



VOLUME II - PART A  
DOSE RECONSTRUCTION FEASIBILITY STUDY

TASKS 1& 2

A Summary of Historical Activities  
on the Oak Ridge Reservation with Emphasis on Information  
Concerning Off-Site Emission of Hazardous Material

Prepared by  
ChemRisk®  
A Division of McLaren/Hart

for the  
Tennessee Department of Health and the  
Oak Ridge Health Agreement Steering Panel



Oak Ridge Health Agreement Steering Panel

# **OAK RIDGE HEALTH STUDIES PHASE I REPORT**

## **Volume II - Part A - Dose Reconstruction Feasibility Study**

**Tasks 1 & 2: A Summary of Historical Activities on the Oak Ridge Reservation with Emphasis on Information Concerning Off-Site Emissions of Hazardous Materials**

Prepared by:

ChemRisk  
A Division of McLaren/Hart  
Environmental Engineering Corporation

for

The Oak Ridge Health Agreement Steering Panel and  
The Tennessee Department of Health  
Division of Environmental Epidemiology  
C1-130 Cordell Hull Building  
Nashville, Tennessee 37247-4913

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*This document was prepared as a team effort by the following ChemRisk employees:*

**Project Manager:**

Stephen R. Ripple, Chief Health Scientist

**Administrative Support:**

Angélica Perea, Secretary

**Principal Authors:**

Gretchen M. Bruce, Associate Health Scientist

John E. Buddenbaum, Senior Health Scientist

Jennifer K. Lamb, Senior Associate Health Scientist

Thomas E. Widner, Principal Environmental Scientist

ChemRisk®  
A Division of McLaren/Hart  
1135 Atlantic Avenue  
Alameda, CA 94501



## CONTENTS OF THE OAK RIDGE HEALTH STUDIES PHASE I REPORT

**Volume I** summarizes the activities of the Oak Ridge Health Agreement Steering Panel, other than the Dose Reconstruction Feasibility Study, during Phase I of the Oak Ridge Health Studies. It includes four major topics:

- **Executive Summary of the Oak Ridge Health Studies Phase I Report**
- **Health Studies Background and Overview**
- **Phase I Goals**
- **Conclusions and Recommendations for Phase I**

**Volume II** documents the study (referred to as the Dose Reconstruction Feasibility Study) to find out if enough data exist to estimate historical doses of chemicals and radionuclides to the public living around the Reservation. It is comprised of four parts:

- **Part A** addressing project Tasks 1 and 2 to identify the historical operations and emissions at each of the complexes and characterize the availability of environmental sampling and research data
- **Part B** addressing Tasks 3 and 4 to identify important environmental exposure pathways and contaminants released from the Reservation
- **Part C** addressing Task 5 to identify information regarding historical locations and activities of off-site populations that could potentially be affected by releases from the Reservation
- **Part D** addressing Task 6 to identify the hazards associated with substances used at the reservation



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## ACRONYMS, INITIALISMS, AND ABBREVIATIONS USED IN THIS REPORT

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AAF	American Air Filter Company
AEC	Atomic Energy Commission
AHRUU	A 1940s research project involving measurements of lanthanum-140 radiations
AIRS	Aerometric Information Retrieval System
ANC	American Nuclear Corporation
ANL	Argonne National Laboratory
ANP	Aircraft Nuclear Propulsion
APPR	Army Package Power Reactor
ARE	Aircraft Reactor Experiment
ARMS	Aerial Radiological Measuring System
ART	Aircraft Reactor Test
ARUU	A 1940s research project involving measurements of lanthanum-140 radiations
ASTR	Aircraft Shield Test Reactor
ATDL	Atmospheric Turbulence and Diffusion Laboratory
ATL	Applied Technology Library
AVID	Accelerated Vendor Inventory Delivery
AVLIS	Atomic Vapor Laser Isotope Separation
BCM	Bear Creek Mile
BMAP	Biological Monitoring and Abatement Programs
BNL	Brookhaven National Laboratory
BSR	Bulk Shielding Reactor
CARL	Comparative Animal Research Laboratory
CDC	Centers for Disease Control and Prevention
CEF	Critical Experiments Facility
CEP	Compliance and Environmental Policy
CERCLA	Comprehensive Environment Response, Compensation and Liability Act
CEW	Clinton Engineer Works
CF	Central Files
CFM	Cubic Feet per Minute
CNF	Central Neutralization Facility
Colex	Column exchange process used in Y-12 lithium separation
CRBR	Clinch River Breeder Reactor
CRF	Curium Recovery Facility
CRM	Clinch River Mile
CRRI	Clinch River Remedial Investigation
CWS	United States Army Chemical Warfare Service
DOE	Department of Energy
DOSAR	Dosimetry Applications Research Facility
EDIS	Engineering Design and Information System
EFPC	East Fork Poplar Creek
EFPCM	East Fork Poplar Creek Mile
EGCR	Experimental Gas-Cooled Reactor
EMD	Environmental Management Division
EPA	United States Environmental Protection Agency

## ACRONYMS, INITIALISMS, AND ABBREVIATIONS USED IN THIS REPORT

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ER	Environmental Restoration
ERDA	Energy Research and Development Administration
ESD	Environmental Sciences Division
F3P	The Fission Product Pilot Plant
FPPP	The Fission Product Pilot Plant
FRC	Federal Records Center
GDP	Gaseous Diffusion Plant
GM	Geiger-Muëller radiation detector
GSA	General Services Administration
HEPA	High Efficiency Particulate Air, as in HEPA filters
HF	Hydrogen Fluoride
HFIR	High Flux Isotope Reactor
HPRR	Health Physics Research Reactor
HRE	Homogeneous Reactor Experiment
HRE-2	Homogeneous Reactor Experiment No. 2 (also called the HRT)
HRT	Homogeneous Reactor Test (also called HRE-2)
ICPP	Idaho Chemical Processing Plant
IDC	Isotopes Development Center
K-25	Code name for the site of the Oak Ridge Gaseous Diffusion Plant
kW	kilowatt (1000 watts of power)
LAM	Local Area Monitoring station, or Laboratory Area Monitoring station
LASL	Los Alamos Scientific Laboratory
LION	Library Information Online Network
LITR	Low Intensity Training Reactor, later the Low Intensity Testing Reactor
LLWDDD	Low-Level Waste Disposal, Development, and Demonstration
MAC	Maximum Allowable Concentration
MED	Manhattan Engineering District
MeV	Million Electron Volts; a unit of radiation energy
MFL	Miller's Fluorinated Lubricating Oil
MIT	Massachusetts Institute of Technology
MMES	Martin Marietta Energy Systems, Inc.
MOU	Memorandum of Understanding
MPC	Maximum Permissible Concentration
mR	Milliroentgen, a unit used to express radiation exposure in air
MSL	Mean Sea Level
MSRE	Molten Salt Reactor Experiment
MTR	Materials Testing Reactor
MW	Megawatt (1 million watts of power)
NCC	National Computer Center
NEPA	Nuclear Energy for Propulsion of Aircraft
NOAA	National Oceanic and Atmospheric Administration
NPDES	National Pollution Discharge Elimination System
OGR	Oak Ridge Graphite Reactor
ORAU	Oak Ridge Associated Universities

## ACRONYMS, INITIALISMS, AND ABBREVIATIONS USED IN THIS REPORT

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ORGDP	Oak Ridge Gaseous Diffusion Plant
ORIC	Oak Ridge Isochronous Cyclotron
ORNL	Oak Ridge National Laboratory
ORR	Oak Ridge Reservation
ORRR	Oak Ridge Research Reactor (also called the ORR; ORRR used in this report)
ORTF	Oak Ridge Task Force
OSTI	USDOE Office of Scientific and Technical information
OUO	Official Use Only
PAH	Polycyclic Aromatic Hydrocarbon
PAHs	Polycyclic Aromatic Hydrocarbons
PAM	Perimeter Area Monitoring Station
PCA	Pool Critical Assembly
PCB	Polychlorinated Biphenyl
PCM	Poplar Creek Mile
PRFP	Power Reactor Fuel Processing
rad	Radiation Absorbed Dose, a unit used to express absorbed dose
RaLa	Radioactive Lanthanum
RAM	Remote Area Monitoring Station
RAP	Remedial Action Program
RCRA	Resource Conservation and Recovery Act
RCW	Recirculating Cooling Water
RHF	Reports Holding File
RIR	A reactor considered as potential source of radioactive barium/lanthanum (Its identity has not been determined)
S-1	The Executive Committee of the Office of Scientific Research and Development's Section on Uranium
S-50	The Oak Ridge liquid thermal diffusion enrichment plant
SCRUP	An ORNL program that processed Chalk River nuclear reactor fuel
SDRC	Site Document Response Center
SF	Source and Fissionable, as in "SF material" accountability
SNAP	Systems for Nuclear Auxiliary Power
SNL	Sandia National Laboratories
SRD	Secret Restricted Data
SRP	Savannah River Plant
STORET	Data Storage and Retrieval System
SWMU	Solid Waste Management Unit
SWSA	Solid Waste Storage Areas
TBP	Tributyl Phosphate
TDEC/DOE-O	Tennessee Department of Environment and Conservation, Department of Energy Oversight Division
TDHE	Tennessee Department of Health and the Environment
TDPH	Tennessee Department of Public Health
TDRH	Tennessee Division of Radiological Health
TDWPC	Tennessee Division of Water Pollution Control



## ACRONYMS, INITIALISMS, AND ABBREVIATIONS USED IN THIS REPORT

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TGFC	Tennessee Game and Fish Commission
TIC	Technical Information Center
TLD	Thermoluminescent Dosimeter
TRE	Trivalent Rare Earths, or Total Rare Earths
TRM	Tennessee River Mile
TRU	Transuranic, or Transuranium Processing Plant
TSCA	Toxic Substance Control Act
TSD	Technical Services Division
TSF	Tower Shielding Facility
TSF-SNAP	A SNAP reactor operated at the Tower Shielding Facility
TSPCB	Tennessee Stream Pollution Control Board
TSR-I	Tower Shielding Reactor Number 1
TSR-II	Tower Shielding Reactor Number 2
TURF	Thorium Uranium Recycle Facility
TVA	Tennessee Valley Authority
TWRA	Tennessee Wildlife Resources Agency
UAVLIS	Uranium Atomic Vapor Laser Isotope Separation
UCC	Union Carbide Corporation
UCCND	Union Carbide Corporation Nuclear Division
UCNI	Unclassified Controlled Nuclear Information
UNH	Uranyl Nitrate Hexahydrate
USAEC	United States Atomic Energy Commission
USAF	United States Air Force
USDI	United States Department of the Interior
USDOE	United States Department of Energy
USEPA	United States Environmental Protection Agency
USGS	United States Geological Survey
USPHS	United States Public Health Service
W, Site W	Code name for the site of the Hanford, Washington plutonium production plant
WOC	White Oak Creek
WOCE	White Oak Creek Embayment
WOCM	White Oak Creek Mile
WOD	White Oak Dam
WOL	White Oak Lake
X, Site X	Code name for the site of the Oak Ridge plutonium production plant (now the site of Oak Ridge National Laboratory)
X-10	Code name for the site of the Oak Ridge plutonium production plant (now the site of Oak Ridge National Laboratory)
Y, Site Y	Code name for the Los Alamos, New Mexico nuclear weapons laboratory
Y-12	Code name for the site of the Oak Ridge electromagnetic enrichment plant (now the site of the Y-12 nuclear weapons plant)

## VOLUME SUMMARY

The Phase I feasibility study has focused on determining the availability of information for estimating exposures of the public to chemicals and radionuclides released as a result of historical operation of the facilities at the Oak Ridge Reservation (ORR). The estimation of such past exposures is frequently called dose reconstruction. The work of Phase I has examined both the feasibility of performing dose reconstruction and also attempted, in a limited way, to examine a portion of the enormous volume of historical data to identify the releases from the facilities in the past having the highest potential to have caused harm to the health of the public.

The project work was composed of a number of individual tasks designed to meet the overall objectives of the Phase I Studies. The study tasks are numbered 1 through 7. The initial project tasks, **Tasks 1 and 2** were designed to identify and collect information that documents the history of activities at the ORR that resulted in the release of contamination and to characterize the availability of data that could be used to estimate the magnitude of the contaminant releases or public exposures. **Task 7** was designed to support the collection of many of the documents and data identified in **Tasks 1 and 2** in a library that could then be used in any future health studies. These three tasks represent the information collection portion of the project and included qualitative evaluations of the potential for activities to have produced significant contaminant releases.

A history of operations that are likely to have generated off-site releases has been documented as a result of **Task 1** activities. The activities required to perform this task involved the extensive review of historical operation records and interviews with present and past employees as well as other knowledgeable individuals. The investigation process is documented in this report. The time period that is covered is the 50-year span from 1942, when the federal government acquired approximately 58,000 acres of land for what became the ORR, through 1992 when the study began. Four large complexes code-named X-10, Y-12, K-25, and S-50 have been in operation at the ORR. Three of the complexes, Y-12, K-25, and S-50 were primarily dedicated to the production of enriched uranium during the early years of operation. S-50 was built near K-25 and operated for only a single year. The uranium enrichment process involves the separation of the uranium-235 required for nuclear weapons from the uranium-238 that is most abundant in nature. Y-12 subsequently became involved in the production of nuclear weapons components.

The major activities at the X-10 complex were much more varied than those of the other complexes, having included development of the world's first full-scale nuclear reactor, a chemical separation pilot plant to recover plutonium, and a wide range of activities related to applied research and development focused primarily on energy and the environment.

This report presents information with respect to each of the major complexes, as well as a number of other off-site areas of concern related to contamination from the ORR. While large volumes of information and documentation were found to be available for each of the major complexes, the nature and quality of the documentation differed considerably between the complexes. The complex that appears to have the most amount of information relevant to dose reconstruction efforts is X-10. While considerable information is available for the K-25 and Y-12 sites, historical activities involving the use and release of hazardous materials do not appear to be as well documented at these complexes. Much of the information that is available for K-25 and Y-12 remains in classified documents, many of which were reviewed for the purposes of the study. One of the objectives of the health studies is to obtain the declassification of the information relating to off-site health impacts.

The Task 1 investigations have led to the documentation of an overview of the activities that have taken place at each of the major complexes, including routine operations, waste management practices, special projects, and accidents and incidents. Historical activities that appear to warrant the highest priority in any further investigations were identified based on their likely association with off-site emissions of hazardous materials as indicated by the documentation reviewed or information obtained in interviews.

Task 1 efforts identified the following activities as having the highest priority for any future studies:

#### X-10 Complex

- The processing of radioactive barium/lanthanum (RaLa processing),
- Processing of short-decayed irradiated thorium by the Thorex process,
- Graphite reactor operations,
- Processing of graphite reactor fuel for plutonium recovery, and
- Waterborne and airborne waste disposal activities.

#### K-25 Complex (including S-50)

- The gaseous diffusion process, which was the primary source of uranium and technetium emissions,
- Feed facility and product and tails withdrawal, which were likely the primary source of release of uranium hexafluoride,
- Abnormal or accidental releases of uranium hexafluoride,
- Liquid waste disposal of a complex waste stream, and
- Some further investigation of the short-lived (1 year) S-50 plant.

### Y-12 Complex

- Electromagnetic separation and enrichment of uranium,
- Lithium separation and enrichment operations, the primary source of mercury release to the environment,
- Uranium weapon component manufacturing,
- Beryllium operations,
- Waste disposal operations, and
- Some further evaluation of the use and release of classified materials.

Information that is available to support the reconstruction of historical releases of hazardous materials and possible off-site exposures for these high priority activities is summarized in this report.

Task 2 focused on the development of an understanding of the environmental sampling and research data that are available to support any future dose reconstruction efforts. Information on the availability of environmental data was obtained from document reviews and personnel interviews. Sources of information included:

- Plant libraries and archives,
- DOE Oak Ridge Operations files,
- The Tennessee Valley Authority,
- The United States Environmental Protection Agency,
- The United States Geological Survey,
- The Tennessee Department of Health,
- The Tennessee Division of Radiological Health,
- The Tennessee Division of Water Pollution Control, and
- Interviews with current and former investigators.

Abstracts were developed to summarize approximately 100 environmental monitoring and research projects that characterize the historical presence of contaminants in areas outside of the ORR. Environmental monitoring data availability is summarized for each of the following environmental media:

- Surface water,
- The air or atmosphere,
- Aquatic and terrestrial food items (biological monitoring),
- Soil, and
- Drinking water and groundwater.

The large volume of information reviewed in the 100 studies is summarized in tables in this report. The tables indicate the time period during which samples were taken, the approximate location of the samples, the contaminants that were measured, the Oak Ridge Facility that was most likely the source of the contaminant, and the relative quantity of sampling data available in the study. These table summaries, as well as the abstracts of the studies, are designed to be used to identify data that can be used to support any further studies to quantify the historical exposure of the public to specific contaminants released from the Oak Ridge facilities.

## 1.0 INTRODUCTION

This report describes historical operations and releases at the Oak Ridge Reservation (ORR) near Oak Ridge in eastern Tennessee. The time period of concern is the 50-year span from 1942, when the federal government acquired approximately 58,000 acres of land for the purposes of building plants to support its secret weapons project, through 1992, when Phase I Health Studies investigations began.

### 1.1 THE OAK RIDGE RESERVATION

Development of the ORR by the Army Corps of Engineers began in 1942, under the names of the Kingston Demolition Range and the Clinton Engineer Works. The ORR quickly became the site of massive construction efforts that resulted in plants for full-scale production of enriched uranium and for the pilot-scale production of plutonium, both being radioactive elements needed for development of the two types of atomic weapons then under development by the top-priority, top-secret Manhattan Project. From a ninety-square-mile parcel of land from which all land-owners had been evicted in 1942, Oak Ridge grew into the fifth largest town in Tennessee at the end of the war, with an industrial complex that consumed one-seventh of all the electrical power being produced in the nation (Nichols, 1987).

The facilities that were developed on the ORR were code-named X-10, Y-12, K-25, and S-50. The X-10 site saw development of the world's first full-scale nuclear reactor, then called the Clinton Pile, to demonstrate the production of plutonium from natural uranium fuel. A chemical separation pilot plant was developed to recover minute quantities of plutonium from fuel discharged from the pile.

Development of processes to enrich uranium in its uranium-235 isotope (half-life of 703,800,000 years<sup>1</sup>) gave rise to the Y-12, K-25, and S-50 Plants. Multiple enrichment processes were concurrently developed because it was not clear which processes would work satisfactorily or which would be most efficient. The Y-12 Plant was built to enrich uranium by the electromagnetic process, using devices called calutrons. The K-25 Plant, including what was then the largest building in the world, was built to perform enrichment by the gaseous diffusion process. The S-50 Plant was built near the K-25 Plant to demonstrate the liquid thermal diffusion process.

After the gaseous diffusion process was chosen as the method of choice for uranium enrichment, the S-50 Plant was shut down. While large-scale electromagnetic enrichment operations also

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<sup>1</sup>Throughout this report, half-lives of radionuclides are frequently given in parentheses after the nuclide name in places where the half-life is a relevant property.

ceased, uranium chemical process development remained active at Y-12. The K-25 or Oak Ridge Gaseous Diffusion Plant (ORGDP) was active for approximately 40 years in uranium enrichment for weapons and power plant applications. After 1960, the K-25 Site was instrumental in development of advanced technologies such as the gas centrifuge and Atomic Vapor Laser Isotope Separation (AVLIS) processes for enrichment of uranium.

After the war-time efforts, the X-10 "Clinton Laboratories" site became the Oak Ridge National Laboratory (ORNL), and developed into the DOE's largest multi-program, non-weapons national laboratory. Research and development activities at ORNL have focused primarily on energy and the environment and basic science supporting that work. Precision machining and chemical processing capabilities in place at the Y-12 Plant led to its transformation in the late 1940s to a high-tech plant for processing of nuclear materials and production of weapons components.

The ORR currently encompasses the land area and facilities depicted in Figure 2-1. In the course of the activities that are summarized in this report, the ORR has seen a diversity of operations and a variety of material uses that likely exceed or rival those of any other facility in the world.

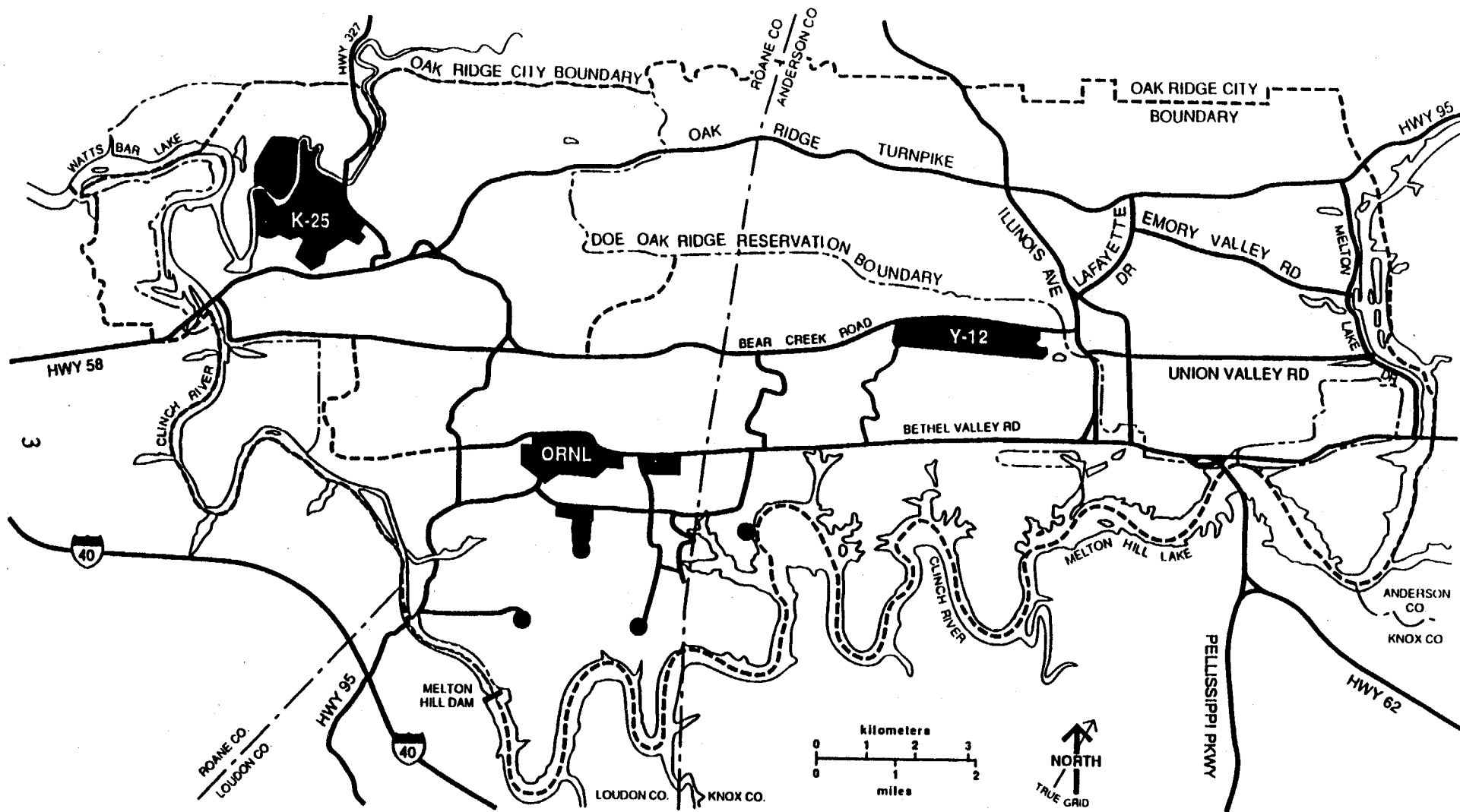
## 1.2 PHASE I HEALTH STUDIES

The State of Tennessee and the U. S. Department of Energy (DOE) in 1991 entered into an agreement for the State to conduct an independent assessment of human health risks to off-site populations that may exist as a result of past or present activities at DOE's Oak Ridge complex. This agreement is administered for the State by the Tennessee Department of Health.

Phase I of the health studies consists of a feasibility study and a screening process that will guide the direction of further efforts to quantify doses and health risks associated with possible exposures to contaminants that were released from the ORR since initial operations in 1943. These studies address doses and risks to off-site populations, not occupational exposures to plant workers, and the focus is on historical releases rather than projection of future doses or risks as is done in some risk assessment projects.

## 1.3 PROJECT TASKS 1 & 2

The one-year Phase I feasibility study is comprised of seven technical tasks, as described in Figure 2-2. The objective of Project Task 1 is to describe historical operations at the ORR and to identify the activities that have likely been associated with significant off-site releases of important materials. Task 2 focuses on identification and description of environmental monitoring and research data that would be available to support a dose reconstruction addressing historical releases of radionuclides and hazardous chemicals from the ORR facilities.



**FIGURE 2-1**  
**THE OAK RIDGE RESERVATION**  
**AND MAIN DOE FACILITIES**



Because Task 1 and Task 2 are similar in the nature of their investigations of historical information concerning ORR material uses, releases, and environmental transport, discussion of the progression and the findings of the two tasks are combined in this report to present a more comprehensive picture of the information that would be available to support a dose reconstruction. Because their missions have been so different, the three main plant areas are discussed separately in this report, followed by a section addressing environmental data and areas of concern that are not associated directly with the plants individually.

**TASK 1 - DESCRIBE HISTORICAL OPERATIONS AND RELEASES**

Activities that have been associated with significant off-site releases were described. This task focused on historical uses and releases of important materials.

**TASK 2 - IDENTIFY AVAILABLE ENVIRONMENTAL DATA**

Sources of environmental monitoring and research data were identified and described. The data that are available were evaluated for potential usefulness in conducting a dose reconstruction.

**TASK 3 - IDENTIFY COMPLETE EXPOSURE PATHWAYS**

Plausible routes of historical contaminant exposure to off-site populations were identified based on the characteristics of each material and the environmental setting at the Oak Ridge Reservation.

**TASK 4 - EVALUATE ENVIRONMENTAL EXPOSURE PATHWAYS**

Various screening methods were used to identify complete exposure pathways that may have been most significant. This task began the process of focusing any future dose reconstruction efforts.

**TASK 5 - CHARACTERIZE POTENTIALLY EXPOSED POPULATIONS**

Historical locations and activities of the populations likely to have been most affected by historical releases were described. General land uses that could have influenced exposures will were also documented.

**TASK 6 - DESCRIBE THE HAZARDS OF IMPORTANT CONTAMINANTS**

Summaries of current knowledge of toxic and hazardous properties were prepared for each of the materials that warrant further detailed study.

**TASK 7 - COMPILE AND INDEX PROJECT DOCUMENTS**

Selected documents relevant to the feasibility study were collected, categorized, summarized, and indexed for future reference.

**FIGURE 2-2: THE PROJECT TASKS OF PHASE I HEALTH STUDIES**

The availability and accessibility of information at the different plants on the Oak Ridge Reservation vary considerably. At the X-10 site, plant activities have been largely centered on research, development and demonstration of technologies dealing with energy, the environment, and related basic sciences. The roots of early activities at Clinton Laboratories and ORNL were in pilot plant operations, where workers set out to confirm the feasibility of proposed processes and obtain quantitative engineering data necessary for the design and operation of production plants. As a result of this emphasis, relatively large quantities of information are available concerning historical activities at the X-10 site. While the K-25 and Y-12 plants have been the sites of significant research and development activities, their primary focuses were to a larger part production oriented. As a result, historical activities at the K-25 and Y-12 sites, including information relevant to historical uses and releases of hazardous materials, were not as rigorously documented as were activities at the X-10 site.

Information is also more accessible at the X-10 site because very few details of historical activities at X-10 remain classified, and much information has been previously reviewed and determined to be publicly releasable. The K-25 and Y-12 plants have more records that remain classified or otherwise restricted, and relatively few documents have been reviewed with regard to their availability for public release. Members of the ChemRisk investigation team were able to gain full access to records for the X-10 site earlier than the K-25 and Y-12 sites. As a result of the greater availability and accessibility of information at the X-10 site, the investigation of historical ORNL operations progressed further during Phase I Health Studies than the parallel efforts at the K-25 and Y-12 plants.

## 2.0 THE X-10 SITE

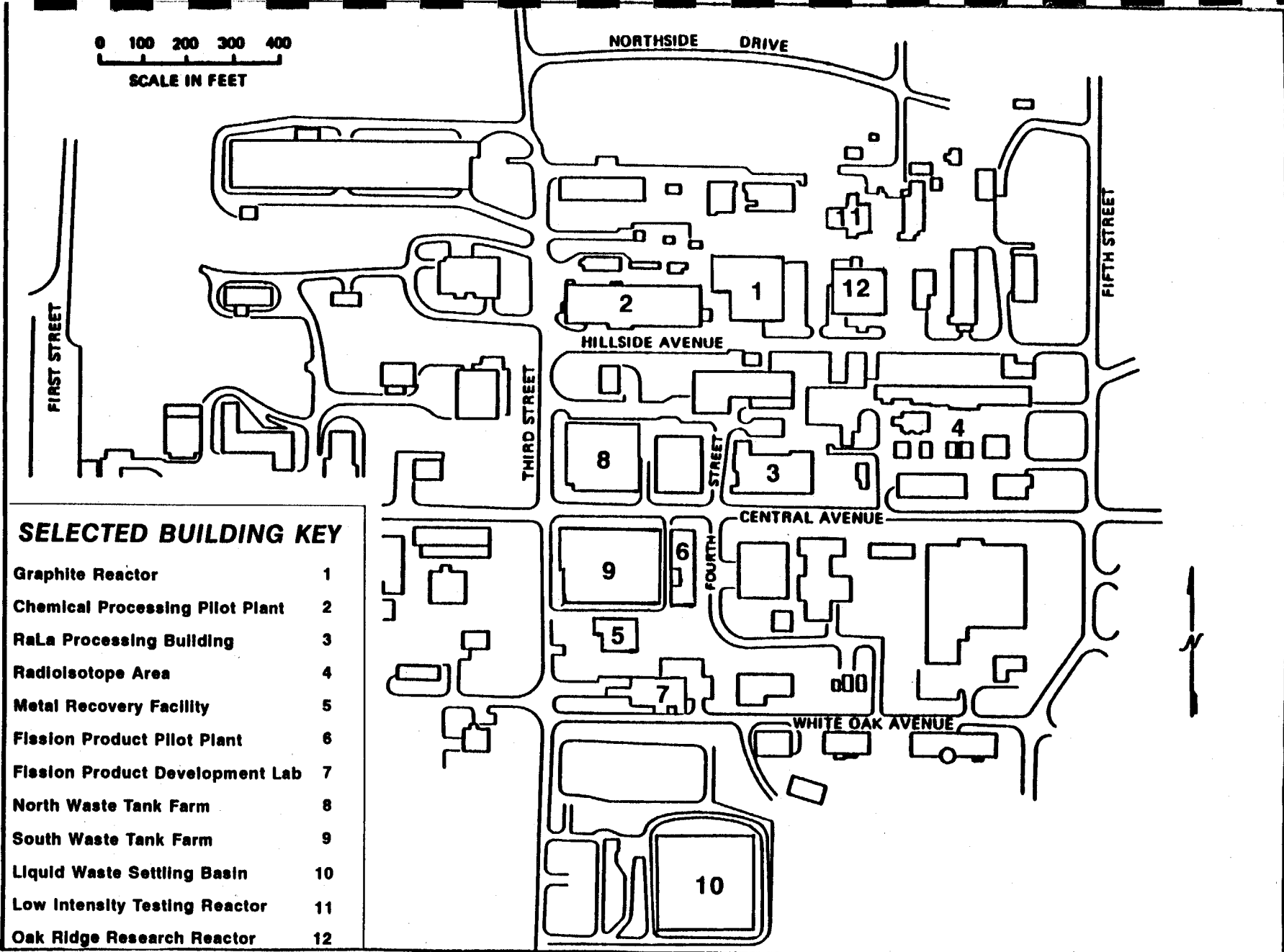
The X-10 site, now known as the Oak Ridge National Laboratory, is a government-owned, contractor-operated facility that has been known by a number of names and has been managed by various contractors, as shown in Table 2-1. Maps of modern-day X-10 facilities are shown in Figure 2-3 and 2-4. The discussion of historical X-10 plant operations and releases in this report first presents an overview of the activities that have taken place at the site, including routine operations, special projects, and accidents or incidents. A summary is given of the investigative process that was used by ChemRisk to review the historical records available at X-10 and to interview active and retired plant workers. Later in this section the historical activities which appear to warrant further quantitative evaluation because they have likely been associated with off-site releases of radionuclides or other hazardous materials are identified based on the qualitative information that has been reviewed. We have summarized the available information that would support reconstruction of associated historical releases and off-site exposures for these activities.

TABLE 2-1

HISTORICAL NAMES AND CONTRACTORS OF THE X-10 SITE

Name	Time Period	Operating Contractor
Clinton Laboratories	1/43 to 6/30/45	E.I. Dupont de Nemours for the University of Chicago
Clinton Laboratories	7/45 to 12/46	Monsanto Chemical Company
Clinton National Laboratory	1/1/47 to 2/28/48	U. S. Atomic Energy Commission
Oak Ridge National Laboratory	3/4/48 through 1949	Carbide & Carbon Chemicals Company, Union Carbide Corporation
Oak Ridge National Laboratory	1950 through 1955	Carbide & Carbon Chemical Division, UCC
Oak Ridge National Laboratory	1956 through 1963	Union Carbide Nuclear Company, Division of UCC
Oak Ridge National Laboratory	1964 - 1974	Union Carbide Corporation, Nuclear Division
Holifield National Laboratory	1975	Union Carbide Corporation, Nuclear Division
Oak Ridge National Laboratory	1976 to 3/30/84	Union Carbide Corporation, Nuclear Division
Oak Ridge National Laboratory	4/1/84 to present	Martin Marietta Energy Systems, Inc.

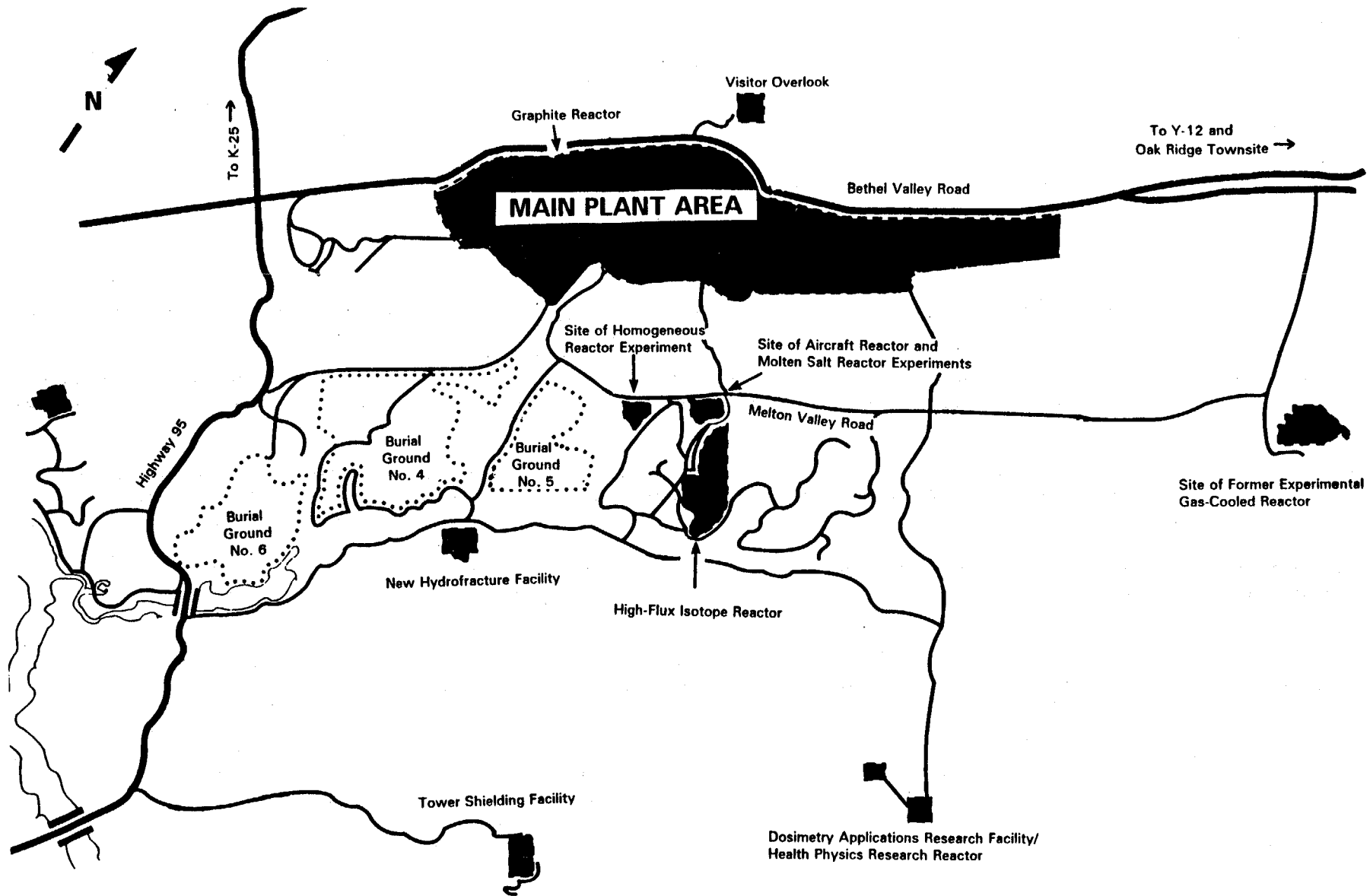
0 100 200 300 400  
SCALE IN FEET



**SELECTED BUILDING KEY**

Graphite Reactor	1
Chemical Processing Pilot Plant	2
RaLa Processing Building	3
Radiolotope Area	4
Metal Recovery Facility	5
Fission Product Pilot Plant	6
Fission Product Development Lab	7
North Waste Tank Farm	8
South Waste Tank Farm	9
Liquid Waste Settling Basin	10
Low Intensity Testing Reactor	11
Oak Ridge Research Reactor	12

FIGURE 2-3  
MAP OF OAK RIDGE NATIONAL  
LABORATORY  
CENTRAL AREA



**FIGURE 2-4  
MAP OF THE OAK RIDGE NATIONAL  
LABORATORY  
OUTLYING AREAS**

## 2.1 SUMMARY OF HISTORICAL ACTIVITIES AT THE X-10 SITE

This section presents a summary of many activities that have occurred at the X-10 site, beginning with the original mission of plutonium production and separation and extending to the modern-day national laboratory focusing on energy and the environment and related physical sciences. A chronology of key programs and activities that have occurred at the X-10 site is shown in Table 2-2. This discussion is not intended to be a complete presentation of all activities that have occurred at the X-10 site. Instead, an effort has been made to provide some description of the major programs or activities at the site that could plausibly have been associated with off-site releases of hazardous materials.

In selecting activities and materials that warrant priority in any further investigations of potential off-site exposures or health risks, the following criteria were applied:

- A material must be sufficiently toxic that it is capable of causing harm to the health of individuals who are exposed,
- The material must have been used in quantities sufficient to result in harmful concentrations after dilution or dispersion in the environment, and
- The manners in which the material was used or stored must have been associated with reasonable potential for release to the environment.
- Or, alternatively, there must be documentation of off-site releases or presence of the contaminant in the offsite environment.

The scope and time limitations of the feasibility study do not permit the evaluation and elaboration on each of the programs described. Additional details and analysis are provided later in this report for those activities that are believed to have the greatest potential for off-site releases based on the initial review. While it is difficult to make any definitive statements regarding the potential impacts from other activities without formal detailed evaluation, those activities that did not fall in the group of primary interest are believed to have significantly lower potentials for the release of contaminants off-site.

Section 2.3 and 2.4 focus on those historical activities that warrant primary focus for detailed investigation of historical release of important materials and the availability of information to support reconstruction of doses from these activities.

TABLE 2-2

## CHRONOLOGY OF MAJOR PROGRAMS AND ACTIVITIES AT THE X-10 SITE

Development of the Oak Ridge Reservation Began	1942
First Nuclear Chain Reaction at the Chicago Pile	December 1942
Clinton Pile Construction Began	February 1943
Chemical Processing Pilot Plant Construction Began	March 1943
Clinton Pile First Achieved a Nuclear Chain Reaction	November 1943
Chemical Separation of Plutonium was Performed	December 1943 through early 1945
Graphite Reactor Operated	November 1943 through November 1963
Materials Testing Reactor Development	Summer 1946 through late 1947
Low Intensity Testing Reactor	1948 through 1968
Bulk Shielding Reactor	1950 through 1991
Homogeneous Reactor Experiment	1951 to 1954
Tower Shielding Facility	1953 to 1993
Army Package Power Reactor Design	1954
Aircraft Reactor Experiment Operated	October 1954 (for 100 hours)
Homogeneous Reactor Test (HRE-2)	1954 to 1961
Project Aquarium Reactor	1955
Aircraft Reactor Test Design	1955 through 1958
Experimental Gas-Cooled Reactor Development	1957 through 1964
Oak Ridge Research Reactor	1958 through 1987
Molten Salt Reactor Experiment	1960 to early 1970s
Health Physics Research Reactor	1960 to 1987
High Flux Isotope Reactor	1961 to present
Bismuth Phosphate Chemical Processing	1943 to 1945
Radioactive Barium/Lanthanum Processing	1944 to 1956
Redox Chemical Processing	1945 to 1951
Hexone Chemical Processing	1946 to 1948
Uranyl Ammonium Phosphate Chemical Processing	1948 to 1949
Metal Recovery Chemical Processing	1948 to 1958
TBP-25 Chemical Processing	1948 to 1953
Purex Chemical Processing	1949 to 1960
Fluoride Volatility Chemical Processing	1949 to 1968
Thorex Chemical Processing	1952 to present
Transuranic Chemical Processing	1961 to 1976

### 2.1.1 The Original Mission

Many of the activities at the X-10 site have been tied in some way to the nuclear fission process. In some cases when a uranium-235 atom is bombarded with a neutron, it splits ("fissions") into two roughly equal pieces as shown in Figure 2-5. These fragments are actually atoms of lighter elements, and are called fission products. When uranium-235 atoms split, they also release enormous amounts of energy and between two and three additional neutrons. These secondary neutrons can in turn collide with other uranium-235 atoms that will release more neutrons, in turn colliding with other atoms, and continuously emitting energy. In this manner, a self-sustaining "chain reaction" occurs.

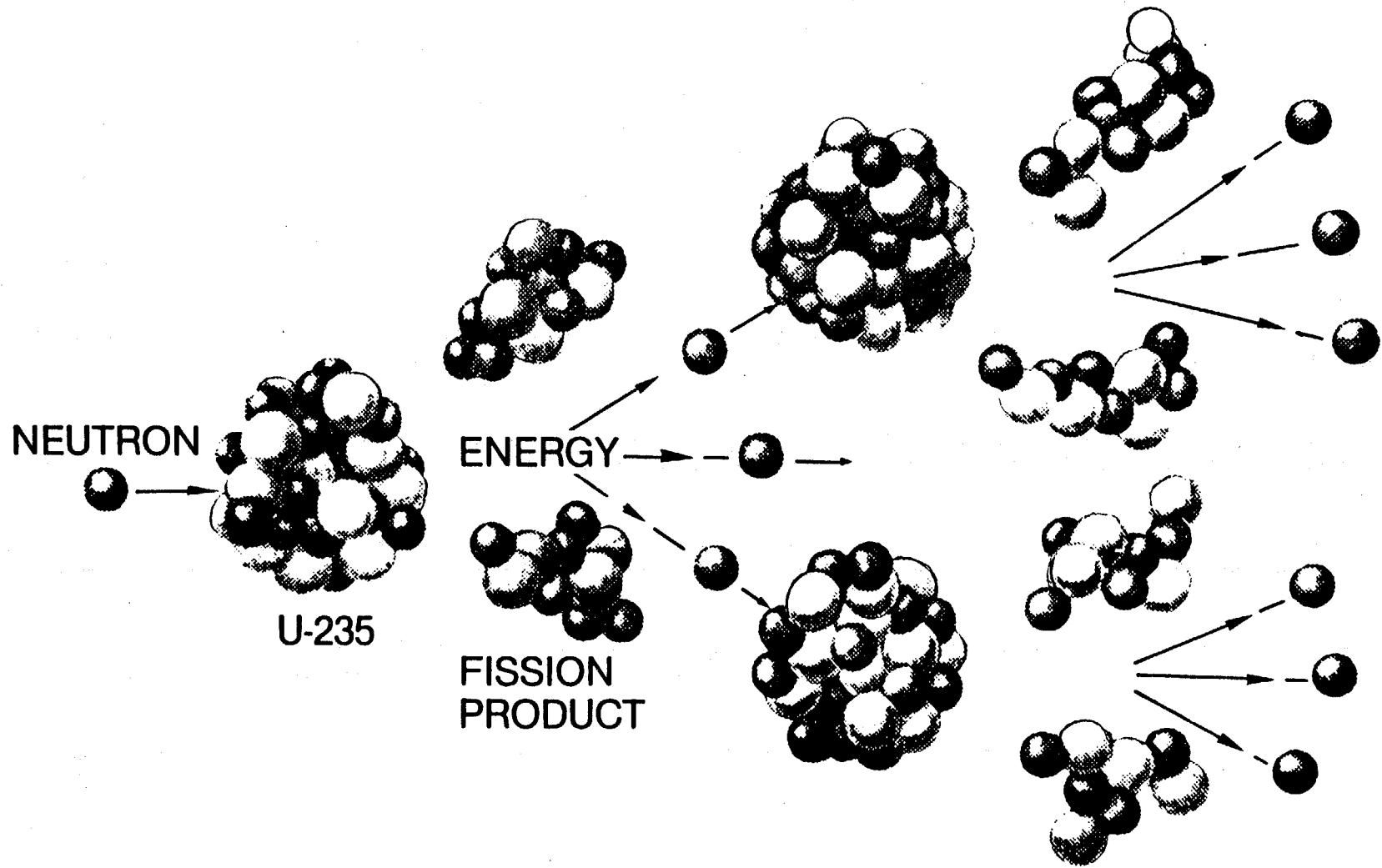
With the successful operation of Enrico Fermi's "Chicago Pile" on December 2, 1942 demonstrating that man could initiate and control a nuclear chain reaction (Allardice and Trapnell, 1967), construction of a plant for the production and chemical separation of plutonium was determined to be justified. The Executive Committee of the Office of Scientific Research and Development Section on Uranium, which was code-named S-1, decided that it would be safer to build the pilot plant and separation facilities in Tennessee than to place these experiments in a populous area such as the Argonne site near Chicago (Gosling, 1990). It was further decided for safety reasons that large-scale plutonium production facilities should be at an even more remote location near Hanford, Washington, where fewer people would be endangered by an accident (Thompson, 1963).

Construction of the Oak Ridge pilot plant facilities was accelerated so as much as possible could be learned and applied to the production facilities at the Hanford, code-named Site W. DuPont agreed to accept the responsibility for construction of the plutonium production plant and for the design and construction of the Tennessee pilot plant. Construction of the pilot plant began on February 1, 1943 on a remarkable rush schedule that placed the "Clinton Pile" in operation on November 4th of the same year (Thompson, 1963).

Because it was the first construction in the Clinton Engineer Works, the name Clinton Laboratories was chosen for the pilot plant being constructed by DuPont for operation by the Metallurgical Laboratory of the University of Chicago. The other main part of the Clinton Laboratories facility was the chemical pilot plant for separation and purification of plutonium. Design was complicated by the fact that chemical operations involving radioactive materials had never been attempted on any scale even approaching that planned for the pilot plant. Construction of the chemical separation plant started in March of 1943 (Gosling, 1990).

While DuPont had chosen an air-cooled design for the Clinton Pile, new neutron multiplication factor calculations by Fermi demonstrated that water cooling was feasible, and a water cooling system was chosen for the Hanford site with its abundant cold water. So although the Clinton





Source: Gosling, 1990.

FIGURE 2-5  
THE URANIUM-235  
FISSION CHAIN REACTION

Pile was not exactly a prototype for the production reactors at Hanford, much of the chemical separation process development work done at Clinton Laboratories saw application at Hanford.

The Clinton Pile has been known by various names, including the X-Pile, the ORNL Graphite Reactor, and affectionate nicknames such as "The Old Lady" and "Grandma" (Cagle, 1953). The term "pile" was used because early reactors were assembled by piling up blocks of graphite with pieces of uranium interspersed within them. The term "reactor" did not come into use until around 1952 (Johnson and Schaffer, 1992).

The following details of the construction of the Clinton Pile, which is depicted in Figures 2-6 and 2-7, were recorded by Cagle (1953). In the pile, graphite served as the moderator; in other words, it slowed down neutrons produced in the induced fissioning of uranium-235 (703,800,000 years) atoms, along with neutrons given off from fission products, so that the neutrons could be absorbed by other uranium-235 atoms to carry on the fission chain reaction. The Clinton Pile was constructed of a 24-foot cube of graphite blocks, four inches by four inches and up to 50 inches in length. These blocks were stacked in layers and keyed together at the edges. Layers that ran east-west had V-cuts in the blocks that formed 1,248 diamond-shaped holes running all the way through the graphite. Between the layers of graphite blocks containing channels were layers of solid graphite blocks. Some of these solid blocks were omitted, resulting in small tunnels called experiment holes in the reactor that were useful for research purposes. The graphite cube was entirely surrounded by a concrete shield 7 feet thick, with openings to give access for fuel loading and discharge and for experiments.

Graphite reactor fuel was in the form of 2½-pound metallic uranium "slugs" 4 inches long and slightly over 1 inch in diameter. They were clad in aluminum jackets approximately 0.0035 inch thick. The aluminum jackets protected the fuel from oxidation. The fuel slugs were loaded into the reactor channels by hand, end-to-end as shown in Figure 2-8, and positioned with measured metal rods to center them in the reactor. Cooling air passed by the slugs in the free space of the channels, drawn by two 900-horsepower centrifugal blowers that forced slightly over 100,000 cubic feet per minute up a 200-foot concrete stack to the environment. Exhausted air was not filtered during initial reactor operations.

In the fall of 1943, after thousands of slugs had been loaded, the reactor first achieved a self-sustaining nuclear reaction ("went critical") in the early morning of November 4th. Beginning in December of 1943, irradiated fuel was discharged from the reactor for use in the chemical processing plant. Discharging of slugs was accomplished, while the reactor was shut down, by pushing the slugs into the exit air manifold with steel rods. The slugs fell upon slanted, neoprene-covered, cadmium-plated stainless steel plates that funneled them into a 20-foot-deep water pit below the ground-level floor (Cagle, 1953). The slugs were then transported under water in buckets through a nine-foot-deep canal to the first shielded cell in the adjacent chemical processing building. The reactor was kept in operation for the purpose of producing plutonium

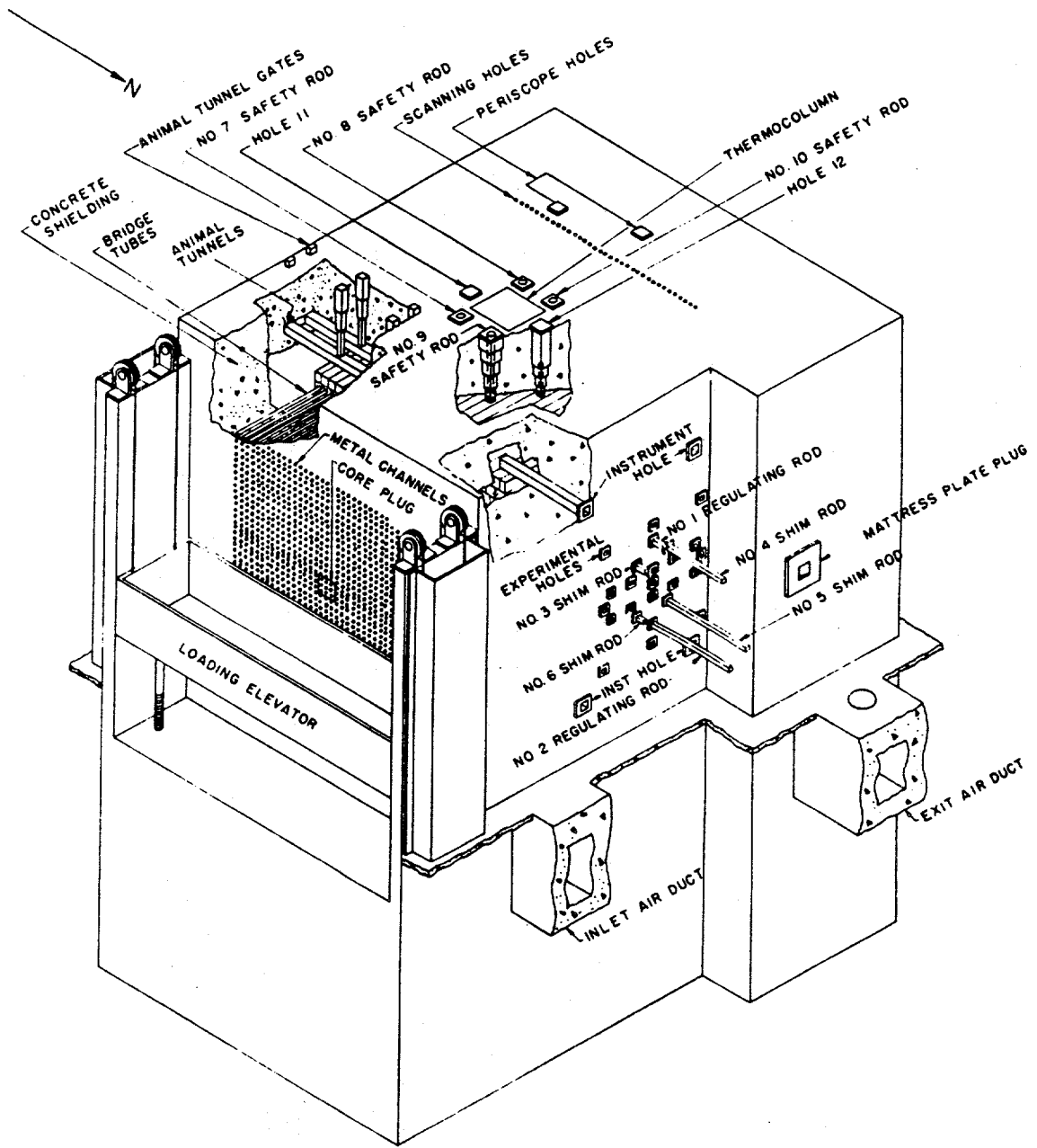


FIGURE 2-6  
 CUTAWAY SKETCH OF  
 THE OAK RIDGE GRAPHITE  
 REACTOR (LOADING FACE)

DWG NO 14587

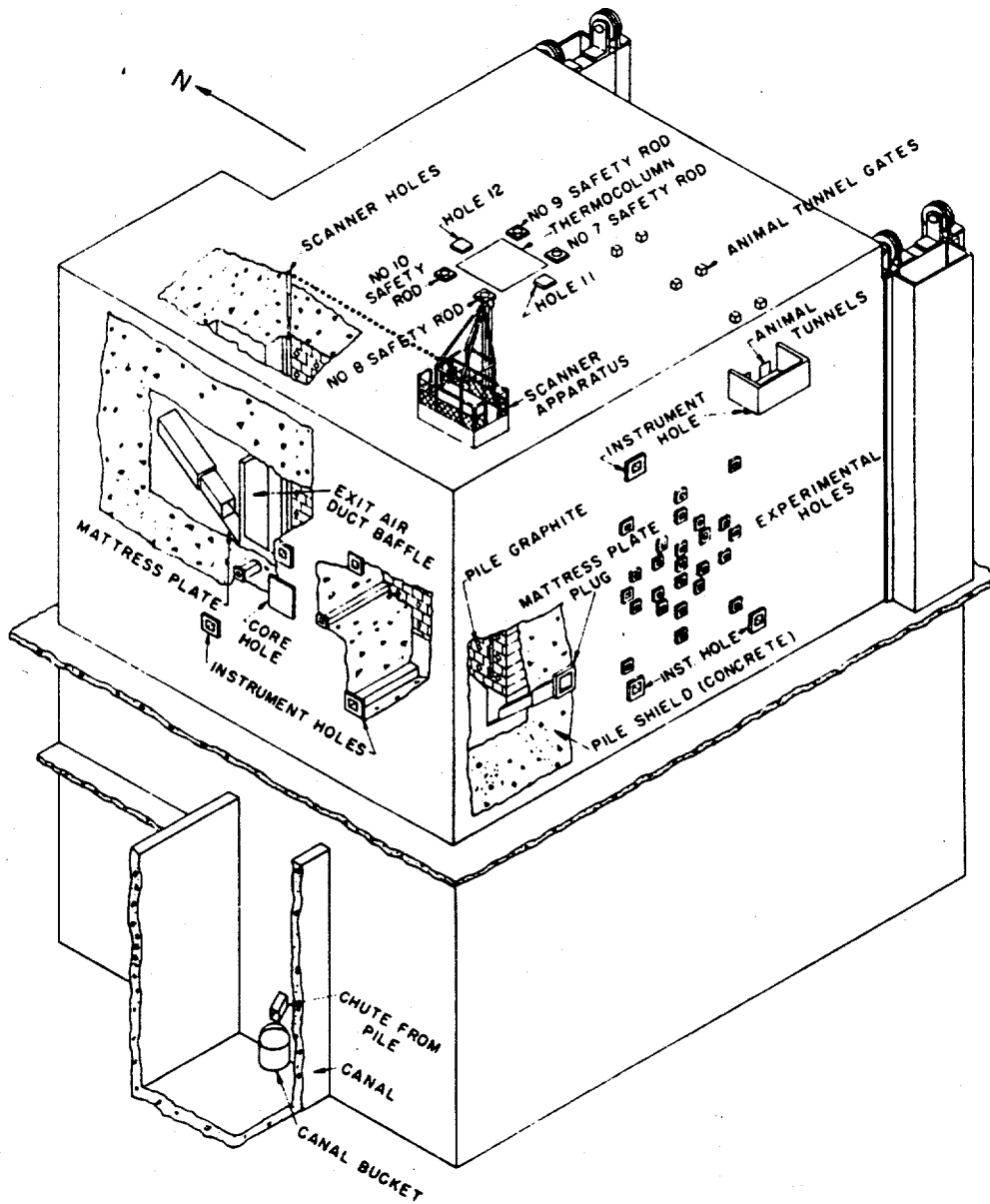


FIGURE 2-7  
CUTAWAY SKETCH OF  
THE OAK RIDGE GRAPHITE  
REACTOR (FUEL DISCHARGE FACE)

Rev. 08-18-93  
OakRdg89372-6



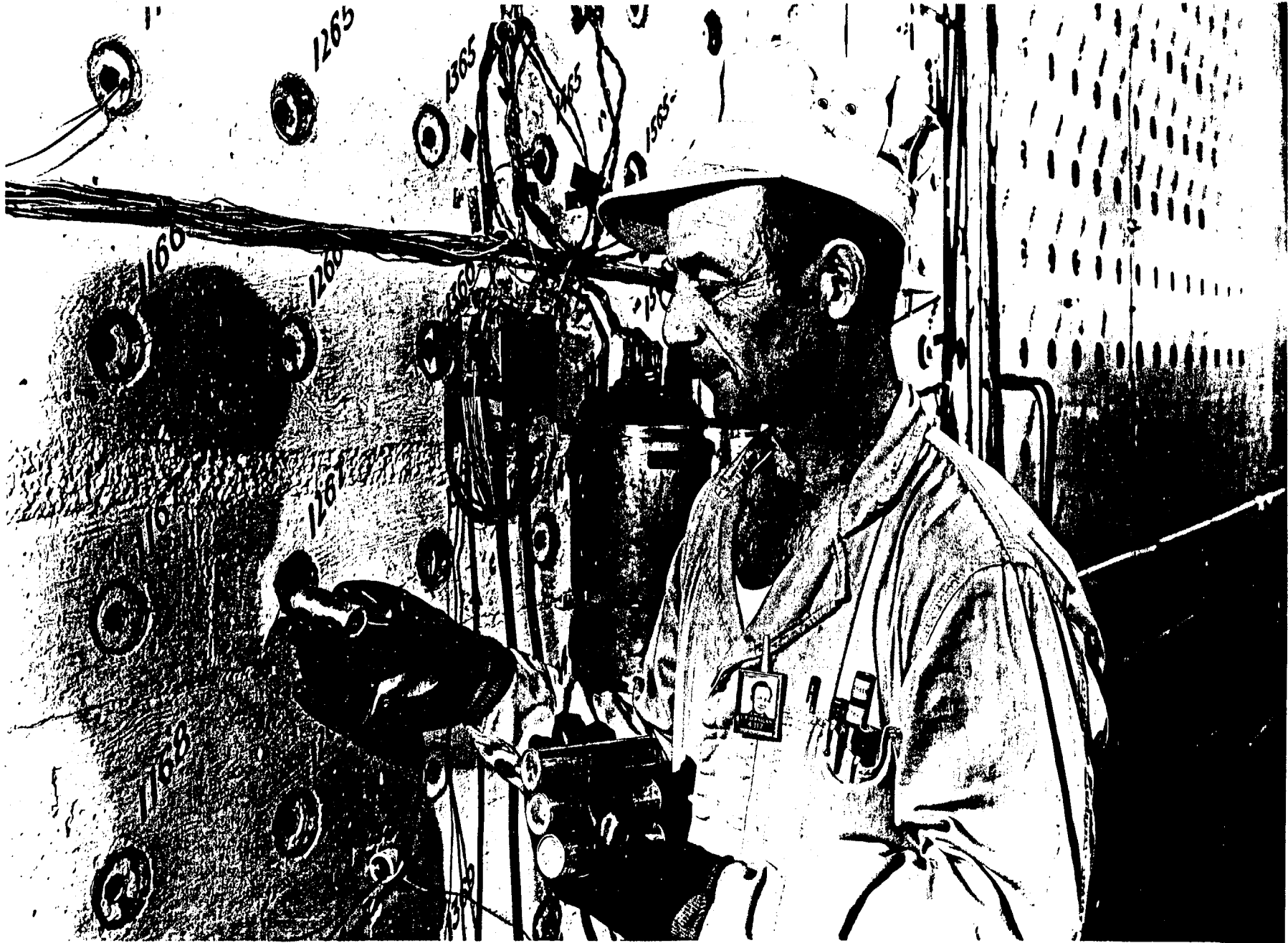


FIGURE 2-8  
WORKER INSERTING URANIUM  
FUEL SLUGS INTO GRAPHITE REACTOR

for experimental purposes until December 1, 1944, when applications of the pile shifted to research and radionuclide production.

Construction of the chemical separation pilot plant was nearing completion before a chemical process was selected (Jones, 1985). The purposes of the "pilot plant" were to test processes for plutonium separation, obtain data needed to design and operate a production plant, and provide plutonium for evaluation at other sites. The method selected for use in the chemical separation pilot plant was a "co-precipitation" process, in which a small quantity of a radioactive element is precipitated (made to separate from solution due to chemical or physical change) with a "carrier" of some other element. Bismuth phosphate was used as the carrier for the initial plutonium separations (Thompson, 1963). A "carrier" is a compound used in precipitation processes to maximize the separation of the desired product from undesirable contaminants, such as fission products. For example, plutonium and fission products were usually not present in concentrations high enough to be precipitated directly (Clark, 1945).

