations (mg L<sup>-1</sup>)

<sup>9</sup> From NLCO analytical data sheets for 1956.

 $^{h}$  These samples were collected onsite above the confluence of the storm sewer outfall ditch (SSOD) with Paddy's Run.

in raduy s run water in 1957 "										
1957		Willey	1957		Willey					
Collection	"Above"	Road	Collection	"Above"	Road					
Date	SSOD h	Bridge ^	Date	SSOD <sup>b</sup>	Bridge <sup>c</sup>					
3-Jan		0.115	26-Mar	0.001	0.047					
6-Jan		0.048	1-Apr	0.001	0.092					
11-Jan		0.043	5-Apr		0.015					
13-Jan	0.009	0.018	6-Apr		0.004					
20-Jan	0.002	0.021			0.036					
22 <i>-</i> Jan		0.582	9-Apr		0.064					
23-Jan	0.008	0.003	8-Apr		0.166					
	0.013	nd <sup>b</sup>		0.003	0.092					
27-Jan	0.043	0.068	12-Apr		0.055					
30-Jan	0.06	1.385	15-Apr		0.036					
31-Jan	0.06	0.026	17-Apr		0.083					
3-Feb	0.002	0.068	18-Apr		0.053					
6-Feb	0.066	0.102	21-Apr		0.032					
9-Feb	0.06	0.051	24-Apr	0.032	0.02					
10-Feb	0.012	0.025	26-Apr		0.017					
12-Feb	0.025	0.019	29-Apr		0.074					
17-Feb	0.004	0.085	2-May		0.149					
18-Feb	0.018	0.323	12-May	0.048	0.137					
21-Feb	0.06	0.051	14-May	0.36	0.033					
24-Feb	0.162	0.049	19-May		0.728					
24-Feb	0.051	0.128	22-May		0.137					
2-Mar	0.048	0.069	25-May		0.073					
3-Mar	0.043	0.74	27-May		0.127					
5-Mar		0.06	28-May		0.081					
8-Mar		0.582	30-May		0.064					
10-Mar		0.028	31-May		0.063					
14-Mar		0.051	3-Jun ·		0.076					
17-Mar		0.582	19-Jun	0.004	0.004					
20-Mar		0.332	20-Dec	0.009	0.021					
23-Mar		0.064	$Avg(mg L^{-1})$	0.15	0.05					
26-Mar		0.873	Stdev (mg $L^{-1}$ )	0.26	0.07					
29-Mar		0.046	Avg (pCi L <sup>-1</sup> )	100	31					
31-Mar	0.012		Stdev (pCi $L^{-1}$ )	170	49					

Table B6S-11. Measured Uranium Concentrations (mg  $L^{-1}$ ) in Paddy's Run Water in 1957 <sup>a</sup>

<sup>a</sup> From NLCO analytical data sheets for 1957.

 $^{b}$  These samples were collected onsite above the confluence of the storm sewer outfall ditch (SSOD) with Paddy's Run.

<sup>c</sup> No samples taken from New Haven Road.

Measured in Faddy's Run in 1959										
1959		Willey	1959	•.	Willey					
Collection	"Above"	Road	Collection	"Above"	Road					
Date	SSOD h	Bridge	Date	SSOD b	Bridge					
1-Jan		0.3	29-May		0.086					
5-Jan		0.06	19-Jul		2.19					
16-Jan		0.34	29-Jul	0.01						
20-Jan		0.032	31-Jul		3.2					
26-Jan		0.07	4-Aug		3.14					
29-Jan		0.04	5-Aug	0.29						
1-Feb		0.08	6-Aug		3.14					
3-Feb		0.29	12-Aug	0.008						
7-Feb		0.32	17-Aug		15.68					
10-Feb		0.06	19-Aug	0.011						
13-Feb		0.12	26-Aug		3.32					
16-Feb		0.04	26-Aug	0.007						
19-Feb		0.39	29-Aug		0.29					
22-Feb		2.25	2-Sep	0.04	0.17					
25-Feb		0.07	5-Sep		0.39					
28-Feb		0.38	9-Sep	0.019						
3-Mar		0.03	16-Sep	0.29						
6-Mar		0.12	23-Sep	0.05						
9-Mar		0.45	30-Sep	0.034	3.61					
12-Mar		0.23	5-Oct	0.015	3.6					
14-Mar		0.07	8-Oct		4.37					
18-Mar		0.09	11-Oct		3.71					
21-Mar		0.04	14-Oct	0.013	3.42					
24-Mar		0.04	21-Oct	0.008	0.22					
27-Mar		0.08	28-Oct	0.056	0.4					
30-Mar		0.04	4-Nov	0.027	1.81					
2-Apr		0.06	11-Nov	0.01	0.59					
1-Apr		0.013	15-Nov		5.16					
5-Apr		0.05	19-Nov	0.013	6.56					
8-Apr		0.19	29-Nov	0.013	0.84					
11-Apr		0.15	9-Dec	0.007	0.52					
14-Apr		0.05	14-Dec	0.01	0.54					
22-Apr		0.57	23-Dec	0.024	0.21					
30-Apr		0.1	30-Dec	0.074	0.28					
3-May		0.14	Avg (mg $L^{-1}$ )	0.04	1.2					
13-May		0.44	Stdev(mg L <sup>-1</sup> )	0.08	2.4					
16-Mav		0.069	Ave (pCi L=b)	30	790					
19-May		0.08	Stdev(pC; I-L)	54	1600					
26-May		0.59	Graeveport 1	<b>~</b> *	2000					
		0.00	1							

Table B6S-12. Uranium Concentrations (mg L <sup>-1</sup> )	
Measured in Paddy's Run in 1959 <sup>a</sup>	

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<sup>*a*</sup> From NLCO analytical data sheets for 1959.

<sup>*b*</sup> These samples were collected onsite above the confluence of the storm

sewer outfall ditch (SSOD) with Paddy's Run.

Measured in Paddy's Run in 1963 a									
1963	Bridge	Willey	New	1963	Bridge	Willey	New		
Collection	North of	Road	Haven	Collection	North of	Road	Haven		
Date	Route 126	Bridge	Bridge	Date	Route 126	Bridge	Bridge		
2-Jan	0.001	1.4	0.04	10-May	0.02		0.03		
5-Jan			0.02	13-May	0.006	0.16	0.08		
8-Jan			0.06	16-May		0.06	0.05		
10-Jan	0.02		0.05	19-May		0.012	0.08		
14-Jan		0.014	0.06	22-May	0.002	0.24	0.14		
17-Jan	0.004		0.07	25-May		0.06	0.05		
23-Jan	0.01		0.03	30-May	0.01		0.02		
26-Jan			0.02	3-Jun <sub>/</sub>	0.003		0.02		
29-Jan		no flow	0.03	6-Jun			0.016		
2-Feb			0.02	9-Jun			0.02		
4-Feb		0.52	0.04	12-Jun	0.01		0.03		
7-Feb	0.01	0.12	0.04	15-Jun			0.03		
10-Feb		0.1	0.04	18-Jun			0.03		
13-Feb	0.004	0.07	0.1	21-Jun	0.01		0.05		
16-Feb		•	0.05	24-Jun	0.01		0.04		
19-Feb	0.005		0.08	27-Jun			0.01		
22-Feb			0.035	30-Jun			0.03		
25-Feb			0.02	3-Jul			0.02		
28-Feb	0.007		0.024	6-Jul			0.02		
3-Mar		0.24	0.18	9-Jul	0.02		0.03		
7-Mar	0.006	0.05	0.15	10-Jul			0.02		
9-Mar		0.035	0.042	11-Jul			0.04		
12-Mar	-	0.018	0.032	12-Jul			0.02		
15-Mar	0.005	0.04	0.1	13-Jul		3.1	0.03		
18-Mar	0.014	0.11	0.05	14-Jul		14.06	0.04		
21-Mar		0.07	0.06	15-Jul			0.02		
24-Mar		0.042	0.035	18-Jul			0.02		
29-Mar	0.011	0.15	0.05	20-Jul		6.6	0.046		
1-Apr		1.0	0.05	21-Jul		1.7	0.68		
4-Apr	0.004	0.044	0.041	23-Jul	0.001		0.13		
7-Apr			0.06	27-Jul			0.03		
10-Apr	0.009		0.042	30-Jul			0.059		
13-Apr			0.11	2-Aug	0.004		0.1		
16-Apr	0.018		0.044	5-Aug	0.004		0.32		
20-Apr		0.13	0.12	8-Aug			0.027		
25-Apr	0.012	0.04	0.018	11-Aug			0.019		
28-Apr		0.07	0.038	14-Aug			0.38		
2-May	0.007	0.2	0.12	16-Aug	0.01		0.045		
7-May			0.03	20-Aug	0.009		0.19		

Table B6S-13. Uranium Concentrations (mg L<sup>-1</sup>) Measured in Paddy's Run in 1963 a

(continued next page)





	Measured in Paddy's Run in 1963 <sup>a</sup> (continued)									
1963	Bridge	Willey	New	1963	Bridge	Willey	New			
Collection	North of	Road	Haven	Collection	North of	Road	Haven			
Date	Route 126	Bridge	Bridge	Date	<u>Route 126</u>	Bridge	Bridge			
23-Aug			0.12	4-Nov			0.053			
26-Aug			0.042	7-Nov	0.003		0.045			
29-Aug			0.35	10-Nov			0.02			
1-Sep	0.023		0.24	12-Nov	0.014		0.007			
4-Sep	0.006		0.054	16-Nov			0.008			
7-Sep			0.036	19-Nov			0.014			
10-Sep			0.051	22-Nov			0.15			
13-Sep	0.008		0.036	25-Nov	0.01		0.05			
16-Sep	0.004		0.037	28-Nov			0.02			
19-Sep			0.034	1-Dec	•		0.01			
22-Sep			0.031	4-Dec			0.017			
25-Sep	0.032		0.12	7-Dec	0.012		0.059			
28-Sep			0.048	10-Dec	0.016		0.028			
1-Oct	0.01		0.036	13-Dec			0.006			
4-Oct			0.033	16-Dec			0.025			
7-Oct			0.041	19-Dec			0.026			
10-Oct	0.012		0.044	22-Dec			0.039			
13-Oct			0.03	26-Dec			0.006			
16-Oct			0.041	29-Dec			0.029			
19-Oct			0.051	Avg (mg $L^{-1}$ )	0.01	1.02	0.06			
22-Oct			0.017	Stdev (mg $L^{-1}$ )	0.01	2.79	0.08			
25-Oct			0.02	$Avg(pCiL^{-1})$	41	690	6.72			
28-Oct	0.019		0.031	Stdev (pCi L <sup>-1</sup> )	56	1900	4.45			
31-Oct			0.02							
" From NLC	O analytical	data sheets	for 1963.	·····						

Table B6S-13. Uranium Concentrations (m	$g L^{-1}$
leasured in Paddy's Run in 1963 <sup>a</sup> (continu	(ed)

		mcasui	cumraut	iy s itun in	1004		
1964	Bridge	Willey	New	1964	Bridge	Willey	New
Collection	North of	Road	Haven	Collection	North of	Road.	Haven
Date	Route 126	Bridge	Bridge	Date	Route 126	Bridge	Bridge
4-Jan			0.1	14-Mar		0.43	nd <sup>b</sup>
7-Jan	0.002		0.028	14-Mar		0.05	
10-Jan			0.042	17-Mar	0.007	0.04	0.048
13-Jan			0.034	20-Mar			0.052
22-Jan			0.18	23-Mar		0.049	0.048
24-Jan			0.054	26-Mar		0.044	0.035
24-Jan			0.056	29-Mar	0.003		0.049
24-Jan			0.13	31-Mar	0.003		
25-Jan		8.8	0.17	1-Apr			0.021
28-Jan			0.033	4-Apr		0.9	0.13
31-Jan	0.018		0.17	7-Apr		0.042	0.042
3-Feb			0.054	10-Apr	0.012	0.08	0.0 <del>9</del>
4-Feb	0.005		0.061	13-Apr			0.14
6-Feb		6.6	0.13	16-Apr	0.001		0.12
6-Feb		7.8	0.036	19-Ap <del>r</del>			0.048
6-Feb		5.6		22-Apr		0.9	0.048
6-Feb		6		25-Apr	0.002	0.028	0.11
9-Feb		0.2	0.047	28-Ap <del>r</del>	0.001	0.11	0.033
12-Feb	0.024		0.2	1-May		0.028	0.041
15-Feb			0.11	4-May			0.057
15-Feb			0.15	7-May	0.003		0.055
15-Feb		6.8	0.04	10-May			0.059
12-Feb		6.7	0.14	13-May	0.005		0.051
15-Feb		6.5	0.061	16-May			0.057
18-Feb	0.022		0.024	19-May			0.049
21-Feb			0.01	22-May	0.005		0.11
25-Feb			0.34	25-May			0.039
28-Feb	0.008		0.029	28-May			0.048
2-Mar			0.22	29-May	0.014		0.037
2-Mar			0.24	3-Jun	0.001	17	0.013
2-Mar			0.44	6-Jun		4.3	0.044
4-Mar		5		9-Jun	0.002		0.04
5-Mar	0.007	3	1.06	12-Jun			0.035
8-Mar			0.14	15-Jun		0.67	0.057
11-Mar	0.01	0.086	0.061	18-Jun	0.005	0.2	0.065
13-Mar		0.28	0.046	21-Jun			0.11
13-Mar		0.65		24-Jun	0.004		0.048
14-Mar		0.051		27-Jun			0.038
14-Mar		0.22	0.48	30-Jun			0.021

Table B6S-14. Uranium Concentrations (mg  $L^{-1}$ ) Measured in Paddy's Run in 1964 <sup>a</sup>

(continued next page)

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Measured in Paddy's Run in 1964 a (continued)									
1964	Bridge	Willey	New	1964	Bridge	Willey	New		
Collection	North of	Road	Haven	Collection	North of	Road	Haven		
Date	Route 126	Bridge	Bridge	Date	Route 126	Bridge	Bridge		
3-Jul	0.009		0.017	13-Oct	0.006		0.012		
6-Jul	0.01		0.025	16-Oct			0.007		
9-Jul			0.09	19-Oct			0.011		
12-Jul			0.11	22-Oct	0.006		0.012		
15-Jul	0.012		0.029	25-Oct			0.011		
18-Jul			0.046	28-Oct			0.011		
21-Jul	0.002		0.026	27-Oct	0.005		0.005		
24-Jul			0.015	6-Nov	0.018		0.014		
5-Aug	0.034		0.061	9-Nov			0.005		
8-Aug			0.049	12-Nov	0.003		0.005		
12-Aug			0.024	15-Nov			0.006		
15-Aug	0.005		0.02	18-Nov			0.016		
18-Aug	0.001		0.017	21-Nov	0.013		0.01		
21-Aug			0.021	24-Nov	0.028		0.045		
24-Aug			0.019	27-Nov			0.019		
28-Aug			0.011	30-Nov			0.016		
31-Aug			0.022	3-Dec			0.001		
2-Sep	0.006		0.011	6-Dec			0.03		
6-Sep			0.017	8-Dec	0.008		0.002		
9-Sep	0.01		0.009	14-Dec	0.008	1.7			
12-Sep			0.006	12-Dec		0.62	0.52		
15-Sep			0.007	15-Dec			0.013		
15-Sep			0.015	18-Dec			0.01		
18-Sep			0.023	21-Dec	0.003		0.001		
21-Sep		2.4	0.11	24-Dec			0.031		
24-Sep	0.009		0.15	27-Dec		0.006	1		
27-Sep			0.031	30-Dec	0.001		0.005		
30-Sep	0.01		0.016	$Avg(mg L^{-1})$	0.01	2.8	0.07		
3-Oct			0.042	Stdev (mg $L^{-1}$ )	0.01	3.9	0.12		
7-Oct	0.005		0.01	Avg $(pCiL^{-1})$	5.8	1900	49		
10-Oct			0.005	Stdev (pCi L <sup>-1</sup> )	5.1	2600	81		
" From NLC	O analytical	data sheets	for 1964.	·······					

Table B6S-14. Uranium Concentrations (mg L<sup>-1</sup>)

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		Measu	ieu in rau	uy s Ruii III	1909		
1965	North of	Willey	New	1965	North of	Willey	New
Collection	Route 126	Road	Haven	Collection	Route 126	Road	Haven
Date	(upstream)	Bridge	Bridge	Date	(upstream)	Bridge	Bridge
3-Jan		0.043	0.09	30-Mar	0.001	0.082	0.21
6-Jan	0.008	0.052	0.04	2-Apr			0.064
9-Jan		0.06	0.03	5-Apr			0.031
12-Jan		0.041	0.021	8-Apr	0.012	0.28	0.27
15-Jan	0.002	0.047	0.02	11-Apr		0.045	0.052
18-Jan	0.002		0.001	14-Apr	0.01	0.019	0.023
21-Jan			0.029	17-Apr		0.19	0.15
24-Jan		0.066	0.035	20-Apr		0.04	0.033
27-Jan		0.63	0.03	23-Apr	0.013		0.036
30-Jan	0.001	0.058	0.013	26-Apr	0.007	0.024	0.0 <b>2</b>
2-Feb	0.005		0.009	29-Apr		0.025	0.034
5-Feb			0.012	2-May		0.036	0.08
8-Feb			0.07	5-May	0.011	0.03	0.038
11-Feb	0.032	0.038	0.12	8-May			0.057
14-Feb		0.025	0.027	11-May	0.003		0.028
17-Feb	0.002	0.038	0.09	14-May			0.025
18-Feb		0.063	0.034	17-May			0.028
19-Feb		0.028	0.022	20-May	0.004		0.11
20-Feb			0.036	23-May			0.026
21-Feb		0.06	0.013	26-May	0.006		0.031
22-Feb			0.013	29-May	0.026		
23-Feb			0.002	1-Jun			0.021
24-Feb			0.03	4-Jun	0.005		0.022
25-Feb	:	0.037	0.038	7-Jun	0.002		0.022
26-Feb	0.001	0.021	0.028	10-Jun			0.017
27-Feb		0.038	0.036	13-Jun			0.11
28-Feb		0.026	0.016	16-Jun			0.025
1-Mar		0.014	0.023	18-Jun	0.002	•	0.031
2-Mar	0.12	0.11	0.08	22-Jun	0.006		0.035
3-Mar		0.017	0.018	25-Jun			0.027
4-Mar		0.36	0.048	28-Jun			0.028
5-Mar		0.015	0.023	1-Jul	0.044		0.017
6-Mar		0.028	0.025	4-Jul			0.008
7-Mar		0.012	0.19	7-Jul			0.017
8-Mar		0.016	0.021	10-Jul	0.013		0.02
9-Mar		0.023	0.036	13-Jul	0.015		0.022
11-Mar		0.022	0.023	16-Jul			0.15
13-Mar	0.002	0.08	0.031	19-Jul		1.9	0.08
16-Mar	0.005			22-Jul			0.026
18-Mar		0.041	0.043	25-Jul			0.017
21-Mar			0.024	28-Jul	nd <sup><i>b</i></sup>		0.009
24-Mar		0.043	0.031	31-Jul			0.006
27-Mar	0.009	0.09	0.033	3-Aug			0.011

Table B6S-15. Uranium Concentrations (mg L<sup>-1</sup>) Measured in Paddy's Run in 1965 <sup>a</sup>

(continued next page)

Measured in Paddy's Run in 1965 <sup>a</sup> (continued)							
1965	North of	Willey	New	1965	North of	Willey	New
Collection	Route 126	Road _	Haven	Collection	Route 126	Road	Haven
Date	(upstream)	Bridge	Bridge	Date	(upstream)	Bridge	Bridge
6-Aug	< 0.001		0.003	24-Oct		0.024	0.026
9-Aug			0.034	27-Oct			0.56
12-Aug			0.011	30-Oct	0.009		0.019
15-Aug			0.017	31-Oct			0.023
18-Aug			0.022	3-Nov	0.002	0.066	0.017
20-Aug	0.009		0.24	6-Nov			0.021
24-Aug	0.01		0.019	9-Nov			0.016
28-Aug			0.034	12-Nov	0.003		0.009
30-Aug			0.022	15-Nov			0.11
2-Sep		2.2	0.37	18-Nov	0.004		0.021
5-Sep			0.032	21-Nov		•	0.011
7-Sep	0.004		0.053	24-Nov			0.007
10-Sep			0,013	27-Nov	0.004		0.017
13-Sep		0.37	0.056	30-Nov	0.006		0.014
16-Sep	0.004	0.17	0.37	3-Dec			0.032
19-Sep		0.07	0.029	6-Dec			0.09
21-Sep	0.012		0.016	9-Dec	0.004		0.024
25-Sep			0.006	12-Dec	0.003		0.01
28-Sep			0.018	15-Dec			0.013
30-Sep	0.005		0.019	22-Dec	0.027		0.023
3-Oct	0.005		0.028	25-Dec	0.004		0.046
6-Oct			0.019	28-Dec			0.013
9-Oct		0.87	0.46	31-Dec			0.027
12-Oct			0.07	Avg (mg $L^{-1}$ )	0.01	0.17	0.05
15-Oct	0.031		0.026	Stdev (mg $L^{-1}$ )	0.02	0.41	0.08
18-Oct	0.006		0.026	$Avg(pCiL^{-1})$	7.4	120	34
21-Oct		0.068	0.036	Stdev (pCi L <sup>-1</sup> )	13	280	56

Table B6S-15. Uranium Concentrations (mg L<sup>-1</sup>)

<sup>n</sup> From NLCO analytical data sheets for 1965.

<sup>h</sup> None detected

#### APPENDIX B -- REGIONAL ENVIRONMENTAL MONITORING

#### PART 7 – GROUNDWATER, CISTERNS, PONDS, AND POOLS

#### INTRODUCTION

Byrne et al. (1991) provides a brief history of the measurement of offsite uranium contamination in groundwater around the FMPC. Sampling by the State of Ohio in late 1981 indicated elevated levels of gross beta radioactivity in three wells south of the FMPC. Subsequent sampling by the FMPC showed that the activity was due to naturally-occurring  $^{40}$ K, and thus not associated with the FMPC. However, the FMPC sampling showed significantly elevated concentrations of uranium in other wells near the site. Because of the elevated uranium concentrations, the FMPC groundwater monitoring program was expanded in 1982 to include many private wells around the site.

The significant offsite uranium contamination in groundwater is south of the site, and is now called the "South Plume." Uranium concentrations in wells in the South Plume remain elevated. There are additional known areas of groundwater contamination on the FMPC site, but only the South Plume area extends outside the site boundary at this time (Byrne et al. 1991). Since this dose reconstruction project is concerned with past doses to people around the site, the groundwater contamination to be considered in this Project is limited to the South Plume. Figure B7-1 shows the estimated areal extent of the South Plume uranium contamination as of the end of 1991, as well as the locations of the private wells monitored (discussed later). The area of the South Plume has been estimated by the FMPC (Schwarzman 1992b), based on monitoring results from the private wells and from other monitoring wells, not shown in Figure B7-1.

In our report of Task 4 of this Project (Killough et al. 1993), 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. For this small group of exposed people, doses will be calculated later in this Project. For years when groundwater uranium monitoring data are available, the measured concentrations in private wells around the FMPC will be used directly in exposure assessments.

For years when groundwater monitoring data are not available, the exposure assessments are more difficult. In our previous source term report (Voillequé et al. 1991), we concluded that uranium contamination in the groundwater had not migrated outside the FMPC boundary by 1962. However, sometime before the end of 1981, uranium contamination had migrated offsite in the South Plume. Recent studies of the groundwater around the FMPC site (Dames and Moore 1985, and DOE 1990) have concluded that the primary source of the uranium contamination in the groundwater is uranium in waters released to the storm sewer outfall ditch and to Paddy's Run Creek. The soils in parts of the outfall ditch and Paddy's Run Creek are very permeable, and apparently allow contaminated water to move directly downward into the aquifer.



Figure B7-1. Approximate area of uranium contamination in the South Plume, as of the end of 1991, and locations of the private wells around the FMPC sampled in the FMPC routine monitoring program. Although well 26 is within the area of groundwater contamination, the uranium concentrations from this well are at background levels.

For years when groundwater monitoring data are not available, the source term work of Tasks 2 and 3 of this Project (in progress) will develop estimates of the uranium concentrations in wells in the South Plume, as a function of time. That work will use two major types of information: measured uranium concentrations in the private wells in the South Plume, and information about releases to the storm sewer outfall ditch and to Paddy's

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Run Creek (the source of the contamination). Estimates of the concentrations of uranium in water released to the storm sewer outfall ditch and to Paddy's Run Creek will be developed in the Tasks 2 and 3 work. Trends in the estimated discharges will be examined and compared to trends in the uranium concentrations in the South Plume, to help determine estimated concentrations in the plume for other time periods.

The historical monitoring data for uranium in private wells are important to the dose reconstruction work of this Project, because they will be used directly for exposure assessments for years when data are available, and will also be used to help estimate concentrations for years when no data are present. This Appendix thus includes compilations of the routine FMPC monitoring data for private wells, monitoring data for private wells obtained by other entities, and data on duplicate analyses of split water samples.

Many residences around the FMPC site have used cisterns. Cisterns are tanks used to store water for household uses, including drinking water. Water for cisterns is obtained from rainwater collection, through roof gutters, from springs or wells, or may be trucked in. Uranium released to the atmosphere from the FMPC may be deposited on rooftops and collected by cistern collection systems. This could then represent a pathway of radiation exposure to people living near the FMPC. The importance of this pathway for potential historical doses to nearby residents has not been fully evaluated. Results of measurements of uranium in cistern water have been compiled in this Appendix. However, since the ultimate uses of these data are not known, summaries are presented, rather than details of the results.

This Appendix also includes a small amount of data on concentrations of uranium in miscellaneous water sources.

In this Appendix, concentrations of uranium in water are presented using both mass units ( $\mu$ g L<sup>-1</sup>) and activity units (pCi L<sup>-1</sup>). Generally the units of the information source are used. To convert from mass to activity (or vice versa), the specific activity of natural uranium has been assumed to apply. The value of  $6.75 \times 10^{-7}$  Ci g<sup>-1</sup> (Rich et al. 1988) has been used.

#### MEASUREMENTS OF URANIUM IN PRIVATE WELLS AROUND THE FMPC

Since the discovery of the uranium contamination of the South Plume, groundwater samples have been taken from existing, private wells by the FMPC, the Ohio Department of Health (ODH), Dames and Moore, and the U.S. Geological Survey (USGS). The majority of the samples of private well water were obtained in the FMPC routine environmental monitoring program.

#### **FMPC Routine Monitoring of Private Wells**

The FMPC began its routine monitoring of private wells around the site in early 1982 (Byrne et al. 1991), although results were not reported in the annual environmental report for 1982 (Fleming and Ross 1983). Results of this routine program have been obtained for 1983-1990 (Fleming and Ross 1983, Fleming and Ross 1984, Facemire et al. 1985, Aas et al.



1986, WMCO 1987, WMCO 1988, WMCO 1989, Dugan et al. 1990, and Byrne et al. 1991). Since the wells sampled were not under the control of the FMPC, inclusion in the program was based on the well owner's request. Samples were generally taken on a monthly frequency, although a few of the wells were sampled less frequently. The annual environmental reports generally provide the minimum, maximum, and annual average uranium concentrations for each well in the monitoring program.

Figure B7-1 shows the locations of the private wells monitored and the estimated areal extent of the South Plume uranium contamination as of the end of 1991. The well locations were obtained from the annual environmental reports and from a detailed drawing obtained from the FMPC (Schwarzman 1992a). The annual average uranium concentrations for 1983-1990 are shown in Table B7-1, along with the long-term averages for each well for all years of monitoring.

The range of background concentrations of uranium in groundwater in the FMPC area has been estimated by the FMPC to be from 0.068 to 2.2 pCi  $L^{-1}$  (Byrne et al. 1991). From Table B7-1, it can be seen that most of the wells exhibit concentrations in this background range. However, three wells, numbers 12, 15, and 17, have significantly elevated concentrations of uranium. These three are all in the South Plume area. Well 26 is also within the areal extent of the South Plume, but its concentrations have been in the background range. Well 26 was installed in 1985 much deeper in the aquifer than the nearby well 12 (Dames and Moore 1985).

The averages for 1984 are actually geometric means (Facemire et al. 1985). Individual monthly results have not been obtained, but Facemire et al. (1985) provide minimum and maximum values for each well. Because the distributions of the individual values are unknown, we assume that the geometric mean can be used as the arithmetic mean. For many of the wells, the spread between the minimum and maximum concentrations is relatively small, so this assumption seems reasonable. For others the spread is greater, indicating a broader distribution for which the arithmetic and geometric means may be significantly different. For our purposes, the use of the geometric mean as an arithmetic mean is probably adequate.

In the mid-1980s investigations of the groundwater contamination around the FMPC were undertaken by Dames and Moore for the FMPC. The report of Task C of their work includes a compilation of the monthly uranium concentrations in wells 12, 15, and 17 from November 1981 through February 1985 (Dames and Moore 1985). The results compiled by Dames and Moore were for samples from the FMPC routine monitoring program. In the report, the designations OS-1, OS-2, and OS-3 are used for the wells that are now called 12, 15, and 17, respectively. The results are given in units mg  $L^{-1}$ . We converted the values to units of pCi  $L^{-1}$ , using the specific activity of natural uranium of  $6.75 \times 10^{-7}$  Ci g<sup>-1</sup>. The monthly sample results are shown in Table B7-2, along with calculated averages for 1982, 1983, and 1984.
Location	1983	1984 ª	1985	1986	1987	1988	1989	1990	Long-term average
1	0.20	0.34	0.30	0.25	0.2	0.15	0.17	0.14	0.22
2	0.20	0.27							0.24
3	0.27	0.34	0.23	0.29	0.2	0.13	0.17	0.14	0.22
4	1.2	1.29	1.08	1.09	1.4	1.2	1.4	1.4	1.3
5	1.4	1.42	1.31	1.09	1.4	1.2	1.5	1.2	1.3
6	1.6	1.29	1.37	1.08	1.2				1.3
7	0.95	0.95	0.95	0.95	1.1	1.1	1.1	1.0	1.0
8	0.54	0.54	0.53	0.55	0.66	0.59	0.60	0.54	0.57
9	0.81	0.81	0.99	0.81	0.97	0.93	1.0	0.88	0.90
10	1.1	0.34	0.38	0.39	0.47	0.49	0.52	0.47	0.52
11	0.81	0.68	0.81	0.91	1.0	0.99	1.1	1.3	0.95
12	140 <sup>b</sup>	165.19	140.00	147	201	170	170	130	160
13	0.41	0.41	0.44	0.41	0.5	0.42	0.37	0.54	0.44
14	0.81	0.74	0.73	0.68	0.89	0.82	0.88	1.0	0.82
15	290	219.35	204.27	193	201	190	190	190	210
16	0.61	0.41	0.67	0.47	0.48	0.43	0.52	0.47	0.51
17	39	36.29	31.15	31	40	38 °	27	30	34
18	0.34	0.34	0.29	0.30	0.4	0.38	0.34	0.27	0.33
1 <del>9</del>	0.14	0.20	0.20	0.21	0.2	0.25	0.12	0.14	0.18
20	0.14	0.20	0.16	0.15	0.2				0.17
21	0.27	0.27	0.29	0.29	0.3	0.27	0.25	0.27	0.28
22		0.74	0.76	0.61	0.80	0.81	0.79	0.61	0.73
23			0.55	0.56	0.56	0.55	0.58	0.61	0.57
24			0.32	0.36 <sup>d</sup>	0.4	0.46	0.40	0.34	0.38
25			0.27 <sup>d</sup>	0.28	0.5	0.19 <sup>d</sup>	0.27 <sup>d</sup>	0.27 <sup>d</sup>	0.30
26			0.24	0.30	0.39	0.17	0.14	0.14	0.23
27				0.38	0.69	0.51	0.50	0.34	0.48
28						0.58 <sup>d</sup>	0.57 <sup>d</sup>	0.51 <sup>d</sup>	0.55
29					1.4	1.3	1.1	1.1	1.2
30					0.4	0.39	0.38	0.34 <sup>d</sup>	0.38
31						0. <b>64</b> °			0.64
32							0.093	0.090	0.09
33								0.29 <sup>d</sup>	0.29
34							0.83	2.8	1.8
35							1.2	1.3	1.3
36								0.81 <sup>d</sup>	0.81
37								0.81 f	0.81
38								0.10 <sup>d</sup>	0.10

Table B7-1. Concentrations of Uranium in Private Well Water (pCi L <sup>-1</sup>	ı)
Around the FMPC; from FMPC Routine Monitoring	

<sup>a</sup> Results for 1984 were geometric means. We use them as if they were arithmetic means.