The steps in the early plutonium production process were as follows:

- Natural uranium fuel slugs were placed in the graphite reactor. Operation of the reactor caused fission products and plutonium to be formed in the uranium fuel slugs.
- After shut-down of the reactor, a number of fuel slugs were pushed out the back side of the reactor and fell to a water-filled canal below. After a period of cooling, batches of fuel slugs were transported through the canal into the first cell of the chemical processing pilot plant in the next building.
- The aluminum jackets were removed from the fuel slugs, typically by dissolving the aluminum in nitric acid with a mercury catalyst.
- The uranium metal was dissolved by addition of nitric acid and heating to 100-115°C and holding at 105°C until the specific gravity of the solution reached 1.75.
- The plutonium in solution was reduced by adding formic acid (HCOOH) at 100-105°C. The solution was diluted with water, and sulfuric acid was added.
- $\text{BiONO}_3$  and  $\text{H}_3\text{PO}_4$  were added to form a bismuth phosphate precipitate, which "carried" the plutonium out of solution.
- The solution was digested for 2 hours, cooled, and centrifuged. The plutonium-containing cake was washed three times with water, and then dissolved in nitric acid.

- Two decontamination cycles were performed to remove fission product contaminants. Each cycle consisted of a by-product precipitation with plutonium in the oxidized state followed by a product precipitation with plutonium in the reduced state.
- A concentration cycle was performed to reduce the volume of the plutonium solution from about 300 gallons to about 8 gallons, and to provide additional decontamination. The concentration cycle included a bismuth phosphate by-product precipitation, a lanthanum fluoride by-product precipitation, a lanthanum fluoride product precipitation, and a step called metathesis which converted fluorides to hydroxides that could be more easily dissolved in nitric acid.
- In final product isolation, hydrogen peroxide was added in two  $\text{PuO}_4$  precipitation steps. The precipitates were dissolved in nitric acid to form a concentrated solution of plutonium product.

(Clark, 1945)

Processing in the plant began in December 1943, using uranium irradiated at the Washington University cyclotron in St. Louis. The first few milligrams of plutonium were shipped to the Metallurgical Project in Chicago on December 30, 1943 (Thompson, 1963). By the end of 1943, chemists had successfully extracted 1.54 milligrams of plutonium from the slugs and sent it to Chicago, apparently by secret courier in a container resembling a fountain pen (Johnson and Schaffer, 1992). By the end of January 1944, the chemical pilot plant was processing one-third ton of irradiated Clinton pile uranium per day (Thompson, 1963). The first plutonium was sent to Los Alamos for weapon design work in February of 1944. By spring, the efficiency of the bismuth phosphate process was improved to the point that ninety percent of the plutonium in the slugs was recovered. By early 1945, when operation of the chemical separation pilot plant ceased at X-10, the graphite reactor and pilot separations plant had yielded a total of 326.4 grams of plutonium (Johnson and Schaffer, 1992).

### 2.1.2 Nuclear Reactor Development and Operations

After operation of the Clinton Pile was successfully demonstrated, nuclear reactors of various designs and purposes were developed at Oak Ridge. In general, nuclear reactors were developed to fulfill these purposes:

- To produce useful radionuclides in the fission process or by interaction of radiations with target materials
- To produce radiation fields useful for testing of radiation effects on materials, shielding tests, tests of other physical properties of materials, and medical treatment

- To generate power for generation of electricity
- To generate power for propulsion of vehicles

Nuclear reactors have the potential for environmental impacts during fuel fabrication, construction, routine operation, incidents, fuel processing, and decontamination and decommissioning. Quite a number of reactors have been designed at the Laboratory. Some of them have operated at the X-10 site, and others that did not actually operate at the Laboratory had significant impacts on the development of Laboratory programs and facilities in the post-war years. After World War II, management of the Clinton Laboratories shifted from General Leslie Groves and the Army Corps of Engineers Manhattan District to David Lilienthal of the newly created civilian Atomic Energy Commission (AEC). Under the AEC, Clinton Laboratories entered into a significant period of nuclear reactor development. Under the direction of Eugene Wigner, the laboratory designed two new reactor types; a reactor with high neutron output ("high flux") useful for testing materials, and the Daniels Pile for demonstrating nuclear energy's potential for producing electricity.

The high-flux reactor became known as the **Materials Testing Reactor (MTR)** because its chief function was to provide intense neutron bombardment for testing materials to be used in future reactors. The main features of the MTR design were parallel uranium fuel plates sandwiched between aluminum plates, with water used as the moderator and the coolant and with beryllium reflectors. The MTR design, which had evolved by the summer of 1946, was the prototype for many university research reactors and was a precursor of all light water reactors that later propelled navy submarines and ships and generated commercial nuclear power (Johnson and Schaffer, 1992). After much design work on the high flux experimental reactor was performed at Clinton Laboratories, the AEC decided on Christmas Eve of 1947 to replace the University of Chicago with Union Carbide as industrial contractor of the laboratory, to centralize reactor development at Argonne National Laboratory, and to construct the reactor at the Arco, Idaho reactor testing station. This period is remembered as "Black Christmas" by many at the laboratory, who felt that they had been double-crossed and that the decisions represented a demotion from national laboratory status to a radioisotopes and chemical processing factory (Johnson and Schaffer, 1992; Thompson, 1963).

A Power Pile Division was formed in July 1946, with its purpose being to design, construct, and operate a pile to produce useful power. The group was directed to proceed with design and construction of an experimental "**Daniels Pile**", named for Farrington Daniels at the Chicago Metallurgical Laboratory, who had in 1944 designed a reactor with a bed of enriched uranium "pebbles" moderated by beryllium oxide and cooled by helium gas. Support for the Daniels pebble-bed reactor waned by 1948, and the Power Pile Division was transferred to Argonne National Laboratory (Johnson and Schaffer, 1992).



While designing the MTR in 1948, the Laboratory built a small mockup of the reactor to test the design of its controls and hydraulic systems. In 1949 uranium fuel plates (made from uranium enriched at the Y-12 plant) were added to the mockup to test the reactor design under critical conditions (Unknown, 1993). The mockup served as a training ground for the Phillips Petroleum Company personnel who would operate the full-scale reactor in Idaho. The AEC later allowed the Laboratory to upgrade the shielding and cooling systems on the mockup, which raised its capacity to 3,000 thermal kilowatts or about 10% of that of the full-scale MTR. Known to some as the "poor man's pile", the mockup became formally known as the **Low Intensity Testing Reactor (LITR)**, and established the feasibility of the boiling water reactor, which later became one of the design prototypes for commercial nuclear power plants. The LITR, which was after a while operated remotely from the graphite reactor control room, remained in operation to support physics experiments until the AEC shut it down on October 10, 1968 (Johnson and Schaffer, 1992; Stapleton, 1992).

In the late 1940s, the U.S. Air Force sought to address the imbalance in air strike capabilities caused by the development of jet engines by the British and Germans. The Air Force urged the development of nuclear powered bombers to counteract the advantage of quick, defensive fighters over slower, long-range offensive bombers. Initial design concepts called for bombers that could fly at least 12,000 miles at 450 miles per hour without refueling; capabilities that would enable nuclear weapons to be delivered anywhere in the world. In 1946 the USAF awarded to Fairchild Engine and Airplane Corporation a contract to direct the Nuclear Energy for Propulsion of Aircraft (NEPA) project (Thompson, 1963; Johnson and Schaffer, 1992). In 1946, General Groves gave the Air Force approval to use the empty S-50 plant near the K-25 plant for investigations of the feasibility of nuclear propelled aircraft.

Initial studies by Fairchild at S-50 showed promise, and the AEC asked the Massachusetts Institute of Technology in 1948 to evaluate the feasibility of nuclear-powered flight. MIT formed the Lexington Project. The MIT group concluded that there was a strong possibility that some version of nuclear-powered flight could be achieved within 15 years if adequate resources and manpower were put into development of reactors, power plants, materials, and other components. The aircraft would require a reactor small enough to fit inside a bomber, adequate radiation shielding to protect the crew, and enough power to get the craft airborne. In September 1949, the AEC approved the Laboratory's participation in the project, and the Laboratory's Aircraft Nuclear Propulsion Project (ANP) was formed. General Electric took over the work of Fairchild soon after, and relocated to Ohio. About 180 Fairchild workers stayed in Tennessee to join the ANP project (Johnson and Schaffer, 1992).

Much of the ANP work at the Laboratory focused on development of effective but lightweight shields to protect aircraft crew members and materials. To support this work, the Laboratory constructed several significant new research facilities. In December 1950, ORNL completed its two-megawatt **Bulk Shielding Reactor (BSR)**, a modified "swimming pool" version of the

earlier MTR design. The reactor's enriched uranium core was submerged in water for both cooling and neutron moderation, and could be moved around in the concrete "swimming pool" tank to test bulk shielding materials in various configurations. In addition, a 10 kilowatt **Pool Critical Assembly (PCA)** was designed and constructed in the Bulk Shielding Reactor pool so that smaller scale experiments could be performed without tying up the larger reactor. Upgraded with a forced cooling system in 1963, the BSR took the place of the graphite reactor, which was retired that same year, as a general purpose research reactor until DOE ordered all research reactors shut down in March 1987 and issued permanent shutdown orders for the BSR on September 10, 1991. (Thompson, 1963; Johnson and Schaffer, 1992; Stapleton, 1992).

In 1953, ORNL completed the **Tower Shielding Facility (TSF)**, which enabled operating reactors to be hoisted nearly 200-feet into the air to enable studies of the behavior of radiations from airborne reactors with a minimum of the scattering experienced when operating on the ground. There was no shielding around the reactor itself. The TSF has housed four different reactors since it was first built, three of which were operated while suspended from the towers. The original Tower Shielding Reactor (TSR-I) was a box-shaped 500-kilowatt MTR-type reactor. For a brief period beginning in 1958, the Aircraft Shield Test Reactor (ASTR), used for shielding research in an operating aircraft, replaced the TSR-I. The TSR-I was replaced in 1960 with a spherical TSR-II, which is still in use. In 1967, the TSF-SNAP (Systems for Nuclear Auxiliary Power) reactor was added to be used alternately with the TSR-II for shielding studies (Thompson, 1963; Johnson and Schaffer, 1992; Stapleton, 1992; Schaeffer, 1992).

In what was known as "**Project Aquarium**", ORNL in early 1955 designed and fabricated a scaled-down (100 kilowatts instead of 30,000) version of the MTR for transport to the Geneva Conference as part of the U.S. "Atoms for Peace" exhibit. The reactor, the first to use uranium dioxide fuel, was tested in the BSR pool, disassembled, and shipped by air from Knoxville to Geneva, where an ORNL team reassembled and tested it. The reactor was designed, built, tested, transported to Geneva, and reassembled in only five months (Winters, 1955; Johnson and Schaffer, 1992; Stapleton, 1992).

Still another modification of the MTR design was the **Army Package Power Reactor (APPR)** developed at ORNL. The unit was designed as a compact nuclear power unit that could be installed at remote or relatively inaccessible locations where nuclear power costs would be competitive and the long-lived fuel would be advantageous. Conceptual design of the system was completed by July 1954. The Army decided to have a full-scale prototype station constructed at Fort Belvoir, Virginia. On April 8, 1957, the APPR-1, constructed by Alco Products Inc. (American Locomotive Company), went critical. The Army later used similar package reactors for power and heat generation in the Arctic and other remote bases (Johnson and Schaffer, 1992).

When basic research in the fields of physics and chemistry, irradiation evaluations of materials, and testing requirements for power reactor fuels demanded a reactor with a higher neutron flux than the graphite reactor, ORNL built the **Oak Ridge Research Reactor (ORRR)**. The ORRR combined the features of the MTR and a swimming-pool reactor. It was cooled and moderated by water, with beryllium and water providing the necessary neutron reflection. It used uranium-aluminum alloy plate-type fuel, similar to that of the MTR, in a core that was enclosed in tank so that it could be cooled with high-velocity water. The entire reactor tank was then immersed in a pool of water. The reactor core tank was cylindrical and flattened on one side. One side had beam hole tubes which extended through the concrete shield; the flat side was clear of obstructions and faced the main pool of water so that experiments or materials could be placed there for irradiation in a zone of high neutron flux. The ORRR reached design power in March 1958. The ORRR operated until it was shut down in 1987 along with the rest of ORNL's reactors. The ORRR supported experiments in many fields of science, including nuclear and solid-state physics, and the reactor was for some time the primary supplier of radionuclides to the free world for research, medicine, and industry applications (Thompson, 1963; Johnson and Schaffer, 1992; Stapleton, 1992).

According to official ORNL reports, during the night of June 30-July 1, 1963, some fission products were released from the ORRR fuel into its water systems. It was discovered later that a large neoprene gasket had blocked cooling water flow to one fuel element. The release from the fuel element was probably less than one or two minutes in duration, with about 1000 curies of volatile fission products (principally xenon-138 (14 minutes) and krypton-88 (2.84 hours)) escaping from the fuel. The curie (Ci) is a unit of radioactivity, with one curie being the quantity of radioactive material in which 37,000,000,000 atoms disintegrate every second. There was some contamination of the building atmosphere (up to 1 picocurie per cubic meter) by the cesium-138 (32.2 minutes) and rubidium-88 (17.8 minutes) formed as decay products of these noble gases. Stack samplers also indicated that 150 to 200 millicuries of iodine were released. The contaminated water was treated by demineralization and eventually discharged to Laboratory waste system. The faulty fuel element was removed from the reactor without difficulty, and the reactor was brought to full power on July 2nd. (Sims and Tabor, 1964; Boyle et al., 1982).

While much work on the aircraft nuclear propulsion project at ORNL centered on design of radiation shielding, another aspect focused on development of a suitable reactor for the application. ORNL constructed a small, high-temperature reactor engine that used circulating molten uranium salts as its fuel. The 1000-kilowatt **Aircraft Reactor Experiment (ARE)** reactor was operated at full design power for approximately 100 hours during October 1954, as planned. While the reactor was operating, every part of the fuel system was red hot, including the reactor core, piping, pumps, valves, heat exchangers, and all other components. The liquid fuel was pumped by a centrifugal pump to a heat exchanger that was cooled by a stream of

circulating, high-velocity helium that in turn went through a water-cooled heat exchanger. (Thompson, 1963; Johnson and Schaffer, 1992).

Following successful operation of the ARE, ORNL began development of a larger, 60 megawatt molten salt **Aircraft Reactor Test (ART)** reactor. The ARE building was expanded to make room for the second, spherical prototype known as the "fireball reactor" to conduct more sophisticated experiments such as those to address effects of flying upside-down or of a mid-air explosion. In early fiscal year 1958, the national program of aircraft nuclear propulsion development came under congressional fire because of its high costs and changing military requirements. President Kennedy canceled major portions of the national ANP program, and design of the ART was completed and "put on the shelf" (Johnson and Schaffer, 1992; Thompson, 1963).

In 1957, the AEC made ORNL responsible for designing fuel elements for an **Experimental Gas Cooled Reactor (EGCR)** to be built in Oak Ridge. By early 1958, the Laboratory had completed a design for a helium cooled, graphite moderated reactor with a core of uranium oxide clad in stainless steel. Graphite-coated particles were also considered for use in fuel elements. With the cooperation of the Tennessee Valley Authority, the AEC began construction of an experimental gas cooled reactor on the shore of Melton Hill Lake near the Laboratory. This reactor was to serve as a power-generating prototype, with eight test-loops also inside the reactor for scientists to test various fuel elements. In 1964, the AEC ordered the project halted, even though construction of the reactor had been completed and its fuel elements had been fabricated and tested. The reactor design had become obsolete before it was operational. (Johnson and Schaffer, 1992; Thompson, 1963).

Based on the success of the molten-salt ANP reactor, ORNL in 1956 began investigating other applications of molten salt technology. Molten salt fuel could function at high temperatures at low pressures in a liquid system that could be continuously cleansed of fission products without shutting down the reactor. On the other hand, molten salts are highly corrosive and present significant materials problems. ORNL developed a nickel-molybdenum alloy, called INOR-8, for containment of molten salts at high temperatures. By 1960, ORNL was designing a **Molten Salt Reactor Experiment (MSRE)** that promised to be an efficient thermal breeder and producer of low-cost nuclear power. A breeder reactor is one that produces more fissionable material during its operation than is destroyed in its fuel. In the case of the Oak Ridge MSRE, the fissionable material was produced from "fertile" thorium contained in a molten mixture in a "blanket" that surrounds the molten salt uranium-235 reactor. "Fertile" materials are nuclides such as uranium-238 and thorium-232 that can be transformed by neutron absorption followed by beta decay into fissionable plutonium-239 and uranium-233, respectively. A "blanket" is a region of fertile material arranged around a reactor to capture neutrons and produce more fissionable material. The molten salt reactor produced neutrons which led to the transformation

of thorium-232 (14,050,000,000 years) into uranium-233 (159,200 years), the same radionuclide used in the reactor fuel.

The thorium breeding concept was attractive because of the virtually inexhaustible supply of thorium found in granite rocks throughout the earth's crust. Use of uranium-233 reactor fuel from thorium was referred to as "burning the rocks." The MSRE, which occupied the same building as the earlier ARE, began operation around 1967 (ORNL, 1965; ORNL, 1968). Molten salt experiments continued into the early 1970s, with the molten salt reactor demonstrating its capability of using the thorium to uranium-233 fuel system in 1969 (Johnson and Schaffer, 1992).

Oak Ridge played a role in an AEC plan in the 1950s for development of five new demonstration nuclear reactors; one being an aqueous homogeneous fuel reactor to be developed at ORNL. A large thorium breeding power reactor was the ultimate goal. The homogeneous reactor was called such because it combined fuel, moderator, and coolant in one water-based solution. Homogeneous reactors had been tried around 1945, but the concept was dropped then because of significant problems, including the formation of bubbles in the liquid fuels and corrosive attack on structural materials. With the new knowledge gained in the ANP program and help from chemical engineers acquired from Y-12, ORNL built a small experimental **Homogeneous Reactor Experiment (HRE)** reactor. The HRE building was completed in 1951, the reactor in January 1952, and a test run began in October 1952. The design power level of one megawatt was achieved on February 24, 1953. High pressure steam from the reactor was fed to a turbine/generator to generate 150 kilowatts of electricity and earn HRE operators the honorary title of "Oak Ridge Power Company." The HRE operated successfully through a program of tests to determine its characteristics and behavior through 1953 and into 1954. Its objectives complete, the HRE was dismantled in 1954 (Johnson and Schaffer, 1992; Thompson, 1963).

ORNL then obtained approval to build a large pilot plant with a "two-region" homogenous reactor core. The aim was not only to produce electrical power, but also to irradiate a thorium "blanket" surrounding the reactor, thereby producing fissionable uranium-233 in the process. The design of the **Homogeneous Reactor Test (HRT)** or HRE-2 began in January 1954 and construction began in July of the same year. Construction was completed in early fall of 1956, and the reactor achieved criticality on December 27, 1957 and full power in February of 1958. The HRT gave operators trouble from the start. In April 1958 a leak developed that permitted fluid to be transmitted from the core region to the blanket region. The reactor was operated for some time before being shut down for inspection. No major hole or crack was found, and the reactor was restarted in June with fuel and heavy water solutions circulating in both the core and blanket systems. The reactor operated intermittently through 1958 and 1959. During 1958 the HRT achieved a record run of 105 days before being shut down because of clear indications that a second hole had developed in the core tank. Uranium was apparently separating from solution

and depositing on the tank walls, leading to excessive heat generation and the burning of holes through the tank wall at these spots. The two holes that had formed were plugged by remote maintenance, but the AEC requested that ORNL operate the HRT at near full power for two or three months and then terminate homogeneous reactor development by July 1, 1961. The HRT was operated at full power in January, February, March, and April until it appeared on April 28, 1961 that the plug in the hole of the core tank wall had disintegrated. The HRT was then dismantled for detailed inspection, and the homogeneous reactor program was closed out (Thompson, 1963; Johnson and Schaffer, 1992).

By the late 1950s the Soviets and Europeans had designed reactors with neutron fluxes that surpassed those available in the United States. ORNL officials argued for construction of a more powerful reactor for scientific studies of the transuranic elements, that is those elements beyond uranium on the periodic table (i.e., with more protons than uranium) and the production of radionuclides. The result was the **High Flux Isotope Reactor (HFIR)**, which was constructed from June 1961 through early 1965 and first achieved criticality on August 25, 1965. The HFIR is a 100 megawatt flux-trap type reactor in which neutrons are "trapped" in a five-inch diameter hole in the center of the highly-enriched uranium-235 HFIR fuel region. Targets, typically containing curium-244 (18.11 years) and other transuranic radionuclides, are placed in the flux trap region for intense thermal (moderated) neutron bombardment in order to form transuranic radionuclides such as berkelium-249 (320 days), californium-252 (2639 years), einsteinium-253 (20.467 days), and fermium-257 (80 days). Californium-252, which undergoes spontaneous fission in a fraction of its decays, is used for nuclear reactor startup neutron sources, compact energy sources in space and other research, and in cancer treatment (ORNL, 1991; Stapleton, 1992; Johnson and Schaffer, 1992).

By December 1973, the HFIR had completed its 100th fuel cycle, each lasting about 23 days. By the time it temporarily shut down in late 1986, the HFIR achieved a record of operation time unsurpassed by any other reactor in the United States. In 1986 tests on irradiation surveillance specimens indicated that the reactor vessel was being embrittled by neutron irradiation at a faster rate than predicted. The HFIR was shut down for extensive reviews and evaluation, and was restarted at a reduced power level on April 18, 1989, only to be shut down again for nine months due to questions of procedural adequacy. Startup was resumed in January 1990, with full power reached on May 18, 1990. In addition to production of transuranic radionuclides, the HFIR has been used for many irradiation experiments using facilities that allow insertion of samples into the flux trap region, into the region of the beryllium reflector, and using beam tubes that allow neutrons to be beamed out to experimental facilities outside the reactor shielding (ORNL, 1991).

In addition to constructing the HFIR in the 1960s, ORNL built a "fast burst reactor", later known as the **Health Physics Research Reactor (HPRR)**. The HPRR was designed and constructed at ORNL, tested in the ORNL critical experiment facility in 1961, loaned to the Nevada Test Site for a few months in 1962, then installed at the ORNL Dosimetry Applications Research Facility (DOSAR) in 1962. The HPRR was a small, unmoderated, unshielded reactor placed in a cylinder nine inches high and eight inches in diameter. Its operation released a neutron pulse useful for health physics and biochemical research. The reactor was designed to operate for very short "bursts" that yielded up to  $10^{17}$  fissions. It could also sustain steady-state operation at power levels from 0.1 watt to 10 kilowatts, and at power levels under 100 watts could operate for any length of time using natural convection air cooling.

The small HPRR is contained in a steel-structure, aluminum sided building and is supported by a large, track mounted positioning device called a transporter. The reactor building is located in a hollow and is surrounded by hills at least 50 feet high to provide natural shielding and to prevent "line-of-sight" viewing of the reactor in all directions. Since the reactor is not shielded, the HPRR control building is located behind a hill 900 feet from the reactor building, and the area is protected by a 1,000-foot-radius antipersonnel fence and a 3,000-foot-radius outer perimeter fence to keep intruders from the area during reactor operation. The HPRR was shut down in 1987 by DOE orders along with the rest of ORNL's research reactors. The reactor had been operated 3,000 times in the steady-state mode and 1,000 times in the pulse mode (Lundin, 1962; Stapleton, 1992; Johnson and Schaffer, 1992).

### 2.1.3 Nuclear Material Separation Processes

A significant activity of the Clinton Laboratories and ORNL has been the development of processes for recovery of product materials from highly irradiated reactor fuel and waste materials. The Laboratory has operated pilot plants for quite a number of chemical separation processes, the first being the bismuth phosphate process that was used in the 1943-1945 period to produce quantities of plutonium for evaluation and use in weapons programs. The objectives of a pilot plant program at ORNL were to:

1. Confirm the feasibility of the proposed process,
2. Obtain data necessary for the design and operation of a production plant,
3. Provide quantities of the product for large scale evaluation at other sites, and
4. Bring out chemical and engineering problems which were not recognized in smaller scale development work.

(Sadowski, 1956)

A number of the processes developed at the Laboratory went on to be widely used throughout the world for processing of nuclear materials. A summary of chemical processing experience at Oak Ridge is shown in Table 2-3, and key processes are discussed below.

#### The Bismuth Phosphate Process

The bismuth phosphate process was the process used in the wartime pilot plant to recover plutonium from Clinton Pile fuel slugs. The process was conducted in six "hot cells" with five-foot-thick concrete walls in Building 205, which is now called Building 3019. The bismuth phosphate operation processed the aluminum-canned uranium slugs that had come from the Clinton Pile in an underground, water-filled canal. The first cell housed a tank for dissolving the uranium slugs in acid, and four other cells had equipment for plutonium precipitation with the bismuth phosphate "carrier" and oxidation and reduction of the uranium left behind. The bismuth phosphate process recovered only plutonium; it did not separate uranium from the process liquid.

#### Radioactive Lanthanum Separation (RaLa)

Large scale production of radioactive barium-lanthanum was requested by Los Alamos on April 8, 1944, reportedly in an urgent request from J. Robert Oppenheimer, the civilian head of the Los Alamos Laboratory weapon development program. In early weapon design efforts at Los Alamos, the G (for "Gadget") Division was directed to perfect the implosion process for creating a critical mass in the center of a weapon and triggering an atomic blast. To study the behavior of test devices as they were caused to implode (burst inward) by use of conventional high explosives, the group incorporated radioactive lanthanum within the test device cores. The very intense gamma radiations from the lanthanum-140 (40.22 hours) were recorded by radiation detectors in order to characterize the movements of the parts of the test device that were completed in fractions of a second. (Thompson, 1949; Rhodes, 1986; Johnson and Schaffer, 1992). As of 1954, the ORNL RaLa product continued to be used for Los Alamos weapon development work (Dorsey, 1954).

RaLa processing involved large quantities of irradiated reactor fuel in which radioactive fission products had been allowed to decay ("cool") for only a short period of time. This is often called "short-decay" fuel. The processing also initially took place at a time when radioactive gases that were released were not efficiently trapped. Because of these factors, RaLa processing at Oak Ridge is described in some detail here.



TABLE 2-3

## ORNL EXPERIENCE WITH CHEMICAL PROCESSING

Period	Process	Building Number
1943-1945	Bismuth Phosphate	3019 (originally called 205)
1945-1951	Redox	3019
1944-1956	RaLa (Radioactive Lanthanum)	3026 (originally called 706-D)
1946-1948	Hexone-25	706-A
1946-1948	Hexone-23	706-A
1948-1949	Uranyl Ammonium Phosphate	706-A
1948-1958	Metal Recovery	3505
1948-1953	TBP-25	3505
1949-1960	Purex	3019, 3505
1949-1968	Fluoride Volatility	3019
1949-1976	Fuel Preparation	3019, 4505, 7930
1951-1976	Raw Materials	4500
1952	TBP-Interim-23	3503
1952-present	Thorex	3019
1953-1959	Feed Materials	4500
1955-1976	Head-End	4500N, 4505, 4507, 7601
1961-1976	TRU	3508, 4507, 7920

Due to the 12.8-day half-life of the parent fission product barium-140, the reactor fuel used for RaLa processing had to be fed to the process shortly after removal from the Oak Ridge or Hanford reactors. While one reference states that the slugs were allowed to cool for about five days after extraction from the pile (Davis et al., 1949) to allow short-lived radioactivities to decay, decay periods as short as three days between discharge and dissolution are documented (Thompson, 1949). One 1946 report indicates that "Slugs for Ba<sup>140</sup> processing are irradiated at least 40 days in the center of the pile and are charged to the 706-D dissolver with less than one day's cooling." (Reynolds et al., 1946). Eister (1949) referred to 40-day irradiation and one-day cooling for X-10 slugs.

Dissolution of short-decay slugs (i.e., 1-5 days cooling) also released four to three times more short-lived (half-lives around 1-5 days) fission product activity than did processing of conventionally (30 days or greater) cooled feed material. These facts are demonstrated by the values of gamma radioactivity content of ORNL graphite reactor "X" slugs as a function of decay time presented in Table 2-4 (Coryell and Levy, 1943). Note, however, that there is only a 33% decrease in the percent of original activity present with 5 days cooling compared to that present after 1 day of decay.

Control devices to limit releases of fission products were minimal in the early days of RaLa processing, but increased after it was recognized that the RaLa operation was a significant contributor to airborne radioactivity in the area of the Laboratory. Activity releases reach a peak about one hour after dissolvings begin, decrease to one-third during the second hour, and diminishes to zero at the end of approximately eight hours (Davis et al., 1949). Based on 150 pounds of slugs in a dissolving batch, one reference indicates that 2500 curies of xenon-133 (5.245 days), 1300 curies of iodine-131 (8.05 days), and under 1 curie of krypton-85 (10.72 years) were released within the dissolver (Davis et al., 1949). RaLa runs in Building 706-D typically saw slugs being dissolved in batches of approximately 50 at a time, with production "runs" involving up to 1,728 slugs (Thompson, 1949). A run could therefore involve up to 35 dissolvings events of approximately 50 of the 2½ pound "X" slugs.

A production process was developed and put into operation in building 706-C, where existing laboratory facilities were converted to barium-lanthanum production in five months. The first RaLa run was completed in September 1944 (Thompson, 1949). Certain steps of the process, such as precipitations, were carried out in glassware in lead-shielded cubicles. The operations were followed visually with periscopes until successive runs (three maximum) had so darkened the glassware by exposure to the intense radiation that observation was impossible and the glass equipment would have to be decontaminated and replaced (Unger, 1951). In all, nine shipments to Los Alamos were made, totalling 3,852 curies produced with the 706-C equipment, which was designed for only small-scale (1-10 curies) fission product separation (Thompson, 1949).

TABLE 2-4

## TIME-DEPENDENCY OF X SLUG GAMMA ACTIVITY

Cooling Time	Curies of Gamma Emitters per Slug	Percent of Original Activity
0	160	100
15 minutes	68	43
1 hour	43	27
6 hours	24	15
1 day	15	9
5 days	10	6
30 days	3.7	2

When it was desired to produce 300 curie batches for shipment on a routine basis, the available facilities were not adequate. Development of equipment more applicable to remote operation was started. During the course of the development work, the production requirement was increased to 2,000 curies per batch for shipment (Eister, 1949). When it became clear that RaLa production was to continue on a long-term basis, Building 706-D, now called Building 3026, was constructed specifically for RaLa production. Operations in 706-D began in May 1945. By March of 1949, shipments to Los Alamos from Building 706-D totalled 31 in number and included 62,412 curies of barium-140 based on measurements at Oak Ridge (Thompson, 1949).

Around 1949 it was decided that Hanford slugs, shipped to Oak Ridge in specially designed lead carriers, would be used permanently as the starting material for RaLa separation in 706-D, as their use had been demonstrated to be feasible and much more economical than X-10 slugs (Thompson, 1949; Unger, 1951). Around 1949 Los Alamos made a request for eight 10,000-curie runs a year to be made starting July 1950. They also indicated interest in receiving 20,000-curie shipments at some later date (Thompson, 1949). The equipment used for RaLa production at ORNL was always operated far above its design capacity; changes to allow for higher levels of production were made only in the final purification stages (Rupp and Witkowski, 1955). In 1950, several ion exchange processes were tested in purification of kilocurie quantities of radiobarium (Higgins et al., 1950). Before the last alterations to the final purification equipment were completed to allow processing of 10,000-curie batches, a new goal of 30,000 curies was set, and 50,000-curie batches were actually produced. After incorporation of the ion exchange process of final separation and purification, batches processed at ORNL grew to as large as 64,805 curies shipped (45,091 curies measured at Los Alamos) in January 1954 (ORNL, 1954; Rupp and Witkowski, 1955).

As stated by Thompson (1949):

"The most outstanding characteristic of the Ba<sup>140</sup> production operations is the unpredictable nature of the process. Because of inherent uncertainties in the operating procedure, such as the decantation of wastes, it has been impossible to maintain a fixed production schedule or even to determine in advance exactly what materials will be required in the processing of a run. Erratic operation of the equipment has made it impossible to adhere strictly to a standard operating procedure; special waste recovery steps or other emergency measures are needed so frequently that it has been impossible to leave the operation of equipment in the hands of Chemical Operators."

It was decided around 1950 that it would not be practical to further alter the Building 706-D equipment to increase production capacity. A proposal to build new facilities at ORNL was submitted to the AEC. An "Overall Economic Survey" of options for continued production of

RaLa was prepared in 1952 (Vaughan, 1951). It presented cost estimates for options that would have RaLa production at ORNL, Hanford, Idaho, or Los Alamos, using Hanford or Savannah River slugs or fuel assemblies from the Idaho Materials Testing Reactor (MTR) or the RIR. The identity of the "RIR" is not evident from records reviewed to date.

When Los Alamos increased requested batch sizes to a minimum of 30,000 curies, MTR fuel assemblies were proposed as a feed source because of their high specific activity. A MTR-RaLa process was developed at ORNL for supporting MTR fuel from Idaho as a feed to RaLa production. The method was developed based on an assumption that MTR assemblies would be processed in Idaho about four hours after discharge from the Idaho reactor (Blanco, 1951; Blanco, 1952a; Blanco, 1952b).

In 1954, after an incident in 706-D resulted in an airborne release, an official of the Research and Development Branch of the Division of Military Application wrote a letter to the Director of Los Alamos Scientific Laboratory asking that Los Alamos "give due consideration to the situation existing with respect to RaLa production and limit requirements on ORNL RaLa sources to those needed for crucial tests in weapon development work." (Dorsey, 1954; Stanley, 1954).

In 1954 the AEC decided to build the new RaLa production facility at Arco, Idaho, with production expected to begin by July 1, 1956 (Rupp and Witkowski, 1955). The final RaLa production run at ORNL was called Run 68, and occurred in 1956. The Laboratory was officially relieved of its RaLa production responsibilities at the end of calendar year 1956, although the AEC asked that facilities remain on standby until March 31, 1957, during initial runs of the new facilities in Idaho (Seagren and Witkowski, 1957).

Over the approximate 12-year period of RaLa production operations at ORNL, there were approximately 87 runs made, including two special runs to provide lanthanum-140 to the X-10 Health Physics group and one special run to provide three containers of lanthanum-140 to the AEC for the ARUU program. The ARUU program (also called the AHRUU program, with the derivation of both acronyms not evident from reviewed records) involved placement of a sample of lanthanum on the ground and measurement of radiation intensities at various distances; the apparent purpose was to determine attenuation factors for lanthanum's gamma rays in air (Morgan, 1948a). A later test used 156 lanthanum sources enclosed in iron pipes and arranged in a 275-by-300-yard plot near the K-25 Site; radiation measurements were made with an electroscope along various lines and compared with predicted exposure rates (Morgan, 1948b). In total, ORNL workers dissolved at least 30,000 slugs in the process of separating over 500,000 curies of radioactive barium-lanthanum for Los Alamos weapons development and several research projects.

### The Redox Process

The Redox process was a solvent extraction process for separation and purification of plutonium and uranium that was developed at Argonne National Laboratory and tested and improved at ORNL after World War II. Solvent extraction methods take advantage of the fact that nitrates of plutonium and uranium are readily soluble in certain organic liquids, while the nitrates of fission products are generally insoluble in these liquids. The Redox process, which used methyl isobutyl ketone (hexone) as the organic solvent, showed great promise to be simpler and more economical than the bismuth phosphate process precipitation process, but it at first yielded uranium of insufficient purity. ORNL workers modified the process and demonstrated on a pilot-plant scale how sufficiently pure uranium could be produced (Thompson, 1963; Johnson and Schaffer, 1992; Brooksbank, 1991).

### The "25" Process

The "25" Process was designed by ORNL chemists and chemical engineers to recover the highly-enriched uranium from used uranium-aluminum alloy fuel elements from the Materials Testing Reactor in Arco, Idaho. The designator "25" originated from uranium-235 (703,800,000 years), the radionuclide of interest. While some of the fissionable uranium-235 contained in the MTR fuel was "burned up" (underwent fission) during operation of the MTR, each MTR fuel assembly contained between 123 and 92 grams of uranium-235 after irradiation times of 12 to 37 days (Blanco, 1952b). The Laboratory designed the Idaho chemical plant for processing of MTR fuel and supervised its construction and its startup in 1952.

### The Tributyl Phosphate (TBP) Process

The tributyl phosphate (TBP) process was developed in 1949 for the recovery and purification of uranium from the metal-bearing wastes that had accumulated in the Laboratory's tank farm from wartime processing efforts. The process used tributyl phosphate as a solvent for extracting uranium, and was later modified to allow recovery of plutonium as well as uranium from ORNL wastes. A Metal Recovery Facility was constructed at ORNL to use the TBP process to recover the estimated 150 tons of uranium and 1 kilogram of plutonium contained in fission product contaminated waste solutions that had accumulated over a period of 10 years in the ORNL tank farm. In operation since January 2, 1953, Metal Recovery also recovered uranium and plutonium from fuel from the Chalk River reactor, Hanford metallurgical waste, the Brookhaven National Laboratory reactor, Argonne National Lab's CP-2 and CP-3 reactors, and 7.8 tons of sand contaminated by weapons tests in Nevada (Thompson, 1963; Brooksbank, 1958; Brooksbank et al., 1953; Brooksbank, 1991).

Metal Recovery processing of Hanford metallurgical wastes (MgO crucible fragments, casting fragments, packing sand, and metallurgical slag) for recovery of plutonium and americium began on March 10, 1954. From March through May, 94 dissolvings yielded 18,558 grams of plutonium (Brooksbank, 1954).

Metal Recovery also recovered 127 grams of neptunium-237 and 6.7 tons of uranium from 11.2 tons of the nonvolatile fluoride "ash" resulting from fluorination of  $UO_3$  to  $UF_6$  at Paducah gaseous diffusion plant (Brooksbank and Hylton, 1958).

In 1958 the Metal Recovery Plant (Building 3505) was linked by underground pipeline to the Building 3019 Thorex Pilot Plant to form the Power Reactor Fuel Processing (PRFP) complex. For some programs where sufficient decontamination could not be achieved in Building 3019's solvent extraction cycle, an additional three cycles were performed in Building 3505 (Brooksbank et al., 1960). The process used in PRFP-3505 was a modified Purex solvent extraction process (Brooksbank et al., 1960), as discussed in the following section.

On October 28, 1959, unusually high levels of gross-beta radioactivity were noted in ORNL process wastewater. Levels increased over the following days, and by November 1st levels were high enough that the gate of White Oak Dam was closed to curtail releases to the Clinch River. The elevated release was eventually attributed to an evaporator leak in Building 3019 that had resulted in release of approximately 2,000 curies (primarily ruthenium-106 (368.2 days) and cerium-144 (284.3 days)) to the waste system. Approximately 45 curies of ruthenium-106, 9 curies of cerium-144, and 0.05 curies of unidentified alpha activity were released to the Clinch River. Portions of the remaining radioactivity that was not released likely decayed away or settled in waste tanks, settling basins, or White Oak Lake. After measures were taken to prevent further leakage, the dam gate was reopened on November 3rd (Morgan, 1961).

On November 20, 1959, a non-nuclear explosion occurred in an evaporator contained in a shielded cell in Building 3019, one of the buildings making up the PRFP. The evaporator was being decontaminated with Turco Decon 4501, a proprietary decontaminating reagent containing phenol, when nitric acid was inadvertently added to the solution and allowed to boil for several hours. The formation of nitrophenol or picric acid caused an explosion, which caused plutonium to be released from the processing cell, probably as an aerosol of fine particles of plutonium dioxide. It was estimated that 1.5 grams of plutonium (about 0.04 curie) entered the cell ventilation system and was removed by roughing and absolute filters. A door was blown open to the outside, resulting in the estimated distribution of about 600 milligrams of plutonium over an area of about four acres south and east of building 3019. The graphite reactor building was most contaminated. An estimated 70 milligrams of plutonium were released to pipe passages and service openings to the interior of Building 3019. The immediate area was evacuated, and steps were taken to remove or "fix" contamination to minimize exposure to persons entering the area. Fixation was accomplished by resurfacing roads and by painting roofs, walls, and

equipment. Paint was even sprayed on grass lawns and sidewalks that were found to be contaminated. Decontamination efforts lasted several months (King and McCarley, 1961; Eisenbud, 1987; Boyle et al., 1982; Parrott, 1979; Morgan, 1961; McDuffee, 1961a).

Around 1961, seven plutonium-aluminum alloy MTR fuel assemblies and one shim rod were processed in the PRFP with the TBP process to recover "high burnup" plutonium rich in the plutonium-240 (6537 years) and plutonium-241 (14.4 years) isotopes. About 500 grams of plutonium were separated. (McDuffee, 1961b; Brooksbank, 1991).

### The Purex Process

Based on the work with the TBP process, a decision was made to investigate use of a similar process for separately recovering plutonium and uranium from Hanford plutonium production metal. The Purex process (from Plutonium Uranium Reduction EXtraction) was shown in 1949 to be feasible and probably more economical than other processes being developed at the time (Lanham and Runion, 1949). Laboratory development of the Purex process at ORNL took place in 1949 and continued through the period of Purex pilot plant operation (Lanham and Runion, 1949; Lanham and Gresky, 1950; Runion and Ellison, 1950; Flanary et al., 1952). The Purex process used solvent extraction with tributyl phosphate in "Varsol" organic diluent and nitric acid as a "salting agent." In other runs, TBP was used in Amsco 123-15 hydrocarbon diluent (Flanary et al., 1952; ChemRisk, 1993). In addition to isolating plutonium and uranium, the Purex process also isolated fission product isotopes of zirconium, niobium, and ruthenium. This feature simplified radioactive waste management. By distilling off the nitric acid for re-use, fission products were made readily available "in a packaged form." In addition to development work on the main Purex solvent extraction process, ORNL developed an ion-exchange plutonium isolation method, a process for recovering plutonium from metallurgical wastes, a method for recovering nitric acid from solvent-extraction wastes, and a fume-less dissolving process for removing radioactive fission gases from dissolver fumes that permitted recovery of the oxides of nitrogen as nitric acid that could be reused (Flanary et al., 1952; Thompson, 1963).

Over a two-year period (approximately May 1951 to May 1953), pilot-plant development of the Purex process was conducted at ORNL to demonstrate that uranium and plutonium could be economically recovered from irradiated natural uranium slugs with continuous operation (as opposed to batch operation). Use of a pulse column contactor for solvent recovery was demonstrated in a total of 34 runs on radioactive material, of which 24 were of four-inch, aluminum-clad Hanford production-level fuel slugs ("W/2 slugs") and the remainder ORNL graphite reactor aluminum-clad slugs. Most runs involved 140 slugs (249 kilograms), except for the first run, which involved 188 slugs. The final 20 runs used continuous processing, involving solvent extraction, solvent recovery, and acid recovery. Irradiation levels of the slugs



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significant forms of radioactive material encountered in the Thorex processing were protactinium-233 and ruthenium. Ruthenium was observed to be a limiting fission product contaminant in the Thorex process (Gresky, et al., 1953). Reduced efficiency of ruthenium decontamination was observed in the short-decay runs (McDuffee, 1959a). The separation of protactinium-233 as a specific product from Thorex process material was an early objective of the program, but was never achieved (Johnson and Schaffer, 1992). With its intense radioactivity during the short-decay runs, protactinium-233 was a significant contributor to radiation levels in the pilot plant building that increased by a factor of five, to particulate releases from the pilot plant stack, and to heat evolution from the feed material that caused operational problems. Because of concerns with personnel exposures in the facility, the short-decay runs were terminated (Winsbro, 1957; Yarbrow, 1958; Albrecht, 1957; McDuffee and Shank, 1960).

#### The Interim-23 Process (INT-23)

The Interim-23 Process, named from uranium-233, was developed to isolate uranium-233 alone from thorium and associated fission products. INT-23 was used to produce kilogram quantities of U-233 for weapons applications from Hanford-irradiated slugs processed at a rate of 70 kilograms of slug material per day. A total of 2.5 kilograms of uranium-233 were isolated in 1952 in Building 3503, the High Radiation Level Chemical Laboratory (Thompson, 1963).

#### The Fluoride Volatility Process

The fluoride volatility process was developed to take advantage of the high volatility of uranium hexafluoride to separate uranium from less volatile fluoride salts. The process was developed at ORNL to recover uranium-235 from molten salt reactor fuels and from other fuels soluble in molten salt. In initial pilot plant operations, 56.6 kilograms of uranium-235 were recovered from aircraft reactor experiment (ARE) molten salt fuel processed in 1958 (Thompson, 1963; Winsbro et al., 1962). The process was used from 1949 to 1968 in Building 3019, including the reprocessing of molten salt reactor experiment (MSRE) fuel and uranium-zirconium alloy fuels containing highly enriched uranium (Brooksbank, 1991; Carr et al., 1971). Runs with aluminum-clad fuel elements from the Oak Ridge Research Reactor (ORRR) and the Low Intensity Testing Reactor (LITR) were conducted, with cooling periods ranging from 25 days to 18 months. During the shortest-decayed run, 24 curies of 10.72-year krypton-85 and 2260 curies of 5.245-day xenon-133 were calculated to be released from the dissolver to air exhausted from the dissolver. Also estimated to be present in releases from the fluorinator were 389 curies of tellurium-127/129 (9.35 hour/69.6 minutes), 2293 curies of iodine-131 (8.05 days), and 4228 curies of ruthenium-103/106 (39.35 days/368.2 days). However, analyses of charcoal-trap samples taken from side-streams of stack gases indicated releases of no noble gases, 20 curies of tellurium, <0.01 Ci of iodine-131, and <0.001 Ci of ruthenium-106 (Carr et al., 1971).

### The Hermex Process

The Hermex process was a method proposed for recycling and purifying scrap uranium, thorium, and other high-melting-point metals or alloys and for decontaminating irradiated uranium or thorium based on the much greater solubility of such metals in hot mercury than in cold. In tests of the Hermex process conducted at ORNL, metallic uranium was dissolved rapidly in boiling mercury and was recovered from the cooled amalgam as uranium mercuride by filtering and pressing. Fission product decontamination factors were 100 to 1,000 for three-year-decayed uranium and 15-28 for 45-day-decayed uranium. Decontamination factors indicate the factor by which concentrations of an undesirable material are reduced in a product stream by a chemical separation process. As of May 1956, the Hermex process had only been tested in laboratory scoping experiments (Blanco et al., 1956). No documentation of larger-scale use of the process has been found.

### Raw and Feed Material Processes

Processes used at ORNL for preparation of raw materials included the Slurex process for recovery of uranium from ore concentrates. It used TBP instead of the more volatile and hazardous ethyl ether that had been previously used (Thompson, 1963; Brooksbank, 1991). This process came into use at Fernald. In addition, the Excer process was developed to convert, by ion exchange, uranyl nitrate from Purex and Redox operations to  $UO_2F_2$ , which was electrolytically reduced to  $UF_4$  in preparation for recycle through the K-25 Plant. The process had promise to be more economical than reduction with hydrogen and hydrogen fluoride (Blanco and Eister, 1956; Thompson, 1963).

The Metallex process was being evaluated around 1956 as a way to reduce thorium chloride to metal using sodium amalgam. During the same time period, the Fluorox process was being evaluated as a method of converting uranium tetrafluoride to uranium hexafluoride. (Blanco and Eister, 1956).

### "Head End" Processes

Various processes have been used and developed at ORNL for dejacketing or segmenting of power reactor fuels in preparation for dissolution and solvent-extraction chemical processing. They have included the DAREX and SULFEX processes for stainless-steel clad fuels and the ZIRFLEX and ZIRCEX processes for zirconium-jacketed fuels. Various mechanical methods, such as shearing, chopping, underwater sawing, or pushing through a die; have also been used at ORNL to expose the fuel cores for dissolution. (Brooksbank, 1991; Goeller et al., 1960; Thompson, 1963)

#### 2.1.4 Radionuclide Production

The Oak Ridge National Laboratory has long been a world leader in supplying radionuclides to the fields of research, medicine, and industry. The radionuclides that have been produced at ORNL can be grouped by their method of production into the following categories:

- Neutron Activation Products — produced by placing materials in a neutron flux,
- Fission Products — separated from nuclear fuel or waste streams from a reactor,
- Transuranic Nuclides — a special class of neutron (reactor) products, produced by neutron bombardment of heavy elements such as curium,
- Cyclotron Products — produced by bombarding materials with energetic charged particles, and
- Calutron Products — produced by electromagnetic enrichment of materials. After World War II, calutrons were used primarily for stable isotope separation.

The Laboratory for many years was involved in development and refinement of methods used for production of radionuclides in each of these categories. In addition, ORNL has been a leader in supporting uses of radionuclides and stable isotopes in many diverse areas of research, medical diagnosis and therapy, and industrial and consumer applications. This section provides a summary of radionuclide development and production work at ORNL. Radionuclide work at the Laboratory began at the Clinton Pile, and in subsequent years also involved the ORRR, the LITR, cyclotrons, calutron facilities for stable isotope separation located at the Y-12 Plant, a fission product development laboratory, the HFIR, the Transuranium Processing Plant, and a group of buildings located in what is known as the isotopes area of the central ORNL grounds. Other facilities that were involved in radionuclide work were the Oak Ridge isochronous cyclotron (ORIC), which began operating in 1963 and later became part of the Holifield heavy-ion research facility, which was completed in 1980 (Johnson and Schaffer, 1992).

On August 2, 1946, the first radionuclide shipment was made under the radioisotope distribution program. It was one millicurie of carbon-14 (5730 years), which was sent to the Bernard Free Skin and Cancer Hospital in St. Louis, Missouri (Thompson, 1963). By 1957, radionuclide shipments grew to over 14,000 per year, and totalled over 166,000 curies (Seagren, 1958).

The first buildings in the Radioisotope Processing Area were occupied in June 1949 (Ramsey et al., 1949). In January of 1962, the AEC announced the establishment of the Isotopes Development Center (IDC) at ORNL. The IDC was created to coordinate the various

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TABLE 2-5

## ORNL RADIONUCLIDE PRODUCTION BY NEUTRON BOMBARDMENT, 1947/1948

Target Material	Radionuclides Produced
Sulfur, U.S.P., Merck	S-35, P-32, Si-31
Monobasic Potassium Phosphate, Reagent, Merck	K-42, P-32
Calcium Carbonate, Powder, C.P., Baker's	Ca-41-45, P-32
Scandium Oxide, from Adam Hilger, Ltd., London	Sc-46
Titanium Oxide, Pure, Amend Drug & Chemical Corp	Ti-51, P-32
Carbonyl Iron Powder, A.D. Mackay	Fe-59, Fe-55, P-32
Cobalt (ic) Oxide, C.P., Baker's	Co-60
Nickel Powder, Belmont Smelting & Refining Works	Ni-65, Co-60, Fe-59, Cu-64
Selenium Powder, General Chemical Company	Se-75
Silver Nitrate, Crystal, Reagent, General Chemical Co.	Ag-110
Indium Nitrate, C.P., A.D. Mackay	In-114
Mossy Cadmium Metal, General Chemical Company	Cd-115, Cu-64
Antimony Metal, Lump, C.P. Baker's	Sb-124
Europium Oxalate	Eu-154 (also Eu-152, Eu-155)
Tantalum Tetraoxide, C.P. City Chemical Corp.	Ta-182
Tungsten Trioxide, C.P., Fisher Scientific Company	W-185
Osmium Powder, Americium Platinum Works	Os-185, Os-191, Os-193
Thallium (ic) Nitrate, C.P., Fordomes Trading Company	Tl-204, 206
99.8% Bismuth Metal, C.P., Baker's	Bi-210
Sodium Carbonate	Na-24
Copper	Cu-64
As <sub>2</sub> O <sub>3</sub>	As-76
Potassium Bromide	Br-82
Rubidium Carbonate	Rb-86
Mo <sub>2</sub> O <sub>3</sub>	Mo-99
Gold	Au-198
Tellurium	I-131
CeO <sub>2</sub>	Ce-141-143
Calcium Nitrate	C-14
IrO <sub>2</sub>	Ir-192

one week. The reactor was also equipped with three pneumatic tubes that allowed samples to be inserted and withdrawn from the reactor, by CO<sub>2</sub> pressure, while the reactor was operating. With the "fast" pneumatic tube, samples could be exposed for less than one second, with removal time also less than one second. From the "large" and "small" tubes, samples were ordinarily discharged to a storage shield to await pickup, while samples from the large tube could also be routed through a shielded tube to a laboratory room for quick analysis.

Devices called "factories" were at times inserted into reactor holes. A status report for December 1944 describes a new carbon-14 factory that was inserted into hole 16 of the pile (Snell et al., 1945). This factory was evidently in the form of tubes that contained a circulating fluid in which carbon-14 (5730 years) was formed by neutron activation. The same report mentions unsuccessful attempts to recover tritium from irradiated lithium-lead alloy slugs irradiated in the pile. An "isotope distribution program" started in August 1946 for commercial supply of radionuclides from the Laboratory. In the first six months of the program, 482 shipments were made. In early 1947, approximately 100 millicuries of sulfur-35 (87.44 days) were separated from bombarded potassium chloride for distribution to radionuclide customers. This was the first sulfur-35 separated at Clinton Laboratories for distribution. (Stuckey et al., 1947a).

Slugs containing calcium nitrate were irradiated in the pile for production of carbon-14. In February, 1947, 130 of these slugs were discharged. One run of 20 slugs in February 1947 yielded 10 microcuries of carbon-14. As of February 1947, there were 106 exposure "cans" in the pile, of which about half were for radionuclide production. Three of the samples removed were CeO<sub>2</sub> for the production of praseodymium-143 (13.8 days). As an indication of development work in progress at the same time, records state that the Technical Division was working on a procedure for separation of calcium-45 (162.7 days) from scandium. (Stuckey et al., 1947a).

The breakdown of the most popular neutron product shipments during the first 7 months of the distribution program was as follows:

<u>Radionuclide</u>	<u>Portion of Shipments</u>
phosphorus-32 (14.29 days)	37%
carbon-14 (5730 years)	19%
gold-198 (2.696 days)	12%
sulfur-35 (87.44 days)	5%
iron-55/59 (2.7 years/44.63 days)	4%
potassium-42 (12.36 hours)	4%
calcium-41/45 (103,000 years/162.7 days)	3%
cobalt-60 (5.271 years)	2%

A process for production of carbon-14 from bombarded  $\text{Be}_3\text{NO}_2$  was developed, using a hydrogen peroxide separation procedure. Plans in 1949 were to use this new procedure in a ten-slug plant being built in the new isotope area (Ramsey et al., 1949).

In the early 1960s, a Curium Recovery Facility (CRF) was established in High Level Chemical Development Facility (Building 4507) hot cells 3 & 4. The original goal of the CRF was to recover and purify americium-243 (7380 years), curium-242 (163.2 days), and curium-244 (18.11 years) from these three feed sources:

- Concentrate received from Savannah River Plant for processing of 10 kilograms of plutonium-239 that had been irradiated in a Savannah River Reactor,
- Eleven Martin  $^{241}\text{AmO}_2$  targets that had been irradiated in the MTR, and
- Thirty-six  $^{241}\text{AmO}_2$  targets that had been irradiated in the ORRR.

Before the CRF got into operation, however, its mission was changed to backing up a private operation (the Martin Co.) to recover curium-242 for the space power program. Curium is useful in space power applications due to its high radiation power (heat), which can be converted to electrical power for space craft systems. In December 1963 the CRF "went hot", and over the next two years several batches of americium-243 and curium-244 were isolated and purified and distributed to various users. A series of irradiated americium-241 (432.2 years) slugs were then processed to recover curium-242 for materials studies and an environmental test of a prototype space power supply was conducted. The final curium-242 campaign was completed in May of 1966. (McDuffee et al., 1970; Genung et al., 1993).

Beginning around 1968, the HFIR was used to produce large quantities of transuranic radionuclides by neutron bombardment, initially as part of the USAEC Heavy Element Production Program. Targets, typically containing curium-244, plutonium-242 (375,800 years), or other transuranic nuclides, were placed in the flux trap region of the HFIR to form transuranic nuclides such as berkelium-249 (320 days), californium-252 (2.639 years), einsteinium-253 (20.467 days), and fermium-257 (80 days). Targets were removed from the HFIR and chemically processed in the Transuranium Processing Plant (TRU; Building 7920) to yield pure transuranic nuclides in the desired chemical and physical forms. Commonly produced sources were often encapsulated in stainless steel or zircalloy. Processes used for separation and purification on transuranic radionuclides have included TRAMEX, CLANEX, BERKEX, and PLURIX. (King et al., 1984; McDuffee and King, date unknown; Brooksbank, 1991).

Building 7920 saw its first hot processing campaign in November 1966 on prototype HFIR targets that had been irradiated at Savannah River; about 180 micrograms of californium-252 were recovered. Subsequent fine-tuning of methods resulted in the output of the process



one week. The reactor was also equipped with three pneumatic tubes that allowed samples to be inserted and withdrawn from the reactor, by CO<sub>2</sub> pressure, while the reactor was operating. With the "fast" pneumatic tube, samples could be exposed for less than one second, with removal time also less than one second. From the "large" and "small" tubes, samples were ordinarily discharged to a storage shield to await pickup, while samples from the large tube could also be routed through a shielded tube to a laboratory room for quick analysis.

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bombarded in the pile (Ramsey et al., 1948). It was rapidly realized, however, that this production method would not be able to keep up with the rapidly growing demand for the nuclide; distribution had increased from 300 millicuries per month in 1946 to 30,000 millicuries per month in 1950 (Rupp et al., 1951).

Because it involved processing of short-decayed reactor slugs in a process that spanned a considerable length of time, fission product iodine production at ORNL is described in some detail here. A sampling of iodine-131 production data is presented in Table 2-6. A method for separation of large quantities of pure, carrier-free fission product iodine from graphite reactor slugs was developed in the late 1940s. An iodine production "plant" went into operation in Building 706-C in September 1948, and in its first three years of operation produced about 1,000,000 millicuries (1,000 curies) of iodine-131 for commercial distribution. Following is a description of the early fission product iodine preparation method, based on a report by Rupp (1951):

The early iodine plant used slugs that had been in the reactor 30 to 60 days, transported as soon as possible after discharge (three at a time) in a 2,500-pound lead and steel slug carrier from the discharge canal in the pile building to the top of the shielded dissolver cell. The slugs were lowered into the dissolver in an aluminum basket, and the aluminum slug jackets and the basket were dissolved in a mixture of sodium hydroxide and sodium nitrate. The slugs were then washed and dissolved in nitric acid. In the dissolver, the isotopic mixture of iodine is approximately as follows:

8-day iodine-131	43%
22-hour iodine-133	13%
long lived/stable iodine-129/127	43%
2.4-hour iodine-132	trace

Some iodine is released from the metal during dissolution, but well over 50% must be removed as elemental iodine by steam-air distillation. Iodine not caught in the distillate (which contained about 0.2 millicuries of iodine-131 plus iodine-133) passed to a NaOH scrubber. Reports state that "practically no iodine gets past the alkaline scrubber", but xenon passes through to the off-gas system. Dissolving took about 20 hours. The distillate was treated with hydrogen peroxide, and the iodine was distilled out to yield a small volume of distillate containing little nitric acid and most of the iodine. This crude iodine distillate was trapped in a NaOH solution and then evaporated down to a small volume suited for final purification in glass equipment. Final purification started out with oxidizing the iodine to iodate with sulfuric acid-potassium permanganate solution, then distillation of volatile substances away from the involatile iodate. Phosphoric acid and hydrogen peroxide are then added to reduce the iodate and

TABLE 2-6

## A SAMPLING OF RADIOIODINE PRODUCTION DATA

Year	Slugs Processed	Millicuries iodine-131 Shipped	Comments
1948			Iodine-131 "Plant" went into service in 706-C.
1949	June: 23 December: 10	June: 10,400 December: 18,428	Reference: Rupp, 1954.
1950	396 from ORNL 12 from Hanford	310,698	Reference: Rupp, 1954.
1951	651 from ORNL 5 from Hanford	467,644	Reference: Rupp, 1954.
1952	644 from ORNL 12 from Hanford	488,231	Reference: Rupp, 1954.
1953	828 from ORNL 4 from Hanford	504,455	New process investigated.
1954		817,879 produced 557,482 sold*	Old processing plant in Building 3026 (706-D) was abandoned. Operations started in new plant in August.
1955		668,578 produced 557,482 sold*	
1956		735,467 sold	
1957		891,029 sold	
1958		4,040,324 made 1,074,128 sold*	
Oct. - Nov. 1969		200,000 (2 batches) Elemental iodine-131: 400	
Feb. 1985			No mention of Iodine-131 production.

Note: Only a portion of available iodine-131 production data have been reviewed to date; where data are not listed, it does not indicate that data are not available.

\* The majority of the difference between quantity produced and quantity sold is attributable to radioactive decay.

permanganate, then the iodine is distilled off and trapped in dilute NaOH. Sodium sulfite is added as a reducing agent, and the solution is filtered and transferred to a shielded container for storage.

The dissolving of slugs and preparation of iodine was done in a heavily-shielded concrete cell. The product was transferred in a lead-covered line to a lead-shielded hood for final purification in glassware. A stainless-steel off-gas line, operated at 40 inches of water negative pressure, removed process gases from the equipment for passage through a wetted-wall precipitron and glass-fiber filter before entering a high-volume stream passing out the 250-foot ORNL central stack.

In February 1947, 21 iodine-131 runs were made in Building 706-C, yielding a total of 1300 millicuries. A method was in preparation at that time for recovery of ruthenium-103/106 (39.35 days/368.2 days) in the RaLa building, 706-D (Stuckey et al., 1947a). The breakdown of the most popular fission product shipments during the first seven months of the distribution program was as follows:

<u>Radionuclide</u>	<u>Portion of Shipments</u>
iodine-131	85%
strontium-89/90 (50.55 days/28.6 years)	5%
yttrium-91 (58.51 days)	2%
barium-140 (12.789 days)	2%

In some cases, fission products were separated from RaLa process wastes; for example, the strontium extracted from alcohol-ether hydrochloric acid wastes from Run 16 (Stuckey et al., 1947b). Radionuclides such as ruthenium-106 were separated from tank farm waste (Ramsey et al., 1948). Much of the fission product separation work in the 1940s was done in Building 706-D "hot hoods."

As of June 1949, fission product iodine-131 production involved 23 slugs per month, yielding shipments of about 10,400 millicuries. In 1949, investigations revealed that the quantity of iodine-131 that was being shipped out as a millicurie was 1.75 times too great and adjustments were made. (Ramsey et al., 1949). Practically all iodine-131 made in 1949 came from the fission iodine process. Hanford-irradiated slugs were used in three runs. In each case, the iodine-131 from two slugs was sufficient to satisfy demand for three weeks. Slightly elevated air contamination sometimes resulted from small leaks during material transfers. At that time, the central exhaust treatment facility and stack had not yet been completed. (Ramsey et al, 1950).

TABLE 2-6

## A SAMPLING OF RADIOIODINE PRODUCTION DATA

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1956		735,467 sold	
1957		891,029 sold	
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Feb. 1985			No mention of Iodine-131 production.

Note: Only a portion of available iodine-131 production data have been reviewed to date; where data are not listed, it does not indicate that data are not available.

\* The majority of the difference between quantity produced and quantity sold is attributable to radioactive decay.

### Calutron Products

In 1950, ORNL acquired Y-12's three research divisions — Isotope Research and Production, Electromagnetic Research, and Chemical Research. The Y-12 stable isotopes program had emerged at the end of the war when Y-12 staff ceased using calutrons to separate uranium isotopes for atomic weapons. Researchers at first used four calutrons salvaged from electromagnetic equipment. Copper isotopes were the first to be collected, followed by isotopes of iron, platinum, lithium, and mercury that were separated and shipped to universities, governments, and national laboratories (Johnson and Schaffer, 1992). As of 1963, the ORNL electromagnetic isotope-separations program was providing more than 250 high-purity, highly enriched isotopes for use in a wide variety of research and technical applications. During 1963, more than 17,000 grams of separated material were collected from 28 of the calutrons allocated for the stable isotope program. In addition, electromagnetic processing of plutonium that was conducted for one year in a containment facility at ORNL provided more than 240 grams of enriched plutonium isotopes 239 through 242 for use in planned research projects (ORNL, 1964). As of late 1985, the stable isotope enrichment program of ORNL (at Y-12) made over 220 isotopes available by direct sale and/or on a loan basis (Bell and Tracy, 1985). Calutrons were last used for stable isotope production at Y-12 in 1990.

### **2.1.5 Support Functions**

One of the areas of potential off-site impacts from X-10 operations concerns disposal practices of the various wastes generated at X-10. In addition, the area of criticality safety is a support function that warrants discussion because it has been associated with several accidents. This section presents a discussion of historical X-10 waste management practices and an overview of the criticality safety program.

### Liquid Waste Management

When the X-10 site was planned, it was believed that the Laboratory would be a temporary facility, and volumes of waste were envisioned to be relatively small. A set of sprayed concrete (gunite) tanks was provided to contain the wastes from pilot operations at X-10. However, in 1943 the mission of ORNL was expanded into one of indefinite duration, and waste volumes soon exceeded the capacity of the gunite tanks. It was then decided to precipitate radioactive material within the tanks by addition of caustics, with the resulting sludges remaining in the tanks and the liquids released, with dilution by large volumes of uncontaminated or low-activity process waste water, into White Oak Creek. To further aid in retention of radioactive material, a dam was built across White Oak Creek about 1.7 miles up from where it meets the Clinch River to provide additional opportunity for any remaining solids to settle out in White Oak Creek. (Spalding and Boegly, 1985).

Apparently the levels of radioactivity entering White Oak Creek were not acceptable, so in June 1944 a 1.5 million gallon capacity settling basin (also called the 3513 pond) was completed to serve as a collection, sampling, and stilling pond for gunite tank liquids to permit radioactive solids to settle from the waste before discharge to White Oak Creek. High activity waste, which was being produced at a rate of about 7,000 gallons a day, were precipitated in the gunite tanks, held up for about one month for decay (of short-lived iodine-131 (8.05 days), cerium-141 (32.5 days), cerium-143 (33 hours), ruthenium-103 (39.4 days), barium-140 (12.8 days), and lanthanum-140 (40.2 hours)), with solids allowed to settle in the tanks, the settling basin, and White Oak Lake. It was calculated that five curies of mixed fission products could safely be discharged per day, and an average of 284 curies per year were released up to 1957. Radioactive waste volumes continued to increase, and in June 1949 an evaporator was put into service to reduce the volume of wastes stored in the gunite tanks and to reduce the volume of waste released to White Oak Creek. From 1949 until retired in 1954, the evaporator reduced 11,560,000 gallons of waste to 423,000 gallons of concentrate. Lack of storage space for this concentrate lead to the first use of a disposal pit in 1951 (Spalding and Boegly, 1985).

Waste pit No. 1 was built in July 1951. While discharges to the pit were terminated on October 5, 1951 when leakage from the pit was discovered, the pit continued to receive discharges from drains in decontamination building 7819 until 1964. While Pit No. 1 leaked, it appeared to mainly leak ruthenium-106 (368.2 days), a radionuclide of less concern than longer-lived strontium-90 (28.6 years) or cesium-137 (30.17 years). Pit 2 was constructed in 1952 and was used until November 1962. In 1954, a pipeline was constructed for transfer of wastes from the gunite tanks to Pit 2, and the waste evaporator was shut down. Prior to the pipeline, wastes were trucked to the pits in 500-gallon tanks outfitted on a Dempster Dumpster truck and after February 1953 in a converted 4,000-gallon tank trailer. Pit 3 was opened in January 1955, and was used until September 1961. Pit 4 was used from April 1956 until the new process waste treatment plant went into service in 1976. In 1959, when large quantities of ruthenium-106 were discharged to the pits, rather severe seepage was noted from Pit 4, and an interceptor trench was dug. For a while, approximately 50 curies per day of ruthenium-106 were pumped back to Pit 4. Waste "Trench" 5 was a new, earth-covered design to reduce radiation exposures to personnel, to avoid needing barriers to keep wild fowl and falling workers from the pits, and to keep rainwater from adding to the volume in the pits. Trench 5 was in service from May 1960 to about 1964. Trench 6 was in service from June 1961 until taken out of service on October 10, 1961 when significant seepage of cesium-137 and strontium-90 was discovered. The early failure of Trench 6 caused Trench 7 to be built in August 1962 and to be used until the hydrofracture facility became operational in 1966 (Spalding and Boegly, 1985).

In the hydrofracture process, hydraulic pressure was used to initiate a crack between layers of shale. Alkaline waste solutions were mixed with cement and other additives, and the mixture was injected under pressure into the cracks at depths of about 700-1000 feet. The "grout" filled the crack to form a thin, horizontal sheet which set within a few hours to fix the waste in the

shale formation. The original hydrofracture facility was used from 1964 to 1979; a total of 26 injections were made. The newer hydrofracture facility, which performed its first injection in June 1982, was designed to inject 140,000 gallons of grout per injection; enough to keep up with ORNL waste generation with one injection per year (Boyle et al., 1982). Injections at the new hydrofracture facility were terminated in 1984 when questions arose about possible leaching to deep groundwater (Ohnesorge, 1986). It was learned that the operators had failed to add cement to a number of injections, and that radioactive material was migrating in the plane of injection (Mobley, 1993).

### Airborne Waste Management

Prior to 1950, air discharges were from stacks serving individual facilities; for example the graphite reactor, the chemical separation pilot plant, and Building 706-D, the RaLa building. The graphite reactor and the pilot plant each had 200-foot-high stacks. The RaLa building exhausted through a local 30-foot-high fan house stack, until 1946 when off-gas was routed to the pilot plant stack. Stack releases at X-10 were initially untreated. Filters were added to RaLa off-gas lines and to the graphite reactor late in 1948. A central off-gas facility with a 250-foot brick stack (sometimes called the "900 area" stack) was built around 1950, and Building 706-D releases were routed there that same year. The new central off-gas system included particulate filters and an electrostatic precipitator at that time but no scrubber for radioiodine. Caustic scrubbers were first installed in the central off-gas facility in April 1961, with an upgraded charcoal filter system to follow around 1970 (Thompson, 1949; Ohnesorge, 1986; Mannesmidt, 1966).

On November 11 and 12, 1959, a particulate radioactivity fallout incident contaminated much of the Laboratory grounds. An investigation established that the fallout was in the form of particles, contaminated almost entirely with ruthenium-106 and its daughter rhodium-106, that had detached from the walls of the electric fan housing and ducts associated with the ORNL central brick stack. A fission product, ruthenium originated in various chemical processing operations associated with pilot plant and radionuclide production programs. Maintenance work on an exhaust damper had evidently loosened the particles, and carried them up the stack in two bursts when the fans were restarted for testing, shut down, and returned to continuous operation the next morning. A rough, upper limit estimate of 16 curies was given as an estimate of the activity on the ground in the vicinity of the central stack as a result of the ruthenium-106 releases. Measured dose rates in the isotopes area were found to be as high as 10 millirem per hour, with some individual particles reading as high as 15 millirem per hour. The first release apparently travelled to the southeast, while the second release the next morning apparently travelled to the northwest. Laboratory streets, surfaces, and cars were decontaminated. No contamination was found off-site. (Borkowski et al., 1960; Ohnesorge, 1986).



Since at least 1962, Building 3019 dissolver and process vessel off-gas has passed through a local absolute filter and then to the 3039 area for scrubbing, additional filtration, and discharge to the 3039 stack. Cell ventilation air is discharged to the 3020 stack after passing through a caustic scrubber and an exhaust (ORNL, date unknown).

While a stack and vent survey published in 1987 indicated the presence of 1197 emitting vents and stacks, major vent sources at ORNL consist of four continuously sampled stacks (Dumont, 1987; Mitchel, 1992). The central "3039" stack receives exhaust from buildings in the radioisotope area, research and administration buildings, and the Oak Ridge Research Reactor buildings. The 7911 stack receives gaseous wastes from the High Flux Isotope Reactor, the Transuranium Processing Plant, and the Thorium-Uranium Recycle Facility. The 3020 stack receives wastes primarily from Building 3019, the chemical pilot plant. Stack 2026 serves only the High Radiation Level Analytical Laboratory.

#### Solid Waste Management

The problem of disposal of solid radioactive waste resulted in early action in a 1944 letter that suggested that radioactive material and contaminated trash be placed in specially-marked metal cans and that "a suitable location for the burying of this material could be provided over on the burning ground. A suitable pit with enclosed fence could be made." (Bates, 1983). This pit became the site of the first solid waste burial ground at the Laboratory. Six different solid waste storage areas (SWSAs) have been used during the life of the laboratory, as summarized in Table 2-7.

The first three were located in the Bethel Valley near the site of the current Laboratory complex based on convenience rather than knowledge of or regard for mobility of the various radionuclides being buried. The later burial grounds were located in the Melton Valley to take advantage of soil properties which tend to immobilize most radionuclides. In the late 1950s to early 1960s, ORNL also served as the major disposal site for wastes from Argonne National Laboratory, Knolls Atomic Power Laboratory, Mound Laboratories, Battelle Memorial Institute, General Electric, and Radiological Service Company (Bates, 1983; Browder, 1959).

#### Criticality Safety

Criticality experiments are controlled tests performed with fissionable nuclear materials to establish or evaluate safe handling procedures for these materials so that criticality accidents do not occur, or to study the behavior of nuclear materials that are intended to achieve criticality in nuclear reactor or weapon applications.

**TABLE 2-7**  
**ORNL SOLID WASTE DISPOSAL AREAS**

Disposal Area	Period of Operation	Status	Acreage
1	1943-1944	Closed	1
2	1944-1946	Closed	4
3	1946-1951	Closed	6
4	1951-1959	Closed	23
5	1959-1973	Open for retrievable storage only past 1973.	50
6	1969-	Current status not indicated.	68 (14.5 usable)

Reference: Bates, 1983

Criticality experiments at ORNL date back to late 1945, when a series of measurements was made on assemblies of fluorocarbon-uranium oxide mixture. A second experiment in 1946 used tubes containing a heavy-water solution of a uranium compound latticed in a heavy water tank. An extended series of experiments supporting design of the MTR was conducted in 1947 and 1948. These early experiments were conducted in cells of Building 3019, the chemical processing pilot plant. Experiments with fluorinated uranium compounds in solid form were performed in 1946 in a small laboratory near the gaseous diffusion plant, and were followed by tests in 1947 with experiments with solutions of uranium salts which continued until mid-1950 when the Critical Experiments Facility was completed (ORNL, 1967).

The ORNL Critical Experiments Facility ("CEF"; Building 9213) is located at a remote site in the southwest portion of the Y-12 area. The facility has performed experiments in association with the ANP program, the Army Package Power Reactor, the HFIR, the EGCR, and fuel elements from Savannah River and Hanford reactors (ORNL, 1967). The following four incidents were identified:

- A capsule containing a polonium-beryllium neutron source, enclosing nearly seven curies of polonium and 0.4 gram of beryllium, was accidentally opened in a CEF assembly area. The results were wide dispersal of the polonium throughout one of the assembly areas and resulting in internal and external contamination of an employee. Initial medical and clinical evaluations showed no health effects (Callahan and Ross, 1952; ORNL, 1967). Further information on potential subsequent effects of this personnel contamination event was not pursued in the Phase I investigation.
- On May 26, 1954 in an experiment with a uranium solution in a vertical cylinder, a "poison" cylinder placed in the center of the larger cylinder to absorb neutrons accidentally became tilted from its proper vertical position, resulting in 2.1% excess reactivity. Approximately  $10^{17}$  fissions occurred. Automatic safety systems functioned and the reaction was terminated. The incident occurred in the shielded CEF facility designed for such tests under remote operation. All personnel were shielded by 5 feet or more of concrete. Personnel exposures indicated on film badges ranged from 0.08 to 0.90 rem, with an average of about 0.3 rem. There appears to have been a "skyshine" component to the exposures. "Skyshine" results when radiation is emitted into the air and is scattered back to the ground off of the molecules that make up the air.

Only a few tens of milliliters of solution were displaced from the cylinder during the incident, and no significant damage to the facility was sustained. (Stratton, 1967; Thomas and Callihan, 1958; Callihan, 1958; ORNL, 1967).

- On February 1, 1956, in an experiment with a uranium solution in a vertical cylinder, there was an unintentional over-addition of solution. The safety system actuated, and

waves caused in the solution by a falling cadmium sheet formed a prompt-critical configuration. About  $1.6 \times 10^{17}$  fissions occurred. The incident occurred in the shielded CEF facility designed for such tests under remote operation. A considerable volume of solution was ejected from the cylinder. Personnel exposures indicated on film badges ranged from 0.140 to 0.575 rem. No significant damage to the facility was sustained. "A favorable wind and the isolation of the laboratory made it possible to purge the test cell in which the accident occurred using ventilating fans installed during construction for that purpose." All uranium was reportedly recovered. (Stratton, 1967; Thomas and Callihan, 1958; Callihan, 1958; ORNL, 1967; Boyle et al., 1982).

- On November 10, 1961, a criticality incident occurred when uranium-235-enriched uranium metal that was paraffin-reflected was being assembled. Too rapid approaching of two pieces of metal caused an excursion that yielded  $10^{15}$  to  $10^{16}$  fissions. There was reportedly no exposure to personnel in the building, and no dispersal of radioactive material beyond the assembly area where the incident occurred. The lab was ready again for normal use in 1.5 hour. (Stratton, 1967; ORNL, 1967; Callahan, 1962; Boyle et al., 1982).

## 2.2 SUMMARY OF THE INVESTIGATION PROCESS AT X-10

The process used to investigate historical operations and releases from the Oak Ridge X-10 Site was based on extensive review of historical records and interviewing of active and retired plant workers. This section describes those information sources that were most useful and summarizes the approaches that were used to locate information relevant to the study. Over 1150 documents have been summarized to date and have been entered into the project repository database; of these, at least 380 specifically address the X-10 site.

### X-10 Lab Records

By far the most information that is relevant to the X-10 portion of this study is located in the X-10 Laboratory Records collection. It is located in Building 4500-N. Because of the development and pilot-plant nature of most of the work that has gone on at X-10, and the diligent recording of activities by those involved, a very valuable historical record exists in the correspondence, progress reports, log books, and various other technical reports contained in the Lab Records vault in Building 4500 North. The records located in Lab Records go back to the Metallurgical Laboratory of the Manhattan Engineering District, and continue on to present day. Many of the useful records are in the form of Central Files (CF) memorandums or reports or documents with report numbers beginning with ORNL or ORNL/TM. The documents are grouped by document type and are arranged by numbers on shelves in the Lab Records vault.

The following methods were used to locate relevant documents in Lab Records:

- Author Cards — There are binders of cards located in the Lab Records vault that list publications sorted by author name. These cards do not continue to present day, but this function is provided by newer systems in latter years. Key authors were identified through interviews and document review.
- Division Document Lists — There are also books in the Lab Records containing publications by each Laboratory division. These listings, which are only available for the earlier years, were useful for identifying areas of investigation for each division and for identification of authors that warranted follow-up.
- The LION System — The Library Information On-line Network (LION) is a computer system that is essentially an on-line catalog of the holdings of the Martin Marietta Energy Systems library system at the Oak Ridge facilities. The LION system was used to search by authors, keywords, and subject areas of interest to identify reports or books that might be relevant to the study. The LION system contains only records entered into the library system after about 1981. However, the LION system does reflect some old records that have for some reason been entered into the database.
- Library Card Catalogs — The ORNL Central Research Library in Building 4500-N contains a card catalog which lists records by author name. Most of the records reflected on these cards are available in Lab Records, in the Library, or on microfilm. Others have been requested from the DOE Office of Scientific and Technical Information (formerly the TIC). One method that was particularly productive was to make note of all the names listed on project summary reports, progress reports, and "run summaries" of key programs; following this branching of individuals and documents often resulted in identification of the key investigators and historical documents relevant to this study.
- Laboratory Records Database — A database is maintained which contains records of ORNL technical reports from the 1980s and 1990s. This database was searched for the following keywords in order to evaluate its usefulness in records searching:

history, summary, progress, effluent, waste, annual, monthly,  
White Oak, Inventory, Accident, Discharge, Monitoring,  
Stack, Solvents, Environmental, Toxic, Hazardous, Off-site,  
Leak, Emissions, Sediments, Pits, Trenches

Because this database is limited to fairly recent technical reports, the Laboratory Records Database was not found to be one of the more useful tools for our investigation of historical operations and releases.

It is estimated that the Phase I investigation team reviewed about 5% of the documents contained in X-10 Lab Records. About 500 reports and records have been obtained from X-10 Lab Records for inclusion in the project repository.

#### The Tiger Team Records Center

A collection of records that were collected to support the DOE "Tiger Team" evaluation of ORNL contained records relevant to the study. "Tiger Teams" were groups of DOE staff and contractors that visited DOE sites and inspected programs for many aspects of compliance with DOE orders and environmental regulations. A listing of the titles of documents in the Tiger Team collection was made available. The listing was reviewed, and documents of potential relevance were provided to us for review. A good number of documents were summarized, and copies of many have been obtained. Most of these records pertain to fairly recent activities at ORNL.

#### ORNL Records Center

The ORNL Records center contains inactive records contained in boxes in a vault in Building 4500-N and in several rooms in the 4500-N attic. These records are tracked by records transmittal and receipt cards that describe briefly what is in each transmittal. The cards are filed by division in a card file in the Records Center Vault. Cards for the divisions most relevant to the study were reviewed as an indicator of the types of records sent to this facility. In addition, the Records Management Database contains records for all documents that are scheduled to be destroyed or transferred to the Federal Records Center that were submitted after about 1976. The Federal Records Center used by the Oak Ridge region is located in East Point, Georgia. The system allows one to search records which contain the basic information contained on the transmittal cards. The keywords that were identified in the Task 1 plan were submitted for searching on this system. In some cases records which have been sent to the Records Center have been destroyed. Documentation of the disposition of the records, for example where they are stored, when they were destroyed, sent to the Federal Records Center, or sent back to the originating division, is located on the cards. While some useful information was located in the searching of Records Center holdings, more information of relevance appears to reside in Laboratory Records.

As stated above, document reviews at the Records Center were directed by keyword searches and card file review for ORNL Divisions of interest. It is estimated that the Phase I

investigation team reviewed between 1% and 5% of the documents contained in the X-10 Records Center. The searching of inactive records in the X-10 Records Center should be continued. Some records of interest that have been identified by manual searching of the card file remain to be located for review. Lists of documents that have been sent to the Federal Records Center from X-10, and contents of the Federal Records Center itself, have not been reviewed.

### Directors' Files

Historical correspondence of the directors and key managers of the Laboratory are maintained in a collection in Building 4500-N and are partially reflected in a database. Some of the earlier Directors' Files material is contained in the ORNL Records Center. The more recent records are assigned to a set of pre-existing categories, rather than filed by originating division, and are entered into a database. A search was performed using the same keywords used for the Laboratory Records database described above. Because this search indicated that the Directors' Files contain mostly administrative correspondence, this information source was not investigated in any further detail.

### DOE Information Resources Center

The Information Resource Center for the Environmental Restoration Program is operated by a contractor for the U.S. Department of Energy. This center, which is located at Jackson Square in Oak Ridge, houses the Administrative Record for ORR environmental restoration activities. The Administrative Record is required by USEPA regulations, and is an ongoing document collection effort designated to contain all documents used in making decisions concerning cleanup of the Oak Ridge Reservation and certain off-site locations. A listing of the holdings of the Information Resource Center holdings was obtained and reviewed. The Center was a convenient place to obtain many environmental documents in the early phases of the study that were later located at the originating facilities' document centers.

### The Office of Scientific and Technical Information (OSTI)

The DOE Office of Scientific and Technical Information (OSTI), formerly called the Technical Information Center (TIC), was used as a source of information that was not obtainable from ORNL sources. OSTI is located at 175 Oak Ridge Turnpike. After some documents of relevance to the study were identified in other documents but could not be obtained at ORNL, the ORNL Technical Information Officer searched a computer database called the Reports Holding File (RHF) that indicates document availability at OSTI. The reports include some DOE Oak Ridge Operations documents, some DOE environmental reports, some Manhattan Engineering District and AEC documents, and an Air Force document pertaining to the Nuclear

Energy for Propulsion of Aircraft (NEPA) project. The project information repository contains approximately 50 documents obtained from OSTI.

### Personnel Interviews

Interviews have been conducted with approximately 50 individuals with extensive experience at the Oak Ridge Reservation. The names of initial interviewees were provided by facility managers for key functional areas identified by ChemRisk. From that point, each interviewee identified additional points of contact. Candidates for interviews were also identified from association with key historical documents. Some desired interviewees have not yet been located, and not all desired interviews have been completed at this time. While several individuals thought it necessary to obtain approval from Martin Marietta, DOE, or their current employer prior to consenting to an interview, only one individual has declined to be interviewed.

Notes taken in the course of the interviews have been reviewed by appropriate classification reviewers. The information obtained in the interviews is being summarized and entered into the project repository and the associated database.

In the course of interviews, active and retired workers were also asked about the existence of historical records that would be relevant to the study. In some cases, records were identified that might not have been received or preserved by central document repositories.

### **2.3 FOCUS AREAS OF POTENTIAL SIGNIFICANCE FOR OFF-SITE EXPOSURE**

Based primarily on the qualitative information reviewed during this feasibility study portion of Phase I Health Studies, the following areas of X-10 site operations appear to be those that should receive the highest priority in any investigations to quantify historical contributions to off-site exposures and health risks to potentially affected populations. Following the brief discussion of the recommended focus areas here, summaries of information that is available to support further investigations are presented in Section 2.4.

- **The Production of Radioactive Lanthanum (RaLa Processing)**

RaLa processing took place at Oak Ridge from 1944 to 1956 in order to supply radioactive lanthanum to Los Alamos for weapons development. RaLa runs involved processing large quantities of short-decayed X-10 and Hanford irradiated reactor fuel. Fission products such as iodine-131 (8.05 days), which normally decay away when fuel is allowed to "cool" before processing, were released. About half of the production runs at Oak Ridge occurred before emission controls were in place, and all of the



approximately 87 runs occurred before routine monitoring of airborne effluents was started at ORNL.

- **Thorex Processing of Short-Decay Irradiated Thorium**

Thorex processing of irradiated thorium metal was conducted at Oak Ridge from approximately 1954 to 1960, with the goal being separation of uranium-233 for weapons applications and thorium for breeder reactor applications. Four Thorex runs during 1956 and 1957 processed short-decayed thorium metal that had been irradiated in Hanford and Savannah River reactors to test pilot plant equipment and the Thorex process under high radioactivity conditions. The first two short-decay runs caused fallout of protactinium-233 on the X-10 area, and equipment failure also resulted in a high radioiodine discharge during one of the runs. Airborne contamination up to  $1.7 \times 10^8$  microcuries per cubic centimeter beta and gamma radioactivity was measured at a laboratory area air monitoring station over a 5-hour period (ORNL, 1958). This value is approximately 2000 times higher than the annual average of weekly data from the perimeter monitors during 1957.

- **Graphite Reactor Operations**

The Clinton Pile, later known as the ORNL graphite reactor, operated exactly 20 years (from November 1943 to November 1963). In addition to the approximate 470 curies of argon-41 (1.827 hours) released per day, the graphite reactor also released radioactive uranium, plutonium, and fission products that escaped with the oxidized uranium powder that was drawn by cooling air from the aluminum-canned fuel slugs as they ruptured at a rate of about 13 per year. Exhausted air went unfiltered for the first five years of graphite reactor operation. No caustic scrubbers or charcoal filters were ever added to the graphite reactor stack.

- **Processing of Graphite Reactor Fuel for Plutonium Recovery**

Supporting what was the original mission of the X-10 site, 299 batches of irradiated uranium slugs discharged from the graphite reactor were processed in the chemical separation pilot plant from around late 1943 to early 1945 for separation and purification of plutonium (Jones, 1985). The bismuth phosphate process used was initially quite inefficient in recovery of plutonium from fission products; workers commented on how they were "throwing the baby out with the dishwater." (Genung et al., 1993). Plutonium recovery operations occurred before routine effluent treatment or monitoring systems were in place. Uranium, plutonium, and various fission products were released.

- **Waterborne and Airborne Waste Disposal**

As the longevity and mission of X-10 site operations have grown significantly since the early Manhattan Engineering District days, so have the challenges of disposal of the accompanying wastes. Releases of waterborne radioactive wastes to the White Oak Creek and subsequently to the Clinch River have occurred since 1943. Airborne releases have also been ongoing since the early period of Clinton Pile operations. Reported effluent measurements do not cover the entire period of X-10 operations, do not always identify the radionuclides present, and likely have considerable uncertainties. While waste disposal has been an important activity in terms of X-10 interfaces with the environment, it is a considerably broader topic than the other focus areas. Further detailed evaluation would likely require focusing on specific issues or aspects of historical waste management.

## 2.4 INFORMATION AVAILABILITY FOR FURTHER QUANTITATIVE EVALUATION

In order to support the direction of any future efforts to quantify doses and health risks associated with contaminants released from the X-10 site, information that is available concerning the identified high priority areas for likely off-site significance has been reviewed. This section describes the information that has been identified during the Phase I feasibility study historical investigations.

### 2.4.1 Production of Radioactive Lanthanum (RaLa Processing)

#### Run Schedules:

Table 2-8 contains a schedule of the RaLa runs that were conducted at the Oak Ridge X-10 site. This summary table was prepared based on various references. The references are not always in agreement with each other or internally consistent with details of some runs. In addition, quantities produced often varied considerably as measured at Oak Ridge and as measured at Los Alamos. Oak Ridge did not have the equipment needed for accurate measurement of the product quantity, so rough estimates were made, followed by more accurate measurements at "Site Y" (Los Alamos) in New Mexico. This table does, however, indicate the extent of RaLa production at Oak Ridge; the escalation of production over the years; the nature of production difficulties experienced; and actions taken in response, such as addition of emission control devices.

Measurements of barium-140 radioactivity in the RaLa product were made at Oak Ridge at the time of final lanthanum separation, using a radiation survey meter placed at a known distance from the product shipping cone. At Los Alamos, similar measurements were made several days after the product arrived there, with ionization chambers that had been calibrated with radium

Table 2-8: Summary of RaLa Runs Conducted at Oak Ridge and Shipments to Los Alamos

RUN NO. AND START DATE	COMPLETION OR SHIP DATE	NO. OF SLUGS; X-10 or W= Hanford	CURIES OF Ba DISSOLVED	CURIES SHIPPED Y = per Los Alamos	YIELD %	COMMENTS
1 9/10/44	9/18/44 by truck	216 X		314 ± 20%		Slugs had been in pile 40 days. Cooled 4-14 days to time of shipment. 15 dissolvings were made.
2 10/10/44	10/17/44	180 X		195		Slugs cooled 3-5 days to time of dissolving. High losses. Measured 1100 milliroentgen per hour through 706-C roof after 16 hours of lanthanum-140 growth from barium-140 decay.
3 Separation Time 14:30 CWT, Nov 23rd	11/23/44	180 X		280	50	45 days irradiation. Slugs discharged Nov 12th. 25% of Ba could not be accounted for.
4	1/23/45	X		160		Line break caused 75% of Ba to be lost. Cell 2 badly contaminated, not usable for further runs in 706-C.
5	1/31/45	X		478		Three equipment breakdowns made decontamination and repair necessary.
6	3/26/45	X		315		Poor yield. Cells and ducts contaminated by leak in dissolver jacket.
7	4/3/45	X		560		No difficulties. After this run 73 curies of Sr-89/90 were recovered from the HCl-ether waste.
8	5/16/45	X		750		Best run yet. No difficulties.
9	5/28/45	X		800		Operations normal. Last preparation in 706-C.
"Dress Rehearsal" in new facility. 5/15/45	Not Shipped	199 inactive, 1 "hot" X				First operations in new 706-D building.
1: 5/26/45 or 5/28/45	Not shipped	208 X	1007	250	25	Not shipped; very low yield and gross contamination of product.
2: 6/5/45		624 X	2185	1,230	54	First successful 706-D run. After this run, added a cyclone separator to the off-gas line. Also put new 4" stainless steel off-gas line to the fan house and a stainless steel fan because of bad corrosion. An ammonia system was installed in the off-gas line after the scrubber to bleed ammonia in to complete the neutralization of the off-gas.
3: 7/12/45		831 X	2385	1,150	50	1,000 curies went up off-gas line. The new fan sucked up an entire batch of solution during transfer from the electrolysis cell.
4: 8/6/45		1144 X	2840	2,100	67	
5: 9/4/45		835 X	4356	2,000	46	After Run #5, installed an iodine condensate collection pot on the A3-A4 line to collect fission iodine from the dissolver off-gas. Highest potency collected: 80 millicuries/liter.
Special Preparation				1,000 La-140		After Run #5 a special preparation of 1,000 curies of lanthanum-140 was made for Health Physics, from the fuming nitric acid waste.
6: 11/23/45	~ 12/7/45	803 X	3329	1,500	45	The first 15 slugs loaded for this run stuck in the slug chute on top of an Argonne slug (larger than an X slug) that had apparently got lodged during dummy test runs just completed. Filled dissolver almost full with nitric acid and heated to 95°C for 5 minutes to dislodge. Air sparging as a means of agitation caused very high losses (up to 1,000 curies). This batch showed only 400 curies at Los Alamos.

Table 2-8: Summary of RaLa Runs Conducted at Oak Ridge and Shipments to Los Alamos (Continued)

RUN NO. AND START DATE	COMPLETION OR SHIP DATE	NO. OF SLUGS; X-10 or W=Hanford	CURIES OF Ba DISSOLVED	CURIES SHIPPED Y = per Los Alamos	YIELD %	COMMENTS
7: 12/14/45	~ 12/20/45	924 X	2760	2,100	70.7	Best run to date. The run took 7 days, as scheduled.
8: 1/2/46	1/13/46	1728 X	4557	2,570	52.4	
9: 3/3/46		894 X	3556	2,040	57.5	Best run to date.
Special Preparation				1,275		Four more dissolvings were made and carried through the regular process; the wastes from Run #9 were added and a total of 1,275 curies were delivered to Health Physics for special tests.
10: 4/7/46	4/17/46	886 X	3665	1,800	49.1	
11: 5/6/46	5/15/45	896 X	3938	2,000	50.8	
12: 6/10/46	6/17/45	897 X	3417	2,500	73.2	Best run to date.
13: 8/20/46		812 X	3521	2,900	82.4	After this run, Cell A was decontaminated for the installation of an off-gas line from the A-16 scrubber to the 205 (chemical processing plant) stack. Prior to this, the A-16 fan had discharged into the 30-foot fan house stack. Because of the amount of activity suspected of being carried out in the A-16 system, it was felt that the air contamination hazard would be reduced by discharging through the 205 stack.
14: 12/9/46?	12/10/46	564 X	2540	1423	56.0	After completion of this run, Operations Division took charge of 706-D and the barium-lanthanum process.
15: ~1/28/47	None shipped	X				The run was a failure due to several operational difficulties. Dissolved an additional 300 slugs, but still couldn't produce usable product.
15A: 2/26/47	3/5/47	1383 X	2783	1925	69.2	
16: 4/2/47		793 X	2667	1020	38.2	High decantation losses caused low yield.
17	4/30/47	892 X	3535	2478	74.1	
18: 6/9/47	6/18/47	912 X	3452	2300	66.8	On June 20th installed a new concrete duct to replace an old metal cell ventilation duct. Was expected to considerably reduce radiation on the northeast side of the building.
19	7/22/47	905 X	3087	1800	58.3	
20	8/19/47	1047 X	3390	2700	79.7	Slugs were found to be low in Ba content. Extra slug loadings and dissolvings were necessary.
21	10/20/47	1166 X	4177	2400	57.4	2,500 curies came up missing. More slugs were dissolved. (The final total curies dissolved is unclear.)
22: 11/9/47	11/22/47	1287 X	3995	2200	55.6	Considerable product loss necessitated dissolving of additional slugs.
22A	11/27/47			1320		After shipment of Run 22, the missing 1,320 curies were found in Tank A9. This material was processed and shipped.
23	1/24/48	862 X	3411	2750	80.62	Multiple slug ruptures in the graphite reactor on November 30th caused this run to be rescheduled.
Special Run		12 W "cold"				Dummy run using Hanford slugs to determine the adaptability of the equipment to this type of slug. It was concluded that Hanford slugs could be used with suitable procedure changes.

Table 2-8: Summary of RaLa Runs Conducted at Oak Ridge and Shipments to Los Alamos (Continued)

RUN NO. AND START DATE	COMPLETION OR SHIP DATE	NO. OF SLUGS; X-10 or W=Hanford	CURIES OF Ba DISSOLVED	CURIES SHIPPED Y = per Los Alamos	YIELD %	COMMENTS
24: 2/24/48	3/13/48	1257 X	4276	3575	83.6	High losses lead to additional dissolvings.
25	7/17/48	522 X	2378	1576	67.7	Special run for the AEC ARUU program. The product was delivered, as requested, in three product containers of 1,409 curies, 120 curies, and 26 curies.
26	7/23/48	971 X	4501	2419	53.7	
27: 8/25/48	9/3/48	851 X	3087	2196	71.1	By this time a carrier had been built for transporting Hanford slugs to Oak Ridge. Plans were made to use Hanford slugs for the next run.
28	11/21/48	W	2619	1380	52.7	Hanford slug run. Low quantity shipped due to low Ba-140 content of Hanford slugs. Operational difficulties at Hanford resulted in lower than normal pile flux. Late in 1948, it was determined that Building 706-D was a major contributor of airborne radioactive particles at ORNL. Air filter houses were installed on the cell ventilation duct, the A16-205 off-gas line, and the A4-205 off-gas line.
29: 1/10/49	1/16/49	38 W	3061	1545	50.5	Hanford slugs again had low Ba assay. Monitoring of an airborne radioactive plume was performed during this run using experimental instruments mounted in an airplane (Davis et al., 1949).
30: 2/14/49	2/27/49	38 W 274 (5857) X	4097	1345	38.4	Had to add additional Oak Ridge slugs because of low Hanford Ba assay.
31: 3/19/49	3/25/49	76 W	6252	3240	51.9	Air activity from drains.
32: 4/17/49	4/22/49	76 W		3800		Two instances of high air activity.
33						Several instances of high air contamination. An investigation disclosed that the A4-205 off-gas line condensate pot was a source of air contamination during Run 33. A small blower has been installed which will vent the air from this pot to above the 706-D roof level. The A-16 off-gas lines and filter box will be relocated by 7/9/49 to provide room for new 900-area off-gas stack equipment (Ramsey et al., 1949)
34: 7/10/49	7/15/49	76 W		4284		High air contamination when product was removed from cell.
35: 8/14/49	8/25/49	74 W		1750		Excessive product decay due to long processing time.
36: 10/9/49	10/14/49	74 W		4275		Some air contamination.
37: 11/20/49	11/25/49	74 W		4240		
38: 1/1/50	1/6/50	74 W		4670		New process being tested. Some air activity.
39: 1/15/50	Not Shipped	74 W		-		Experimental run on new process.
40: 3/13/50	3/18/50	73 W		3440		First run exhausted through 900 area (central) stack.
41: 4/9/50	4/13/50	76 W		4800		One air activity incident.
42: 6/11/50	6/16/50	74 W		4140		
43: 4/2/51		69 X		157		A low level run.

Table 2-8: Summary of RaLa Runs Conducted at Oak Ridge and Shipments to Los Alamos (Continued)

RUN NO. AND START DATE	COMPLETION OR SHIP DATE	NO. OF SLUGS; X-10 or W=Hanford	CURIES OF Ba DISSOLVED	CURIES SHIPPED Y = per Los Alamos	YIELD %	COMMENTS
44: 5/13/51		73 W		3157		Losses over 6,100 curies. The fate of the "lost" material is not evident from records reviewed to-date.
45: 8/12/51		150 W		8722		Losses over 10,500 curies. The fate of the "lost" material is not evident from records reviewed to-date.
46: 1/12/52		176 W		22368		
Test Run		65 X				A test run with 65 X slugs was made during March 1952. Losses were 137.6 curies.
Test Run		65 X				A test run with 65 X slugs was made during May 1952.
47: 5/21/52		231 W		27832		Contamination incident of personnel and operating area. Losses over 10,200 curies.
48: 6/28/52		214 W		23,322		Some contamination of operating area.
49: 7/28/52	Shipped 8/52	226 W		26,615		
50: 8/18/52	None shipped					Equipment failures caused loss of all but 2,000 curies of the product. Not shipped.
51						
52: 5/23/53	6/4/53	71 W		7,000 to 13,000		8" Hanford slugs (first mention). Instrument troubles. Completion date per CF 54-11-186.
52A		Run 52 material		3,700		Run 52A was a rerun to recover losses from Run 52.
53: 7/1/53		125 W		17,000		8" Hanford slugs.
53A				34,000		
54: 11/4/53	11/25/53			none		Run was a total failure. Release of gases contaminated building. Completion date per CF 54-11-186.
55 & 55A: 1/15/54		192 W	84,683	64,805 45,091 per Y		Biggest and most successful run to date. 8" Hanford slugs. Testing new slug coating and bonding removal.
56: 4/26/54				none		Run was a total failure. None shipped. The operation of equipment far above its capacity caused the "most serious accidental release of activity ever experienced in the history of the process." Slugs were thermally hotter than normal. Dissolver solution was blown up the slug chute into the operating area. Building shut down for decontamination for 11 weeks.
57: in 1954			37,020	22,800 per Y		
58: in 1954			42,664	19,942 per Y		

Table 2-8: Summary of RaLa Runs Conducted at Oak Ridge and Shipments to Los Alamos (Continued)

RUN NO. AND START DATE	COMPLETION OR SHIP DATE	NO. OF SLUGS; X-10 or W=Hanford	CURIES OF Ba DISSOLVED	CURIES SHIPPED Y = per Los Alamos	YIELD %	COMMENTS
59: in 1955				1955: 15,100 avg per run; excluding one special run of 700 Ci		
60: in 1955						
61: in 1955						
62: in 1955						
63: in 1955						
64: in 1956			49,124	16,300		
65: in 1956			54,662	17,400		
66: in 1956			50,900	15,600		
67: in 1956			48,050	14,900		
68: in 1956			47,869	15,000		

References: Thompson, 1949; Rupp and Witkowski, 1955; Seagren and Witkowski, 1957; Ramsey et al., 1949; Bradshaw and Cottrell, 1954).

sources to indicate the "radium equivalent curies" of lanthanum-140 that were received. Values measured at Los Alamos were reduced by 10% to account for barium-140 gamma radioactivity, and were used to calculate the amount of barium-140 that had been present at last separation time at Oak Ridge. Experience showed that if Oak Ridge estimated that 1000 curies of barium-140 were present at last separation time in a shipment, Los Alamos would usually measure 1000 "curies" of lanthanum-140 about 5 days later. (Spence, 1947; Thompson, 1949) Subsequent analyses by absolute beta counting of samples using end-window Geiger-Muëller detectors indicated that the curie size of RaLa runs prior to April 1951 had been underestimated by a factor of 1.45 (Larsen, 1951).

#### RaLa Operational Details:

RaLa process development work is well documented. (Baldwin and Savolainen, 1947; Eister, 1949; Higgins, 1951; Blanco, 1951; Blanco and Kibbey, 1951; Schubert, 1945; Higgins et al., 1950; Blanco, 1952a; Culler et al., 1949; Blanco, 1952b; Unger, 1951; Thompson, 1949)

The mainline flowsheet for the RaLa process included the following steps:

1. Dissolution of the Metal Slugs - The uranium slugs were dissolved in nitric acid after their aluminum jackets had been removed with nitric acid and  $\text{NaNO}_3$ .
2. Extraction - The barium-140 was extracted from the solution by coprecipitation with  $\text{PbSO}_4$  carrier.
3. Metathesis - The  $\text{PbSO}_4$  and  $\text{BaSO}_4$  were converted to the carbonates by treatment with  $\text{K}_2\text{CO}_3$ .
4. Electrolysis - The lead was removed from the nitric acid solution of the metathesis cake by electrolysis to  $\text{PbO}_2$ .
5. Volume Reduction - The solution was evaporated to a small volume.
6. Barium Nitrate Precipitation - The barium as the nitrate was precipitated by adding fuming nitric acid.
7. Barium Chloride Precipitation - For further purification from iron, lead, strontium, etc., the barium was precipitated as  $\text{BaCl}_2$  from a hydrochloric acid-ether mixture. The precipitate is evaporated to dryness for shipment.

(Baldwin and Savolainen, 1947)



The 706-D equipment was operated far above its designed capacity of 500 curies in the dissolver and extraction section and 10,000 curies in the purification section (Rupp and Witkowski, 1955).

We have obtained a copy of a log book that contains a listing of secret documents issued by the Operations Division between April 1945 and March 1958. The log book identifies, in part, report numbers of run summaries for the RaLa runs that were conducted by the Operations Division. These extend from Runs 14 in December 1946 through the last run in 1956. This log book identifies run reports, run analytical reports, health physics reports for the RaLa building, area and waste monitoring reports, and various correspondence concerning RaLa production. These reports should be consulted for the most detailed operational information concerning the RaLa runs.

As discussed earlier in this report, because of the short half-life of barium-140, the reactor fuel used for RaLa processing had to be fed to the process shortly after removal from the Oak Ridge or Hanford reactors. While one reference states that the slugs were allowed to cool for about five days after extraction from the pile (Davis et al., 1949), decay periods as short as three days between discharge and dissolution are documented (Thompson, 1949). One 1946 report indicates that "Slugs for Ba<sup>140</sup> processing are irradiated at least 40 days in the center of the pile and are charged to the 706-D dissolver with less than one day's cooling." (Reynolds et al., 1946) Eister (1949) referred to 40-day irradiation and one day cooling for X-10 slugs. As pointed out earlier based on Table 2-4, 5-day cooling reduces the activity contained in irradiated slugs by about 33% compared to 1-day cooling. It is not yet clear how closely the irradiation and cooling histories of individual slugs or groups of slugs were tracked or recorded. Examination of RaLa run summaries should clarify this.

An evaluation of iodine-131 distribution in wastes from the RaLa process in 1946 revealed the following:

- Approximately 3 curies of iodine-131 (8.05 days) per slug are theoretically available. This is based on 900 slugs processed in Run 10, with curies calculated "to time of pushing" (discharge from reactor), and a calculation that iodine present would be equal to 46% of the barium-140 (12.789 days) initially present. The total iodine-131 "available" in this run was approximately 2800 curies. The total found by analysis was 2100 curies.
- About one-third of the total found (670 curies) showed up in waste from the dissolver scrubber. The volume of this waste was about 58,000 gallons, giving an iodine-131 concentration of "only" 12 millicuries per gallon.
- Two-thirds of the total found (1400 curies) remained in the dissolver solution and went through the extraction into the metal waste solution without significant diminution.

- No attempt was made to determine the amount of iodine escaping from the scrubber to the stack "as the amount is believed to be small and no good method of sampling was available."

(Reynolds et al., 1946)

In late 1948 and early 1949, airborne measurements of radioactivity were taken in an attempt to demonstrate the feasibility of certain instruments for airborne measurements and to study meteorological effects on plume dispersion. The instruments used included an atmospheric conductivity chamber, a high-pressure ionization chamber, an NRL Dual-Channel Airborne Radiation Detection Unit, and an alpha counter for particulate filters. The most significant flight occurred on January 11, 1949, while RaLa Run 29 dissolving was in progress. This run was noted to have involved Hanford slugs that had lower barium content than expected; their content of other fission products could also have been lower than usual. The airborne plume was detected out to a distance of 17 miles when flying at 1300 feet above sea level, or about 300 feet above the ridges. (Davis et al., 1949). As shown in Table 2-8, RaLa Run 29 was documented to have involved Hanford slugs with low barium content; content of other fission products could also have been lower than normal. The fact that levels of airborne radioactivity from routine Oak Ridge operations were not high enough to fully test the airborne detection equipment in this series of flights apparently provided motivation, at least in part, for the deliberate airborne release of radioiodine from Hanford, in what has become known as the "green run."

During 1948, RaLa releases were evaluated based on sampling through K-25 barrier filters, sampling with CWS filters, and evaluation of filter dust burden. The 706-D cell ventilation line released about 15,000 millicuries (15 curies) of gamma active particle material (reported as 1 MeV per photon, one photon per disintegration) during Run 28 and 2,600 millicuries during Run 29. The dissolver off-gas line discharged 360 and 420 millicuries, respectively. The vessel off-gas line discharged about 2,200 and 2,500 millicuries, respectively. (Miller, 1949).

Another sampling study in 1948 evaluated particulate releases from RaLa dissolver off-gas, vessel off-gas, and cell ventilation lines during Run 28 and Run 29. Sampling equipment consisted of a CWS filter mounted behind Aerotec cyclones. Prior to Run 29, a filter house was installed on the cell ventilation line; during Run 29, measurements were taken before and after the filter house. (Coughlen, 1950; ORNL, 1948).

A summary of modifications made to air handling practices for RaLa is as follows:

- After Run #5 in September 1945, an iodine condensate collection pot was installed on the A3-A4 line to collect fission-product radioiodine from the dissolver off-gas. Highest potency collected: 80 millicuries/liter.
- After Run 13 in August 1946, 706-D off-gas (from the A-16 scrubber) was rerouted to the 205 (chemical processing pilot plant) stack. Prior to this, the fan had discharged to the local 30-foot fan house stack. Because of the amount of activity that was suspected of being carried out in this system, it was felt that the air contamination hazard would be reduced by discharging into the 205 stack.
- Late in 1948, it was determined that Building 706-D was a major contributor of airborne radioactive particles at ORNL. After Run 28, a temporary filter building containing two American Air Filter Co. AAF#FG50 fiberglass filters backed by U.S. Army Chemical Warfare Service CWS #6 asbestos-base paper filters, was constructed and installed on the cell ventilation duct. Run 29 sampling showed a release of 120 millicuries, indicating 97% removal efficiency.
- An investigation disclosed that the A4-205 off-gas line condensate pot was a source of air contamination during Run 33 in mid-1949. A small blower was installed to vent the air from this pot to above the 706-D roof level. The A-16 off-gas lines and filter box will be relocated by 7/9/49 to provide room for new 900-area off-gas stack equipment. (Ramsey et al., 1949)
- Run 40 in March 1950 was the first run to be exhausted out the new "900 area" stack (the central off-gas, or isotope area stack). In the 900 area off-gas treatment facility (as proposed in 1949) 3,000 cubic feet per minute of exhausted air from 706-D process cells passed through Trion stainless steel electrostatic precipitator collecting cells, American Air Filter Co. AAF#FG50 deep-bed pocket filters containing "fiberglass filterdown", and then U.S. Army Chemical Warfare Service CWS #6 asbestos-base paper filters before discharge up the new 250-foot-high red brick stack.

(Thompson, 1949; Miller, 1949)

When Hanford slugs were being used in RaLa production in 1949, the activity levels in wastes from Building 706-D to gunite waste tank W-12 were between 100,000 and 1,000,000 beta counts per minute per milliliter (c/m/ml) and 10,000 gamma c/m/ml (Browder, 1949). These values convert to between approximately 1,700 and 17,000 beta counts per second per milliliter (c/sec/ml) and about 170 gamma c/sec/ml. A study in 1954 correlated airborne particulate

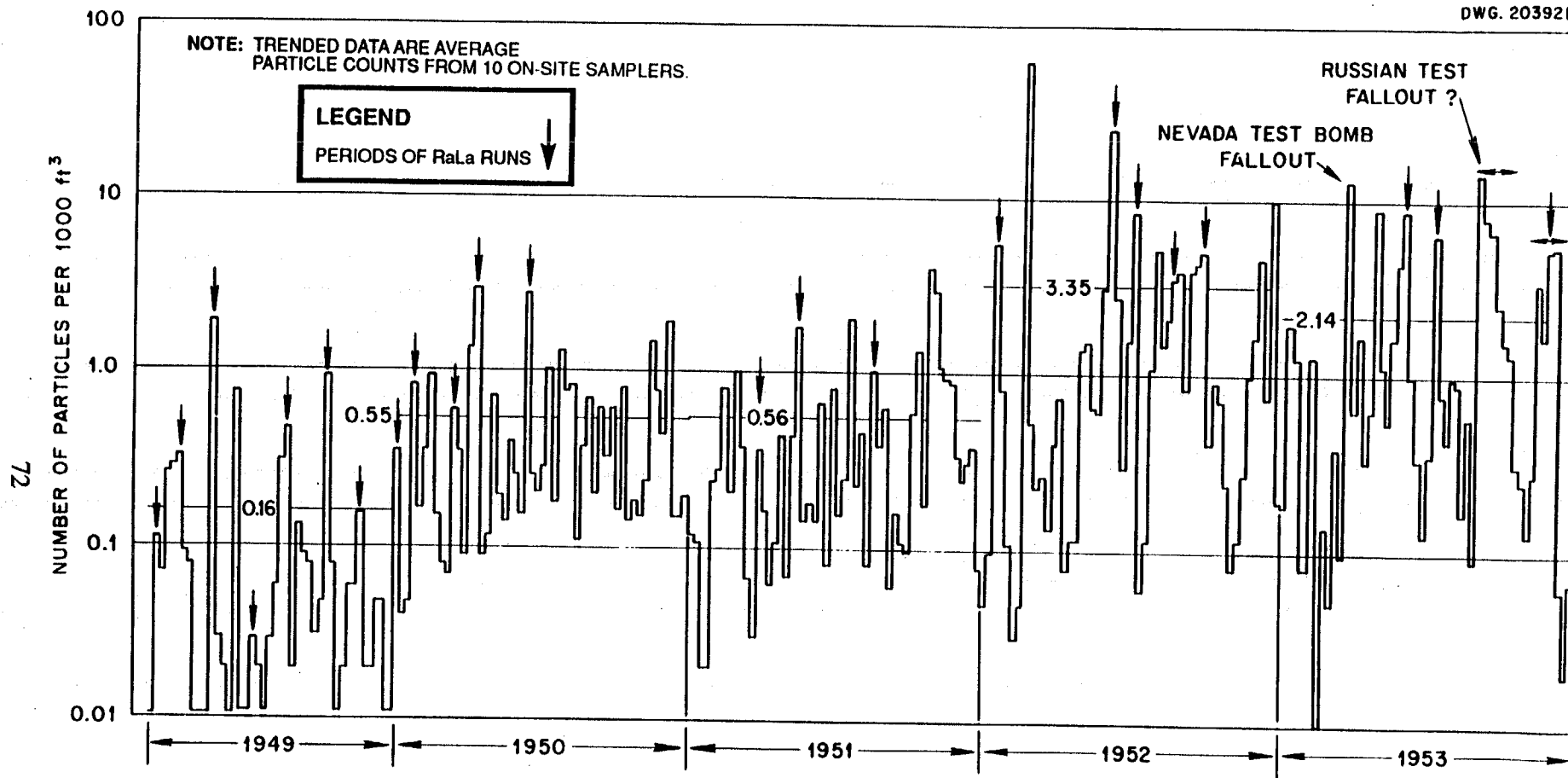
contamination measurements with laboratory processes and reactor operations over the period from March 1949 to June 1954 (Bradshaw and Cottrell, 1954). The study looked at existing ambient radioactivity measurement data and also involved taking special measurements. Over the five-year study period, almost every RaLa runs coincided with a peak in the number of radioactive particles collected per cubic foot of air by continuous air monitors located about the Laboratory site. The particles were counted by autoradiography and visual inspection of the photographic film under a microscope. The times of RaLa runs are depicted as vertical arrows in the Figure 2-9 plot of continuous air monitor data. The cause of the highest peak in Figure 2-9 is not stated in the Bradshaw and Cottrell report, and was not evident from the records reviewed during the Phase I investigations. Bradshaw and Cottrell (1954) concluded the following:

- In spite of corrective measures taken in 1949 and 1950 to reduce air contamination from the RaLa process, this process was found to be the major contributor to Laboratory area airborne particulate contamination during the November 1953 run.

Note: The RaLa run during November 1953, Run 54, was listed as a total failure, with releases of gases contaminating the building and no product shipped.

- The process liquid waste storage tank, not the process stack, was the major source of air contamination from the RaLa process during the period. Jetting and sparging operations in the tank were the most consistent particle producing processes.
- The activity level of krypton and xenon discharged by the stack was about  $10^3$  microcuries per cubic centimeter when averaged over the entire November 1953 RaLa run, corresponding to a total release of about 100,000 curies. About 30 curies of other radionuclides, mainly gaseous iodine-131, were also discharged.
- Mixed fission product activity near the vent in the waste tank was about  $2 \times 10^6$  microcuries per cubic centimeter of air when averaged over the duration of the second run. Most of this activity was in the form of particulates.
- The continuous air monitoring method in place at the time would not detect all particles present, based on higher particle counts obtained with membrane filters.

On April 29, 1954, a large amount of activity was released from the top of the RaLa process cell in Building 706-D. The activity was released during addition of 390 pounds of 60% nitric acid to the dissolver, where 101 Hanford slugs had been loaded. Three successful dissolvings



SOURCE: BRADSHAW AND COTTRELL, 1954

of about 18 slugs had already been made. The slugs were allowed to set dry in the dissolver for about 28 hours. The partially dissolved slugs, because of their exceptionally intense radioactivity, had become very (thermally) hot during the time that they were not covered by liquid, and the reaction with nitric acid was more violent than expected. When the nitric acid was added, the solution containing the radioactivity was blown into the operating area through the slug loading chute and the solution addition lines. Release of the activity continued for about two hours until the regular "hot" (high-activity) off-gas system could handle it. The release occurred during the 4 p.m. to 11 p.m. shift. Winds were mostly from the south during the release. High airborne activity (principally short-lived iodine) occurred in Building 706-D (3026), all buildings in the radioisotope area, the rolling mill (3012), Bulk Shielding Reactor (3010), LITR (3005), Hot Research Shop (3006), and later to some extent in the Graphite Reactor (3001). (Rupp and Witkowski, 1955; Stanley, 1954)

Building 3026, the RaLa process building, was out of service for 11 weeks for decontamination. After the release ended, exposure rates at building windows 20 feet above the process cells exceeded 10 roentgen per hour; inside, exposure rates exceeded 100 roentgen per hour. The short-lived radioactivity was allowed to decay for ten days before the building could be entered for decontamination. Samples taken from the settling basin appeared to be significantly elevated at 11:00 p.m. The air monitor at the rock quarry on Bethel Valley Road showed a very slight rise after the accident. Processing of the first part of the RaLa run proceeded on April 30th. (Rupp and Witkowski, 1955; Stanley, 1954).

The RaLa process building was also blamed for general contamination of the Laboratory grounds on several occasions during 1954. On one occasion, high stack discharges were caused by a plugged caustic line to a dissolver off-gas scrubber. In another case, the scrubbers operated normally but not effectively enough to avoid general area contamination under atmospheric inversion conditions that existed at the time. (Rupp and Witkowski, 1955).

#### 2.4.2 Processing of Short-Decay Thorium Metal (Thorex Processing)

As described earlier, Thorex processing of irradiated thorium metal was conducted at Oak Ridge from approximately 1954 to 1960. The goals of the Thorex pilot plant were to recover uranium-233 (159,200 years) for weapons applications and thorium-232 (14,050,000,000 years) for breeder reactor uses from fission and activation products, including the highly-radioactive protactinium-233 (27.0 days).

Laboratory development of the Thorex process took place at ORNL in the early 1950s (ORNL, 1952; Gresky et al., 1953a; Gresky et al., 1953b; Gresky et al., 1953c).

Four Thorex runs during 1956 and 1957 processed short-decayed Hanford and Savannah River fuel to test pilot plant equipment and the Thorex process under high radioactivity conditions. Radiation levels in the building were considerably higher than normal. Table 2-9 summarizes the Thorex runs for which run summaries have been reviewed to date.

The ORNL Thorex flowsheet used in most runs involved the following steps:

- Dissolution of the irradiated metal in excess nitric acid
- Feed adjustment and acid recovery
- Solvent extraction of thorium and uranium, partitioning, and stripping. (Thorex used tributyl phosphate as the solvent and kerosene as a diluent )
- Continuous solvent recovery
- Semi-continuous sorption of uranium on ion-exchange resin
- Batch elution of sorbed uranium from the resin bed

(McDuffee et al., 1956)

#### Operational Details:

The Thorex flowsheet was designed to process thorium which had been irradiated until levels of 1500 grams of uranium-233 per metric ton of thorium (abbreviated "1500 g/t") were produced and then allowed to decayed for 180 days. Around 1957, interest increased in processing thorium irradiated to levels of 3,000 to 4,000 g/t and decayed 20 to 30 days. Short-decay runs were planned in order to determine the chemical suitability of the ORNL Thorex flowsheet for processing such feeds and to define the problems with such processing. (McDuffee, 1957a).

The heat output from the intense radiation given off by the short-decayed Savannah River metal limited loading of the transport carriers to less than 50% of their normal capacity. While the average decay times reported for runs SD-1, SD-2, and SD-3 were 33, 60, and 32 days, respectively, one report indicates that dissolvings were begun approximately 10 days earlier than the average decay time (Winsbro, 1957; Yarbro, 1958).

TABLE 2-9  
THOREX PILOT PLANT RUNS

Run Designation	Dates	Cooling Time (days)	Average Grams U-233 per Ton*	Comments
HD-7, -8	Separated 4/15/55	160	560	Hanford Metal
HD-6R, -9	Separated 5/10/55	215	1292	Hanford Metal
HD-10	Separated 5/23/55	166	1044	Hanford Metal
HD-11, -12A	Separated 9/12/55	260	1117	Hanford Metal
HD-13, -14A	Separated 10/11/55	253	1082	Hanford Metal
HD-12B, -14B	Separated 10/15/55	268	1083	Hanford Metal
HD-15	Run Summary 6/6/56	305	1374	Hanford Metal
HD-16	Run Summary 10/1/56	349	1440	Hanford Metal
HD-17	Run Summary 3/4/57	246	1285	Savannah River Metal
HD-18	Run Summary 6/12/57	303	1432	Savannah River Metal
HD-19	7/11 - 7/27, 1956	lot 1: 109 lot 2: 29	lot 1: 3340 lot 2: 2260	SHORT DECAY RUN; Hanford Metal
SD-1	8/15 - 8/20, 1957	24 - 31	3000	SHORT DECAY RUN; Savannah River Metal
SD-2	9/8 - 9/21, 1957	65	3500-4000	SHORT DECAY RUN; Savannah River Metal
SD-3	10/31 - 11/4, 1957	16 - 28	4000	SHORT DECAY RUN; Savannah River Metal
HD-20	Run Summary 10/30/57	396	3210	Hanford Metal
HD-23	Run Summary 11/12/57	468	3089	Hanford Metal
AWD-1	Run Summary 1/6/58	346 days after HD-19		Processed first-cycle raffinate from HD-19
SSD-1	Run Summary 4/1/59	2 yrs	2300	"Simulated short-decay run." Recycle thorium product spiked with SD-3 solution plus 2y-decayed metal
HD-27	Run Summary 5/11/59	700 and 70	2000 and 3500	
AWD-2	Run Summary 3/1/60	1 yr		Processed raffinate from SD runs.

\* An indicator of time spent in the reactor.



Run HD-19 results, compared to longer-decay runs, were summarized as follows:

- Additional solvent extraction cycles were needed.
- Ruthenium removal efficiency dropped by a factor of 1000.
- Operator exposure increased five-fold.
- General radiation levels in the building went up by a factor of 5.

(McDuffee and Shank, 1960)

Prior to Runs SD-1 through SD-3 a caustic scrubber was added on the dissolver off-gas system for iodine removal, and the dissolver off-gas line was routed to the graphite reactor filter house prior to Run SD-3. (McDuffee and Shank, 1960).

Observations and conclusions regarding Runs SD-1 through SD-3 included the following:

- Gamma activity of the product exceeded specifications by a factor of 200.
- Significant radiation-induced solvent degradation occurred.
- The caustic scrubber removed about 99% of the iodine but none of the particulate activity, which was primarily protactinium.
- Radiation intensity at certain process filters increased to about 200,000 roentgen per hour within several hours after start of operation. One operator received an estimated 63 rem exposure when he accidentally entered the protactinium-233 decay-storage cell. This accident could have been fatal if he had not immediately realized his location. A clerk also received a 13 rem exposure.
- A high iodine discharge to the environment occurred during run SD-1 when a remote-operated bypass valve on the caustic scrubber failed in its open position.

(McDuffee and Shank, 1960)

Following the second high personnel exposure, the short-decay runs were terminated. Some dissolved uranium was allowed to remain in the feed preparation system. In preparation for resumption of operations several weeks later, sparging of this solution resulted in a burst of airborne contamination throughout the pilot plant building (3019). Continuous air monitor filters showed the activity to be mostly iodine-132 (2.3 hours). The activity initially suggested that a

neutron excursion (criticality incident) had occurred, but further investigation indicated that tellurium-132 (78.2 hours) decay to iodine-132 was the source. (McDuffee and Shank, 1960).

#### Environmental Impacts:

An increase in radioactive fallout on the X-10 area during the first two short-decay runs indicated an increase in area contamination resulting from short-decay runs. A program was established prior to SD-3 to determine the source and nature of the fallout. Also, prior to SD-3, the dissolver and vessel off-gas streams from the Thorex pilot plant were routed through the 3002 filter building and to the graphite reactor stack. Cell ventilation air continued to be routed to the pilot plant stack. (Yarbro, 1958).

During the first two short-decay runs, increased fallout was reportedly detected as far away as Berea, Kentucky (McDuffee and Shank, 1960). Berea is approximately 120 miles to the north of Oak Ridge.

Data from Berea that were located during the Phase I investigation are limited to an annual average of  $9 \times 10^{-13}$  microcuries per cubic centimeter of beta and gamma radioactivity, which is approximately a factor of 1.6 less than the average seen at the DOE boundary perimeter monitors and a factor of eight less than the average seen at the laboratory area monitors (ORNL, 1958). In the 1957 applied health physics annual report (ORNL, 1958), a majority of the activity at the perimeter stations was attributed to weapons test fallout. Further evaluation of data from the Berea sampler will be necessary before the statements about remote fallout from Thorex processing can be fully addressed.

More useful data for air monitors in the laboratory area are contained in the 1957 applied health physics annual report. The highest single incident of air contamination during 1957 reportedly occurred on September 12th. This was during Thorex short-decay run number 2. A sample collected by a laboratory area continuous air monitor over 5 hours on that day indicated an average contamination level of  $1.7 \times 10^{-8}$  microcuries per cubic centimeter of beta and gamma radioactivity. This is compared to a maximum weekly average from a single laboratory area monitor from a single week during 1957 of  $5.6 \times 10^{-10}$  microcuries per cubic centimeter and an annual average of weekly data from all laboratory area monitors of  $8.2 \times 10^{-12}$  microcuries per cubic centimeter. The elevated releases on September 12th resulted from the faulty operation of a Building 3019 off-gas system scrubber that was mentioned earlier. The activity was reported to primarily have been radioiodine, and persisted "for only a short time" (ORNL, 1958).

A group of 18 fallout trays was installed in a cross-pattern with the pilot plant stack as the center. The trays were placed 75 feet to 425 yards from the base of the stack. The data from

these trays indicated a steady decrease in fallout following the conclusion of Run SD-3. Data are presented for October 28 through December 31, 1957. The particle activity, broken down into three groups, indicated the greatest reduction occurred in the high-activity particles. Analysis of the particles collected showed that the particles were iron oxide and that the activity was greater than 99% protactinium-233, the parent of uranium-233. (Yarbro, 1958; McDuffee and Shank, 1960).

To determine more precisely the source of the fallout, gas samplers were installed in the cell ventilation duct, in the duct between the graphite reactor filter house and its stack, and in the 10-inch duct that carries vessel off-gas from the pilot plant to the graphite reactor filter house. The samplers were designed to deliver an isokinetic sample to a cyclone separator followed by a glass-fiber filter. The equipment was installed on December 10, 1957. Results from samples between December 11th and December 31st, 1957 are presented in ORNL Central Files memorandum 58-2-149. All three samplers indicated that over 99% of the activity was protactinium-233. The data were interpreted to indicate that the major source of airborne contamination was the cell ventilation header, in which rust was flaking off and carrying protactinium-233 out the pilot plant stack. (Yarbro, 1958).