<sup>b</sup> It appears, from information discussed later, that this value may be erroneous.

<sup>c</sup> The pump for well 17 was inoperable for part of the year; only eight samples were obtained.

<sup>d</sup> Sampled on a quarterly basis only.

<sup>e</sup> Well 31 was withdrawn from the program; only six samples were obtained.

<sup>f</sup> Well 37 was scheduled for annual sampling only.

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Date	Well 12	Well 15	Well 17	Date	Well 12	Well 15	Well 17
Nov-81	130			Sep-83	180	260	46
Dec-81	110	220	36	Oct-83	180	260	42
Jan-82				Nov-83	170	270	36
Feb-82	160	350	34	Dec-83	160	250	28
Mar-82	160	280	47	Jan-84		240	36
Apr-82	190	300	48	Feb-84	160	240	30
May-82	210	300	51	Mar-84	170	240	35
Jun-82	170	300	53	Apr-84	180	230	34
Jul-82	180	300	67	May-84	180	210	34
Aug-82	160	320	31	Jun-84	180	210	40
Sep-82	160	320	41	Jul-84	170	200	37
Oct-82	190	330		Aug-84	160	1 <del>9</del> 0	32
Nov-82	150	340	36	Sep-84	170	200	
Dec-82	160	370	45	Oct-84	150	210	46
Jan-83	170	360	44	Nov-84	160	240	39
Feb-83	210	390	37	Dec-84	130	210	36
Mar-83	160	330	30	Jan-85	130	240	32
Apr-83	150	310	41	Feb-85	160	200	28
May-83	170	280	38	] .			
Jun-83	190	280	45	mean 1982	170	320	45
Jul-83	190	250	38	mean 1983	180	290	39
Aug-83	190	250	40	mean 1984	170	220	36

Table B7-2. Monthly Uranium Concentration (pCi L<sup>-1</sup>) in the Three Contaminated Private Wells, November 1981 through February 1985 °

The average concentration for well 12 in 1983, from the monthly results in Table B7-2, is 180 pCi  $L^{-1}$ , which is significantly different from the average of 140 pCi  $L^{-1}$  reported in the environmental report for 1983 and shown in Table B7-1. For the other averages, the results from the environmental reports (Table B7-1) agree with the averages computed from monthly results (Table B7-2). Thus, it appears that the concentration listed in the environmental report (Fleming and Ross 1984) for well 12 for 1983 may be erroneous. Until additional information is located, we assume this to be the case, and assume that the correct average is 180 pCi  $L^{-1}$ , based on the monthly results.

Additional results for a limited number of private well samples collected in 1982 are provided in an undated FMPC memorandum (Thiesen circa 1983). Table B7-3 shows the average uranium concentrations for the period March-August 1982, from this memorandum. The memo identified the wells by the initials used by the FMPC at that time. However, it was possible to determine the well number that has more recently been used by the FMPC, based on the well locations shown in a drawing attached to the memo (Thiesen circa 1983), well identification information from the FMPC (Kraps 1992), and a drawing in the 1983 FMPC environmental report (Fleming and Ross 1984). For identification here, only the well numbers are used, and these are shown in Table B7-3. The concentrations were

<sup>&</sup>lt;sup>a</sup> Results of FMPC monitoring, compiled in Dames and Moore (1985).

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given in units mg  $L^{-1}$ , but are converted, in Table B7-3, to  $\mu g L^{-1}$  and pCi  $L^{-1}$ , for convenient comparisons with other data. Locations of the wells are shown in Figure B7-1.

	Uranium concentration			
FMPC well number <sup>a</sup>	(µg L <sup>-1</sup> )	(pCi L <sup>-1</sup> )		
16	1.1	0.74		
14	1.6	1.1		
10	. 1.0	0.68		
18	0.9	0.6		
22	1.7	1.1		
15	440	300		
12	260	180		
20	0.5	0.3		
11	1.8	1.2		
17	73	49		
21	0.8	0.5		
19	0.5	0.3		
13	1.2	0.81		

Table B7–3. Average Uranium Concentration in Offsite Wells, for March-August, 1982

<sup>a</sup> Determined in this present work, based on a drawing of locations in Thiesen (circa 1983), and on other information.

As for the previously discussed data sets, the average concentrations for March-August 1982 show significant uranium contamination only at wells 12, 15, and 17, which are south of the site in the South Plume area. The concentrations for these wells agree with the average of the monthly concentrations shown in Table B7-2. The concentrations for the other (uncontaminated) wells are generally similar to annual averages for the same wells for different years, shown in Table B7-1.

Figures B7-2 and B7-3 are plots, against time, of the monthly concentrations and annual average concentrations for wells 12, 15, and 17. In Figure B7-3, the averages from the annual environmental reports (from Table B7-1) are supplemented with the calculated averages for 1983, based on the monthly results (from Table B7-2).

The plots in Figures B7-2 and B7-3 indicate no significant trends in the concentrations for wells 12 and 17. However, it appears that concentrations in well 15 gradually increased in 1982, and then gradually decreased in 1983 and the first half of 1984.

Concentrations in the other wells are evaluated to estimate typical background concentrations of uranium in well water around the FMPC. As mentioned earlier, the FMPC has estimated that background concentrations around the site range from 0.068 to 2.2 pCi  $L^{-1}$  (Byrne et al. 1991). From the average concentrations, in Table B7-1, it appears that all of the wells, except for 12, 15, and 17, are within this range and are not significantly different from each other. However, two wells deserve a closer look. In 1990, well 34 had an average concentration of 2.8 pCi  $L^{-1}$ , which is higher than for other wells (except 12, 15, and



Figure B7-2. Monthly measurements of uranium concentration in well water for the three contaminated private wells, for November 1981 through February 1985.





17). This concentration was a large increase over the value of 0.83 pCi  $L^{-1}$  for 1989, and may indicate a significant change. Because the individual monthly results for well 34 have not been obtained, it is difficult to evaluate the significance of any trend. This well does not appear close to the known extent of the South Plume, but it is in the general direction of the Plume's movement, and is close to Paddy's Run Creek, which was a potential source of uranium infiltration to the aquifer. For now, it seems reasonable to assume that well 34 is potentially contaminated, and thus should not be considered representative of background.

Well 11 appears to have a trend of increasing concentrations with time (Table B7-1), although the maximum annual average concentration does not seem significantly elevated.



However, this well is located very close to the estimated boundary of the South Plume, so increasing concentrations are not unexpected. To investigate the significance of the upward trend in concentrations, the Mann-Kendall nonparametric test for trend was applied (per Gilbert 1987). The test results indicate that the probability is less than 0.3% that the test statistic would have been observed if no trend were present. We thus conclude that a significant upward trend exists at well 11. Because this well is close to the estimated area of the South Plume, it is reasonable to assume that it is contaminated and is not representative of background.

If wells 11, 12, 15, 17, and 34 are excluded, the long-term (up to eight years) average concentrations for the other wells range from 0.09 to 1.3 pCi  $L^{-1}$ , with a grand average of 0.6 pCi  $L^{-1}$  for all of these other wells. It is thus concluded that a reasonable estimate of the long-term average, background concentration of uranium in well water (averaged over many locations) around the FMPC is 0.6 pCi  $L^{-1}$  and a reasonable estimate of the range of long-term average, background concentrations for individual wells is 0.09 to 1.3 pCi  $L^{-1}$ .

### **Other Monitoring of Private Wells**

Although the routine monitoring data reported above is the most comprehensive data set for uranium in private well water, additional data have also been obtained. These data are from studies by the U.S. Geological Survey (USGS), Dames and Moore, and the Ohio Department of Health (ODH).

USGS study. The USGS study (Sedam 1984) was undertaken in 1982 for the U.S. Department of Energy (DOE), to try to determine the source of the elevated uranium concentrations in wells south of the FMPC site (what is now known as the South Plume area). The USGS study included sampling some of the wells that had been sampled by the FMPC. The USGS samples were taken in August 1982, and were compared by Sedam (1984) to FMPC results from December 1981 samples. Table B7-4 shows this comparison of USGS and FMPC results. The USGS identified the wells by its own well number and by the initials used by the FMPC at that time. However, it was possible to determine the well number that has more recently been used by the FMPC, based on the well locations shown in a drawing in Sedam (1984), well identification information from the FMPC (Kraps 1992), and a drawing in the 1983 FMPC environmental report (Fleming and Ross 1984). For identification here, only the well numbers are used, and these are shown in Table B7-4. For wells 12, 15, and 17, data from the FMPC program for August 1982 are available (see Table B7-2), and these are also included in Table B7-4 for comparison. These last data have been converted from units of pCi  $L^{-1}$  to units  $\mu g L^{-1}$ , using the specific activity of natural uranium of  $6.75 \times 10^{-7}$  Ci g<sup>-1</sup>.

Based on the data in Table B7–4, the results obtained by the USGS are generally similar to those obtained by the FMPC. However, for wells 22, 14, and 11 the FMPC results appear to be significantly higher than the USGS results. These results can also be compared to the FMPC routine results compiled in Table B7–1. With conversion of units to  $\mu$ g L<sup>-1</sup>, the long-term average concentrations in wells 22, 14, and 11, based on the FMPC routine monitoring, are 1.1, 1.2, and 1.4  $\mu$ g L<sup>-1</sup> (from Table B7–1). These values agree much better with the USGS results.

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Well iden	tification	Uranium	concentration	(µg L <sup>-1</sup> )
USGS well	FMPC well	December 1981	August 1982	August 1982
number <sup>a</sup>	number <sup>b</sup>	FMPC <sup>a</sup>	FMPC <sup>c</sup>	USGS <sup>a</sup>
H-101 H-103 H-106 H-107 H-108 H-109 H-110 H-111 H-1116 H-117	10 22 14 13 12 16 18 15 11 20	<3.0 6.0 6.0 <3.0 190 <3.0 <3.0 320 6.0 <3.0	240 470	<2.0 1.4 1.5 0.7 250 d <0.4 430 1.5 <0.4
H-118	19	<3.0	46	<0.4
H-119	21	<3.0		<0.5
H-121	17	54		46

Table B7-4. Comparison of Uranium Concentrations in Private Wells Around the FMPC Determined by the USGS and the FMPC

<sup>a</sup> From the USGS report (Sedam 1984).

<sup>b</sup> Determined in present work, based on location drawing in Sedam (1984).

<sup>c</sup> From Table B7–2, this Appendix, with units conversion.

<sup>d</sup> Sample not used, due to excessive bleach in water supply (Sedam 1984).

Sedam (1984) concludes that wells containing elevated concentrations of uranium extended in a line 2000 feet south from the southern boundary of the FMPC. This location of contaminated groundwater agrees with the current estimates of the areal extent of the South Plume (see Figure B7-1). Sedam also concludes that the plume of higher uranium concentrations is inconsistent with groundwater flow patterns and conjectures that it is possibly due to storm overflow of materials from the FMPC into Paddy's Run, with infiltration through the stream bottom. This path is now the generally accepted source of the elevated uranium concentrations in the South Plume area.

Dames and Moore sampling. As part of the groundwater investigations performed by Dames and Moore, many monitoring wells and some private wells were sampled and analyzed for uranium concentration (Dames and Moore 1985). The field work was performed from December 1984 to March 1985. In the report, the designations OS-1, OS-2, and OS-3 are used for the wells that are now called 12, 15, and 17. Samples were split with the FMPC, so two results are available for each sample. The results for wells 12, 15, and 17 are shown in Table B7-5. For these wells, data from the routine FMPC monitoring are available (see Table B7-2), and the average from December 1984 through February 1985 (March was not available) is also included in Table B7-5. These last data have been converted from units of pCi L<sup>-1</sup> to units  $\mu$ g L<sup>-1</sup>, using the specific activity of natural uranium of 6.75 × 10<sup>-7</sup> Ci g<sup>-1</sup>. The results from the Dames and Moore sampling agree relatively well with averages from the FMPC routine sampling.

Well iden	tification	Uraniu	1m concentration	(µg L <sup>-1</sup> )
FMPC well number	Other well designation	D & M sample and analysis <sup>a</sup>	D & M, FMPC analysis <sup>a</sup>	FMPC routine monitoring <sup>b</sup>
12	OS-1	300	227	210
15	OS-2	350	302	320
17	OS-3	43	41	59

 Table B7-5. Comparison of Uranium Concentrations in Private Wells

 12. 15. and 17 Determined by Dames and Moore and the FMPC

<sup>a</sup> From the Dames and Moore (1985) report. Abbreviated "D & M" here.

<sup>b</sup> Average of December 1984, January 1985, and February 1985 results, from Table B7-2, this Appendix, with units conversion.

Samples collected in late 1984–1985. Between December 1984 and August 1985, water samples were collected for uranium content analysis. These samples are described in draft documents (Spenceley circa 1985a and Spenceley circa 1985b). Most were collected during December 1984 and January 1985. Well water samples were collected from wells with Hamilton and Harrison, Ohio, addresses, and from wells in other towns. Unfortunately, the data sheets were handwritten, and the copies were not always clear. The data presented here represent only material for which the location and result could be read—about 10% of the entries were illegible.

Uranium in all well water samples collected at Harrison and Hamilton have a mean value of  $1.2 \ \mu g \ L^{-1}$ , a median of  $0.9 \ \mu g \ L^{-1}$ , and a standard deviation of 2.6  $\ \mu g \ L^{-1}$ . However, there are two "outliers" collected on Willey Road, just south of the FMPC, which yielded results of 18.9  $\ \mu g \ L^{-1}$  and 189  $\ \mu g \ L^{-1}$  (this latter result was the average of two samples). These locations are believed to be the same as well 12, which has shown elevated concentrations, similar to the higher result, since the FMPC monitoring began (see Tables B7-1 and B7-2). Samples collected at two other locations, determined here to be wells 15 and 17, also gave results far above the mean, but similar to concentrations given in Table B7-1. If these elevated samples (all from Hamilton) are not included in calculations of the statistics, the mean uranium concentration is  $0.9 \ \mu g \ L^{-1}$ , the median is  $0.9 \ \mu g \ L^{-1}$ , and the standard deviation is  $0.5 \ \mu g \ L^{-1}$ . Thus, in general the concentrations for wells near the FMPC are similar to those from the FMPC routine monitoring.

Well water collected at Harrison yielded a mean value of 0.9  $\mu$ g L<sup>-1</sup>, a median of 0.8  $\mu$ g L<sup>-1</sup>, and a standard deviation of 0.5  $\mu$ g L<sup>-1</sup>. These are the same statistics as for the group as a whole (without the outliers). For Hamilton, the values are the same without the outliers as for the group as a whole; with them, the mean, median, and standard deviation are 1.6  $\mu$ g L<sup>-1</sup>, 1.0  $\mu$ g L<sup>-1</sup>, and 3.3  $\mu$ g L<sup>-1</sup>, respectively.

Hence, without the high samples at Hamilton, the statistics for the two towns are the same. Indeed, a Student's T-test with or without the outliers indicates that the means are not significantly different.

**Ohio Department of Health sampling 1985–1988.** In 1985, the Ohio Department of Health (ODH) initiated environmental sampling programs around the FMPC and the Portsmouth Gaseous Diffusion Plant (PGDP), to respond to community concerns about

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contaminated groundwater around the FMPC and unanticipated releases of uranium from the FMPC to the atmosphere. The program was terminated in January 1988 (though ODH continued some monitoring around the FMPC), and results are given in an ODH report (Steva 1988). The primary purpose of the sampling program was to sample drinking water used by residents living close to the two DOE facilities, although measurements were also made for uranium in soil, exposure rate, outdoor radon, radon in homes, radon in water, and uranium in surface waters. Many of the sampling locations were based on requests from residents living around the facilities. Most of the samples were obtained in 1985 and 1986, although precise dates are not given for individual samples.

For private wells around the FMPC, the ODH sampled about 245 wells (Steva 1988). Of these, only three contained uranium concentrations above background. The ODH report (Steva 1988) does not provide any cross-reference of its sample log numbers to the well numbers used by the FMPC. However, the locations of the three contaminated wells (log numbers 107, 289, and 49) are shown on drawings, and appear to be the same as wells 12, 15, and 17 (FMPC designation), respectively, that are routinely sampled by the FMPC. The concentrations of uranium measured in water samples from these three wells were 150 pCi L<sup>-1</sup> for log 107 (average of two samples), 250 pCi L<sup>-1</sup> for log 289, and 27 pCi L<sup>-1</sup> for log 49 (Steva 1988). For comparison, the average uranium concentrations measured by the FMPC routine monitoring for 1985 and 1986 were 140 pCi L<sup>-1</sup> for well 12, 200 pCi L<sup>-1</sup> for well 15, and 31 pCi L<sup>-1</sup> for well 17 (see Table B7-1). Given that the FMPC averages are based on many more samples over two years, while the ODH averages are based on one to three samples, the concentrations measured by ODH agree well with the FMPC results.

#### FMPC/State of Ohio Split Sample Analyses

Analyses of split well water samples analyzed for uranium can provide information about the quality of the results of the FMPC analyses. Well water samples have been split with the Ohio Department of Health (ODH), and analyzed in duplicate, with an ongoing program established in 1987. Some results of these split sample analyses have been obtained. The ultimate uses of these data for this dose reconstruction work have not been determined, so at this point we mostly summarize the results. Additional evaluations of the data may be performed later.

In 1985, nineteen split well water samples underwent duplicate analysis by NLO (FMPC) and the ODH (Anonymous, circa 1986). These data were analyzed to test the agreement between the analytical laboratories. Summary statistics for the duplicate analyses are shown in Table B7-6. A Student's T-test (two tailed) for paired samples indicated that the sample means were not significantly different, indicating good agreement between the duplicate analyses.

As part of the ODH environmental sampling program around the FMPC in 1985-1988 (see also page B7-11), every fifth water sample was split with the FMPC (Steva 1988). For this period, 55 samples were analyzed by both ODH and the FMPC for uranium concentration, of which 48 were private well samples and 8 were cistern water samples. Steva (1988) determined that at the 99% confidence level, all of the paired results showed no

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of Uranium in Well Water ( $\mu$ g L <sup>-1</sup> )					
Statistic	NLO (FMPC)	ODH			
Mean	2.7	2.1			
Median	0.5	0.5			
Standard deviation	9.5	6.8			

Table B7-6. Results of 1985 Split Analyses
of Uranium in Well Water ( $\mu g L^{-1}$ )

significant difference between the ODH result and the FMPC result. Steva provides analytical uncertainties for the ODH sample results only.

In 1987, the FMPC and ODH established an ongoing program of routine split sample collection (WMCO 1988). This program primarily involved water samples, both from surface waters and from private wells, but also included sediment and milk samples. Results of the split sample analyses for 1987, 1988, and 1989 are reported in the environmental reports of 1987 (WMCO 1988), 1989 (Dugan et al. 1990), and 1990 (Byrne et al. 1991).

For 1987, 31 water samples were split between FMPC and ODH for uranium analyses, of which 14 were surface water samples and 17 were private well water samples. The FMPC (WMCO 1988) concludes that the results were "...very similar with no significant discrepancies." The results provided (WMCO 1988) only included average concentrations for each location, and analytical uncertainties were not given.

For 1988, 51 surface water samples and 59 private well water samples were split between FMPC and ODH (Dugan et al. 1990). For 1989, 49 surface water and 57 private well water samples were split (Byrne et al. 1991). For these two years, the FMPC evaluated the split analyses by first calculating a range for each individual FMPC and ODH result, by adding and subtracting the "±" uncertainty term, provided with each result by the respective analytical laboratories. If the resultant FMPC and ODH ranges for an analysis overlap, the FMPC considered the results to be equivalent. The FMPC determined that 92% and 94.3% of the uranium in water (surface and well) analyses were equivalent in 1988 and 1989, respectively (Dugan et al. 1990 and Byrne et al. 1991). Unfortunately, the precise meaning of the "±" uncertainties reported is not provided by Dugan et al. (1990) or by Byrne et al. (1991).

For the surface and well water results performed by the FMPC in 1988 and 1989, that were reported for split sample comparisons, essentially all of the "±" uncertainty results are 34% to 35% of the reported result (Dugan et al. 1990 and Byrne et al. 1991). In one case, for surface sampling location W7 in January 1989, the reported uncertainty was 2.1 times the reported result (Byrne et al. 1991). This may have been an error, as the reported result is less than the minimum value reported for location W7 in 1989 in the report for 1989 (Dugan et al. 1990). If it is determined that these uncertainty values may be useful for further work, such as on the estimation of concentrations of uranium in groundwater in years before 1981 (in the report of Tasks 2/3 of this Project, in preparation), more information about these uncertainties will be sought.

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### MEASUREMENTS OF URANIUM IN CISTERN AND POND WATERS

Results of uranium in cistern water analyses have been obtained for a small number of samples. In addition, a few results of uranium in pond and miscellaneous waters have been obtained. Uses of these data for the dose reconstruction have not been determined. Summaries of the data are provided in this section.

### Uranium in Cistern Water

Results of uranium in cistern water samples taken in Cincinnati, Cleves, Harrison, Hamilton, and Miamitown in 1984 and 1985 are reported in a draft memorandum (Spenceley circa 1985b). These results are summarized in Table B7–7 below. Although the mean and median values are different between Cincinnati samples and samples collected elsewhere, the difference in the sample means is not statistically significant.

		-F (+-8	
Statistic	All samples	Cincinnati samples	Other samples
Number	11	5	6
Mean	1.1	0.4	1.8
Median	0.4	0.4	1.0
Standard deviation	1.7	0.8	2.4

Table B7-7. Concentrations of Uranium in Cistern Water Samples (µg L<sup>-1</sup>)

As part of the ODH environmental sampling program around the FMPC in 1985–1988 (see also page B7–11), water from cisterns in the FMPC area was also sampled (Steva 1988). A total of 54 cisterns were sampled, with the water sample analyzed for uranium. For 53 cisterns, the uranium concentrations ranged up to 1.2 pCi L<sup>-1</sup>. The highest uranium concentration found was an average of 25 pCi L<sup>-1</sup> (two samples of the same cistern), for a cistern located just north of the FMPC, on State Route 126. This cistern collected rainwater from roof gutters. The cistern had been disconnected from the collection system for two years prior to sampling, so the water had been undisturbed for about two years. Immediately prior to sampling, the owner agitated the cistern water. The source of the elevated uranium concentration in this cistern is not absolutely known, but it seems likely that airborne uranium released from the FMPC was deposited on the rooftop. These data may be evaluated in more detail, if they are determined to be useful to the dose reconstruction effort.

### Uranium in Pond and Miscellaneous Waters

Results of uranium concentrations in water collected from miscellaneous ponds, pools, and drinking water are reported in a draft memorandum (Spenceley circa 1985b). These results are summarized in Table B7–8. The results are generally similar to background concentrations of uranium in groundwater, with the exception of one slightly elevated concentration, from a sample collected at a pond in Hamilton.

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Other wat	er Sources
Location	Concentration ( $\mu g L^{-1}$ )
Pond, Hamilton	3.8
Pool, Cincinnati	0.4
Pool, Harrison	0.7
City Water, Cincinnati	0.9
Pond, Hamilton	0.6

# Table B7–8. Uranium Concentrations in Other Water Sources

### CONCLUSIONS

Annual average concentrations of uranium in private wells around the FMPC, from the FMPC routine monitoring program, have been compiled. These data show that uranium concentrations are significantly elevated above background in three wells, 12, 15, and 17, which are located within the South Plume area. Concentrations in wells 12 and 17 show no significant trends, but concentrations in well 15 gradually increased in 1982 and then gradually decreased in 1983 and the first half of 1984. Detailed data, showing individual results (rather than just annual averages), are apparently available from the FMPC, and should be forthcoming. As appropriate, these detailed monitoring results will be discussed in the report of Tasks 2 and 3 of this Project.

Data from the uncontaminated private wells indicate that long-term average background concentrations of uranium in groundwater in the FMPC area range from 0.09 to  $1.3 \text{ pCi L}^{-1}$ .

Monitoring of private wells around the FMPC for uranium has also been performed by entities other than the FMPC. Though these data are much less comprehensive, the results have also been compiled. Results of these other sampling programs corroborate the findings based on the FMPC routine monitoring.

Results from duplicate analyses of water samples split between the FMPC and the Ohio Department of Health (ODH) have been summarized. These data show generally good agreement between FMPC and ODH results. Uncertainties reported with the FMPC data were 34% to 35% of the reported results for essentially all reported analyses, including those at higher concentrations. The results of split analyses and uncertainties of FMPC concentrations may be used for further work in this Project, in which case additional evaluations of the data may be performed.

Uranium concentrations in cistern water, pond water, and miscellaneous water sources have also been summarized. Concentrations in cisterns were generally in the range of background groundwater concentrations. One cistern, located just north of the FMPC, showed significantly higher uranium concentrations, that may be due to deposition of airborne uranium released from the FMPC.

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### APPENDIX C – PARTICLE SIZE OF AIRBORNE EFFLUENTS

### PART 1 — NKES STUDY-METHODOLOGY QA

### INTRODUCTION

The scope of Part 1 of Appendix C is a quality assurance (QA) of particle size determinations for releases from stacks at the Feed Material Production Center. The 1985 study described in "A Study Of The Particle Size Distribution Of The Stack Emissions At Fernald" prepared by the Northern Kentucky Environmental Services. October 31, 1985 (hereafter referred to as NKES) was reviewed in order to check the validity of the results and to evaluate uncertainties in the particle size determinations.

In Part 1 of this Appendix. the methodology employed by the Northern Kentucky Environmental Services is compared to that recommended in the "operating manual" for the Andersen Mark III stack sampler (Andersen 1984) This manual will be referred to in the present report as the "ANDERSEN manual." To investigate the raw data and calculations from the NKES study, raw data from about 10 percent of randomly selected sampling runs were analyzed and compared with the reported results. The conclusions gleaned from these recalculations are presented herein. Additionally, other information of importance to the Fernald dose reconstruction project presented in the NKES report is noted and comment is made on further particle size work required for environmental modeling.

The quality assurance activity of the RAC project at Fernald is not intended as a critique of previous work. Its goal is to evaluate how previous work can be employed to determine radiation doses to residents around Fernald and to estimate the uncertainties that accompany results of the prior studies.

### METHODOLOGY

The evaluation of methodology is based on a reading and interpretation of the descriptive material in the NKES report. No attempt was made to contact the Northern Kentucky Environmental Services, other than to obtain the raw data. nor to interview individuals responsible for the NKES work. The procedures employed by NKES as reported in their reference document were compared to those described in the ANDERSEN manual.

### **Operation of the Mark III Sampler**

The Andersen Mark III sampler used in the NKES study is an in-stack multistage impactor designed for isokinetic sampling. (Under isokinetic conditions, the velocity of the gas through the sampler inlet equals the velocity of the gas in the stack. Thus, the streamlines of the gas are not disturbed by the sampling orifice, and representative particle size sampling occurs.) The device is calibrated with unit density (1 g cm<sup>-3</sup>) spherical particles so that the aerodynamically equivalent sized particles collected on each stage are always identical for any given set of sampling conditions. (Aerodynamic diameter is defined

as the diameter of a spherical particle with a density of 1 g cm<sup>-3</sup> that has the same gravitational settling and other aerodynamic properties as the particles in question, in this case uranium particles.)

The setup and operation of the sampler is as follows:

- Stack gases enter the inlet nozzle at or near isokinetic conditions. The gases then flow through the cascade impactor stages, backup filter and the inner liner of the electrically heated sampling assembly.
- A pitot tube is located on the probe sheath to measure the stack differential pressure  $(\Delta P)$  in the area of the sampling assembly nozzle. Using the  $\Delta P$ , a nomograph provided with the instrument can be used to calculate the desired nozzle flow rate, expressed as a differential pressure across a calibrated orifice.
- The stack gases continue their flow from the probe liner through a cyclone trap (used if the impactor is not mounted on the sampling assembly) and a glass fiber filter contained in the hot side of the sampling unit. Impingers in an ice bath cool the gases before they enter the umbilical cord.
- The umbilical cord carries the filtered. cooled stack gases from the sampling site to the control unit.
- The control unit utilizes a vacuum pump to draw the stack gases through the sampling train. A dry gas meter records the volume of gas sampled. Sampling rates are controlled by a coarse and fine value adjustment as indicated by a calibrated orifice and Magnehelic or inclined tube differential pressure gauges.

The Mark III sampler comes in a six or eight stage mode. The data sheets in the NKES report indicate that an eight stage model was employed. A preseparator (the cyclone trap) may be used to remove larger particles when conditions warrant. It does not appear to have been used in the NKES work.

The ANDERSEN manual provides calibration curves which plot "sampler flow" versus "cut point" (in terms of aerodynamic diameter of spherical particles) for the cyclonic preseparator. a preimpactor and each of the impactor stages. The impactor stages were followed by a backup filter in order to insure total particulate collection. Gases were not sampled during the NKES study and hence the impingers are not mentioned further in this report.

### Comparison of Andersen Manual Recommended Methodology with NKES Study

Sampling Locations. Sampling location within the duct is important in that particle segregation can occur due to flow of the gas-particulate material through a horizontal duct or following a bend in a duct. The most important aspect of selection of sampling location is to select sampling points whose effluents are representative of the entire stack. Dust loading





across a transverse of the flow can be employed as an indicator of the representativeness of the size distribution.

<u>The ANDERSEN Manual</u>. The ANDERSEN manual suggests consideration be given to flow orientation, vertical or horizontal, and nearness to bends, obstructions, and changes in cross-section. Dust loading tests, fully traversing the flue, are suggested prior to any impactor tests. This is to establish a concentration profile and avoid unrepresentative impactor runs.

The NKES Study. The NKES report indicates that sampling points were selected in keeping with EPA Method 1 and 2 (EPA 1990). These are based on transverse measurement of  $\Delta P$  as an indicator of flow at the measurement point in the dust. The NKES report states the  $\Delta P$  did not vary more than 10 percent in the transverse. The data sheets do indicate the performance of transverse tests to measure  $\Delta P$ . Adequate attention appears to have been given in the NKES work to ensure the representativeness of samples across a sampling transverse and positioning of the sample. In a few cases, a diagram indicated the configuration of the duct if other than circular.

**Dust Loading**. Dust loading is an important parameter in insuring proper operation of the impactor and hence the representativeness of the particle sizes on each impactor stage.

<u>The ANDERSEN Manual.</u> The ANDERSEN manual suggests that "An attempt should be made to sample as long as possible without overloading the plates. With extremely dry. non-adhesive particles. a maximum recommended catch weight on any one plate is 10 mg. Beyond this limit. particle reentrainment begins, resulting in a shifting of the size distribution toward the smaller end. Experience has shown that this upper limit may be exceeded by a factor of two or three without adversely affecting the results with most stack effluents because most exhibit some degree of adhesion. Visual observation of the collected sample on each stage is the preferred method for determining if over sampling has occurred."

<u>The NKES Study</u>. A check of all the data sheets indicates that the duct loading on any single plate exceeded 10 mg in about three cases (some data sheets did not duplicate well, and the exact number is in question) out of the 256 stages weighed in the entire study (32 sampling runs). Two of the above dust loading weights were less than 12 mg, the other was less than 34 mg.

Sampling times for inlet samples were 20 to 30 min and for emission (outlet) samples were an average of 30 hr.

The NKES study does not appear to have violated the intent of the ANDERSEN manual regarding dust loading.

### Conclusions on Sampling Methodology in the NKES Study.

The sampling methods employed in the NKES study appear to be within the context of the directions contained in the ANDERSEN Operating Manual. The techniques employed should not have added to the existing uncertainties inherent in the sampling methodology.

### QA CHECK OF RAW DATA AND CALCULATIONS

About ten percent of the hand written analytical laboratory sheets were inspected in detail to evaluate the raw input data and calculations. English and International (SI) units denoted the pressure and temperature inputs in Pascals, inches, cm or mm of water or mercury for pressure; and °C, °F, °K and °R for temperature. Except in a single case, conversions were rounded off and performed correctly. Stack velocity and flow were expressed in units of feet or meters. No explanation was given for the diversity of units employed.

An error was noted in copying total dust loading in one case. There was a single instance where the volume of air sampled was off by a factor of two.

With the exceptions noted above, the discrepancies were generally small. For the most part, the errors led to erroneous emissions concentrations, and did not impact directly on the particle size determinations.

### **RESULTS OF NKES STUDY**

For the purposes of this evaluation, "results" are considered as the outcome of the NKES work, as distinguished from raw data or the calculations noted above.

Labeling of Results. Several clarifications would have been useful in the NKES report. For example, a table of total dust MMAD (mass median aerodynamic diameter) is labeled as AMAD Total Dust. The term AMAD usually refers to the "activity median aerodynamic diameter" and infers that the results came about through counting of radioactivity. However, the material analyzed was total dust and uranium, determined by weighing and fluorimetry respectively. There is, however, no reason to suspect that the uranium activity is not proportional to the uranium mass. It may be deduced that the AMAD in the NKES stood for an "average" median aerodynamic diameter, but no assurance can be attributed to this assumption.

Confusion sometimes arises regarding the relationship with the familiar activity median aerodynamic diameter (AMAD) used by the International Commission on Radiological Protection (ICRP) in connection with its respiratory models. The ICRP AMAD refers to the median of a distribution, with respect to radioactivity, of (equivalent) aerodynamic particle diameters: moreover, the ICRP Task Group on Lung Dynamics restricted consideration to distributions with geometric standard deviation less than 4.5 (ICRP 1979). Most of the distributions of aerodynamic diameter with respect to uranium mass measured by NKES fail to be lognormal. Part 2 of this Appendix and Appendix D of the Task 4 report (Killough et al. 1993) discuss particle size distributions for releases of uranium as employed by this study.

**Resultant Particle Size Values.** Data from the NKES report have been extracted and organized to reflect inlet and emission (outlet) particle sizes for each stack studied. These appear in Table C1-1 below. The "sigma" term is not defined. The magnitude of the values suggests they are Geometric Standard Deviations (GSD), but the use of the " $\pm$ " would be inappropriate. In addition, since many of the distributions are not lognormal, the terminology is not correctly employed, if that is what was intended.



Although generally, there was no check of the tabulated data in the reference report against the data sheets, except for those noted in Part 2 of this Appendix, the MMAD value of 138  $\mu$ m for Total Dust Run #9, stack #5–251 is not found in the data sheets. The value for uranium particle size in the data sheets for this run is 0.48  $\mu$ m and given in the report as 0.45 $\mu$ m (a minor discrepancy).

By inspection of the results (Table C1-1), the Total Dust MMAD and Uranium AMADs are similar. The data for #4-G4-2 and #4-G4-7 (total dust and uranium). #5-251 (uranium). #5-253 (total dust) and #8-G43-27 (uranium) appear to be reversed: that is, the particle sizes for the outlets (emissions side) is greater than the inlet (prior to the dust collector). Dust collector efficiencies usually are higher for larger particles than smaller ones, and the outlet particle sizes are normally smaller than those of the inlet. Samples for #5-251 and #5-253 appear to be associated by anomalies either in recording the data or in the analyses themselves.

	Total Dust "MMAD" and Uranium "AMAD"								
Stack	Designation	Total D	ust MMAD	Uraniu	m AMAD				
			(μm)	()	um)				
R	luns	Inlet	Outlet	Inlet	Outlet				
#9 #9	(Runs 1&2) (Runs 3&4)	$7.0 \pm 2.2$ $7.3 \pm 2.6$	$0.57 \pm 7$ 1.13 + 4.0	$7.0 \pm 2.0$ 5.0 + 2.0	$2.0 \pm 2.0$ $1.0 \pm 4.4$				
#5-261	(Runs 8&5)	$5.3 \pm 2.0$	$3.5 \pm 4.0$	$10.8 \pm 2.0$	$6.6 \pm 2.0$				
#5-260	(Runs 10&6)	6.6 <u>+</u> 2.5	$1.05 \pm 1.6$	$7.0 \pm 2.1$	$1.1 \pm 1.6$				
#5-251	(Runs 9&7)	138.0 ± 4.4	0.63 ± 2.0	0.45 <u>+</u> 1.5	$8.6 \pm 2.7$				
<b>#5-25</b> 3	(Runs 19&11)	$7.4 \pm 2.2$	8.0 ± 5.5	9.0 ± 2.0	$0.67 \pm 1.6$				
#5-249	(Runs 20&16)	$10.5 \pm 2.4$	4.5 ± 1.9	10.3 ± 2.0	$6.7 \pm 7.9$				
#5-250	(Runs 21&17)	$16.5 \pm 2.2$	$1.5 \pm 2.6$	$16.3 \pm 2.0$	$8.3 \pm 2.5$				
#5-254	(Runs 31&22)	8.1 ± 1.2	$3.7 \pm 4.2$	$7.0 \pm 2.7$	5.0 ± 2.5				
#5-256	(Runs 25 <b>&amp;</b> 24)	9.6 <u>+</u> 1.6	8.2 ± 4.2	8.5 ± 1.6	6.5 ± 3.5				
<b>#8-</b> G43-27	(Runs 23 <b>&amp;18</b> )	8.6 ± 2.2	7.8 ±2.3	$7.6 \pm 2.2$	8.8 ± 2.0				
#4-G4-14	(Runs 13&12)	13.9 <u>+</u> 1.9	$2.1 \pm 1.6$	$14.0 \pm 2.0$	9.0 ± 2.5				
#4-64-2	(Runs 14&15)	7.2 ± 2.0	12.0 <u>+</u> 3.9	7.5 <u>+</u> 2.0	$10.0 \pm 2.0$				
#4-64-7	(Runs 26&27)	$1.66 \pm 2.3$	8.0 <u>+</u> 4.4	1.9 <u>+</u> 2.0	8.5 ± 4.2				
#4-034-12	(Runs 28&29)	10.5 ± 2.0	NA	10.5 ± 2.3	8.0 ± 2.0				
#4-64-15	(Runs 30&32)	$3.1 \pm 1.8$	0.3 <u>+</u> 9.6	4.4 <u>+</u> 3.0	$3.2 \pm 5.2$				

### Table C1-1. Data From the Northern Kentucky Environmental Services Report (Organized by Stack)

a Uncertainty terms are not defined in the NKES report. The magnitude of the values suggest they are geometric standard deviations, but if so, the use of a "±" is inappropriate.

### CONCLUSIONS

Uranium Outlet Particle Diameters. Because of differences in the various reports of particle size data. namely NKES and material or reports derived therefrom, extreme care

should be exercised prior to using information on particle size without first checking the original data sources. Variation between data sources indicates that the Total Dust MMAD and uranium AMAD are within one micron for any set of runs. However, "sigma" values may vary by a factor of two and the nature of the particle size distribution is not always clear.

Significance for Dose Reconstruction. The most significant result from the above studies which impacts on dose reconstruction is that the median aerodynamic diameters varied from stack to stack at a particular plant. Any attempt to group stacks into plant groupings, or plants into a single or multiple source must consider the particle size of the "unusual" stack at a plant and the emissions from "uncharacteristic" stacks. In addition to the variations in particle sizes among stacks, the sigma values require careful consideration in that they could influence the overall behavior of the uranium discharged in terms of its deposition or inhalation. Finally, possible bimodal or other distributions need to evaluated as to their effect on environmental transport and inhalation.

It should be kept in mind that the particle size studies reviewed above characterize uranium in terms of its aerodynamic diameter, whereas most meteorological distribution models consider the physical diameter, and perhaps settling velocity, in performing atmospheric transport. In any case the two are not the same and need to be related one to the other prior to making deposition and diffusion calculations.

Further Work Required. There is a need to investigate the effect of particle density and the relationship between the aerodynamic and physical diameters of the uranium particles. The effect of particle size on deposition of uranium and inhalation with distance from the plant should be the subject of a limited investigation.

It may be possible to group stacks, plant or processing sources in terms of a characteristic particle size(s). Again care needs to be taken in doing this. This is discussed further in the Task 4 Appendix C report (Killough et al. 1993). and Part 2 of this Appendix.

Other Useful Data In The NKES Report. The NKES report contains useful information for other aspects of the study, namely: traverse data. stack temperature and relative humidity. and stack diameter.

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### APPENDIX C — PARTICLE SIZE OF AIRBORNE EFFLUENTS

### PART 2 — FINAL PARTICLE-SIZE DISTRIBUTIONS

### MEASURED PARTICLE SIZES

The particle-size distributions of uranium stack emissions are needed in order to calculate both the gravitational settling of uranium-containing particulates in airborne plumes and radiation exposures via the inhalation pathway. In addition, a knowledge of the particle-size distributions is necessary if corrections of uranium stack releases need to be made to account for losses through particle deposition in sampling lines. The only measurements of the particle sizes of stack emissions from the FMPC were conducted by Northern Kentucky Environmental Services (NKES) during 1985. An unpublished report is available on this work (Reed 1985). In the NKES study, measurements were made for both the inlet ducts and the outlet ducts of 15 major uranium-emitting stacks with dust collectors. The particle-size distributions determined in the study are listed in an FMPC report. FMPC-2082 (Boback et al. 1987).

George G. Killough, of the RAC research team, generated a series of plots containing distributions of the uranium species for both the inlet and outlet ducts of each of the 15 dust collectors with the use of a procedure he had developed for interpolating and extrapolating the FMPC-2082 values. The plots and procedure are reported in Appendix F of the RAC Task 2 and 3 report on Fernald dose reconstruction (Voillequé et al. 1991).

Appendix D of the Task 4 report contains the final particle-size distributions as used in this study (Killough et al. 1993) Particle sizes for the outlet ducts (or emission stacks) are representative of emissions from stacks with intact bag filters in the dust collectors. The values for the inlet ducts, however, may be assumed to represent emissions from the same stacks during those periods in which the bag filters had failed in a manner that allowed unfiltered inlet air to escape to the atmosphere.

Particle-size distributions measured for stack emissions during the year 1985 can apply to all other years in which the same plant operations served the same stacks, because plant operations have not changed significantly from 1953 to 1985. The hydrofluorination process for producing  $UF_4$  (green salt), for example, has remained basically the same over the years with respect to conditions which might affect the particle sizes of the product. The various plant operations which produce  $U_3O_8$  particulates also have not changed in a manner which would significantly alter particle sizes.

The predominant uranium species emitted from each stack was identified from FMPC reports and engineering drawings of process equipment. In some cases, more than one uranium species was determined to be emitted from a stack. Either  $UF_4$  or  $U_3O_8$  was emitted from all of the stacks of the NKES study except for one stack which emitted a mixture of  $UO_2$  and  $UO_3$ .

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### Verification of Particle-Size Measurements

The values as listed in FMPC-2082 were verified by comparisons with information from the original data sheets of NKES. Verified values reported herein are those for which the original data are consistent with values listed in FMPC-2082 and which meet the test of physical reality. The latter test is simply the question of whether, as expected, the particle sizes for the outlet ducts of specific dust collectors are less than those for the inlet ducts over the entire range of measurements.

Most of the particle sizes listed in FMPC-2082 were verified in accordance with the criteria above, but discrepancies and omissions were found in the cases listed below:

- The original data sheets for the inlet duct of G4-5 and the outlet duct of G4-12 were not included in the original data file, but these data sheets were later obtained from Michael Boback of FMPC. Since these two data sheets appeared to contain original data taken by NKES, the particle sizes for these ducts as reported in FMPC-2082 were verified.
- Discrepancies included outlet ducts of G5-251. G5-253. and G5-260; the particle-size distributions as reported in FMPC-2082 for these cases were not consistent with the original NKES data sheets. The FMPC-2082 values for these cases had been derived from modified data sheets. Examination of original analytical data sheets helped to resolve the questions of the source of the modifications, but indications of possible sample misidentifications were found for these outlet ducts. Accordingly, the calculated particle sizes were not verified.

Also, the calculated particle sizes of the inlet duct of G5-251 were not verified because they were unrealistically much smaller than those for the outlet duct and were therefore suspect.

- It was observed that measured particle sizes for the outlet ducts of G4-7 and G43-27 were greater than those for the corresponding inlet ducts, which is physically unrealistic. No additional data sheets for these stacks could be located, so the particle sizes listed in FMPC-2082 were regarded as suspect and were not verified.
- It was also found in the verification process that reported values for the larger particle sizes for the inlet ducts of G5-254 and G5-256 as reported in FMPC-2082 seem to contain relatively small systematic errors of 5% and 10%. respectively. These errors were corrected, however, and the corrected values are included as a part of the verified source-term data.

### Calculation of Averages of Verified Values

The average particle-size distributions for both the inlet ducts and the outlet ducts for stacks emitting  $UF_4$  and  $U_3O_8$  were calculated from the Killough plots (Voillequé et al. 1991, Appendix F). Tables C2-1 through C2-4 list particle-size distributions by similar size ranges





for stack emissions of the same uranium species  $(UF_4 \text{ or } U_3O_8)$  and duct type (inlet or outlet) which had been verified as described previously. The particle-size values are given as equivalent aerodynamic diameters in micrometers. The average distributions for the same size ranges are given in the same tables.

The equivalent aerodynamic diameter is defined as the diameter of a sphere of unit density (1 g cm<sup>-3</sup>) which has the same gravitational settling velocity as the particle if the particle is spherical. The physical diameter is equal to the equivalent aerodynamic diameter divided by the square root of the particle density in g cm<sup>-3</sup>.

Table C2-5 lists the median values for the average distributions, which are AMAD (activity median aerodynamic diameters) values required by the ICRP for its respiratory model. The GSD values listed may be used with caution to determine approximate curves only since the distributions are not generally lognormal.

### Table C2-1. Particle Sizes for Airborne UF<sub>4</sub> Made by the Hydrofluorination Process in Plant 4 (Outlet Ducts)

			I	Percent UF	4 for Particl	e-Sizeª Ran	ge	
Plant	Stack	0-2.5	2.5-5	_5-7.5	7.5-10	10-15	15-20	20-40
4	G4-2	8	12	15	16	29	15	5
	G4-5	25	17	14	8	13	7	16
	G4-12	5	15	22	23	25	8.5	1.5
	G4-14	11.5	13.5	15	15	21	14	10
5	G5-249	25	13	15	13	19	10.5	4.5
	G5-250	16	14	15	15	16	14	10
Avera	ge	15.1	14.1	16.0	15.0	20.5	11.5	7.8

Equivalent aerodynamic diameter in micrometers

### Table C2-2. Particle Sizes for Airborne UF<sub>4</sub> Made by the Hydrofluorination Process in Plant 4 (Inlet Ducts)

	Percent UF <sub>4</sub> for Particle-Size <sup>a</sup> Range								
Plant	Stack	0-2.5	2.5-5	5-7.5	7.5-10	10-15	15-20	20-40	
4	G4-2	5	17	26	22	22	5.5	2.5	
	G45	23	27	14	10	11	5	10	
	G4-12	3.5	8.5	18	24	29	14	3	
	G4-14	0.8	3.2	8	14	34	22	18	
5	G5–249	4.5	9.5	15	20	29	15	7	
	G5–250	0.7	2.8	6.5	12 -	28	30	20	
	G5–253	12	10	17	18	27	12	4	
Avera	ge	7.1	11.1	14.9	17.1	25.7	14.8	9.2	

<sup>a</sup> Equivalent aerodynamic diameter in micrometers

			(0	v Ducto/				
Percent U <sub>3</sub> O <sub>8</sub> for Particle-Size <sup>a</sup> Range								
Stack	0-2.5	2.5-5	5-7.5	7.5-10	10-15	15-20	20-30	
G5-254	24	22	21	15	10	7.2	0.8	
G5-256	32	16	16	13	17	5	1	
G5-261	13	18	23	19	19	6	2	
ige	23.0	18.7	20.0	15.7	15.3	6.1	1.3	
	Stack G5-254 G5-256 G5-261 ge	Stack         0-2.5           G5-254         24           G5-256         32           G5-261         13           ge         23.0	F <u>Stack 0-2.5 2.5-5</u> <u>G5-254 24 22</u> <u>G5-256 32 16</u> <u>G5-261 13 18</u> <u>ge 23.0 18.7</u>	$\begin{array}{c c c c c c c c c c c c c c c c c c c $	Control 2 detsy           Percent $U_3O_8$ for Partic           Stack         0-2.5         2.5-5         5-7.5         7.5-10           G5-254         24         22         21         15           G5-256         32         16         16         13           G5-261         13         18         23         19           oge         23.0         18.7         20.0         15.7	Percent U <sub>3</sub> O <sub>8</sub> for Particle-Size <sup>a</sup> Rat           Stack 0-2.5         2.5-5         5-7.5         7.5-10         10-15           G5-254         24         22         21         15         10           G5-254         24         22         21         15         10           G5-254         24         22         21         15         10           G5-256         32         16         16         13         17           G5-261         13         18         23         19         19           oge         23.0         18.7         20.0         15.7         15.3	Percent $U_3O_8$ for Particle-Size <sup>a</sup> Range           Stack 0-2.5 2.5-5 5-7.5 7.5-10 10-15 15-20           G5-254 24 22 21 15 10 7.2           G5-256 32 16 16 13 17 5           G5-261 13 18 23 19 19 6           age 23.0 18.7 20.0 15.7 15.3 6.1	

Table C2-3. Particle Sizes for Airborne U<sub>3</sub>O<sub>8</sub> from Foundry Operations in Plant 5 (Outlet Ducts)

" Equivalent aerodynamic diameter in micrometers

Table C2-4. Particle Sizes for Airborne  $U_3O_8$  from Foundry Operations in Plant 5 (Inlet Ducts)

	Percent U <sub>3</sub> O <sub>8</sub> for Particle-Size <sup>a</sup> Range							
Plant	Stack	0-2.5	2.5 - 5	5-7.5	7.5–10	10-15	15-20	2035
5	G5-254	16	22	14	14	14	8	12
	G5-256	5	16	24	17	26	10	2
	G <b>5-2</b> 60	11	20	22	18	18	6.5	4.5
	G5-261	4	10	13	16	27	16	14
Avera	ge	9.0	17.0	18.3	16.3	21.3	10.1	8.1

<sup>a</sup> Equivalent aerodynamic diameter in micrometers

Table C2-5. Mediar	Values of Average	<b>Particle-Size</b>	Distributions	of UF <sub>4</sub>	and	$U_3O_8$
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		Emissions		
Species	Source	Duct	• Median <sup>a</sup>	GSD <sup>b</sup>
UF <sub>4</sub>	Hydrofluorination in Plant 4	Outlet	8.1 µm	2.0
		Inlet	9.5 μm	1.9
U.3O8	Foundry Operations in Plant 5	Outlet	6.0 μm	2.1
		Inlet	8.3 μm	2.0

<sup>a</sup> These median values are read from the 50th percentile on log-probability graphs. The median values of equivalent aerodynamic diameters are the same as AMAD values (activity median aerodynamic diameters) if it is assumed that mass medians and activity medians are equal

 $^{b}$  The geometric standard deviation (GSD) reported in this table has a precise meaning only with respect to lognormal distributions, which should not be assumed for these data. The GSD values were obtained with use of a linear representation of points in the middle regions of log-probability graphs.

## Comparisons of Emission Distributions from Stacks with Different Types of Bag Filters in Dust collectors

Early in the history of FMPC operation, all of the bag filters used in the dust collectors for emission stacks were made from wool felt. There was a change to bag filters made from Gore-Tex<sup>TM</sup> in later years, but the change took place gradually over a period of years. The change was taking place during 1985 when the NKES particle-size measurements were made. The type of bag filter used during the NKES measurements was identified from plant records. Therefore, it was possible to make some comparisons between wool and Gore–Tex<sup>™</sup> bag filters in efficiency of removal of particles of different sizes. A wool bag filter was used in G4-5 while the others in Table C2-1 were made from Gore-Tex<sup>™</sup>. There appears to be a higher percentage of small particles (< 2.5 micrometers AMAD) in the outlet from this stack. The inlet particle-size distribution for this stack (Table C2-2), however, was not greatly different from its outlet distribution. In Table C2-3, the bag filter used for G5-261 was made from Gore-Tex<sup>™</sup> while the other two stacks used wool bag filters in their dust collectors. Comparisons within this table indicate also that there was a higher percentage of small particles in the emissions from wool bag filters. Calculations made for efficiencies of removal of the larger sizes by using particle sizes in the inlet duct (Table C2-4), however, do not show much difference between wool and Gore-Tex<sup>™</sup>. It was concluded on the basis of the observations above that any differences in removal efficiency between wool and Gore-Tex<sup>™</sup> bag filters as calculated from data in Tables C2-1 through C2-4 are not significant. Examination of data from a much larger number of stacks with different types of bag filters would be required to determine whether any real differences exist.

### How the Inclusion of Unverified Data would Affect Averages

If the unverified distribution listed in the FMPC-2082 report for the outlet duct of G5-253 had been included in Table C2-1 for UF<sub>4</sub> emissions, the average values would have been skewed somewhat toward smaller sizes. The average percentage contribution of particles less than 2.5 micrometers AMAD would have been 25.5% instead of 15%. Contributions of the other six size-groups would have been less than the average values in Table C2-1. Inclusion of the unverified distribution for  $U_3O_8$  emitted from stack G5-260 listed in FMPC -2082 would have also resulted in a similarly skewed average for Table C2-3. For particles less than 2.5 micrometers AMAD, the average contribution would have been 36.8% instead of 23%, and the contributions for each of the other six size-groups would have been correspondingly smaller.

If the unverified G5-251 values listed in the FMPC-2082 report had been included in Tables C2-1 and C2-2, the averages for these tables would not have been greatly different except for the particles less than 2.5 micrometers AMAD for the inlet duct. The contribution of this size-group would have been 17.7% instead of 7.1%. Differences would have ranged between 1% and 3% for the other six size-groups for the inlet duct. There would have been differences of less than 1% for each of the seven size-groups of the outlet duct.

The averages for  $U_3O_8$  in Tables C2-3 and C2-4 would not have changed greatly if the unverified particle-size distributions of the inlet and outlet ducts of G43-27 had been included. The percentage contribution of particles less than 2.5 micrometers AMAD in the outlet duct would have been 19% instead of 23% with much smaller changes in

contributions of the other six size-groups. The change for the inlet duct would have been less than 1% for each of the seven size-groups.

The G4-7 stack emitted a mixture of  $UO_3$  and  $UO_2$ . This is the only stack in the group studied by NKES in which the predominant emitted species was neither  $UF_4$  nor  $U_3O_8$ . Therefore, it was not possible to compare the unverified particle-size distribution for this stack with other distributions.

### INFERRED PARTICLE SIZES FOR STACKS FOR WHICH NO MEASUREMENTS HAD BEEN MADE IN THE 1985 NKES STUDY

The particle-size distributions for emissions from stacks for which no measurements had been made may be inferred from the results obtained from the other stacks. The particlesize distributions of the stacks which emitted  $UF_4$  produced by the hydrofluorination process were averaged as shown in Tables C2-1 and C2-2, and this average distribution is assumed to apply to all stacks emitting  $UF_4$  also produced by hydrofluorination but for which reliable measured values are not available. Airborne  $U_3O_8$  is produced in the FMPC as a result of the oxidation of uranium metal surfaces by air. There are two general types of plant operations which can produce airborne  $U_3O_8$  particles: (1) foundry operations such as melting and casting of uranium metal, crucible breakout of uranium derbies and ingots, and cleaning of metal surfaces, and (2) the machining of uranium derbies and ingots. The stacks which exclusively emitted U<sub>3</sub>O<sub>8</sub> in the 1985 NKES study served only foundry operations in Plant 5. Hence, the average particle-size distribution for  $U_3O_8$  emissions in this study as shown in Tables C2-3 and C2-4 is assumed to apply to all stacks exclusively serving foundry operations which emitted  $U_3O_8$  and for which no measurements had been made in 1985. Surface oxidation of uranium scrap in high-temperature furnaces such as those in Plant 8 was also assumed to be in the same category as foundry operations.

### Inferred Particle Sizes for U<sub>3</sub>O<sub>8</sub> Produced During Machining

Machining operations such as cutting and milling of uranium metal ingots and derbies were conducted in Plant 6 and Plant 9. No particle-size measurements for  $U_3O_8$  produced during machining operations were made in the 1985 NKES study, however, so comparisons with similar operations at other facilities were used to estimate particle-size distributions from machining at the FMPC.

A 1959 paper reported an average value of 2.5 micrometers for the mass median diameter of  $U_3O_8$  particles produced in the machining of uranium at Los Alamos (Hyatt et al. 1959). This value corresponds to an AMAD of 6.7 micrometers for an assumed density of 7.0 g cm<sup>-3</sup> for the  $U_3O_8$  particles. A mean particle size of 6.9 micrometers was recently reported for similar operations at AWE in the United Kingdom (Vallis 1991). An average for the two facilities (6.8 micrometers) may be assumed to apply to inlet ducts to dust collectors serving machining operations in Plant 6 and Plant 9 at the FMPC. An average value of about 5.1 micrometers is estimated to apply to the outlet ducts. This value would represent a 25% reduction in median particle size as a result of filtering, which is about the average reduction observed in measurements at the FMPC.

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### Particle Sizes of U<sub>3</sub>O<sub>8</sub> from Foundry Operations at Other Facilities

Particle sizes of  $U_3O_8$  in air during foundry operations were measured at Los Alamos and at two facilities in the United Kingdom. The results provide some confirmation of the FMPC data for similar operations. The AMAD values in micrometers were as follows:

Los Alamos (Hyatt et al. 1959)	7.3
AWE in UK (Vallis 1991)	11
Springsfields in UK (Fishwick 1991)	8

The average value (8.8 micrometers) compares favorably with the average inlet value of 8.3 micrometers for FMPC foundry operations.

### Particle Sizes for Emissions from Plant 1 and Plant 2/3

A mixture of particles of  $U_3O_8$ ,  $UO_3$ , and  $UO_2$  is assumed to be emitted from stacks of Plant 1 and Plant 2/3 as a result of handling of ores and various other feedstocks to provide feed to digestors. Since the 1985 NKES study did not include any stacks for these plants, particle-sizes for these emissions must be inferred from measurements made for similar operations elsewhere. A study was carried out on particle sizes of uranium containing dust from mining and milling operations in the Elliot Lake Area of Canada (Duport and Edwardson 1985, Duport and Horvath 1989).

AMAD values (micrometers) were reported for mill atmospheres for the following processes: jaw crushing, 9.5; cone crushing, 9; screening, 7.5; grinding, 8; acid precipitation, 6; filtering, 10; concentrate drying, 8; and concentrate packing, 7.5. The corresponding GSD values ranged between 3 and 5. The average AMAD for mills (possibly a weighted average) was reported to be about 7 micrometers. A mean particle size of 7 micrometers with a GSD of about 4 may be inferred for the  $U_3O_8$  dust emitted from Plant 1 and Plant 2/3 as a result of ore handling if it may be assumed that the ore-handling processes in these plants were similar to those in the Elliot Lake Area. This inferred value would apply to inlet ducts of the dust collector stacks. A mean value of 5.3 micrometers would apply to the corresponding outlet ducts as a result of a reduction of 25% in the median particle size during filtration.

# Particle Sizes for UF<sub>4</sub> Produced by Reduction of UF<sub>6</sub> Vapor by Hydrogen Gas in Dissociated Ammonia

One of the stacks in the FMPC Pilot Plant served a process for making  $UF_4$  by reduction of  $UF_6$  by hydrogen gas. There is no particle-size information available on emissions from this process at present.

### OTHER PROPERTIES OF PARTICULATES

In addition to particle sizes, the particle densities and shape factors are also used in calculations of the gravitational fall velocities of large or dense particles. These other parameters are discussed below.

### Densities

Emitted particulates are produced rapidly during FMPC processing, and hence they would be expected to be imperfectly formed and contain voids. Therefore, their densities would be less than theoretical or "handbook" values, which would represent maximum values. The only information found in FMPC reports or other records concerning particulate density of emitted materials was a value of 6.4 g cm<sup>-3</sup> for UF<sub>4</sub> produced at the FMPC (Freitag 1964). The "handbook" value for UF<sub>4</sub> is 6.7 g cm<sup>-3</sup>. The value used in Los Alamos particle-size studies for U<sub>3</sub>O<sub>8</sub> particulates produced by foundry operations and by machining is 7.0 g cm<sup>-3</sup> (Hyatt et al. 1959). The "handbook" value listed for U<sub>3</sub>O<sub>8</sub> is 8.30 g cm<sup>-3</sup>.

### Shape factors

The calculated value for the gravitational fall velocity of a particle should be divided by a shape factor if the particle is not spherical. Values of shape factors applicable to cylindrical shapes are listed in Table C2–6 (Chamberlain 1975).

Table C2–6. Shape Factors versus Axis Ratio		
Ratio of Axes	Shape Factor	
1	1.06	
2	. 1.14	
3	1.21	
4	1.32	

Only a few memoranda or FMPC plant reports containing photomicrographic information on plant products have been located. These reports contained photomicrographs of UF<sub>4</sub> produced by the Winlo Process, which was carried out in Plant 8 from 1962 to 1964. The average measured ratio of axes of this product was found to be about 1.5. Photomicrographs of  $U_3O_8$  dust from Los Alamos foundry operations (Hyatt et al. 1959) show irregular particles with length to width ratios generally ranging from 1 to 2. A value of 1.5 represents an approximate average. In lieu of better information, it appears likely that the use of a shape factor of 1.1, for instance, would not lead to serious error in calculation of gravitational fall velocities.

#### SUMMARY AND CONCLUSIONS

The particle-size distributions measured for stack emissions in 1985 by NKES can be applied to emissions from the same stacks serving the same plant operations or processes in other years since specific plant operations have not changed significantly over the years. Averaged distributions for  $UF_4$  emissions and for  $U_3O_8$  emissions as calculated from the NKES measurements can be applied to stacks for which there were no measurements made in 1985 for all cases in which the emitted species was produced through similar operations. Inferred values for particle sizes of  $U_3O_8$  from machining operations in other facilities can

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be applied to stacks serving FMPC machining operations. Similarly, uranium oxide particle sizes reported by other facilities can be applied to similar ore-handling processes carried out in Plants 1 and 2/3.

An assignment of particle sizes for uranium releases over all of the years of operation of the FMPC requires identification of both the predominant species and its generating plant process for each major emission point for each year. For the few cases for which no reliable information on particle size can be obtained, particle sizes can be assigned at midpoints of expected maximum uncertainty ranges.

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# APPENDIX D -- COMPARISON OF THE RAC MODELS WITH OTHER MODELS PART 1 -- COMPARISON OF MODELS FOR AIRBORNE URANIUM AND RADON

### INTRODUCTION

The modeling methodology of Task 4 (Killough et al. 1993) identifies two air transport models to be applied to releases of particulate uranium and to radon and radon daughters released from the FMPC site. The well-known GAUSSIAN PLUME model (Hanna et al. 1982) is used for releases of uranium from the old solid waste incinerator on the east boundary of the site, and from the oil burner, which was located in the production area during the period (1960–1962). For this three-year period, these sources accounted for less than 1% of the uranium released to the atmosphere from the site. For rooftop releases of uranium from the production plants, we used a variant of the Gaussian plume, called the TIME-DEPENDENT model (Ramsdell 1990). designed to account for building wake effects. We have also applied a specially-coded version of this model to releases of radon and radon daughters from the K-65 silos west of the production area. on the assumption that wake effects from the silos should be considered.