### 2.4.3 Graphite Reactor Operations

#### Operational Data:

The Clinton Pile discharged radioactivity in the form of activation products formed by irradiation of the cooling air, uranium oxide particles released after failures of the aluminum jackets encasing the uranium metal fuel slugs allowed air to contact the uranium, and gaseous and particulate fission products formed in the uranium fuel and released from ruptured slugs. Argon-41 (1.827 hours) was formed by activation of the 0.9% of argon gas that is present in the atmosphere. The cooling air passed through the reactor only once, then was exhausted up the 200-foot stack. The release rate of argon-41 has been estimated to be approximately 470 to 500 curies per day (Cagle, 1955; Ohnesorge, 1986). The 200-foot pile stack was provided primarily to allow for dispersion and decay of this gas before it touched ground.

As of August 31, 1948, 163,661 aluminum-canned natural uranium "slugs" had been loaded ("charged") into the ORNL graphite reactor; 40,639 were in the pile at that time. Of the slugs used through that date, 48, or 0.03% of the total, experienced failure of their aluminum jackets. When secondary failures caused by the slug ruptures are included, the number rose to 67 (Cagle and Emler, 1948). The graphite reactor filter house was put into service shortly after, at 2 a.m. on November 15, 1948 (McLain, 1948c).

Uranium fuel slugs in the graphite reactor began to rupture around September 27, 1944, and continued to fail at a rate of about 13 per year from 1944 through 1948 (Morgan, 1948).

During removal of ruptured slugs from the graphite matrix, visible clouds of dust were swept from the channels by the cooling air (Cheka and McAlduff, 1949).

On Sunday evening, November 30, 1947, a substantial increase in the activity of the cooling air exiting from the pile indicated that the jacket on a slug had ruptured. The pile was shut down, and the row containing the slug located by measurement of radioactivity leaving the individual fuel channels. The uranium oxide formed had not only ruptured the aluminum jacket, but had also wedged the slugs into the graphite slug so tightly that they could not be pushed out with a long rod as had been the usual practice. After applications of force by means of a sledge hammer and later an air hammer did not clear the channel, it became evident that sufficient force could not be exerted to dislodge the slugs without endangering the stability of the graphite structure. It took nine days using core drills, chisels, and probes to discharge the 13 distorted slugs that were completely oxidized. About 32 pounds of uranium were released from these 13 slugs; "most" of this material was estimated to have fallen into the canal and the bottom of the exit air plenum chamber. (Emlet, 1947; Cagle and Emlet, 1948).

As a result of the November 1947 slug ruptures, some of the released uranium oxide particles were distributed throughout the pile matrix and along the exit air ducts, as was evidenced by unprecedented activities in the air of the exit duct for more than a month after the event (Unknown, 1949).

Records are available of the channel numbers of slugs placed in the pile, the dates inserted into the reactor and approximate dates of rupturing, total age in days at time of rupture, accumulated kilowatt-hours of exposure in the reactor, position in row, and temperature range (Cagle and Emlet, 1948).

Records of graphite reactor operations are contained in log books which are archived in the ORNL Records Center. Items documented in the log books include:

- Reactor power levels
- Insertion of samples in the reactor for irradiation
- Health physics survey operations
- "Probe" readings of particulate activity in cooling air
- Discoveries of ruptured slugs
- Slug discharging and charging operations

During the period of early plutonium production and separation at Clinton Laboratories, the graphite reactor was required to operate at the highest possible power level without exceeding the temperature limitations of the fuel jackets. This practice was carried over after plutonium separation ceased and up until 1952. Because of diurnal variations of the temperature of the pile

cooling air, power levels during the course of a day varied from 3,400 to 4,200 kilowatts. An approximately constant power level was typically experienced from around 10:00 p.m. until daylight. The pile power level varied from daylight until 2:30 or 3:00 p.m., when peak temperatures typically occurred, and from that point until around 10:00 at night. (Cagle and Ramsey, 1955)

For some time after the slug ruptures that occurred in the fall of 1948, the pile was operated only when there was a wind up the valley of over five miles per hour and when there was no inversion. It was stated that if further slug ruptures occurred before the filter house was complete, the pile would have to be down a large percentage of the time due to these limiting conditions of operation. (McLain, 1948b).

#### Monitoring of Pile Exhaust Air:

Air sampling for particulates in the air exhausted from the ORNL pile began in 1943. Initial monitoring was primarily performed to indicate uranium fuel slug jacket failures.

Cooling air that was drawn through the graphite reactor contained particulate matter from a number of sources. Analyses of dust samples performed around 1948 indicated the presence of uranium oxide from slug jacket failures, graphite dust from the pile matrix, fibers from the rear wall cellamite heat shield, silica from the concrete shielding, and general atmospheric dust that had made it through the inlet air filters (Cagle, 1949).

When the pile first began operation, the only instrument present for measurement of cooling air activity was an ionization chamber instrument ("stack gas monitor") that monitored and recorded only the activity leaving the pile in gaseous forms. The sample line to this monitor originated on the exhaust side of the fans and had a device called a precipitron in it prior to the ionization chamber to remove particles. No measurement of particulate matter was intended, as it was assumed that releases of noble gas fission products would be sufficient to cause indication of slug ruptures with this instrument (Cagle, 1949; Cagle and Emlet, 1948).

The ducting and fans in the pile exhaust system were coated with oil, and would accumulate particulate matter and become radioactive. Exhaust fan housings generally read from 1,000 to 20,000 millirem per hour after a slug rupture. A procedure of monitoring the activity of the exhaust fan housing was adopted, and it was by this method that the first slug jacket failure was detected on September 27, 1944. At that point it was realized that the stack gas monitor could not be relied upon as an indicator of slug jacket failures, and that it was desirable to have an early indicator of ruptures that would allow operators to discharge the failing slugs before the fans had been significantly contaminated. (Cagle, 1949; Cagle and Emlet, 1948; Cagle, 1955).

After the multiple slug ruptures of November 1947, two instruments for the detection of particulate matter were introduced into the pile exhaust system. The first was a continuous air sampler that drew an air stream from between the pile and the fans and passed sampled air through a "milk filter" and back into the exhaust system. An ionization chamber was located near the filter, with recorders placed at the instrument and at the pile control desk. This instrument performed well at the time because of the low normal particle content of the exhaust air stream. (Cagle, 1949; Cagle and Emlet, 1948).

The other monitoring technique added around late 1947 was "the probe", which was simply an oil-soaked cloth attached to a length of pipe and suspended in the air stream. The probe was periodically withdrawn and radiation readings taken at one foot with an electroscope. At the time of initial installation, the background reading was less than 5 milliroentgen per hour. (Cagle, 1949). Up until the end of November 1947, a probe reading of 10 milliroentgen per hour obtained within one hour after inserting a clean probe was a positive and infallible indicator of a ruptured slug (Cagle and Emlet, 1948). Operators would wait for the probe to return to a constant reading of at least 10 milliroentgen per hour before attempting to locate the rupture.

Once indications of slug jacket failure were received, a device called the "scanner" was used to locate the channel containing the failed slug(s). The scanner consisted of a telescoping sampler tube that could be extended through vertical holes in the pile's top shielding to measure radioactivity exiting the rear of individual fuel channels. The air samples passed through an ionization chamber, and channels having bad slugs would have unusually high activities. (Cagle, 1949).

By using the milk filter apparatus, the probe, and the scanner, and extreme care in discharging damaged fuel, 32 slug failures were detected and disposed with relatively small releases of particles to the pile stack (Cagle, 1949).

Values of probe readings were recorded in graphite reactor log books that are archived in the ORNL Records Center. Values were normally taken once per shift, with the frequency increased to hourly at times of heightened interest in possible slug ruptures. Charts of probe readings from November 16, 1947 through September 17th, 1948 were included in a 1948 report on ORNL pile slug ruptures (Cagle and Emlet, 1948).

When the severe slug rupture incident occurred on November 30, 1947, probe readings rose from the normal 4 to 5 milliroentgen per hour to 20 to 30 milliroentgen per hour. When the pile was restarted about nine days later, the probe readings increased very rapidly, making it necessary to put new cloth on the probe as often as three times per day. The slow release of particles from the graphite matrix of the pile severely limited the options for future detection of slug failures. Frequent bursts of radioactivity left the pile, even when there was no rupture. Past

monitoring methods became inadequate due to the high background of particles; probe background readings jumped up to 20 milliroentgen per hour and the milk filter apparatus became so contaminated after each filter changing that its utility was gone. The milk filter apparatus was taken out of service. The scanner gave elevated readings for almost a fourth of all channels and could no longer be relied upon. A method of visual scanning was instituted on a weekly basis that necessitated shutting down the pile and looking through each channel, toward lights suspended in the exit plenum chamber, to locate distorted slugs. (Cagle, 1949; Cagle and Emlet, 1948).

At one point during removal of ruptured slugs, the probe was removed after being in the effluent air duct for four days. It was measured with a "cutie pie" ionization chamber (through the side wall of the chamber) and read 200 milliroentgen per hour at 1.5 inches; the activity decayed to 20 milliroentgen per hour in 24 hours. (Cheka and McAlduff, 1949).

When traditional air monitoring methods failed to provide early detection of slug ruptures, two new stack monitors were developed and placed into operation. The first was a radioactive noble gas indicator that used a constantly moving charged wire that traveled down the center of a pipe carrying a flow of filtered exhaust air. Any radioactive noble gases in the air which decayed to radioactive daughter products would adhere to the wire and be pulled through a counter chamber. The instrument operated well, providing a continuous record of the relative areas of exposed uranium in the pile, but was of little value due to the relatively large amount of uranium oxide in the pile compared to the small surface of a newly ruptured slug. (Cagle, 1949; Livingston, 1948).

The second type of new monitor attempted was an active particle detector consisting of a moving paraffin-impregnated cloth tape which unwound from a roll and was drawn through a tube through the concrete shielding of the exit duct to be exposed to the hot passing air. There the paraffin softened, and particles striking it would adhere and be drawn through a counter chamber to be measured and recorded. Like the probe, frequent bursts of active particles made the paraffin tape monitor unreliable as an indicator of slug ruptures. (Cagle, 1949).

In September 1948, sampling by the Health Physics Division indicated that as much as 900 grams of particles, ranging in size from 600 micrometers to less than 1 micrometer, might be passing from the pile to the fans each day. While not all of the particles were radioactive, these results precipitated immediate action on the construction of a filter house for pile exhaust. By mid-1949, analyses of stack gases indicated that the particle content had decreased to about 230 grams per day, and the milk filter was put back into service with an improved filter and detection instruments. (Cagle, 1949).

Sampling performed over the period from September 23rd to November 7th, 1948 with an Aerotec cyclone placed in the pile exhaust duct between the pile and the fan house indicated that

the rate of discharge of particles from the pile usually averaged around 1 to 3 grams per hour, with peak rates as high as 120 grams per hour for fractional hour periods. The samples contained a great deal of concrete, and the specific activity was very low. The gross beta activity of particles leaving the pile varied from less than 0.1 millicurie per hour to over 12 millicuries per hour, while the gamma activity varied from less than  $10^5$  to over  $10^8$  counts per minute. The uranium content of the samples was around 1%. Only a few percent of the particles were highly radioactive. Cyclone sampling backed up with particulate filters performed later in 1948 demonstrated the presence of short-lived noble gas fission products in the pile gas stream. Follow-up experiments yielded noble gas release estimates as follows:

xenon-139 (41 seconds)      45 millicuries/minute

krypton-81m (9.8 seconds)    140 millicuries/minute

(Livingston, 1948)

After the pile filter house was in place, ionization chambers were in place directly in front of the F.G. #50 filter and the CWS filters (McLain, 1948c). It is not clear if these detectors were permanently mounted or inserted portable detectors.

#### Effluent Treatment History:

On August 25, 1948, a second episode of multiple stuck slugs resulted in release of uranium oxide from five slugs. The November 1947 major slug failure incident had been considered an isolated event and not likely to happen again. However, after the August 25, 1948 episode, every effort was directed to removal of particles from pile cooling air. A high-priority project to construct a pile filter house was begun.

Air started flowing through the pile filter house at 2 a.m. on November 15, 1948 (McLain, 1948c). The filter house, as described by Tench and Ramsey (1952) was a large reinforced concrete structure composed of four identical cells, each containing a roughing filter and a polishing filter. The air entered the top of the filter house, passed downward through the roughing filter, then horizontally through the polishing filter into the exit air duct. The roughing filters were American Air Filter Company deep pocket filters, with each pocket containing two layers of filter media. Each cell contained 30 frames of 5 pockets each. The filter media was one layer of A.A.F. glass wool Filterdown F.G. #25 and one layer of Filterdown #50. The polishing filters were U.S. Army Chemical Warfare Service No. 6 paper or AEC No. 1 paper. The CWS paper was primarily composed of specially screened Bolivian asbestos combined and strengthened with other fibers. The CWS filter paper was folded into 11-inch pleats and fastened into plywood frames.



Electrostatic precipitators and cyclones, which had originally been considered for use on pile exhaust air, (McLain, 1948a) were never installed.

The design efficiency of the filter house was stated to be 99.9% or better for particles greater than 0.1 micrometer in diameter. In 1949, an evaluation of the filtering efficiency of the FG material used in the filter house was performed. Two layers of FG #25 material captured only 43% of the particles in the air. The FG #25/FG #50 combination performed as well as two layers of FG #50 material, and was projected to have longer operating life. (Tench and Ramsey, 1952; Cagle, 1955; Savage, 1949). An evaluation of F.G. #25 and F.G. #50 Filterdown in series with CWS #6 paper, with a second layer of CWS #6 as a reference, indicated a collection efficiency of 99.9%, and tests with a miniature duplicate of the pile filter house (possibly the filter house placed on Building 706-D, the RaLa building) indicated an efficiency of 99.7% at a contaminated dust flow of 10 millicuries per hour. (Cagle, 1955).

After the pile filter house was operational, no further particle showers were experienced from the graphite reactor stack (Cagle, 1955).

#### Environmental Monitoring:

The X-10 Health Physics Division observed as early as 1944 that the radiation background in the area down-wind from the pile (to distances of several thousand feet) increased considerably following slug bursts, and this added contamination in such areas decreased during each subsequent rain. However, at that time, the contamination was thought to consist mostly of short-lived fission products that had escaped from pin-hole leaks that suddenly developed in the slug jackets. (Morgan, 1948).

When a problem with radioactive particle contamination from the Hanford Works chemical operations was first reported around early 1948, ORNL Health Physics surveyors began to look for similar problems at X-10. Many "specks" were found, ranging in frequency from one per 50 square feet to only a few scattered particles at 2,000 feet and in size from 90 micrometers to about 500 micrometers. Special analyses showed that the alpha activity due to plutonium ranged from 75% to 90% of the total activity in the several specks tested. Tests also indicated the presence of a considerable amount of ruthenium-106/rhodium-106 (368.2 days/29.92 seconds), and showed the maximum activities of the specks to be about one microcurie of beta activity and 0.1 microcurie of alpha emitters. (Morgan, 1948).

On July 9th, 1948, "Operation Particle" was designated as a special project to identify the sources of airborne radioactive contamination at the X-10 site and to initiate appropriate corrective actions. Health physicists were called away from other operations to survey large areas of the X-10 site by various methods. (Cottrell, 1948a; Morgan, 1948).

Monitoring techniques used in Operation Particle included (Cheka and McAlduff, 1949; Morgan, 1948):

**Sedimentation Frames** were 3-foot by 4-foot by 5-inch plywood boxes, the bottoms of which were lined with kraft paper, which were set out to collect particles falling from the air. At first, a mat of fiberglass was placed over the paper to immobilize the particles. The frames were checked from time to time with Geiger-Muëller counters, but no activity was found for about a month, on July 20th, when specks were first found. While a majority of the dust sifted through the mat, it was later found to retain a fraction of the dust, and use of the mats was discontinued. The tray liners were taken to the laboratory, where all dust was concentrated into an area that could be covered by a 14" by 17" x-ray film for autoradiography to indicate the number of radioactive particles collected. Sedimentation frames were placed at 22 locations.

**Airway "Sanitizer" vacuum cleaners** were later used to remove the collected dust from each paper liner into a removable paper filter bag, which was then sealed and autoradiographed. The Sanitizers were also used as direct air sampling equipment. They were placed on three-foot metal stands to prevent their exhaust air from stirring up dust.

**Filtrons** were air samplers that used constant displacement pumps to draw 5 cubic feet per minute of air through a cylindrical cartridge lined with Hollingsworth and Vose 9 mil filter paper. Collection efficiencies for these filters were not known, but were known to be much higher than those of the Airway filter bags.

**U.S. Public Health Service large capacity air samplers** were supplied to ORNL on loan from the USPHS. These samplers had a rated flow of 275 cubic feet per minute through 32 square-feet of filter paper. These were modified to take 10-square-foot CWS #6 filters. This paper was reported to have an efficiency of 99.97% for the most penetrating particle size. The revised filter paper cut the sample flow rate to 240 cubic feet per minute.

**Constant Air Monitors** were installed about the plant, three of which were located at outdoor locations at the time. These devices had air-flow systems and filters similar to the filtrons, but also had G-M tube radiation detectors monitoring the filter cartridges. The radiation detectors were connected to recording devices. The filters from the outdoor locations were also periodically autoradiographed, as were the filters from the other filtering devices described above.

**Electrostatic Precipitators** that drew 6.5 cubic feet per minute samples were tried, but were soon discontinued due to voltage breakdown and low efficiency and reliability.

Results of Operation Particle measurements using these techniques were summarized by Cheka and McAlduff (1949). Results of environmental monitoring during this time frame are also available in reports that were prepared by W.D. Cottrell. These reports were initially issued as Waste Monitoring Section weekly reports, and were later renamed to Area Monitoring Reports. These reports described instances of air contamination, meteorological data (such as number of inversions experienced and wind direction frequency distributions), and routine monitoring results of waterborne discharges. (Cottrell, 1948a-h).

#### 2.4.4 Processing of Clinton Pile Fuel for Plutonium Recovery

The X-10 chemical separation pilot plant used the bismuth phosphate "co-precipitation" process to recover plutonium from irradiated Clinton Pile uranium fuel. Processing in the plant began in December 1943, using uranium irradiated at the Washington University cyclotron in St. Louis. By the end of January 1944, the chemical pilot plant was processing one-third ton of Clinton Pile uranium per day. By the time operations of the chemical pilot plant ceased at X-10 in early 1945, a total of 326.4 grams of plutonium had been separated from 299 batches of irradiated uranium fuel slugs. Process improvements along the way increased the efficiency of the bismuth phosphate process for separating plutonium from fission products from about 40% to about 90%. (Johnson and Schaffer, 1992; Thompson, 1963; Jones, 1985).

Very little information has been located to date concerning the early plutonium separation operations of the chemical separations pilot plant. This is primarily due to the early occurrence and short duration of the program, which essentially was in full operation less than two years.

Waterborne effluents would have been minimal during the plutonium separation program itself due to the use of the gunite storage tanks, which were designed to hold all the waste from the pilot plant operations. Discharges occurred primarily in later years, when supernatant from these tanks were treated and released and sludges were processed in the metal recovery plant.

Airborne effluent monitoring was not routinely performed in the 1940s. However, there are records of discharge monitoring practices. Some information has been located in Metallurgical Project reports issued by Clinton Laboratories in 1943, 1944, and 1945. Some of these reports are available in ORNL Laboratory Records. Other documents are available through OSTI in Oak Ridge. An example would be M-CE-1887, a semi-monthly report of the Engineering Development Section (Leverett, 1944). This report discusses stack monitoring for the "204 stack" associated with the pilot plant, then called Building 205. Problems were being encountered with formation of bubbles in an ionization chamber used to measure iodine given off from the slug dissolver unit. Readings were difficult to obtain at that time because slugs being dissolved recently had been decayed for approximately 60 days.

It is not known at this time whether "run summaries" were prepared for the plutonium separation program like they were for RaLa, Thorex, and similar programs. A search is being conducted for these documents.

ORNL Laboratory Records has log books of individuals assigned to early Clinton Laboratories functions. These log books, along with interviews of individuals known to have been involved in early pilot plant operations, will likely be the best source of information concerning early plutonium recovery activities. A recently completed history of the Chemical Technology Division includes interviews with some early pilot plant workers, some of which have not yet been interviewed in conjunction with this project.

#### 2.4.5 Waterborne and Airborne Waste Disposal

##### Airborne Effluent Monitoring Information:

Before 1956, monitoring of airborne waste discharges from X-10 facilities was limited to non-routine grab sampling. Routine monitoring was started by the Health Physics Division in 1956, when a filter tape monitor was placed on the graphite reactor stack. A similar monitor was placed on the chemical separation pilot plant stack in 1957 and on the central off-gas system stack in 1959. The first monitor used a two-inch filter tape, through which was drawn a measured stream of stack gas. A side-window Geiger-Muëller (G-M) radiation detection tube was located adjacent to the tape to reflect the buildup of activity during the collection period. A local strip-chart recorder indicated detector response. The filter tape was changed daily by manually pulling tape through the collection block and cutting off the piece containing the sample. The sample was allowed to decay for 72 hours, after which a beta-gamma count was performed. The monitors were mainly for operational trending and incident detection; there was no nuclide identification and no attempt made to calculate stack discharge. Should the rate of buildup on the monitored filter exceed a preset limit, a local alarm sounded (Manneschmidt, 1962).

As an indicator of analytical techniques available in the early 1950s, a summary of counting equipment and techniques used at ORNL includes descriptions of alpha proportional counters, alpha pulse analyzers, beta proportional counters, alpha and beta proportional counters, end-window G-M tube counters, ion chambers, a gamma scintillation spectrometer, a gamma scintillation counter, coincidence counting equipment, and x-ray counting equipment (Brown, 1952).

The early sampling probe was a one-inch stainless steel pipe inserted across the diameter of the stack. In the case of the central stack, it was at the 50-foot level. The sample was withdrawn through perforations that were along the entire length of the pipe inside the stack (pointed in all

directions, both upstream and downstream) and carried to the monitor located at ground level (Manneschmidt, 1962).

Browder (1959) indicates that in early 1959, continuous stack monitors were in place at the isotope area (the central stack), the "hot" (chemical processing) pilot plant, and the graphite reactor. The equipment at the central stack consisted of a conventional LaPine motor blower unit which drew air from the stack gas stream through millipore® (membrane) filter. At the pilot plant stack, the sample was drawn through a membrane filter by means of a steam-jet arrangement. The sample at the graphite reactor was drawn from the duct down-stream from the filter house and uses the pressure differential across the fan to draw the sample through Hollingsworth-Vose (H.V.) 70 filter paper. Mounted in a shielded box below the H.V. 70 filter paper is a G-M tube which monitors the activity collected on the filter. The counting rate is recorded on an Esterline Angus recorder. Whether this equipment was in addition to the filter tape monitors described by Manneschmidt (1962) is not clear.

An experimental "filter-monitor" of unspecified design was put in place on the central stack in the Fall of 1959. It took its sample from the same sample line that the routine sampler used at that time, but collected samples over one-week periods instead of daily samples. The experimental apparatus was being used (by J.W. Youngblood) to determine the kinds of activity which are released from the stack and to test the efficiency of various methods of removal (Borkowski, 1960).

In the course of the 1959 ruthenium fallout incident, which was described earlier, the routine daily stack monitoring samples failed to show a marked increase in particulate releases during the periods of ruthenium release. Samples taken from the experimental sampling apparatus in place at the time indicated increased ruthenium-106 releases by approximately a factor of 1000 during the weeks ending November 2nd and November 9th, 1959, compared to levels before October 26th, 1959 (Borlowski, 1960). After that incident, a series of sampling and monitoring improvements were initiated at the principal stack (Manneschmidt, 1962; Manneschmidt, 1966):

1. The first improvements were provision of remote surveillance of the tape monitor described earlier and addition monitors.
2. The first device to be added was a monitored charcoal trap. It consisted of an aluminum cylinder which was filled with 750 grams of 14-mesh charcoal and had a central well for insertion of a Reuter Stokes type RSG-1 ion chamber radiation detector. The cylinder was attached to the sample line described earlier, and approximately 0.5 cubic foot per minute of stack gas was drawn through the charcoal. The signal from the ion chamber was amplified and recorded. A similar ion chamber was inserted in the discharge from the off-gas clean-up facility (which treats a relatively low volume of highest activity airborne wastes)

to provide continuous indication of the contribution of this source to stack releases.

3. A third improvement was acquisition of a moving tape particulate monitor. In this monitor, the sample stream passed through a 1½-inch tape which automatically moved step-wise according to a preset cycle. After exiting the sampling area, the tape was positioned under an end-window G-M tube which measured the collected radioactivity.

The monitors described to this point were intended to serve mainly as indicators of abnormal releases or operational trends. In order to better estimate release totals, "inventory" samplers were added in the early 1960s. Apparently the first to be added was a unit in the central stack that included a Gelman filter holder that contained a 2" membrane type filter (Gelman Green 7) followed by a holder containing a small charcoal cartridge holding about 3 grams of charcoal. The associated pump (Gast Model 0211) had a rated capacity of 1 cubic foot per minute, and a 2 cubic feet per minute purge-type rotameter was provided for sample flow indication. As of 1962, flow totalizer units were being considered. The filter/charcoal cartridge combinations were changed daily, analyzed with a single-channel gamma spectrometer, and alpha counted. Similar samplers were attached to a number of the larger ventilation ducts discharging into the central stack; however, they contained only the filter paper, which is beta-counted but not gamma scanned. (Manneschmidt, 1962).

In early 1962, tests were done to determine how stack sample withdrawal could best be done. An experiment was conducted in which three probes were inserted in the central stack at the 50-foot level; one in the center, one about five feet from the wall, and one midway between the other two. Step-moving tape monitors were attached to each probe. Based on the information collected, a withdrawal system evolved that consisted of a three-probe configuration. Two of the tubes were sample extraction tubes that led to monitoring devices outside the stack. The third probe contained an in-stack sampler that used a filter/charcoal cartridge. (Manneschmidt, 1962; Manneschmidt, 1966).

Moving-tape particulate monitors went through four designs by 1966:

1. The manual tape monitor described earlier.
2. The step-moving tape monitor with end-window G-M tube.
3. A 1961 moving-tape model with detector at the point of collection (G-M tube or alpha scintillation detector).

4. A 1966 design with membrane filter for minimum self-absorption and surface barrier detectors for discrimination of radon and thoron daughter products. (Thoron is a traditional name for radon-220 (55 seconds)).

(Manneschmidt, 1962; Manneschmidt, 1966)

The early aluminum-cylinder iodine monitor was replaced around 1962 with another charcoal trap design that consisted of an eight-ounce polyethylene, wide-mouth bottle half-filled with charcoal and closed with a perforated cap; attached to the cap is a plastic centrifuge tube with holes in its end. Sampled gas enters through the centrifuge tube, passes through the charcoal, and out the perforations in the cap. The jar arrangement is placed inside a cluster of G-M tubes and inside a lead shield. The G-M tubes are connected in parallel to a count-rate meter in the monitoring building. As of October 1966, this monitor was in place on several stacks, including the central ("3039") stack. (Manneschmidt, 1966).

A flow-through inert gas monitor was also constructed in the 1960s. It was similar to the jar-type iodine monitor, but had an end-window G-M tube inside instead of charcoal; incoming gas impinged on the detector window. As of 1966, this monitor had not been calibrated, but served to indicate passage of activity. (Manneschmidt, 1966).

As a "second line of defense", a "top-of-stack" monitor was installed on the ORNL central stack around 1966. A G-M tube encased in styrofoam was attached to an endless rope which passes through two pulleys mounted atop the 250-foot stack. When the detector package was raised as high as it would go, it was believed to be in position to "see" the plume coming from the stack without being in the plume or in the "shine" (direct radiation) from the stack's interior walls. A scintillation detector was also being tested for top-of-stack monitoring, to provide continuous gamma spectrometric analysis of releases (Manneschmidt, 1966).

As of 1980, radiation and flow monitoring devices were similar for all the stacks and monitored ducts. The central stack (3039) monitoring system was typical of those used on other stacks. The system used a three-probe sampler; one probe contained an in-stack filter/charcoal sampler, the second is attached to a beta-gamma particulate monitor; and the third is attached to an alpha particulate monitor and an iodine monitor, which operate in series. A sample is also directed to an inert gas monitor located at ground level. (Gilbert/Commonwealth, 1980).

Summaries of effluent data were reported in ORNL Applied Health Physics annual reports from 1961 through 1976, Industrial Safety and Applied Health Physics Annual reports from 1977 to 1982. Responsibility for effluent reporting then shifted to the Environmental and Occupational Safety Division.

Raw data for early effluent measurements have been located for only a portion of the period for which measurements were taken. Some raw data has been located for airborne releases for the period from June 23, 1963 through December 19, 1968. The data are contained on "Summary of Waste Discharges" forms, a sample of which is shown in Figure 2-10. Stack discharges in 1963 are indicated on a daily basis for the graphite reactor, chemical pilot plant, and central off-gas stacks. Values are daily release totals, in millicuries for a particulate filter and charcoal cartridge for each stack. Activity was assumed to be  $I^{131}$ , unless otherwise noted." Radionuclides specifically identified over the six-years of data located to-date include strontium-90 (28.6 years), mercury-197 (64.14 hours), cesium-137 (30.17 years), antimony-125 (2.77 years), mercury-203 (46.6 days), cesium-127 (6.2 hours), tellurium-132 (78.2 hours), iodine-132 (2.3 hours), iodine-133 (20.8 hours), and tellurium-127 (9.35 hours), -128 (stable), and -129 (69.6 minutes). As time passed, data for additional stacks were added as follows:

- Building 2026 (High Radiation Level Analytical Facility) filter and charcoal added around February 10, 1965, with 7-day totals given.
- MSRE filter and charcoal added around April 20, 1965, with 7-day totals given.
- HFIR filter and charcoal added around February 1st, 1966.

Around June 1966, chemical pilot plant stack data changed to weekly reporting; after that point, only the central stack had daily data reported. Some HFIR stack releases are attributed to the TRU facility in the comments fields (HFIR, TRU, and TURF share a stack). Recording of gross alpha measurements for the HFIR stack began around December 1967, but was not included on all forms.

Quantities of airborne iodine releases from ORNL were first reported in 1961. Noble gas releases (assigned to krypton-85 (10.72 years) and xenon-133 (5.245 days)) were first reported in 1970. Gross alpha measurements of airborne effluents were first reported in 1972. Airborne tritium release estimates were for the first time based on monitoring data, rather than calculations based on radionuclide inventory, in 1984 (Ohnesorge, 1986).

Records transmittal cards have been located in the X-10 Records Center which describe records pertaining to daily logs and chart papers from the graphite reactor. Efforts are ongoing to locate these records and evaluate their content, because observation of the graphite reactor control room in February of 1993 showed chart recorders with the following labels:

"Stack Gas Activity"  
"Exit Duct Particulate Radioactivity; Particulator Bldg 3001"  
"Stack Particulate Radioactivity; Particulator Fan House"  
"Stack Charcoal Trap Monitor; Charcoal Trap Fan House"  
"LITR Off-Gas"



# SUMMARY OF WASTE DISCHARGES

WEEK ENDING  
*June 16, 1968*

## INTERMEDIATE LEVEL WASTE VOLUME

BUILDING OR AREA SERVED	TANK NUMBER	GALLONS							
		MON.	TUES.	WED.	THURS.	FRI.	SAT.	SUN.	TOTAL
ISOTOPE	WC-1	1080	2557	2190	990	2385			9504
REACTORS	WC-19	1025			1150			800	2975
3019	W-1	3850	4600	4550	3200	4700		5000	25900
4500	WC-13				560				560
4501	WC-17				400			400	800
4500	WC-11				2300			2200	4500
3517	W-5	2320	400	-	250	2280	972		6222

## PROCESS WASTE

BUILDING OR AREA SERVED	MANHOLE NUMBER	VOLUME, M Gal							
		MON.	TUES.	WED.	THURS.	FRI.	SAT.	SUN.	TOTAL
3019	25	29	27	25	26	16	16	16	155
REACTORS AND 3036	114	101	98	92	96	99	101	101	688
3025, 3026, AND 3550	149	55	64	63	64	62	57	57	422
4500	190	264	259	274	291	270	223	223	1804
3517	209	5	5	5	4	3	3	3	28
3508, 3503	229	45	51	58	55	57	58	58	382
ISOTOPE	234	26	21	18	18	17	8	8	116
3525	235								89
SETTLING BASIN DISCHARGED (Beta Curies)		0.014	0.006	0.025	0.008	0.009	0.009	0.007	0.08

## STACK DISCHARGES

STACK NUMBER	ACTIVITY, mc							
	MON.	TUES.	WED.	THURS.	FRI.	SAT.	SUN.	TOTAL
3018 FILTER	0.4	0.4	0.4	0.4	0.4	0.3	1.3	4 <sup>Ⓢ</sup>
CHAR.	2	2	3	7	1	2	7	24 <sup>Ⓢ</sup>
3020 FILTER	LOW	LOW	LOW	LOW	LOW	LOW	LOW	X
CHAR.								
3039 FILTER	44 <sup>Ⓢ</sup>	0.4/1 <sup>Ⓢ</sup>	0.6	0.1	0.1 <sup>Ⓢ</sup>	0.1 <sup>Ⓢ</sup>	0.1 <sup>Ⓢ</sup>	1/55 <sup>Ⓢ</sup>
CHAR.	894	901	901	74	66	61	54	

\*1<sup>251</sup>, unless otherwise noted.

Remarks:

- ①  $Co^{137}$ : discharged through the cell ventilation system from FPD.
- ② ALSO TRACE AMOUNTS OF  $Co^{137}$
- ③ TRACE AMTS. OF  $Te^{132}$  AND  $I^{132}$

UCN-4748  
(3 8-68)

FIGURE 2-10  
SAMPLE "SUMMARY OF WASTE DISCHARGES" FORM

Whether these charts still exist has not been verified, however, the cards indicate that these records should be preserved because of their historical value. The time span of the existence of these recorders has not yet been documented.

Waterborne Effluent Monitoring Information:

Most early ORNL discharges to White Oak Creek were via the settling basin, with flow measured by a 90° V-notch weir in conjunction with a standard stillingwell and Stevens water level or head recorder. The waste stream was sampled by a Trebler Proportional sampler equipped with a revolving dipper so shaped that individual dips collect a volume proportional to the flow through the weir at the time of each dipping. The sampler was equipped with a timer to limit the number of dips per hour, thereby limiting the sample to a volume suitable for handling and analysis. (Browder, 1959).

As of 1944, Health Physics and the Analytical Section both took readings to determine the radioactivity in waste solutions. Health Physics took measurements each day to estimate the gamma-activity of the settling pond and of White Oak Lake. The Analytical Section measured the beta count rate of an evaporated 5 milliliter sample from the settling pond once per shift. Once a month, an analysis is made of an composite White Oak Lake sample to determine the radioactive elements present. This elemental analysis was used to determine the average beta and gamma energies of the radionuclides in the water. At the time, the external dose that would be received by the body of a person submerged in the water appeared to be more restrictive than doses from drinking the water; this was thought would remain the case while the principle nuclides in the water were columbium (now called niobium) and zirconium rather than barium or strontium. (Morgan, 1945).

Gamma readings of settling basin and White Oak Lake water were taken by three methods (Morgan, 1945):

1. The Two-Vessel Method - which involved submersion of a G-M tube in sampled water contained in cylinders of 14-inch diameter and of 7-inch diameter.
2. The Single-Vessel Method - which involved submersion of a G-M tube in a single cylinder of water.
3. The Submarine Counter Method - in which a portable submersible counter is submerged directly in White Oak Lake (background radiation from the bottom and top of the settling basin was too high for the method to work there).

After 1948, radiochemical analyses identified the species and quantities of radioactivity discharge to the Clinch River (Ohnesorge, 1986).

As of 1959, grab samples were collected daily at stream stations on Melton Branch and White Oak Creek. These samples were used primarily to determine whether the activity detected is from the Laboratory proper or from the Melton Valley area. At White Oak Dam, located downstream from the confluence of White Oak Creek and Melton Branch, continuous samples were taken for daily analysis. Daily samples taken from the settling basin and from White Oak Creek were evaluated for gamma activity to determine submersion exposure, counted for gross beta radioactivity, composited for weekly analysis of plutonium content, and composited for monthly radiochemical analysis of specific long-lived fission products. The long-lived radionuclides analyzed for by the Analytical Chemistry Division were radioisotopes of strontium, cerium, ruthenium, iodine, zirconium, trivalent rare earths (TRE), niobium, barium, and cobalt. (Browder, 1959). It is not known at this time why cesium-137 was not listed as an analyte by Browder (1959).

One of the greatest uncertainties in waterborne effluent measurements has been in determining flow rates, especially at periods of high flow. In 1984, weirs were completed at White Oak Creek, Melton Branch, and White Oak Lake to provide more accurate flow measurements over a wide range of flows. (Ohnesorge, 1986).

Raw data for early effluent measurements have been located for only a portion of the period for which measurements were taken. Some raw data has been located for waterborne releases from the settling basin for the period from June 23, 1963 through December 19, 1968. While the settling basin has not been the final release point off-site, these data might be useful for trending or source term evaluation. The data are contained on "Summary of Waste Discharges" forms, a sample of which is shown in Figure 2-10. The forms present daily values of settling basin discharges, in "beta curies."

Records transmittal cards have been located in the X-10 Records Center which describe records pertaining to early White Oak Dam flow and radioactivity measurements. The cards indicate that these records were returned to the originating division. Efforts are ongoing to locate these records and evaluate their content.

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### 3.0 THE K-25 GASEOUS DIFFUSION SITE

By late 1942, the Military Policy Committee, a committee established under the atomic project to formulate policies on research and development, construction and production, and strategic and tactical matters, had authorized the development of four technically and theoretically different uranium enrichment processes. These processes - electromagnetic, gaseous diffusion, liquid thermal diffusion, and gas centrifuge - were considered potential methods for producing sufficient enriched uranium of a quality to be militarily useful in World War II (Jones, 1985). After studying the options, the Military Policy Committee decided to give priority to full-scale development of the gaseous diffusion process (Jones, 1985).

The gaseous diffusion process works by first, chemically combining uranium atoms with fluorine atoms to produce molecules of uranium hexafluoride ( $UF_6$ ).  $UF_6$  is a gas at certain temperatures and pressure. Each molecule of the gas will have an energy related to the temperature of the gas. The energy associated with each molecule is expressed by its velocity. For equal energies, lighter gas molecules have higher velocities than the heavier gas molecules. Thus in a container, the lighter gas molecules, which move faster, will strike the walls of the container more frequently than the heavier molecules. If the walls of the container are made of a porous material, called a barrier, the lighter molecules will have a greater probability of passing through one of the pores than a heavier molecule because they strike the walls more frequently.

Natural uranium, uranium found in the earth's crust, consists of 99.3% uranium-238 (U-238) and 0.7% uranium-235 (U-235). A  $UF_6$  gas made from natural uranium will have molecules of different masses; hence, different velocities. At each barrier, the relative percentage of U-235 molecules that pass through the barrier will slightly exceed the relative percentage of U-238 molecules. However small this difference, the percent of U-235 in the mixture of molecules will increase beyond each barrier. After many barriers, the percent of U-235 molecules in the gas will become significantly higher than in the original gas. The goal of the gaseous diffusion process was to obtain uranium enriched in U-235.

As the enriched uranium was purged at one end of the process, depleted uranium or tails, consisting mostly of U-238, was removed at the other end.

It is important to note that natural uranium also consists of 0.006% U-234. Since the U-234 is lighter than both U-235 and U-238, it is enriched at the same time U-235 is enriched. This point is significant when analyzing source terms and was considered in the Task 3/4 calculations.

The Military Policy Committee selected the M.W. Kellogg Company to design and engineer the production of a plant. The plant was designated K-25. K for the Kellex Corporation (a subsidiary of M.W. Kellogg) and 25 for the code name of U-235.

In May 1943, the J.A. Jones Construction Company signed contracts to build the K-25 powerhouse, cascade building, and administration area. The cascade building would be required to hold thousands of diffusion stages, the largest of them of 1,000 gallon capacity. The building design was monumental—four stories high, almost a half of a mile long in the shape of a U, a fifth of a mile wide, and two million square feet in area.

In early 1943, with the construction of the K-25 site about to begin, several theoretical and practical problems in the gaseous diffusion plant design had not yet been solved. The worst problem was that a suitable barrier material had not yet been developed. In February 1943, because of the continuing problems with the barrier development, General Groves, Officer in Charge for the Manhattan Project, decided to limit the enrichment potential of the K-25 plant to 36.6 percent U-235 and use the enriched material to feed the Beta calutron electromagnetic plant at Y-12 (Rhodes, 1986; Jones, 1985).

After completion of the cascade design (for 36.6 percent enrichment), Kellex researchers and engineers continued work on developing diffusion equipment that could achieve a higher product enrichment. When data showed a greater product output could be achieved by increasing the amount of uranium of a lower enrichment for feeding into the electromagnetic plant, Groves directed Kellex to design and engineer a 540-stage side-feed unit, K-27, in which the tails (material depleted in U-235) from the main K-25 cascade could be combined with natural uranium to produce a slightly enriched product. By feeding the K-27 output into the higher stages of K-25, plant designers estimated the total production of U-235 could be increased by 35 to 60 percent.

The first diffusion stages in the K-25 building became operational in January 1945, and the first enriched uranium was shipped from K-25 to Y-12 on March 19, 1945 (ORGDP, 1985b). The K-27 building became fully operational in December of 1945. Three additional cascade buildings, K-29, K-31, and K-33 were operational by 1954. These additional buildings brought the total number of stages to 5,100. This allowed for the enrichment of normal uranium at an assay of 0.711% U-235 to an enrichment level of about 93% (Jones, 1985). When requirements for military applications were satisfied in 1964, the top of the cascade, and buildings K-25 and K-27 were shut down. This reduced the separating stages to 1440, which limited the enrichment level of the plant to under 10%. This provided the full range of enrichment assay to supply civilian nuclear power reactors (ORGDP, 1985a). In 1985, the remaining gaseous diffusion process buildings were placed on standby and then shut down.

### 3.1 SUMMARY OF HISTORICAL ACTIVITIES

In addition to the gaseous diffusion process, a variety of support activities and research and development activities have been a part of the operations at the K-25 site. A map of the K-25 site is provided in Figure 3-1. This section describes the gaseous diffusion process, the support activities associated with the gaseous diffusion process, the laboratories, the historical waste disposal methods, the research and development associated with the development of alternative uranium enrichment methods, and the S-50 Liquid Thermal Diffusion Plant. The facilities discussed in this section are shown in Figure 3-1 and Table 3-1 provides a summary of major activities at the site. Each section provides a brief account of the monitoring data availability for a specific process. A more detailed evaluation of the availability of airborne and waterborne effluent data is provided in Section 3.4.

#### 3.1.1 Gaseous Diffusion Process, Bldgs. K-25, K-27, K-29, K-31, and K-33

As described in Section 3.0, the gaseous diffusion process relies on the theory that if uranium gas is pumped through a porous barrier, the lighter U-235 isotope will pass through the barrier more rapidly than the heavier U-238 isotope. Figure 3-2 illustrates a single stage of this basic concept. The uranium, which exists as gaseous uranium hexafluoride ( $UF_6$ ), is forced to flow through the inside of a tubular porous membrane (barrier). The pressures are controlled so that about half the gas diffuses through the barrier and is subsequently introduced to the next higher stage, while the remaining undiffused portion flows to the next lower stage. The diffused stream is slightly enriched in U-235, and the undiffused stream is depleted in U-235 to the same degree. Figure 3-3 depicts how the individual stages are connected to accomplish the desired enrichment.

This figure also indicates the basic process equipment components. Axial flow compressors driven by electric motors compress the  $UF_6$  to maintain interstage flow (ORGDP, 1985; USDOE, 1979).

A process gas cooler, which used a liquid fluorocarbon, was provided for each stage because gas compression generates heat that must be removed. The diffuser, or converter, was a large cylindrical vessel that contains the barrier material. The entire process was an enclosed system which operated below atmospheric pressures (below 14.7 pounds per square inch) (ORGDP, 1985a; USDOE, 1979).

A number of stages were connected together to form cells. The diffusion cascade was made up of a number of cells. To achieve any appreciable separation of the uranium isotopes, a large number of series-connected cells were required because the degree of isotopic separation accomplished from stage to stage was only about 0.2% per stage (ORGDP, 1985a). Over the forty years of operation, the K-25 plant processed varying U-235 enrichments. Until the shutdown of the K-25 building in 1964, the plant produced enrichments as high as 93%, after the shutdown U-235 enrichments were below 10%.

TABLE 3-1

HISTORICAL SUMMARY OF THE MAJOR ACTIVITIES AT THE K-25 SITE

PROGRAM	DATES
S-50 Liquid Thermal Diffusion	1944-1945
Gaseous Diffusion up to 93% U-235 enrichment	1945-1964
Gaseous Diffusion less than 10% U-235 enrichment	1964-1984
Gas Centrifuge Development	1960-1988
Atomic Vapor Laser Isotope Separation	1988-1992
Toxic Substance Incinerator, K-1435	1989-present

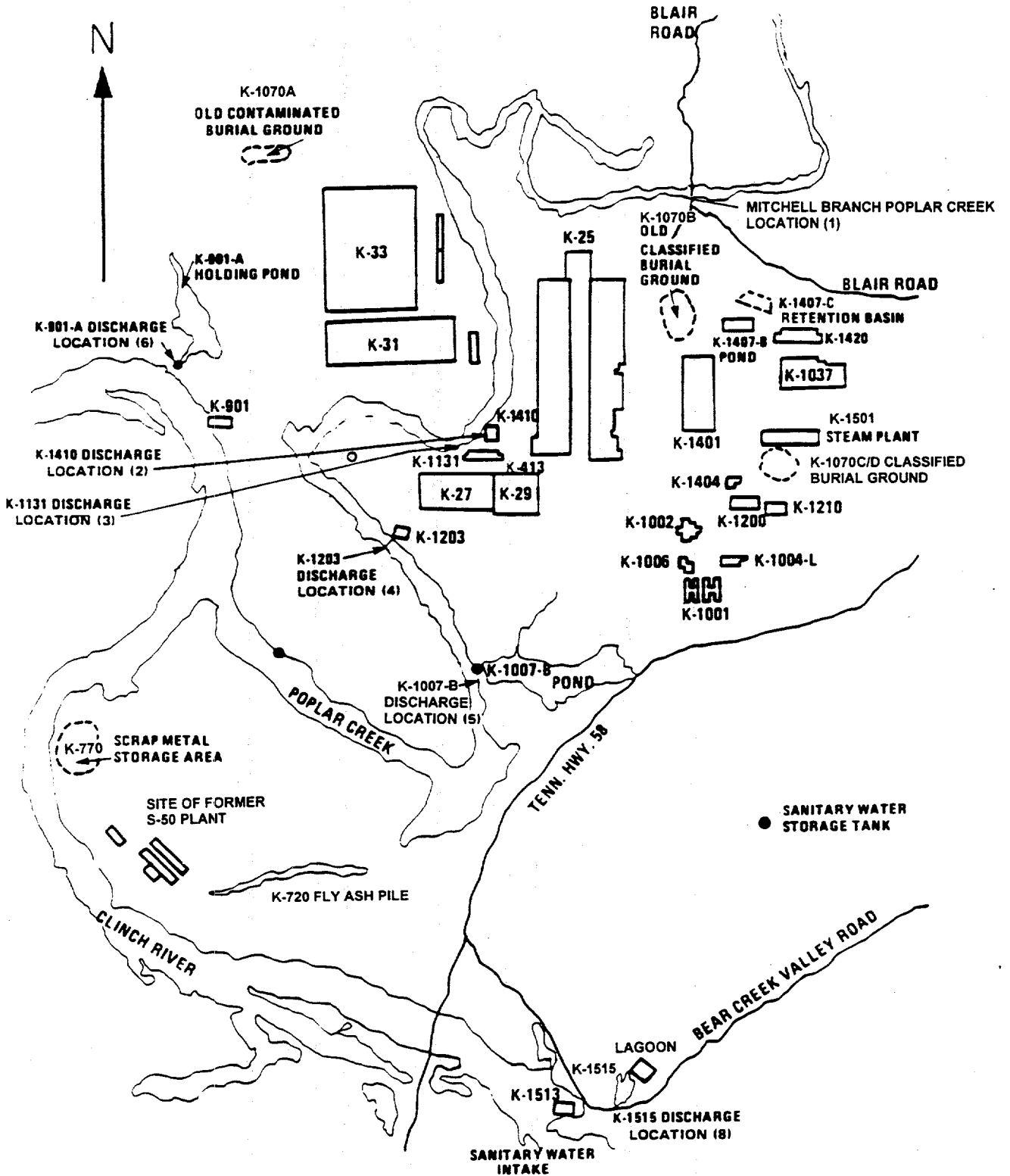
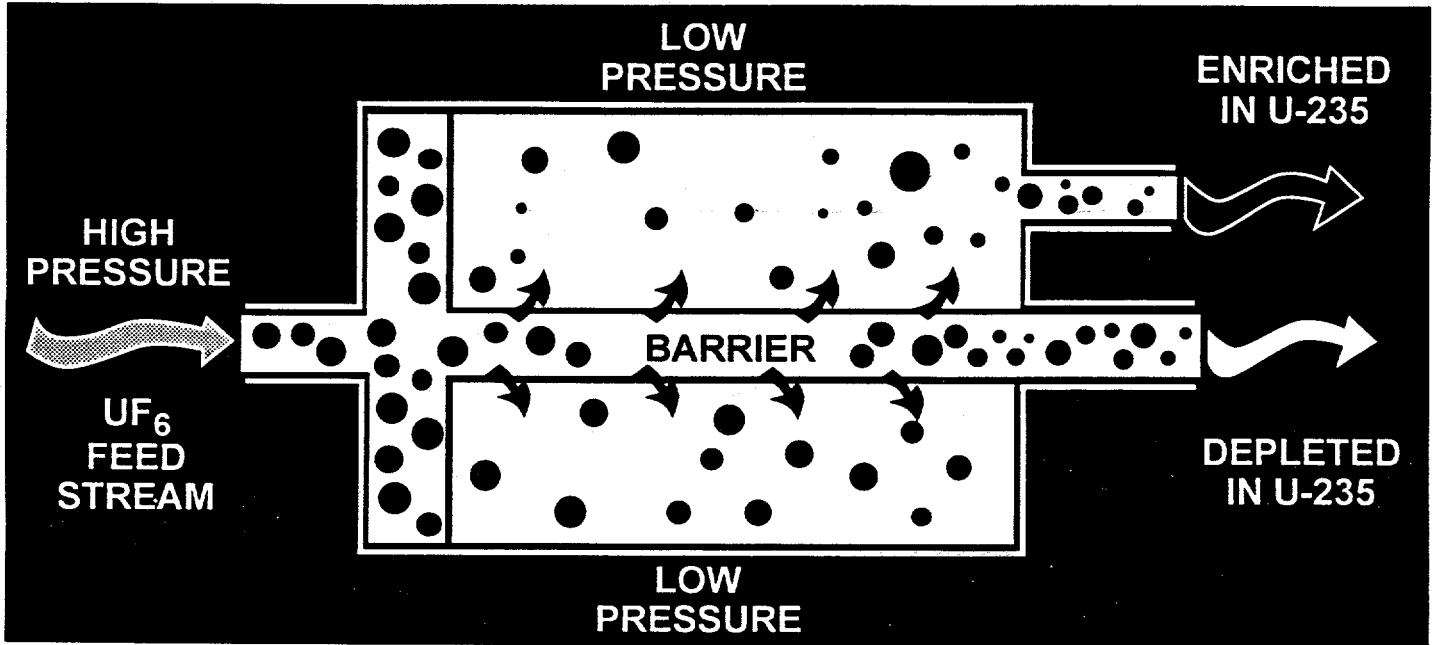


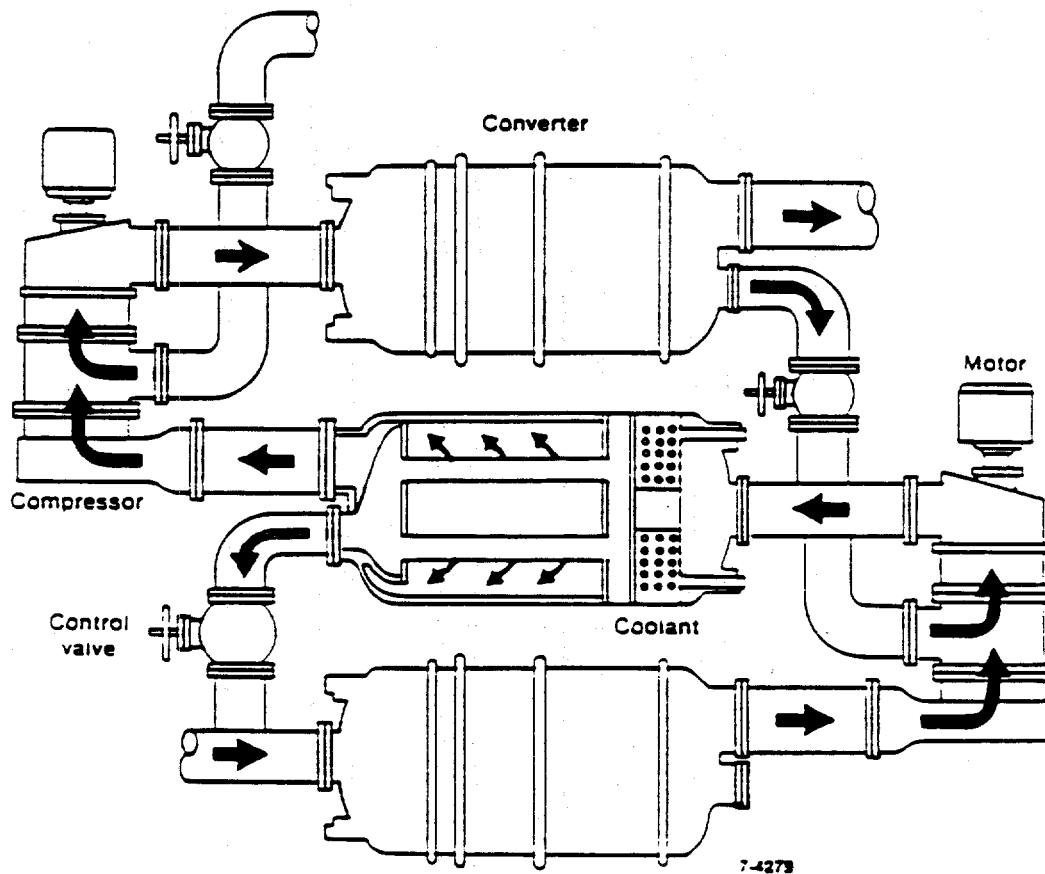
FIGURE 3-1  
 OAK RIDGE GASEOUS  
 DIFFUSION PLANT (ORGDP)  
 SITE MAP.



SOURCE: USDOE, 1979

FIGURE 3-2  
GASEOUS DIFFUSION  
STAGE





SOURCE: USDOE, 1979

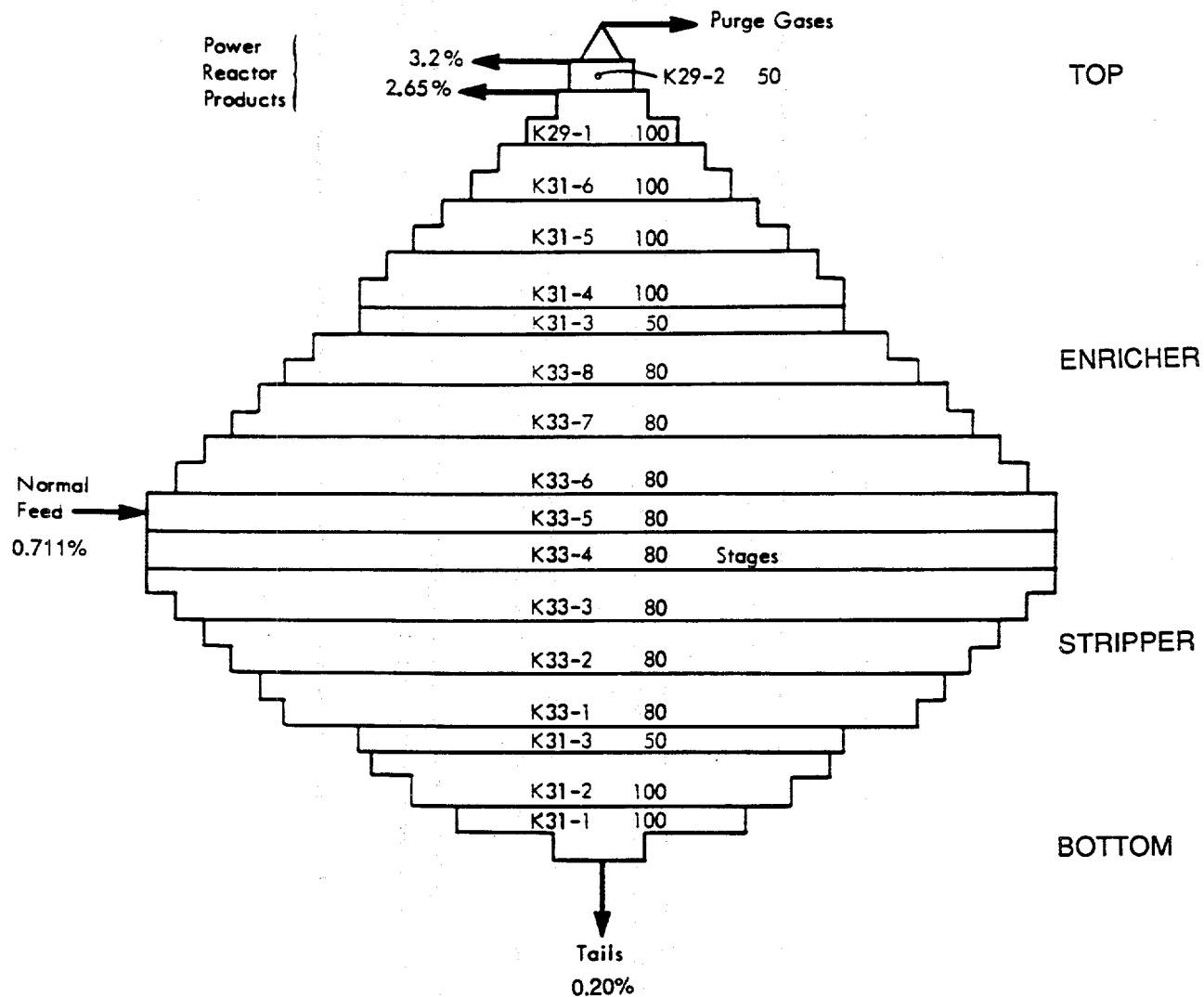
FIGURE 3-3  
DIFFUSION STAGE  
ARRANGEMENT

Figure 3-4 depicts a simplified flow taper for the gaseous diffusion cascade. Gaseous uranium hexafluoride was fed into an intermediate point in the cascade. Product material enriched in the U-235 isotope was withdrawn through one end of the cascade (the top); tails material depleted in the U-235 isotope was withdrawn from the other end (the bottom). The portion of the cascade between the feed point and the product point was called the enriching section; whereas the portion between the feed point and the tails point was called the stripper section. The taper is a reflection of the fact that the stages in the middle of a cascade must have higher flow rates than those near the end of the cascade (ORGDP, 1985a).

Because the diffusion cascade operated below atmospheric pressure light gases accumulated at the top of the cascade. If these light gases were not vented to the atmosphere they would accumulate at the top of the cascade and block the flow of the enriched uranium hexafluoride. In order to avoid accumulation of light gases, purge facilities are operated just above the top of the product withdrawal point. These light gases come from several sources (USDOE, 1979):

- Nitrogen - Mainly from the in-leakage occurring at every compressor shaft seal
- Hydrogen Fluoride - From reaction of in-leaking moist air with the uranium hexafluoride
- Oxygen, Argon - From in-leaking air
- Chlorine Fluorides - Used to condition and dry metal surfaces
- Fluorine - Used to condition metal surfaces
- Coolant Vapor - From leaks in the process gas cooler coolant system

The facility used for venting of light gases was called the purge cascade. The purge cascade consisted of equipment similar to that used in the rest of the cascade, but it was designed to handle the lighter gases. Since the coolant and chlorine fluoride gases are heavier than nitrogen and air but lighter than uranium hexafluoride, they were withdrawn through a parallel or side purge. The effluents from both systems were passed through chemical traps to reduce residual levels of uranium and fluorine compounds before discharge to the atmosphere. The magnesium fluoride traps located within the system removed technetium-99, a fission product which entered the K-25 diffusion cascade when production reactor returns material was processed (Section 3.1.7) (ORGDP, 1985a; USDOE, 1979). When the irradiated fuel was reprocessed, most of the fission and activation products were separated from the uranium; however, a small amount of some products remain with the uranium. One of these elements is technetium. When the uranium is converted to  $UF_6$ , the technetium is also converted. When the technetium enters the



SOURCE: ORGDP, 1985

FIGURE 3-4  
ORGDP CASCADE  
FLOW TAPER

gaseous diffusion plant it readily passes through the barriers because of its smaller mass and higher velocity and collects in the purge end of the system (Cloutier, R.J., 1993).

Over the 40 years of operation, the purge cascade was located in several different locations. In 1964, when the K-25 and K-27 units were shut down, K-311-1 (in the K-25 building) was converted from a side purge unit to a top purge unit, and K-310-3 (in the K-25 building) was converted to a side purge cascade for the removal of the intermediate-molecular-weight gases. Then in 1976 new purge facilities were provided by the refurbishment of K-402-9 (in the K-27 building) to a purge unit and later K-402-8 (in the K-27 building) to a companion side purge. A wet potassium hydroxide scrubber was added for final removal of uranium and fluorine compounds before discharge to the atmosphere (USDOE, 1979).

Because of historical uranium accounting requirements at K-25, uranium release data for the purge cascade are fairly extensive. Effluent data for the purge cascade are available from 1946 until shutdown in 1985. Data for other radionuclides that entered the cascade when production reactor return material was processed are limited.

### 3.1.2 Gas Centrifuge Development Facilities, K-1200 Area

From 1960 to the 1980s, the gas centrifuge program at the K-25 site pursued the development of an alternative technology for the enrichment of uranium. Much like the diffusion process, the centrifuge process handled uranium as gaseous uranium hexafluoride, and through the cascading of a few centrifuges, the desired enrichment level could be attained.

In the gas centrifuges, uranium hexafluoride was fed into a rotor which rotated at high speed inside an evacuated casing. The gas accelerated to approximately the speed of the rotor. Centrifugal force caused the heavier U-238 molecules to move closer to the wall of the rotor, producing partial separation of the U-235 and U-238 isotopes. Feed is introduced near the middle of the rotor, and enriched and depleted streams are removed near the ends. Similar to the gaseous diffusion process, the desired enrichment is not obtained in a single stage; several centrifuge machines must be connected in a series known as a cascade. The most significant difference between the gaseous diffusion and gas centrifuge processes was power usage. A centrifuge plant required only 4% of the power needed for a diffusion plant (USDOE, 1977).

The primary efforts of the K-25 program were aimed at machine development, reliability, productivity, and operability. To achieve the desired results, several facilities were constructed for developing machine production techniques, long-term reliability, and equipment cascading. The facilities included:

- The Equipment Test Facility which began operation in 1971 for examining the reliability of four types of high-capacity centrifuges (USDOE, 1977),
- The Component Preparation Laboratory which began operation in 1974 and aided in the development and demonstration of techniques for manufacturing centrifuges (USDOE, 1977),
- The Component Test Facility which began initial operation in 1975 and was used to test the reliability and operability of substantial numbers of centrifuges. This facility also served as a pilot plant cascade (USDOE, 1977),
- The Advanced Machine Development Laboratories which were used to improve and test centrifuges (USDOE, 1977),
- The Advanced Equipment Test Facility which was used to test the reliability of production centrifuges (USDOE, 1977), and
- The Centrifuge Plant Demonstration Facility which was used to test centrifuges and other cascade equipment and systems (USDOE, 1977).