Implementations of these models for specific purposes involve complexities that have been discussed elsewhere (Killough et al. 1993). This discussion is confined to tests to confirm our interpretation of the basic form of each code — by comparisons to an independent code. For the Gaussian plume model, we have compared results calculated by our program with similar numbers computed by MICROAIRDOS<sup>TM</sup> (Moore et al. 1989). In the case of the time-dependent model, we have used a graph from Ramsdell (1990) as our standard.

### The Gaussian Plume Model

The RAC Gaussian Plume Model was compared against an assumed source term and MICROAIRDOS<sup>TM</sup>. The assumed source terms were selected *a priori*, the only selection criterion being that they be workable with both models. The hypothetical source term consisted of 1000 kg of natural uranium, and 1000 Ci of radon ( $^{222}$ Rn) in equilibrium with its daughters. A particle-size diameter of 1 micron Activity Median Aerodynamic Diameter (AMAD) was employed. The curie equivalents for 1000 kg of natural uranium assumes 99.3 g of  $^{238}$ U per 100 g of natural uranium, and a specific activity for  $^{238}$ U of  $33.5 \times 10^{-8}$  Ci g<sup>-1</sup>. This calculates to  $3.33 \times 10^{-1}$  Ci of  $^{238}$ U per 1000 kg of natural uranium. The daughters of  $^{238}$ U,  $^{234}$ U and  $^{234}$ Th were assumed to be in equilibrium with  $^{238}$ U, and therefore the same curie amounts were present for each decay product as for the parent  $^{238}$ U. Assumptions for  $^{235}$ U were 0.0072 g per 100 g of natural uranium and a specific activity of 2.16 x  $10^{-6}$  Ci g<sup>-1</sup> to yield the equivalent of 1.56 x  $10^{-4}$  Ci per 1000 kg of natural uranium.

For radon, the RAC GAUSSIAN PLUME program calculates the dynamic build up of radon daughters as the plume moves downwind. Any level of equilibrium at the source can be assumed. The equilibrium between radon and its daughters is assumed to be 0.5 by MICROAIRDOS<sup>TM</sup>. This would be important if one were calculating doses, but is not

pertinent to this comparison because the model output parameters that we compared were air concentrations (for radon and uranium) and ground deposition for uranium.

Other pertinent input parameters to the models for uranium were a physical release height of 30 m with no plume rise. For radon, a physical release height of 12 m with no plume rise was employed.

Initially, calculations were made for the NE sector where one would expect the highest air concentrations and ground depositions for releases from the FMPC. We calculated results at 500 m, and then at 1000 m intervals out to 8000 m (Tables D1–1 and D1–2). Following this initial comparison, results were compared for various wind directions to insure that this variable did not skew the results. Only <sup>238</sup>U and radon were compared in the latter case, since no variations with uranium isotopes were observed (Table D1–3). However, for ground concentrations <sup>234</sup>Th did give about one-tenth the ratio between MICROAIRDOS<sup>TM</sup> and the RAC model. This is because MICROAIRDOS<sup>TM</sup> assumes that the radionuclides are released over a year and decay on the ground for a year after deposition. The RAC model employs instantaneous release depositions. For long-lived radionuclides such as <sup>238</sup>U, <sup>234</sup>U, and <sup>235</sup>U with half-lives of 4.468 x 10<sup>9</sup>, 2.445 x 10<sup>5</sup>, and 7.038 x 10<sup>8</sup> years (Shleien, 1992).the discrepancy would be unnoticed because the radionuclide decay over a year is minimal. However for <sup>234</sup>Th, with a half-life of 24.1 days (Shleien 1992) the discrepancy due to decay is considerable.

	Concentration in Air (Ci m <sup>-0</sup> )						
Distance (meters)	MICROAIRDOS™	RAC	Ratio MICROAIRDOS™/RAC				
1000	8.32x10 <sup>-11</sup>	7.10x10 <sup>-11</sup>	1.17				
2000	$3.09 \times 10^{-11}$	$2.57 \times 10^{-11}$	1.20				
3000	$1.69 \times 10^{-11}$	$1.40 \times 10^{-11}$	1.20				
4000	$1.11 \times 10^{-11}$	9.15x10 <sup>-11</sup>	1.21				
5000	$8.07 \times 10^{-12}$	6.63x10 <sup>-11</sup>	1.21				
6000	$6.25 \times 10^{-12}$	$5.12 \times 10^{-11}$	1.21				
7000	$5.05 \times 10^{-12}$	4.14x10 <sup>-11</sup>	1.22				
8000	$4.22 \times 10^{-12}$	3.46x10 <sup>-11</sup>	1.21				

Table D1-1. Radon (222Rn) Concentrations in Air at Various Distances in the NEDirection: Comparison of MICROAIRDOS and RAC Models



	•	MICF	ROAIRDOS	STM and RAC Mo	dels		
	Radio- nuclide	Concentra (Ci		n in Air Concentration on Ground		- <u></u>	
Distance (meters)		MICROAIRDOS	RAC	MICROAIRDOS™	RAC	R <u>MICROAIR</u> Air	atio <u>DOS™/RAC</u> Ground
500	238U	1.53x10 <sup>-14</sup>	1.14x10 <sup>-14</sup>	9.16x10 <sup>-9</sup>	6.92x10 <sup>-9</sup>	1.34	1.32
500	$234_{U}$	$1.53 \times 10^{-14}$	1.14x10 <sup>-14</sup>	9.16x10 <sup>-9</sup>	6.92x10 <sup>-9</sup>	1.34	1.32
500	$^{234}$ Th	$1.53 \times 10^{-14}$	1.14x10 <sup>-14</sup>	8.73x10 <sup>-10</sup>	6.92x10 <sup>-9</sup>	1.34	0.13
500	$235_{U}$	7.16x10 <sup>-18</sup>	5.34x10 <sup>-18</sup>	$4.29 \times 10^{-12}$	3.24x10 <sup>-12</sup>	1.34	1.32
1000	$238_{U}$	9.57x10 <sup>-15</sup>	6.69x10 <sup>-15</sup>	5.74x10 <sup>-9</sup>	4.08x10 <sup>-9</sup>	1.43	1.40
1000	234U	$9.57 \times 10^{-15}$	6.69x10 <sup>-15</sup>	5.74x10 <sup>-9</sup>	4.08x10 <sup>-9</sup>	1.43	1.40
1000	$^{234}$ Th	$9.57 \times 10^{-15}$	6.69x10 <sup>-15</sup>	5.46x10 <sup>-10</sup>	4.08x10 <sup>-9</sup>	1.43	0.13
1000	$235_{U}$	4.49x10 <sup>-18</sup>	3.14x10 <sup>-18</sup>	$2.69 \times 10^{-12}$	1.91x10 <sup>-12</sup>	1.42	1.40
2000	$238_{U}$	5.34x10 <sup>-15</sup>	$3.50 \times 10^{-15}$	3.20x10 <sup>-9</sup>	2.12x10 <sup>-9</sup>	1.52	1.50
2000	234U	$5.34 \times 10^{-15}$	$3.50 \times 10^{-15}$	$3.20 \times 10^{-9}$	2.12x10 <sup>-9</sup>	1.52	1.50
2000	<sup>234</sup> Th	5.34x10 <sup>-15</sup>	3.50x10 <sup>-15</sup>	$3.05 \times 10^{-10}$	2.12x10 <sup>-9</sup>	1.52	0.13
2000	$235_{U}$	2.50x10 <sup>-18</sup>	1.64x10 <sup>-18</sup>	$1.50 \times 10^{-12}$	9.93x10 <sup>-13</sup>	1.52	1.50
3000	238U	3.18x10 <sup>-15</sup>	$2.09 \times 10^{-15}$	1.90x10 <sup>-9</sup>	1.27x10 <sup>-9</sup>	1.52	1.50
3000	234 <sub>U</sub>	3.18x10 <sup>-15</sup>	$2.09 \times 10^{-15}$	1.90x10 <sup>-9</sup>	1.27×10 <sup>-9</sup>	1.52	1.50
3000	$^{234}$ Th	3.17x10 <sup>-15</sup>	2.09x10 <sup>-15</sup>	$1.81 \times 10^{-10}$	1.27x10 <sup>-9</sup>	1.52	0.14
3000	$235_U$	1.49x10 <sup>-18</sup>	9.75x10 <sup>-19</sup>	8.91x10 <sup>-13</sup>	$5.94 \times 10^{-13}$	1.52	1.50
4000	$238_{U}$	$2.01 \times 10^{-15}$	1.37x10 <sup>-15</sup>	1.21x10 <sup>-9</sup>	8.34x10 <sup>-10</sup>	1.47	1.44
4000	234 <sub>U</sub>	$2.01 \times 10^{-15}$	1.37x10 <sup>-15</sup>	1.21x10 <sup>-9</sup>	8.34x10 <sup>-10</sup>	1.47	1.44
4000	<sup>234</sup> Th	$2.01 \times 10^{-15}$	1.37x10 <sup>-15</sup>	1.15x10 <sup>-10</sup>	8.34x10 <sup>-10</sup>	1.47	0.14
4000	$235_{U}$	9.42x10 <sup>-19</sup>	6.43x10 <sup>-19</sup>	5.64x10 <sup>-13</sup>	3.91x10 <sup>-13</sup>	1.46	1.44
5000	$238_{U}$	$1.39 \times 10^{-15}$	9.69x10 <sup>-15</sup>	$8.32 \times 10^{-10}$	5.88x10 <sup>-10</sup>	1.48	1.42
5000	234U	$1.39 \times 10^{-15}$	9.69x10 <sup>-15</sup>	$8.32 \times 10^{-10}$	5.88x10 <sup>-10</sup>	1.48	1.42
5000	<sup>234</sup> Ть	$1.39 \times 10^{-15}$	9.69x10 <sup>-16</sup>	$7.92 \times 10^{-11}$	5.88x10 <sup>-10</sup>	1.48	0.13
5000	235 <sub>U</sub>	$6.51 \times 10^{-19}$	4.54x10 <sup>-19</sup>	3.90x10 <sup>-13</sup>	$2.76 \times 10^{-13}$	1.43	1.41
6000	238 <sub>U</sub>	9.93x10 <sup>-16</sup>	7.19x10 <sup>-16</sup>	5.95x10 <sup>-10</sup>	4.34x10 <sup>-10</sup>	1.38	1.37
6000	234 <sub>U</sub>	9.93x10 <sup>-16</sup>	7.19x10 <sup>-16</sup>	5.95x10 <sup>-10</sup>	4.34x10 <sup>-10</sup>	1.38	1.37
6000	<sup>234</sup> Тһ	9.93x10 <sup>-16</sup>	7.19x10 <sup>-16</sup>	$5.67 \times 10^{-11}$	4.34x10 <sup>-10</sup>	1.38	0.13
6000	$235_{U}$	4.65x10 <sup>-19</sup>	3.37x10 <sup>-19</sup>	2.79x10 <sup>-13</sup>	2.03x10 <sup>-13</sup>	1.38	1.37
7000	$238_{U}$	7.25x10 <sup>-16</sup>	5.54x10 <sup>-16</sup>	4.34x10 <sup>-10</sup>	3.36x10 <sup>-10</sup>	1.31	1.29
7000	234 <sub>U</sub>	$7.25 \times 10^{-16}$	5.54x10 <sup>-16</sup>	4.34x10 <sup>-10</sup>	3.36x10 <sup>-10</sup>	1.31	1.29
7000	$^{234}$ Th	$7.24 \times 10^{-16}$	5.54x10 <sup>-16</sup>	4.13x10 <sup>-11</sup>	3.36x10 <sup>-10</sup>	1.31	0.12
7000	235 <sub>U</sub>	3.39x10 <sup>-19</sup>	$2.60 \times 10^{-19}$	$-2.03 \times 10^{-13}$	1.58x10 <sup>-13</sup>	1.30	1.27
8000	238 <sub>U</sub>	5.63x10 <sup>-16</sup>	4.40x10 <sup>-16</sup>	$3.37 \times 10^{-10}$	2.66x10 <sup>-10</sup>	1.28	1.27
8000	234 <sub>U</sub>	5.63x10 <sup>-16</sup>	4.40x10 <sup>-16</sup>	3.37x10 <sup>-10</sup>	2.66x10 <sup>-10</sup>	1.28	1.27
8000	234Th	$5.62 \times 10^{-16}$	4.40x10 <sup>-16</sup>	$3.21 \times 10^{-11}$	2.66x10 <sup>-10</sup>	1.28	0.12
8000	235 <sub>U</sub>	2.64x10 <sup>-19</sup>	2.06x10 <sup>-19</sup>	$1.58 \times 10^{-13}$	1.24x10 <sup>-15</sup>	1.28	1.27

### Table D1-2. Uranium Particulate Concentrations in Air and on the Ground at Various Distances in the NE Direction: Comparison of Predictions of MICROAIRDOS<sup>TM</sup> and RAC Models

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	23	<sup>8</sup> U Release qu	antity (3.33 x 1	0 <sup>-1</sup> Ci)		
		Air (Ci	m <sup>-3</sup> )	Ground (Ci m <sup>-2</sup> )		
	· MICH	ROAIRDOS	RAC	MICROAIRDOS <sup>TH</sup>	RAC	
NW	4.2	6x10 <sup>-16</sup>	3.01x10 <sup>-16</sup>	2.56x10 <sup>-10</sup>	$1.82 \times 10^{-10}$	
SW	4.7	0x10 <sup>-16</sup>	$3.27 \times 10^{-16}$	$2.82 \times 10^{-10}$	$1.98 \times 10^{-10}$	
SE	6.9	4x10 <sup>-16</sup>	4.91x10 <sup>-16</sup>	4.16x10 <sup>-10</sup>	$2.09 \times 10^{-10}$	
			Ratio			
		MICROA	IRDOS™/RAC			
	Aiı	2		<u>Ground</u>		
	1.4	1		1.40		
	1.4	3		1.42		
	1.4	1		1.99		
		<sup>222</sup> Rn Release	quantity (1000	) Ci)		
	Air (C	i m <sup>-3</sup> )				
	······································			Ratio		
	MICROAIRDOS™	RAC	Μ	ICROAIRDOS™/RAC	2	
NW	2.18x10 <sup>-12</sup>	1.97x10 <sup>-12</sup>		1.10		
SW	$2.52 \times 10^{-12}$	$2.08 \times 10^{-12}$		1.21		
SE	$3.49 \times 10^{-12}$	$3.61 \times 10^{-12}$		0.96		

## Table D1-3. Comparison of Predicted <sup>238</sup>U and <sup>222</sup>Rn Concentrations in Air and on the Ground at 4000 m in Different Directions.

### RESULTS

A comparison of results obtained from the two programs for radon and uranium concentrations in the NE sector are presented in Tables D1-1 and D1-2, respectively. The results show reasonable agreement. The ratio between MICROAIRDOS™ and the RAC program has a range of 1.27 to 1.52 in both comparisons of air concentrations and of ground depositions for <sup>238</sup>U, <sup>234</sup>U, and <sup>235</sup>U (Table D1-2). For radon air concentrations the variation is somewhat less, from 1.17 to 1.21 (Table D1-1). For the shorter lived <sup>234</sup>Th the ground concentration ratio ranged from 0.12 to 0.14 due to its decay on the ground over a year. MICROAIRDOS™ employs one year's decay on the ground while the RAC model does not.

A similar comparison (Table D1-3) for <sup>238</sup>U was run for three different wind directions. The directions chosen were: NE, SW; and SE. The results show reasonable agreement between the programs with a MICROAIRDOS<sup>TM</sup>/RAC ratio of 1.4 to 2.0.

Differences between the RAC air dispersion model and MICROAIRDOS™ may be due to assumptions used for each model. The principal cause for discrepancy is that the RAC model corrects wind speeds from the height at which they are measured (typically 10 m) to the release height (30 m in this case). This correction would increase the wind speed by different amounts for different stability classes, and it would correspondingly decrease the estimate of ground-level air concentration. The results for each model were in reasonable agreement

indicating that algorithms and methods of coding employed by RAC and MICROAIRDOS<sup>TM</sup> are comparable.

### The Time-Dependent Model

For the time-dependent model, we employ output from a study by Ramsdell (1990). Ramsdell (1990) used a graph (Figure 9 of his paper) to summarize a model comparison involving the time-dependent model, and we have digitized the appropriate curve from that graph to serve as our standard for comparison. It was impractical to use any of the computer programs that we have written for the comparisons reported by Killough et al. (1993). because these programs were designed to use meteorological joint frequency tables, and they calculate deposition and plume depletion. The version used by Ramsdell (1990) for the graph used a single wind speed  $(3 \text{ m s}^{-1})$ , a single stability category (D), a standard building area  $(1,000 \text{ m}^2)$ , and did not account for plume depletion due to deposition. In addition, a calibration parameter K, was set equal to 1.0 by Ramsdell for the curve shown in the graph. We prepared a simple program that represents our interpretation of the algorithm derived in the paper (Ramsdell 1990). It is the same interpretation that has been written into the programs for the comparisons, except that parameter values have been set to agree with the choices that produced the curve. Figure D1-1 shows the digitized curve from Ramsdell (1990) and a curve plotted from values calculated with our program. The two curves practically coincide.

We note one difference between the standard curve and ours. Although the figure in Ramsdell's paper did not specify which formulas were used for the dispersion coefficients  $\sigma_y$  and  $\sigma_z$ , the references NRC (1982) and Bander et al. (1982) were cited in that connection elsewhere in the paper. In our calculation for Figure D1-1, we continued to use Briggs formulas (Hanna et al. 1982) as we have done for the comparisons. The difference appears to be unimportant.



**Figure D1-1.** Comparison of computer implementation of the time-dependent model by *RAC* with a curve published by Ramsdell (1990).

Such a comparison as the one summarized in Figure D1-1 is, of course, very narrow in scope. It tests our interpretation of the published algorithm and our method of coding the algorithm. It cannot test directly our more elaborate implementations of the model.

### SUMMARY

Tests of algorithms and coding methods of the RAC GAUSSIAN PLUME model indicates good agreement with a commercially available independently derived air model.

A test of the algorithm and coding method for the TIME-DEPENDENT model showed our method of implementation is correct.

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### APPENDIX D—COMPARISON OF THE RAC MODELS WITH OTHER MODELS

### PART 2—SURFACE WATER PATHWAYS

#### INTRODUCTION

The surface water modeling methodology for the transport and dispersion of radioactive materials from the FMPC is based on a simple monthly dilution (MD) model. In Task 4 (Killough et al. 1993), we described this model for calculating concentrations of radionuclides in surface waters near the FMPC. We will ultimately use this model to calculate radiation doses from releases of radioactive materials from the FMPC. This part of Appendix D compares our MD model with an independent, surface water dispersion model, GENII (Napier et al. 1988).

We present the results of such a comparison, based on our monthly source term estimates for 1960 to 1962 (Voillequé et al. 1991), in Task 4. We compared our basic MD model, including uncertainty estimates from a statistical risk management program called CrystalBallÔ, with the GENII model developed by Battelle for the Hanford Environmental Project (Napier et al. 1988). In this appendix we provide the details of this comparison.

#### **GENII MODEL**

The GENII code allows one to calculate the concentration of radionuclides in water, and radiation doses resulting from releases of radionuclides to nontidal rivers and near-shore lake environs. GENII incorporates the internal dosimetry models recommended by the International Commission on Radiological Protection (ICRP) in updated versions of the environmental pathway analysis models used at the Hanford Laboratories in Richland, Washington (Napier 1988). The surface water program in GENII solves for radionuclide concentrations in a river under the following assumptions:

- constant flow depth
- constant downstream longshore velocity
- straight river channel
- constant lateral dispersion coefficient
- continuous point discharge release of effluent
- constant river width.

For the GENII runs, we used representative values for the river in the vicinity of the FMPC for water depth, velocity, and channel width reported by IT Corporation (1988), and based on the HEC-2 computer model developed by the Hydrological Engineering Center of the U.S. Army Corps of Engineers. These values are listed below:

- channel width = 345 feet (105 m)
- water depth = 5.4 feet (1.65 m)

• flow velocity = 2.1 feet per second (0.64 m s<sup>-1</sup>).

#### **MONTHLY DILUTION MODEL (MD)**

Our dilution model requires that we account for dilution and transport of the material in the receiving body of water, i.e., either the Great Miami River or Paddy's Run. These calculations assume that the radionuclide concentration at the downstream receptor of interest in a receiving surface water  $(C_m)$  is equal to the radionuclide concentration at the point of radionuclide release, i.e., the radionuclide concentration at the discharge point into the Great Miami River  $(C_m)$  divided by the dilution factor (S):

$$C_m = C_m / S \tag{R-1}$$

where

 $C_m$  = radionuclide concentration in the receiving surface water (Bq m<sup>-3</sup>),

- $C_0 =$  radionuclide concentration of an effluent at the point of release (Bq m<sup>-3</sup>), and
- S = the dilution factor, a ratio of the flow rate of the receiving body of water to the flow rate of the waste effluent.

The effluent concentration  $(C_0)$  is the radionuclide release rate divided by the effluent flow rate:

$$C_0 = W_0 / Q_0 \tag{R-2}$$

where

 $W_0 =$  radionuclide release rate at the point of release (Bq s<sup>-1</sup>), and

 $Q_0 = -$  flow rate of the effluent discharge at the point of release (m<sup>3</sup> s<sup>-1</sup>).

The dilution factor, S, is based upon the river flow characteristics of the surface water body (Great Miami River or Paddy's Run), and FMPC discharge volumes from the site to the body of water. The averages of the flow rates, reported in Task 4 (Killough et al. 1993), are shown in Table D2-1.

The MD model uses a statistical uncertainty analysis computer program (CrystalBall<sup>TM</sup>) to provide bounds around our central estimates. We assumed a distribution of values for monthly discharge of uranium and monthly discharge volume. For the monthly dilution factors, we assumed a distribution of flow rates based upon daily measurements by United States Geological Survey (USGS).

For the MD and GENII model comparison, we used the uranium source term and effluent volume estimates for 1960, 1961 and 1962. (Voillequé et al. 1991). Although we estimated monthly uranium source terms for this time period, this comparison is based on annual estimates. Table D2-1 presents the source term and flow rate estimates from the Task 2 and 3 report (Voillequé et al. 1991). Using a conversion factor of  $6.8 \times 10^{-7}$  Ci U per g U. the table lists the source term in curies (Ci). Similarly, the effluent volume, recorded by the FMPC in gallons, is converted to cubic meters using the conversion factor of 264 gallons per cubic meter.



Oreat mann rever and Fauly's Run										
Year	Source Term (kg) <sup>n</sup>	Source TermEffluent Flow RateSurface Water $(kg)^n$ $(m^3 s^{-1})^n$ Flow Rate $(m^3 s^{-1})^h$		Dilution Factor (unitless)						
Gre	at Miami River									
1960	5600 (5400-5800)	0.054	53	990						
1961	7300 (7000-7600)	0.053	95	1790						
1962	6200 (5700-6700)	0.042	78	1840						
]	Paddy's Run									
1960	1300 (800-1800)	0.0033	0.059	18						
1961	1400 (1000-1600)	0.0050	0.059	12						
1962	1500 (1100-2100)	0.0072	0.059	8						
" From	Voillequé et al. 1991	; the median annual re	elease estimate with 5 <sup>th</sup>	to 95 <sup>th</sup>						

## Table D2-1. Uranium Source Term and Flow Rate Estimates for the Great Miami River and Paddy's Run

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" From Voillequé et al. 1991; the median annual release estimate with 5<sup>th</sup> to 95<sup>th</sup> percentile range.

<sup>b</sup> Average flow rate for the Great Miami River is based on daily USGS measurements, and for Paddy's Run on occasional measurements made at the site (Pennak 1973).

The mass-to-activity conversion for natural uranium is based on the fractional abundance of  $^{238}$ U in natural uranium of 0.993, and a specific activity of  $3.33 \times 10^{-7}$  Ci g<sup>-1</sup> (Shleien 1992). If we assume the decay products are in equilibrium with  $^{238}$ U, then the activities for  $^{234}$ Th and  $^{234}$ U will be equivalent. For  $^{235}$ U, the fractional abundance is 0.0072 with a specific activity of 2.16 x  $10^{-6}$  Ci g<sup>-1</sup>. The source term estimates in Ci for these radionuclides for 1960 to 1962 are listed in Table D2-2. Based on these values, we calculated the annual average uranium concentration downstream in the Great Miami River and in Paddy's Run with the RAC MD Model and with the GENII Model.

# RESULTS

The results of the comparisons for the Great Miami River are displayed in Tables D2-2 and for Paddy's Run in Table D2-3. The agreement between the models for uranium concentrations in both the river and in Paddy's Run is good, and an analysis of variance indicates no significant difference between the models. The ratio between the GENII and the MD models varies from 0.87 to 1.08 for the river, and from 0.98 to 1.08 for Paddy's Run. This agreement suggests that the methods we have developed to determine surface water concentrations of uranium and other radionuclides based on our monthly source term data are reasonably congruent with other models developed for similar purposes.

In Task 4, we also compared our model-calculated values for uranium concentrations with actual environmental sampling measurements that were done in the Great Miami River and in Paddy's Run (Killough et al. 1993). Figure R-6 in Task 4 compares monthly average uranium concentrations measured in the river with those calculated with our *RAC* model results. In a similar fashion, Figure R-7 in Task 4 shows monthly average uranium concentrations measured in Paddy's Run downstream of the FMPC for 1960, 1961 and 1962. When we compare the model-predicted (P) to observed (O) or measured concentrations in

the Great Miami River, the median P/O ratio for this period is 1.3, indicating very good agreement. The median P/O ratio for Paddy's Run is approximately 3, indicating that the MD model overpredicts the measured uranium concentrations in Paddy's Run somewhat. This overprediction is probably due to the extreme seasonal variation in flow, causing difficulty in estimating an average flow in the creek over an extended time period. These comparisons of our model-calculated uranium concentrations to those derived from the GENII code, and to the observed concentrations in the river and Paddy's Run for a threeyear period support our methods, and provide a measure of proof that our model of calculating environmental concentrations, and ultimately radiation doses, is reasonable.

	Great Miami River									
		_	U Concentration (pCi L <sup>-1</sup> )							
		Source Term	GENII	RAC	Ratio					
Year	Radionuclide	(Ci)	Model	MD Model	(GENII/MD)					
1960	<sup>238</sup> U	1.86	1.10	1.25	0.88					
	<sup>234</sup> Th	1.86	1.10	1.25	0.88					
	$^{234}$ U	1.86	1.10	1.25	0.88					
	$^{235}$ U	0.08	0.052	0.06	0.87					
1961	<sup>238</sup> U	2.35	0.80	0.74	1.08					
	<sup>234</sup> Th	2.35	0.80	0.74	1.08					
	$^{234}$ U	2.35	0.80	0.74	1.08					
	$^{235}$ U	0.11	0.037	0.036	1.03					
1962	<sup>238</sup> U	2.10	0.85	0.83	1.02					
	<sup>234</sup> Th	2.10	0.85	0.83	1.02					
	$^{234}$ U	2.10	0.85	0.83	1.02					
	<sup>235</sup> U	0.10	0.039	0.04	0.98					

Table D2-2. Model Comparisons of Radionuclide Concentrations in the
Great Miami River

 Table D2-3. Model Comparisons of Radionuclide Concentrations in

Paday's Run									
•			<u>U Concentra</u>						
		Source Term	GENII	RAC	Ratio				
Year	Radionuclide	( <u>Ci</u> )	Model	MD Model	(GENII/MD)				
1960	2:38U	0.42	230	218	1.06				
	<sup>2:34</sup> Th	0.42	230	218	1.06				
	<sup>234</sup> U	0.42	230	218	1.06				
	$^{235}U$	0.020	11	10	1.10				
1961	$^{238}$ U	0.45	240	238	1.01				
	<sup>234</sup> Th	0.45	240	238	1.01				
	$^{234}\mathrm{U}$	0.45	240	238	1.01				
	$^{235}$ U	0.022	12	12	1.00				
1962	$^{238}$ U	0.48	260	265	0.98				
	<sup>234</sup> Th	0.48	260	265	0.98				
	<sup>2:34</sup> U	0.48	260	265	0.98				
	$^{235}$ U	0.023	14	13	1.08				



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# MONITORING DATA FOR RADON IN AIR AND EXPOSURE RATE: WITH COMPARISONS TO PREDICTIONS

#### INTRODUCTION

In addition to the particulate releases from the FMPC stacks, there are two types of releases from the waste storage silos, located in the waste disposal area west of the FMPC production area, that are evaluated. First, there is the release of  $^{222}$ Rn and its short-lived daughters from the K-65 Silos, Silos 1 and 2. This release was described in our previous source term report (Voillequé et al. 1991). Second, there is gamma radiation that is emitted from the K-65 Silos and the Metal Oxide Silo, Silo 3. This gamma radiation represents a potential source of direct radiation exposure to people living near the Silos. Calculations of direct exposures from radiation emitted from the Silos are described in the Task 4 report (Killough et al. 1993) and final Tasks 2 and 3 report (in preparation) of this Project. In our previous source term work (Voillequé et al. 1991), we determined that the Metal Oxide Silo is not an important source of radon releases. However, because it contains high concentrations of radioactive materials, it does represent a potentially significant source of direct radiation exposure. Figure E-1 shows the location of the waste storage silos.



Figure E-1. Location of the waste storage silos on the west side of the FMPC site.

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In this Appendix we discuss the environmental monitoring data related to both the radon releases and the direct gamma exposures. Where these environmental data are sufficient, we also use our source term and transport models to predict levels expected in the measurements, and compare the predictions to the measurements. The results of these comparisons will be used later in this Project to make final determinations about the performance of our models. In addition, some of the data evaluated here have not been published previously, and it is important to make these data available. Of particular interest are the early radon monitoring data, for measurements taken before the K-65 Silos were sealed in 1979.

# MONITORING DATA FOR RADON IN AIR, WITH COMPARISONS TO PREDICTIONS

A number of sources of environmental monitoring data for <sup>222</sup>Rn concentration in air have been located. The earliest monitoring in the FMPC environs appears to have been initiated in 1978 by the FMPC. A set of handwritten notes (Boback circa 1984) indicates that these early measurements consisted primarily of grab samples, both of particulates to be analyzed for radon daughters and of air to be analyzed for radon. Some longer-term samples were taken using passive radon monitors. These early samples were taken at the FMPC site boundary air monitoring stations, primarily at boundary station 6, which was at the site boundary west of the K-65 Silos, and at locations very close to the K-65 Silos. The measurements continued into 1980.

Environmental monitoring data for <sup>222</sup>Rn concentrations in air are also provided in the FMPC annual environmental reports, which present the results of environmental monitoring performed by FMPC staff. Radon monitoring is first mentioned in the 1979 environmental report (Boback and Ross 1980). This report provides maximum concentrations measured during "preliminary sampling," and indicates that the methods to be used for monitoring radon were still under investigation at that time. We assume that this "preliminary sampling" and the early sampling described above are the same.

The FMPC established a routine radon monitoring program in July 1980 at the (then) six boundary air monitoring stations (Boback and Ross 1981). Alpha track detectors, configured as passive radon gas detectors and supplied by a commercial vendor, were used. The routine program was intended to provide quarterly monitoring (i.e., the detectors were to be exposed for three-month periods), although there were significant variations in actual exposure times. In 1981 the program was expanded to include sampling at the (new) seventh boundary station and two background locations (Fleming et al. 1982). The initial results of the routine radon monitoring program are included in the 1980 report. However, this report provides only the ranges of the measured concentrations. The 1981–1985 reports (Fleming et al. 1982, Fleming and Ross 1983, Fleming and Ross 1984, Facemire et al. 1985, and Aas et al. 1986) tabulate the maximum, minimum, and average concentrations measured at the seven boundary stations and at the background stations.