The gas centrifuge operations were shut down in 1985.

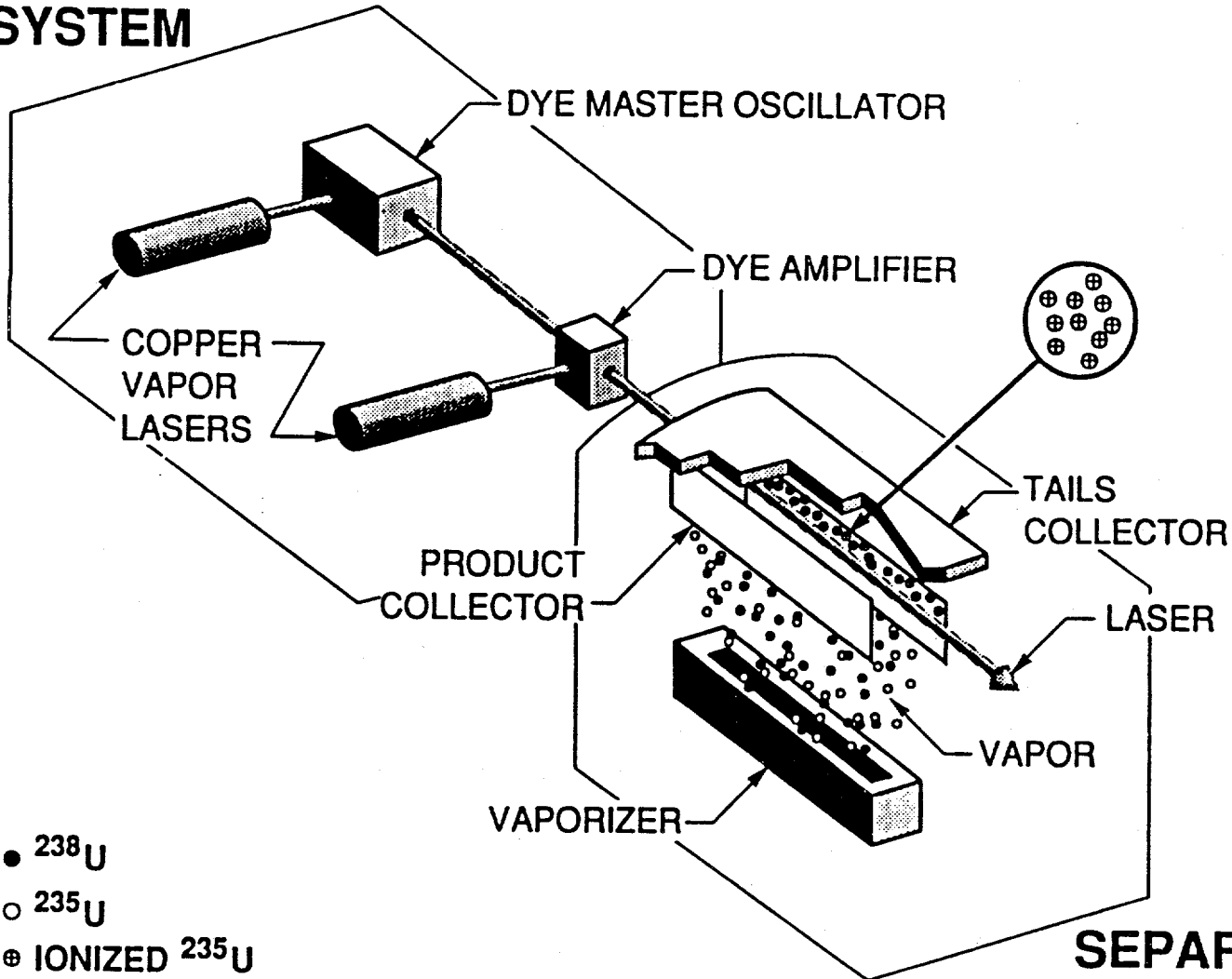
### 3.1.3 Atomic Vapor Laser Isotope Separation

The Atomic Vapor Laser Isotope Separation (AVLIS) process separates the atoms of U-235 from U-238 by electrostatic extraction of laser produced U-235 ions. Figure 3-5 is a simplified example of the AVLIS separation technique. In the first step, metallic uranium is melted and vaporized. The vapor is then illuminated by precisely tuned laser lights that are selectively absorbed by U-235 atoms. The absorption of this energy causes individual U-235 atoms to emit an electron.

The loss of an electron from the U-235 atom creates a charged U-235 ion. The U-238 atoms, which are unaffected by the laser beams, pass through the product collector to condense on the tails collector. The enriched uranium liquid metal condensate flows out of the separator to the cast and is stored in solid metallic form for eventual conversion to uranium oxide for reactor fuel rods (Saylor et al., 1990).

AVLIS activities at the K-25 site consisted of research and development of the chemical processing of uranium to produce an AVLIS feed material and the chemical processing to convert the AVLIS metal product to  $UO_3$ . These processes are known as feed and product conversion, respectively (USDOE, 1991).

# LASER SYSTEM



- $^{238}\text{U}$
- $^{235}\text{U}$
- ⊕ IONIZED  $^{235}\text{U}$

# SEPARATOR SYSTEM

SOURCE: USDOE, 1991

FIGURE 3-5  
THE ATOMIC VAPOR LASER  
ISOTOPE SEPARATION  
(AVLIS) PROCESS

It is difficult to characterize the effluents and wastes generated from research and development. However, based upon the description of the feed and product conversion process in the Environmental Assessment for the Demonstration of Uranium-Atomic Vapor Laser Isotope Separation (U-AVLIS) the likely effluents would have been off-gases (water vapor, carbon monoxide, and carbon dioxide), nitrogen oxides, particulates uranium, uranium trioxide and uranium tetrachloride, nitric acid, ammonia, chlorine gas, magnesium chloride, potassium chloride, potassium hydroxide, and paints and solvents from the maintenance operations (USDOE, 1991).

### 3.1.4 S-50, Liquid Thermal Diffusion Plant

In June 1944, it became apparent that the development problems with the K-25 barrier might stall the production of enough enriched uranium for an atomic weapon. The Military Policy Committee decided to pursue the liquid thermal diffusion uranium enrichment process. The goal of the project would be to produce uranium with concentrations of U-235 ranging from 0.9 to 3 percent. This slightly enriched product could then be used as feed for the electromagnetic process at Y-12 (Jones, 1985). This enrichment would increase the efficiency of the electromagnetic process by about 20 percent (Jones, 1985).

Thermal diffusion relied on the tendency of lighter isotopes (for example uranium-234 and uranium-235) to diffuse toward a hotter region while heavier isotopes (such as uranium-238) diffused toward a colder region. The mechanism for driving such diffusion was a hot pipe inside a cold pipe with liquid uranium hexafluoride flowing between the two pipe walls. Depending on the difference in temperature and the spacing between the two pipes more or less diffusion would occur. At the same time the heating and cooling of the uranium hexafluoride would start a convection current flowing up the hot pipe wall and down the cold, that would bring the U-235-enriched fluid to the top of the column where it could be tapped off. To increase enrichment, a number of columns could be connected in series to make a cascade (Rhodes, 1986).

Within days of the Military Policy Committees decision to pursue liquid thermal diffusion, the H.K. Ferguson Corporation had been contracted to build a 2,100 column thermal-diffusion plant beside the K-25 power plant on the Clinch River in ninety days or less (later Ferguson formed the Fercleve subsidiary to run the plant). Ferguson built a 500-foot barn of black metal siding and began operating the first rack of columns in sixty-nine days, by September 16, 1944 (Rhodes, 1986).

The S-50 plant consisted of 2,142 columns, each 48 feet in height distributed in twenty-one racks. Each of the columns had three concentric tubes, comprised of a 1 1/4-inch nickel pipe inside; a slightly larger copper pipe in the middle; and a 4-inch galvanized iron jacket on the outside. In the one one-hundredth of an inch annular space between the outer wall of the nickel

pipe and inner wall of the copper pipe the diffusion process would occur. Steam, under pressure of 100 pounds per square inch and at a temperature of 545 degrees Fahrenheit, circulated downward through the nickel pipe while water at 155 degrees Fahrenheit circulated upward through the iron jacket; simultaneously, uranium hexafluoride would flow into the base of each column. At the top of each column was a system of freezing coils, this allowed plant employees to draw off or milk small amounts of the enriched product at frequent intervals (Jones, 1985).

In the summer of 1944, when the crews began start-up operations, large amounts of steam leaked from the rack. Under ordinary conditions, such defective equipment would have resulted in immediate shutdown, but the time constraints of the project forced Fercleve to proceed with the start-up. In the confined spaces of the diffusion columns, high-pressure steam and uranium hexafluoride, which expanded 35 percent in passing from a solid to a liquid, created highly explosive forces. In October of 1944, only 10.5 pounds of uranium containing 0.852 percent U-235 had been produced. While production increased to 171.8 pounds in November, it fell back to 20 pounds in December, when steam leaks forced numerous shutdowns. By February, production reached a total of 3,158 pounds. In the spring and early summer of 1945, plant output went up rapidly, reaching a peak of 12,730 pounds in June. At first the slightly enriched uranium was sent to the electromagnetic plant. In April 1945, S-50 began sending the product to K-25. In August of 1945, when K-25 became operational, the S-50 plant was no longer needed. On September 9, 1945 the S-50 plant ceased all operations (Jones, 1985).

Very little information regarding the tear down of the original process building is available; however the fate of two support buildings (K-722 and K-725) has been documented. After the S-50 project was discontinued in 1945, the S-50 site became the research and development area for the Nuclear Energy for Propulsion of Aircraft (NEPA) Project (Goddard, 1991). From 1946 to 1951, the Fairchild Engine and Aircraft Corporation used the site for NEPA research. From 1951 to 1952, General Electric took over all operations at the site. After General Electric left, Union Carbide Corporation took over all operations in the S-50 area, which was renamed the K-700 area (ORGDP, 1988). Figure 3-1 roughly depicts the K-700 area as it appears today. Currently, some of the facilities are under RCRA and CERCLA investigations. These are described below.

#### K-722 Property Sales Building

In the mid 1940s this building was part of the support facilities for the S-50 thermal diffusion plant. Approximately 90% of the building is a warehouse which is used to conduct public sales of excess government property from X-10, Y-12, DOE, Oak Ridge Associated Universities and K-25. The north end of the building houses the property sales office. During a radiological survey of the building in 1952, a few radioactively contaminated (uranium) areas were discovered. Most of these areas were decontaminated at the time of the discovery; however,



decontamination of a few areas was not possible. In order to remove the contamination, the concrete floor was chipped away and removed. After chiseling about 5 inches through the floor, another floor, or subfloor was encountered. The subfloor appeared to be the source of radioactivity (Goddard et al., 1991).

### K-725 Beryllium Building

This building was originally part of the support facilities for the S-50 thermal diffusion plant. Early in the building's operations, a second concrete pad was placed over the original floor because of high alpha counts from depleted uranium handled in the building. After the S-50 project was discontinued in 1945, the building was used from 1946 to 1952 for the NEPA Project. The building is contaminated with beryllium, uranium, and mercury (Goddard et al., 1991).

#### **3.1.5 K-1435 TSCA Incinerator**

The incineration facility, K-1435 includes structures, systems, and components for receiving, sorting, storing, preparing, and burning PCB materials, uranium-contaminated PCBs, and other hazardous and nonhazardous wastes in a rotary kiln incinerator (Joyner, 1991). Preoperational testing of the K-1435 incinerator began in 1988 and continued intermittently until March of 1990 when all regulatory permitting requirements were fulfilled. The incinerator began to burn waste in May 1990 (MMES, 1991). The nuclides incinerated in the K-1435 incinerator include: uranium, technetium-99, cesium-137, neptunium-237, protactinium-234m, plutonium-238, plutonium-239, thorium-228, thorium-230, thorium-232, and thorium-234 (MMES, 1991). The radionuclides, carbon monoxide and carbon dioxide airborne releases from this facility have been continuously monitored.

#### **3.1.6 Processing of Recycled Uranium**

Initially, all uranium fed to the gaseous diffusion process consisted of natural uranium. Later, methods were developed to extract the uranium from spent nuclear reactor fuels. This extracted uranium was shipped primarily from Savannah River and Hanford to K-25, where it was converted to uranium hexafluoride in the feed plant. This material was then fed to the gaseous diffusion process. The processing of spent nuclear reactor material was intermittent, and three distinct campaigns were noted: 1952 - 1964; 1969 - 1974; and 1976 - 1977 (Egli et al., 1985).

The uranium hexafluoride from spent reactor fuel feed contained traces of fission products and transuranics. The fission products and transuranics identified in this feed to the gaseous diffusion process included technetium-99, neptunium-237, and plutonium-239. These fission products and transuranics were present in the K-25 airborne and waterborne effluents as a result of being introduced to the gaseous diffusion process (Lay and Rogers, 1986).

### 3.1.7 Work for ORNL

Between the years 1976 and 1980, research and pilot plant operations were conducted at K-25 for ORNL. These operations simulated the removal of krypton-85 from certain gaseous waste streams for nuclear fuel reprocessing operations. Over the 5-year time period that the experiment was conducted, approximately 110 curies of krypton-85 were released to the atmosphere (Lay and Rogers, 1986).

### 3.1.8 Support Facilities for the Gaseous Diffusion Process

The gaseous diffusion process requires several different types of support facilities. The functions of these operations range from providing direct support to the diffusion process to research and development to increase the efficiency of the plant. The facilities are listed below.

1. Enrichment Technology Facilities
2. Analytical laboratories, K-1004
4. Fluorine production plant, including hydrogen fluoride (HF) storage facilities
5. Barrier Plant
6. Feed Plant and Tails Withdrawal
7. Facilities for:
  - a. receiving, shipping, and sampling of  $UF_6$
  - b. decontamination of equipment and subsequent recovery of uranium
  - c. cleaning of metals used throughout the diffusion process
  - d. plating of metal parts
  - e. rebuilding of motors
8. Recirculating process cooling water system
9. Power Plant

A brief description for some of the facilities is provided in this section. This list is not intended to be inclusive of all support facilities. However, it is an attempt to provide a focus for future dose-reconstruction efforts of the K-25 site.

### K-1004L Enrichment Technology Facilities

The Enrichment Technology Facilities contained equipment and facilities for the development and testing of cascade barrier material. Three different groups - Barrier Preparation, Barrier Science, and the Cascade Pilot Plant - all conducted research in this facility (ORGDP, 1985a). The primary duty of the Barrier Preparation group was to flow test experimental barrier material and hardware. The wastes generated by this group included nitrogen, hydrogen, helium, and argon.

The primary duty of the Barrier Science group was to laboratory test equipment for determining gas-flow properties of experimental barrier material. The waste streams generated by this group included fluorine, chlorine trifluoride, nitrogen and hydrogen fluoride for conditioning and drying of metals (ORGDP, 1985a).

The Cascade Pilot Plant was a small multistage cascade that was used to determine the separative quality of barrier material using uranium hexafluoride under controlled conditions (ORGDP, 1985a). The maximum quantity of uranium hexafluoride contained in the system at any one time was about 25 pounds (USDOE, 1979). Airborne releases from this operation consisted primarily of gaseous fluorides which were passed through alumina traps prior to release to the atmosphere (USDOE, 1979). Information reviewed to date has not indicated when the alumina traps were placed on the building.

### K-1037 Barrier Manufacturing Facility

The barrier manufacturing facilities included the main production facility, K-1037 and the K-1401 Converter Retubing and Assembly Area. These facilities were used to manufacture barrier material and were placed on standby in 1981. The facility was unique in that it was the only location in the United States where the barrier material was manufactured (USDOE, 1979). These facilities did not utilize radioactive materials (ORGDP, 1985a). With the exception of nickel, the materials used in the manufacture of the barrier are classified. A short summary of the materials used in the barrier has been prepared by ChemRisk and is available for review by Q-cleared personnel. Further evaluation of materials used in the barrier manufacturing process can be performed. However, any evaluations would be required to remain classified. The work could be evaluated by the proper Q-cleared individuals.

### K-1420 and K-1303 Decontamination and Uranium Recovery Facilities

Equipment used in the gaseous diffusion process was subject to gradual deposition of uranium-bearing compounds (USDOE, 1979). When equipment was removed from the system, it was decontaminated to meet radiation standards before maintenance functions were performed (USDOE, 1979). At the K-25 site, decontamination facilities were located in Buildings 1303 (1945-1953) and K-1420 (1953 -1985).

The primary method for cleaning the process equipment included a form of mechanical removal in combination with cleaning solutions of water, steam, weak nitric acid, or sodium carbonate (USDOE, 1979). Large parts were decontaminated in three spray booths. The first spray booth system consisted of an acid wash booth which used a 1% to 3% nitric acid solution for cleaning, and the second and third spray booths consisted of rinse water. Aluminum nitrate was added to the nitric acid solution to chemically react with the fluorides for corrosion protection of the stainless steel piping. The cleaning solutions from the first two spray booths were collected in tanks and transferred to the uranium recovery area of K-1420. After being sampled for uranium content, the water used in the second rinse booth was normally discharged to the K-1407B holding pond. The waste cleaning solutions contained uranium in addition to other metals such as aluminum, copper, iron, magnesium, manganese, and nickel (USDOE, 1979).

The acid solution from the decontamination operations contained uranium and were sent to the uranium recovery facilities. The uranium recovery process consisted of a solvent extraction purification step followed by concentration, drying, and calcining which produced uranium oxide (as  $U_3O_8$  and  $UO_3$ ). This oxide could then be reintroduced into the uranium fuel cycle. An indication of the recovery capabilities was provided by the 1979 Environmental Assessment which notes that in 1979, 2,400 kg of recovered uranium was shipped to off-site uranium hexafluoride production facilities (USDOE, 1979).

The primary liquid effluents from the recovery facility emanated from the solvent extraction step. The waste stream from this purification operation was highly concentrated with nitrates and contained uranium and technetium compounds. These effluents were sampled for uranium content and then released to the K-1407B holding pond.

For most of the building history the airborne effluents from the decontamination and recovery facilities were not monitored. The primary constituents of the airborne releases were uranium-bearing particulates and oxides of nitrogen.

### K-1420 Cylinder Cleaning Facility, K-1420

The cylinder cleaning facility was used to remove residual quantities of non-volatile uranium compounds that remained in uranium hexafluoride storage cylinders after they were emptied by heating in an autoclave, thus transforming the uranium hexafluoride to vapor so that it could be fed to the gaseous diffusion cascade. The cylinder cleaning facility featured a stainless steel floor with drain connections to the K-1407 B holding pond. Normal cleaning solutions included water and commercial acidic cleaning agents. The wash solution was siphoned from the cylinder and the uranium bearing solutions were drained to bottles and sent to the uranium recovery facility. Spills of the liquid used to clean the cylinders occurred but the uranium lost through this spillage was never recorded (Goddard et al., 1991).

### K-1420 and K-1024 Mercury Recovery Room

The K-1420 mercury recovery room was used from the early 1960s to the early 1980s. Prior to that time the mercury recovery room was located in the instrument shop in K-1024. The room contained equipment that was used to clean and recover mercury that was used in plant instruments. Drain lines flowed through the building's north side process lines to the K-1407B Holding Pond (Goddard et al., 1991). Mercury was found in the center floor drain of the K-1420 Mercury Recovery Room. This is an indication that mercury spills likely occurred in this room. Anecdotal evidence also indicates this to be true. However, at this time, no reliable estimates on the amount of mercury spilled have been located. Additionally, mercury has not been identified as a contaminant in the K-1407B sludge (Baer, 1993).

### K-1420 and K-1303 Oil Decontamination and Recovery Facility

The oil decontamination facility located in K-1420 was used to remove uranium and sludge from waste hydrocarbon lubricating oils. During the early years of plant operations, lubricating oil was mostly obtained from the uranium enrichment process buildings' cold trap rooms. Initially, the lubricating oil was processed in the K-1303 building. When K-1420 became available (around 1953 or 1954) the recovery process was moved. When the use of the cold trap rooms, which condensed uranium hexafluoride gas to a solid, ceased in the mid-1940s, generation of uranium contaminated oil slowed (Goddard et al., 1991).

Contaminated oil was placed in the mixing tank and mixed with carbon tetrachloride to decrease the oil's viscosity. From the mixing tank, the oil flowed to the pressure filter tank, where the uranium-contaminated oil sludge was separated from the oil's clean component. The sludge was packaged in drums pending eventual treatment through the K-1420 uranium recovery process. The clean oil was transferred to the mixing tank of the second equipment assembly, where cobalt fluoride was added to refluorinate the oil and restore its inertness to fluorination agents in the uranium enrichment cascade. The resultant mixture moved to the pressure filter tank, where the

cobaltous fluoride component was removed and stored for later treatment. The clean Miller's Fluorinated Lubricating (MFL) oil was reclaimed for continued use. The oil recovery batch process handled uranium-bearing oil, carbon tetrachloride, and cobalt fluoride (Goddard et al., 1991).

#### K-1420 and K-1410 Electroless and Electrolytic Nickel Plating

Steel surfaces exposed to fluoride gases in the diffusion cascade were protected from corrosion with nickel plating, both electroless and electrolytic (ORGDP, 1985a). Electroless plating was used when uniformity of coat and coverage of unusual shapes was desired. The electrolytic process was used when simple nickel-plated parts were to be constructed by welding or when special tensile strengths were demanded of the coating (ORGDP, 1985a).

The electroless facility in K-1420 consisted of an 8000-gallon electroplating tank, heat exchangers, pumps, reagent addition systems, and immersion cleaning or pickling tanks (USDOE, 1979 and ORGDP, 1985a). In addition to the plating process, the operation involved the cleaning of parts to be plated. Cleaning solutions normally used included an aqueous solution of 25% hydrochloric acid, 75% sulfuric acid, detergents, carbon tetrachloride, trichlorethylene, and perchloroethylene (USDOE, 1979; Goddard, 1991; and ORGDP, 1985a). Vapor degreasers and vapor blasters were part of the cleaning equipment (ORGDP, 1985a). Rinse tanks were provided to rid the parts of the cleaning and plating solutions (USDOE, 1979). The rinse flows were piped to the K-1407A neutralization pit for pH adjustment then sent on to K-1407B holding pond. It is likely that in the early years of the facility's operation the plating solutions were also discharged to K-1407A, however this has not been confirmed at this time. In the late 1970s, however, the plating bath and cleaning solutions were periodically transported to the Y-12 biodegradation facility (USDOE, 1979).

From 1963 to 1979 the K-1410 building was used for electrolytic nickel plating operations (Goddard et al., 1991). The building contained acid and plating solution tanks for electroplating large parts (ORGDP, 1985a). Liquid effluents from the electroplating facility included alkaline cleaners, acids, nickel sulfate, and fluoride. These liquids were discharged into the building's process drains (Goddard et al., 1991). No stack monitoring data were located for this facility, however indoor monitoring data are available.

#### K-1401 Maintenance Facility

From 1944 to 1985, the K-1401 building was the primary maintenance facility at the K-25 site. Various cleaning, conditioning, and assembly operations were conducted in this building. During construction and installation of the gaseous diffusion equipment and throughout the K-25 site operational period, there was an emphasis on cleanliness. This was because uranium

hexafluoride reacts violently with hydrocarbons. All oil and grease had to be removed from parts that might be exposed to the uranium hexafluoride. Part of the maintenance facility was a metals cleaning and conditioning operation that was used to prepare various metals for fabrication and assembly operations. The primary method for cleaning involved the use of four 10-by-50-foot cleaning baths. Over the years various acids and solvents were used in the metals cleaning operation. The chemicals utilized included hydrochloric acid, sodium hydroxide, 1,1,1-trichloroethane, trichloroethylene, and carbon tetrachloride (Goddard et al., 1991).

In order to combat the corrosive nature of uranium hexafluoride, process equipment required conditioning with fluorine gas prior to installation. The K-1401 facility contained an area where process equipment was conditioned with fluorine gas prior to installation in the gaseous diffusion cascade (USDOE, 1979).

Other maintenance activities in building K-1401, such as equipment fabrication, assembly and painting required the use of a variety of materials. These materials include: chromium, mercury, cutting oil, paints, epoxy, and methyl-ethyl ketones. No data were located on the quantities of the chemicals used in the maintenance operations. The waste solutions from the cleaning operations in K-1401 were neutralized in the K-1407A neutralization pit prior to discharge to the K-1407B pond. The fluorine gas was vented to the atmosphere (Goddard et al., 1991). No stack monitoring data were located for this facility, however, indoor air monitoring data are available.

#### K-1423 Toll Enrichment Facility

The Toll Enrichment Facility, K-1423 was placed in operation in January, 1969. It was the shipping and receiving point for private industry owners of uranium hexafluoride who sought uranium enrichment services from ORGDP for the fueling of light water nuclear power reactors. The uranium hexafluoride feed for the diffusion plant was shipped by truck to the K-1423 Toll Enrichment Facility at subatmospheric pressure in 2.5, 10, and 14 ton cylinders, then transported to the cascade feed facility at Building K-1131. From 1969 through 1983, a total of 13,297 two and one half ton product cylinders were shipped to private industry (ORGDP, 1985a).

Under normal operating conditions, the only release of liquid effluent from the Toll Enrichment Facility to the environment was the condensate from the steam-heated autoclaves. This condensate was routed to Poplar Creek. Likely gaseous effluents from the Toll Enrichment Facility were a result of routine purging of cylinder connections after sampling and transfer, and from the cylinder venting operations. These gases were normally evacuated through a K-25 pipeline to K-27 (ORGDP, 1985a).

### K-1131, Feed Vaporization and Tails Withdrawal Facility

Feed Vaporization and Tails Withdrawal facilities have been located in Building K-1131. The function of the Feed Vaporization facility was to heat cylinders containing solidified uranium hexafluoride to convert the material to pressurized vapor for feeding to the cascade. The feed system consists of autoclaves for vaporizing uranium hexafluoride, feed headers and the piping network for transporting and distributing the feed flow, and instrumentation and equipment necessary to control the flow into the cascade feed lines and to protect the cascade from sudden pressure rises.

The function of the Tails Withdrawal facility was to remove depleted uranium hexafluoride from the enrichment cascade in 14-ton cylinders. Continuous operation of the enrichment cascade required that the combined rates of uranium hexafluoride product and tails withdrawal equaled the uranium hexafluoride feed rate. The uranium hexafluoride tails, depleted in the U-235 isotope, were removed from stripping stages at the bottom of the cascade and piped through interplant headers to the tails withdrawal system. The primary system for tails withdrawal was compression-liquefaction. Compression-liquefaction is based on the principle of increasing the pressure of the uranium hexafluoride gas stream above the triple pressure point and then cooling the gas stream to condense the uranium hexafluoride as a liquid which was then drained into storage cylinders. In an alternate withdrawal method, the uranium hexafluoride gas was passed through water-cold traps and condensed as a solid. The cold traps were then isolated and the traps are heated to convert the uranium hexafluoride to the liquid phase for draining into storage cylinders. This method was utilized in the early years of plant operation.

A potential feed for the enrichment cascade was "reactor return feed." The source of this material is spent fuel that has been removed from a power reactor. Although the spent fuel was processed to remove fission products by the reactor facilities, experience indicated that trace contamination by transuranic elements such as plutonium-239 and neptunium-237 remained. The neptunium-237 was found to deposit on the plant equipment surfaces and the plutonium-239 was found to concentrate in the cylinders containing the recycled  $UF_6$ . Absorbent traps in the reactor return feed stream were used to trap the transuranics. The traps were vertically mounted model pipes charged with cobaltous fluoride pellets and contained in a heated housing. The traps were not needed in the later years of gaseous diffusion operations because all reactor return feed was sampled and free of significant transuranics prior to being used for feed.

In both tails withdrawal methods, the noncondensables in the withdrawal stream were recycled back to the enrichment cascade along with small amounts of uranium hexafluoride.



Records indicate that feed vaporization operations were the source of several "abnormal" uranium hexafluoride releases. These release records will be discussed in Sections 3.3 and 3.4.

#### K-1131 Fluorine Plant and Uranium Oxide Conversion

The K-25 site produced fluorine, which was used primarily to treat various pieces of equipment in Building K-1131. The process used an aqueous solution of potassium bifluoride and hydrogen fluoride in five electrolytic cells with direct current electricity and carbon electrodes. The electrolysis produced two gaseous products - hydrogen and fluorine - which were withdrawn separately. Each product stream also contained from 0 to 14% HF. The fluorine was passed through filters to remove electrolyte, a condenser to remove HF, and was distributed to plant users. The hydrogen was vented directly to the atmosphere.

From 1950 until 1968, the K-1131 feed plant was to convert uranium dioxide to uranium hexafluoride. The K-25 site received uranium oxide from outside vendors. Materials used in the process included dry ice, trichloroethylene, and carbon dioxide. These release records will be discussed in Sections 3.3 and 3.4.

#### K-413 Product Withdrawal Facility

The function of the K-413 Product Withdrawal Facility was to remove enriched uranium hexafluoride from the cascade into 10-ton and 14-ton cylinders. The facility was placed into operation around 1947. For most of its operating history, the Product Withdrawal Facility was comprised of two independent withdrawal systems that could simultaneously remove from the cascade two product streams having different assays. The uranium hexafluoride product stream, which is enriched in the U-235 isotope, was removed from the enricher stages at the top of the cascade and piped through interbuilding headers to K-413. The system used for product withdrawal is compression-liquefaction. Over the years, the K-413 facility was the site of several uranium hexafluoride releases (ORGDP, 1985a).

#### K-1303 Mercury Recovery and Decontamination and Recovery Operations

The K-1303 facility accommodated a variety of major operations including units for fluorine production, decontamination and recovery of fluorinated lubricating oils, vacuum distillation and recovery of mercury, decontamination of uranium enrichment process equipment, and a research compressor. At one time this facility contained an "incinerator" which consisted of a 55-gallon drum under a hood exhaust (Visner, 1947). With the exception of the mercury distillation unit, little information is available about the operations in this building. The Mercury Distillation and Recovery Unit operated from 1948 to 1956. At that time, operations were transferred to K-1024. In 1948, the exhaust system in the mercury distillation area was modified to direct and discharge mercury fumes to the atmosphere above the building's roof. A former mercury unit

worker recalled that globules of mercury used to collect on the ground along the roof's drip line prior to the addition of the northeast wing over the area. The globules may be explained by the presence of the exhaust system installed in 1948. The system discharged mercury fumes into the atmosphere above the roof. Some of the fumes may have condensed onto the roof and eventually washed by rainfall onto the ground alongside the building (Goddard et al., 1991). No effluent monitoring data were located for this building, however indoor air monitoring data are available.

#### K-1413 Research and Development Facility

The K-1413 building operated as a research development facility in the early 1950s. Originally, the K-1413 building consisted of only the K-1413 building and one waste disposal pit (east pit). In the late 1960s, an annex was added to K-1413, and a second disposal pit (north pit) was constructed. In 1974-1975 a third disposal pit (south pit) was constructed to isolate the treatment and disposal of classified waste (waste which may contain materials whose presence is classified for security reasons). A pumping station, located just north of the K-1413 building was also built at this time (Goddard et al., 1991).

A variety of research and development activities were conducted in the K-1413. One of the early K-1413 development projects involved the fluorination of uranium metal chips to uranium hexafluoride. The chips were immersed in perchloroethylene and shipped to the K-1413 facility in 55-gallon drums. Interviews revealed that spills of perchloroethylene occurred at the facility. Uranium hexafluoride was accidentally lost to the stack in at least one incident (Goddard et al., 1991).

Other development projects included the following:

- Fluoride volatility using depleted and slightly enriched uranium oxide in zirconium-clad fuel elements,
- Investigation of compressors for pumping fluorine and uranium hexafluoride,
- Reduction of uranium trioxide to uranium dioxide using hydrogen and vibrating trays,
- Conversion of  $UO_2$  to  $UF_4$  using HF,
- Tower fluorination of  $UO_2$  or  $UF_4$  to uranium hexafluoride,
- Calcination of uranyl nitrate to  $U_3O_8$ ,

- Hydration of normal assay  $UO_3$  with solutions of Ammonium hydroxide for specific crystal formation,
- Separation of boron isotopes,
- Trapping of radioisotopes of elements such as antimony, tantalum, titanium, and ruthenium from uranium hexafluoride reactor returns (These fission and neutron activation products were present in uranium recovered from reactor fuel at Hanford), and
- Separation of radioactive noble gases (e.g., krypton and xenon) in pilot plant.

The disposition of the wastes from this facility varied with time. Prior to 1974 -1975 the wastes generated in the east and north pits were discharged into storm drains, later, the wastes were pumped from the pits to K-1407A or K-1407B. Then still later, when the need arose to treat and dispose of classified waste from the gas centrifuge operations on-site, wastes were transferred from the north pit to the south pit, pumped from the south pit into a tanker truck, and transported to the K-1407-C Retention Basin (Section 3.1.3) (Goddard et al., 1991).

The east and north pits at the K-1413 site were originally separated into two halves by 4-inch thick baffles within four to six inches of the bottom of the pits. Both halves of the pits were filled with limestone gravel. The waste stream entered near the top of the pit and was forced by the baffle to percolate down through one half and back up the other half of the pit before it was discharged. Wastes generated in the laboratory on the mezzanine and in the ground floor areas A and B of the K-1413 building were treated in the east pit, and wastes generated in area C were treated in the north pit using this limestone neutralization process. The limestone was removed from the pits when the pumping station was installed. No records are known to exist concerning the disposal of the limestone.

The south pit was also used to dispose of wastewater transported by tankers from the Y-12 Plant. This wastewater was treated in the south pit. Records indicate that one shipment of Y-12 wastewater contained 2.7 mg/L of mercury (Goddard et al., 1991).

The volume and composition of the chemicals processed during the facility's life cycle operations varied with time. Records of the chemicals used in the various activities along with quantities and concentrations have not been located; however, a partial list of possible contaminants includes (Goddard et al., 1991):

Uranium tetrafluoride	Uranium hexafluoride
Uranium oxide	Perchloroethylene
Antimony	Tantalum
Titanium	Zirconium
Fluorine	Uranyl nitrate
Ammonium hydroxide	Anisole
Nickel	Ruthenium
Diethylene glycol	Dibutyl ether
Sulfuric acid	Hydrochloric acid
Nitric acid	Organic acids
Nickel	Copper
Mercury	Chromium
Hydrogen fluoride	Sodium fluoride
Tungsten hydroxide	Rhenium hydroxide
Uranium hydroxide	Sodium hydroxide
Calcium hydroxide	Potassium hydroxide
Tungsten hexafluoride	Rhenium hexafluoride

#### Other Support Facilities

Little information was located for some process buildings utilized in the early years of plant operation. Some information regarding the use of certain buildings was found in health physics activities monthly reports for 1947 and 1948 (Visner, 1947). The following is a list of buildings for which information is sparse:

- K-1405 Lab and Pilot Plant
- K-1301 C-216 Manufacture and C-616 Recovery; Fluorine Production
- K-1212 K-1213 Shipping and Receiving
- K-1031 Carbon Storage and Sampling
- K-631 Tails Withdrawal
- K-131 Feed Purification Building
- K-101 C-816 Stripping
- K-1030 Electrical Maintenance Building
- K-1062 Hot Salvage Yard
- K-413 Chemical Operation Laboratory

Further research would be required to identify the operations in each of these buildings. Possible locations of information regarding the above buildings may be the Site Quarterly Progress Reports, the K-1034A Site Records, and employee interviews.

### 3.1.9 Laboratories

The laboratories provided technical support both for the gaseous diffusion plant and research and development for other advanced isotope separation systems. There were two types of laboratories at the K-25 site, production laboratories and development laboratories. The production laboratory facilities were primarily located in K-1004A, B, C, D, and K-1006 (ORGDP, 1985a). The development laboratories were primarily located in K-1413; K-1405-6; K-1303; K-1006; K-1004A, B, C, D and part of K-1401.

In both the production and development laboratories, a variety of analytical techniques were utilized, including organic and inorganic chemical analysis, mass spectrometry, spectrochemical analysis, microanalysis, and X-ray analysis. The analytic work included the routine monitoring of production processes, development of analytical procedures, and on-line instrumentation development and monitoring.

Materials used in the laboratories included uranium hexafluoride samples from the processing areas, which were analyzed for U-235 and purity (K-1004A). Occasionally, tungsten hexafluoride and rhenium hexafluoride were processed. Other materials used in the laboratories included fluorine gas, hydrofluoric acid, chlorine trifluoride, and hydrogen gas. Additionally, numerous small quantities of laboratory reagents and chemicals were found throughout this area (Goddard et al., 1991).

Uranium was also utilized in experiments located in development laboratories in K-1006, where the physical and chemical properties of the uranium hexafluoride in combination with materials found in the various processes were determined. The various analytical procedures generated liquid solutions of uranium. These solutions were stored in 5-inch diameter cylinders and eventually transferred to K-1420 for uranium recovery. Some quantities of uranium hexafluoride that were produced by the centrifuge enrichment development facilities were transferred in the laboratory to standard uranium hexafluoride cylinders for transport to the diffusion cascade area for sampling and refeeding (Goddard et al., 1991).

### 3.1.10 K-1501 Steam Plant

The K-25 steam plant generates steam used for process purposes and for space heating. This facility has seven natural gas boilers with a combined steam-producing capacity of 370,000 pounds of steam per hour. Prior to 1989 the boilers were coal-fired. At that time gaseous releases from the steam plant consisted of fossil fuel combustion products, specifically, sulfur dioxide (SO<sub>2</sub>), oxide of nitrogen (NO<sub>x</sub>), hydrogen fluoride (HF), and particulates. The primary liquid effluents from the steam plant consisted of the acidic discharge from the supply water treatment operation and the caustic boiler water blowdown. These discharges were combined

and neutralized before being released to the K-1407B holding pond. Settleable solids were collected in the pond and removed about every ten years (USDOE, 1979).

A weak sulfuric acid runoff from the coal storage yard occurred after heavy rains. Metals were also leached from the coal, the metals included arsenic, iron, copper, nickel, and manganese. This solution was collected and piped to the K-1407A neutralization pit or diverted around the pit into Poplar Creek, depending on the solution's pH and flow rate. However, this system was not in place until after 1985. Originally, the leachate from the north coal pile was routed to the Mitchell Branch of Poplar Creek discharge point (USDOE, 1979).

The ash that resulted from coal combustion was the only solid waste generated by the steam plant. This material was collected and dry deposited in local off-site landfill areas (USDOE, 1979).

### 3.1.11 Recirculating Cooling Water System, K-822, K-801-H, K-802-H, K-832-H, K-861, K-892-G, K-892-H, K-892-J

Excess heat in the gaseous diffusion cascade is removed by a recirculating cooling water (RCW) system and subsequently dissipated to the atmosphere via mechanical draft cooling towers. This type of heat dissipation has been used since the startup of K-25 in 1945 (USDOE, 1979). Raw water was taken from the Clinch River through the K-901 pumphouse and pumped to the K-892 clarification facility where lime, soda ash, and organic coagulants are added to remove calcium and magnesium as well as ordinary suspended solids. After clarification, the water used directly by the diffusion process was, until 1977, treated with a hexavalent chromium ( $\text{Cr}^{6+}$ ) compound to inhibit corrosion of the heat transfer equipment. After 1977, the RCWs were converted to a phosphate system. This treated water was subsequently transferred to the cooling loops. Through recycling within each loop, process heat is removed and dissipated to the atmosphere via the cooling towers. The coagulant sludge is discharged into the K-901A holding pond. Other liquid effluent from these systems is that from the K-1037 cooling tower, which enters Poplar Creek through the K-1700 discharge (Goddard et al., 1991).

To better understand the impacts of the airborne effluents from the K-25 cooling towers, a study was conducted in 1972 and 1973 to determine drift and associated chemical losses from the cooling towers, the extent of drift deposition on the terrestrial environs, and plume characteristics (USDOE, 1979). Drift losses were found to be in general agreement with losses reported from similar towers of the same age and ranged from about 0.001% of the recirculating flow for the K-33 tower to about 0.12% of the recirculating flow for the K-31 tower. Most of the drift deposition was found to be within about 400 meters of the towers (USDOE, 1979).

The study of the cooling towers' water vapor revealed that typical plume height was about 100 to 200 meters and downwind reach was about 100 to 200 meters. The maximum downwind extension was observed to be about 600 meters (USDOE, 1979).

### 3.1.12 Electrical Switchyards, K-732, K-762, K-792, K-709

The diffusion process required large amounts of electricity. The major use of electricity in the process buildings was to power the motors that turn the stage compressors. Beginning in 1945, power was transmitted to the switchyards of the plant and reduced by transformers. These transformers contained PCBs. The record of releases of oil containing PCB for these switchyards is not complete. However, it is likely that all of the switchyards experienced releases of oil containing PCBs (Goddard et al., 1991). Surface water and sediment monitoring data (Section 5.0) indicate that oil leaks from the switchyards may have migrated to the surrounding waterways. However, at this time, no estimates of historical releases are available.

When the K-762 and K-792 switchyards were constructed, french drains to collect and divert groundwater were installed under the gravel beds. In 1975, a series of french drains were installed under the K-732 gravel beds (Goddard et al., 1991).

### 3.1.13 Waste Disposal

A variety of methods have been utilized for the disposal of wastes generated by the gaseous diffusion process, support facilities, and laboratories. This section characterizes the different waste disposal sites at the K-25 Plant.

#### K-1203 Sewage Treatment Plant

The K-1203 Sewage Treatment Plant in its original configuration was placed in service in 1943. The plant consisted of two tanks, a pump house, a series of sludge-drying beds, a chlorine contact basin with a chlorine feed system, a sewage lift station, and a high-water lift station. In September 1976, the present configuration was placed in service. The tanks were removed and replaced with a clarifier/aeration unit. The process consists of two main stages: aeration and clarification (settling) (USDOE, 1979).

Prior to 1976, sludge from the drying beds was disposed of at the K-1070B and K-1070C/D Classified Burial Grounds, but this practice was discontinued because uranium was found in the sludge (USDOE, 1979).

Trace quantities of radioactive contamination entered the sewage system through the laundry and various changehouse facilities. The treated effluents from this facility were likely to contain uranium, technetium, and chlorine (Goddard et al., 1991).

### K-1407A Neutralization Pit

The K-1407A neutralization pit has operated since the 1940s. Originally the pit was used for neutralizing corrosive wastewater and received influent from: K-1420 uranium recovery operation, K-1501 zeolite regenerations, K-1420 plating operations, K-1413 laboratory solutions, K-1401 acid line, K-1420C floor pan cleaning solutions and coal pile runoff (Goddard et al., 1991). Corrosive wastewaters were neutralized with either powdered lime or concentrated sulfuric acid. Neutralized wastewater was then discharged to the K-1407B Holding Pond for settling if the water was contaminated with radioactive or hazardous wastes. Coal pile runoff was discharged to either the K-1407-E or the K-1407-F Settling Ponds. Since the opening of the Central Neutralization Facility in 1987, only coal pile runoff goes into the pit for neutralization. A variety of organics - aromatic hydrocarbons, acetone, freons, paints, epoxies, carbon tetrachloride, and other degreasers - were discharged into the neutralization pit through the K-1401 acid line. Transuranics in Building K-1420 were discharged into process drains, which flowed into the K-1407A Neutralization Pit and K-1407B Holding Pond (Goddard et al., 1991).

### K-1407B Holding Pond

The K-1407B holding pond was an unlined settling basin that received wastes for more than 40 years. The pond received organics and metal hydroxide precipitates after neutralization in the K-1407A neutralization pit. Additionally, the pond received direct discharge of wastewater from the following locations: K-1302 recirculating cooling water (RCW) supply, K-1503 neutralization pit, K-1421 incinerator drain, K-1420. The following wastes were likely discharged into the pond from various operations in K-1420: uranium compounds, transuranics, organic degreasers, and oils, including some containing polychlorinated biphenyls (PCBs) (Goddard et al., 1991).

In March of 1973, an estimated 8,158 kilograms of sludge contaminated with about 2.9 curies of slightly enriched uranium were dredged from the holding pond and pumped to the K-1407C retention basin (Goddard et al., 1991). In 1988, all water was discharged from the holding pond and the sludge was removed and fixed in concrete. These drums were stored at the K-1417 Block Casting/Drum Storage Area.

### Mitchell Branch of Poplar Creek

The K-1407B Holding Pond and the K-1407E and K-1407F Settling Ponds discharged to the Mitchell Branch of Poplar Creek (also known as the K-1700 stream). This stream is a transport mechanism for releasing hazardous and/or radioactive constituents to Poplar Creek (Goddard et al., 1991). Samples collected in June 1986 indicated the presence of chromium, lead, nickel, arsenic, and uranium in streambed sediments (Goddard et al., 1991).



The outfall of the K-1700 stream at Poplar Creek was routinely monitored for uranium and fluorides throughout most of the sites history.

#### K-1407H Central Neutralization Facility

Since its construction in 1987, the K-1407H Central Neutralization Facility (CNF) has provided pH adjustment and chemical precipitation for several aqueous streams throughout the K-25 site (Goddard et al., 1991).

#### K-1407C Retention Basin

The K-1407C Retention Basin was an unlined impoundment built in 1973 and closed in 1988. The Retention Basin was built in order to store the sludge that was dredged from the K-1407B Holding Pond. This sludge contained organics and corrosive metal hydroxide sludges that were radioactively contaminated (Goddard et al., 1991). Analytical results indicate that radioactive materials are present at depths exceeding eighteen inches (Goddard et al., 1991).

In 1988, all water was discharged from the holding pond and the sludge was removed and fixed in concrete. These drums were stored at the K-1417 Block Casting/Drum Storage Area.

#### K-1417 Block Casting/Drum Storage Area

Since 1987, this site has been used for storage of the concrete-fixed pond sludge from the K-1407B and C holding ponds. At one time there were as many as 46,000 drums of pond waste in this area. (Goddard, 1991).

In November of 1989, some of the drums were found corroded and leaking. The runoff from the drums was discharging directly to the Mitchell Branch of Poplar Creek. An investigation of the runoff area and sediment samples of the Mitchell Branch were collected to determine the extent of contamination caused by the leaking drums. The results of this investigation are available in Golliher, 1990.

#### K-1503 Neutralization Pit

Beginning in the mid-1940s, the K-1503 Neutralization Pit received waste from a chemical water softening process at the K-1501 Steam Plant. In 1973, a synthetic zeolite process was installed at the steam plant. The pit was upgraded, lined, and placed in operation as a neutralization pit. Contents of the pit initially flowed to a nearby catch basin and then on to the K-1407B Holding Pond (Goddard et al., 1991).

K-1007 B Holding Pond

From the 1940s to the present, process drains from laboratories in buildings K-1005, K-1006, and the K-1004 drained to the K-1007B Pond. These laboratories supported the various uranium enrichment and environmental monitoring programs. Prior to 1985, when a program to eliminate the disposal of laboratory wastes through process drains was implemented, an estimated 2200 gallons of laboratory wastes were discharged through lab drains each year. The chemicals that may have been present in liquids from the lab drain are as follows (Goddard et al., 1991):

Acetone	Diethylene glycol
Acetic acid	Dibutyl ether
Acetonitrile	Ethylene glycol
Benzene	Freons
Bromoform	Hexane
Cadmium	Hydrochloric acid
Carbon tetrachloride	Hydrofluoric acid
Dichloropropane	Hypophosphoric acid
Ethanol	Isopropyl alcohol
Ammonium hydroxide	Mercury
Chloroform	Methyl alcohol
Chloric acid	Methyl ethyl ketone
Chromates	Methylene chloride
Nickel compounds	Sodium hydroxide
Nitric acid	Sulfuric acid
Phosphoric acid	Technetium
PCBs	Tetrachloroethylene
Photographic solutions	Toluene
Potassium dichromate	Trichlorethylene
Potassium hydroxide	Tungsten
Pyridine	Uranium
Rhenium	

The K-1007B pond discharged directly to Poplar Creek. Monitoring data for the discharge location are available starting in 1974, when the outfall was National Pollution Discharge Elimination System (NPDES) permitted.

### K-901A Holding Pond

Discharge of wastewater into the marsh-like area that eventually became the K-901A Holding Pond area started in the late 1950s. A dam was constructed to create the holding pond in 1965-1966. Discharge to the holding pond was discontinued in 1985 (Goddard et al., 1991). Over most of its operating cycle the K-901A area and pond received sludge and blowdown from the K-25 recirculating cooling water (RCW). The holding pond also served as a disposal ground for the contents of compressed gas cylinders with inoperable valves (Goddard et al., 1991).

The RCW was treated with a corrosion inhibitor consisting of chromate, zinc, and a polyphosphate (a proprietary material marketed by Betz Laboratories, Inc.). These were discharged to the K-901A Holding Pond until 1974, when an electrochemical metal reduction unit was installed and the use of the Betz 1100 anionic polymer began. The electrochemical reduction unit (ANDCO) reduces soluble hexavalent chromium to insoluble trivalent chromium. In addition to RCW blowdown, various sizes and types of steel compressed gas cylinders that were used in laboratory work, research, and operations at ORGDP were disposed of in the K-901A Holding Pond. Cylinder contents included uranium hexafluoride, hydrogen fluoride, combinations of halides (bromine, fluorine, chlorine) and various fluorinated/chlorinated hydrocarbons. These cylinders were disposed of by suspending them over the K-901A pond and shooting them from a distance with a high-powered rifle to empty them of their contents, and then immersing them in the pond. Although no records were kept, it is estimated that from the mid-1960s to 1975, the contents of about 200 cylinders were discharged into the K-901A pond in this fashion. No records were located regarding the contents of the cylinders which were disposed in this manner, thus making it difficult to quantify releases from this operation (Goddard et al., 1991).

The K-901A pond discharged directly to the Clinch River. The outfall of the pond has been monitored for pH, flowrate, and metals since the mid-1970s. Monitoring data from 1973-1992 were located.

### K-1024 Dilution Pit

From 1945 to 1963 the K-1024 dilution pit served the instrument shop in building K-1024. From 1970 to 1985, the pit received waste from the centrifuge development laboratory. In 1985 the pit went to standby status (Goddard et al., 1991).

The design of the pit was such that the acid/solvent waste stream from the instrument shop, and later from the centrifuge development laboratory, was diluted by sanitary water entering the head of the acid waste line. The acid waste line then flowed through a diluting pit before discharge into the K-25 Site storm drain system. In the instrument shop, discharges were a result of the cleaning of GDP instruments. The cleaning involved the use of solvents and acids, including

trichloroethylene, isopropyl alcohol, Freon TF, hydrochloric acid, aqueous hydrochloric acid, nitric acid, and chromic acid. The volume and composition of the chemical wastes handled by the diluting pit varied with time. Additionally, when processing the instruments from the uranium enrichment cascades, the cleaning cycle could have contributed uranium to the waste discharge from the instrument shops (Goddard et al., 1991).

#### K-1515 Lagoon

Since its formation in 1943, the lagoon served as a sludge holding pond for the water treatment plant. Sludge was generated by the addition of a flocculating chemical (aluminum sulfate) to the incoming raw water from the Clinch River. Resultant solids were allowed to settle in the lagoon prior to discharge into the Clinch River. A corrosion inhibitor (sodium hexa-meta-phosphate) and a chlorine system completed the water treatment process (Goddard et al., 1991).

The effluents from the lagoon have been monitored under NPDES since the late 1970s. The monitoring effort included parameters such as flow rate, pH, and various metals. All monitoring data are available from K-25 plant personnel (Shoemaker, 1993).

#### K-1421 Incinerator

A brick incinerator was operated on the site from the 1950s to late 1960s to burn low-level contaminated combustible materials such as gloves, coveralls, wood, paper, plastic, and waste oil sludge. In the late 1960s, that unit was upgraded with a secondary burner and higher stack. In 1982, the upgraded unit was replaced with a unit modified to meet EPA standards. Ash was collected, and uranium leached and processed through the K-1420 Recovery Facility. The K-1421 Incinerator was shut down in 1982 because releases exceeded EPA standards for uranium. Uranium sludge from the MFL oil recycling project was burned in the incinerator from 1980 to 1982 (Goddard et al., 1991).

Information regarding pollution control equipment were not located. Monitoring data are available from K-25 plant personnel.

#### K-1064 Drum Storage and Burn Area

From the 1950s to 1960, the K-1064 area was used for the open burning of waste solvents. From 1960 to 1979, the area was used for storage of drums filled with waste solvents, organics (including polychlorinated biphenyls), and radioactively contaminated waste oil (ORGDP, 1989). Wastes burned at the site include waste paints, organic wastes, and radiologically contaminated waste oils. An accounting of the quantity of wastes burned has not been located to date (Goddard et al., 1991).

Records indicate that 1838 drums, with a total capacity of 90,000 gallons, were stored at the

facility prior to 1979 when drum storage was discontinued in the area (Goddard et al., 1991).

#### K-1085 Old Firehouse Burn Area

An existing farmhouse on the site was converted to a firehouse in 1944 and served as such until 1951. A fuel station and garage were also used on the site from 1944 to 1947. Open burning of contaminated oil took place on the concrete pads of three buildings until 1960. Also, waste oil was burned in an unlined pit until 1951, when the pit was filled. No reports of the quantities of waste burned at this unit have been located. Available information indicates that various waste solvents such as trichloroethylene, acetone, perchloroethylene, carbon tetrachloride, tetrachloroethylene, methylene chloride, PCB-contaminated oil, and paint wastes were burned in an open container at the unit. The waste-handling operations at this unit were not controlled. Pit burning was extinguished with water at the end of each day, and water was periodically pumped from the pit (Goddard et al., 1991). At this time, no information has been located regarding the disposal of the water pumped from the pit.

#### K-1070A Old Contaminated Burial Ground

From the late 1940s through 1975 this burial ground was used for disposal of low-level contaminated waste and mixed chemical waste. A summary of the location of wastes buried at K-1070A can be found in the Historical Investigations (Section 3.2.1.8). Wastes include MFL oil and a wide range of metals, including thorium, lead, and uranium. Much of the material, including scrap metal, empty barrels, and leached aluminum trioxide was contaminated with uranium (Goddard et al., 1991).

#### K-1070B Old Classified Burial Ground

The Old Classified Burial Ground was used from the early 1950s to 1976. The burial ground was formed by filling a large swampy area that drained into a small branch creek which, in turn, emptied into Poplar Creek (TSD, 1977). This site was used for the disposal of a wide variety of wastes. The burial ground contains classified materials such as hardware and equipment. Lead, uranium, aluminum, copper, beryllium, bronze, and brass are among the metals buried at this site. Asbestos is also buried at this site. Contamination from liquid organics or hydrocarbon oils is thought to be minimal. Radioactive material and surrounding dirt were reportedly cleaned from an area southeast of the burial ground in the mid-1970s (Goddard et al., 1991).

### K-1070C/D Classified Burial Ground

This burial ground operated from 1972 to 1989. The burial ground contains classified materials such as hardware and equipment. Also located in the same general area are pits used to dispose of hazardous chemicals and solvents (Goddard et al., 1991). Approximately 9,100 gallons of solvents and 1,600 pounds of chemicals were disposed of in these pits. Lead and uranium are among the metals buried in the burial ground's trenches (Goddard et al., 1991). Detailed information on the disposal of chemicals is available in a classified document (Perry, 1984).

### K-1070D1, D2, and D3 Storage Dikes

From 1980 through April, 1985, these dikes stored a total of 912 drums of hazardous waste liquids. An inventory of the wastes that were stored was kept, and the following represents a portion of the list: trichloroethane and water, waste oil greater than 5 ppm PCBs but less than 50 ppm PCBs, paint waste, trichloroethylene and water, methylene chloride, perchloroethylene, acetone, xylene, and freon. There are no known releases from the area (Goddard et al., 1991).

### K-1070A Landfarm

Between 1979 and 1985 about 190,000 gallons of diffusion cascade lubricating oil from the K-33 diffusion process was recycled using Fuller's Earth. The Fuller's Earth served to remove concentrated acids, sludges, and other degradation products from the cascade oil. Once saturated, the Fuller's Earth was sampled and analyzed for PCBs. The PCB concentrations in the Fuller's Earth had to be less than 5 ppm before the Fuller's Earth could be applied to the surface of the K-1070A landfarm. Runoff or leachate data from the site are not available. However, preliminary analyses of soil for total petroleum hydrocarbons, total inorganic elements, and semivolatile organics indicate that all levels are less than or equal to background values (Goddard et al., 1991).

### Contaminated Scrap Metal Dumpsters K-1401-2W, 3E, and 4W, K-1420, K-1004L, K-1006, K-1225, and K-1030

The scrap metal dumpsters received contaminated solid metal scraps generated by various operations around the site. Estimates of releases from the dumpsters are not available.

### K-901A North Disposal Area and K-901A South Disposal Area

The K-901A Sanitary Waste Disposal Area began receiving waste sometime in the mid-1940s and the K-901 Waste Disposal Area began receiving waste in the early 1950s. Both received waste from on-site contractors and maintenance activities until the mid-1970s (Goddard et al., 1991). The disposal areas are separated by approximately one quarter of a mile.

No records of the materials disposed of in the two disposal areas exist, and information reported here is based on interviews with K-25 employees and site observations (Goddard et al., 1991). The two sites probably received varying quantities of road bedding materials, paint cans and buckets, wallboard, lumber, soil, rock, roofing and guttering materials, piping, concrete, asphalt, and steel. Some of this construction material is probably interlaced with small amounts of low-level uranium-contaminated roofing removed from the enrichment buildings. Additionally, chemically treated wood removed from repaired cooling towers also may have been disposed of at the site (Goddard et al., 1991).

### K-770 Scrap Metal Yard

The K-25 plant began storing low-level contaminated scrap metal at K-770 in the 1960s. This waste consisted of various types of metals generated from operations at the K-25 site. The scrap metal was contaminated with radioactive materials, primarily uranium. Asbestos from pipe insulation was also present. Samples collected in a storm drain in the storage area revealed detectable concentrations of radioactive constituents. Soil samples collected in the past also revealed detectable concentrations of radioactive constituents (Goddard et al., 1991). Runoff from the metal yard was not controlled and drained through the storm drain mentioned previously. The effluents from this storm drain were not monitored until October, 1992.

### K-720 Fly Ash Pile

From the 1940s through the 1960s, fly ash from the coal-fired steam plant was disposed of in a waste pile south of the K-25 site and north of the Highway 58 bridge over the Clinch River. Runoff and leachate from this fly ash pile were not controlled (Goddard et al., 1991).

## **3.2 SUMMARY OF THE INVESTIGATIVE PROCESS USED AT K-25**

This section describes the document centers and repositories utilized in the search for historical information or data. Keyword searches were utilized at a number of the repositories. The keywords used are listed below:

Accident	Monitoring
Annual	Monthly
Bear Creek	Off-Site
Biological Monitoring	Pits
Clinch River	Poplar Creek
Contaminated	Progress
Discharge	Release
East Fork Poplar Creek	Sediments
Effluent	Soil
Emissions	Solvents
Environmental	Stack
Exposure	Summary
Hazardous	Surface Water
History	Toxic
Incident	Trenches
Inventory	Waste
Leak	Watts Bar
Melton Hill	White Oak

### 3.2.1 Unclassified Document Sources

The unclassified information repositories located at the K-25 site are described in this section.

#### Site Document Response Center (SDRC), K-1200

The SDRC was created in January of 1991. The purpose of the center was to provide a gateway to all site information to make it readily accessible to outside auditors. Originally, the SDRC was set up to respond to DOE Tiger Team questions, however the center has evolved to serve all employees and outside auditors. At the time of review, the Center contained approximately 2,500 documents. The documents in the Center consist of unclassified formal reports, correspondence, and photographs which address issues in which the Tiger Team or other auditors might be interested. Few documents in the SDRC predate the 1980s. ChemRisk reviewed titles of the entire contents of the SDRC and reviewed the contents of approximately 10% of the documents in the Center.

#### Environmental Management Division Document Center (EMD), K-303-8

The Environmental Management Division (EMD) document center was created in September, 1990 to provide a repository for all correspondence, data, or formal reports generated by personnel in the Environmental Management Division. This document center is a mixture of



formal reports and correspondence. The vast majority of the documents are from the 1980 to 1990 time period, however, a significant portion of documents are from the 1970s. The center contains few documents from the 1960s time period and almost no documents from the 1950s. ChemRisk requested a keyword search of the database. The search produced a listing of approximately 2,000 documents. The document lists were searched for document titles that may be relevant to the Health Studies. About 10% of the documents contained in the lists were selected and reviewed by ChemRisk.

The EMD center also has approximately 40 cubic feet of unindexed records in storage. A cursory review of those documents revealed that these records were a mixture of correspondence, draft documents and analytical data. Because these records are not indexed, a search of them would be quite time consuming and it is likely that the information contained in these records could be found in summary documents in other document repositories.

#### Compliance and Environmental Policy Document Center (CEP), K-1001

The Compliance and Environmental Policy (CEP) document center was created in March of 1991, in order to create a central repository for documents and correspondence relating to Environment and Safety regulations. At the time of review, the Center contained approximately 33,000 documents. The majority of these documents are correspondence or small reports with letters as attachments. The time period covered by the documents in the Center is approximately 1950 to the present. ChemRisk reviewed the list of 33,000 documents. Through that review, approximately 700 documents were selected for a more detailed review.

Similar to the EMD Center, the CEP Document Center has 50 cubic feet of unindexed records. A search of these unindexed records was not conducted.

#### Environmental Restoration Document Center, K-1210

The Environmental Restoration Document Center was formed in 1990 to give the ER Division a repository to store and maintain documents or correspondence relating to environmental restoration efforts at the Oak Ridge Reservation. The Document Center receives documents for all five DOE Oak Ridge Operations sites (K-25, Y-12, X-10, Paducah, and Portsmouth). Additionally, the Center receives documentation from the files of employees that retire from the K-25 site. At the time of review, the Environmental Restoration Document Center contained approximately 4,000 formal reports and approximately 40,000 internal correspondence documents. The distribution of the documents is approximately 30% from the time period prior to 1975, approximately 40% from the 1975 to 1985 time period, and approximately 30% after 1985. ChemRisk reviewed approximately 400 of the formal reports, but, because the internal correspondence was considered internal use only, was not allowed access to this file until late in the project. Consequently, time constraints did not allow a review of this file.

### Waste Management Document Center (WMDC), K-1037

The WMDC was created in 1990 to provide the Waste Management Division with a central repository to store and maintain documents. At the time of review, the Document Center contained approximately 10,000 documents. The majority of these documents were correspondence. A formal keyword search of the document database was performed. This search revealed that there was considerable overlap between the holdings of the WMDC and the EMD, which had been reviewed previously. Because many of the documents in the WMDC were duplicates of documents in the EMD, only about 20 documents were reviewed.

### Records Analysis Department, Townsite

The Records Analysis Department is in the process of creating a database of all records retention schedules. The development of this database is in response to a DOE mandate which states that all sites must have a tabular listing of all records retention schedules. Prior to this database, no such listing was available for any of the ORR sites. A record schedule is a series of records for each division that have a particular retention period. Data entry of the record schedules has been completed, however, the locations of records within a series have not been determined. Furthermore, it is not always known whether the records within a series have been destroyed as retention periods elapsed. This database appears to have limited usefulness for the study at this time. It appears that its best use is to identify record series located at the site records vault.

### Environmental Monitoring and Surveillance Reports

Environmental Monitoring and Surveillance Reports for the Oak Ridge Reservation have been published annually since 1971. These reports provide data for the ambient air monitoring, surface water monitoring, and groundwater monitoring. Information regarding the availability of ambient air monitoring and surface water monitoring data is discussed in Section 5.0. The monitoring reports provide data for both airborne and waterborne discharges of radionuclides. Until the mid-1980s, these data are presented as annual totals for the entire Oak Ridge Reservation. In the mid-1980s, annual radionuclide discharge totals are provided for each Oak Ridge facility.