In 1986, the radon monitoring program was expanded slightly to include sampling at two onsite air monitoring stations (AMS 8 and AMS 9) and three offsite locations (AMS 10,

11, and 13), in addition to the seven boundary stations (then called AMS 1-7) and two background stations (WMCO 1987). Maximum, minimum, and average concentrations were reported for each monitoring station. In 1987 the program was expanded greatly to include sampling at 16 locations on the site boundary, 16 locations on the fenceline around the K-65 Silos, two other onsite locations on the west side of the production area, four background stations, a few residences near the FMPC site, and air monitoring stations AMS 1-13 (WMCO 1988). The program continued with only minor changes through at least 1990 (WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991). The 1987-1990 reports only give results for the air monitoring stations and the site boundary stations; results for the K-65 Silos fenceline and the other two onsite locations are not reported (WMCO 1988, WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991). The 1987-1990 reports provide average results only.

Additional, more detailed results have been obtained for some periods of the FMPC radon monitoring. A handwritten spreadsheet (Anonymous circa 1984) provides a compilation of the individual detector results of the routine FMPC monitoring for June 13, 1980, through December 27, 1983. Computer file copies of the FMPC alpha track monitoring data for 1987–1992 (only part of 1992) have also been received, directly from the site (Byrne 1992). These computer files include the individual measurements for locations reported in the environmental monitoring reports and also for the K-65 Silo fenceline locations. Starting in 1988, continuous radon gas monitoring has been performed on the K-65 Silos fenceline and at other locations using active, powered, flow-through instruments. Computer file copies of the data for these continuous radon monitors for 1988–1992 (again, only part of 1992) have also been received (Byrne 1992).

Environmental radon monitoring on and around the FMPC site has also been conducted by entities other than the FMPC operating contractor. The Mound facility, which is a DOE facility in Miamisburg, Ohio, established a radon monitoring network at the FMPC in September 1984 (Hagee et al. 1985). Mound used Passive Environmental Radon Monitors (PERMs) with one- to two-week exposure periods. Mound initially monitored at six onsite locations, and later expanded to 17 onsite locations at varying distances from the K-65 Silos. The monitoring was performed through early October 1986. A Mound report summarizes the results for September 20, 1984 through February 5, 1985 (Hagee et al. 1985). A letter with attached tables provides detailed results for July 2, 1985, through October 3, 1986 (Jenkins 1986). An Oak Ridge National Laboratory report (Berven and Cottrell 1987) summarizes results for the entire monitoring period.

The Ohio Department of Health performed environmental radon monitoring at 12 locations on the FMPC site boundary and at four control locations, from June 1985 to October 1989 (Steva 1988 and Anonymous circa 1989). This monitoring used alpha track detectors that were changed after 3.5 to 8 months of exposure.

In the Task 4 Report of this Project (Killough et al. 1993), we discussed the Mound monitoring data and made comparisons of those data to predictions based on our source term and dispersion model. In this Appendix, we examine the other data sets, after first reviewing the methods we use for calculating the dispersion of radon released to the air, and the resultant radon concentrations outside the FMPC.

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## Methods for Calculating Dispersion of Radon Released from K-65 Silos

Details are given in the Task 4 report of methods used to calculate both the dispersion of radon in air and radon concentrations at offsite receptors from releases from the K-65 Silos (Killough et al. 1993). Details about the quantities of radon released from the Silos, and information about the characteristics of the Silos, were initially discussed in the draft report of Tasks 2 and 3 of this Project (Voillequé et al. 1991), and final information is given in the final report of Tasks 2 and 3 (in preparation). The following is a brief review of some of the information from those reports that is pertinent to model predictions discussed later in this Appendix.

The calculations of the dispersion of radon in air from releases from the K-65 Silos are performed using the same methods as are used for particulate releases from rooftop stacks. This includes accounting for wake effects produced by the presence of the Silos and surrounding berms. Of course, the physical characteristics of radon and radon daughters are different from those of the uranium particulate materials released from processing plant stacks, and these differences are accounted for through the use of appropriate parameter values for the dispersion model. After the middle of 1979 the radon releases are estimated to consist of a continuous release component and a daylight-only release component. For calculations for the daylight-only component, special meteorology data sets are generated to represent daylight conditions.

As discussed in the reports of Tasks 2 and 3, it was determined that the K-65 Silos, Silos 1 and 2, are the only significant contributors to releases of  $^{222}$ Rn to air from the FMPC, and are thus the only sources considered. As discussed in the report of Task 4, for receptor locations outside the site, the two K-65 Silos can reasonably be treated as a single release point. For the building wake effects module of the radon dispersion model, the obstacle (the Silos and surrounding berms) is modeled with height 11 m and width 55 m.

Estimated radon release rates from the K-65 Silos are the following:

- For 1959 to mid-1979, the median release estimate is 6200 Ci y<sup>-1</sup> released continuously.
- For mid-1979 to 1987, the median release estimates are 800 Ci y<sup>-1</sup> released during daylight hours and 130 Ci y<sup>-1</sup> released continuously.

## Early Radon Monitoring Data

As indicated above, the earliest monitoring of <sup>222</sup>Rn in air around the FMPC was apparently initiated in 1978 by the FMPC. This early monitoring continued through early 1980, after which time a routine program was implemented. The majority of the information related to this early monitoring that we have located was compiled by Boback (circa 1984). This compilation is a file folder kept by Boback, which contains mostly handwritten documents, including handwritten summaries of data, Analytical Data Sheets (ADSs) of the FMPC Health and Safety Division's analytical laboratory, hand-drawn plots of data, and worksheets for the analysis of TLDs in the passive radon monitors. To our knowledge, the only data from this early work that was published was an indication of the maximum concentration, given in the annual environmental report for 1979 (Boback and Ross 1980). Some of the data was described in FMPC internal memoranda (Heatherton 1979, Ross 1979, and Ross 1980). Another internal memorandum (Boback 1979) briefly described plans for

# Appendix E Monitoring Data for Radon in Air and Exposure Rate: with Comparisons to Predictions

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radon monitoring on the site boundary. The information from this early monitoring is the only data we have located that includes radon concentrations in air prior to the mid-1979 sealing of the K-65 Silos. For this pre-sealing period, we estimate (in the final report of Tasks 2 and 3) that the radon release rate was about six times higher than after the Silos were sealed, and any corroborating (or contradictory) evidence for this higher pre-sealing release estimate is very important for our dose reconstruction work.

This early radon monitoring included measurements of three different types. First, integrated measurements of <sup>222</sup>Rn were made using passive radon monitors (Boback 1979 and Boback circa 1984). Although not specifically stated, from information in Boback (circa 1984) it appears that these monitors were the same as those commonly called Passive Environmental Radon Monitors (PERMs). The operating principle of the PERM involves, first, diffusion of radon from ambient air through a porous barrier into a sensitive volume of the instrument (George 1977). Inside the sensitive volume, the positively charged <sup>218</sup>Po ions, formed from the decay of <sup>222</sup>Rn, are collected on a negative electrode. The cumulative alpha activity collected on the electrode is detected by a thin LiF thermoluminescent dosimeter (TLD) chip, which is very sensitive to alpha radiation, but relatively insensitive to beta and gamma radiation. After exposure, the TLD chip is removed and read in a TLD analyzer. The measured cumulative alpha activity is directly proportional to the time-integrated radon concentration.

The passive radon monitors were exposed for periods from one day to three weeks. All measurements except two were made at the boundary air sampling station BS-6, on the west side of the FMPC, during the period April 1979 through January 1980. The exceptions were measurements at boundary station BS-1, on the north side of the site, and near waste pit 5, both made during May 1978. Figure E-2 shows the location of the boundary air monitoring stations BS-1 through BS-6, at which most of the measurements from this early period were made. Detailed results from these passive, integrated measurements are given in Table ES-1 ("S" for Special) at the end of this Appendix. No information has been found regarding the accuracy and precision of the specific monitors used.

The second type of radon measurement was grab samples using scintillation cells (or flasks). Scintillation cells are closed containers with the interior surfaces coated with ZnS (the scintillator) (NCRP 1988). Ambient air is drawn into the cell by the vacuum of an evacuated cell or by drawing air in with a pump, dependent on the specific type used. The information in Ross (1980) indicates that Lucas cells, which are evacuated prior to use and opened to fill by the vacuum, were used. The simplicity and reliability of Lucas cells made them appropriate for field sampling (NCRP 1988). The daughters of radon collect on the interior surfaces of the scintillation cell, and alpha radiations emitted from their decay cause scintillations in the ZnS. The actual analysis, usually performed in a laboratory, uses a photomultiplier tube mounted on the end of the cell to detect the scintillation light.

The grab samples using scintillation cells were taken between August 1978 and April 1980, although it did not appear that a regular schedule was followed. On most occasions, samples were taken at all six of the boundary air monitoring stations (BS-1 through BS-6), within about an hour of each other. On a few occasions in June through August 1979 a number of samples were taken on or very close to the K-65 Silo domes. Figure E-2 shows

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the boundary sampling locations. The detailed data are given in Table ES-2, at the end of this Appendix.

The third type of measurement was actually the measurement of radon daughters. The analytical data sheets (ADSs) in Boback (circa 1984) indicate that millipore filters were used, with a sampling time of 30 min at flow rates generally from 17 to 21 L min<sup>-1</sup>, although a few samples used lower flow rates. These ADSs also showed that gross alpha counting was performed 30, 60, 90, and 120 min after the end of sample collection. At these times after sample collection, essentially all of the <sup>218</sup>Po would have decayed, so that the counts registered would be from the only other alpha-emitting, short-lived daughter, <sup>214</sup>Po, which is formed on the filter from the decay of the previous daughters (NCRP 1988). The calculated concentrations from each counting interval were then plotted on semi-logarithmic graph paper, and the concentration at time zero after sample collection was extrapolated from a line drawn through the data. On a summary sheet, the results were then reported as <sup>222</sup>Rn concentrations, indicating that 100% equilibrium of the daughters had been assumed. We note that no decay correction was applied to account for the long sampling time (30 min) relative to the half-lives of the radon daughters; thus it was implicitly assumed that the

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sample was collected instantaneously at the end of the 30-min sampling period. The lack of this decay correction means the reported results would be systematically underbiased.

These radon daughter grab samples were taken during September and October 1978, primarily at the boundary air sampling station BS-6. A few samples were also taken on the Silo 2 dome, one sample was taken at BS-5, and one sample was taken near the clearwell, in the waste pits area. Figure E-2 shows the boundary sampling locations. The detailed data are given in Table ES-3, at the end of this Appendix.

We are interested in comparing the measured radon concentrations to predicted concentrations based on our source term and radon dispersion model. As discussed in the report of Task 4 of this Project (Killough et al. 1993), meteorology data specific to the time of this early monitoring are not available, and we use the composite FMPC meteorology data set, based on data from 1987–1991. The longer the time period considered, the more representative the composite data are. Thus, for comparisons with predicted concentrations, the integrated measurements, using the passive radon monitors, are the preferred data source, because the integrated measurements provide a much better indication of the long-term average radon concentration.

Since all but two of the integrated measurements were taken at boundary station BS-6, we focus on data for this location. The data from the integrated measurements for BS-6 are summarized in Table E-1, where we have calculated average concentrations for exposure periods when multiple passive radon monitors were used. An FMPC internal memorandum (Boback 1980) indicates that in June 1979 the gooseneck vent pipes were removed and the openings were sealed, and the metal covers for the manholes and fill pipes were gasketed and bolted shut. The measured radon concentrations at BS-6 show a significant and lasting decrease around the beginning of July 1979. We thus assume that the sealing of the Silos was completed around the end of June 1979. So far we have been unable to locate maintenance records from the FMPC to confirm this date. We have additionally calculated the average concentrations for the before-sealing and after-sealing periods, where for the before period we average samples through June 22, and for the after period we average samples from July 5 onward. Figure E-3 shows the average concentrations for individual measurement periods and the averages before and after the Silo sealing. This plot shows that a very significant decrease in the radon concentration at BS-6 occurred after the Silos were sealed.

Before comparing the average measured concentrations to our predicted concentrations, we need to subtract the background radon concentration to estimate the concentration that is due to releases from the K-65 Silos. Unfortunately, this early monitoring did not include any measurements of background radon concentrations. However, in Appendix A we compiled the background radon monitoring from the routine FMPC monitoring from 1981– 1990, and addressed the seasonal variation of background radon concentration, based on monitoring performed by Mound. Those data, from Appendix A, are the best available data to provide reasonable estimates of the background during this early monitoring.

From Appendix A, the mean of the annual average background radon concentrations around the FMPC was estimated to be 0.58 pCi  $L^{-1}$ . When different locations and different years are considered, the standard deviation of the annual averages was 0.17 pCi  $L^{-1}$ . In

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Monitoring period	Number of monitors	Exposure time (h) <sup>b</sup>	Average concentration <sup>222</sup> Rn (pCi L <sup>-1</sup> )
04/12/79-04/19/79	3	168	5.8
04/19/79-04/27/79	3	192	2.3
05/04/79-05/07/79	1	72	1.8
05/07/79-05/14/79	1	168	0.8
05/21/79-05/29/79	1	192	1.6
05/29/79-06/04/79	1	144	2.38
06/11/79-06/15/79	1	96	3.8
06/18/79-06/22/79	1	96	2.3
07/02/79-07/05/79	1	72	1.6
07/05/79-07/09/79	1	96	0.6
07/13/79-07/30/79	2	408	0.35
07/30/79-07/31/79	2	24	0.75
11/15/79-11/21/79	2	144°	0.6
11/28/79-12/05/79	2	165°	0.2 <sup>d</sup>
12/05/79-12/12/79	2	170 <sup>c</sup>	0.48
12/12/79-01/02/80	2	504°	0.25
04/12/79-06/22/79 <sup>e</sup>			2.6
07/05/79-01/02/80 <sup>e</sup>			0.36

Table E-1.	Summary o	of Integrated	Measureme	ents of <sup>222</sup>	Rn in Air
at Boun	dary Station	BS-6, from	April 1979 t	o January	y 1980 a

<sup>a</sup> Detailed results are shown in Table ES-1.

<sup>b</sup> Unless indicated otherwise, calculated in this work from the monitoring period.

<sup>c</sup> Provided in the original data in Boback (circa 1984).

<sup>d</sup> One of the results was <0.1 pCi L<sup>-1</sup>, which we assumed equal to 0.05 pCi L<sup>-1</sup> for purposes of calculating the average.

<sup>e</sup> Time-weighted averages for the before- and after-sealing periods.

Appendix A we also calculated the ratios of monthly average concentration to annual average concentration for three pseudo-background locations. For April, May, and June the average ratio was 1.0. For July, November, and December the average ratio was 0.77. We thus estimate the background concentrations for BS-6 to average  $0.58 \pm 0.28$  pCi L<sup>-1</sup> for April, May, and June, and to average  $0.45 \pm 0.22$  pCi L<sup>-1</sup> for July, November, and December, where the "±" values define a 90% confidence interval (± 1.645 standard deviations).

From the data in Table E-1, the standard error of the mean concentration before the Silo sealing is calculated to be 0.5 pCi L<sup>-1</sup> and the standard error of the mean after the sealing is 0.08 pCi L<sup>-1</sup>. Thus, 90% confidence intervals for the net concentrations can be calculated as follows. For the period before the sealing:  $(2.6 \pm 0.8) - (0.58 \pm 0.28) = 2.0 \pm 0.9$  pCi L<sup>-1</sup>. For the period after the sealing:  $(0.36 \pm 0.13) - (0.45 \pm 0.22) = -0.09 \pm 0.26$  pCi L<sup>-1</sup>.

We use our radon dispersion model (RNCHIQ4) to estimate the ratios of air concentrations of radon to release rates of radon ( $\chi/Q$ ). The distance from the center of the two K-65 Silos (recall we model the two as a single silo) to the boundary station BS-6 is

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Figure E-3. Concentrations of <sup>222</sup>Rn in air at boundary air monitoring station BS-6, before and after the penetrations in the K-65 Silos were sealed.

estimated to be 1100 ft, or 330 m, based on scaling from a detailed map of the FMPC site (WMCO 1989b). Station BS-6 is west-southwest from the K-65 Silos, so the wind direction is from the east-northeast. The results of these calculations are shown in Table E-2.

	$\chi/Q$ (pCi m <sup>-3</sup> per Ci y <sup>-1</sup> )					
Month	Continual release	Daylight-only release				
April	0.304					
Мау	0.242					
June	0.171					
July	0.112	0.141				
November	0.251	0.178				
December ·	0.288	0.267				
Average for April, May, and June	0.239					
Average for July, November, and December	0.217	0.195				

## Table E-2. Predicted $\chi/Q$ for Radon at Boundary Station BS-6, from K-65 Releases

From the values of  $\chi/Q$  and the radon release rates (see page E-4), our predicted concentrations of radon at BS-6 due to radon releases from the K-65 Silos are 1.5 pCi L<sup>-1</sup> for April, May, and June 1979, before the sealing of the Silos, and 0.18 pCi L<sup>-1</sup> for July, November, and December 1979, after the sealing. For the period before the sealing, our predicted concentration is within the 90% confidence interval about the mean measured concentration, and the predicted to observed (P/O) ratio is 0.75, indicating good agreement.

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For the period after the sealing, our predicted concentration is slightly above the 90% confidence interval about the mean measured value. However, the net measured concentration was negative, indicating that the background that we assumed was not entirely appropriate. The uncertainty associated with the application of background values from other studies is unknown. At such low concentrations, large uncertainty is expected. Thus, no definitive conclusions can be made regarding the discrepancy between the measured and predicted concentrations for the period after the Silo sealing.

We also briefly consider the radon daughter grab samples described by Boback (circa 1984). There were 15 samples taken at the boundary station BS-6 in September and October 1978 (see Table ES-3). Of these, one result was about seven times higher than all other results. If this extreme value is neglected, the average reported concentration is 1.9 pCi  $L^{-1}$ . If the extreme value is included, the mean concentration is 4.3 pCi  $L^{-1}$ . As mentioned earlier, these results were reported as concentrations of <sup>222</sup>Rn, but are really estimates of the average concentration of each of the short-lived daughters of <sup>222</sup>Rn, assuming equilibrium conditions existed. Since the measurements were only 30-min grab samples, it does not seem reasonable to compare the results directly to predicted values, which would be based on a longer time resolution. However, a qualitative comparison may be of some use.

If no short-lived daughters were released with the radon released from the K-65 Silos, we would expect the net radon daughter concentrations at BS-6 to be only about one-tenth the net concentration of radon. This is because station BS-6 is relatively close to the Silos and the transport time is short, so the ingrowth of daughters during transport is very slight. Of course, if the daughters are released in equilibrium with radon, their concentrations at BS-6 would be almost equal to that of radon (there are some losses due to deposition). The average measured daughter concentration of 1.9 or 4.3 pCi L<sup>-1</sup> is significantly above the expected background and in the range of measured concentrations of  $^{222}$ Rn. This tends to support the release of radon daughters in appreciable fractions of equilibrium concentrations. This provides some corroboration (admittedly weak) of the determination, in the final report of Tasks 2 and 3 of this Project (in preparation), that the radon daughters were released in equilibrium with  $^{222}$ Rn for this period prior to the sealing of the Silos.

In summary, these data from the early radon monitoring at the FMPC are important as the only radon monitoring data from the period before the penetrations in the K-65 Silos were sealed (in mid-1979). The integrated radon measurements, at one boundary station, BS-6, for this period before the Silos were sealed, agree well with our predicted concentrations at this location. The difference in the radon concentrations at BS-6 before and after the sealing provide strong evidence of a significantly higher radon releases prior to the sealing, and thus qualitatively corroborate the significant difference in before and after release rates determined in our source term work of Tasks 2 and 3 of this Project (Voillequé et al. 1991 and the final report, in preparation).

#### **FMPC Routine Radon Monitoring**

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As discussed earlier, the FMPC established its routine radon monitoring program in July 1980 at the six boundary air monitoring stations, then called BS-1 through BS-6. Alpha-track detectors, configured as passive radon gas detectors have been used in this

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monitoring from the beginning. The detectors are typically exposed for about three months, although variations in exposure times occur.

In 1981 the program was expanded to include sampling at the new, seventh boundary station, BS-7, and two background stations (Fleming et al. 1982). In 1986, the seven boundary air monitoring stations were renamed AMS 1 through AMS 7, and further expansion added monitoring at two onsite locations, AMS 8 and AMS 9, and three offsite locations, AMS 10, 11, and 13 (WMCO 1987). In 1987 the program was expanded greatly to include sampling at 16 locations on the site boundary, called FMPC A through FMPC P, 16 locations on the fenceline around the K-65 Silos, called K65 A through K65 P, two other onsite locations on the west side of the production area, four background stations, and a few residences near the FMPC, in addition to the air monitoring stations AMS 1-13 (WMCO 1988). The program continued with only minor changes through at least 1990 (WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991). The locations of the boundary air monitoring stations, AMS 1-7, and the two onsite air monitoring stations, AMS 8 and 9, at which radon monitoring was performed, are shown in Figure E-4. Stations AMS 1-7 are the same as the former stations BS-1 through BS-7.



Figure E-4. Locations of the boundary and onsite air monitoring stations. The boundary stations, AMS 1-7, were the primary radon monitoring locations for the FMPC routine monitoring program through 1986. These boundary stations are the same as the former stations BS-1 through BS-7.

Radiological Assessments Corporation "Setting the standard in environmental health" UUU269 Much of the available data for the routine radon monitoring has been obtained from the annual environmental monitoring reports. However, additional, more detailed results have been obtained for some periods of the FMPC radon monitoring. A handwritten spreadsheet (Anonymous circa 1984) provides a compilation of the individual detector results of the program for June 13, 1980, through December 27, 1983. Computer file copies of the FMPC alpha track monitoring data for 1987–1992 (only part of 1992) have also been received directly from the site (Byrne 1992). These computer files include the individual measurements for locations reported in the environmental monitoring reports and also for the K-65 Silo fenceline locations (not reported in annual reports). Starting in 1988, continuous, real-time (hourly results) radon gas monitoring has been performed on the K-65 Silos fenceline and at other locations using passive, scintillation cell instruments (Pylon monitors). Computer file copies of the data for these continuous radon monitors for 1988–1992 (again, only part of 1992) have also been received (Byrne 1992).

In the report of Task 4 of this Project (Killough et al. 1993), we determined that the Mound monitoring data were the best for comparisons to our predicted radon concentrations, because those data were collected at locations at a greater variety of distances from the K-65 Silos, and thus included a more complete range of radon concentrations than the other available data sets. However, the data from the FMPC routine monitoring program represent a much longer period of monitoring, and the average concentrations over a long period may be useful for comparisons to predictions. In addition, the monitoring at the K-65 Area fenceline, for 1987–1990, provides information on the change in radon concentrations that occurred after the foam layer was applied to the Silo domes (in late 1987), which we determined to cause a significant decrease in the radon releases from the Silos (Voillequé et al. 1991 and final report of Tasks 2 and 3, in preparation). These aspects of the routine monitoring of 1980–1983. Since a particular need for the real-time radon monitoring has not been identified, we do not discuss those data further.

Detailed monitoring results for 1980–1983. A handwritten spreadsheet (Anonymous circa 1984) has been obtained that provides a compilation of the individual alpha-track detector results of the routine radon monitoring program for June 13, 1980, through 1983. Although summarized results from these years of monitoring were presented in the annual environmental monitoring reports, these detailed data apparently are not readily available. The detailed radon monitoring results from this period are provided in Table ES-4.

Average boundary station concentrations. The boundary air monitoring stations, originally called BS-1 through BS-7 and later called AMS 1 through AMS 7 (see Figure E–4), provide the longest continuous record of radon monitoring around the site. The long-term average concentrations at these locations can thus be used for comparisons with predicted concentrations. The annual average radon concentrations at these boundary stations for the period 1981–1990 are given in the annual environmental monitoring reports for those years (Fleming et al. 1982, Fleming and Ross 1983, Fleming and Ross 1984, Facemire et al. 1985, Aas et al. 1986, WMCO 1987, WMCO 1988, WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991), and are shown in Table E–3. For 1981, background measurements were only made

for the last three quarters of the year (see Table ES-4). Thus, for 1981 we show the average concentrations based on these three quarters only. Average background concentrations were calculated from the data for all background locations, as presented in Appendix A. The values reported for 1984 were geometric means, but the individual results were not available; so we use the values as if they were arithmetic means.

Year	AMS 1	AMS 2	AMS 3	AMS 4	AMS 5	AMS 6	AMS 7	Average for boundary stations	Average background <sup>b</sup>
1981°	0.78	0.80	0.47	0.48	0.23	0.65	0.76	0.60	0.59
1982	0.7 <del>9</del>	0.91	0.66	0. <del>9</del> 0	0.94	1.01	1.07	0.90	0.61
1983	0.65	0.77	0.76	0.65	1.05	0.82	0.91	0.80	0.69
1984 <sup>d</sup>	0.917	0.801	0.843	0.591	0.970	0.584	0.717	0.775	0.596
1985	0.81	0.82	0.28	0.56	0.80	0.66 <sup>e</sup>	1.01	0.71	0.48
1986	0.64	0.84	0.68	0.55	0.58	0.65	0.96	0.70	0.58
1987	0.54	0.46	1.12	1.02	0.60	1.26	0.66	0.81	0.66
1988	0.7	1.0	1.2 <sup>f</sup>	0.7	1.7 <sup>f</sup>	1	1.5	1.11	0.6
1989	0.6	0.7	0.7 <sup>f</sup>	0.7	0.7 <sup>f</sup>	0. <b>9</b>	0.6	0.7	0.5
1990	0.4	0.6	0.7 <sup>f</sup>	0.4	1.1 <sup>f</sup>	0.6	0.5	0.6	0.5

# Table E-3. Annual Average Radon Concentrations (pCi L<sup>-1</sup>) at FMPC Boundary Air Monitoring Stations and Background Locations<sup>a</sup>

<sup>a</sup> Results obtained from annual environmental monitoring reports, except as noted.

<sup>b</sup> Average for all background locations, as shown in Appendix A of this report.

<sup>c</sup> Results for 1981 are based on the last three quarters of data, to be compatible with the background concentrations, which were only measured for these three quarters (see also Table ES-4).

- <sup>d</sup> The "average" values for 1984 were geometric means, but we treat them as arithmetic means.
- <sup>e</sup> This is the average of values given for two stations, BS6A and BS6B.
- <sup>f</sup> In 1988 radon monitoring at stations AMS 3 and AMS 5 ceased. However, the new location FMPC-E was the same as AMS 3, and FMPC-I was the same as AMS 5. Thus, the results from FMPC-E and FMPC-I are given here.

The average concentrations for the boundary stations and the average background concentrations are plotted in Figure E-5. In all of the annual environmental monitoring reports for 1981-1990 except for the 1989 report, the authors conclude that the average boundary concentration of radon was not significantly different from the background concentration. However, the long-term persistence of average boundary concentrations greater than average background, as seen in Figure E-5, indicates that the average boundary concentrations are significantly higher than background.

For comparisons of the measured concentrations to predicted concentrations, it seems that an average should be used, because there is great variability in the individual annual average concentrations. From our source term work in Tasks 2 and 3 (Voillequé et al. 1991), we estimated that the radon release rate from the K-65 Silos was constant from 1980 through 1987. Thus it seems reasonable to average concentrations over the period 1981-



Figure E-5. Average radon concentrations at FMPC boundary monitoring stations and background stations, from FMPC annual environmental monitoring reports. The average boundary concentrations are the average of results for stations AMS 1 through AMS 7. The average background concentrations are the average for all background locations for the given year.

1987. Table E-4 shows the annual average net radon concentrations (after subtraction of background) for the boundary stations AMS 1 through AMS 7. These values were calculated from the gross concentrations of Table E-3. The average net concentrations for the period 1980-1987 are also shown in Table E-4.

Year	AMS-1	AMS-2	AMS-3	AMS-4	AMS-5	AMS-6	AMS-7
1981ª	0.19	0.21	-0.12	-0.11 ·	-0.36	0.06	0.17
1982	0.18	0.30	0.05	0.29	0.33	0.40	0.46
1983	-0.04	0.08	0.07	-0.04	0.36	0.13	0.22
1984	0.321	0.205	0.247	-0.005	0.374	-0.012	0.121
1985	0.33	0.34	-0.20	0.08	0.32	0.19	0.53
1986	0.06	0.26	0.10	-0.03	0.00	0.07	0.38
1987	-0.12	-0.20	0.46	0.36	0.06	0.60	0.00
1988	0.10	0.40	0.60	0.10	1.10	0.40	0.90
198 <b>9</b>	0.10	0.20	0.20	0.20	0.20	0.40	0.10
1990	-0.10	0.10	0.20	-0.10	0.60	0.10	0.00
Average 1981–1987	0.13	0.17	0.0 <del>9</del>	0.08	0.14	0.20	0.27

 Table E-4. Annual Average Net Radon Concentrations (pCi L<sup>-1</sup>)

 at FMPC Boundary Air Monitoring Stations

<sup>a</sup> Results for 1981 are based on the last three quarters of data, to be compatible with the background concentrations, which were only measured for these three quarters (see also Table ES-4).

For predictions of radon concentrations, we first determine the distances and directions of the boundary monitoring stations from the K-65 Silos. The locations of these stations are shown in the annual environmental monitoring reports, and in Figure E-4. These locations were plotted on large-scale drawings of the FMPC site that included the Ohio State Plane (OSP) coordinate system (Schwarzman 1992). The approximate OSP coordinates of each location were then scaled from the drawings. The coordinate locations of the K-65 Silos were determined in the final report of Tasks 2 and 3 of this Project (in preparation). Simple trigonometric relationships were used to calculate, from the coordinates, the distances and directions of the monitoring locations from the point between the two Silos. The radon dispersion model requires as input the direction from which the wind would have to blow to expose the receptor to radon from the Silos, expressed as one of the sixteen compass directions. (As an example, exposure of a receptor to the northeast of the Silos occurs with wind blowing from the southwest.) Table E-5 shows the results of these calculations.

	OSP Coord	linates (ft)	From center of two Silos				
Location	East	North	Distance (m)	Wind from <sup>a</sup>			
Silo 1	1,378,484	480,400					
Silo 2	1,378,486	480,522					
Center of	1,378,485	480,461	•				
two Silos							
AMS 1	1,380,920	483,810	1260	SW			
AMS 2	1,383,550	484,120	1900	SW			
AMS 3	1,383,300	480,500	1470	W			
AMS 4	1,382,930	476,770	1760	NW			
AMS 5	1,378,390	477,430	920	N			
AMS 6	1,377,430	480,190	330	ENE			
AMS 7	1,376,620	483,630	1120	SSE			

# Table E-5. Estimated Coordinate Locations of Boundary Air Monitoring Stations, with Distances and Directions from the K-65 Silos

<sup>a</sup> The "wind from" direction is the directions from which the wind would have to blow to expose the receptor (the monitoring station) to radon from the Silos. This direction format is used for consistency with our radon dispersion model.

Other input parameters for the radon dispersion model (which is called RNCHIQ4) were described in an earlier section of this Appendix (see page E–4). Annual average values of the ratio of predicted radon concentration to radon release rate  $(\chi/Q)$  were calculated for both continuous release conditions and daylight-only releases. Table E–6 shows these results.

The estimated radon release rates (see also page E-4) are multiplied by the predicted  $\chi/Q$  values to predict the radon concentrations due to releases from the K-65 Silos. We then divide the predicted concentrations by the net measured concentrations for the 1981–1987 period, to form predicted to observed (P/O) ratios. The net measured concentrations, predicted concentrations, and P/O ratios are given in Table E-7.