### Historical Investigations

This is a six volume set of binders which details the background information used to create the document entitled "Site Descriptions of Environmental Restoration Units at the Oak Ridge K-25 Site, Oak Ridge, Tennessee K/ER-47" (Goddard et al., 1991). The majority of the information located in each binder was collected by two K-25 retirees that researched the history of each Environmental Restoration (ER) Unit. The volumes contain a summary of historical operations

at each ER unit and details the data gathering techniques used to generate the historical summary. In general, the information provided was gathered from historical documents in the Applied Technology Library or through personal interviews with current and retired K-25 personnel. The historical investigation documents occasionally provide information concerning the types and quantities of chemicals used at particular facilities at the K-25 site. The information provided in the historical investigation is the basis for a significant portion of the information provided in this report.

#### RCRA Facility Investigation Documents

The RCRA Facility Investigation documents provide geographical, historical, operational, geological, and hydrological data for the solid waste management units (SWMUs) at the K-25 site. The SWMUs are subject to assessment by the U.S. Environmental Protection Agency (USEPA), as required by the 1984 Hazardous and Solid Waste Amendments to the Resource Conservation and Recovery Act (RCRA). In general, the historical information found in the RCRA documents is well-researched and comprehensive. These documents provide an excellent starting point for site historical information.

#### Federal Records Center (FRC), East Point, GA

Through communication with the supervisor of K-25 Site Records, ChemRisk learned that K-25 does not have any records stored at the Federal Records Center. All records which had been previously stored at the FRC were destroyed in accordance with their retention schedules (McCollum, 1993a). Additionally, no records are available concerning the contents of the records that were destroyed (McCollum, 1993b).

#### K-25 Site Photography, K-1004J

The K-25 site photography department has a collection of negatives and photographs of the K-25 site and its surroundings that date from 1964 to the present. All negatives and photographs prior to that time were destroyed. The photography department also has a collection of historical photographs from the 1945-1950 time period that was obtained from DOE photography.

#### DOE Photography

The DOE photography collection includes photographs and negatives of both the K-25 and S-50 sites from the early 1940s to the present.

### 3.2.2 Classified Information Sources

ChemRisk team members with Q clearances were given access to all areas of the plant, and all information sources. This section describes the classified repositories at the K-25 Site.

#### Central Files, K-1001

Central Files contains a record copy of classified and unclassified reports generated by K-25. The File contains approximately 79,000 classified reports and an unknown number of unclassified reports. All classified report numbers are available on a database. However, a printout reviewed by ChemRisk indicated that while report numbers are listed, the majority of the report titles have not been entered into the database. The unclassified reports have never been indexed. Therefore, it is not known how many, or which, reports are part of the Central Files. Because the classified index is incomplete and there is not an index for the unclassified documents, the usefulness of this repository for the study is limited. The Central Files could be utilized only when a document is already known to exist and a document number is available. If the document number is available, the Central Files personnel could then search the record shelves to determine if the desired document is part of the Central Files collection.

#### K-1034A Site Records

The Site Records vault contains 12,200 cubic feet of inactive records from all past or present divisions. The records are indexed by box. The index consists of a four-by-six inch card, or retention schedule, for each box. The cards are filed by the division from which the documents originated and contain a general description of the box contents. For example, a retention schedule may state that a certain box contains Inventories and Distributions for 1963. The card also provides the location of the box in the 1034A vault and the length of retention for the box. However, at this time, under DOE order destruction of records useful to epidemiologic or health studies is not permitted. These records contain information concerning industrial hygiene, worker identification, site organization, configuration, and operation (Barrow, 1991).

ChemRisk was permitted access to all retention schedules and records in the 1034A vault. The retention schedules were reviewed and approximately 75 boxes representative of various divisions and time periods, were selected for detailed review. The records in the 1034A vault are of great historical value and a more detailed investigation of this center would be necessary in a Phase II investigation.

### Applied Technology Library, K-1002

The Applied Technology Library (ATL) was formed about 1946 to provide a place where employees would have access to scientific literature generated at K-25 and other DOE sites including, but not limited to, X-10, Y-12, Paducah, and Portsmouth. A classified card catalog that covers the period from the early 1940s to the mid-1960s is available for review. For the mid-1960s through the mid-1980s time period, there is a classified microfiche index of documents. The microfiche index is separated into the three uranium enrichment technologies; gaseous diffusion, gas centrifuge, and AVLIS. The separation of the index into the three technologies was carried out in order to maintain control over the number of people who reviewed the information on the various technologies. Formally, this form of control is called the "need to know". ChemRisk was allowed access to all three indices. Neither the card catalog nor the indices are computerized, and it is time-consuming to do a thorough search. Reviews of the card catalog were made of the keywords listed in Section 3.2. The most valuable set of information that was identified with regard to historical activities at the K-25 site are the classified plant quarterly progress reports that range in date from 1948 to 1985. These reports will be discussed at greater length below.

The microfiche index was placed on a computerized system, however, the software is no longer available to utilize the information on the computer tape. It may be possible for a computer programmer to design software to access the information on the computer tape.

### Engineering Design and Information System, K-1001

The Engineering Design and Information System (EDIS) is an index of engineering drawing titles for the K-25 site. Facility Records is the organization responsible for maintaining the index. There are both classified and unclassified microfiche versions of the EDIS. The EDIS is organized by building, and subheadings within each building include subjects such as ventilation, electrical, etc. The index provides the reference drawing number for each drawing of interest. The drawing can then be retrieved for review. Ninety-five percent of the K-25 drawings are on microfilm and can be readily retrieved by Facility Records personnel.

Facility Records has a small collection of engineering drawings for the original S-50 site. The collection primarily consists of civil, structural, and architectural drawings. Only one process drawing for the S-50 site was located. Because, these records were not part of the Union Carbide Collection, these drawings were not indexed or microfilmed. Facility Records personnel are still in the process of searching old files to try to locate more S-50 drawings.

### Inventory

At K-25 there are two methods by which a division may purchase chemicals or radionuclides. The first is through the K-25 site stores, and the second is through direct material purchase from the manufacturer. The stores purchases have been on a computer database at least since the early 1970s. The software used to create these databases is not available, however the computing and telecommunications group at K-25 is working with ChemRisk to retrieve the stores purchase files. The data are input to the stores system by a stores catalog number, not by chemical name, and the catalog number is required to query the database. ChemRisk provided the K-25 Computing and Telecommunications group with a list of catalog numbers that were obtained from an April 1979 Consolidated Stores Catalog for Chemicals and Gases. Catalog numbers were searched on a 1983 stores inventory. The information gathered from this search permits the determination of whether there was any purchasing activity for a particular chemical or gas for the year. This database provides information for any chemicals that have been ordered through stores, but provides no information or inventory data for chemicals that may have been ordered by a division directly from a manufacturer. The extent of direct ordering is not known at this time, however based on preliminary evidence it is likely that the number of chemicals ordered directly by a division is quite significant.

Other inventory information is available in the Plant Records Vault (K-1034A). This inventory information was found in the General Accounting and Finance Division Records (Boxes 6,3,7,38 and 7A,4,2,10) and is classified. Inventories were found from July of 1959 through July of 1964. The inventories were reported by stores either quarterly or monthly. The inventory records had the following format:

Catalog #	Description	Unit	Balance
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The types of chemicals and materials for which information is available are solvents, coolants, acids, bases, and various other metals. In some instances the materials are referred to by catalog number only, or are listed as classified. However, when inventory material from several months or quarters are compiled, a material referred to by catalog number in one month may be referred to by its name another month, thereby making it possible to determine the identity of the compound.

The database from the 1970s and 1980s only captures the chemicals, radionuclides or gases that were ordered through stores, however, at this point in time it is not known whether the Finance and Accounting inventory information only reflects the stores information or is inclusive of direct charge purchases. In order to determine what quantity of certain materials may have been released, it would be important to determine the quantity of chemicals purchased via direct charge.

Other inventory information that would capture the direct charge items is available only for very recent years in the form of the Accelerated Vendor Inventory Delivery (AVID) system. This system is an order entry system (called an up-front system) because when the material is first requested for purchase, it goes into this system. The system contains both hazardous and non-hazardous material. The system first went into operation about 1987, but records were never archived until 1989. Since the plant was placed in standby condition in 1985, inventory information from 1989 would not be very useful.

### Quarterly Progress Reports

The K-25 Site Quarterly Progress Reports summarize the activities each quarter of the fiscal year. These reports are available for the time period 1948 - 1985. Throughout the 35 years of Plant Operation, the structure of the progress reports essentially remained the same. The following is an example of the contents of the quarterly progress reports.

- A. Summary
- B. Fiscal Affairs
- C. Production
  - 1. U-235 Production
  - 2. Electrical Power
  - 3. Auxiliary Chemical Production
  - 4. Auxiliary Operations
  - 5. Barrier Production
  - 6. Material Usages
- D. Engineering and Maintenance
- E. Health and Safety
- F. K-25 Industrial Relations
- G. Research and Development
- H. SF Accountability
- I. Plant Expansion

The quarterly progress reports are an attempt to summarize the important activities at the K-25 site. The reports cover Production, Research and Development, Health and Safety, Labor Relations, and Uranium Accountability. These reports are classified and contain so much detailed technical information that it would be very difficult to have them declassified. However, there are sections, such as Auxiliary Production, Material Usage, and Health and Safety, that likely could be extracted from the classified Quarterly Progress reports of the documents and declassified.

### Division Progress Reports

At the K-25 site individual divisions wrote weekly, biweekly, monthly, or quarterly progress reports. These reports described the on-going activities of the division. Many of these documents are located in the Applied Technology Library Vault. Most of these documents remain classified. However, some of these documents have the potential for declassification. Generally, these reports are of historical value and declassification of some of the reports (i.e., health physics and industrial hygiene reports) may be beneficial.

### **3.2.3 Areas not Covered**

Because of the time constraints of Phase I, a number of document repositories were not reviewed. This section describes each repository that was not reviewed.

#### Records Coordinators

A list of records coordinators for each division was provided to ChemRisk (Repository number 1207). A limited number of Coordinators for divisions with records that might be relevant to off-site releases were contacted in an attempt to track records for certain divisions. Preliminary impressions were that the records coordinators contacted were unfamiliar with records disposition and were of limited assistance. Although, these coordinators were unfamiliar with records disposition, they were generally able to provide contacts within the division that could provide further information. Apparently, the records coordinator's responsibilities were in addition to other full-time activities.

#### Plant Shift Superintendent Records

The Plant Shift Superintendent keeps a record of all accidents occurring on the K-25 Site. Because of time constraints ChemRisk was not able to review these records. However, major accidents would also be reflected in the Plant Quarterly Progress Reports.

#### Unindexed Records

Both the Compliance and Environmental Policy and Environmental Management Division document centers have a significant quantity of records that have not been indexed. Therefore, any review of these records would prove to be quite time consuming. ChemRisk conducted a cursory review of the unindexed records in the EMD center. This review revealed that these records were primarily correspondence, draft documents, and analytical data. Although it appeared likely that many of these records contain information that could be located elsewhere, a more thorough review of the unindexed records would be required to establish this with any certainty.



### 3.3 RECOMMENDED FOCUS AREAS OF LIKELY OFF-SITE SIGNIFICANCE

A number of areas with the potential for off-site release were described in Section 3.1. However based on the qualitative information reviewed during this feasibility portion of the Phase I Health Studies, a few processes were likely the most significant contributors to off-site releases. These focus areas are described in this section.

The main mission of the K-25 site was clearly the handling and production of various forms of uranium. The qualitative information reviewed to date suggest that the release of uranium from production and handling facilities should be the initial focus of any further investigation of off-site impact from K-25. The primary focus for the releases should be:

#### **Gaseous Diffusion Process**

The gaseous diffusion process operated continuously from 1945 to 1985. For those forty years, the purge cascade, (the gaseous diffusion process vent), was one of the primary airborne sources of uranium and technetium-99 releases from the diffusion plant (Lay and Rogers, 1986).

#### **Feed Facility and Product and Tails Withdrawal**

The Feed Facility (including uranium oxide conversion) and Product and Tails Withdrawal facilities handled uranium hexafluoride in the natural, enriched, and depleted stages. Over the years the facilities existed in several different buildings including K-306-7, K-131, K-413, K-631, K-1131, and the K-33 Feed Room. Based on the information in Table 3-2, releases from the product and tails withdrawal facilities and the feed facilities were responsible for approximately 60% of the total accidental uranium hexafluoride releases from the gaseous diffusion plant from 1946-1982. It is evident that these facilities were a major source of uranium releases from the K-25 site.

#### **Abnormal Releases**

Although the K-25 site did not have a criticality or other catastrophic accident, there were many instances of accidental uranium hexafluoride releases. The site kept what appears to be a fairly accurate record of abnormal uranium hexafluoride releases from the process facilities. These abnormal releases may prove to be quite significant when compared to routine releases of uranium from the purge cascade.

TABLE 3-2

**URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982**

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
04-28-45	K-305-5	UF <sub>6</sub>	9,200	Product	Product drain line valve bellows ruptured.
11-09-46	K-306-7	UF <sub>6</sub>	1,183	60.00	Release from cylinder valve during cap removal.
07-26-48	K-1405	Solution	1,492	NA	Failure to plug opening in equipment.
09-28-48	K-1303	Solution	2,132	NA	Corroded equipment leaked.
09-28-48	K-1405	UO <sub>2</sub>	4,517	NA	Powder blown to atmosphere.
04-29-49	K-1405	Solution	1,599	NA	Corroded tank leaked.
05-02-49	K-1405	Solution	1,592	NA	Corroded tank leaked.
09-04-49	K-631	UF <sub>6</sub>	45,081	Tails	Kerotest cylinder valve broke off near weld.
04-14-50	K-1405	UF <sub>6</sub>	9,200	NA	Cold trap leak due to defective valve.
04-17-50	K-1405	UF <sub>6</sub>	3,373	NA	Cold trap inlet valve leaked.
06-28-50	K-1405	UF <sub>6</sub>	122,668	0.71	Hydraulic rupture of over filled cylinder.
04-28-51	K-1405	UF <sub>6</sub>	28,046	Depleted	Valve leak on by-pass to exhaust stack.
06-03-51	K-1405	UF <sub>6</sub>	10,735	NA	Line leak when purged.
07-12-51	K-1405	UF <sub>6</sub>	3,067	0.71	Leaking gaskets in pilot plant.
07-02-51	Vault 16A	Solution	79,582	0.71	Leak from corroded drums.
07-20-51	K-1131	UF <sub>6</sub>	43,612	0.71	Leaking valve seat on purge line.
08-05-51	K-1131	UF <sub>6</sub>	61,334	Depleted	Fracture of cylinder end plate.
08-09-51	K-1405	UF <sub>6</sub>	12,269	0.71	Plugged line leaked when opened.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
08-24-51	K-602-4	UF <sub>6</sub>	1,227	0.49	Seal failure.
09-14-51	K-1405	UF <sub>6</sub>	3,401	0.71	Spilled from barrier tube.
09-22-51	K-631	UF <sub>6</sub>	14,008	Depleted	Thermowell leak on charge line to AC pump.
11-05-51	K-1405	UF <sub>6</sub>	3,067	0.71	Loose flange connection.
11-08-51	K-1405	UF <sub>6</sub>	3,401	NA	Unknown.
11-14-51	K-1405	UF <sub>6</sub>	3,066	0.71	Line cut to release plug.
11-21-51	K-402-8	UF <sub>6</sub>	1,534	0.52	Valving error.
01-03-52	K-631	UF <sub>6</sub>	1,165	0.69	Plugged line vaporized when opened.
01-16-52	K-413	UF <sub>6</sub>	3,665	0.71	Opened valve, safety cap removed.
04-01-52	K-1004A	UF <sub>6</sub>	1,108	0.45	Overfilled sample cylinder leaked.
05-19-52	K-631	UF <sub>6</sub>	1,533	4.83	Faulty gasket on cylinder valve leaked.
05-20-52	K-1401	UF <sub>6</sub>	7,666	0.71	Stokes pump casing burst.
05-27-52	K-1401	UF <sub>6</sub>	7,688	0.71	Stuck valve to Stokes pump.
06-52	Laboratory	NA	1,108	NA	NA.
07-18-52	Vault 16A	Solution	1,815	NA	Corroded drum leak.
08-52	Cascade	UF <sub>6</sub>	1,534	NA	NA.
08-52	Development	UF <sub>6</sub>	7,011	NA	NA.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
09-09-52	K-131	UF <sub>6</sub>	20,672	0.71	Ruptured feed pigtail.
09-19-52	K-1131	UF <sub>6</sub>	306,718	0.71	High pressure in cold trap due to HF <sub>2</sub> , caused rupture disk to release pressure.
10-03-52	K-631	UF <sub>6</sub>	15,335	NA	Bad gasket on pigtail connection.
10-06-52	K-1131	UF <sub>6</sub>	15,333	0.71	Ruptured cold trap tube.
10-10-52	K-631	UF <sub>6</sub>	4,600	0.43	Pressure instrument on feed line broke.
11-03-52	K-413	UF <sub>6</sub>	3,067	0.46	Cut into process line.
12-01-52	K-1303	Uranyl Nitrate	7,488	Enriched	Leak from product evaporator.
12-03-52	K-1301	UF <sub>6</sub>	1,626	0.43	Leak from cylinders on transfer operation.
12-29-52	K-306-7	UF <sub>6</sub>	1,534	Product	Ruptured bellows on a B-4 pump.
12-30-52	K-306-7	UF <sub>6</sub>	6,133	Product	Ruptured sample line.
12-30-52	K-402-1	UF <sub>6</sub>	768,520	0.56	Valve failure on a liquid feed cylinder.
12-52	Coded Chem.	Solution	1,626	NA	Leak from corroded drum.
01-08-53	K-131	UF <sub>6</sub>	59,193	Feed	Defective cylinder valve.
01-13-53	K-631	UF <sub>6</sub>	138,015	0.65	Hose blew off drain line.
04-15-53	K-631	UF <sub>6</sub>	1,534	0.65	Bearing failure on compressor.
05-10-53	K-631	UF <sub>6</sub>	15,336	0.65	NA.
05-10-53	K-633	UF <sub>6</sub>	6,133	NA	Cold trap vented to alumina trap which over loaded and vented to atmosphere.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
05-25-53	K-413	UF <sub>6</sub>	37,849	0.71	Cylinder exploded, oil UF <sub>6</sub> mixture.
06-01-53	K-1131	UF <sub>6</sub>	269,143	0.67	Valve retaining nut split, upper part of valve blown from body.
08-02-53	K-413	UF <sub>6</sub>	5,000	0.80	Release when alumina traps opened.
09-06-53	K-402-4	UF <sub>6</sub>	23,003	0.71	Valve opened in error.
09-07-53	K-131	UF <sub>6</sub>	3,067	1.21	Cylinder valve broke.
09-09-53	K-1401	UF <sub>6</sub>	5,521	0.71	Plugged line caused cylinder rupture.
09-13-53	Vault 16A	Solution	14,156	0.67	Corroded container leaked.
09-30-53	K-402-8	UF <sub>6</sub>	29,730	0.88	Cold trap heated up and vaporized UF <sub>6</sub> .
10-01-53	Vault 15A	Solution	3,288	0.67	Corroded drums leaked.
10-01-53	Vault 16A	Solution	7,339	0.84	Corroded drums leaked.
10-30-53	K-1131	UF <sub>6</sub>	15,335	0.67	Cold trap warmed up vaporizing UF <sub>6</sub> .
12-19-53	K-1131	UF <sub>6</sub>	3,067	0.71	Burned gasket on pigtail.
12-22-53	K-1131	UF <sub>6</sub>	2,147	0.71	Over pressure in F <sub>2</sub> scrubber.
12-29-53	K-101	UF <sub>6</sub>	3,067	21.62	Broken sight glass.
2-12-54	K-131	UF <sub>6</sub>	1,534	1.83	Pressure control valve ruptured.
3-8-54	Vault 15	Solution	3,149	0.67	Leak from corroded storage drums.
3-31-54	Vault 16A	Solution	22,047	0.83	Corroded containers leaked.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
4-5-54	K-131	Solution	55,987	0.75	Corroded drum leaked.
4-18-54	K-1301	Solution	1,198	0.92	Corroded drum leaked.
6-15-54	K-1401	UF <sub>6</sub>	1,000	0.71	Ruptured cylinder.
6-23-54	K-633	UF <sub>6</sub>	15,333	0.66	Pigtail broke on feed cylinder.
6-54	K-633	UF <sub>6</sub>	15,336	0.67	Leaked from valve when removed from system.
8-30-54	K-631	UF <sub>6</sub>	3,067	0.71	Seal change-out on Beach-Russ pump, leak from oil system.
10-10-54	K-1004L	UF <sub>6</sub>	3,067	0.65	Defective seal in pilot plant leaked.
12-22-54	K-413	UF <sub>6</sub>	3,067	1.30	Leaked from Beach-Russ pump.
3-10-55	K-33	UF <sub>6</sub>	144,815	0.71	Cylinder and pigtail explosion and rupture.
3-16-55	K-631	UF <sub>6</sub>	3,012	0.50	Seal failure on Elliott compressor.
4-19-55	K-633	UF <sub>6</sub>	15,333	Depleted	Copper feed line ruptured.
9-12-55	K-1420	Solution	2,756	0.44	Piped to holding pond in error.
10-30-55	K-33	UF <sub>6</sub>	1,840	PPF	Cylinder valve leak.
11-8-55	K-1131	UF <sub>6</sub>	3,067	0.65	Plugged line caused pressure blow back to tower.
11-20-55	Vault 16	Solution	1,555	3.47	Corroded drum leaked.
12-14-55	Vault 16	Solution	9,117	0.69	Corroded drum leaked.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
12-26-55	K-413	UF <sub>6</sub>	2,147	1.44	Cylinder valve leak.
1-15-56	Vault 15A	Acid Sol.	36,642	Depleted	Corroded drum leaked.
1-20-56	K-33 Feed Rm	UF <sub>6</sub>	1,533	0.71	Leak at valve threads on feed cylinder.
3-19-56	K-33	UF <sub>6</sub>	1,208	1.49	NA.
4-4-56	Vault 15A	Solution	1,734	0.88	Corroded drum leaked.
4-9-56	Vault 15A	Solution	2,571	0.86	Corroded drum leaked.
4-9-56	Vault 16A	Solution	4,886	2.87	Corroded drums leaked.
4-9-56	Vault 16A	Solution	1,897	1.74	Corroded drums leaked.
4-9-56	Vault 16A	Solution	11,583	2.61	Corroded drums leaked.
5-6-56	K-633	UF <sub>6</sub>	1,534	0.33	Blown gasket in test loop.
5-31-56	Vault 15A	Solution	1,286	0.86	Corroded drums leaked.
6-13-56	K-303	UF <sub>6</sub>	1,534	0.33	NA.
8-31-56	K-1131	UF <sub>6</sub>	30,672	0.67	Gasket on UF <sub>6</sub> pump failed.
9-09-56	K-1004L	UF <sub>6</sub>	3,066	0.71	Cylinder connection failed.
9-20-56	K-601	UF <sub>6</sub>	4,673	0.71	Rupture disk blown out.
12-56	Vaults 15, 16, 7A	Solution	40,752	1.65	Corroded drums leaked.
1-7-57	K-1131	UF <sub>6</sub>	1,840	0.67	Powder seal lost in process.
2-22-57	K-1131	UF <sub>6</sub>	1,534	0.65	Faulty cylinder valve.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
2-57	Vault 15A	Solution	39,682	NA	Leak from corroded drums.
10-10-57	K-413	UF <sub>6</sub>	3,041	1.51	Copper tube burned from a reaction in oil mist filter.
1-12-58	K-33 Feed	UF <sub>6</sub>	4,600	0.71	Faulty pigtail connection.
1-58	Vault	UF <sub>6</sub>	3,041	NA	Hole burned in mist filter.
1-25-58	K-631	UF <sub>6</sub>	4,600	0.39	Valve bellows rupture.
2-9-58	K-631	UF <sub>6</sub>	1,533	Depleted	Faulty pigtail connection.
3-20-58	K-33	UF <sub>6</sub>	22,080	0.71	Cylinder valve failure.
4-58	Vault	UF <sub>6</sub>	4,601	0.65	Valve bellows rupture.
9-2-58	K-1004L	UF <sub>6</sub>	7,666	0.71	Seal failure, pilot plant.
9-11-58	K-1004A	UF <sub>6</sub>	1,043	0.71	Faulty cylinder connection.
9-58	Development	UF <sub>6</sub>	7,666	0.65	Leak from pilot plant.
9-58	K-1131	UF <sub>6</sub>	374,212	0.71	Loss in transferring material.
9-58	K-1131	UF <sub>6</sub>	799,679	0.71	Ruptured filler in hydrogen reduction system.
2-27-59	K-1420	Acid Solution	2,819	0.67	Corroded base caused pump flange failure.
4-3-59	K-304-2	Solution	6,400	NA	Drain line leak.
6-15-59	K-1059	NA	2,105	0.62	NA
2-24-60	K-1131	UF <sub>6</sub>	214,960	0.71	Ruptured coil in E cold trap.



TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
2-27-60	K-1131	UF <sub>6</sub>	460,008	0.69	Ruptured tube in cold trap.
2-29-60	K-631	UF <sub>6</sub>	38,894	0.71	Cylinder valve leaked.
5-7-60	K-1131	UF <sub>6</sub>	184,003	0.65	Ruptured tube in F cold trap.
6-9-60	K-1420	UF <sub>6</sub>	4,453	1.50	Drain line leak.
1-2-61	K-631	UF <sub>6</sub>	1,533	Tails	Pigtail connection bad.
4-13-61	K-1131	UF <sub>6</sub>	3,067	0.71	NA
5-10-61	K-1004A	UF <sub>6</sub>	1,860	0.71	Valve bonnet broke off sample bottle.
5-13-61	K-1131	UF <sub>6</sub>	92,000	Depleted	Secondary cold trap tube rupture.
5-19-61	K-1131	UF <sub>6</sub>	6,100	Depleted	Secondary cold trap bypassed intermittently in trying to remove plug from process line; allowed release to vent stack.
6-3-61	K-33 Feed Rm	UF <sub>6</sub>	1,840	0.71	Purged valve left open in error.
7-26-61	K-1131	UF <sub>6</sub>	6,133	Depleted	Cracked pigtail.
9-29-61	K-1131	UF <sub>6</sub>	1,533	0.71	Jet assembly burnt out on reactor.
11-13-61	K-1413	UF <sub>6</sub>	193,200	Normal	Cylinder valve packing gland failed.
3-2-62	K-1413	UF <sub>6</sub>	NA	0.22	One-half inch process line ruptured.
3-1-62	K-1413	UF <sub>6</sub>	19,933	Depleted	Ruptured copper tubing.
3-16-62	K-1420	UF <sub>6</sub>	6,078	40.97	Cold trap valve failure.
3-2-64	K-1131	UF <sub>6</sub>	3,067	0.36	Bad gasket on pigtail connection.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
7-64	Vault	Solution	20,743	Enriched	NA.
11-23-65	K-631	UF <sub>6</sub>	4,600	Tails	Bellows rupture on transmitter.
8-67	K-1025	UF <sub>6</sub>	1,239	72.00	Leaking cylinder in storage.
1-68	NA	UF <sub>6</sub>	1,589	0.25	Tails rupture test.
5-5-69	K-1423	UF <sub>6</sub>	6,746	0.71	Valve seat on cylinder leaked.
5-24-70	K-33 Feed Rm	UF <sub>6</sub>	4,600	1.49	Control valve bellows in feed header cracked.
9-26-70	K-413	UF <sub>6</sub>	1,533	NA	Drain line leakage.
12-16-70	K-1131	UF <sub>6</sub>	153,314	0.71	Probable pigtail leak. Material contained in autoclave drained to Popular Creek.
4-26-71	K-1401	UF <sub>6</sub>	11,040	0.71	Copper Pigtail ruptured.
8-24-71	K-311-1	UF <sub>6</sub>	7,663	3.40	Casing of Cell 5--1B pump ruptured.
11-25-73	K-633	UF <sub>6</sub>	3,066	0.33	Block valve leak to air ejector.
7-16-73	K-1004A	UF <sub>6</sub>	3,600	0.71	Ruptured valve on sample cylinder.
9-17-75	K-1423	UF <sub>6</sub>	5,519	3.25	Cracked valve and cylinder wall (Nukem 8).
3-77	K-413	UF <sub>6</sub>	3,070	2.90	Accidental release, pigtail pinch-off incomplete.
5-77	K-33	UF <sub>6</sub>	1,533	0.79	Accidental release, leak around pump exhaust gasket.

TABLE 3-2  
(Continued)

URANIUM MATERIAL RELEASES ONE KILOGRAM OR GREATER  
1945 - 1982

Date	Plant Location	Material Type	Uranium Grams	Assay Wt. % U-235	Cause
3-19-78	K-1210	UF <sub>6</sub>	4,302	NA	Gasket failure from overheating.
7-28-80	K-1413	UF <sub>6</sub>	5,827	0.71	Valving error involving an MD cylinder.
12-01-80	K-1302	UF <sub>6</sub>	1,533	3.18	Stack release caused by valving error.
5-27-81	K-29	UF <sub>6</sub>	2,000	3.2	Release from cell following high temperature reaction.

NA = Not Available

Source: Legaey, G. 1982.

Two other areas that are likely to have contributed significantly to off-site releases are the **Liquid Waste Disposal** and the **S-50 Thermal Diffusion Plant**, these areas should also receive priority in investigations to quantify historical contributions to off-site exposures.

**Liquid Waste Disposal** is another area of plant activity having the potential for significant off-site transport of contaminants. The liquid waste streams from the many K-25 facilities represent an extremely complex source to evaluate. However, the majority of the liquid wastes were ultimately discharged off-site via the K-1407B holding pond. Any further investigation of off-site impacts of this liquid waste stream should begin by focussing on the evaluation of releases from this point of release.

Initial efforts to review the **S-50, Liquid Thermal Diffusion Plant** suggest that the operation of this facility and support buildings may also warrant some further investigation to determine its potential for offsite release of contaminants. Although this facility only operated from September 1944 to September 1945, most accounts of the plant operations detail many mechanical problems, especially with leakage of process fluids (Rhodes, 1986; Jones, 1985; Legeay, 1993). In fact one source indicates that at times there was as much as 3,000 pounds per month of uranium for which the plant could not account (Dwyer, 1945).

### 3.4 INFORMATION AVAILABILITY FOR DOSE RECONSTRUCTION AND FURTHER QUANTITATIVE EVALUATION

In order to support the direction of any future efforts to quantify doses and health risks associated with contaminants released from the K-25 site, information that is available concerning the identified high priority areas for potential off-site releases has been reviewed. This section describes the information that has been identified during the Phase I feasibility study historical investigations.

#### **Gaseous Diffusion Process**

Because the gaseous diffusion process was enclosed and operated below atmospheric pressure the purge cascade was, historically, the only portion of the process which was monitored for uranium hexafluoride or technetium-99. From 1946 to 1985, the Operations Division was responsible for monitoring uranium effluents from the purge cascade. In the early years, uranium accountability was the primary reason for monitoring the purge cascade; there was not an interest in monitoring for gases or chemicals being vented through the purge cascade. Raw data for uranium concentrations in the purge gases dating back as far as September 1945 were located in the Shift Superintendent Logs located in the Site Records Vault (Box H,H,5,7,37). From 1947 to 1954, the K-25 Site Quarterly Progress Reports (Section 3.2.2.7) reported the average total purge rate per day and the average uranium hexafluoride concentration in the purge gases. Beginning in 1955, the Quarterly Progress Reports contain a section entitled Nuclear

Materials Management, this section includes an Accounting Memorandum table. This table, provides a summary of the disposition of the uranium material that was unaccounted for during a particular quarter. The table has estimates for the amount of uranium deposited in the cascade, in the holding ponds, in Poplar Creek, on the ground near the Feed Plant, on drums or cylinders, and the amount of uranium vented to the atmosphere. At this time, it is not clear if the reported value for the amount of uranium vented to the atmosphere is a total for the purge cascade only, or if effluents from other K-25 buildings are a part of the reported total. Technetium-99 was monitored in the purge cascade effluents beginning in 1974, and data are reported in the Plant Quarterly Progress Reports, as well as, the site Historical Release Reports.

Because the diffusion cascade was an enclosed process that operated below atmospheric pressure, the likelihood of fugitive emissions was low. However, at least one interviewee indicated that occasional leakage, due to seal failure within the cascade may have occurred (Widner et al., 1992). Information on ventilation flow in process buildings to support the analysis of fugitive releases can be found in the Engineering Design Information System (Section 3.2.1). The engineering drawings provide historical information about the ventilation system of each building.

Other information sources that may be helpful in source term evaluation are the operating manuals for the early cascade operations. These manuals are available at the Site Records vault, Box G,3,6,33. This box contains Kellex Research Files which include 12 volumes of an operating manual series for the K-25 site. Below is a list of the available operating manuals:

- Control Room
- Process Control
- C-616 Recovery
- Case I Operation
- Mass Spec. and Line Recorder
- Surge and Waste System
- Case II Operation
- Case III Operation
- Purge Cascade
- Case IV and V Operations
- Space Recorder

Although the monitoring data for the gaseous diffusion are not complete, there appears to be enough uranium accountability information to develop a source term for uranium effluents from the gaseous diffusion process. Source term development for other chemicals and radionuclides emitted from the process would likely be considerably more difficult.

### **Feed Facility and Product and Tails Withdrawal**

Beginning in the mid-1950s selected vents from the K-1131 and K-413 buildings were monitored for uranium, hydrogen fluoride and fluorine. A complete set of monitoring data was not located, however, isolated time periods of monitoring data are available in Box 6A,8,9,51 at Site Records.

Additionally, these facilities had many abnormal releases. The Quarterly Progress Reports detail "abnormal release" during each quarter of operation. Additionally, in the uranium accountability sections of the Quarterly Progress Reports there are estimates of the amount of material "lost" to the ground around the feed plant.

As previously discussed, health physics and industrial hygiene indoor air sampling data are available for these facilities.

### **Abnormal Releases**

In part because of the stringent uranium accountability procedures, the K-25 facility kept a fairly accurate record of uranium hexafluoride abnormal releases. Table 3-2 presents a list of uranium material releases over 1 kilogram. This list was compiled in 1982 by a former K-25 employee. The list was compiled from the Plant Quarterly Progress reports which contained a section for documentation of abnormal releases over a specific quarter. In general, this section not only listed the estimated release, but also provided a short explanation on the cause of the release.

### **Liquid Waste Disposal**

The uranium concentrations in the discharges from the K-1420 process building, (the major source of uranium in liquid effluents) were monitored, as were the uranium concentrations discharges in the K-1700 stream at the outfall to Poplar Creek.

The uranium concentration data for discharges from the process buildings to the K-1407B holding pond, which discharged to the K-1700 stream, are available in the Uranium Accountability sections of the Quarterly Progress Reports. This section reports the quantity of uranium discharged to the K-1407B settling pond.

Prior to 1971, the compiled waterborne effluent for the K-1700 stream outfall data are located in the Plan Quarterly Progress Reports and the Health Physics Activities Monthly Reports. After 1971, compiled data are available in the Annual Environmental Surveillance documents. Beginning in the mid-1970s analytical data records for the NPDES discharge points at the site are available at Site Records (Row F, Tier 3). Although some of the monitoring data will be

useful in evaluating source terms for uranium and fluorides, inventory information and environmental monitoring data may be required in order to develop a source term for other chemicals and radionuclides.

### S-50 Liquid Thermal Diffusion

Two factors make it difficult to locate documentation regarding the S-50 site. First, the site was constructed and operated by the Fercleve Corporation which had no obligation to file reports with the K-25 site. The second factor is that the site was shutdown nearly 50 years ago. The investigation of the document sources discussed in Section 3.2 revealed little documentation of the activities at the S-50 plant. Currently, ChemRisk is working with K-25 plant historians to determine whether or not documentation of S-50 operations might be available at the DOE Archives near Washington, D.C.

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#### 4.0 THE Y-12 SITE

The Oak Ridge Y-12 Plant is a government-owned nuclear weapons facility, currently operated by Martin Marietta Energy Systems, Inc. under contract with the U.S. Department of Energy. The discussion of historical operations and releases in this report presents an overview of important activities including routine operations, special projects, and accidents or incidents. As part of Task 1 of the feasibility study, ChemRisk has identified Y-12 activities that may be associated with off-site releases. The available documentation and information resources that would support further quantitative evaluation of historical releases from these operations is also summarized.

Discussion of Y-12 activities in this report are presented with varying levels of detail, which is primarily a function of the quality and quantity of information that was uncovered during the investigation process. The availability of information is a function of factors such as the scheduled, routine destruction of records, the inability to find or identify relevant records during the feasibility study phase of the project, or the fact that some processes were not extensively documented and therefore the information is limited.

This section is written to serve as a working document for further investigation in that it identifies key documents and information resources that would be valuable in characterizing historical off-site releases of important chemicals and radionuclides from the Y-12 Plant.

#### 4.1 SUMMARY OF HISTORICAL ACTIVITIES AT Y-12

This section describes historical operations and materials utilized at the Y-12 Plant over the time period from 1943 to 1992. The Y-12 Plant has used numerous materials throughout the last 50 years in conducting its production, research and development, and support activities for the U.S. government. The following discussion describes how some of these materials were used in fulfilling Y-12's original mission, the evolution of the plant into a nuclear weapons component fabrication plant, ORNL activities located at the Y-12 site, engineering design and testing support activities, special projects, and waste management activities. The discussion is not intended to be a comprehensive presentation of all activities that have occurred at the Y-12 site, but rather attempts to identify activities that could plausibly have been associated with off-site releases of hazardous materials. Sections 4.3 and 4.4 identifies those activities that may warrant further investigation from the standpoint of historical releases.

##### 4.1.1 Site Description and Operations Summary

The Oak Ridge Y-12 Plant, located in Oak Ridge, Tennessee, was built for the U.S. Army Corps of Engineers in 1943 as part of the Manhattan Project under the name Clinton Engineer

Works. The Y-12 Plant is located at the eastern end of the Bear Creek Valley. The facility is within the corporate limits of the City of Oak Ridge and separated from the city by Pine Ridge. The plant is located approximately 1/2 mile from the center of the City of Oak Ridge. The plant is bordered on the southern side by Chestnut Ridge and on the north side by Bear Creek Road and Pine Ridge (UCC-ND, 1983). The Scarboro community lies just north of Pine Ridge, with separation from Y-12 buildings of as little as 0.3 mile. The main production area is about 0.6 miles wide by 3.2 miles long and covers roughly 825 acres, 600 acres of which are enclosed by a perimeter security fence. The plant and its fenced buffer area total about 4860 acres. A map of the plant site is shown in Figures 4-1 and 4-2. The site contains roughly 240 principal buildings with about 6.5 million square feet of floor space. Approximately 25% of this floor space is utilized by ORNL staff and operations; the remaining portion is dedicated to Y-12 operations, of which 66% is allocated to production-related functions. The remaining space is devoted to offices and various support activities (UCC-ND, 1983).

Tennessee Eastman, a subsidiary of the Eastman Kodak Company, was the original Y-12 site contractor under agreement with the U.S. Army Corps of Engineers. In accordance with the Atomic Energy Act of 1946, on January 1, 1947, all atomic energy activities including the Y-12 facility were turned over to the United States Atomic Energy Commission (U.S. AEC). During that same year, the Manhattan Engineer District disbanded and Tennessee Eastman was replaced by Carbide and Carbon Chemicals Corporation as the Y-12 site contractor. The name "Y-12" was a code name given to the plant during the Manhattan Project and has not changed since that time. The following list summarizes changes in the prime site contractors throughout Y-12's history.

<u>Operating Contractor</u>	<u>Time Period</u>
Tennessee Eastman, Subsidiary of Eastman Kodak Company	1943 to early 1947
Carbide and Carbon Chemicals Corporation	1947 to 1950
Carbide and Carbon Chemicals, Division of Union Carbide and Carbon Corporation	1951 to 1956
Union Carbide Nuclear Company, Division of UCCC	1957 to 1963
Union Carbide Corporation, Nuclear Division	1963 to 1984
Martin Marietta Energy Systems, Incorporated	1984 to present

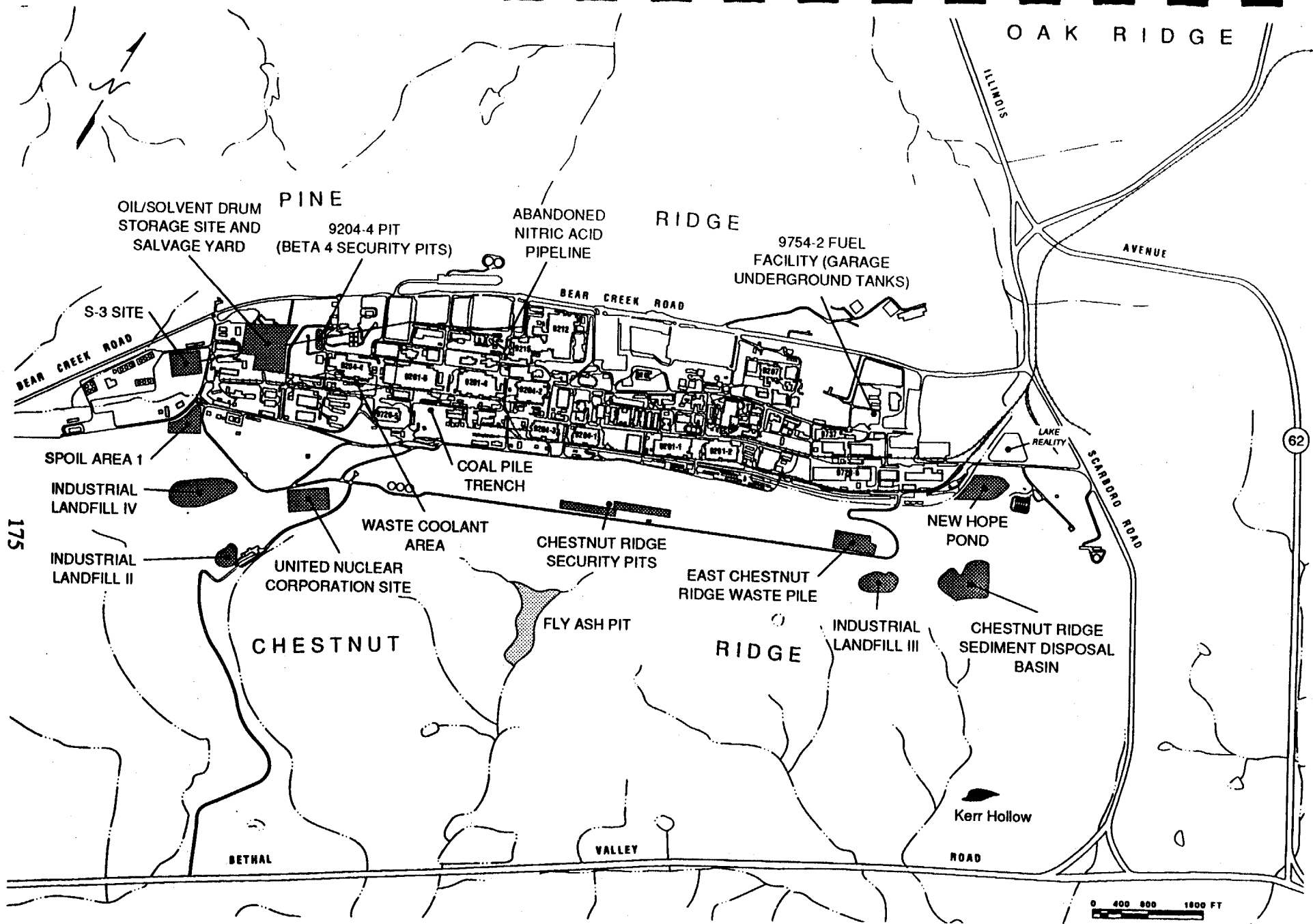


FIGURE 4-1  
 MAP OF THE Y-12 PLANT SITE-  
 MAIN PRODUCTION/SUPPORT AREA

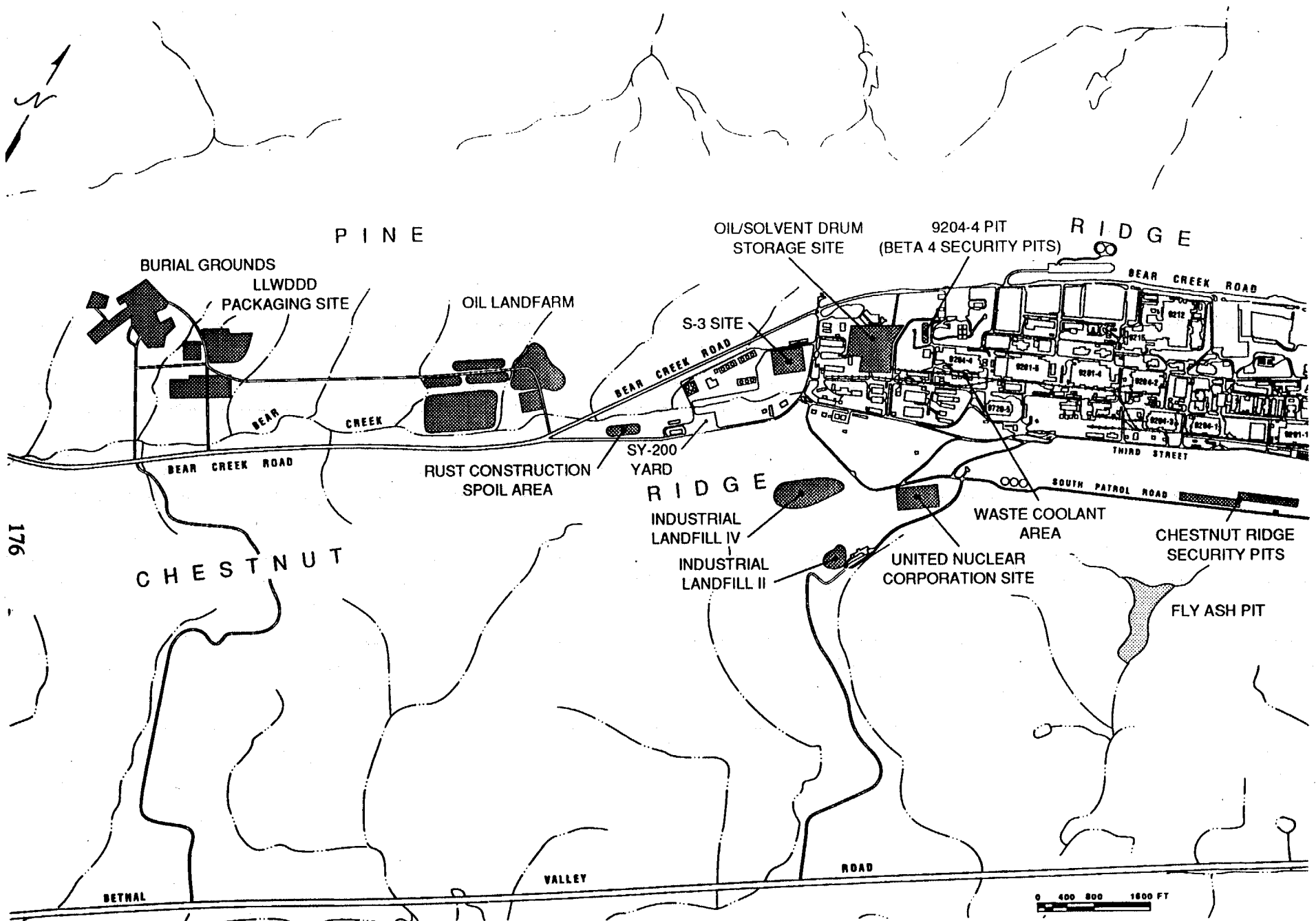


FIGURE 4-2  
 MAP OF THE Y-12 PLANT SITE-  
 WESTERN END OF THE PLANT

The Y-12 Plant had five principal responsibilities:

- Development of the electromagnetic separation process for uranium,
- Production of nuclear weapon components,
- Fabrication support to nuclear weapon design agencies,
- Support for the Oak Ridge National Laboratory, and
- Support to other governmental agencies and facilities.

Important materials associated with the above activities have included enriched uranium, depleted uranium, uranium alloys, beryllium and lithium compounds, specialty steels, lead, precious metals, niobium, tantalum, and other refractory metals, thorium, plutonium, mercury, and zirconium. Routine operations involving some of these materials have included chemical recycle and purification, melting, casting, forging and rolling, grinding, and highly precise machining of sophisticated weapon components. Although changes have occurred over the years in the production and fabrication of end products, uranium of various isotopic composition has been the major process material at Y-12 (UCC-ND, 1983).

Other missions at Y-12 included fabrication support to the DOE weapon design facilities at the Los Alamos Scientific Laboratory (LASL), the Lawrence Livermore National Laboratory (LLNL), and the Sandia National Laboratories (SNL). For these laboratories, Y-12 produced components for design evaluation and nuclear test devices fired at test sites such as the Nevada Test Site. Y-12 also performed uranium recovery and recycle from reactor fuel returns received from DOE's Savannah River Plant (SRP) in Aiken, South Carolina, and Idaho Chemical Processing Plant (ICPP) in Idaho Falls, Idaho. Also, several ORNL activities carried out at Y-12 have included those of the ORNL Biology Division, Engineering Technology Division, Stable Isotope Production, and Fusion Research.

Y-12's original mission in supporting the World War II effort was the separation and enrichment of the fissile isotope of uranium-235 (U-235) from natural uranium by the electromagnetic process. This effort, involving the use of approximately 1200 electromagnetic separation units and many support buildings, required an original investment of approximately \$550 million and a maximum operating workforce of 24,000 people (Gosling, 1990). Electromagnetic separation was one of three techniques used for uranium-235 enrichment operated under the Manhattan District project. The others were liquid thermal diffusion and gaseous diffusion (Compere et al., 1991).

Several other important materials have been used to support Y-12 weapon component fabrication and assembly operations. Starting in the early 1950s, nuclear weapons production increased dramatically and modifications to weapon designs required different mixtures of uranium and other materials to be processed at the Y-12 Plant. To support some of these changes Y-12,

around 1950, embarked on a large-scale lithium isotope separation and enrichment process requiring large quantities of mercury (Wilcox et al., 1983b). Similar to the World War II uranium enrichment effort, the lithium separation project required a crash construction program, the development of new technology, and overcoming numerous technical difficulties.

Research and development at Y-12 has been an important function provided by the plant. Much of the focus has been to provide engineering design and testing support to Y-12's production activities. Their first important contribution to the plant was the development and improvement of process chemistry and material recycle during the early electromagnetic separation and enrichment program. This same type of support was continued on into the weapon component manufacturing operations at Y-12 and extended to other DOE facilities and government agencies such as the military.

A chronological listing of major programs and activities at Y-12 is presented in Table 4-1.

#### 4.1.2 Electromagnetic Separation and Enrichment Operations

##### The Original Mission

In 1940, Alfred O. Nier of the University of Minnesota used a mass spectrometer to send a stream of charged particles through a magnetic field. Atoms of the lighter isotope were deflected more by the magnetic field than those of the heavier isotope, resulting in two different streams that were subsequently collected in different receivers (Gosling, 1990). In early 1942, the Radiation Laboratory at the University of California at Berkeley, headed by Professor E.O. Lawrence, demonstrated that it was possible to separate and enrich uranium-235 from the naturally more abundant isotope uranium-238 by converting a 37-inch cyclotron to a production mass spectrometer, later to be called a **calutron** (Compere et al., 1991).

The electromagnetic method as it existed in 1940, however, would have taken twenty-seven thousand years in a single spectrometer to separate one gram of uranium-235 (Gosling, 1990). Further research continued, and in the fall of 1942, Lawrence and his staff refined the design of the first Y-12 calutron. By December, Lawrence, General Leslie Groves, and the general contractor Stone and Webster agreed on the general size and materials required to build the Y-12 electromagnetic facilities (Compere et al., 1991). The revised conceptual designs included large numbers of electromagnetic separation units in order to produce sufficient quantities of enriched uranium in a short time. Since the war effort required kilogram quantities of enriched uranium, the new facilities were designed to support multiple banks of calutrons capable of enriching large quantities of uranium. Highly enriched uranium produced at Y-12 was later used in the "Little Boy" bomb dropped on Hiroshima, Japan, on August 6, 1945.

TABLE 4-1

CHRONOLOGY OF MAJOR PROGRAMS AND ACTIVITIES AT THE Y-12 SITE

Electromagnetic Separation of Uranium-235	1943 - 1948
Electromagnetic Separation of Stable Isotopes	1947 - 1990
Production of Uranium Weapon Components	1948 - 1992
ELEX Separation of Lithium Isotopes (Using Mercury)	1950 - 1956
Production of Thorium Weapon Components	1950s - 1975
Waste Disposal in S-3 Ponds	1951 - 1990
Production of Lithium and Beryllium Weapon Components	1950s - 1992
COLEX Separation of Lithium Isotopes (Using Mercury)	1956 - 1963
Waste Disposal in New Hope Pond	1963 - 1990



On February 18, 1943, under the direction of the Manhattan Engineering District of the U.S. Army Corps of Engineers, construction began on the Y-12 facility. Secluded in the hills of eastern Tennessee, this site seemed well suited for a highly secret project and provided adequate protection in the event of a serious accident (Jones, 1985). The site was remote, yet readily accessible by rail or water and had adequate power supply from the Tennessee Valley Authority (TVA). For the Y-12 Plant, the initial wartime effort resulted in 18 plant process buildings being built between 1943 and 1946. Most of these buildings were constructed along the East Fork Poplar Creek. A listing of initial dates of operation of key buildings is presented in Table 4-2. The urgency of this mission to develop an atomic bomb required a large workforce and specialized training in handling enriched uranium using new and developing technology under trial and error conditions. The physical theory, process chemistry, and production design for each building were developed concurrently with construction and operation, so that each group of units served as a pilot for the next group (Compere et al., 1991). These buildings, referred to as individual "plants," housed the electromagnetic separation equipment, chemical processing and recycle, and uranium salvage operations.

The original mission of the Y-12 plant was highly classified, as have been most of its subsequent missions. It was kept a secret from most plant workers and known by only those who had a need to know or those who could surmise what Y-12 was manufacturing (Googin, 1993). It was not until the first atomic bomb had been dropped on Japan that the workers at the plant and the rest of the world finally knew what Y-12's massive production and development effort had been set out to accomplish. The calutron units used to separate uranium-235 from uranium-238 were highly protected from espionage during the war and were given, as part of that effort, the code name "D" units (Overholt, 1987). The name "D" represented the 180° shape of the calutron. The uranium that was fed to the calutron units was also considered highly secret and therefore given the code name "T" material (Overholt, 1987). The name "T" referred to tuballoy, a name given to natural or normal uranium feed material. Most aspects of early Y-12 operations were kept highly secret and were not declassified until a number of years later (Compere et al., 1991; Googin, 1993).

Early operations of the calutrons were plagued with equipment and material problems which forced frequent shutdown of operations, often times requiring rework of the production equipment. Some of the problems included contamination of process oil and loss of electromagnetic field strength due to dust and oil contamination on the large separation magnets (Compere et al., 1991). Most of these problems were overcome and by the end of February 1944, 200 grams of 12% uranium-235 product material had been produced and sent on to Los Alamos and some retained for further enrichment, often called "beta" feed material (Gosling, 1990). Y-12 would go on to process thousands of pounds of uranium to obtain sufficient quantities of enriched uranium for the highly secret project. Electromagnetic separation was the only process that produced enriched uranium for the atomic bomb used during World War II, and by June 1947, Y-12 had halted all uranium enrichment operations and put the equipment in

TABLE 4-2

## COMPLETION DATES FOR Y-12 PLANT PROCESS BUILDINGS

Building Number	Date of Initial Operation	Use
9202	October 1943	$\alpha$ and $\beta$ chemical preparation and initial chemical recovery processing
9203	October 1943	$^{235}\text{U}$ analysis (mass spec.), control analyses, and initial product processing
9205	October 1943	Uranium isotopic analysis
9201-1	January 28, 1944 March 3, 1944	Track 1, $\alpha$ - I calutrons Track 2
9204-1	March 15, 1944 June 30, 1944	Track 1, $\beta$ - calutrons Track 2
9201-2	March 20, 1944 April 13, 1944	Track 3, $\alpha$ - I calutrons Track 4
9201-3	June 15, 1944	Track 5, $\alpha$ - I/5 calutrons
9201-4	July 27, 1944 August 27, 1944	Track 6, $\alpha$ - II calutrons Track 7
9204-2	September 12, 1944 November 2, 1944	Track 3, $\beta$ calutrons Track 4
9201-5	September 12, 1944 October 28, 1944	Track 8, $\alpha$ - II calutrons Track 9
9206	November 1944	$\beta$ chemical recycle and product processing
9733-3	November 1944	Chemical control analysis
9204-3	December 13, 1944 January 30, 1945	Track 6, $\beta$ calutrons Track 5
9207	January 1945	Uranium salvage
9733-4	February 1944	Chemical control analysis
9212	November 1945	$\beta$ product processing
9204-4	November 15, 1945 January 1946	Track 8, $\beta$ calutrons Track 7
9211	January 1946	Uranium salvage

Source: Compere, 1991.

a standby mode for future needs. The entire Y-12 uranium enrichment program for producing the first atomic bombs lasted from 1944 to 1947.

The following were the four primary steps in the Y-12 enrichment process:

- Preparation of uranium feed material,
- Electromagnetic separation of the feed material,
- Uranium recovery, purification and recycle, and
- Uranium salvage operations.

Eventually, most of the equipment was disassembled and the technology of the electromagnetic separation process was declassified and widely disseminated. Since the war, continued research and development efforts focussed on improving the efficiency and capability of the calutron separation units, many of which have been located in some of the original production buildings such as 9201-4, 9204-3, and 9731 (Livingston, 1948; Salvage et al., 1950; Kite, 1985; Hemphill, 1950). Some of the equipment and operations have remained in operation, even today, for the purposes of separating stable isotopes. Copper and chromium were some of the first important isotopes separated with this equipment (Salvage et al., 1950; Compere et al., 1991). The separation of stable isotopes, managed by ORNL, became an important program in identifying and studying important physical and chemical characteristics of isolated (enriched over natural abundance) isotopes of various elements.

#### Electromagnetic Separation Equipment

The calutrons (derived from the names University of California and cyclotron) were production mass spectrographs with essentially the same operating principles of a mass spectrometer (Jones, 1985). The calutrons were primarily constructed of stainless steel, copper, graphite, and ceramic components. Large magnets which provided the magnetic focussing field were wound in silver and positioned between back-to-back calutron tanks. Y-12 borrowed almost 15,000 tons of silver from the United States Treasury (Gosling, 1990). The silver was shipped to outside manufacturing facilities, where it was fabricated into magnetic coils and then later shipped to Y-12. A highly precise magnetic field was required for proper operation of the calutrons. It was reported that a deviation of 0.6 % could result in the collection of the wrong isotope. To prevent this from happening, monitoring and magnetic regulation circuits were installed to maintain uniformity across the magnetic fields (Compere et al., 1991).

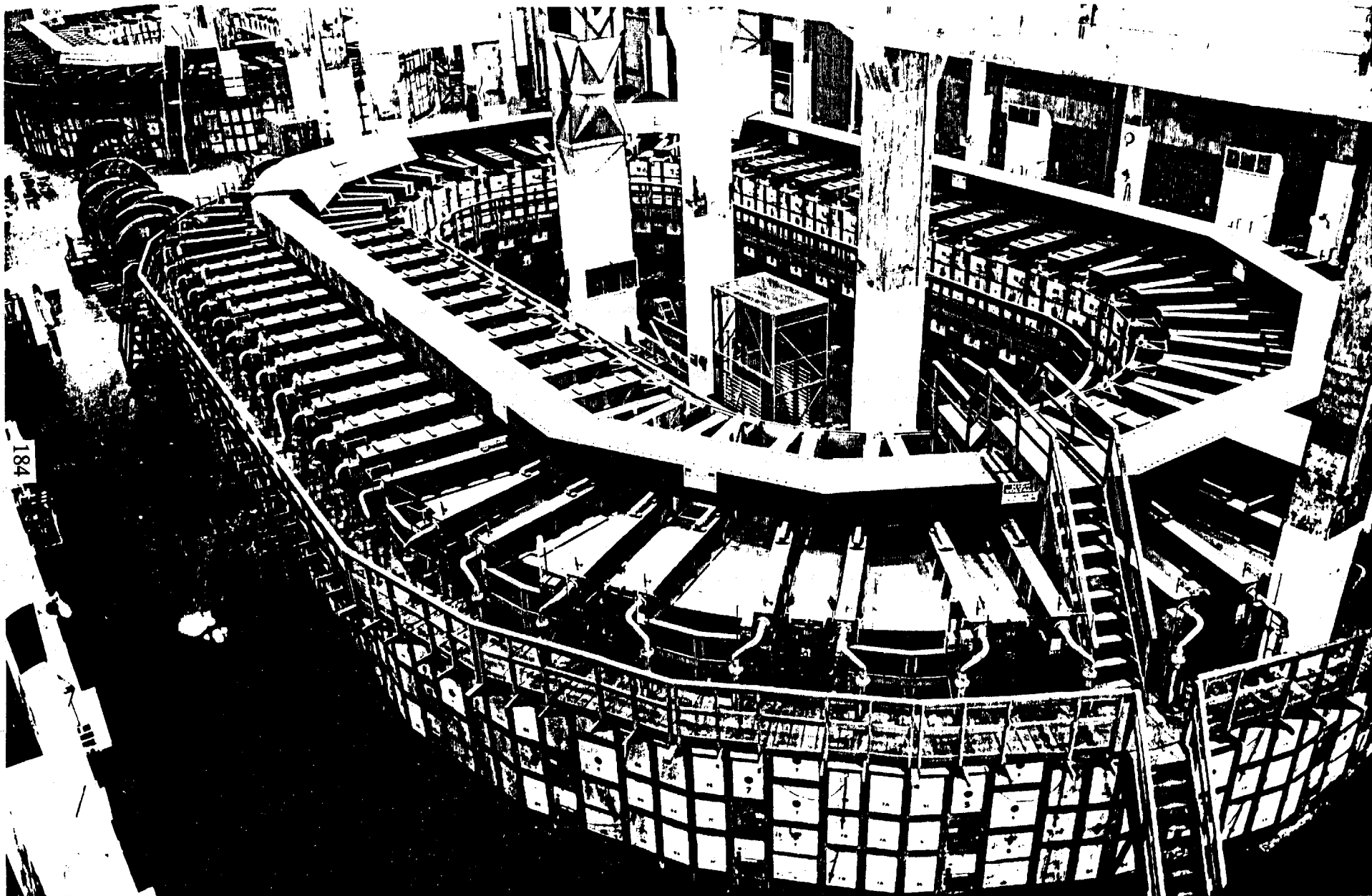
The production-scale separation and enrichment of uranium-235 necessary for use in an atomic bomb required two separate types of dual-ion source calutrons, an "alpha" unit for the first stage enrichment, and "beta" unit for the second or final stage unit. These were very complex assemblies with many intricate parts. The original alpha calutron, which was the first of two types of alpha units to be built and used at Y-12, was designed to process large quantities of

normal uranium feed material. These were called alpha-1 calutrons. Normal or naturally-occurring uranium consists primarily of 99.3% <sup>238</sup> isotope and 0.7% uranium-235 isotope and was prepared as uranium tetrachloride for use in the calutrons. Large numbers of alpha-1 units had to be used because of their poor collection efficiency of uranium-235, which was usually less than 10% (Compere et al., 1991). The second alpha unit was called alpha-2 and is discussed below. Beta separation units were the final stage in the enrichment process and capable of enhancing uranium-235 levels to over 90% of the total uranium content. This beta-enriched uranium was commonly referred to as the "top product", and had the desired enrichment for use in nuclear weapons (Harmatz et al., 1948). In separate buildings, the alpha and beta units were arranged in multiple banks called "racetracks"; the units were arranged in pairs, back-to-back, either in an oval or a rectangular shape, and the buildings were referred to as alpha or beta buildings. Figure 4-3 depicts an oval-shaped racetrack arrangement of 96 operating alpha-1 calutrons. Each calutron unit was separated by a large magnet and other than sharing a common magnet, these side-by-side units operated independently of each other. Rapid construction continued, and eventually Y-12 ended up with five alpha-enrichment buildings and four beta-enrichment buildings (Compere et al., 1991). The alpha buildings were designated by special numbers (e.g. 9201-1, 9201-2, etc.). The beta buildings were designated 9204-1, 9204-2, etc.