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	From center b	etween Silos <sup>a</sup>	$\chi/Q$ (pCi m <sup>-3</sup> per Ci y <sup>-1</sup> )		
Location	Distance (m)	Wind from direction <sup>b</sup>	Continuous releases	Daylight-only releases	
AMS 1	1260	SW	0.176	0.0542	
AMS 2	1900	SW	0.127	0.0325	
AMS 3	1470	W	0.135	0.0355	
AMS 4	1760	NW	0.0908	0.0187	
AMS 5	920	Ν	0.0557	0.0406	
AMS 6	330	ENE	0.281	0.241	
AMS 7	1120	SSE	0.0385	0.0139	

Table E	-6.	Predicted	Annu	al Ave	rage ;	∕∕Q fo	or Ra	don at	Ł
Boundary	Ai	· Monitori	ing Sta	ations.	from	K-65	Silo	Relea	ses

<sup>a</sup> Relative to a point centered between the two K-65 Silos.

<sup>b</sup> The direction from which the wind would have to blow to expose the receptor (monitoring station) to radon released from the Silos.

Table E-7. Comparison of Measured and Predicted <sup>222</sup>Rn Concentrations at FMPC Boundary Air Monitoring Stations for 1981–1987, Due to Rn Releases from the K-65 Silos

	From center b	etween Silos	Rn concentrat		
Location	Distance (m)	Wind from	Net measured	Predicted	P/O ratio
AMS 1	1260	SW	0.13	0.066	0.50
AMS 2	1900	SW	0.17	0.043	0.25
AMS 3	1470	W	0.09	0.046	0.53
AMS 4	1760	NW	0.08	0.027	0.34
AMS 5	920	Ν	0.14	0.040	0.29
AMS 6	330	ENE	0.20	0.23	1.1
AMS 7	1120	SSE	0.27	0.016	0.060
GMª					0.33
GSD⁴					2.5

As seen in Table E-7, the geometric mean P/O ratio is 0.33, and the geometric standard deviation is 2.5. It is difficult to determine the reasons for the great range in P/O ratios. However, the gross measured concentrations were very close to the background measured concentrations, so the uncertainty in the difference (the net) would be quite large. In 1987, the FMPC started placing many detectors at one of the site fenceline monitoring locations, to assess analytical precision of the alpha track detectors used (WMCO 1988). In 1987, five duplicate detectors were used, and the relative standard deviation for the results of these detectors, for the four quarters of monitoring, ranged from 74% to 158% (Byrne 1992). These extremely high relative standard deviations occurred at measured concentrations of 2.7 to  $5.0 \text{ pCi L}^{-1}$ . Measurements of a similar nature do not appear to have been performed prior

to 1987. For 1988, the relative standard deviations were much lower, ranging from 16% to 48%, for measured concentrations from 0.4 to 1.9 pCi  $L^{-1}$ . In later years of the monitoring program, at least two detectors were used at each monitoring location (WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991). However, as shown by the data in Table ES-4, monitoring in 1980–1983 generally employed only one detector per location. The data do not exist to allow a thorough investigation of the analytical uncertainty associated with the alpha-track results for years prior to 1987. But, based on the information just described, it does appear that these uncertainties are substantial. Thus, the P/O ratios we have calculated (in Table E-7) also have large, unknown uncertainties that cannot be quantified.

**Radon concentrations on the K-65 Area fenceline.** In 1987, the expansion of the FMPC routine radon monitoring program included the addition of sixteen monitoring locations on the fenceline of the K-65 Area (WMCO 1988). The monitoring locations, called K65 A through K65 P, are shown in Figure E-6. Results for these locations are not provided in the annual environmental monitoring reports, but are included in the computer spreadsheet files obtained directly from the FMPC site Byrne 1992).





The K-65 fenceline monitoring generally utilized two Type F detectors and a single Type M detector at each location for each quarter of monitoring. The Type F detectors are sensitive to  $^{222}$ Rn and  $^{220}$ Rn (the latter in the thorium decay series), while the Type M detectors are sensitive to  $^{222}$ Rn only (Byrne 1992). The half-life of  $^{220}$ Rn is 55.6 s (Walker et al. 1989) (versus the 3.8 d of  $^{222}$ Rn), so it does not persist in the air. There is some  $^{232}$ Th in the K-65 and Metal Oxide Silos, but the concentrations are about 400 times lower than the concentration of  $^{226}$ Ra (final report of Tasks 2 and 3, in preparation). Thus, significant

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concentrations of  $^{220}$ Rn in air around the K-65 Silos are not expected. For this reason, and because two Type F detectors were generally used, we choose to use the results of the Type F detectors. Table ES-5, at the end of this Appendix, provides the average results for each quarter of monitoring for each location on the K-65 Area fenceline.

These K-65 Area fenceline radon concentrations are potentially useful for determining how the radon release rate from the K-65 Silos changed after the foam layer was applied to the Silo domes at the end of 1987 (see reports of Tasks 2 and 3; Voillequé et al. 1991 and in preparation). The release rate was estimated to remain essentially constant for 1988–1991, after which another change to the Silos occurred. Thus, we are interested in average concentrations for 1987 and for 1988–1991. The average K-65 Area fenceline concentrations (averaged over all locations), by year and by period, are summarized in Table E-8. In Figure E-7 the quarterly averages and the period averages are plotted.

$(pCi L^{-1})$ at the K-65 Area Fenceline					
Year	Radon concentration				
1987ª	6.3				
1988	6.5				
1989	5.1				
1990	2.5				
1991	7.3				

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<sup>a</sup> Second, third, and fourth quarters of 1987.

5.5

<sup>b</sup> Average for this time period.

1988-1991<sup>b</sup>



Figure E-7. Quarterly and long-term average radon concentrations at the K-65 Area fenceline for 1987 through 1991.

Table E-8 and Figure E-7 show the significant variability of the measured radon concentrations, both on a quarter-to-quarter and year-to-year basis. This variability adds uncertainty to the average concentration for the period before 1988, because measurements were made for only three quarters in 1987, with none prior to 1987.

## Radon Monitoring by the Ohio Department of Health

The Ohio Department of Health (ODH) also performed radon monitoring around the FMPC, from June 1985 through October 1989. Information about this monitoring and results through November 1987 are provided in an ODH report (Steva 1988) and additional results are given in a table (Anonymous circa 1989). The ODH monitoring used Terradex Type F Track-Etch detectors (a specific brand of alpha-track detector), exposed from 3.5 months to one year. The program included 12 monitoring stations on the boundary of the site, and four control (background) locations. Figure E-8 shows the locations of the boundary stations. The control stations were located within about five miles of the FMPC, with two stations northeast and two southeast from the site. Detectors were generally installed at about 3 to 4 ft above the ground. Results of the monitoring are shown in Table E-9.



Figure E-8. Locations of the Ohio Department of Health radon monitoring stations on the boundary of the FMPC. The four control locations are not shown here.

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		Onto Dep			ountoring		
Location	06/06/85– 01/14/86	01/14/86 04/29/86	04/29/86 08/12/86	08/12/86– 04/08/87	04/08/87– 11/06/87 <sup>a</sup>	11/06/87– 10/18/88	10/18/88– 10/30/89
1	0.69	0.57	0.43	0.2	0.0	0.6	0.9 <sup>b</sup>
2	0.79	0.59	1.19	1.0	0.0	1.3	$1.1^{b,c}$
3	1.89	0.35	0.47	d	0.0	0.8	0.8
4	d	0.33	4.69 <sup>c</sup>	0.2	$0.1^{b,c}$	$1.4^{b,c}$	d
5	1.04	0.45	0.89	0.2	0.4 <sup>b,c</sup>	$1.5^{b}$	$1.0^{b}$
6	0.55	0.31	0.66	0.3 <sup>c</sup>	0.0 <sup>c</sup>	0.3	$1.3^{b}$
7	0.47	0.16	0.23	0.3	0.0	0.0	0.5
8	0.52	0.45	0.33	0.1	0.0	0.0	0.5
9	0.47	0.14	0.40	0.1	0.0	0.3	$0.3^{b}$
10	0.17	0.21	0.82	0.2	0.0	b,c	d
11	0.63	0.28	0.26	0.2	0.0	d	d
12	0.47	0.26	0.55	0.1	0.0	b,c	е
13⁄	1.31	0.95	0.28	$0.3^{b,c}$	0.0 <sup>c</sup>	0.0	0.5
14⁄	0.70	0.38	0.35	0.6	0.0	0.4	0.4 <sup>b</sup>
15⁄	0.41	0.11	0.09	0.2	0.0	0.2	d
16⁄	0.37	0.33	0.28	0.5	0.0	0.4	d

Table E-9. Radon Concentrations (pCi L<sup>-1</sup>) around the FMPC from the Ohio Department of Health Monitoring

<sup>a</sup> The detectors for this period were analyzed with less analytical sensitivity, so the numerous zeros do not reflect a true decrease in radon levels (Steva 1988).

<sup>b</sup> The detector was damaged or the filter paper was punctured.

<sup>c</sup> The detector was found on the ground at the end of the exposure period.

<sup>d</sup> The detector was missing at the end of the exposure period.

<sup>e</sup> This detector was removed.

f Control (background) locations.

For a number of reasons, we consider the data from this ODH radon monitoring to be less desirable than the data from the FMPC routine monitoring program. First, as seen in Table E-9, many of the detectors were damaged or found on the ground, making the results questionable. The paper filters are designed to exclude dust and radon daughters from the detector's sensitive volume, and when this filter is damaged, the measurement may include contributions from radon daughters that enter the detector, or from other alpha-emitting radionuclides. Second, only a single detector was used at each location for each monitoring period. This significantly increases the uncertainty of the measured concentrations. In addition, the monitoring locations are generally similar to those of the FMPC routine program, but the ODH monitoring was only for 4.5 years (versus about 10 years for the FMPC program). We thus consider the data from the FMPC routine monitoring to be more useful, and we perform no further analyses with these ODH monitoring data.

## Conclusions

The early radon monitoring data, from 1978–1980, are very important in relation to the estimated radon release rates for the 1959 to mid-1979 period. These early data appear to be the only environmental radon monitoring performed prior to the sealing of the K-65 Silos in

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mid-1979. The estimated radon release rate from the K-65 Silos for the period 1959 to mid-1979 (from our source term work in the final report of Tasks 2 and 3, in preparation) is about seven times higher than the estimated release rate for the period mid-1979 to 1987, after the Silos were sealed. Because the estimated release rate for this earlier period is much higher than later periods, it is especially important to have corroborating environmental data. From our analysis of the integrated radon measurements from April, May, and June 1979, the radon concentrations in air at the boundary station BS-6 prior to the sealing of the Silos agree well with our predicted concentrations. The data also show a significant decrease in radon concentration after the sealing. Thus, these data provide strong evidence for the general magnitude of our estimated release rate, and for the release rate being significantly lower after the Silos were sealed.

The grab measurements of radon daughters at boundary station BS-6 taken in September and October 1978 had an average concentration of 1.9 or 4.3 pCi L<sup>-1</sup> (dependent on whether the extreme value is excluded or included). This concentration was significantly greater than the expected background, and is thus assumed to be due to releases from the K-65 Silos. The average is also in the range of the average radon concentration before the Silos were sealed. This general agreement between radon daughter and radon concentrations provides some corroboration of our determination in the source term work (in the final report of Tasks 2 and 3, in preparation) that for the period prior to the sealing of the Silos, radon daughters were released in equilibrium with  $^{222}Rn$ .

For the period mid-1979 to 1987, we have made comparisons of predicted radon concentrations in air to measured concentrations for two data sets: (1) the monitoring performed by the Mound facility in 1985 and 1986 (in our report of Task 4, Killough et al. 1993), and (2) the FMPC routine monitoring at boundary air monitoring stations. In both of these comparisons, the predicted and measured concentrations agree relatively well, considering the significant uncertainties in the radon release rates, air dispersion model, and in the measurements. The comparisons did show some underbias in our predicted concentrations. The results of these comparisons will be used later in this Project for final determinations about the performance of our radon dispersion model.

Data for radon concentrations measured on the fenceline around the K-65 Area in the FMPC monitoring program, from 1987 through 1991, were also presented. Because these measurements bracket the end of 1987, when the foam layer was applied to the K-65 Silo domes, they may be useful for our development of the radon release rate for 1988 (in the final report of Tasks 2 and 3, in preparation).

# MONITORING DATA FOR EXPOSURE RATE, WITH COMPARISONS TO PREDICTIONS

Only a few sources of environmental monitoring of penetrating radiation exposure rates have been located. The most obvious source is exposure rate monitoring reported in FMPC environmental reports. External radiation monitoring was initiated in late 1975, using thermoluminescent dosimeters (TLDs) at locations along the FMPC site boundary (NLCO 1976). The first results of this penetrating radiation monitoring were presented in the 1976

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annual report (Boback et al. 1977), which reported the minimum, maximum, and annual average exposure rate for each of the six boundary air monitoring stations, BS-1 to BS-6, based on quarterly TLD measurements. The monitoring continued at the same locations through 1980 (Boback et al. 1978, Boback and Ross 1979, Boback and Ross 1980, and Boback and Ross 1981). In 1981, a new air monitoring station was added, BS-7, and exposure rate monitoring was also extended to this location (Fleming et al. 1982). This boundary monitoring continued unchanged through 1990, although four offsite locations were added in 1985, background measurements were added in 1986, and measurements at two additional onsite air monitoring stations, AMS 8 and AMS 9, were added in 1987 (Fleming and Ross 1983, Fleming and Ross 1984, Facemire et al. 1985, Aas et al. 1986, WMCO 1987, WMCO 1988, WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991).

In 1957, a survey of gamma exposure rates around the K-65 Silos was performed, to provide background information about potential personnel exposures that might result from the construction of an additional waste storage tank in the K-65 area (Ross 1957). Measurements were made at regular intervals in eight compass directions from each of the K-65 Silos, but only out to maximum distances of 320 ft or less.

In 1986 and 1987, exposure rate surveys were performed along Paddy's Run Road, along the west side of the FMPC site, near the K-65 Silos. We have obtained daily survey forms for the June 1987 measurements (FMPC 1987) and monthly spreadsheet summaries of the daily measurements for all of 1987 (Anonymous circa 1987). As far as could be determined, these data have not been published by FMPC.

From September 1985 through September 1986, the Ohio Department of Health (ODH) also performed exposure rate monitoring around the FMPC (Steva 1988). This monitoring utilized TLDs at 31 monitoring stations around the FMPC boundary, eight control (background) locations within about five miles of the site, and one control location in Columbus, Ohio. The TLDs were exposed for six-month periods. For the first six-month monitoring period, only eight of the 80 TLDs installed had results that should be used. Most of the results were "below minimum measurable quantity," and a few others are invalid due to damage to the detector or because the TLD was found on the ground. We believe that the uncertainty in results of this monitoring are much too great to use the results for any quantitative comparisons. However, we do note that Steva (1988) concludes that the exposure rates on the western boundary of the FMPC, nearest the K-65 Silos, may have been slightly elevated compared to the other monitoring locations.

In the Task 4 Report of this Project (Killough et al. 1993), we discussed the Paddy's Run Road survey data and made comparisons of those data to predictions of our models. In this Appendix, we examine the data sets from the FMPC routine monitoring and from the 1957 survey, after first summarizing the methods we use for direct exposure calculations. The data from the ODH monitoring are not considered further.

## Methods for Calculating Direct Exposures Due to Silos Sources

Details of the methods to be used for calculations of direct exposures and doses from gamma radiation emitted from the waste storage silos are given in the report of Task 4 of this Project (Killough et al. 1993). For information related to model predictions that are discussed later in this Appendix, a brief summary of some of the information from that report follows.

The K-65 Silos, Silo 1 and Silo 2, and the Metal Oxide Silo, Silo 3, are considered the only FMPC sources of radiation that are significant for offsite, direct exposures of people. We chose to use a readily available computer software package, MicroShield 4 (Negin and Worku 1992), to calculate exposure rates due to radioactive material in the three Silos. For evaluating sources within the Silos, we model the contents as cylindrical sources. Figure E-9 shows the source and shield geometries to be used. For calculations for offsite receptors, which are all more than 1000 ft from the Silos, the two K-65 Silos can be treated as a single source, but with height twice the physical height. For calculations for receptors closer to the Silos, this does not necessarily hold.



Figure E-9. Source and shield geometry models used for estimation of direct exposure rates from the K-65 and Metal Oxide Silos. For offsite receptors, the two K-65 Silos are modeled as a single Silo, but with twice the actual height.

A number of parameters needed to perform the MicroShield calculations, describing the geometry, some properties of the sources, properties of the shielding, and fineness of the numerical integration, apply to all the calculations. The values to be used for these parameters are shown in Table E-10. Also required as input are parameters describing the

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Cylinder geometry		Source properties		Sh				
designation	height (ft)	radius (ft)	material	density (g cm <sup>-3</sup> )	material	thickness (in)	density (g cm <sup>-3</sup> )	quadrature order <sup>a</sup>
·····			K-65 Silos	before Ber	ms Added			
dome head space	18.67 <sup>b</sup>	28.5	air	0.001293	concrete	9.805	2.35	10, 10, 10
cylinder air space	10.8 <sup>6</sup>	40	air	0.001293	concrete	8	2.35	10, 10, 10
waste	$42.6^{b}$	40	concrete	variable	concrete	8	2.35	10, 10, 10
			K-65 S	Silos with I	Berms			
dome head space	18.67 <sup>b</sup>	28.5	air	0.001293	concrete	9.805	2.35	10, 10, 10
			Me	tal Oxide S	Silo			
waste	31.4	40	concrete	variable	concrete	8	2.35	10, 10, 10

<sup>a</sup> Integration quadrature orders for radial, circumferential, and axial directions.

<sup>b</sup> As mentioned in the text, this height is twice the physical height, to allow the treatment of the two K-65 Silos as a single Silo, for offsite receptors. This does not apply to receptors at very close distances, or to the Metal Oxide Silo.

## 1957 Survey around the K-65 Silos

As indicated above, the 1957 survey around the K-65 Silos was conducted because of radiation safety concerns related to the addition of another waste storage tank in the K-65 Area. The memorandum of Ross (1957) reports few details about the methods used in the survey. The measurement method is not given, and the exact date of the survey is not given. However, there is indirect information about the date of the survey. The memorandum indicates that the proposal to build another tank was made in a letter dated June 11, 1957. The memorandum (Ross 1957) was dated July 17, 1957. Thus, the survey was made between these two dates.

Figure E-10 shows the approximate locations of the survey measurements, except for those closer than 10 ft from the Silos. This figure is based only on the directions and distances given by a table in Ross (1957). Ross refers to a drawing that was attached, but that drawing was not attached to the copy that we located. The measurement results are shown below in Table E-11.

For comparisons of predicted exposure rates to these measured exposure rates, we are most interested in the measurement locations farthest from the Silos, since those are more representative of the offsite members of the public with which we are ultimately concerned. In addition, the MicroShield documentation (Negin and Worku 1992) warns that the point kernel model used in MicroShield should be considered approximate for receptor points

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Appendix E Monitoring Data for Radon in Air and Exposure Rate: with Comparisons to Predictions





points "close" to the source. For these reasons, we choose, somewhat arbitrarily, to only use the measurements at distances of 10 ft or greater for comparisons with model predictions.

As can be seen in Figure E-10, some of the survey locations are totally hidden from one of the Silos (hidden behind the other Silo). For these cases, we consider the exposure rate to be due only to the Silo in view. There are also some survey locations that are partly hidden from one of the Silos. For these locations, an additional geometry factor would be needed to account for the partial shielding. As we are uncertain about the precision with which the survey locations were determined (the memorandum of Ross (1957) does not discuss this), we choose to eliminate these locations with partial shielding from consideration for model comparisons. The two symbols used in Figure E-10 differentiate between measurement locations for which we do and do not perform model comparisons. Out of the 139 locations for which measurements were made, we are left with 52 for which we make model comparisons.

For the MicroShield calculations, we first gather information for input parameters required. Table E-10, presented earlier in this Appendix, shows a number of the parameters

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	Direction from the Silo							
Distance (ft)	N	NE	E	SE	S	SW	W	NW
	Meas	suremen	its arou	nd Sout	n Tank	[Silo 1]		
contact	7.5	8.9	7.2	8.2	7.5	8.5	8.0	9.1
1	7.0	8.0	7.2	8.0	7.0	7.5	7.7	8.8
3	7.0	7.5	5.5	7.0	6.8	6.3	6.0	8.2
5	7.0	6.9	4.2	6.1	6.1	5.7	5.0	6.5
10	6.0	5.5	3.0	4.5	4.5	4.3	3.5	5.2
20	5.5	3.8	2.0	2.9	3.0	2.7	2.8	3.5
40		2.5	1.9	1.7	1.6	1.6	1.8	2.5
80		1.3	1.0	0.7	0.7	0.8	0.7	1.5
160		0.7	0.4	0.3	0.3	0.3		0.7
320		0.2	0.1					
	Meas	suremen	its arou	nd Nortl	h Tank	[Silo 2]		
contact	4.8	4.8	5.5	6.5	6.8	6.8	6.3	<b>5.8</b> ·
1	4.5	4.5	5.3	6.0	6.3	6.2	6.2	5.2
3	4.0	4.2	4.6	5.8	6.3	5.8	5.2	4.8
5	3.8	3.7	3.9	5.0	6.0	5.0	4.2	4.2
10	3.0	2.7	3.3	4.2	5.5	3.8	3.5	3.2
20	2.0	1.8	2.2	3.5	5.0	3.0	2.2	1.9
40	1.2	1.5	1.3	2.5		2.4	1.2	1.0
80	0.5	<b>0.6</b>	0.7	1.6		1.6	0.6	0.5
160			0.5	0.7		0.7		0.2
320			0.1	0.3		0.2		

# Table E-11. Exposure Rate Measurements (mR h<sup>-1</sup>) from1957 Survey around the K-65 Silos

to be used. Because the receptor points are fairly close to the Silos, it is not reasonable to treat the two K-65 Silos as a single Silo. Thus, the heights of the cylinders used to model the K-65 Silos are different from those given in Table E-10.

These exposure rate measurements were made either in June or July of 1957, which was before the filling of the Silos was completed. Thus, we have to account for Silo 2 being only partially full at the time of the survey. In the final report of Tasks 2 and 3 (in preparation) of this Project we discuss a procedure to estimate the fractional filling of the K-65 Silos as a function of time. Based on information presented in that report, filling of Silo 1 would have been completed well before 1957. The filling of Silo 2 is estimated to have proceeded at a uniform rate between June 1953 and September 1958, a total of 63 months. We assume that this 1957 survey occurred around July 1, 1957, which would be 48.5 months into the filling of Silo 2. This results in a silo filling factor (the fraction of the maximum material emplaced) of 77% for Silo 2 at the time of the survey. This is applied to the height of the cylinder used to model the K-65 material in Silo 2, and then the height of the cylinder head space is adjusted to compensate. For Silo 1, the heights to be used are 9.33 ft for the dome head space cylinder, 5.4 ft for the cylinder head space, and 21.3 ft for the K-65 waste material cylinder, which are all just the physical values for a single silo. For Silo 2, the

heights to be used are 9.33 ft for the dome head space cylinder, 10.3 ft for the cylinder head space, and 16.4 ft for the K-65 waste material cylinder.

The densities and moisture content of the K-65 and metal oxide materials to be used are those given in the final report of Tasks 2 and 3 of this Project (in preparation). For the K-65 material, these are a density of 0.85 g cm<sup>-3</sup> and moisture content of 56% dry weight. The moisture content is translated into a volumetric water content of 0.476 g cm<sup>-3</sup>, for input into MicroShield. For the metal oxide material, in Silo 3, the density is 0.64 g cm<sup>-3</sup> and the moisture content is negligible.

The final report of Tasks 2 and 3 of this Project (in preparation) also discusses the radionuclide concentrations in the K-65 and metal oxide materials, based on measurements of samples obtained in 1989 and 1991. For the K-65 material, that report combines the two K-65 Silos to determine average concentrations that can be applied when the two Silos are modeled as a single source. That is not done for these comparisons, so the concentrations to be used are the averages for each individual Silo. The report of Tasks 2 and 3 does provide average concentrations for each Silo, but they are not converted to volumetric concentrations as required for MicroShield. We perform the conversion by multiplying the mass concentrations by the material density to obtain volume concentrations. These concentrations, to be used for comparisons of the 1957 survey, are shown in Tables E-12 and E-13. The concentrations for the metal oxide material are those reported in the report of Tasks 2 and 3, and are shown in Table E-14.

Radionuclide	Concentration (µCi cm <sup>-3</sup> )	Radionuclide	Concentration (µCi cm <sup>-3</sup> )	Radionuclide	Concentration (µCi cm <sup>-3</sup> )
<sup>227</sup> Ac 228Ac <sup>212</sup> Bi <sup>214</sup> Bi	$7.18 \times 10^{-3} \\ 7.29 \times 10^{-4} \\ 7.29 \times 10^{-4} \\ 4.46 \times 10^{-1}$	212P0 214P0 216P0 218P0	$\begin{array}{c} 4.66 \times 10^{-4} \\ 4.46 \times 10^{-1} \\ 7.29 \times 10^{-4} \\ 4.46 \times 10^{-1} \end{array}$	228Th 230Th 231Th 232Th	$7.29 \times 10^{-4} \\ 5.88 \times 10^{-2} \\ 3.89 \times 10^{-5} \\ 7.29 \times 10^{-4}$
<sup>231</sup> Pa <sup>234</sup> Pa <sup>234m</sup> Pa <sup>212</sup> Pb <sup>214</sup> Pb	$7.18 \times 10^{-3} \\ 7.12 \times 10^{-7} \\ 5.48 \times 10^{-4} \\ 7.29 \times 10^{-4} \\ 4.46 \times 10^{-1}$	<sup>224</sup> Ra 226Ra 228Ra 220Rn 222Rn	$7.29 \times 10^{-4}$ $4.46 \times 10^{-1}$ $7.29 \times 10^{-4}$ $7.29 \times 10^{-4}$ $4.46 \times 10^{-1}$	<sup>234</sup> Th <sup>208</sup> Tl <sup>234</sup> U <sup>235</sup> U <sup>238</sup> U	$5.48 \times 10^{-4}$ $2.62 \times 10^{-4}$ $7.32 \times 10^{-4}$ $3.89 \times 10^{-5}$ $5.48 \times 10^{-4}$

Table E-12. Radionuclide Concentrations in K-65 Material of Silo 1: for Use in MicroShield Calculations

Concentrations of radionuclides in the head space of the K-65 Silos are also given in the final report of Tasks 2 and 3. For 1957, the concentrations to be used are  $2.4 \times 10^{-3} \,\mu\text{Ci cm}^{-3}$  of  $^{222}\text{Rn}$ , and  $2.4 \times 10^{-3} \,\mu\text{Ci cm}^{-3}$  of each of the short-lived daughters,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , and  $^{214}\text{Po}$ .

Distances from the centers of the Silos to the measurement points are also needed. The final report of Tasks 2 and 3 provides coordinate locations, in the Ohio State Plane system (OSP), for the three Silos, and these coordinates are repeated in Table E-15. The distances of the survey points given by Ross (1957), and shown in Table E-11, are assumed to be distances from the outer wall of the Silo. From these distances from the wall of the Silo and

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Radionuclide	Concentration (µCi cm <sup>-3</sup> )	Radionuclide	$\begin{array}{c} Concentration \\ (\mu Ci\ cm^{-3}) \end{array}$	Radionuclide	Concentration (µCi cm <sup>-3</sup> )
227Ac 228Ac 212Bi 214Bi 231Pa 234Pa 234Pa 234mPa 212Pb 214Pb	$5.72 \times 10^{-3}$ $1.25 \times 10^{-3}$ $1.25 \times 10^{-3}$ $2.54 \times 10^{-1}$ $5.72 \times 10^{-3}$ $1.26 \times 10^{-6}$ $9.72 \times 10^{-4}$ $1.25 \times 10^{-3}$ $2.54 \times 10^{-1}$	212Po 214Po 216Po 218Po 224Ra 226Ra 228Ra 220Rn 2222Rn	$\begin{array}{c} 8.01\times10^{-4}\\ 2.54\times10^{-1}\\ 1.25\times10^{-3}\\ 2.54\times10^{-1}\\ 1.25\times10^{-3}\\ 2.54\times10^{-1}\\ 1.25\times10^{-3}\\ 1.25\times10^{-3}\\ 2.54\times10^{-1}\\ 2.54\times10^{-1}\\ \end{array}$	228Th 230Th 231Th 232Th 234Th 208Tl 234U 235U 235U 238U	$\begin{array}{c} 1.25\times10^{-3}\\ 5.56\times10^{-2}\\ 7.86\times10^{-5}\\ 1.25\times10^{-3}\\ 9.72\times10^{-4}\\ 4.51\times10^{-4}\\ 1.03\times10^{-3}\\ 7.86\times10^{-5}\\ 9.72\times10^{-4} \end{array}$

 Table E-13. Radionuclide Concentrations in K-65 Material of Silo 2:

 for Use in MicroShield Calculations

Table E-14. Radionuclide Concentrations in Metal Oxide Material of Silo 3: for Use in MicroShield Calculations

Radionuclide	$\begin{array}{c} Concentration \\ (\mu Ci\ cm^{-3}) \end{array}$	Radionuclide	$\begin{array}{c} Concentration \\ (\mu Ci\ cm^{-3}) \end{array}$	Radionuclide	$\begin{array}{c} Concentration \\ (\mu Ci\ cm^{-3}) \end{array}$
227Ac 228Ac 212Bi 214Bi 231Pa 234Pa 234mPa 212Pb 214Pb	$\begin{array}{c} 3.72 \times 10^{-4} \\ 5.01 \times 10^{-4} \\ 5.01 \times 10^{-4} \\ 1.90 \times 10^{-3} \\ 3.57 \times 10^{-4} \\ 1.25 \times 10^{-6} \\ 9.60 \times 10^{-4} \\ 5.01 \times 10^{-4} \\ 1.90 \times 10^{-3} \end{array}$	212Po 214Po 216Po 218Po 224Ra 226Ra 228Ra 220Rn 2222Rn	$\begin{array}{c} 3.21\times10^{-4}\\ 1.90\times10^{-3}\\ 5.01\times10^{-4}\\ 1.90\times10^{-3}\\ 5.01\times10^{-4}\\ 1.90\times10^{-3}\\ 5.01\times10^{-4}\\ 5.01\times10^{-4}\\ 1.90\times10^{-3}\\ \end{array}$	228Th 230Th 231Th 232Th 234Th 208Tl 234U 235U 238U	$\begin{array}{c} 5.01\times10^{-4}\\ 3.28\times10^{-2}\\ 6.39\times10^{-5}\\ 5.01\times10^{-4}\\ 9.60\times10^{-4}\\ 1.80\times10^{-4}\\ 9.46\times10^{-4}\\ 6.39\times10^{-5}\\ 9.60\times10^{-4}\\ \end{array}$

Table E-15. Approximate CoordinateLocations of the Waste Storage Silos

	OSP Coor	dinates (ft)
Silo	East	North
Silo 1 (K-65)	1,378,484	480,400
Silo 2 (K-65)	1,378,486	480,522
Silo 3 (Metal Oxide)	1,378,492	480,730

the coordinate locations, the distances of the survey points from the centers of the three Silos are easily calculated using trigonometric relations. Table ES-6, at the end of this Appendix, shows the calculated distances to the centers of the three Silos.

Using the input parameters described above, exposure rate calculations were performed with MicroShield. Preliminary calculations were done to assess the importance of exposures

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from the metal oxide material in Silo 3. Results indicate that for the survey point closest to Silo 3, where the relative contribution of Silo 3 would be maximized, the exposure rate due to Silo 3 is predicted to be only 4.5% of the exposure rate due to the two K-65 Silos. This small contribution is not significant for the comparisons in which we are interested, so exposure rates due to Silo 3 were neglected in further calculations. Detailed results of the predicted exposure rates due to the three different sources (K-65 material, head space in cylinder, and head space in dome) of the two K-65 Silos are shown in Table ES-7, at the end of this Appendix. Summarized results for the survey locations are given in Table E–16.

						-		
	Direction from the Silo							
Distance (ft)	N	NE	E	SE	S	SW	W	NW
		Survey	Locatio	ns arou	nd Silo	1		
10	19.2	17.0			14.5			16.8
20	17.6	13.7	11.9		11.0		11.9	13.5
40		8. <b>93</b>	6.72		5.8 <del>9</del>		6.68	8.68
80		4.75	2.76		2.19		2.73	4.50
160		1.50	0.89		0.63			1.45
320		0.30	0.22					
		Survey	Locatio	ns arou	nd Silo	2		
10	9.23			13.3	17.3	13.5		
20	6.64		8.26	11.1	17.6	11.3	8.29	
40	3.42		4.76	8.22		8.65	4.83	
80	1.28		2.19	5.26		5.70	2.23	
160			0.79	1.76		1.86		
320			0.20	0.34		0.34		

# Table E-16. Predicted Exposure Rates $(mR h^{-1})$ for Locations of 1957 Survey around the K-65 Silos

The 1957 survey apparently did not include measurement of the background exposure rate (Ross 1957). However, the background exposure rate around the FMPC site has recently been estimated to be roughly  $0.01 \text{ mR h}^{-1}$  (Byrne et al. 1991 and others). Since the lowest measured exposure rates were 0.1 mR  $h^{-1}$ , subtraction of the background exposure rate would not significantly change the measured values. Therefore, we neglect contributions of background.

For comparison of the predicted to measured exposure rates, we form predicted to observed (P/O) ratios. These P/O ratios are shown in Table E-17. For the survey locations around Silo 1, the geometric mean (GM) P/O ratio is 3.1, with geometric standard deviation (GSD) 1.3. For those around Silo 2, the GM P/O ratio is 2.9, with GSD 1.4. For all locations (around both Silos), the GM P/O ratio is 3.0, with GSD 1.3. The P/O ratios are plotted against the measured exposure rate in Figure E-11.

The plot in Figure E-11 indicates a trend in P/O ratios with measured exposure rates. At the lowest exposure rates, which occur at the greatest distances from the Silos, the P/O ratios are generally between 1 and 2.5, compared to the typical P/O ratios of between 3 and

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Distance (ft)	Direction from the Silo							
	N	NE	Е	SE	S	SW	W	NW
		Survey	Locatio	ns arou	nd Silo	1		
10	3.2	3.1			3.2			3.2
20	3.2	3.6	6.0		3.7		4.3	3.9
40		3.6	3.5		3.7		3.7	3.5
80		3.7	2.8		3.1		3.9	3.0
160		2.1	2.2		2.1			2.1
320		1.5	2.2					
		Survey	Locatio	ns arou	nd Silo	2	_	
10	3.1			3.2	3.1	3.6		
20	3.3		3.8	3.2	3.5	3.8	3.8	
40	2.8		3.7	3.3		3.6	4.0	
80	2.6		3.1	3.3		3.6	3.7	
160			1.6	2.5		2.7	•	
320			2.0	1.1		1.7		

# Table E-17. Predicted to Observed (P/O) Ratios for 1957 Exposure Rate Survey around the K-65 Silos

4 at greater measured exposure rates. From Table E-17, there does not appear to be any significant trend in P/O ratios with direction from the Silos. And as seen in the plot, there is no significant difference between the survey points around Silo 1 and those around Silo 2.

These comparisons of predicted to measured exposure rates for the 1957 survey around the K-65 Silos indicate that our predicted exposure rates are overbiased relative to the survey measurements, and the degree of overbias seems to decrease with decreasing exposure rate, which occurs for increasing distance from the Silos. Sources of the bias and trend are not known. Contributing factors might include inaccuracies in our source term, inaccuracies of the way we model the Silo sources, inaccuracies in the MicroShield software, and inaccuracies in the measurements. The accuracy of the measurements is unknown. The memorandum describing the survey results (Ross 1957) does not indicate how the measurements were made. It is well known that the responses of different gamma radiation survey instruments can vary over an order of magnitude because of nonlinear energy response and different energy response curves for different detector types. At the closer distances to the Silos, an additional concern is the directional response of the measurement method. At close distances, the radiation field will consist of photons from many different directions, rather than a parallel beam of photons.

For perspective on the importance of direct exposures due to the K-65 Silos, we calculated the exposure rate at a distance of 1100 ft from the Silos, which is about the distance to the closest point on the western boundary of the FMPC. At this point, the predicted exposure rate for the estimated conditions at the time of the 1957 survey would be  $6.8 \ \mu R h^{-1}$ , above background. The dose to a maximally exposed individual spending the


Figure E-11. Comparison of predicted to measured exposure rates for 1957 survey around the K-65 Silos. The P/O ratios are the predicted exposure rate divided by the measured exposure rate. The geometric mean P/O ratio is 3.0, with geometric standard deviation 1.3.

whole year at this point could then be about 60 mR  $y^{-1}$ . Of course, doses would decrease very quickly with increasing distances from the Silos.

#### **FMPC Routine Exposure Rate Monitoring**

As discussed earlier, the monitoring of penetrating radiation exposure rates around the FMPC was initiated in late 1975, with the results for 1976 being the first presented in the annual environmental monitoring reports. The monitoring was performed using quarterly exposures of thermoluminescent dosimeters (TLDs). The summary data from this program for 1976-1990 were obtained from the FMPC annual environmental monitoring reports (Boback et al. 1977, Boback et al. 1978, Boback and Ross 1979, Boback and Ross 1980, Boback and Ross 1981, Fleming et al. 1982, Fleming and Ross 1983, Fleming and Ross 1984, Facemire et al. 1985, Aas et al. 1986, WMCO 1987, WMCO 1988, WMCO 1989a, Dugan et al. 1990, and Byrne et al. 1991). This monitoring was performed at the stations of the FMPC air sampling program. From 1976 through 1985, the stations monitored for exposure rates were primarily on the FMPC boundary, and were then called boundary stations (abbreviated BS). Starting in 1986, the stations were renamed air monitoring stations (AMS), reflecting the expansion of the monitoring to locations not on the site boundary. Figure E-12 shows the locations of the onsite monitoring. In this Figure and the rest of this section the monitoring stations are referred to using the AMS abbreviation. The locations of stations BS-1 through BS-7, used through 1985, are the same as the replacement stations AMS-1 through AMS-7, respectively.

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The annual average exposure rates from 1976–1990, from the annual environmental monitoring reports, are given in Table E–18. For comparisons with predicted exposure rates, we first focus on the data from stations AMS-1 through AMS-7 and the background locations. We eliminate stations AMS-8 and AMS-9 from consideration because they are close enough to the production area that they may be influenced by radiation sources in the production area. Stations AMS-10 through AMS-13 are all far enough from the Silos that they should not be influenced by radiation from the Silos, but only three years of data are available, so their usefulness as background is hard to assess. The data from stations AMS-1 through AMS-7 and the background are plotted in Figure E–13.

A few important features can be observed from Figure E-13. The exposure rates at AMS-6 are clearly elevated above other locations for all years of the monitoring, although the difference is greater for years after 1979. These results are expected, since AMS-6 is the closest monitoring station to the waste storage silos. Thus, station AMS-6 is the only location for which we will compare predicted exposure rates.

Next, the results for the other boundary stations, AMS-1 through AMS-5 and AMS-7, show that no single station is clearly distinguishable from the others. Among these six stations, five had the lowest and four had the highest exposure rates at different times. In addition, for the six years of the background monitoring, the exposure rates at the

					Air	monito	ring stat	ion (AM	IS) numl	er <sup>a</sup>				
Year	1	2	3	4	5	6	7	8	9	10	11	12	13	BKG <sup>b</sup>
1976	8	10.	10.	9	9	12								
1977	9	9	10.	10.	9	12								
1978	8	9	10.	9	9	12								
1979	9	10.	9	9	-8	15								
1980	10.	11	12	11	11	19								
1 <b>98</b> 1	10.	12	12	11	11	18	12							
1982	10.	12	12	11	12	19	12							
1983	11	12	12	11	12	18	13							
1 <b>984</b>	9.7	10.3	10.3	9.7	10.5	15.5	11.4							
1985	10.78	11.51	11.06	11.10	11.10	16.95	12.44							11.43 <sup>c</sup>
1986	8.9	<del>9</del> .3	8. <del>9</del>	8.7	9	13. <b>6</b>	8.7							9 <sup>d</sup>
1987	7.71	8.78	8.07	8.21	8.02	12.54	8.44							8.05 <sup>e</sup>
1988	9.38	10.62	10.34	9.58	9.50	15.30	9.72	9.23	12. <b>97</b>	8.09	10.57	9.02	8.47	9.91/
1989	11.71	11.49	12.49	10.6	11.02	16.81	11.33	11.29	14.01	9.13	9.46	8.80	8.73	10.04
1990	7.3	7.6	7.2	7.3	7.1	13	7.1	6.9	9.4	5.4	7.1	6.8	6.3	6.3 <sup>g</sup>

Table E-18. Annual Average Exposure Rates from FMPC Routine, Quarterly TLD Monitoring (µR h<sup>-1</sup>)

<sup>a</sup> Locations AMS-1 through AMS-7 were called BS-1 through BS-7, respectively, through 1985.

<sup>b</sup> "BKG" refers to the background monitoring stations.

<sup>c</sup> The average of results from four offsite locations.

<sup>d</sup> The average of results from two offsite locations, based on pressurized ionization chamber measurements.

<sup>e</sup> The average of results from AMS-BK1 and AMS-BK2, four to six miles from the site.

<sup>f</sup> The average of results from two locations, 25 and 40 km from the site.

 $^g$  The average of results from four locations, 10 to 40 km from the site.

background locations are not significantly different from those at these six boundary stations. Because of this similarity in exposure rates, we assume that the background exposure rate can be reasonably represented by the average of the exposure rates at the six boundary stations, AMS-1 through AMS-5 and AMS-7. This would give us a consistent basis for comparison of the exposure rates at AMS-6. We think this may be an improvement over the FMPC background locations, which changed locations often.

It will be shown later that concentrations of radon daughters at most of the boundary stations due to releases of radon and radon daughters from the K-65 Silos are estimated to cause significant exposure rates for the 1976–1978 period. The estimated average exposure rate for the six stations, AMS-1 through AMS-5 and AMS-7, due to radon daughters from Silo releases is 0.67  $\mu$ R h<sup>-1</sup> (see page E–37). Thus, this contribution is first subtracted from the average measured exposure rate for AMS-1 through AMS-5 and AMS-5 and AMS-7, to more accurately represent background exposure rates. Because radon and especially radon daughter releases decreased substantially in the 1980–1987 period, a similar correction is not required for this later time period.

Table E-19 shows the estimated net exposure rates at AMS-6, after subtraction of the representation of background exposure rates. This table also shows the average net exposure rates for the periods 1976-1978 and 1980-1990. These are periods before and after, respectively, the sealing of the K-65 Silos in 1979, which caused a great change in the radon concentrations in head space air, as well as in the quantities of radon released from

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Figure E-13. Annual average exposure rates from FMPC routine, quarterly TLD monitoring.

the K-65 Silos. These average net exposure rates are those to which we will compare predicted exposure rates. We will also predict exposure rates for the boundary stations other than AMS-6, but mainly to show that there is little impact of the Silos at those locations.

We next consider input parameters needed for the MicroShield calculations. Table E-10, presented earlier in this Appendix, shows a number of the parameters to be used. Because the receptor points considered here are relatively far from the Silos, we use our standard model for the K-65 Silos, which treats the two Silos as a single source. Thus, all of the parameter values given in Table E-10 are used for these calculations.

The density and moisture content of the metal oxide material in Silo 3 are the same as values given in the final report of Tasks 2 and 3 of this Project (in preparation). These are a density of 0.64 g cm<sup>-3</sup>, and negligible moisture content. The radionuclide concentrations in the metal oxide material are those reported in the report of Tasks 2 and 3, which are shown in Table E-14.

For the K-65 Silos, the only source to be considered for these time periods is radon and daughters in the head space air. Concentrations of these radionuclides in the head space are given in the final report of Tasks 2 and 3 (in preparation). For 1976–1978, the concentrations to be used are  $2.4 \times 10^{-3} \,\mu\text{Ci cm}^{-3}$  of  $^{222}\text{Rn}$ , and  $2.4 \times 10^{-3} \,\mu\text{Ci cm}^{-3}$  of each of the short-lived daughters,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , and  $^{214}\text{Po}$ . For 1980–1990, the concentrations to be used are  $2.62 \times 10^{-2} \,\mu\text{Ci cm}^{-3}$  of  $^{222}\text{Rn}$ , and  $2.62 \times 10^{-2} \,\mu\text{Ci cm}^{-3}$  of each of the short-lived daughters,  $^{218}\text{Po}$ ,  $^{214}\text{Pb}$ ,  $^{214}\text{Po}$ .

The distances from the Silos to the monitoring locations were determined by first plotting the monitoring locations on a detailed engineering drawing of the FMPC site (WMCO 1989b). The distances between the monitoring locations and Silo 3, and the midpoint between Silos 1 and 2, were scaled from the drawing (see also Figure E-12). The

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Year	AMS-6 gross	backgrounda	AMS-6 net
1976	12	8.5 <sup>b</sup>	3.5
1977	12	8.7 <sup>b</sup>	3.3
1978	12	8.3 <sup>b</sup>	3.7
1979	15	9	6
1980	19	11	8
1981	18	11.3	7
1982	19	11.5	8
1983	18	11.8	6
1984	15.5	10.3	5.2
1985	16.95	11.33	5.6
1986	13.6	8.9	4.7
1987	12.54	8.21	4.3
1988	15.30	9.86	5.4
1989	16.81	11.44	5.4
1990	13	7.3	6
.976–1978			3.5
iverage			
.980–1990			<b>5.9</b>
verage			

Table E-19. Net, Annual Average Exposu	ıre
Rates ( $\mu R h^{-1}$ ) at AMS-6	

AMS-7, to represent background.

<sup>b</sup> Contribution due to radon daughter concentrations from K-65 Silos releases has been subtracted.

distances determined are given in Table E-20. As seen in Figure E-12, buildings in the production area shield station AMS-3 from the Silos, and Silo 1 shields station AMS-5 from Silos 2 and 3. Since we are primarily interested in predicted exposure rates at station AMS-6, for calculations we ignore the shielding of these other stations.

Using the parameters described above, exposure rate calculations were performed with MicroShield. The results for the boundary monitoring stations are shown in Table E-21. Based on these predicted exposure rates, direct exposures from the waste storage silos do not appear to contribute significantly to the measured exposure rates at the boundary stations (in Table E-18), except for station AMS-6.

For station AMS-6, we also consider the penetrating radiation that would result from elevated concentrations of radon daughters at that station, due to releases of radon and daughters from the K-65 Silos. Calculations of the radon concentration due to releases from the K-65 Silos, discussed earlier in this Appendix, indicated a significant concentration prior to mid-1979 at station AMS-6 (then called BS-6) (see page E-9). For these calculations, however, we also must use the quantities of the radon daughters released from the K-65 Silos. In the source term work of this Project, we have estimated release rates of radon and radon daughters (see final report of Tasks 2/3, in preparation). For 1976-1978, the release

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	Distance from monitoring station (ft) to:				
Monitoring station	Point between Silos 1 and 2	Center of Silo 3			
AMS-1	4100	3900			
AMS-2	6200	6100			
AMS-3ª	4800	4800			
AMS-4	5800	6000			
AMS-5 <sup>a</sup>	3000	3300			
AMS-6	1100	1200			
AMS-7	3700	3400			

Table E-20. Distance between FMF	<b>C</b> Boundary
<b>Exposure Rate Monitoring Station</b>	ns and Silos

<sup>a</sup> Note that buildings in the production area provide shielding for station AMS-3, and Silo 1 provides shielding of Silos 2 and 3 for station AMS-5.

rates are estimated to be 6200 Ci y<sup>-1</sup> of each of  $^{222}$ Rn,  $^{218}$ Po,  $^{214}$ Pb,  $^{214}$ Bi, and  $^{214}$ Po, continuously released. For 1980–1987, the release rates are estimated to be 130 Ci y<sup>-1</sup> of  $^{222}$ Rn continuously released; 800 Ci y<sup>-1</sup> of  $^{222}$ Rn released during daylight hours only; and 170 Ci y<sup>-1</sup> of each of  $^{218}$ Po,  $^{214}$ Pb,  $^{214}$ Bi, and  $^{214}$ Po, released during daylight hours only.

	F	For 1976–1978	3	For 1980–1990			
Station	From K-65 Silos	From Silo 3	Total	From K-65 Silos	From Silo 3	Total	
AMS-1	$2.96 \times 10^{-4}$	$9.51 \times 10^{-5}$	$3.9 \times 10^{-4}$	$3.24 \times 10^{-3}$	$9.51 \times 10^{-5}$	$3.3 \times 10^{-3}$	
AMS-2	$4.24 \times 10^{-6}$	$1.85 \times 10^{-6}$	$6.1 \times 10^{-6}$	$4.64 \times 10^{-5}$	$1.85 \times 10^{-6}$	$4.8  imes 10^{-5}$	
AMS-3	$6.93  imes 10^{-5}$	$1.79  imes 10^{-5}$	$8.7  imes 10^{-5}$	7.58 × 10 <sup>-4</sup>	$1.79 \times 10^{-5}$	$7.8 \times 10^{-4}$	
AMS-4	9.31 × 10 <sup>-6</sup>	2.19 × 10 <sup>-6</sup>	$1.2 \times 10^{-5}$	$1.02 \times 10^{-4}$	$2.19 \times 10^{-6}$	$1.0 \times 10^{-4}$	
AMS-5	3.29 × 10−3	$3.10 \times 10^{-4}$	$3.6  imes 10^{-3}$	$3.60  imes 10^{-2}$	$3.10 \times 10^{-4}$	$3.6  imes 10^{-2}$	
AMS-6	$5.33  imes 10^{-1}$	$5.06 imes10^{-2}$	$5.8  imes 10^{-1}$	$5.83  imes 10^{0}$	$5.06 imes10^{-2}$	5.9 × 10 <sup>0</sup>	
AMS-7	$6.96 \times 10^{-4}$	$2.54 \times 10^{-4}$	9.5 × 10 <sup>-4</sup>	$7.61\times10^{-3}$	$2.54 \times 10^{-4}$	$7.9  imes 10^{-3}$	

Table E-21. Predicted Exposure Rates ( $\mu R h^{-1}$ ) at FMPC Boundary Monitoring Stations, Due to Direct Exposures from Waste Storage Silos

Monitoring station AMS-6 is west-southwest from the K-65 Silos, so the "wind from" direction is east-northeast (see Figure E-12). The distance from the center of the K-65 Silos to AMS-6 is 1100 ft, or 330 m (see Table E-20). With these values, we used our radon dispersion model (RNCHIQ4) to calculate the ratios of air concentrations of radon and radon daughters to release rates of radon ( $\chi/Q$ ). Table E-22 shows the predicted values of  $\chi/Q$  at AMS-6, for continuous releases and daylight-only releases from the K-65 Silos.

To estimate exposure rates, we require dose conversion factors. From a DOE compilation of external dose-rate conversion factors (DOE 1988), we obtained conversion factors for immersion in a semi-infinite cloud containing radioactivity. These dose-rate conversion

## Appendix E Monitoring Data for Radon in Air and Exposure Rate: with Comparisons to Predictions

	1976-1978	1980–1987		
Radionuclide	Continuous releases <sup>a</sup>	Continuous releases <sup>b</sup>	Daylight-only releases <sup>c</sup>	
<sup>222</sup> Rn	0.281	0.281	0.241	
<sup>218</sup> Po	0.278	0.140	0.130	
<sup>215</sup> Pb	0.273	0.00843	0.0537	
<sup>214</sup> Bi	0.272	0.00051	0.0501	
<sup>214</sup> Po	0.272	0.00049	0.0500	

Table E-22. Predicted $\chi/Q$ (pCi m <sup>-3</sup> per Ci <sup>222</sup> Rn y <sup>-1</sup> )
at AMS-6, for Releases from K-65 Silos

<sup>a</sup> Daughters released in equilibrium with <sup>222</sup>Rn.

<sup>b</sup> No radon daughters released.

<sup>c</sup> Radon daughter releases equal 0.21 times radon releases.

factors are shown in Table E-23. The conversion factors are for effective dose equivalent to a person immersed in the radioactive cloud. We assume that the effective dose equivalent is about equal numerically to the exposure in air (that is, an exposure of 1 mR in air results in an effective dose of 1 mrem to an exposed person). Thus, the conversion factors can be used as exposure rate conversion factors.

Table E-23. Dose-Rate Conversion Factors for Exposure to a Semi-Infinite Cloud Containing Radioactivity

Radionuclide	Dose-rate factor (mrem y <sup>-1</sup> per µCi m <sup>-3</sup> )
<sup>222</sup> Rn	1.95
<sup>218</sup> Po	0.0
<sup>214</sup> Pb	$1.25 \times 10^{3}$
<sup>214</sup> Bi	$8.11 \times 10^{3}$
214Po	$4.34 \times 10^{-1}$

The concentrations of the radon daughters are calculated by multiplying the  $\chi/Q$  values by the release rates. The exposure rates are then calculated by multiplying the concentrations by the appropriate exposure-rate (dose-rate) conversion factor. Based on the dose-rate factors shown in Table E-23, <sup>214</sup>Pb and <sup>214</sup>Bi are the only significant contributors to the exposure rates, and so we only consider these two radionuclides. Table E-24 shows the results of these calculations. For 1976-1978, the predicted exposure rate at station AMS-6 due to K-65 Silos releases is significant, and it seems reasonable that the exposure rates at the other boundary stations due to these releases may also be significant. Using the same methodology, we estimate the average exposure rate for the six other boundary stations, AMS-1 through AMS-5 and AMS-7, due to K-65 Silos releases, to be 0.67  $\mu$ R h<sup>-1</sup> for 1976-1978. This exposure rate is significant enough that it should be subtracted from our pseudo-background concentration. That has been done (see page E-33). For 1980-1987,

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oncentration (μCi m <sup>-3</sup> )	Exposure rate $(\mu R h^{-1})$	Concentration (µCi m <sup>-3</sup> )	Exposure rate (µR h <sup>-1</sup> )
$1.69 \times 10^{-3}$	0.24	$4.41 \times 10^{-5}$	0.00629
$1.69 \times 10^{-3}$	1.56	$4.01  imes 10^{-5}$	0.0371
-	$\frac{(\mu \text{Ci m}^{-3})}{1.69 \times 10^{-3}}$ $1.69 \times 10^{-3}$	( $\mu$ Ci m <sup>-3</sup> )       ( $\mu$ R h <sup>-1</sup> )         1.69 × 10 <sup>-3</sup> 0.24         1.69 × 10 <sup>-3</sup> 1.56	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$

however, the exposure rate is significantly less, because radon daughter releases are significantly reduced, and a similar correction is unnecessary.

Table E-25 summarizes our calculations of exposure rates at monitoring station AMS-6 due to direct exposures from the waste storage silos, and to radon and daughter releases from the K-65 Silos. We also calculated ratios of the predicted to observed (P/O) exposure rates, and these are also shown.

	Predict	ed exposure rates	due to:		
Period	Direct exposures	Rn and daughter releases	Total Predicted	Average measured	P/O Ratio
1976–1978 1980–1987	0.58 5.9	1.8 0.043	2.4 5.9	3.5 5.9	0.69 1.0

Table E-25. Summary of Predicted and Measured Exposure Rates  $(\mu R h^{-1})$  and Predicted to Observed (P/O) Ratios for Station AMS-6

As shown by the P/O ratios, our predicted exposure rates agree well with the measured exposure rates for FMPC monitoring station AMS-6.

#### Conclusions

In relation to direct exposures from gamma radiation emitted from materials in the K-65 and Metal Oxide Silos, we have compared predicted and measured exposure rates for three major studies of exposure rate measurements: (1) surveys along Paddy's Run Road in 1987 (in Task 4 of this Project, Killough et al. 1993), (2) a 1957 survey relatively close to the K-65 Silos, and (3) the FMPC routine exposure rate monitoring at the site boundary air monitoring stations. For the Paddy's Run Road surveys, the predicted exposure rates were about one half the measured values. For the 1957 survey, the geometric mean predicted to observed ratios (P/O) was 3.0, although P/O values were generally less than 2.5 for greater distances from the Silos (in this case 160 ft or more). For the FMPC routine monitoring, P/O ratios were about 1 for the short period prior to the 1979 sealing of the Silos (1976–1978) and for the period after the sealing (1980–1987). These comparisons indicate reasonably good agreement between our predictions and the environmental measurements. These results will be further evaluated later in this Project, before making final determinations about the performance of our direct exposure model.

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Location	Sampling period	Exposure time (h)	Concentration <sup>222</sup> Rn (pCi L <sup>-1</sup> )	References <sup>a</sup>
				· · · · · · · · · · · · · · · · · · ·
Pit 5°	05/18/78-05/31/78		0.75	handwritten summary, Heatherton 1979.
BS-1	05/18/78-05/31/78		0.28	handwritten summary, Heatherton 1979.
BS-6	04/12/79-04/19/79		6.9	handwritten summary tables.
BS-6	04/12/79-04/19/79		4.8	handwritten summary tables.
BS-6	04/12/79-04/19/79		5.7	handwritten summary tables.
BS-6	04/19/79-04/27/79		2.8	handwritten summary tables.
BS-6	04/19/79-04/27/79		2.4	handwritten summary tables.
BS-6	04/19/79-04/27/79		1.8	handwritten summary tables.
BS-6	05/04/79-05/07/79 <sup>c</sup>		1.8	handwritten summary, handwritten summary tables.
BS-6	05/07/79-05/14/79	•	0.8	handwritten summary, handwritten summary tables.
BS-6	05/21/79-05/29/79		1.6	handwritten summary table.
BS-6	05/29/79-06/04/79		2.38	handwritten summary, handwritten note.
				handwritten summary tables.
BS-6	06/11/79-06/15/79		3.8	handwritten summary table.
BS-6	06/18/79-06/22/79		2.3	handwritten summary table.
BS-6	07/02/79-07/05/79		1.6	handwritten summary table.
BS-6	07/05/79-07/09/79		0.6	handwritten summary table.
BS-6	07/13/79-07/30/79		0.4	handwritten summary table.
BS-6	07/13/79-07/30/79		0.3	har.dwritten summary table.
BS-6	07/30/79-07/31/79		0.9	handwritten summary table.
BS-6	07/30/79-07/31/79		0.6	handwritten summary table.
BS-6	11/15/79-11/21/79	144	0.7	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	11/15/79-11/21/79	144	0.5	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	11/28/79-12/05/79	165	<0.1	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	11/28/79-12/05/79	165	0.4	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	12/05/79-12/12/79	170	0. <b>46</b>	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	12/05/79-12/12/79	170	0.50	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	12/12/79-01/02/80	504	0.2	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.
BS-6	12/12/79-01/02/80	504	0.3	"Radon Monitor TLD Analysis"
				worksheet, handwritten summary tables.

Table ES-1. Integrated Measurements of <sup>222</sup> Rn in Ambient Air Using Passive Radon
Monitors, from May 1978 through January 1980

<sup>a</sup> All references cited are part of Boback circa 1984, except for those specifically noted otherwise.

<sup>b</sup> Location described as north of Pit 5 [we assume waste pit 5], about 20 ft from east end.

<sup>c</sup> The handwritten summary and one table give the start date as 5/4/79. A second table gives the date as 5/3/79. We assume that 5/4/79 is correct.

Location	Date	Time	Concentration <sup>222</sup> Rn (pCi L <sup>-1</sup> )	Comments (References <sup>a</sup> )
BS-1	08/22/78	9:40 am	0.25	(handwritten summary and Heatherton 1979)
BS-2	08/22/78	9:48 am	0.77	(handwritten summary, and Heatherton 1979)
BS-3	08/22/78	9:09 am	0.38	(handwritten summary, and Heatherton 1979)
BS-4	08/22/78	9:17 am	0.65	(handwritten summary, and Heatherton 1979)
BS-5	08/22/78	9:26 am	1.93	(handwritten summary, and Heatherton 1979)
BS-6	08/22/78	9:32 am	3.98	(handwritten summary, and Heatherton 1979)
BS-1	08/23/78	9:07 am	0.40	(handwritten summary, and Heatherton 1979)
BS-2	08/23/78	9:14 am	0.80	(handwritten summary, and Heatherton 1979)
BS-3	08/23/78	8:35 am	0.29	(handwritten summary, and Heatherton 1979)
BS-4	08/23/78	8:43 am	0.50	(handwritten summary, and Heatherton 1979)
BS-5	08/23/78	8:51 am	0.40	(handwritten summary, and Heatherton 1979)
BS-6	08/23/78	8:58 am	1.74	(handwritten summary, and Heatherton 1979)
BS-1	04/26/79		0.8	(handwritten summary table)
BS-2	04/26/79		<0.1	(handwritten summary table)
BS-6	04/26/79		0.1	(handwritten summary table)
BS-4	04/27/79		<0.1	(handwritten summary table)
BS-5	04/27/79		0.1	(handwritten summary table)
BS-6	04/27/79		<0.1	(handwritten summary table)
Ь	06/25/79	11:30 am	82	Flask N10. (handwritten summary table)
Ь	06/25/79	11:30 am	335	Flask N11. (handwritten summary table)
Ь	06/25/79	11:30 am	99	Flask N12. (handwritten summary table)
Ь	06/25/79	11:30 am	411	Flask N13. (handwritten summary table)
Ь	06/28/79	12:30 pm	45	Flask N10. (handwritten summary table)
Ь	06/28/79	12:30 pm	268	Flask N11. (handwritten summary table)
Ь	06/28/79	12:30 pm	54	Flask N12. (handwritten summary table)
Ь	06/28/79	12:30 pm	261	Flask N13. (handwritten summary table)
Ь	07/03/79	12:20 pm	44	Flask N10. (handwritten summary table)
Ь	07/03/79	12:20 pm	211	Flask N11. (handwritten summary table)
ь	07/05/79	12:10 pm	54	Flask N10. (handwritten summary table)
ь	07/05/79	12:10 pm	249	Flask N11. (handwritten summary table)
Ь	07/05/79	12:10 pm	58	Flask N12. (handwritten summary table)
Ь	07/05/79	12:10 pm	240	Flask N13. (handwritten summary table)
Silo 1, east <sup>c</sup>	07/06/79	12:55 pm	21	Flask N10. (handwritten summary table)
Silo 1, west <sup>c</sup>	07/06/79	12:55 pm	67	Flask N11. (handwritten summary table)
Silo 2, east <sup>c</sup>	07/06/79	12:55 pm	591	Flask N12. (handwritten summary table)
Silo 2, west <sup>c</sup>	07/06/79	12:55 pm	38	Flask N13. (handwritten summary table)
Ь	08/08/79	9:22 am	0.25	Flask N7. (handwritten summary table)
Ь	08/08/79	9:45 am	2.65	Flask N9. (handwritten summary table)
ь	08/08/79	10:10 am	0.34	Flask N15. (handwritten summary table)
ь	08/08/79	10:32 am	3178.7	Flask N13. (handwritten summary table)

### Table ES-2. Grab Measurements of <sup>222</sup>Rn in Ambient Air Using Lucas Cell, Scintillation Flasks, from August 1978 through April 1980

<sup>a</sup> All references cited are part of Boback circa 1984, except for those specifically noted otherwise.

<sup>b</sup> Locations not indicated. We think they were probably on or very near K-65 Silo domes, based on very high concentrations and inclusion in same table as samples of 7/6/79.

<sup>c</sup> We assume these are on or very near the Silos, based on very high concentrations.