In the fall of 1942, J. Robert Oppenheimer, who was managing the construction of the first atomic bomb at Los Alamos National Laboratory (LANL), reported that more fissile material would be required to build the bomb (Gosling, 1990). The original alpha unit was only capable of enrichments up to roughly 20% uranium-235, and the throughput with the two-ion-source unit was too limited for the increased production requirements (Compere et al., 1991). In order to meet these new goals, Berkeley was forced to design a second unit, called alpha-2, which employed four ion sources, doubling the throughput of the earlier alpha-1 units. The alpha-2 units were then added to Y-12 operations specifically to meet Oppenheimer's revised requirements. These enhanced alpha units were faster at preparing uranium feed sufficient in uranium-235 levels for the beta separation process, and as a consequence, resulted in dramatic increased production capabilities (Jones, 1985). Y-12 workers were asked to work harder and longer to complete this secret mission. The typical workers who operated the calutrons were local women who had graduated from high school (Overholt, 1987).

The following passage from a former Y-12 worker is real evidence of the continual effort by the Y-12 workers and others to enhance the process efficiency and throughput of the calutrons:

"... One evening Professor E. O. Lawrence from Berkeley came by the XBX (area which housed a Beta calutron track) operating cubicle to observe an experimental run. We had already started and everything was going smoothly. The collection meters on the



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FIGURE 4-3  
AN ALPHA-1  
CALUTRON RACETRACK

U-235 collectors indicated a steady reception of the desired isotope. We were quite content to let the run continue without interruption. Seeing this, Professor Lawrence sat down at the controls. He began working the knobs, turning up the accelerating voltages, increasing the temperature of the oven containing the uranium tetrachloride charge material. As we watched him, he gently increased reception at the collectors, edging it higher and higher. The run changed from one of smooth, low productivity to one of sparking unsteadiness, one requiring continual attention. But the production rate of U-235 increased greatly. Professor Lawrence personified a more aggressive approach to our experiment, attempting something we should have been trying. He then prepared to leave, but not without first admonishing us to do everything possible to increase production, even to the destruction of the equipment. We were deservedly humiliated. Shortly after Professor Lawrence left the building, the equipment in the vacuum tank literally erupted; vacuum was lost and the run automatically terminated itself. We followed customary procedure: removing all equipment from the tank, examining it, and determining the cause of the failure. It was obvious: wild sparking had broken heavy ceramic insulators supporting the high voltage ion source which propelled the uranium ions from source to isotopes receiver. Our next job was to try to find a way to prevent this heavy sparking or, at least, to protect the insulators from being bombarded. This is what we should have always been doing, and through experimental failure we could and did find improved methods."

(Overholt, 1987; p.71)

The calutrons were operated under heated vacuum conditions which were required to maintain proper separation of the uranium isotopes. The vacuum conditions also provided the extra benefit of controlling airborne releases of uranium, although some losses of uranium did occur when vacuum was lost in the system, generally these were fairly small losses (Googin, 1993).

After the alpha feed, consisting of natural uranium, was sent through the first stage of enrichment, every part including bolts and screws inside the calutron had to be disassembled and cleaned to remove the uranium and prepare it for recycle. The further along the material got in the enrichment process, the more valuable it became and the more critical it was to recover as much of the uranium as possible (Compere et al., 1991; Googin, 1993). As a consequence, routine releases of uranium were more significant during the initial, alpha preparation and recovery stage than those losses that occurred during the beta stage. Loss of material to process components, building structures, and the outside environment during the beta processing was far

more detrimental to the production schedule and the war effort and, as a result, the objective was to keep losses to a minimum (Compere et al., 1991; Griffith, 1957).

The calutrons continued to enrich uranium for atomic bomb production until shortly after the war. On September 22, 1945, the alpha separation units, which had produced the low-level enriched uranium-235 material for the beta stage, were shutdown and replaced by a supply of partially-enriched feed material from the K-25 Gaseous Diffusion Plant. K-25 material became the feed material for the final stage of the enrichment process at Y-12 (Compere et al., 1991). The flow of uranium feed material into the enrichment process and uranium recovery for both alpha and beta operations is depicted in Figure 4-4. Eventually all the calutrons used for enriching uranium were shutdown and placed in a stand-by mode. The K-25 Plant and, later on, other DOE gaseous diffusion plants were capable of supplying Y-12 with sufficient quantities of weapon-grade enriched uranium.

#### Feed Material Preparation

The preparation of uranium feed and the subsequent recovery and recycle of the uranium from the calutrons were carried out in numerous small batches under large laboratory-type equipment and conditions. These processes played a vital role in the enrichment process.

By October of 1943, Buildings 9202, 9203 and 9205 were completed. The starting material shipped to Y-12 was natural uranium primarily in the form  $UO_3$ . These buildings were used to convert uranium trioxide ( $UO_3$ ) into uranium tetrachloride ( $UCl_4$ ) which was then loaded into calutrons and electromagnetically separated. Once a run was completed, uranium was recovered and recycled for further enrichment or reduced to uranium hexafluoride for shipment to Los Alamos. The  $UCl_4$  feed material was commonly referred to as "charge material" and was loaded into specially-designed, stainless steel containers called "charge bottles" (Compere et al., 1991). The  $UCl_4$  feed material was generally in a crystalline form as it was loaded into the calutron tanks. Y-12 used large quantities of carbon tetrachloride and tons of natural uranium to make  $UCl_4$  feed material for the electromagnetic separation units (Griffith, 1957). The conversion of uranium into  $UCl_4$  involved chemical processes and reactions which had the potential for loss of uranium compounds through air dispersion or liquid releases. Most of the chemical handling and process operations were performed in large versions of laboratory-type hoods or some type of containment unit which minimized the potential for airborne releases of uranium into the workplace air. These exhaust systems were not filtered for the normal uranium, although some attempt was made to filter higher enriched material. Occasionally there were spills and equipment leaks which often led to the release of uranium-bearing materials to the off-site environment primarily through storm sewers (Griffith, 1957; Owings, 1986; Googin, 1993).

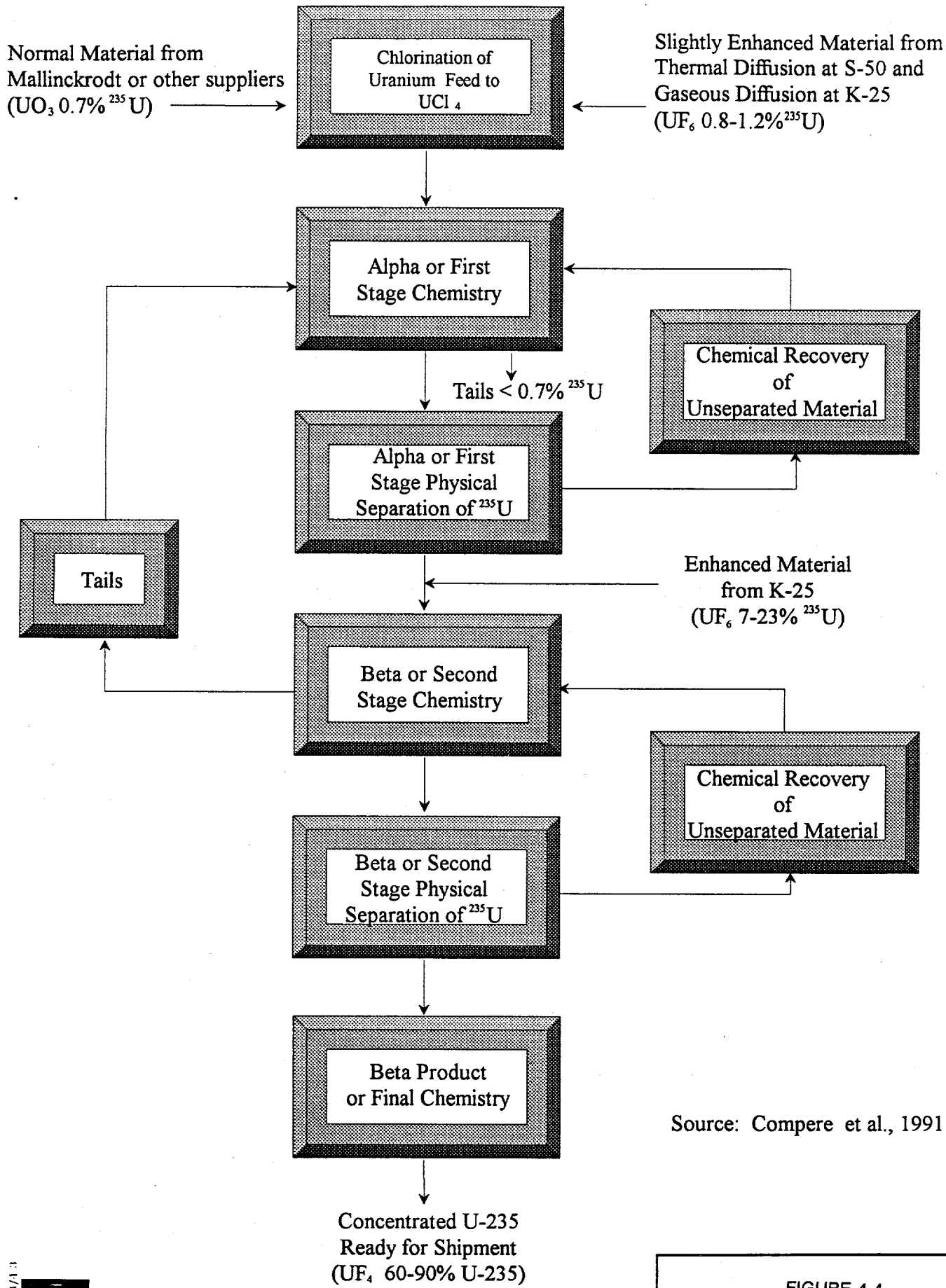


FIGURE 4-4  
FLOW OF MATERIAL IN  
THE ELECTROMAGNETIC PLANT



Two methods were used to produce  $UCl_4$ . The first method was liquid-phase chlorination in which uranium and liquid carbon tetrachloride ( $CCl_4$ ) were heated under pressure. After the chlorination was completed and the uranium tetrachloride crystals were collected and placed in the charge bottles, the process equipment was then washed and all washings were collected for recycle (Compere et al., 1991). Although no documents were found during the Phase I investigation that specifically discussed releases of any material from these processes, one report mentioned that solutions of raffinates (waste products in solution) and wastewater were released through drains to East Fork Poplar Creek (Griffith, 1957). Many of these effluents from this first method contained carbon tetrachloride and chlorine and low levels of uranium. A second method used was vapor-phase chlorination, in which carbon tetrachloride was gradually added to uranium trioxide under heated conditions for approximately 8 hours inside a chemical reactor bowl. The compound was then subjected to a nitrogen purge inside the reactor bowl to remove and exhaust phosgene vapors from the system (Compere et al., 1991). The process vapor also contained small amounts of uranium, which were sent through caustic and vapor scrubbers and then released to the atmosphere through stacks that were periodically monitored (Compere et al., 1991; Googin, 1993). Particular attention was paid to controlling and eliminating releases of the highly-toxic phosgene gas that was given off as a by-product of this process (Googin, 1993; Compere et al., 1991). Periodic grab air samples were taken to ensure some degree of control over the process. Effluent monitoring is discussed further in a later section of this report.

#### Material Recovery and Recycle

The electromagnetic separation itself was only 7 to 8% efficient, which meant that most of the uranium had to be recovered and made into new charge material and recycled back through the enrichment process until the desired enrichment levels were obtained (Griffith, 1957; Compere et al., 1991). The overall chemical recovery of enriched uranium involved the following basic steps:

- Washing of the electromagnetic equipment units (calutrons) with nitric acid,
- Precipitation of uranium from the washing solutions with hydrogen peroxide,
- Separation of the uranium peroxide precipitate,
- Calcination of precipitate and chlorination of uranium trioxide,
- Transfer of uranium tetrachloride into charge bottles, and
- Charge bottles loaded into calutron units for further enrichment.

(Adams et al., 1947)

Y-12 constructed elaborate recovery facilities to accomplish efficient recovery of uranium from the calutron components. The alpha calutron units were rather large, complicated structures of metal, largely copper, carbon and ceramics. After a production run, the different parts of the calutron contained a variety of uranium compounds, including tetrachloride, oxychloride,

carbides, oxides, and metal (Compere et al., 1991). After use, the various parts of the machine were vacuumed and washed in concentrated and dilute nitric acid solutions and brushed with stainless steel brushes. Since the uranium was in the form of chlorides, oxides, uranyl nitrates and  $UCl_4$ , the washings generated a rather corrosive solution from which the uranium had to be recovered and remade into charge material. Various processes were used at different times, but most of the material was recovered by one in which impurities were removed from the solutions by pH adjustment and filtration (Griffith, 1957; Compere et al., 1991). The uranium was precipitated as a peroxide and the solutions were filtered (Compere et al., 1991). The filtrates from the peroxide step contained mostly natural uranium, which was passed to the sewer system. The uranium peroxide was calcined to uranium trioxide in rotary furnaces, from which there were some airborne releases. The uranium trioxide was converted to the chloride form by heating with liquid  $CCl_4$  under pressure or by a vapor phase process as previously discussed. The chloride was further purified by sublimation when required, and the wastes generated were generally released through the drains into East Fork Poplar Creek. It is estimated that the airborne losses were probably not more than a few percent of the total losses as shown in Table 4-3 for this process (Griffith, 1957).

The uranium from carbon parts was leached in a strong nitric acid solution or recovered by burning the carbon which generated airborne releases (Griffith, 1957). Rejected carbon parts containing low levels of uranium deposits were crushed and burned in stainless steel furnaces. The airborne effluents were passed through a cotton filter and then on to a rotoclone system which passed the material to the exhaust stack (Compere et al., 1991). The carbon burning was a source of airborne releases. Some stacks or general ventilation systems had glass wool filters which were periodically leached with nitric acid to recover the uranium (Googin, 1993).

In the case of the beta cycle the charge material preparation and uranium recovery operations were performed much more carefully than that of the alpha cycle because of the enhanced value of the material. The beta calutron units were smaller and more efficient. The chemical recycle had the same objective of reconvertng unused material into new  $UCl_4$  feed material but was done in a more quantitative manner and carefully tracked in a material accounting system (Compere et al., 1991; Googin, 1993). The uranium from the various precipitation steps was recovered by solvent extraction and more careful attention was paid to the prevention of airborne uranium releases. Special attention was given to the recovery of uranium collected on the beta receivers. The receiver was the portion of the beta calutron which contained the desired isotope uranium-235 for use in the weapons. According to one report, of the total losses from the beta recovery process steps, airborne or dust losses were the most significant whereas with the alpha recovery processes, most of the losses were from liquid releases (Griffith, 1957). For both alpha and beta processes, the estimated losses through both the airborne and liquid pathways

TABLE 4-3

## ESTIMATED EARLY URANIUM RELEASES FROM THE Y-12 PLANT

Source	Period	Uranium (kg)*	Basis for Estimate
<b>I. Alpha Process</b>	1943-1945	40,000	Known losses and unaccounted-for material
<b>II. Beta Process</b>	1944-1947	400	Known losses and unaccounted-for material
<b>III. Product Processing</b>			
<b>A. Building 9206</b>			
Beta Product	1944-1947	0.4	Unaccounted-for material
<b>B. Building 9212</b>			
1. Process Vents from UF <sub>6</sub> Reduction, D-Wing	1954 to 1956	0.3	Effluent analysis and operating experience
2. Process Vents from UF <sub>4</sub> Preparation by Batch Process	1945 to 1952 1953 to 1956	0.5 1.5	Effluent analysis
3. Discards from Chemical Processing to Sewer	1953 to 1956	5.2	Accountability and operating records
4. Airborne Ventilation Losses from Product Processing	1945 to 1952 1953 to 1956	1.7 1.7	Air sample analysis
5. Losses Outside Plant Area on Shoes and Clothing	1945 to 1956	1	Estimated
<b>IV. Other Chemical/Metallurgical Processing</b>			
<b>Building 9206</b>			
A. Process Vents from UF <sub>4</sub> Preparation	1955 to 1956	0.1	Effluent analysis and operating experience
B. Airborne Ventilation Losses	1955 to 1956	0.5	Operating experience in similar operations
C. Airborne Salvage Area Losses	1946 to 1952 1953 to 1956	0.8 5.2	Unknown
D. Discards from Chemical to Sewer	1946 to 1952 1953 to 1956	2.1 13.2	Accountability records
E. Accidental UF <sub>6</sub> Release to the Atmosphere	5/11/56	2	Measured by difference after cleanup
F. Losses Outside Plant Area on Shoes and Clothing	1946 to 1956	0.5	Estimated

TABLE 4-3  
(Continued)

ESTIMATED EARLY URANIUM RELEASES FROM THE Y-12 PLANT

Source	Period	Uranium (kg)*	Basis for Estimate
<b>V. Normal and Depleted Assay Processing</b>			
A. <u>Building 9212</u>			
1. Airborne Ventilation and Hood Discharge	1948 to 1953	1500	Unaccounted-for losses
2. Airborne Ventilation and Hood Discharge	1954 to 1956	5,000 to 10,000	Present recovery figures. (Losses since July, 1955, are approximately 0.1 kg/day)
B. <u>Building 9206</u>			
1. Combustible Salvage Burning	1948-1955	3,200	Operating experience
2. Discards from Salvage to Sewer	1946 to 1956	1,435	Accountability records
3. Waste Flush to Sewer	1955 to 1956	60	Operating experience
C. <u>Building 9211</u>			
1. Airborne Salvage Burning and Kiln Losses	1953 to 1956	120	Estimated to December 1955; calculated from effluent analysis December 1955 to Date
2. Airborne Ventilation Losses	1953 to 1956	10	Operating experience
3. Discards to Sewer	1953 to 1956	900	Accountability records
<b>VI. Discards to Disposal Pits and Burial Grounds</b>	1943 to 1956	9,000	Accountability and operating records

\* The material lost varied in composition from natural to highly enriched in U-235 composition.

Source: Griffith, 1957.

were to a large extent based upon material balance information, known losses from accidental releases, or from actual measurements (Herndon et al., 1947; Murray, 1947; Griffith, 1957).

A summary of estimated early uranium releases that occurred at Y-12 including estimates of significant releases up to 1956, is presented in Table 4-3. Some of the estimates shown in Table 4-3 do not agree with later estimates of historical uranium releases reported in the mid 1980's (Butz, 1986; Owings, 1986; DOE, 1988). The reasons for these differences were not determined during this investigation. Examples of some of these later estimates can be found at the end of this report in Tables 4-9 and 4-10.

Some of the beta product was also collected on carbon parts of the calutrons. These parts were vacuum-cleaned to recover the bulk of the uranium product, which was in the form of a loose powder, and the remaining carbon was burned to insure complete recovery. Some parts, such as the charge bottles and ion source subassemblies, were removed, weighed and put into a hot concentrated nitric acid and hydrogen peroxide bath, later to be scrubbed with brushes and rinsed (Compere et al., 1991). The uranium input to these processing steps was monitored very closely and, although there were undoubtedly uranium releases to the off-site environment, all of the operations were done with great care and uranium material balances indicated the losses to be fairly low (Compere et al., 1991; Griffith, 1957).

The final step in the Y-12 electromagnetic enrichment process was accomplished when the desired enriched uranium material was collected, put into a nitric acid solution, purified by ether extraction, precipitated as the peroxide, converted to the oxides, and made into  $UF_4$  with  $H_2$  and  $HF$ . The  $UF_4$  was then shipped to Los Alamos for incorporation into the first atomic bombs. U.S. scientists learned a great deal about uranium chemistry during these early calutron washing and recycle operations (Compere et al., 1991). It is believed that some of the dust losses were never detected in the beta recovery and recycle chemical processes and, as a consequence, would have appeared as losses in the final beta separation and material recovery processes (Griffith, 1957).

Starting in 1945, Y-12 began to receive partially enriched  $UF_6$  from K-25 for use in the beta units. These new shipments effectively eliminated the need to continue the alpha separation and enrichment operations for producing beta feed uranium-235 for weapons material. The  $UF_6$  was converted to  $UCl_4$  and loaded into the calutrons. Experiments were carried out using uranium hexafluoride ( $UF_6$ ) as calutron feed material but the method was never proved useful due to the corrosive nature of fluoride on the calutron components (Patton et al., 1963). For a time in 1945, Y-12 received 0.8% to 1.2% enriched uranium in the form uranium hexafluoride from the S-50 Liquid Thermal Diffusion Plant. However, once K-25's gaseous diffusion plant became fully operational and was shipping product to Y-12, it was decided to close the S-50 plant due to high costs and technical deficiencies (Googin, 1993).

### 4.1.3 Weapons Production Facilities and Operations

The extensive experience that Y-12 gained in chemical processing of numerous uranium compounds during the electromagnetic separation program led to Y-12's involvement in enriched uranium nuclear weapon parts manufacturing after World War II. Since 1948, Y-12's primary mission has been the fabrication and assembly of various types of nuclear weapon parts. Complicated material processes ranging from basic chemical conversions of raw materials to highly-sophisticated machining of weapon parts have been carried out at Y-12. Due to security concerns, much of the information that describes these historical weapon production operations has been kept classified throughout the years at Y-12.

#### Enriched Uranium Operations

Weapon-grade enriched uranium has been one of the predominant materials processed within the Y-12 production areas for the fabrication of nuclear weapon components. Other weapon component materials have included depleted uranium, lithium, beryllium, and thorium, but enriched uranium has been the material processed most often at Y-12. The plant has handled large quantities of enriched uranium dating back to the Manhattan Project's electromagnetic uranium enrichment plant. It was reported that Y-12 began limited high-precision machining of enriched uranium weapon parts back as early as 1948. During this period the plant was undergoing a change in an attempt to establish itself as a supplier of nuclear weapons.

As the cold war developed between the U.S. and the U.S.S.R., and the postwar nuclear arms race heated up, it became apparent that Los Alamos Scientific Laboratory was not capable of keeping up with production requirements for nuclear bombs. By 1950, Y-12 had assumed the role as the lead U.S. manufacturer of weapon components made from enriched uranium. Y-12 produced a wide variety of weapon parts made of enriched uranium and other materials associated with these parts. Enriched uranium has properties which make it quite suitable for nuclear weapon applications. At the appropriate enrichments, it is capable of sustaining a nuclear chain reaction of sufficient magnitude to produce a nuclear explosion. It also has the advantage over plutonium, another material used for nuclear weapons, of being much less radiotoxic and easier to handle and process from a health and safety standpoint.

Total quantities of enriched material handled at any one time throughout the history of the plant have been kept classified. In order to keep track of this material at Y-12, the plant has always maintained a detailed accounting system (Mabe, 1993). Accounting and control measures were critical because these materials are extremely expensive, potentially harmful to the environment, not readily available for purchase, and controlled for national security reasons and the threat of nuclear proliferation. The Y-12 accounting system is described in more detail in Section 4.4 of this report.

The following sections describe the general chemical and metallurgical processes used with enriched uranium at Y-12. This discussion of enriched uranium operations is not intended to cover every process step, rather, it is intended to provide a brief overview of important activities and describe how the material might be released to the outside environment. The operations conducted since the early 1950s have not changed much, and most of the chemistry and process steps used today were developed during that time period (Patton et al., 1963). The primary operations for enriched uranium processing and weapon parts fabrication have been located in Buildings 9201-5, 9206, 9212, 9215, 9204-2, and 9204-2E. Various chemical and metallurgical processes have been involved in these operations and have been associated with routine airborne and waterborne releases. Control mechanisms used in the majority of the enriched uranium processes have included multi-stage roughing (larger particle collection) and HEPA filtration systems (Sanders, 1992; Butler, 1993). Although examples of these control mechanisms and some quantitative evaluations were described in several reports, specific comprehensive historical details regarding the types, quantities and location of these effluent filtration systems were not identified during the investigation (UCC-ND, 1971a; UCC-ND, 1971b).

Historical enriched uranium operations have included: 1) reduction of  $UF_6$  to  $UF_4$ , 2) reduction of  $UF_4$  to metal, 3) casting or forging, 4) rolling and forming in mill operations, 5) machining of metals, 6) wet chemistry purification, recycle, and salvage, and 7)  $UF_4$  extraction. Shortly after World War II, Y-12 began to receive routine shipments of highly enriched uranium hexafluoride ( $UF_6$ ) from the gaseous diffusion enrichment plants, which was then reduced to  $UF_4$  and uranium metal for fabrication into nuclear weapon components. Y-12 continued to receive  $UF_6$  from these plants for a number of years in order to meet production schedules for weapon parts. The  $UF_6$  was placed in heated chemical reactors and converted to  $UF_4$  with the addition of hydrogen. The chemical reaction produced  $UF_4$  which was then reduced to metal and hydrogen fluoride (HF) which was given off as a by-product of the reduction.  $UF_4$  is called "green salt" because of its color. After 1964, Y-12 stopped receiving  $UF_6$  and began to get its supply of enriched and depleted uranium from retired weapons, a source which was capable of sustaining the required U.S. stockpile of enriched uranium for weapon parts fabrication (Googin, 1993). The uranium recovered from old weapons as well as other DOE facilities was purified and recycled back through the Y-12 production processes. For example, the enriched uranium from retired weapons was recovered, usually remelted in an induction furnace, made into a casted part, and sent to machining. Sometimes, however, the material was cast into long semi-cylindrical or cylindrical shapes and sent to a rolling/pressing mill and then on to a machining shop to be turned and cut to the desired design specifications (MMES, 1988).

On May 11, 1956, a hydrogen line to the Building 9206  $UF_6$  reduction area broke. This caused incomplete conversion of the  $UF_6$  to  $UF_4$ . The chemical traps which were provided to capture the  $UF_6$  in such an emergency were filled to capacity at the time of the break, and  $UF_6$  was allowed to escape to the atmosphere. The escaping  $UF_6$  was visible as  $UO_2F_2$  smoke as it was emitted from the vent stack. It was estimated that approximately 2 kilograms of enriched

uranium were released to the atmosphere (Griffith, 1957). Detailed information regarding this accident was not identified during this investigation. Other accidental releases of  $UF_6$  may have occurred at Y-12, but none were identified during this investigation.

### Enriched Uranium Recovery, Purification and Recycle

Recovery and recycle of uranium has been performed, when economically and technically feasible, on a wide variety of process wastes and by-products generated by the various manufacturing processes that has taken place in the main production buildings. Much of the recovery operations take place in Buildings 9212, 9206, and adjacent buildings. Numerous chemical operations have been necessary in the recovery and purification of uranium from various materials that come from Y-12 enriched uranium processes. The enriched uranium facilities have included a uranium recovery process that recycles almost 100% of its scrap material (Butler, 1993).

Recovery operations consisted basically of 1) burning combustibles, 2) dissolving or leaching solids, and 3) purifying the uranium bearing solutions by chemical extraction. Generally, purified uranyl nitrate hexahydrate (UNH) solutions were produced from these operations and then denitrated to uranium trioxide ( $UO_3$ ), which was then reduced with hydrogen to uranium dioxide ( $UO_2$ ). The  $UO_2$  was subsequently converted to uranium tetrafluoride ( $UF_4$ ) by reaction with gaseous anhydrous hydrogen fluoride. Finally, the  $UF_4$  from the process was reduced under high temperatures to yield uranium metal, which was then cast into desired mold configurations. At almost any point in this operation, the enriched uranium could be withdrawn to meet a particular material need. These reduction processes involved dry powder compounds handled in glove boxes, with local exhaust due to the great potential for airborne suspension. Historically, these exhaust systems were heavily filtered to prevent or minimize airborne uranium releases to the environment. These releases might have included several different compounds of uranium (MMES, 1988; Butler, 1993).

Among the safety precautions that have been strictly observed are those safety steps to significantly reduce the possibility of a nuclear fission excursion or "criticality accident" from occurring due to the processing of certain uranium of high enrichments at Y-12. A criticality accident involves a chain reaction in which many uranium-235 nuclei absorb neutrons and undergo fission, giving off a sudden burst of nuclear fission energy and nuclear radiations. Such a fission chain reaction will occur and sustain itself if concentrations of enriched uranium and the physical geometry of the material are maintained under the proper conditions. So care must be taken in storage of solid, liquid, and gas forms of highly enriched uranium. Workers must be properly trained to understand the hazards and work under safe-operating procedures to prevent such an accident from occurring. Uranyl nitrate, various uranium compounds, and uranium metal all have the potential to undergo and sustain this sudden nuclear reaction.



On Monday, June 16, 1958, at approximately 2:05 p.m., an accidental nuclear fission excursion occurred in Building 9212. A solution of 93% enriched uranium, containing approximately 50 gm uranium-235/liter, flowed through a valved pipeline from an extraction product "nuclear-safe" tank and partially filled a 55 gallon drum which was a "nuclear-unsafe" container for those enriched solutions (Patton et al., 1958). The tank was undergoing leak tests with water at the time of the accident. Criticality alarms were sounded immediately throughout the plant (McClendon, 1958).

Following the initial nuclear burst, which did not discharge the contents of the drum, the uranium solution went critical at least two more times. It was reported that the estimated range of nuclear fissions that occurred during this accident was between  $2.2 \times 10^{18}$  and  $3.7 \times 10^{18}$ . The nuclear chain reaction was eventually stopped by additional water that flowed into the drum. Based upon an evaluation of a strip-chart recorder located in another building, which recorded results from a continuous radiation monitor, it is believed that one or more nuclear excursions occurred within an approximate 20-minute period (Patton et al., 1958).

Immediately following the initial sounding of local criticality alarms, Y-12 emergency personnel equipped with portable radiation survey meters measured direct radiation levels that exceeded 100 mR/hr at the building control center, a distance that was about 350 feet from the location of the accident. The Y-12 Plant emergency squads surveyed the west and north ends of the building, and found levels which ranged between 50 to 100 mR/hr. During this same period, elevated radiation levels were detected by laboratory supervisors at the north end of the Analytical Laboratory located in Building 9995, approximately 400 feet east of the accident scene. Initially these levels were fluctuating in intensity up to about 1000 mR/hr and shortly thereafter decreased down to 500 mR/hr. It was noted that Building 9212 is oriented in such a manner that the corridors and operating areas extend in an east-west direction (McClendon, 1958). It was believed that less attenuation had occurred in the east and west direction due to the lack of walls and large equipment. The areas north and south of the accident were shielded by more building structure and large amounts of heavy equipment which may have reduced the radiation readings. These radiation surveys indicated that the incident had occurred within Building 9212. This was later confirmed by 9212 personnel who reported the initial details of the accident. Surveys taken along the outside of the 9212 perimeter fence showed that radiation levels that were up to 50 mR/hr for a period of twenty minutes after the excursion began. Radiation levels then dropped to between 5 and 10 mR/hr by approximately 2:25 p.m. (20 minutes after the beginning of the excursion) (McClendon, 1958).

Radiation surveys were made of the plant area to obtain an overall evaluation of the seriousness of the accident. Within hours after the incident, it was reported that there was no direct radiation or significant contamination outside the perimeter fencing, which formed the security area in which Building 9212 is located. This area, approximately 800 feet by 1000 feet, was subsequently marked off as an initial restricted area. Efforts were made to survey the areas

around the plant and detect the release, or subsequent fallout, of fission product activity. High-volume air samplers were set up outdoors at five locations ranging from about 700 feet to about 3000 feet downwind from the accident. However, sampling was not begun until about 50 minutes into the incident and 20 minutes after the ventilation supply and exhaust fans in the accident areas had been turned off. The samples were collected on Whatman No. 41 filter paper and counted for alpha and beta-gamma activity. These samples indicated a maximum concentration of  $2.5 \times 10^{-11}$  microcuries/cm<sup>3</sup> beta-gamma activity at the time of sample collection. This was below the  $10^{-9}$  microcuries/cm<sup>3</sup> permissible level of airborne activity specified by radiation protection standards. However, there was significant airborne alpha contamination detected in the same areas sampled (Patton et al., 1958; McLendon, 1958).

Some indication of airborne contamination released to the atmosphere was obtained from inspection of two continuously recording beta-gamma air monitors which were located outside Building 9212 next to Building 9207 and 9204-1 which were east to southeast of 9212. These monitors were Geiger-Muëller tube survey instruments which were capable of measuring and recording the beta-gamma emissions from particulates collected on a fixed filter paper. The tubes were surrounded by the filter paper and were shielded by 1-1/2 inches of lead to reduce external background radiation. Both instruments detected the initial direct gamma radiation from the actual excursion and both showed subsequent increases in the level of atmospheric beta-gamma contamination. Some of the following observations were made from inspection of the charts from these instruments, and were believed to be an indication of the radioactive fallout from the accident (McClendon, 1958).

- The level of initial direct radiation recorded at Building 9204-1 was higher than that observed at Building 9207 because of the distance involved.
- The airborne contamination reached Building 9207 at higher concentrations on the order of a factor of three, since it was directly downwind.
- Because of the short half-lives of the fission products (less than 2 hours) and the relatively short length of exposure of any persons to the contaminated atmosphere, it was believed that the concentrations detected did not constitute an acute health hazard to individuals on the plant site or off-site.

- Weather conditions associated with wind velocity and wind direction that existed at the time of the accident were favorable and minimized contamination of nearby populated areas. Although earlier air samples were negative, they were taken some time (approximately 50 minutes) after the accident and may have missed any potential radioactive fall-out.

No further information was obtained by ChemRisk's investigation to support these conclusions. Further evaluations may be warranted to confirm or reject the validity of these observations. These Y-12 conclusions are presented in this report as potential useful information and as a guide in any future studies of historical Y-12 off-site releases.

Between 3:00 and 4:00 p.m. on the day of the accident, radiation surveys were conducted of the parking lots along the north side of the plant site, which were located east and west of the accident areas. Spot checks were made on the ground, paved areas and automobiles. No evidence of beta-gamma contamination was detected and the automobiles were thought to be free of any contamination (McClendon, 1958; Patton et al., 1958).

At approximately 9:30 p.m., on June 16, 1958, the drum involved in the incident was "poisoned" with the insertion of a cadmium scroll as a neutron absorbing material to prevent any further nuclear excursions. The term "poisoned" in this application meant that the cadmium scroll was used as a poison to effectively absorb the neutrons in the drum which prevented any further nuclear reaction. Clean-up activities within Building 9212 areas except C and C-1 wings began during the night of June 16. A nuclear-safe tank was fabricated and installed in a shielded radiography cell in Building 9212 to hold the contents of the drum. The empty drum was then transported to ORNL for radiochemical analysis. Clean-up activities were continued and by the morning of June 19, all of the Building 9212 chemical recovery facilities with the exception of those in the central and east portion of C-1 wing were placed back into operation (Patton et al., 1958; McClendon, 1958).

Eight Y-12 employees were in the vicinity of the drum at the time of the incident. Their external radiation doses ranged between 28.8 rem and 460 rem. The neutron and gamma doses to personnel were determined by measuring the neutron-activated sodium-24 in human blood of those who were exposed during the accident. A comparison of the activation of blood from these individuals with that from a donkey exposed to a mock-up of the criticality, and for which the dose was carefully determined, resulted in the best dose estimates. Evidence of other accidents of this type at Y-12 were not discovered during our investigation.

### Fabrication and Machining

Weapons fabrication and machining are highly-sophisticated operations at Y-12. These processes are housed in 14 production shops. Historically, the main responsibility of these operations has been to machine rough shapes to precise configurations. Capabilities have spanned a wide spectrum of machine types and manufacturing methods. Rough-shaped components have been received for final machining and assembly of the specified parts (UCC-ND, 1983).

Enriched uranium machining was carried out primarily in Building 9215. Parts of various sizes are cut to final dimensions on cutting lathes. Over the years, large quantities of machine coolant and metal cutting oils have been used in these machining operations. Dating back to the early years, large volumes of PCBs (polychlorinated biphenyls) were used as an effective cutting oil for machining these parts. Chlorinated oils such as PCBs were ideal for cutting enriched uranium because of their neutron-absorbing characteristics. PCBs acting as neutron poisons were an additional safety factor in assuring that no accidental criticality would result from the build up of machine cutting. Y-12 typically used a series of PCBs called "Aroclors." After machining, most of the uranium was removed from the oil, and the oil was then disposed of in large tanks located outside the production buildings. Limited capacity in the tanks required Y-12 to eventually dispose of the oils in the Bear Creek Burial Ground area (Napier, 1992). Documents which describe quantities of PCBs used earlier were not identified during the investigation

Perchloroethylene-based oil, also called "perc," was also used in large quantities as a machine coolant for a number of years in the enriched uranium machine shops. Perchloroethylene is also called tetrachloroethylene. Around 1976, Y-12 replaced the PCB cutting oils with this coolant due to potential health hazards associated with PCBs. During the early 1980s, up to 20 kilograms or more per year of "perc" were used as coolant for machining enriched uranium. Quantities for earlier years were known to be higher as a function of production levels (Napier et al., 1982; Napier, 1992). Y-12 did recycle some of these coolants when it was economically feasible (Napier, 1992; Mabe, 1993). Documents which describe perc quantities were not identified during this investigation.

In the enriched uranium areas, the machines have been equipped with local hoods and exhaust systems which pass airborne materials through a series of filters, including HEPA (high efficiency particulate air) filtration. After filtration, most of these effluents are vented to the atmosphere through exhaust stacks which have been routinely monitored since the 1950s. Custodians (e.g., individual departments) of the enriched material have always been required to track the amounts of uranium losses for accountability purposes.

Uranium is very pyrophoric in the metallic state and, therefore, Y-12 has always had to take the necessary steps to minimize the occurrences of uranium metal fires, particularly those involving uranium cuttings or chips. The use of water and coolants for metal chips has helped to avert potential fires. However, there have been numerous small chip fires at Y-12 over the years (Napier, 1992; Sanders, 1992; Hunt, 1993). Reportedly, most of the small fires were confined to the uranium chips and did not spread to other parts of the machining equipment or operations. Under proper operating conditions, the HEPA filtration is believed to have controlled a significant fraction of the airborne uranium effluent that might have been generated from these fires.

Enriched uranium chips immersed in coolant were routinely transferred on dolly carts to recycling operations. The coolant was used to prevent chip fires. There have been some exceptions, for example, a fire in 1985 occurred when metal chips loaded on a dolly cart caught fire and released some uranium to the outside without any air filtration through a small wall fan located nearby. It was estimated that a small amount of enriched uranium was released through this fan (Butz, 1985).

In late 1985, perchloroethylene was replaced with a aqueous solution of propylene glycol with borax (sodium tetraborate) for use as the machine coolant for enriched uranium parts (Napier, 1992; Butterini, 1992). Sodium tetraborate was added as a neutron absorber to prevent criticality accidents. This new coolant was not as effective at preventing chip fires, but had the benefit of not being as volatile as perchloroethylene. According to one employee interviewed during our investigation, it was stated that the use of the new coolant effectively reduced the uranium airborne releases from the higher levels observed when the uranium was machined with the perchloroethylene (Shelton, 1993). The specific reasons for these reduced releases were not clearly defined during our investigation. This new coolant is still used as the machining coolant for enriched uranium. Perchloroethylene continued to be used as a degreaser of machined parts in Building 9204-2E through the 1980s.

Machining operations also included various solvents and degreasing agents for cleaning production parts and assemblies. Materials used in this application included trichloroethylene, trichloroethane, and a few types of freon. According to some conservative estimates, as much as 80 to 100% of these materials were released to the air (Fee, 1986). A partial historical chemical release document describing quantities of various chemicals used at Y-12 was located during the investigation (Fee, 1986). Further discussion of this documents can be found in Section 4.4 of this investigation report.

#### Uranium Salvage Operations

Salvage operations involved the recovery of enriched uranium from all non-product components or byproducts of the operations. Everything from machine turnings and nitric acid solutions to

mop water and hand rags were sent through processes which recover trace amounts of weapon-grade uranium. These operations involved a number of major material process streams, and detailed accounting records are kept at each step of the process (Butler, 1993). The economic and strategic value of enriched uranium required the reclamation of most uranium side streams generated from fabrication and production processes. Enriched uranium salvage operations used at Y-12 are characterized by the following functions:

- Concentration of uranium by ignition of gross bulks of combustibles,
- Dissolution in nitric acid,
- Concentration and purification of dissolved uranium, and
- Conversion of the purified uranium to a form suitable for re-entry into main line production operations.

Chemistry operations for the extraction and purification of enriched uranium which comes from various salvage and by-product streams are primarily located in Buildings 9212 and 9206. Typically, the materials are washed in nitric acid baths to dissolve or leach the enriched uranium or carbon items are burned in a muffle-type furnace and the uranium is recovered later. The recovered uranium was usually in the form of uranyl nitrate which was sent through purification and extraction columns to remove the impurities from the solutions. The enriched uranium could then be precipitated with ammonium hydroxide and filtered from solution as ammonium diurate (Patton et al., 1963). The ammonium diurate was then dried in a kiln to produce  $U_3O_8$  and then converted to  $UF_4$  ("green salt"), reduced to metal, and was ready for melting, rolling, or casting into rough shapes for machining and weapon parts fabrication (Patton et al., 1963; MMES, 1988). Most of the effluents from these operations were treated or filtered to prevent airborne or waterborne releases of the valuable enriched uranium to off-site environments (Butler, 1993). Historically, the acidic processes were difficult to filter and were a source of airborne releases (Hunt, 1993; Butler, 1993). Liquid wastes from these processes, low in uranium content, were sent to outdoor seepage ponds.

Destructive distillation was used as another way to recover uranium from salvage materials; this was an air oxidation process carried out in a muffle furnace and incineration unit. The use of this method depended on the type and quantity of salvage material that contained recoverable amounts of uranium-235. In those circumstances where the uranium-235 content of the uranium was approximately 20% or above, economic considerations usually warranted that this or other recovery methods be used on items such as discarded graphite molds, absorbent paper, old gloves, clothes, shoes, filter paper, filter cloth, oily rags, and etc (MacKay, 1948; Patton et al., 1963).

A principal advantage of destructive distillation in the processing of highly enriched uranium salvage was that it allowed tars, organic acids, hydrocarbons and nitric acid to be removed at exhaust gas flow rates low enough that the amount of uranium was small in the effluents, thus allowing volatiles to be condensed and discarded and nonvolatiles to be vented to exhaust stacks without filtering (Patton et al., 1963). On the other hand, combustion in an incineration unit involved turbulent and non-uniform flow of combustion gases which contained significant quantities of uranium that were filtered in exhaust stack gases (Sanders, 1992). These exhaust gases were typically filtered with HEPA filtration systems to contain or minimize the uranium releases from Y-12 (Butler, 1993).

For high uranium-235 concentrations, the amount of combustible material generated from a single source was usually too small to warrant an incinerator installation. However, in circumstances where a long-term, large-scale generation from a single source prevailed, incineration has been used (Patton et al., 1963). For salvage materials containing uranium of low uranium-235 concentrations, incineration typically has been used at Y-12. Treatment such as washing was all that was economically warranted for gloves, clothes, shoes, and many other sources of volatile organics. For destructive distillation, combustibles were loaded and sealed in the electrically heated stainless steel retort (a vessel in which substances are decomposed by heat), which was then brought up to temperature. When all the volatiles had been driven off, the remaining charred residue was oxidized by the introduction of oxygen and then cooled by room air drawn into the furnace (Patton et al., 1963). The ash, which contained a fair amount of uranium, was removed from the furnace by means of a vacuum hose which was manually operated. The hose was connected to a filtering system consisting of a cyclone separator, a porous metal filter, a cloth pack filter, and a vacuum blower connected in series. The ash from the cyclone separator dropped into some type of dust collector. The effluents from this system were also filtered with HEPA filters. The ash was then removed and leached with nitric acid and then sent to other operations for extraction and purification of the recovered uranium (Patton et al., 1963). The high enrichment levels of this uranium required Y-12 to keep track of all inventories very meticulously through each process step.

Back in the 1950s, combustible organic salvage materials were burned in a furnace prior to leaching in Building 9211. Routine exhaust gases from the furnace contained  $\text{UO}_3$  and  $\text{U}_3\text{O}_8$  dust. The ash was leached and precipitated as ammonium diuranate. This was calcined to oxide in an indirectly fired rotary kiln. Considerable dust losses occurred from this operation. In December, 1955, a scrubber was put into operation which reduced the losses from both operations to about 0.3 kg/day of uranium when the salvage burning was being carried out. These airborne losses were primarily dust and fumes as  $\text{UO}_2(\text{NO}_3)_2$ , uranium oxides, and ammonium diuranate. The material processed was primarily old salvage which had been in storage at Y-12 (Griffith, 1957).

On October 23, 1956, at 11:54 am, a fire was discovered in the exhaust and filtering system of the Uranium Metal Burning Furnace in Building 9211. The exhaust and filtering system was located outside of the east end of the building, adjacent to Building 9211 (Elam et al., 1956). The system removed combustion products from a uranium metal burning furnace that was located in Building 9211. The fire destroyed 32 orlon cloth filter bags and 20 absolute filters with fiberglass media. The metal filter housing was also damaged by the fire. The fire was confined to the filter housing, with combustible material limited to the filter media, and no attempt was made to extinguish the fire (Elam et al., 1956). The fire apparently was started from a hot piece of metal which had been exhausted to the cloth bag filters. The fire spread through the cloth filters and into the absolute filters, destroying most of the equipment. The enrichment level of uranium used in this operation was not determined during our investigation, although one report indicates that these operations involved normal or depleted uranium (Griffith, 1957). Specific estimates of uranium releases that may have occurred during the fire were not identified during this investigation.

The uranium metal burning furnace was designed to operate at 1,600 °F to oxidize uranium metal by burning. Automatically-controlled gas burners were used to control the temperature in the furnace during the metal burning process. The exhaust systems were designed to exhaust products of combustion from the furnace, provide sufficient room exhaust to minimize contamination in the area, to filter gases from the recovery of oxide, and to reduce stack releases of radioactive material. Since little was known of the characteristics of airborne  $U_3O_8$  particles that were produced in a furnace of this type, the dust loading in the effluent gases leaving the furnace was assumed to be heavy and to consist of particles of varying sizes with a larger proportion in the small micron sizes. In the final design, the exhaust system consisted of room exhaust and dilution air at two points, a cyclone type dust collector, a self-cleaning, bag type dust collector, an absolute filter bank, an induced draft fan, and an 84-foot exhaust stack (Elam et al., 1956).

### Depleted Uranium

Depleted uranium operations for the production of weapon components have been in existence at Y-12 since the early 1950s. Depleted uranium is a form uranium consisting of over 99.8% uranium-238 and less than 0.7% uranium-235. The depleted uranium and depleted uranium alloy operations typically included melting and casting, forging and rolling into plates, forming or extruding into shapes, and final heat treating to obtain desired properties. The depleted forms were then machined into the final design specifications for weapons components (UCC-ND, 1983b). Buildings 9998, 9215, 9201-4, 9201-5 and 9204-4 housed the majority of Y-12's depleted uranium process operations (Hunt, 1993).



Depleted uranium has been handled in large quantities at Y-12 and the limited economic and strategic value of the material has minimized the need to reclaim or recycle this material from the production streams. As a result, large quantities of solid and, to a lesser extent, liquid wastes containing depleted uranium have been generated over the years at Y-12. The H-1 foundry, located in Building 9998, received depleted uranium from other DOE facilities and retired weapons since the 1950s. The foundry remelted the uranium metal and cast different shapes using large crucibles. The shapes were then rolled, cut, and pressed into rough shapes with multi-ton hydraulic presses located primarily in Buildings 9404-4 and 9201-5. Rough-shaped pieces were then sent to machine shops for final cutting to desired specifications. Losses of uranium from these operations have been primarily in the form of uranium oxide or uranium metal. Up until late 1953, almost all of the foundry and machining operations were not filtered (Griffith, 1957). Particle or dust collectors such as cyclones and bag houses were used in most of the processes to capture the larger-sized particles (Sanders, 1992; West, 1992; Hunt, 1993).

Airborne losses from the foundry (which was at times located in H-1 and in other buildings) and other operations were fairly large from 1948 to 1956. Production levels increased and additional filters were installed sometime in 1955, although initial problems reduced their effectiveness in filtering releases for the first year or so of operation. It has been reported that between January 1954 and July 1956, potentially large uranium losses may have occurred, since little dust was collected or filtered compared with the amount of uranium collected after the new filtering equipment was operating efficiently. According to one report, the amount of uranium released could possibly be estimated from production recovery figures, making adjustments for level of production at any given time. The losses that occurred during this period were often calculated from material balance information (Sanders, 1992). Specific material balance information for these operations was not identified during this investigation. The losses during 1956 were also estimated from stack monitoring to be about 0.1 kg of depleted uranium per day. Eventually, continuous monitors were installed on the exhaust stacks sometime in the mid 1950s (Griffith, 1957). The validity of these release estimates was not established during this investigation and, therefore, warrants further evaluation in future studies.

### Lithium Operations

In the mid 1950's, Y-12 began to process large quantities of lithium-6 compounds for use in thermonuclear weapons. Lithium deuteride has been the primary material used in these weapon systems. Thermonuclear weapons obtain their energy from the combination or fusion of heavy hydrogen atoms into heavier atoms. For this reason, they became commonly known as hydrogen bombs. Deuterium and lithium are necessary materials for this fusion reaction to take place.

Y-12 lithium operations have involved several manufacturing process steps to meet the necessary design specifications and production schedules of the weapon components. The amount of lithium handled at Y-12 is classified. Primary lithium operations and support activities have

been located in Buildings 9204-2, 9805, and 9204-2E (Hunt, 1993). Y-12's lithium process steps have included the following:

- Separation and enrichment of lithium isotopes (1950-1963),
- Production of lithium hydroxide,
- Neutralization of lithium hydroxide to lithium chloride (LiCl),
- Evaporation, crystallization, and reduction of LiCl to lithium metal,
- Conversion of lithium metal to lithium deuteride (LiD) or lithium hydride (LiH),
- Crushing, pulverizing, and demetalizing of LiD or LiH into a powder, and
- Pressing and molding of rough shapes and final machining.

The hygroscopic nature of lithium has required that lithium operations be confined to special, environmentally-controlled glove boxes and rooms which provide the proper atmospheric conditions.

Deuterium is required for the production of lithium deuteride or lithium hydride. Y-12 has produced its own source of deuterium by electrolysis of heavy water; these processes have been located in building 9805. Y-12 historically received its supply of heavy water from the Savannah River Plant (SRP) nuclear reactor return operations, located in Aiken, South Carolina. Shipments of heavy water are known to have been a source of the radioactive isotope of hydrogen known as tritium ( $^3\text{H}$ ) which has been introduced into Y-12's electrolysis operations since the 1950's (Hunt, 1993). Y-12 has taken routine tritium concentration measurements of SRP heavy water shipments dating back to the 1950's. Tritium concentrations have ranged between 0.002 and 0.4 microcuries per milliliters of water (Hunt, 1993). Up until 1985, most of the tritium concentrations measured in the shipments were at the low end of this concentration range because of the type of reactor returns that were being shipped from the Savannah River Plant. After 1985, the tritium concentrations of SRP heavy water remained at the higher end of this range. Information that describes tritium releases from Y-12 was not reviewed in detail during the investigation because sufficient time did not remain once associated records were obtained. The Y-12 Health Physics Department and the Y-12 Plant Analytical Laboratory have provided monitoring services to these operations for a number of years (Hunt, 1993).

#### Beryllium Operations

Y-12 has handled beryllium since the early 1950s as part of its weapons component manufacturing operations. Beryllium powders and metals have been heated, pressed, shaped or molded, and machined to the designed specification according to a particular weapon system. The primary beryllium operations have been located in Buildings 9201-5 and 9766.

Some of these beryllium operations released small particles into the air, requiring proper process containment such as sealed glove boxes and ventilation filtration systems. A significant amount of the airborne beryllium consisted of small particles. Inhalation of beryllium at various levels appears to cause a lung disease known as berylliosis in some individuals. It also appears that traditional dose response relations do not appear to hold for this immune system disease. Beryllium is also classified as a probable human carcinogen.

This potential toxicity of beryllium has required Y-12 to properly control and monitor airborne releases to the workplace or the outside environment. Routine workplace and stack monitoring has been performed in most of the beryllium process areas. ChemRisk identified examples of airborne monitoring results in Y-12 Health Physics retired records, located in a Y-12 document repository. These sample results which included sampling dates, locations and beryllium concentration values, were reviewed on a random basis but were too numerous to cover in much detail during this investigation. Most of the data randomly sampled pertained to air sampling of beryllium operations during the 1950s and 1960s. Y-12 Summary reports in the 1970s and 1980s (e.g. Y-12 Plant Quarterly Reports) described similar results in summary form. No evidence was uncovered during the investigation that indicated that liquid releases have been significant at Y-12. Interviewees have reported that solid waste contaminated with beryllium has been disposed of through land burial.

It has been reported that much of the beryllium operations were routinely monitored and controlled for airborne contamination (Sanders, 1992; Googin, 1993). Dating back to the 1950s, HEPA filtration was often used to capture airborne beryllium and minimize the releases to the off-site environments. Specific information about the types and quantity of beryllium operations that were filtered was not uncovered during this investigation. Over the years, Y-12 Industrial Safety and Hygiene and Health Physics Departments provided safety oversight to the beryllium process controls (e.g., exhaust filtration) and administered and reported the workplace and exhaust stack airborne monitoring programs. During this investigation, it was observed that many of the details to beryllium process operations have been classified for security reasons.

### Thorium Operations

Starting in the early 1950s, the Y-12 plant began thorium processing and fabrication of weapon components in operations similar to those that were involved with depleted uranium. Y-12 primarily handled metallic forms of thorium, which was processed in a multitude of operations such as arc-melting, casting, forging, rolling, and pressing into rough shapes and configurations. Based on information that was gathered, it is believed that the majority of these thorium production-scale operations took place in Building 9201-5 and 9204-4 and lasted until the early 1970s. As with the depleted uranium, the rough-shaped thorium parts were then submitted to the machine department for precise turning and cutting of weapon components. The thorium operations at Y-12 generated fairly significant activity amounts of solid and liquid waste as

shown in Tables 4-8 and 4-9. These included metal degreasing solvents such as tetrachloroethylene and others, which were recycled or analyzed for thorium and then buried at the west end burial grounds.

Discharges from the Y-12 production facilities came primarily from liquid and solid waste generated in processing weapons parts. These waste materials associated with salvage materials were stored or disposed on-site. Thorium process solutions discharged to the storm sewer system originated from ORNL research programs in Y-12 and from Y-12 production programs. The discharged ORNL solution included thorium oxide slurries from corrosion testing experiments and cleanup of test equipment used by ORNL Reactor Engineering (Owings, 1986).

### Lead Operations

Numerous application of lead, some of which were not directly involved with weapons production, have been carried out at the Y-12 Plant since the early 1950s. Applications ranging from the use of lead for radiation shielding to the fabrication of sophisticated weapon parts have been performed at Y-12. Y-12 usually received most of its lead from commercial suppliers. Some of the lead has had to meet very stringent specifications for isotopic composition and purity. Y-12 has often used lead to produce various lead-alloy materials for weapon applications. The major lead process operations have been located in Buildings 9998, 9201-5, and 9204-4 and have included arc-melting, forging, rolling and milling, and machining. Based on interviews conducted during this investigation, lead operations have involved relatively small quantities of material in comparison with Y-12 uranium operations (Butterini, 1992; Googin, 1993).

Process controls to minimize the release of lead or lead-bearing materials from Y-12 operations have been similar to those for depleted uranium operations. Information gathered during the investigation suggests that early airborne effluents were not extensively filtered or monitored, although quantities of lead that may have been released were not identified during this investigation. Large, bag-type filters were used for some of these process operations. It was not determined during this investigation whether or not better airborne effluent controls have been implemented in more recent years. Further evaluations may be warranted in any future studies to determine the potential off-site significance from historical lead releases.

#### **4.1.4 Lithium Separation and Enrichment Operations**

In the early 1950s, a massive national effort was undertaken to develop thermonuclear fusion weapons. Unlike earlier fission bombs that derived their energy from the splitting of uranium atoms, these new weapons obtained their energy primarily from the combination or fusion of

heavy hydrogen atoms into heavier atoms. For this reason, they became commonly known as hydrogen bombs.

The chemical compound lithium deuteride is a form of hydrogen fuel for use in fusion bombs, having desirable density and machinability characteristics (Wilcox et al., 1983a). Natural lithium contains about 7% of the lithium-6 isotope; the remainder is lithium-7. In the 1950s, a decision was made to separate high-purity lithium-6 from natural lithium to produce lithium-6 deuteride for use in more powerful and efficient weapons. The Y-12 Plant in Oak Ridge was given the assignment to develop, design, construct, and operate a production process to accomplish this task (Wilcox et al., 1983a). Most of these processes and equipment are still classified today.

The separation process that produced the majority of the enriched lithium-6 material was called the **Colex** operation, a name that was derived from the column-exchange method. This column exchange is a chemical exchange process in which lithium isotopes were partially separated as the material was transferred between two chemical phases. One of these phases was an aqueous solution of lithium hydroxide and the other phase was a lithium amalgam, a solution of lithium in mercury (Wilcox et al., 1983b). Many millions of pounds of mercury were required to meet production schedules. Directives signed by President Eisenhower allowed Y-12 to use a significant portion of the mercury that was available from the National Stockpile (Wilcox et al., 1983b). It was the use of this mercury in the Colex process from 1955 to 1963 that led to large releases (Wilcox et al., 1983a).

On May 17, 1983, Y-12 published a declassified version of a mercury inventory report, Y-12 Report Y/AD-428, which generated a great deal of interest by the public about the use and impacts from mercury at the Y-12 Plant (Wilcox et al., 1983a). On May 20, 1983, Y-12 management organized a team called the Mercury Task Force, made up of Y-12 employees and others to study the use of mercury, salvage and recovery operations, and to evaluate the potential impacts on worker health and the environment (Wilcox et al., 1983a).

### Lithium Operations

The separation of lithium isotopes on an industrial scale was based on the fact that, under certain conditions, the lithium-6 isotope will dissolve more readily in mercury than will the lithium-7 isotope. Lithium dissolved in mercury solvent is referred to as lithium amalgam and will remain in a stable state while in contact with an aqueous solution under the proper conditions. If lithium amalgam is allowed to flow in contact with a fluid containing another lithium compound, the lithium-6 atoms will migrate to the amalgam and the lithium-7 atoms will adhere to the lithium compound in the aqueous fluid. The most productive mixture of materials used in this isotope separation process was lithium hydroxide dissolved in water (Wilcox et al., 1983a).

Experiments were carried out by the Oak Ridge National Laboratory (ORNL) in 1951 and 1952 on an organic solvent to take the place of water. This was known as the organic exchange (or **Orex**) process. The **Orex** process was not pursued past the pilot plant stage due to technical difficulties (Napier, 1992). Initially, three separation techniques were considered and tested on a pilot scale; the **Elex** process, the **Orex** process, and the **Colex** process. Separate pilot plants were built to house the pilot studies. Buildings 9733-1 and 9202 were operated between 1951 and 1954 by ORNL as a development facility for the **Orex** process. The **Elex** Pilot Plant was housed in building 9733-2 and 9201-2 between 1950 and 1951, and used electrically-driven chemical exchange processes to separate the lithium isotopes. A production-scale plant for the **Elex** process started up in building 9204-4 and ran successfully from 1953 to 1956. However, this process was later found to be less efficient and was replaced by the **Colex** process (Napier, 1992).

**Colex** operations were located in buildings 9201-4 and 9201-5 and became the mainstay of Y-12's lithium separation operations. **Colex** proved to be the most efficient process at separating lithium isotopes, a process which required millions of pounds of mercury and which resulted in significant releases of mercury to East Fork Poplar Creek and to the air. The **Colex** operations were active from 1956 to 1963 and produced enough enriched lithium to fulfill future needs in the weapons program. Y-12 shut down the operations and scrapped the program in 1963. Several years were involved in the tear-down of production equipment and separation and recovery of mercury from the production facilities and equipment (Wilcox et al., 1983b). Most of the equipment still remains in Building 9201-4 (Napier, 1992).

Throughout the lithium isotope separation programs, large quantities of mercury were lost from the process equipment to the environment. Mercury loss estimates are shown in Table 4-4. The majority of the losses were a result of chronically leaky pipes and valves, equipment exchanges, accidental spills and transfers of mercury from storage flasks to process equipment. Many of these losses resulted in substantial quantities of mercury being released to the environment through the air, water, and sediment. The plant tried to control some of these releases by installing metal traps and trays in the drains and sumps inside the process buildings with some success (Napier, 1992). Mercury vapors were a constant problem inside the buildings, mostly from spills and off-gassing from the process equipment. Heated or warm mercury in process equipment was particularly susceptible to vaporization when not fully contained. Production operators installed large fans at the ends of the process buildings to increase the air flow rate through the process areas and thus reduce mercury airborne concentrations in the buildings. These fans are shown in Figure 4-5. This accelerated the mercury losses to air but provided a safer working environment for Y-12 employees (Napier, 1992). These fans were so large that the people living in Oak Ridge and surrounding communities could hear them running twenty-four hours a day. One study indicated that these fans were capable of 100 building air exchanges per hour (Perry et al., 1957). Another measure that was reportedly used to reduce

TABLE 4-4

## COMPARISON OF Y-12 MERCURY MATERIAL BALANCE ESTIMATES IN POUNDS

Source of Material Inventory and Losses	Estimate from UCC-ND Mercury Task Force, June 20, 1983	Estimate from Y-12 Report, June 9, 1977
Vouchered to Y-12	*	*
Accounted-for quantities:		
Returned unopened or rebottled and stored/sold	*	*
In lithium hydroxide tails, sold and stored	*	*
In Building 9201-5 scrap, sold	14,000	10,000
In Building 9201-5 sludge, removed and sold	174,000	111,000
As flasking overage given to GSA	*	*
In Building 9201-4 equipment, still in place	200,000	*
In sludges and sumps in Alpha-4 Building	250,000	100,000
In Building 9201-2 sewer pipe	800	**
Accounted-for total	*	*
Known lost and not accounted-for quantities total:		
Known lost to air	51,300	30,000
Known lost to East Fork Poplar Creek	238,944	470,000
Known lost to New Hope Pond sediment, Chestnut Ridge	6,629	7,200
Known lost to New Hope Pond sediments now in place	8,475	**
Known lost to ground, Building 9201-5 spill accident	49,853	49,853
Known lost to ground, seven other spills	375,000	**
Known lost to ground, Building 81-10 operations	3,000	**
Known lost total	733,201	557,053
Not-accounted-for total	1,291,855 <sup>c</sup>	1,880,699

\* These data are classified for security reasons

\*\* Data not available in 1977 report.

The numbers from the report are not accurate down to the one pound level.

Source: Wilcox et al, 1983.