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Location	Date	Time	Concentration <sup>222</sup> Rn (pCi L <sup>-1</sup> )	Comments (References <sup>4</sup> )
<u>ь</u>	08/08/79	10:45 am	787.5	Flask N10. (handwritten summary table)
ь	08/08/79	11:09 am	2858.1	Flask NO. (handwritten summary table)
ь.	08/08/79	11:22 am	29.9	Flask N8. (handwritten summary table)
6	08/08/79	11:47 am	54.3	Flask N14. (handwritten summary table)
ь	08/08/79	12:09 pm	2.94	Flask N16. (handwritten summary table)
6	08/08/79	12:32 pm	1925.0	Flask N12. (handwritten summary table)
ь	08/08/79	12:52 pm	7.33	Flask N2. (handwritten summary table)
ь	08/08/79	1:15 pm	-0.28 <sup>d</sup>	Flask N11. (handwritten summary table)
BS-1	08/23/79	13:56	bkge	Cloudy. (handwritten summary tables)
BS-2	08/23/79	14:05	bkge	Cloudy. (handwritten summary tables)
BS-3	08/23/79	13:33	0.23	Cloudy. (handwritten summary tables)
BS-4	08/23/79	13:43	0.34	Cloudy. (handwritten summary tables)
BS-5	08/23/79	14:27	0.06	Cloudy. (handwritten summary tables)
BS-6	08/23/79	14:18	0.14	Cloudy. (handwritten summary tables)
BS-1	08/27/79	10:45 am	0.58	Cloudy. (handwritten summary tables)
BS-2	08/27/79	10:52 am	bkge	Cloudy. (handwritten summary tables)
BS-3	08/27/79	10:35 am	b <b>kg</b> e	Cloudy. (handwritten summary tables)
BS-4	08/27/79	11:12 am	bkg <sup>e</sup>	Cloudy. (handwritten summary tables)
BS-5	08/27/79	11:06 am	0.15	Cloudy. (handwritten summary tables)
BS-6	08/27/79	11:01 am	0.44	Cloudy. (handwritten summary tables)
BS-2	08/29/79	9:40 am	0.36	Foggy, wind from west at 4 mph. (handwritten
				summary tables)
BS-6	08/29/79	9:50 am	0.17	Foggy, wind from west at 4 mph. (handwritten summary tables)
BS-2	10/25/79	9:15 am	0.3	Flask N7. (handwritten summary, and summary table)
BS-2	10/25/79	9:17 am	0.2	Flask N2. (handwritten summary, and
BS-6	10/25/79	9:25 am	0.1	Flask N15. (handwritten summary, and summary table)
BS-6	10/25/79	9:30 am	0.3	Flask N8. (handwritten summary, and summary table)
<b>BS-1</b>	11/13/79	ſ	0.7	(Ross 1979, handwritten summary tables)
BS-2	11/13/79	ſ	<0.18	(Ross 1979, handwritten summary tables)
BS-3	11/13/79	ſ	0.5	(Ross 1979, handwritten summary tables)
BS-4	11/13/79	ſ	0.1	(Ross 1979, handwritten summary tables)
BS-5	11/13/79	ſ	0.5	(Ross 1979, handwritten summary tables)
BS-6	11/13/79	ſ	0.6	(Ross 1979, handwritten summary tables)
BS-6	04/02/80	12:00	<0.1	Two samples with same results. (Ross 1980)

Table ES-2. Grab Measurements of 222Rn in Ambient Air Using Lucas Cell, ScintillationFlasks, from August 1978 through April 1980 (continued)

 $^{d}$  Reported as negative because the gross count rate was less than the background rate.

<sup>e</sup> Reported as "not significantly greater than background," or "not measurably above background" in one table, and as "<0.1" pCi L<sup>-1</sup> in the other table.

All taken between 8:15 and 9:05 am.

<sup>g</sup> Reported as "ND" in Ross (1979) and in one summary table, and as "<0.1" in the other table.

- 000302

			Concentration	
Location	Date	Time	<sup>222</sup> Rn (pCi L <sup>-1</sup> ) <sup>4</sup>	Comments (References <sup>b</sup> )
BS-5			0.4	(handwritten summary only)
BS-6	09/12/78	8:30 am	0.2	SW wind. (summary, ADS, and plot)
BS-6	09/13/78	8:31 am	5.1	Calm, hazy. (summary, ADS, and plot)
BS-6	09/14/78	8:14	0.2	SSW wind. (summary, ADS, and plot)
BS-6	09/15/78	8:24	0.4	Calm, foggy. (summary, ADS, and plot)
BS-6	09/18/78	8:36	0.4	SW wind, clear. (summary, ADS, and plot)
BS-6	09/19/78		1.8	W wind, light fog. (summary, ADS, and plot)
BS-6	09/20/78	9:01	1.7	NNW wind, light fog. (summary, ADS, and plot)
BS-6	09/21/78	8:32	2.1	"No wind (if any wind south-southwest)," light fog.
BS-6	09/27/78	8:30 am	3.8	Light WSW wind, clear. (summary, ADS, and plot)
Clearwell <sup>c</sup>	09/28/78		0.2	N wind, 5–10 mph. (summary, ADS, and plot)
BS-6	10/03/78	8:33	5.0	Calm. (summary, ADS, and plot)
BS-6	10/05/78	8:35	1.5	NW wind. (summary, ADS, and plot)
BS-6	10/06/78	8:38	0.2	SW wind. (summary, ADS, and plot)
BS-6	10/10/78	8:46	2.1	Light SE wind, light fog. (summary, ADS, and plot)
BS-6	10/11/78 <sup>d</sup>	8:35	36	NE wind, rainy. Filter still damp while counting. (summary, ADS, and plot)
Silo 2 dome	10/16/78	9:04	190°	SW win, clear. (ADS and plot only)
Silo 2 dome	10/17/78	8:54	8"	Calm, foggy. (ADS and plot only)
Silo 2 dome	10/18/78	8:45	21 <sup>e</sup>	Light SW wind. (ADS and plot only)
Silo 2 dome	10/23/78	8:53	f	Calm, overcast. (ADS only)
Silo 2 dome	10/25/78	8:51	ſ	WNW wind. (ADS only)
BS-6	10/27/78	8:53	ſ	W wind, foggy. (ADS only)
BS-6	10/30/78	9:04	ſ	ENE wind. (ADS only)

Table ES-3. Estimated Concentrations of <sup>222</sup>Rn in Ambient Air Based on Radon Daughter Grab Samples, from September and October 1978 <sup>a</sup>

- <sup>3</sup> Radon daughter samples were collected on millipore filters, with pump flow rates generally from 17 to 21 L min<sup>-1</sup> (except samples of 10/16 through 10/25, which used rates of 1.3 and 2.5 L min<sup>-1</sup>), and sampling time of 30 min (from ADSs). Gross alpha counting was performed at 30, 60, 90, and 120 min after end of sampling (from ADSs). Calculated concentrations (before decay correction) were plotted on semi-logarithmic paper, and the concentration at zero time after the end of sampling was extrapolated from a line through the data (from plots). Results were reported (in the summary sheet) as <sup>222</sup>Rn, based on 100% equilibrium of the radon daughters.
- <sup>b</sup> All references cited are part of Boback circa 1984. The "summary" is a handwritten summary of results. The "ADS" are Analytical Data Sheets of the FMPC Health and Safety Division's analytical laboratory. The "plots" are hand-drawn graphs of the concentrations on semi-log paper.
- <sup>c</sup> Location was "on rail near NE corner of Clearwell pump house."
- <sup>d</sup> The summary sheet gives the date as 10/1/78, but the ADS indicates 10/11/78 for collected, received, and reported dates. We assume that 10/11/78 is correct.
- <sup>e</sup> Results not included on summary sheet. We read values off the plot, for time zero.
- <sup>f</sup> Results not plotted by Boback (circa 1984). Individual results for 10/23/78 sample were reported as 0.692, 0.405, 0.249, and 0.136 pCi L<sup>-1</sup>, at times 30, 60, 90, and 120 min, respectively, after end of sampling. Similarly, reported results for 10/25/78 sample were 24.360, 14.817, 8.713, and 5.201 pCi L<sup>-1</sup>, for 10/27/78 sample were 0.190, 0.101, 0.061, and 0.035 pCi L<sup>-1</sup>, and for 10/30/78 sample were 1.477, 0.865, 0.448, and 0.241 pCi L<sup>-1</sup>.

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Monitoring period	BS-1	BS-2	BS-3	BS-4	BS-5	BS-6 <sup>b</sup>	BS-6 <sup>b</sup>	BS-7	K-65 <sup>c</sup>	NMOT	8 mi ENE <sup>e</sup>	5 mi WSW <sup>e</sup>
06/13/80-10/03/80						0.24	1.91 <sup>f</sup>		180.22			
07/03/80-10/03/80	0.44	0.54	0.49	0.19	0.65 <sup>g</sup>							
10/03/80-03/13/81	0.11	0.17	0.22	0.05	0.08	0.34			80.08			
03/13/81-05/26/81	h	0.44	0.13	0.18	0.40	0.63	0.54	0.30	135.75 <sup>g</sup>			
04/07/81-05/26/81											0.60	
05/26/81-09/28/81	i	1.35	0.53	0.66	1.48	0.94	0.66	1.07			0.80	0.948
09/28/81-02/02/82	0.85	0.48	0.60	0.48	0.36	0.73	0.36	j	120.73		0.60	0.36
02/02/8205/13/82	0.46	0.92	0. <b>6</b> 1	0.46	0. <b>6</b> 1	1.08	1.70	k	126.22		0.30	0.30
05/13/8208/25/82							•				0.42	0.95
05/13/82-08/24/82	1.508	1.07	0.64	1.18	1.28	1.18	0.648	1.39	1 <b>6</b> 0.15			
08/25/82-11/02/82											1.34	0.87
08/24/82-11/02/82	0.87	1.32	0.87	1.10	0.87	0.87	1.32	1.76	167.39			
11/02/82-01/06/83	0.33	0.33	0.50	0.84	1.01	0.33	0.33	0.50	63.86	0.84	0.16	0.50
01/06/83-03/29/83	0.39	0.75	0.37	0.37	1. <b>69</b>	0.37	0.56	0.56	181.15	0.37	0.75	0.568
03/29/83-06/28/83	0.23	0.60	0.60	0.36	0.23	0.96	0.48	0.48	131.88	0.84	0.36	0.48
06/28/83-09/27/83	0.72	1.09	0.48	0.93	0.93	0.72	1.78	0.62	180.59	2.1	0.72	0.51
09/27/83-12/27/83	1.25	0.62	1.57	0. <b>93</b>	1.36	0.72	0. <b>93</b>	1. <b>99</b>	112. <b>49</b>	1.67	1.25	0.878

 Table ES-4. Detailed 222 Rn Concentration Results (pCi L<sup>-1</sup>) from the Routine FMPC

 Radon Monitoring Program for 1980-1983<sup>a</sup>

<sup>a</sup> Ref. Anonymous circa 1984.

<sup>b</sup> Station BS-6 is listed twice because for most monitoring periods duplicate detectors were exposed.

<sup>c</sup> The location of this station was not described; it is probably very close to the K-65 Silos (based on the levels seen).

<sup>d</sup> The location of this station was not described.

<sup>e</sup> These are offsite locations, used for background concentrations.

<sup>f</sup> Detector cup was open.

000001

<sup>g</sup> Splits in the filter were noted.

<sup>h</sup> Duplicate results for this period were 0.44 and 0.35 pCi  $L^{-1}$ .

<sup>*i*</sup> Duplicate results for this period were 1.48 and 0.39 pCi  $L^{-1}$ .

<sup>1</sup> Duplicate results for this period were 0.97 and 0.48 pCi  $L^{-1}$ .

<sup>k</sup> Duplicate results for this period were 0.77 and 0.46 pCi  $L^{-1}$ .

F = 7339

Nominal period	Monitoring Dates	K65 A	K65 B	K65 C	K65 D	K65 E	K65 F	K65 G	K65 H
2nd qtr 1987	03/04/87-06/10/87	9.6	16.2	14.9	10.6	8.3	11.3	4.2	2.7
3rd gtr 1987	06/11/87-09/05/87	5.4	10.8	11.2	11.5	6.7	11	4.3	2.6
4th qtr 1987	09/06/87-01/07/88	3.2	7.2	6.1	6.8	4.5	3.7	1.8	2.9
1st qtr 1988	01/08/8803/05/88	4.55	4.75		3.1	5.2	2.1	1.9	4.25
2nd qtr 1988	03/06/8806/02/88	3.3	3.2	3.8	5.1	3. <del>9</del>	3.3	2.45	3.4
3rd qtr 1988	06/03/8809/03/88	9.4	9.25	4.2	13.5	4.0	3.55	2.6	4.35
4th qtr 1988	09/04/8802/01/89	4.25	6.4	9.15	16.95	17.25	12.85		5.8
1st gtr 1989	02/02/8903/05/89	3.9	3.95	8.3	7.1	9.45	15.2	3.6	3.65
2nd qtr 1989	03/06/8906/11/89	2.15	2.15	3.0	4.6	3.9	4.35	1.85	2.25
3rd gtr 1989	06/12/8909/11/89	3.3	2.45	4.7	6.25	5.1	5.05	3.0	7.2
4th qtr 1989	09/12/89-01/24/90	4.0	8.65	7.25	6.35	3.35		7.9	5.4
1st gtr 1990	01/25/90-03/02/90	4.05	11.45	6.0	4.4		2.25	3.3	1.25
2nd qtr 1990	03/03/90-07/04/90	2.95	4.3		3.85	2.75	2.85	2.15	2.55
3rd gtr 1990	07/04/90-10/12/90	2.05	2.5	3.1	3.65	3.75	3.75	2.95	1.65
4th gtr 1990	10/12/90-01/07/91	2.7	4.55	2.75	3.05	2.25	3.5	2.5	1.7
1st gtr 1991	01/08/91-03/08/91	2.8	4.7	4.3	5.4	4.7	5.1	3. <b>9</b>	2.6
2nd qtr 1991	03/09/91-07/02/91	8.7	6.6	12.6	6.8	4.7	5.5	3.5	3.3
3rd qtr 1991	07/02/91-10/01/91	8.7	42.9	32	6.2	8.6	18	1.9	2.2
4th atr 1991	10/01/91-01/07/92	16.9	12.6	15.1	15.5	15.5	14.5	8.9	3.8

 Table ES-5. Average 222 Rn Concentrations (pCi L<sup>-1</sup>) at K-65 Area Fenceline Monitoring Stations; Results from FMPC Routine Monitoring<sup>a</sup>

 Table ES-5. Average 222Rn Concentrations (pCi L<sup>-1</sup>) at K-65 Area Fenceline Monitoring Stations; Results from FMPC Routine Monitoring (continued)<sup>a</sup>

Nominal period	Monitoring Dates	K65 I	K65 J	K65 K	K65 L	K65 M	K65 N	K65 O	K65 P	Mean <sup>b</sup>
2nd qtr 1987	03/04/87-06/10/87	3.4	4.7	5.8	12.5	12.1	10.2	7.8	6.8	8.82
3rd qtr 1987	06/11/87-09/05/87	3.7	3.1	3.7	5.0	4.2	3.9	2. <del>9</del>	2.2	5.76
4th qtr 1987	09/06/87-01/07/88	3.6	4.1		5.1	4.5	6.3	6.7	5.1	4.77
1st qtr 1988	01/08/8803/05/88	4.8	5.9	3.85	6.55	4.1	4.45	3.3	2.3	4.07
2nd qtr 1988	03/06/88-06/02/88	5.0	3.6	4.6	6.75	5.4	4.0	3.05	2.95	3. <b>99</b>
3rd qtr 1988	06/03/8809/03/88	2.55	3.15	4.3	8.6	5.35	3.65	4.0	2.8	5.33
4th qtr 1988	09/04/88-02/01/89	5. <b>65</b>	4.05	3.45	26.5	1 <b>9</b>	9.55	6.75	4.7	10.15
1st qtr 1989	02/02/8903/05/89	4.7	4.95	7.3	10.65	10.95	14.2	5.85	3.75	7.34
2nd qtr 1989	03/06/89-06/11/89	2.75	3.0	3.5	7.5	5.25	3.5	2.85	2.8	3.46
3rd qtr 1989	06/12/89-09/11/89		3.0	4.95	9.5	4.35	4.6	2.9	1.45	4.52
4th qtr 1989	09/12/89-01/24/90	5.7	6.55	5.5	10.85	7.5	4.4	5.15	2.05	6.04
lst qtr 1990	01/25/90-03/02/90	1.8	1.5	2.9	2.3	2.9	2.6	2.05	1.8	3.37
2nd qtr 1990	03/03/90-07/04/90	2.2	1.15	1.45	2.25	2.25	2.35	1.8	1.185	2.40
3rd qtr 1990	07/04/90-10/12/90	1.7	1.15	1.25	2.1	1.95	2.2	1.65	0.63	2.25
4th gtr 1990	10/12/90-01/07/91	2.1	1.85	1.6	2.35	2.9	3.7	2.05	1.4	2.56
1st qtr 1991	01/08/91-03/08/91	4.0	1.7	1.9	3.0	4.5	3.0	6.9	2.1	3.79
2nd qtr 1991	03/09/91-07/02/91	2.4	1.9	1.5	10.3	6.7	5.1	3.5	1.5	5.29
3rd qtr 1991	07/02/91-10/01/91	2.0	1.7	2.0	2.2	2.5	4.2	4.1	1.0	8.76
4th qtr 1991	10/01/91-01/07/92	4.4	3.0	3.7	8.2	10.8	15.2	15.3	5.5	10. <b>56</b>

<sup>a</sup> Ref. Byrne 1992. Concentrations given here are the average of (typically two) results for Type F detectors.

<sup>b</sup> Mean concentration for all locations for the given quarter.

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Survey Points around Silo 1						Survey Poin	nts aroun	d Silo 2		
Survey	location	Distan	æ (ft) fron	n centers	Survey location Dis		Distan	ance (ft) from centers		
direction	distance (ft)	Silo 1	Silo 2	Silo 3	direction	distance (ft)	Silo 1	Silo 2	Silo 3	
N	10	51	71	279	N	10	173	51	157	
N	20	61	61	269	N	20	183	61	147	
NE	10	51	92	295	N	40	203	81	127	
NE	20	61	89	289	N	80	243	121	87	
NE	40	81	85	277	E	20	137	61	215	
NE	80	121	91	256	E	40	148	81	221	
NE	160	201	142	231	E	80	173	121	238	
NE	320	361	286	258	E	160	237	201	285	
Е	20	61	136	334	E	320	383	361	411	
E	40	81	145	338	SE	10	94	51	246	
E	80	121	170	349	SE	20	91	61	254	
Е	160	201	233	382	SE	40	88	81	270	
Е	320	361	379	483	SE	80	95	121	304	
S	10	51	173	381	SE	160	146	201	376	
S	20	61	183	<b>39</b> 1	SE	320	290	361	526	
S	40	81	203	411	S	10	71	51	259	
S	80	121	243	451	s	20	61	61	269	
S	160	201	323	531	SW	10	92	51	248	
W	20	61	137	337	SW	20	89	61	256	
W	40	81	148	342	SW	40	85	81	273	
W	80	121	173	354	SW	80	91	121	308	
NW	10	51	94	297	SW	160	142	201	380	
NW	20	61	91	291	SW	320	286	361	532	
NW	40	81	88	280	w	20	136	61	219	
NW	80	121	95	262	w	40	145	81	225	
NW	1 <b>60</b>	<b>20</b> 1	1 <b>46</b>	240	W	80	170	121	244	

Exposures Due to Silo 1 Sources						Exposures	Due to Sil	o 2 Sources	
	Calculated exposure rate $(mR h^{-1})$ due to					Calculate	d exposure	rate (mR h	1 <sup>-1</sup> ) due to
distance from center (ft)	K-65 material	Head space in cylinder	Head space in dome	Total	distance from center (ft)	K-65 material	Head space in cylinder	Head space in dome	Total
51	13.62	0.6101	0.2398	14.47	51	7.674	1.320	0.2398	9.234
61	10.20	0.5345	0.2178	10.95	61	5.306	1.119	0.2178	6.642
71	7.420	0.4430	0.1881	8.051	71	3.636	0.9016	0.1881	4.726
81	5.373	0.3595	0.1589	5.891	81	2.542	0.7185	0.1589	3.419
85	4.755	0.3305	0.1483	5.234	85	2.229	0.6572	0.1483	3.034
88	4.353	0.3104	0.1407	4.804	88	2.028	0.6155	0.1407	2.784
<del>89</del>	4.230	0.3041	0.1383	4.672	89	1.967	0.6022	0.1383	2.707
91	3.997	0.2918	0.1335	4.422	91	1.853	0.5767	0.1335	2.563
92	3.887	0.2859	0.1312	4.304	92	1.799	0.5644	0.1312	2.495
94	3.680	0.2746	0.1267	4.081	94	1.698	0.5409	0.1267	2.366
95	3.582	0.2691	0.1246	3.976	95	1.651	0.5301	0.1246	2.306
121	1.945	0.1656	0.08103	2.192	121	0.8773	0.3218	0.08103	1.2801
1 <b>36</b>	1.453	0.1294	0.06459	1.647	136	0.6514	0.2504	0.06459	0.9664
137	1.426	0.1274	0.06366	1.617	137	0.6394	0.2464	0.06366	0.9495
142	1.305	0.1180	0.05924	1.482	142	0.5839	0.2279	0.05924	0.8711
145	1.238	0.1128	0.05683	1.408	145	0.5539	0.2177	0.05683	0.8285
146	1.217	0.1111	0.05600	1.384	146	0.5444	0.2145	0.05600	0.8149
148	1.177	0.1079	0.05448	1.339	148	0.5260	0.2081	0.05448	0.7886
170	0.8332	0.07951	0.04081	0.9535	170	0.3710	0.1530	0.04081	0.5648
173	0.7976	0.07647	0.03932	0.9134	173	0.3550	0.1471	0.03932	0.5414
183	0.6932	0.06739	0.03484	0.7954	183	0.3082	0.1295	0.03484	0.4726
201	0.5479	0.05448	0.02836	0.6307	201	0.2432	0.1045	0.02836	0.3761
203	0.5344	0.05325	0.02774	0.6154	203	0.2372	0.1022	0.02774	0.3671
237	0.3611	0.03713	0.01953	0.4178	233	0.1671	0.07406	0.02031	0.2615
243	0.3388	0.03500	0.01843	0.3922	243	0.1501	0.06704	0.01843	0.2356
286	0.2226	0.02365	0.01254	0.2588	286	0.09849	0.04525	0.01254	0.15628
290	0.2147	0.02286	0.01213	0.2497	323	0.07151	0.03346	0.009315	0.11429
361	0.1201	0.01320	0.007043	0.1403	361	0.05308	0.02522	0.007043	0.08535
383	0.1022	0.01132	0.006052	0.1196	379	0.04650	0.02224	0.006214	0.07495

#### Table ES-7. Calculated Exposure Rates at Distances of 1957 Survey around K-65 Silos

Note: The individual results shown are presented as output from the MicroShield computations. The significant figures shown in the total values are used only for intermediate calculations, and do not imply this degree of certainty in the results.

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#### APPENDIX F — OTHER RADIONUCLIDES IN AIRBORNE AND LIQUID EFFLUENTS

#### SOURCES OF OTHER RADIONUCLIDES

Several radionuclides other than uranium and thorium were released to the atmosphere in small quantities during FMPC operations. Radioactive decay of the isotope <sup>238</sup>U in natural uranium, for example, produced the decay products <sup>234</sup>Th, <sup>234m</sup>Pa, <sup>234</sup>Pa, <sup>234</sup>U, <sup>230</sup>Th, and <sup>226</sup>Ra. Also, decay of <sup>232</sup>Th, the predominant nuclide in natural thorium, produced <sup>228</sup>Ra, <sup>224</sup>Ra, <sup>228</sup>Ac, <sup>228</sup>Th, <sup>212</sup>Pb, <sup>212</sup>Bi, and <sup>208</sup>Tl.

In addition, recycled uranium, which was processed at the FMPC beginning in fiscal year 1961, introduced small amounts of fission and activation products into process streams.

Trace concentrations of transuranic radionuclides such as plutonium isotopes and <sup>237</sup>Np were introduced as contaminants in purified uranium received from DOE reprocessing sites.

#### QUANTITATIVE INFORMATION REPORTED FOR OTHER RADIONUCLIDES

#### **Airborne** Releases

There were no measurements made of the other radionuclides in airborne releases until 1985. Results of measurements made at that time are given in Table D-1 of the interim Task 2/3 report (Voillequé et al. 1991). The measurements were made for bulk dust samples from dust collectors serving Plants 1, 4, 5, 8, 9, and the Pilot Plant and for the Plant 8 scrubbers. These data were carefully examined in an attempt to verify the results. The data from Table D-1 are summarized below:

- Thorium-234 and its daughter, <sup>234m</sup>Pa, are present in the largest quantities by far. With the exception of <sup>230</sup>Th in Plant 1 dusts, all other radionuclides made only very small contributions to the total activity for each plant.
- The <sup>234</sup>Th/<sup>234m</sup>Pa ratio theoretically should be one, in all cases since secular equilibrium is attained very quickly for decay of <sup>234</sup>Th, as was determined through use of the RADDECAY computer code (Grove Engineering 1987). The measured ratio averaged about 1.4 for Plants 1 and 4, about 0.7 for the dust collectors of Plant 8, about 1.15 for the Pilot Plant, and about 2.8 for Plant 5.
- The maximum activity of <sup>234</sup>Th in the samples should have been about 333 microcuries per kg uranium, since this is the value at secular equilibrium. Values listed in Table D-1 of the Task 2/3 report (Voilleque et al. 1991) for Plant 1 and Plant 4 are about 43% higher than what would be expected at secular equilibrium. The value for Plant 8 is about 30% low, and the value for the Pilot Plant is almost exactly as expected at equilibrium. The mean value for <sup>234</sup>Th activity in samples from Plant 5, however, was about 15,000 microcuries per kg uranium as contrasted with the expected equilibrium

value of 333. It is clear that the very high analytical values for this plant are a result of a different mechanism than one which might explain discrepancies in values for the other plants.

#### Wastewater Discharges

Concentrations of plutonium isotopes and  $^{237}$ Np relative to that of uranium were measured in FMPC wastewater discharges over the period from 1976 through 1984. The results are listed in Table D-2 of the Task 2/3 report (Voillequé et al.1991). The mean concentration of  $^{239}$ Pu and  $^{240}$ Pu, combined, was 0.34 microcuries per kg U. The highest value was 2.1 in 1980. The mean concentration for  $^{238}$ Pu was 0.0094 microcuries per kg U. The highest value was 0.026 in 1984. The  $^{237}$ Np mean value was 0.16, and the highest value was 0.40 in 1982.

Concentrations of the radium isotopes,  $^{226}$ Ra and  $^{228}$ Ra, were also measured in the wastewater discharges and are also listed in Table D-2. Mean values for  $^{226}$ Ra and  $^{228}$ Ra were 5.6 and 15 microcuries per kg uranium, respectively. The highest value for  $^{226}$ Ra was 8.6 in 1976, and the highest value for  $^{228}$ Ra was 65 in 1977.

Relative concentrations of the fission products,  $^{137}Cs$ ,  $^{106}Ru$ ,  $^{99}Tc$ , and  $^{90}Sr$ , which had also been measured in liquid waste discharges from 1976 through 1984, are reported in Table D–3 of the Task 2/3 report in units of microcuries per kg uranium. Mean values for  $^{137}Cs$ ,  $^{106}Ru$ , and  $^{90}Sr$  are 19, 2.0, and 14, respectively. The mean value for  $^{99}Tc$  is 9.6 x  $10^3$ , which is higher than the other values by factors ranging from 500 to 4800.

#### VERIFICATION OF ANALYTICAL DATA

#### **Resolution of Questions Arising from Data Discrepancies for Airborne Releases**

Copies of the original analytical data sheets used in the construction of Table D-1 of the Task 2/3 report were inspected carefully. Gamma spectrometry was used to analyze for  $^{234}$ Th and its daughter  $^{234m}$ Pa. Radiochemical analysis was employed in the determinations of the other radionuclides.

It was found that the analytical data for  $^{234}$ Th, the major component of the bulk collector dust from the plant operations, were corrected to account for decay from the date sampled to the date analyzed. The time lag ranged from 36 to 63 days. No correction was made for  $^{234m}$ Pa. The  $^{234}$ Th values were corrected forward to the date of determination in order to compare them realistically with the  $^{234m}$ Pa values.

Table F-1 lists both the values for  $^{234}$ Th corrected to the date of determination and the reported values for  $^{234m}$ Pa. Table D-1 of the Task 2/3 report contained extremely low values for  $^{234m}$ Pa in the Plant 8 scrubber liquid. Examination of the analytical data sheets for scrubber liquid revealed that there were errors in transcription of the data to the table for the Plant 8 scrubbers which accounted for these very low reported values. Table F-1 lists the correct values for  $^{234m}$ Pa.

The table lists  $UF_4$  and  $U_3O_8$  dusts separately for Plant 5 because they represent data for dust collectors servicing different stages in the Plant 5 processing. Similarly, Plant 8 dust collectors are listed separately from the Plant 8 scrubbers.

As can be seen by inspecting the results in the table, the  $^{234}$ Th/ $^{234m}$ Pa ratio is much closer to 1 than was presented in Table D-1 of the Task 2/3 report.

Table F-1. Corrected <sup>234</sup> Th	Values and <sup>234m</sup> Pa	Values (Mean	Values for	Each
	Plant)			

Plant	<sup>234</sup> Th Concentration (microcuries per g U)	<sup>234m</sup> Pa Concentration (microcuries per g U)	Ratio <sup>234</sup> Th/ <sup>234m</sup> Pa
1	408	418	0.98
4	399	410	0.97
$5 (UF_4)$	463	603	0.77
5 $(U_3 \dot{O}_8)$	4902	6080	0.81
8 (dust)	303	331	0.92
8 (scrubber)	336	307	1.09
9	3112	729	4.27
Pilot Plant	332	287	1.16

The corrections made in the  $^{234}$ Th concentrations to produce Table F-1 answered some of the questions about the analytical results, but two remaining concerns needed to be addressed. First, almost all of the  $^{234}$ Th concentrations are somewhat higher than the value expected for secular equilibrium with  $^{238}$ U (333 microcuries per kg uranium). Second, the  $^{234}$ Th concentrations for Plant 5 and Plant 9 dusts are extremely high relative to the expected value for secular equilibrium.

The <sup>234</sup>Th concentrations for Plants 1, 4, 5 (UF<sub>4</sub> only), 8 (dusts only), 8 (scrubbers), and the Pilot Plant average about 18% higher than the expected value at secular equilibrium. The explanation for these somewhat high values may lie in the fact that other thorium nuclides as well as <sup>235</sup>U present in the samples, interfere in the analytical procedure (Weaver 1992, Condra 1992). These interferences would have produced high values for <sup>234</sup>Th.

The extremely high concentrations for  $^{234}$ Th reported for Plant 5 and Plant 9 dust may possibly be explained by the fact that the dust collectors served processes in these plants that involved melting and solidification of uranium metal. Two of the main operations in Plant 5 were (1) reduction of UF<sub>4</sub> with magnesium metal to produce derbies of uranium metal after solidification of liquid uranium, and (2) melting of derbies of uranium metal to produce ingots. Casting of large ingots was carried out in Plant 9. The U<sub>3</sub>O<sub>8</sub> solids accumulated on the surfaces of the uranium both before and after solidification as a result of oxidation by air. Some of the solid as small particulates became airborne and was exhausted to the atmosphere through plant stacks. The thorium daughters along with other impurities in the liquid uranium metal are reported to have separated from the liquid uranium and to have migrated to the surface (Dugan 1992). This migration resulted in higher than expected <sup>234</sup>Th concentrations in the U<sub>3</sub>O<sub>8</sub> solids which accumulated on the metal surface.

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#### **Resolution of Questions Concerning Wastewater Discharges**

The very high reported concentrations for  $^{99}$ Tc in wastewater may be explained by the fact that this radionuclide, unlike other fission products and transuranics, is present in anionic form (as TcO<sub>4</sub><sup>-</sup>) rather than as a cation. Cations are subject to retention on clays and humic acids in soils via ion exchange processes. Technetium is reported to be very soluble and mobile in soils, with low K<sub>d</sub> values relative to other fission products (Vandecasteele et al. 1989). Most of the wastewater from the FMPC stemmed from runoff from FMPC ground surfaces where it was in contact with soils (Voillequé et al. 1991).

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