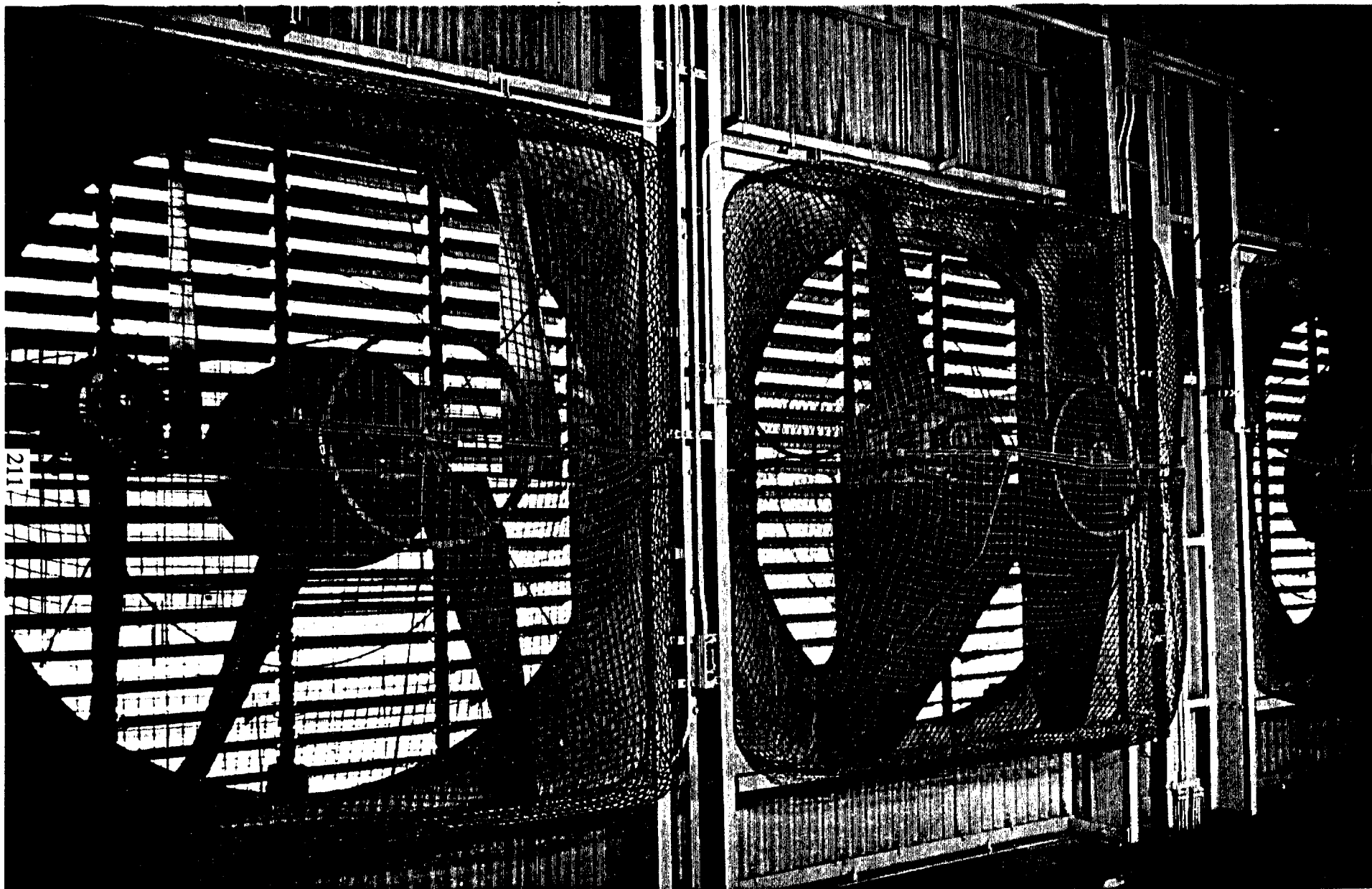


FIGURE 4-5  
LARGE VENTILATION FANS  
INSTALLED TO CONTROL  
AIRBORNE MERCURY LEVELS IN  
BUILDING 9201-5

REV: 04-30-93  
TDH-Okmg/4-93/4-4



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airborne concentrations was to keep the floors moist with water in areas of the buildings where there was prevalent mercury contamination.

The mercury and amalgam solutions were transferred under pressure through numerous pumps, pipes, flanges, valves, connectors, and seals (Perry et al., 1957). Numerous leaks occurred, and large spills were common as a result of the high frequency of equipment change-out. Mercury that spilled out onto the floors would readily vaporize into the air. The urgency for speed in the design of the plants did not allow for engineering of optimal features to handle leaks and spills. Limited time was available for mercury recovery, housekeeping and control of the mercury vapor contamination. By early January 1956, the air contamination of the Colex Plant buildings indicated a need to reduce the mercury vapor contamination in the Colex buildings and determine reasonable operating procedures and techniques to allow normal operation of the process while maintaining the mercury vapor concentration in the air below the accepted Maximum Allowable Concentration (MAC) of  $0.1 \text{ mg/m}^3$  (Perry et al., 1957). As described previously, large ventilations fans were added to reduce the airborne concentration.

Through various cleanup efforts, the average mercury vapor concentrations in both Colex buildings was reduced to levels below the MAC by March 1956. The 9201-4 building average dropped to below  $0.05 \text{ mg/m}^3$  in April of that year, and it seemed that an effective permanent solution had been found. The 9201-5 building average remained below the MAC for the remainder of 1956 and dropped below  $0.05 \text{ mg/m}^3$  in January 1957 (Perry et al., 1957). However, it has been mentioned that chronic leaks and spills existed throughout the Colex operations and were never completely corrected (Googin, 1993).

Since around 1984, a few studies have been conducted by Martin Marietta personnel, from predominantly ORNL and Y-12, to evaluate mercury contamination in the off-site environment that was caused by Y-12 operations. Some of these studies focused on characterizing the sources of mercury coming from the Y-12 plant and mercury releases into East Fork Poplar Creek (EFPC). One report evaluated mercury release rates from Y-12 process buildings that are known to have large deposits of mercury in and around the buildings and in the drainage system at Y-12 (Turner et al., 1985; Sanders, 1992). The largest discharges have been reported to be from buildings 9204-4, 9201-5, 9201-4, and 81-10. For example, measured release rates as high as 55 grams per day of mercury have been reported (Turner et al., 1985). These discharges have come from residual deposits of metallic mercury that eventually have been released through drainage systems that feed into EFPC and the former New Hope Pond. Some studies showed that New Hope Pond acted to filter or holdup 50% of the mercury that was being released to EFPC (Turner et al., 1985). Some of these discharge measurements are subject to considerable uncertainty, on the order of  $\pm 50\%$  or higher (Turner et al., 1985).

### Mercury Material Balance

The 1983 Mercury Task Force also developed estimates for the accounted-for quantities and known or unaccounted losses of mercury as a result of the Y-12 lithium operations. In an earlier 1977 report (Case, 1977), it was stated that 2.4 million pounds of mercury was lost or spilled from the Y-12 Plant (Wilcox et al., 1983b). However, it was stated in a later Y-12 report that the number should be correctly referred to as the amount of known mercury losses plus losses that are not-accounted-for. The distinction being the amount discharged to the air or into the creek is the known or accounted-for losses of mercury that were released to the environment. The unaccounted-for losses amount was estimated from inventory records, operation reports, and best guesses and represent the amount that may or may not have been released to the off-site environment.

The task force revised the 1977 data on the amount of mercury that could be accounted for based on updated information. Extensive reviews of available records of mercury transfers to Y-12 and return shipments from Y-12 were performed by the 1983 Mercury Task Force. The mercury-receiving operation was carried out by Rust Engineering for AEC, and all their records, which were transferred to the Federal Records Center in Atlanta, were subsequently destroyed as scheduled. Rust did not weigh the mercury upon arrival at Y-12 from the General Services Administration (GSA). Some original GSA flasks are shown in Figure 4-6. Mercury was sent through an underground pipeline that led to the Colex process buildings (Wilcox et al., 1983b).

The differences between the 1977 and 1983 figures are not large, as shown in Table 4-4. The earlier 1977 report was produced in just two weeks and was reportedly the best that could be accomplished in that time period. The revised 1983 number of 2.0 million pounds lost or not-accounted-for is lower than the 1977 figure (2.4 million pounds) not because of a change in the estimated receipts, but because of net increases in several of the accounted-for categories such as mercury holdings in some of the buildings. It is reported that the great majority of the accounted-for material is that quantity of mercury actually put into flasks and weighed, which is well known by those involved in the task force. The other numbers are the best estimates that can be established from records and inspections (Wilcox et al., 1983a).

The 0.7 million pounds in known losses includes losses to the air, water, and land. The losses to air and land are much higher than estimated in the 1977 report (494,000 vs. 87,000 pounds), and the losses to the water (creek) are lower (239,000 vs. 470,000 pounds). The rationale for each of the new estimates were developed in detail by the task force, and believed to be better estimated than the 1977 numbers (Wilcox et al., 1983b). The 1.3 million pounds not-accounted-for was established by the task force (shown in Table 4-4). One possible explanation for the differences is an estimate of 60,000 pounds that might be contained within the production building structures (walls, ceilings, floors, insulation, etc.). Since there is no analytical



FIGURE 4-6  
TYPICAL Y-12  
MERCURY FLASKS



sampling basis for such an estimate, the approach used was to follow the pattern of an Environmental Protection Agency (EPA) study of the chlor-alkali industry, which showed substantial losses of mercury each year caused by mercury being held-up inside building structures (Wilcox et al., 1983b).

Another explanation for an even larger part of the not-accounted-for mercury is the big uncertainty in the amount of mercury actually received at Y-12 (Wilcox et al., 1983b). An intensive study of the available records of mercury purchases was carried out during June 1983. Individuals who had worked in the Colex process were interviewed and GSA offices in Washington, D.C. were visited to try to get information on which to base a better mercury material balance. GSA furnished the mercury from the government's strategic material stockpile. However, no hard data on the actual amounts shipped or received could be located. It has been speculated that the difference between the quantity of mercury Y-12 was charged with and the quantity actually received might have been about 500,000 pounds, perhaps half of the 1.3 million pounds not-accounted-for. One former AEC official interviewed said "the shortage might be as high as the equivalent of 900,000 pounds." No additional information was located by ChemRisk, and may not be available (Wilcox et al., 1983a). One interviewee mentioned that the differences could be explained by the hot process used by the suppliers to fill each flask with mercury prior to shipment to Y-12. Each flask was filled with roughly 76 pounds of hot mercury and then weighed. Some mercury may have escaped as each flask was allowed to cool before it was sealed and shipped to Y-12 (Napier, 1992).

Each flask of mercury received by Y-12 was supposed to contain 76 pounds of mercury. Rust Engineering received and transferred the mercury into the pipeline, and Y-12 filled the cascades from the pipeline. Much of the salvage and transfer operations were located in Building 81-10. Interviews conducted by the 1983 Task Force with workers involved in the original operation, where mercury was transferred to the Colex operations, indicate that many flasks leaked, some were only partially filled, and some were even empty (Wilcox et al., 1983b).

#### **4.1.5 SRP and ICPP Recovery Operations (Transuranics)**

Two major material flows received by the Y-12 Plant from other DOE facilities originated from the Savannah River Plant (SRP) and the Idaho Chemical Processing Plant (ICPP). The Y-12 Plant began receiving enriched uranium for processing as described earlier in 1953 from the ICPP and in 1955 from the SRP. Estimates of total recycle material receipts from these sources up to 1984 are shown in Table 4-5.

Enriched uranium from the SRP was received at the Y-12 Plant in tanker trailers, usually containing 3,800 - 5,000 gallons in the form of UNH solution. After unloading and sampling, the SRP material was evaporated to some desired concentration level. Enriched uranium from

TABLE 4-5  
Y-12 ESTIMATED RECEIPTS OF RECYCLE MATERIALS (kg U)

Fiscal Year	SRP	ICPP	Total
1953	0	101	101
1954	0	217	217
1955	3	828	831
1956	0	744	744
1957	201	797	998
1958	258	898	1,156
1959	270	3,741	4,011
1960	6,395	769	7,164
1961	2,305	0	2,305
1962	2,701	775	3,476
1963	6,461	0	6,461
1964	2,977	771	3,748
1965	3,546	425	3,971
1966	3,467	1,408	4,875
1967	2,604	0	2,604
1968	2,097	394	2,491
1969	4,121	427	4,548
1970	2,045	108	2,153
1971	3,805	1,660	5,465
1972	4,716	415	5,131
1973	5,051	563	5,614
1974	4,599	0	4,599
1975	5,110	1,702	6,812
1976	4,320	195	4,515
1977	4,497	1,333	5,830
1978	2,070	525	2,595
1979	4,591	535	5,126
1980	1,510	0	1,510
1981	4,918	905	5,823
1982	5,728	577	6,305
1983	6,682	1,041	7,723
1984	5,776	2,868	8,644
<b>Total</b>	<b>102,824</b>	<b>24,722</b>	<b>127,546</b>

Source: Egli, 1985.

the ICPP was received as  $UO_3$ . After unloading and sampling, the ICPP material was dissolved in nitric acid to produce a UNH solution for further processing. At Y-12, the UNH was purified by solvent extraction, processed to metal, and returned to the SRP for fabrication into new fuel elements. The enriched uranium received from the ICPP was processed in a manner similar to that from the SRP and was eventually returned to the SRP for further use (Egli et al., 1985).

Y-12's use of materials from SRP and ICPP allowed for trace quantities of transuranics to accumulate in the uranium purification process operations. A significant portion of associated waste materials were transferred in a liquid solution to the S-3 seepage ponds. Recent studies of the S-3 ponds have shown various concentrations of these transuranics in the bottom sediments (Napier, 1982; Sanders, 1992).

#### 4.1.6 Zirconium Production Operations

The first successful hafnium-free zirconium separation facility was developed and operated by Y-12 Plants in early 1950. The nuclear reactor industry developed a need for zirconium which was free of the contaminant hafnium for reactor fuel development. Zirconium resists corrosion in water of high temperatures. However, in a reactor, hafnium absorbs neutrons and "poisons" nuclear fission reactions. Separation of zirconium from hafnium by solvent extraction of the metal thiocyanate complex was achieved in the laboratory and demonstrated on a pilot plant scale in January 1956. Because of developments made by the Production and Research Divisions during the production period, substantial cost reductions were realized by modification of the original process that increased the percentage of zirconium recovered in extraction by the addition of stripping columns and the substitution of phthalic anhydride for salicylic acid in the purification of zirconium oxide. This facility had the capacity to produce 150,000 to 200,000 pounds per year of essentially hafnium free zirconium as purified oxide.

Zirconium tetrachloride was the most expensive single item in the plant operations and it was essential that the zirconium loss be kept to an absolute minimum. Extraction columns, using 2% hafnium-zirconium, were capable of recovering between 97% and 99% of the zirconium.

Increased extraction yields improved the recovery of the by-product of highly enriched hafnium by 90%. The hafnium left over from the processes was considered to be of sufficient value to be reclaimed and therefore was recovered and stored. Some of the hafnium may have been sold to outside buyers. As authorized by the Atomic Energy Commission, hafnium tailings collected from the zirconium extraction plant from May 1, 1950 until March 25, 1951 were re-run in order to concentrate the hafnium to a minimum of 95% of the total metals. Based on 2% contained hafnium in the crude tetrachloride feed, a quantity of approximately 5,500 pounds of hafnium was expected. Because no precise analytical methods had been developed for the analysis of hafnium, an exact measure of the quantity was not possible. Estimated gross weight

of the collected hydroxide cake and analyses of spot samples indicated that the quantity available should be approximately 5,000 pounds. Assuming that 80% recovery could be obtained during processing, the Commission was informed that they could expect to recover approximately 4,000 pounds of hafnium as hafnium tetrachloride (Googin et al., 1951).

After rewashing the hafnium hydroxide cake to remove sulphates, the estimated net weight and spot analyses indicated in excess of 5,000 pounds of contained hafnium. The washed cake was dissolved for feed to the extraction plant. Analyses of solutions indicated that an excess of 4,000 pounds of hafnium was fed to the extraction system for recovery (Googin et al., 1951).

#### 4.1.7 Y-12 Waste Disposal Operations

Y-12's production activities have generated a significant amount of liquid and solid waste. Chemical and metallurgical processes have resulted in large quantities of waste such as nitric acid, PCB oils, perchloroethylene (or tetrachloroethylene), depleted uranium, trace amounts of transuranics in the S-3 seepage ponds, and trace metals such as arsenic found in coal. The generation of these wastes required Y-12 to use several areas located in and around Y-12 for liquid and solid waste burial. Burial grounds west of the plant were used extensively for burying solvents, depleted uranium, and other materials. This section describes some of the major waste disposal operations used by Y-12 for handling their waste.

##### Early Waste Activities

During the first years of Y-12 operations, liquid disposals such as nitric acid solutions were screened for uranium concentrations and then released with little or no treatment into drains and sumps which fed directly or indirectly into East Fork Poplar Creek on the south side of the plant (Griffith, 1957). During these years, Y-12 attempted to recover as much uranium as economically or practically feasible. Most of these releases involved normal or depleted uranium which had limited value. If enriched uranium concentrations were less than 10 ppm (parts per million) in a waste or raffinate solution, then the solution was released to the East Fork Poplar Creek (Googin, 1993).

Later on in the 1950s, during uranium salvage recovery operations, one report mentions that if uranium concentration in the liquid waste were 50 ppm or less, then the material was released to the sanitary or storm sewers (Pletz, 1954). A large portion of these releases came from the chemical recovery and salvage operations that were used to recover and recycle uranium from the early calutron enrichment program and early weapons production operations. Buildings 9202, 9203, 9206, 9212, and the alpha and beta buildings were the primary facilities for some of these operations, and handled large quantities of uranium compounds such as uranyl nitrate and uranium oxides. Some of the largest releases as shown in Table 4-3 occurred during the first year (1944), which led to better controls to minimize liquid releases. For the higher

enriched uranium operations, tight controls were strictly adhered to since they did not want to lose such a valuable product. Collection traps were added to the drains and containment berms were added to some of the floors to better contain uranium solutions that were released from spills and leaking equipment (Googin, 1993; Griffith, 1957).

#### Depleted Uranium Volume Reduction

Y-12 depleted uranium operations have generated a large volume of industrial waste, including unreclaimable materials from uranium salvage operations and other materials which were of little value. As part of the depleted uranium fabrication, large quantities of metal turnings and chips needed to be treated and disposed of in one of the Y-12 burial grounds west of the plant, off of Bear Creek Road. Due to the sheer volume of the material, Y-12 undertook a study to examine three techniques that could be used to significantly reduce the volume of disposed material. They first considered pressing the uranium chips into briquettes and then recasting these into uranium metal. This approach was not considered cost effective (Sanders, 1961). The second approach was to dump the dry chips into a large burial pit and cover them with soil. However, this approach presented a serious fire threat during handling operations due to the pyrophoric nature of the uranium chips. A third and final approach studied eventually became the accepted method and was used for a number of years up into the 1980s. This approach to reduce the volume involved the transfer of the chips to the Bear Creek Burial Grounds submerged in water, reducing the threat of the chips spontaneously catching fire from sparking. The chips were then placed in shallow burial pits, ignited, and allowed to burn. This technique was very effective in substantially reducing the volume of depleted uranium waste (Sanders, 1961).

Since these burnings were conducted outside without the possibility of filtering the effluents, Y-12 decided to run experiments to determine if the burning of these chips caused significant airborne contamination and potential dispersion of the contamination to off-site locations. Several air sampling devices were set up around the area during test burnings to determine the airborne concentrations. Samples were collected on filter papers and radioactive particulate fallout trays. Test results over a three month period indicated that most of the airborne radioactivity stayed within a 100 foot radius of the burning uranium pits (Sanders, 1961). ChemRisk was unable to find further information substantiating these conclusions or the techniques involved in performing the air sampling. These burnings were routinely performed, so therefore health physics personnel performed periodic grab air samples to assess the extent of airborne contamination. Sample results from these measurements were not located during the investigation. However, one retired employee maintained that some of the results do exist somewhere at Y-12 (Sanders, 1992).



### S-3 Ponds

In 1951, Y-12 constructed four seepage ponds to provide a holding place for millions of gallons of nitric acid and other chemicals containing low-levels of radioactivity, such as uranium, plutonium, technetium, and neptunium. These were called the S-3 ponds. These ponds were built at the west end of the Y-12 plant and covered approximately three acres. The typical annual quantity of liquid waste entering the four interconnected ponds was estimated in one report to be approximately 2.7 million gallons. An additional 4 to 6 million gallons of rain water were deposited in the ponds each year. The nominal volume of all four ponds was approximately 10 million gallons, with each pond containing about 2.5 million gallons. Liquid wastes that were pumped or dumped into these ponds consisted of contaminated wastewaters, dilute acids, coolants, caustic solutions, and bionitrification sludges. The northeast pond contained mainly nitric acid, other concentrated acids, aerosol cans, and small propane containers. Wastes were transferred to the ponds by surface dumping from large tank trucks or through a series of underground pipes connected with production salvage areas. The eastern ponds were connected to the western ponds by overflow pipes. The S-3 ponds are believed to have been a source of groundwater contamination of organic solvents and radionuclides.

One report that was reviewed indicated that three million gallons per year evaporated from the ponds, and that an additional 4.7 million gallons per year were removed from the ponds through ground seepage. Each pond was 200 feet wide, 200 feet long, and 15 to 20 feet deep. In the early 1970s, a process was installed to recycle some of the nitrate wastes and reduce the volume that was transferred to these ponds (UCC-ND, 1983c).

Around 1971, the Y-12 Plant developed processes for recycling some nitrate wastes from the uranium purification process. It was agreed that a process would be developed to biologically destroy some of the nitrate wastes. The remaining plant wastes from other operations were not recyclable and were to be discharged into the S-3 ponds until other disposal processes could be developed. Laboratory tests indicated that 30% of the nitrate could be recycled as nitric acid and 35% could be recycled as aluminum nitrate. The remaining nitrates (35%) were biologically destroyed in a stirred-tank denitrification process. The nitric acid waste came from condensates produced in evaporation processes within the Y-12 Plant. The waste stream contained 3 to 10% nitric acid, and the major impurities were approximately 100 mg/L of chlorides, 100 mg/L of fluorides, and 300 to 1,000 mg/L of organics. To recycle the waste, the chlorides and fluorides had to be removed. The waste was passed through a vaporizer containing a mixture of 23.8 weight % aluminum nitrate nonahydrate, 53.9 weight % calcium nitrate tetrahydrate, and 23.3 weight % water, which removed the fluoride ions and vaporized the feed to a distillation column (Napier et al., 1987).

In summary, wastes originally entering the ponds included the following:

- Acidic nitrates from uranium recycle,
- Plating and pickle bath wastes,
- Mop waters from operating areas, and
- Aqueous machine coolants such as PCBs and perchlorethylene.

From 1973 to 1978, hazardous material minimization and substitution efforts at the Y-12 Plant included these seven major projects:

- Recycle of nitric acid,
- Recycle of aluminum nitrate,
- Substitution of chromium in cooling tower water,
- Reduction of the number of machine coolants used,
- Extension of the lives of acid pickle baths,
- Minimization of depleted uranium release to East Fork Poplar Creek, and
- Minimization of hydrogen fluoride usage.

(Napier, 1989)

#### New Hope Pond

In early 1960, the Y-12 Plant recognized the need to equalize the pH of effluents from the plant and to develop a settling basin for heavy metals and solids (e.g., uranium and mercury) from Y-12's industrial complex. The conceptual design which was developed was a water distribution system for East Fork Poplar Creek which would permit a 24-hour mixing time of the plant's 24-hour water discharge of approximately 8 million gallons. Prior to this, the liquid effluent from the Y-12 Plant discharged into the head waters of the East Fork Poplar Creek, which in turn flowed directly through the city of Oak Ridge (Sanders, 1972).

In 1963 a 5.2-acre pond known as "New Hope Pond" was constructed at the east end of the plant at a cost of \$150,000. The pond was constructed with a proportional sampling system on the inlet and outlet side to monitor for pH, gross alpha and mercury concentrations. Early health physics and Y-12 quarterly reports list sampling results for the ponds. For example, when the pond was first built, it was equipped with a continuous pH recording monitoring system telemetered into the Plant Emergency Control Center where abnormal conditions of the pond could be monitored (Sanders, 1972).

In the early 1970s, additional analytical capabilities to improve the control of pollution were added to the pond for continuous automatic analysis. These additional analytical tests were for chemical oxygen demand, hexavalent chromium, conductivity, and nitrates (Sanders, 1972).

The flow of East Fork Poplar Creek was recorded continuously by a depth recorder which was calibrated against a specially designed weir. The pond design also included a water distribution system with fifteen separate outlets for the equal dispersion of liquid into the main body of the pond. It was hoped that this approach would disperse any contamination in the inlet water and lead to higher dilution prior to release to EFPC (Strohecker, 1993).

The pond also had an oil-removal system consisting of a small basin of approximately 25 x 40 feet at the inlet of the large pond. The upstream end of the basin was fitted with a weir which floated about one inch below the surface of the creek. Oil and small debris floated over the weir into the basin, where oil and trash were retained by a skimmer weir at the basin outlet. The excess water flowed under the outlet weir through a culvert into the large pond. Accumulated oil and debris could then be periodically pumped to a trailer for disposal by an oil biodegrading technique (Sanders, 1972).

By 1971, control had become less effective as a result of decreased retention time of the water caused by silt generation primarily as a result of numerous construction activities at the plant site (Sanders, 1972). In 1972, Y-12 decided to dredge the pond sediments to remove the large amounts of silt and some of the contamination that was present. The debris removed from the pond was buried on the top of Chestnut Ridge and covered with topsoil.

The construction of New Hope Pond was an attempt to allow for pH adjustment to the East Fork Poplar Creek and to reduce releases from both steam plant operations and mercury processes, and is known to have been a significant pathway for off-site migration of hazardous materials (Sanders, 1972; Napier, 1992; Strohecker, 1993).

### Burial Grounds

The low-level radioactive waste burial areas are located in Bear Creek Valley about 1.8 miles west of the main plant site. The areas are designated Burial Ground 1-A, Burial Ground 2-B, and Burial Ground 2-C. The grounds are enclosed by a fence with entrance gates that are kept locked at all times and are opened only by authorized persons as required for entry or exit (Sanders, 1977). Trenches in Burial Grounds 1-A and 2-C are about 18 feet deep by 26 feet wide. The excavated material has been used to cover the deposited contaminated waste material frequently so as to avoid prolonged exposure of the waste-filled trenches. The excess dirt provides a final trench cover that varies in thickness from 5 to 15 feet. When a trench is filled to capacity, it is leveled and is seeded with grass, and pine seedlings are planted on it for additional erosion control.

Burial Ground 1-A has been used for the burial of low-level radioactive material. Solid waste material disposed of in this area includes materials contaminated with depleted uranium, for example: wood, paper, plastics, particulate filters, carbon, mixed-metal machine turnings, metal

drums, and small quantities of metal with low economic value. This burial site is 3 to 30 feet above the water table, depending on weather conditions. The plant roughly generated about 100 tons (760 m<sup>3</sup>) per year of the low-level radioactive waste that is deposited in Burial Ground 1-A (Sanders, 1977).

Pure, depleted uranium, which is accountable and recoverable, is stored at Burial Ground 2-B. The trenches are about 15 feet deep and 10 feet wide. The material is compacted and covered daily. The area is 80 to 110 feet above the water table the year round. The plant has roughly generated about 800 tons (608 m<sup>3</sup>) per year.

Contaminated materials such as enriched uranium and natural thorium have been disposed of in Burial Ground 2-C. The waste materials are the same types as those listed for Burial Ground 1-A. This burial area is from 2 to 6 feet above the water table, depending on the season. The plant has roughly generated a quantity of about 500 tons (380 m<sup>3</sup>) dumped in the trench (Sanders, 1977).

The classified-material disposal area is inside the main plant. It has been used to dispose of classified materials that are radioactive. The trenches are about 19 feet deep by 8 feet wide. The materials disposed of in this pit have consisted of classified forms made from uranium, thorium, and other materials. The materials were covered on the same day of disposal. The minimal thickness of the final cover is 6 feet. The operation required the use of about 0.03 acre per year. A total of 250 tons (160 m<sup>3</sup>) of classified materials have been disposed of per year. This area is 80 to 100 feet above the normal water table (Sanders, 1977).

Some radiological monitoring of surface and underground water in and around the burial ground has been performed on a routine basis (Sanders, 1977). The types of monitoring and frequency of samples taken in these areas have increased over the years. Today, numerous groundwater monitoring wells are located throughout and around these old disposal areas. These are potentially a good source of data for estimated off-site releases of contaminants.

#### Y-12 Steam Plant Burial

During 1965 and 1966, classified materials were buried in the coal yard area and are now underneath the Y-12 coal pile. Available information indicated that disposal took place in a pit 161 feet long, 14.5 feet wide and 15 feet deep. The centerline of the pit is located approximately 85 feet south of the center of Second Street, with the eastern end of the pit located approximately 130 feet from the west end of 9401-3. The long axis of the pit parallels Second Street. An estimated 2,000,000 kg of material is buried in a trench under the coal pile. Approximately 1,750,000 kg of the above is depleted uranium and depleted uranium alloys. One report mentioned that the location of a classified waste pit under the coal pile may present

environmental problems because of its proximity to an acid waste source and its probable location below the groundwater table. This report also mentioned that a follow-up study was planned to evaluate the classified waste pit to determine the condition of the uranium waste and its impact on the surrounding earth and groundwater (MMES, 1983). Additional information regarding these studies was not identified or found during our investigation.

#### Y-12 Disposal at ORNL Burial Site

Use of ORNL burial sites by the Y-12 Plant predates Y-12's available records that start in February 1957. Since the Y-12 Plant Bear Creek Burial Ground didn't operate at a large capacity until 1955, disposals from Y-12 to ORNL may have been initiated during the Manhattan Project. It would appear from the available records that disposals were at a high annual rate (> 500 tons/yr) until early 1961, at which time the Y-12 Plant began making beryllium, thorium, and considerable uranium disposals in the Bear Creek Burial Ground area located west of the plant. Many disposals to the ORNL burial site contained classified materials associated with the Y-12 Plant's weapons production program. According to one report, numerous documents were reviewed by plant personnel to develop inventories of the quantitative and material descriptions of those items buried at ORNL up until 1984. Information regarding possible disposal after 1984 was not identified (MMES, 1984).

Materials sent to ORNL for disposal are predominantly suburanic and transuranic radionuclides, biological specimens, and contaminated debris. The total quantity sent to ORNL for which records are available is approximately 4800 tons. Quantities predating available records could be greater (MMES, 1984). The materials disposed of in ORNL burial sites were categorized into twelve major categories, which was consistent with those categories reported in other disposal reports. The twelve major categories were:

Acids	Debris	Radionuclides
Asbestos	Heavy Metals	Thorium
Beryllium	Inorganics	Unknown
Biological	Organics	Uranium

(MMES, 1984)

#### 4.1.8 ORNL Operations at Y-12

##### Stable Isotope Production

The earliest reported separation of stable isotopes was in 1945. The 2.5-hour half-life isotope of nickel, nickel-65, was produced by the irradiation of stable copper-65. In order to do this, the stable isotopes of copper, copper-63 (69.1%) and copper-65 (30.9%), were

electromagnetically separated in a calutron unit. The copper-65 was subjected to radiation to produce nickel-65. Use of stable isotopes in positively determining the mass of artificially produced radionuclides has been a major contribution to modern physics and chemistry, with over 80 isotope masses confirmed (Compere et al., 1991).

After World War II, nuclides other than uranium were separated with the few calutrons left in operation. The successful experiment noted above that separated the isotopes of copper led to a subsequent interest in separating, according to their atomic masses, pure isotopes of various stable elements in sufficient quantities to determine their physical properties and to provide for their use in fundamental research (Keim et al., 1951; Compere et al., 1991).

No isotopes other than deuterium had been separated in appreciable quantities by the mid-1940s. This meant that many of the physical constants of different isotopes were subject to conjecture, but not to physical verification, including such fundamental properties as the measured mass, natural occurrence or abundance, physical properties, and neutron cross sections of different nuclides and the radionuclides they produce upon irradiation. Currently, these matters are well understood and have been subjected to considerable experimental verification. Between 1946 and 1982, isotopes of the following elements were electromagnetically separated and purified using the last remaining alpha and beta calutron units at Y-12:

Lithium	Beryllium	Boron	Carbon	Nitrogen	Oxygen
Magnesium	Silicon	Sulfur	Chlorine	Potassium	Calcium
Titanium	Vanadium	Chromium	Iron	Nickel	Copper
Zinc	Gallium	Germanium	Selenium	Bromine	Rubidium
Strontium	Zirconium	Molybdenum	Ruthenium	Palladium	Silver
Cadmium	Indium	Tin	Antimony	Tellurium	Barium
Lanthanum	Cerium	Neodymium	Samarium	Europium	Gadolinium
Terbium	Dysprosium	Erbium	Ytterbium	Lutitium	Hafnium
Tantalum	Tungsten	Rhenium	Osmium	Iridium	Platinum
Mercury	Thallium	Lead	Bismuth	Uranium	

In 1945 and 1946, runs were used to separate small amounts of copper, iron, chromium, nickel, carbon, lead, magnesium, molybdenum, silver, calcium, zirconium, cadmium, lithium, strontium, indium, zinc, silicon, selenium, bromine, uranium, and bromine. With additional experience, both the range and purity of isotopes separated was increased as chemical techniques and equipment improved, and by June 1950, 550 samples of isotopes, including 173 isotopes of 43 elements, had been collected. Most of these were very small samples, usually on the order of a few milligrams. Because of the very small amounts of material, special handling and recycling techniques were developed and a large number of effective microtechniques grew out of the methods used (Compere et al., 1991).

A major portion of the work performed in the late 1940s and early 1950s was directly related to developing nuclear technologies, including the specific information needed for engineering background and production of small amounts of very pure stable isotopes as starting materials for the production of radioactive isotopes. In 1948, scientists studied the preparation of high specific activity iron-59 using iron-58 as the starting material. Previous preparations were characterized by a low specific activity, due to the very low abundance of iron-58. The material was activated using the Clinton pile at X-10. The work was continued as techniques improved until gram quantities of high-purity iron-58 were produced. A significant amount of effort was directed towards the development of purification techniques for important materials, such as beryllium or hafnium and zirconium. In the case of beryllium-10, this was important because it permitted accurate definition of the beta ray spectrum of beryllium-10, a factor reasonably important in the design of nuclear reactors, in which beryllium can be neutron activated (Compere et al., 1991).

Y-12 separated cadmium-113, which has a neutron capture cross section of 24,000 "barns," compared with the one to two "barn" values for other cadmium isotopes. A barn is a unit of measure of the effective cross-sectional area of an atom that is available for some type of nuclear interaction with an incoming neutron. A barn is defined as  $10^{-24}$  cm<sup>2</sup>.

A number of laboratories requested separation of the isotopes of lead in order to determine whether lead-204 was an  $\alpha$  emitter and if it had an isomeric state. Several preparation runs at Y-12 were made to make these determinations. The isotopes of zinc were also separated for both fundamental and radioisotopic studies. A large number of production runs were devoted to the separation of chromium isotopes for research into nuclear engineering properties, radionuclide production, and fundamental physical and chemical properties. A number of production runs were also performed to collect platinum isotopes, including one isotope which was believed to be present as platinum-200 (Compere et al., 1991).

Chemical separations were a major consideration in several parts of the stable isotopes program. Substantial amounts of charge material had to be prepared and recycled in a very pure form. Sometimes, as in the case of the rare earths, it was impossible to purchase these materials in a chemically pure form (Compere et al., 1991). Because electromagnetic separations were inherently inefficient, that is, significant amounts of the feed material being separated were deposited on calutron inner surfaces, efficient methods for purifying and recycling materials were necessary. Product materials which deposited in collection pockets had to be recovered, separated from the pocket materials, and purified in a suitable chemical form for storage. Additionally, isotopes which were "borrowed" by researchers were generally returned. Returned materials were purified and returned to the inventory. It was reported that Y-12 published a list of the various recovery, refining, and recycling methods commonly used for separating isotopes of various elements (Compere et al., 1991). This report was not located during this investigation.

A number of naturally-occurring radionuclides, including potassium-40, rubidium-87, tin-124, lutetium-176, rhenium-187, and samarium-146, were also separated electromagnetically. This permitted identification of the radionuclide as well as precise definition of its half life.

In 1958, the building containing the remaining two beta calutron tracks (Building 9204-3, Tracks 5 & 6) became available and was assigned to stable isotopes separation. One track of calutrons was modified with heavy iron yokes so that individual sections could be operated to separate the isotopes of different elements. Sophisticated chemical and machining facilities were provided to facilitate chemical separations of isotopes. Improved equipment decreased the size of the staff required to maintain and operate the equipment, providing better separations at a lower cost. Use of the pilot units was gradually abandoned (Compere et al., 1991).

As compared with other Y-12 operations, the stable isotope separation program handled relatively small quantities of hazardous materials. It was found that most of the elements processed were handled in milligram or gram quantities. Relevant information regarding the potential for any significant off-site releases were not identified during the investigation. None of the information that was reviewed suggested that these operations were a significant source of off-site releases of chemicals or radionuclides.

Currently, the stable isotopes program is the Department of Energy's largest involvement with commercial markets; stable isotopes are used in preparing the specialty isotopes used in medical research, as tracers, and in a number of physics applications. Every year, the ORNL program produced and sold millions of dollars worth of these materials (Compere et al., 1991). Calutrons were last used for stable isotope production at Y-12 in 1990, when several special experiments were conducted (Googin, 1993).

#### ORNL Biology Division at Y-12

The Biology Division operated by the ORNL historically has studied the potential adverse health effects of hazardous substances. Almost all the work of the division has been experimental and utilizes mammalian and sub-mammalian systems to obtain data for predicting and understanding hazards to human health through laboratory animal exposure studies. The Biology Division of the ORNL consists of Buildings 9207, 9210, 9211, 9220, 9224 and 9208 and many smaller buildings located within the Y-12 Plant Area (Moore, 1986).

By autumn of 1946, the scope of a proposed biological and biomedical program for Oak Ridge led to the organization of the Biology Division; and Alexander Hollaender, a radiation biologist on the staff of the National Institutes of Health (NIH) in Washington, D.C., was appointed as the Division's first Director. Although the Division's main objective was to do fundamental research in radiation biology, the division was involved in research-related educational programs,



interdisciplinary cooperative projects, and prompt communication of scientific data (White, 1966).

A major turning point in the history of the Biology Division came during the 1960s. With rapid growth characterizing the development of the nation's space and health-research efforts, it became apparent that the Division was uniquely qualified to contribute to the biomedical aspects of programs administered by both the National Institutes of Health (NIH) and the National Aeronautics and Space Administration (NASA). Interagency agreements between the USAEC and other government agencies led to three cooperative projects: Space Biology (NASA), Molecular Anatomy (NIH), and Basic Carcinogenesis (NIH) (White, 1966). Since that time, the division has been involved in numerous other projects of this magnitude such as radiation microdosimetry. It was decided that the most important contributions the division could make to the AEC-NASA projects would be to assess the importance of the radiobiological aspects of space flight and to determine the possible additive or synergistic action of the attendant physical factors, such as weightlessness and vibration, with low doses of radiation. Thus, the division began two main projects: assessment of the relative biological effectiveness (RBE) of protons of various energies in a systematic series of biological materials ranging from microorganisms to human cells, and development of in-flight experiments for satellites which could answer the question of whether there are, in fact, any synergistic effects of radiation or of the other mechanical and physical parameters encountered in space flight (White, 1966).

The first of two major National Cancer Institute NCI-AEC projects, the Zonal Centrifuge Development Program, was initiated in 1962. This was a program aimed at developing virus-isolation techniques related to the study of the causes of human cancer and of the relation of radiation to human disease, especially leukemia. In 1963, the NCI-AEC had a second joint project which was the Carcinogenesis Program. Its purpose was to expand research already under way in the division on the interrelationships of chemical agents in the production of tumors (White, 1966).

Hazardous substances which have been investigated are both physical and chemical. Among the physical agents, major interest has been focused on the health effects of gamma rays and fission neutron radiations on animals with particular attention to the carcinogenic responses to low dose levels and to the relative biological effect. Among the chemical agents, special emphasis is placed on problems associated with the emerging energy technologies such as nuclear power. Since the substances to which people may be exposed tend to be complex mixtures of chemicals, the division's activities concentrated on evaluating and understanding the toxicological interactions when mammals are exposed to multiple substances, either concurrently or successively (White, 1966).

Laboratory work within the division includes the use of hundreds of chemicals that are classified as hazardous or toxic and are handled in small or laboratory-size quantities. Small quantities

of short-lived radionuclides are used as tracers. Most of the animal experimentation consists of exposing mice and rats to known amounts of carcinogens, mutagens, and teratogens. Additional work in the division includes research bacteria viruses, and recombinant DNA. The Biology division has maintained more than 200 active laboratories that have carried out a great variety of scientific research studies in microbiology, biochemistry, genetics, and work with small-animal tissue. Laboratory equipment may include microscopes, centrifuges, analytical devices, extraction equipment, constant temperature baths, sterilizers, refrigerators, freezers, glove boxes, and many specialized pieces of equipment designed for specific research activities. Hoods and biological safety cabinets are provided in laboratories. Building 9210 is used for mouse living quarters, maintenance, and breeding for genetic research. This building is referred to as the "mouse house" and contains a very limited quantity of hazardous materials, most of these have been of a biological nature, such as animal excrement (Triplett, 1993).

Table 4-6 depicts the radiation sources utilized within the Biology Division at Y-12 over the last few decades. These facilities are used mainly for experimental irradiation of mice, rats, insects, and mammalian tissue. The sources are cesium-137, cobalt-60, and californium-252 sealed sources which consist of welded metal capsules. The greatest potential hazard is a direct exposure to an unshielded capsule. This is unlikely to have presented a contamination hazard except in the event of a broken capsule weld. No information was found during our study to indicate that this type of accident has ever occurred in these facilities. The source rooms have lead-lined walls and doors at all locations where there is any potential for personnel or public exposure, and appropriate alarms (Moore, 1986).

Three inhalation facilities in the division have been used for exposing mice and rats to toxic gases, vapors, or aerosols inside closed chambers. The inhalation facility is on the fourth floor of Building 9211, occupying a 3,000 ft<sup>2</sup> portion of building. There are eight large inhalation chambers and four small chambers in one of the rooms. Each room containing inhalation chambers has an associated room in which animals are maintained prior to and after inhalation. The inhalation chambers are all made from stainless steel and are all of similar design. They are four-sided, box-like chambers which are pyramid-shaped on the top and bottom. A door with a large plexiglass window covers almost the entire front face of each chamber. Neoprene gaskets seal the doors when closed. Animal cages are placed on shelves in the bottom part of the chambers. The air-inhalant mixture enters through the top of a chamber, passes over the animal cages, and leaves through the bottom. Exit air passes through a small, tested HEPA filter and is then exhausted through tested HEPA-filtered roof vents (Moore, 1986). The small chambers and some of the large chambers use room air. A long perforated steel tube about 2 feet long with a valve at the bottom allows room air to enter through the top of each of these chambers. A substance inlet tube below the valve allows the inhalant vapor or aerosol to enter and mix with the inlet air stream. These rooms operate under 1/2 to 1-1/2 inches negative pressure of water with respect to the room (Moore, 1986). Other inhalation facilities have

TABLE 4-6

## CHEMICAL RADIATION SOURCES IN BIOLOGY DIVISION AT Y-12

Building	Room	Radioactive Source	Quantity (curies)	Dose Rate (at 1 m, unshielded) (rad/hr)	Type of Radiation	Main Irradiation Use
9207	4034A	$^{137}\text{Cs}$ $^{137}\text{Cs}$	1300 65	430 21.5	$\gamma$	mice, rats mice, rats
9207	4040	$^{137}\text{Cs}$	0.83	0.3	$\gamma$	mice, rats
9210	341	$^{137}\text{Cs}$	2.5	0.8	$\gamma$	mice
9770-2		$^{137}\text{Cs}$	80	26.5	$\gamma$	mice
9207	127	$^{60}\text{Co}$	3.75	5	$\gamma$	insects, mammalian tissue
9983-17	Trailer	$^{252}\text{Cf}$	3.91	0.8	neutrons and $\gamma$	mice, rats, mammalian tissue

Source: Moore, 1986.

involved the use of ethylene oxide, 1-3-butadiene, cadmium containing aerosol, beryllium compounds. Again, these materials are handled in small quantities and these facilities used HEPA filtration in their ventilation systems (Moore, 1986).

Many volatile organic solvents have been used in the Biology Division laboratories. Most of these solvents are toxic, flammable, or both. They have been used normally in pint or quart bottles except for some one-gallon bottles of commonly used chemicals such as acetone, toluene, ethanol, methanol, and a few others. Volatile hazardous solvents are used only in hoods which exhaust to the roof of the building. Radionuclides are commonly used for research. Table 4-7 lists the type, typical quantity purchased annually, and the properties of radionuclides which are frequently used as tracers in the division (Moore, 1986).

No significant accidents or incidents involving the release of hazardous materials associated with the Biology Division activities were identified during this investigation. In the past, small quantities of laboratory-type chemicals (i.e., dilute acids, organic, and possibly low levels of radionuclides) have been disposed of in sinks and drains and then to the storm sewers.

According to one employee interviewed, these disposals were routinely monitored and to their knowledge, water samples never exceeded administrative concentration action guidelines which have also been set below regulatory standards. However, the nature of the sampling techniques would need to be investigated further to render any valid conclusion about the significance of off-site releases from these past disposal practices.

Routine releases of chemicals or radionuclides from the Biology division do not appear to have contributed significantly to off-site releases from the Y-12 site.

Ventilation supply systems in the Biology division buildings provide a flow of cooled or heated filtered air through all rooms, offices, and laboratories in the division. Many exhaust systems serve laboratory-type hoods and biological safety cabinets in the laboratories in which room air is swept past partially enclosed work areas and exhausted to the outside. Since a large part of the research consists of processes in which biological or chemical hazards may be involved, the ventilation and exhaust systems play an important role in reducing hazards from airborne contaminants (Moore, 1986).

Air intake vents are located on the roofs and sides of the Biology division buildings. Each of these vents is equipped with single tested HEPA or Hospital-Grade filters. Some of these have been recently installed. Hoods, laboratories, offices, and other rooms in each building exhaust through ducts leading to roof vents. Some areas in the buildings are designed specifically for handling chemical carcinogens, mutagens, teratogens, and radioactive materials. The exhaust air from these areas and from other rooms and hoods where these materials are handled passes

TABLE 4-7

## RADIOISOTOPES USED BY THE BIOLOGY DIVISION AT Y-12

Radioisotope	Type of Emission	Radioactive Half-Life (days)	Approximate Annual Quantities (mCi)
<sup>3</sup> H	beta	4.47 x 10 <sup>3</sup>	350
<sup>14</sup> C	beta	2.1 x 10 <sup>6</sup>	1.5
<sup>24</sup> Na	beta, gamma	0.625	NA
<sup>32</sup> P	beta	14	93
<sup>35</sup> S	beta	88	30
<sup>51</sup> Cr	gamma	28	NA
<sup>59</sup> Fe	gamma, beta	46	NA
<sup>125</sup> I	gamma	60	185
<sup>131</sup> I	gamma, beta	8	60
<sup>109</sup> Cd	x-rays	453	1.0
<sup>7</sup> Be	gamma	53.6	3
<sup>45</sup> Ca	beta, gamma	165	15
<sup>239</sup> Pu	alpha, beta, gamma	8.9 x 10 <sup>6</sup>	1 *

NA Information not available at the time of this report.

\* Every few years 1 mCi of <sup>239</sup>Pu was injected into mice and studied. The <sup>239</sup>Pu is prepared at X-10 and shipped to Y-12. <sup>239</sup>Pu normally is not stored by the Biology Division at the Y-12 site.

Source: Moore, 1986.

through tested HEPA filters and charcoal filters on the roof. Laboratories that handle volatile radioactive compounds such as iodine-125 are vented and filtered with special charcoal adsorbing filters (Moore, 1986). Relevant records that describe early operations were not found during this preliminary investigation.

HEPA filters on the air intake vents in the division are rated at a 1000 cfm flow rate with an efficiency of 99.97% for particles 0.3 micrometers in diameter. As of 1986, these filters were replaced with 2000 cfm tested Hospital-Grade filters with an efficiency of 95% except for intake filters for Building 9210. Air intake filters for that building have continued to operate with a 1000 cfm, 99.97% efficient HEPA filters. All exhaust filters in the division are rated at 1000 cfm with a pre-installation efficiency of 99.97%. HEPA filters are tested using an aerosol of polydispersed (0.3-1 $\mu$ m) dioctylphthalate (DOP), and are required to have efficiencies of at least 99.95% after testing. The Hospital-Grade filters are tested similarly for at least 95% efficiency (Moore, 1986).

#### ORNL Engineering Technology Division at Y-12

Although the name of the ORNL Engineering Technology Division has changed over the years, the primary function of the division has been nuclear reactor research and development. Its early beginnings date back to 1943 when it was involved in development of the X-10 Graphite Reactor and the Chemical Pilot Plant 3019. In the 1950s it was involved in the Aircraft Nuclear Propulsion project. In 1951, the Reactor Technology Division was reorganized into two divisions, the Aircraft Nuclear Propulsion (ANP), and the Reactor Experimental Engineering (REE). Later that same year, the ANP division moved to Y-12. Building 9201-3 and REE moved to Building 9204-1. Discussions regarding other activities associated with this division were covered in detail in Section 2.1 of this report, "Summary of Historical Activities at the X-10 Site." There was little indication from the limited information reviewed that this Division used substantial amounts of material. While there were likely small releases of both chemicals and radionuclides, it doesn't appear that these were significant in terms of off-site releases.

#### **4.1.9 Y-12 Rover Project**

For approximately 13 years between 1960 and 1972 Y-12 was involved in a nuclear rocket program called the Rover Project. The project was a joint effort funded by AEC and the National Aeronautical and Space Agency (NASA) in which Y-12 made fuel elements that were designed to be use in a nuclear-powered space vehicle. It was believed that if this project had been completed, that a nuclear-powered spacecraft would have been able to reach sufficient speeds in outer space to have increased the feasibility of sending a vehicle and a small crew of humans to planets as far away as Mars (Napier, 1992). However, due to a lack of consistent

government support this project was halted sometime around 1970. Y-12's primary role in the Rover Project involved the following activities:

- Developing raw materials (i.e., enriched uranium alloys and compounds),
- Developing manufacturing methods for mass production of extruded fuel elements,
- Developing manufacturing methods for mass deposition of niobium carbide,
- Developing inspection procedures and computerized quality control systems to support mass production,
- Producing 10,000 to 20,000 fuel elements (rods) per year for about 13 years, and
- Extending design-basis reactor performance from several minutes to 1 hour by improvements in manufacturing processes.

#### 4.1.10 Y-12 Steam Plant

The Y-12 Steam Plant, designated as Building 9401-3, was constructed during 1954-1956. It has four boilers of the two-drum, bent tube design. Each boiler is designed for 250,000 lb/hr continuous steam generation at 250 psig and 500°F at the superheader outlet. The boilers were downrated to 200,000 lb/hr with the addition of baghouse dust collectors in 1985. Two boilers have the capability to fire with natural gas for emergency purposes only, but all four boilers are presently firing pulverized coal. Steam loads vary from 150,000 lb/hr in summer to 600,000 lb/hr peak in winter. Each boiler is equipped with the usual auxiliaries, including an air preheater, electrostatic precipitator, forced draft fan, and induced draft fan. The electrostatic precipitators were constructed in 1967 (MMES, 1983).

Steam is distributed at 235 psig and 500°F to the Y-12 buildings with pressure reduction at each building. Normal building usage is 15 to 30 psig. Heating, ventilation, and air conditioning condensate is returned from the major buildings to the steam plant for boiler feedwater. From 25 to 50% of the condensate is recovered. The remaining boiler feedwater is obtained by treatment of filtered water from the Oak Ridge Water Treatment Plant. Approximately 120,000 tons of mine-run coal are burned each year at the Y-12 Steam Plant. The coal is procured from local mines and is delivered by truck (MMES, 1983).

The Y-12 Plant coal pile is located just west of the Steam Plant. A minimum inventory of 50,000 tons of coal is maintained throughout the year, and at maximum capacity the pile contains 80,000 tons. The pile is compacted and sloped resulting in a top surface that slows the

penetration of rainwater into the body of the pile. The direct surface runoff constitutes most of the runoff, with some leachate percolating through the uncompacted sides of the pile. Historically, runoff was removed from the coal pile area through storm drains discharging directly into EFPC. The main impacts upon receiving waters resulting from the coal pile runoff are suspended solids (coal fines), low pH, and metallic and sulfate ions such as arsenic. The Y-12 Steam Plant ash slurry is currently pumped over the crest of Chestnut Ridge where the pipeline empties into a wet weather creek, McCoy Branch. From that point on, the stream flows by gravity through a filled ash impoundment, past a spillway in the ash impoundment, and through McCoy Branch that empties into the northwest corner of Rogers Quarry. The quarry is used as a sedimentation basin (MMES, 1983).

#### 4.2 SUMMARY OF THE INVESTIGATIVE PROCESS USED AT Y-12

The process used to investigate historical operations and releases from the Oak Ridge Y-12 site was based on extensive review of historical records and interviewing of active and retired plant workers. This section describes all the information resources that were examined and highlights those areas that were most useful in putting together a historical profile of Y-12. Also discussed are the approaches that were used to locate information relevant to the study.

Approximately 1,600 documents and photographs pertaining to Y-12 were pulled from various document repositories, roughly 420 of which were relevant to the study and summarized in the project repository database. Of the approximate 420 relevant documents and photographs, copies for 371 of these were requested by ChemRisk for the project repository, the remaining 49 documents were classified at the time of the request. Of the 371 reviewed and requested, ChemRisk received copies of 359 documents. The balance of the documents were made available for review by ChemRisk investigators, but copies could not be taken from the Y-12 site for several reasons. For example, in the case of some photographs, the negatives were not located or the negatives were located but determined to be classified. Also, some documents that were reviewed were not released because of their classification. Of the 359 documents sent to ChemRisk, 11 documents were marked as "Unclassified Controlled Nuclear Information."

Author and keyword searches proved to be the most valuable in searching for relevant documents that characterize the major Y-12 operations. Keyword searches were performed at some of the information resource repositories, at other locations it was not possible to do such searches. Keywords used in this approach included:

Monthly	Sediments	Beryllium
Inventory	Soil	Uranium
Discharge	Surface Water	Off-Site
Effluent	Solvents	New Hope Pond
Leaks	Chemicals	S-3 Pond
Spills	Poplar Creek	Pits
Waste	East Fork Poplar Creek	Burial Grounds



Airborne	Bear Creek	Stacks
Accident	Contamination	Vents
Incident	Health Physics	Calutron
Environmental	Industrial Hygiene	Alpha
Hazardous	History	Beta
Radioactive	Summary	Enrichment
Emissions	Progress	Rover Project
Release	Annual	Quarterly
Monitoring	Reports	Radiation Safety
Exposure	Mercury	

### Y-12 Central Files

The single largest document repository containing documents relevant to Y-12 is the Y-12 Central Files, in building 9711-5 below the main plant cafeteria. This document resource area contains roughly 100,000 active records, as estimated by Central File personnel. These active records go back to the Manhattan Project in 1943, and continue up to the present day. Active records can be identified by searching a database called the "Y-12 Information Management Database." Many of the useful active records are in the form of memorandums, reports, or other miscellaneous documents with report numbers that begin with the letter Y (e.g., Y/TS-001 or Y-10). Each document is called a "Record Copy" and is grouped by number sequence and stored in the Y-12 Central File vault.

With the majority of Y-12 activities focused towards production, diligent recording and archiving of historical information was not always a priority. This was especially true of active records. One of the determinants of whether Central Files received a copy of a report was whether the author placed it on the distribution list. Written plant policies and procedures that required submittal of reports to Central Files existed in one form or another throughout the years at Y-12, but they were not consistently adhered to; submittal of a document was left to the discretion of the author.

Central Files did prove to be a valuable resource for information during the investigation. Central Files contains a large number of classified and unclassified documents, some of which were reviewed and found relevant to the study. Numerous valuable historical records were identified such as progress reports, correspondence, monitoring data records, logbooks, and other technical reports. The following methods were used to locate relevant documents in Central Files:

- **Master Index Listing:** This is a computerized listing of all documents which were officially submitted to Y-12 Central Files. Most of these documents are Y reports, including a large number of classified documents. This listing is organized in semi-numerical order. This listing was randomly searched to

identify relevant titles for detailed review. Numerous relevant documents were reviewed, and copies of these documents were requested.

- **Author Index Listing:** This is a computerized alphabetical listing of Y reports by an author's last name. This listing contains the same documents listed in the Master Index List. Based on interviews and other documents, key names were searched in the listing and relevant documents were identified and reviewed by ChemRisk. Relevant documents previously obtained from other resources were sometimes not listed. This again is indicative of the lack of adherence to Y-12 policies of submitting reports or documents to Central Files. Nevertheless, several documents relevant to the study were identified from the listing that would be valuable resources in a follow-up study. This listing proved to be much more valuable than the Master Index Listing because the names were listed alphabetically, while the Master Index is not sorted by any parameter that facilitates effective searching.
- **Keyword Index Listing:** This is an alphabetical listing by keywords. It contains the same documents found in the Master Index Listing and the Author Index Listing.

#### Y-12 Records Center

The Y-12 Records Center contains retired or "inactive" records, generally stored in cardboard boxes placed in a vault in Building 9771-5 and in several rooms in Building 9987. These records are typically older documents that a division or department has decided to store for some defined period of time, but are too numerous to store in their areas. The boxes may contain both classified and unclassified records. Each box is assigned a retention period which defines how long the records are kept prior to destruction (e.g., 75 years for health physics records). Periodically, DOE institutes moratoriums on destruction of certain types of records, such as those potentially relevant to epidemiological studies. These records are tracked by records transmittal and receipt cards that briefly describe what is in each transmittal. A database does not exist that would allow someone to search for relevant documents. The cards are filed by division in a card file in the Records Center Vault.

These retired records are not on a database. Any review of these records would require a search of the numerous file cabinets which contain the Retired Records Index Cards. These cards provide department or division numbers assigned to each box and the box location, but lack information about the box contents. This approach was not attempted for the purpose of this study, due to limited time. Individual Departmental Retired Records Listings, which identify

box locations, dates of documents, subject matter of documents (e.g., stack release data), and retention schedules, could also be reviewed to identify relevant documents.

Departmental Retired Records Listings are bibliographies retained by individual departments or divisions at Y-12 that describe in detail the types of retired records that are stored in boxes at the Records Center vaults. The location of each box is identified by a 3-digit number which describes the box number, section, and tier within the vaults. These listings are typically compiled by a systems analyst, and are retained on miscellaneous databases. These listings provide important information necessary for identifying inactive records. As previously mentioned, department personnel assign the retention period for each box. Assistance from individual departments or divisions would be useful for searching these records. Some records of interest were identified, such as mercury, beryllium and uranium air sampling data but these are only a sample of what is likely to be available in this repository. Some of the old Health Physics records reviewed during the investigation appear to be quite valuable in providing analytical data that would help characterize stack releases from various Y-12 processes. Although not listed as a separate resource, we found in our investigation that some active employees have kept fairly extensive personal files, some of which were shared with ChemRisk and found to be useful for this study.

The Y-12 Records Center also contains a set of documents called the "Classified Mercury Files." These files contain numerous letters, progress reports, inventory data, production data, and monitoring data that describe the historical usages of mercury at Y-12 that were gathered during the 1983 Mercury Task Force investigation. As mentioned before, the task force focused on finding relevant information that would help quantify mercury losses to the environment from the Y-12 site. The Task Force has stored over 1000 documents in classified file cabinets, which are located at the Y-12 Records Center vault in Building 9711-5. There are 4 file cabinets, or approximately 19 file drawers, full of mercury-related documents. At one time, Y-12 had a database which contained most of these documents found in these file cabinets. This database is no longer available, but Y-12 retained a hard copy printout which lists available documents in the cabinets. ChemRisk performed a limited search of these cabinets and identified and reviewed some relevant documents. These files would be useful in supporting any follow-up study.

The Y-12 Records Center also contains Plant Shift Superintendent's (PSS) Logbooks. The Y-12 PSS Office is the central communications center for the entire plant. This office monitors the plant's activities and provides emergency coordination and response functions for all Y-12 operations. All accidents, fires, or unusual occurrences are reported to the PSS office. The PSS office has maintained detailed phone logs which include brief descriptions of the nature of the call that have been received by an operator at this office. These logbooks have been generated since the late 1940s. Most of the older logbooks are stored as inactive records at the Y-12 Records Center vault. A random search was performed on some of these logbooks during the

investigation. This review suggested that these documents could be helpful in identifying significant occurrences that have taken place in the Y-12 Plant. Each entry includes the name or initials of the person who took the call, date of incident or call, source of call such as a person's name or department name, and a brief description of the occurrence, including location and materials or equipment involved in the event.

#### Y-12 Document Response Centers

Individual departments or divisions are beginning to set up their own centers to hold and archive important documents in order to better support the various audits and appraisals that are routine at Y-12. Most of the documents retained in these centers are fairly recent, typically going back 10 to 15 years. An occasional old document may be found, but these are not typical of what is retained in these centers. The Health, Safety and Environment Division, the Environmental Restoration Division, and the Quality Division each have document centers that were reviewed during our study. Computerized database listings were reviewed for each center and a fair number of documents were found to be relevant to the study. Many of the relevant documents can also be found in the Y-12 Central Files vault. The majority of these documents are unclassified. All three centers are located at the Y-12 Plant Site.

#### The Tiger Team Records Center

The Y-12 Tiger Team Records Center is located in Building 9711-5. A collection of records that have been gathered to support DOE Tiger Team investigations are stored here and were reviewed for relevant documents. Typically, Tiger Team investigations are carried out by DOE contractors other than Martin Marietta and have involved, as their primary purpose, a technical appraisal of all areas of Y-12, including health and safety and hazardous material monitoring programs. A listing of the titles of unclassified and classified documents in the Tiger Team collection was made available. The listing was reviewed, and documents of potential relevance to the study were provided to us for review. Approximately 20 documents were evaluated, and copies of others were obtained by ChemRisk. Most of these records pertain to fairly recent Y-12 activities, however, some historical information was found in these documents.

#### Y-12 Technical and Engineering Library

Y-12 has two libraries which were found to be additional resources for relevant documents. The Y-12 Technical Library and the Y-12 Engineering Library are located at the Y-12 Plant site. The Y-12 Technical Library contained classified and unclassified documents. It is not known if this is true for the Engineering Library. Relevant documents from these libraries were identified in the following ways:

- Various searches were performed on the Library Information On-Line Network (LION), which is a computer system that is an on-line catalog of document holdings of the Martin Marietta Energy Systems libraries and can be accessed at each of the individual libraries throughout the Oak Ridge Reservation. The LION System will allow searches by authors, keywords, and subject areas of interest to identify reports or books. The LION system only contains documents that were entered into the database after 1981. Many of the documents identified had already been reviewed at the Y-12 Central Files area.
- Documents identified through book or report references were often located at one of these libraries. The ORNL Research Library was also used during the Y-12 Site investigation to locate relevant documents.
- A Library Card Catalog was searched at the Y-12 Technical Library. The associated file cabinets were searched by authors' last names, subject matter, or a document title if known.

#### DOE Information Resources Center

The Information Resource Center for the Environmental Restoration program is operated by a contractor for the U.S. Department of Energy. This center houses the Administrative Record for ORR environmental restoration activities and is located at Jackson Square in Oak Ridge. The Administrative Record is required by USEPA regulations, and is an ongoing document collection effort designated to contain all documents used in making decisions concerning cleanup of the Oak Ridge Reservation and certain off-site locations. A listing of the holdings of the Information Resource Center holdings was obtained and reviewed. The Center was a convenient place to obtain many environmental documents in the early phases of the study that were later located at the originating facilities' document centers.

#### The Office of Scientific and Technical Information (OSTI)

The DOE Office of Scientific and Technical Information (OSTI), formerly called the Technical Information Center (TIC), was contacted for documents that could not be located at Y-12. Reports found at OSTI include some DOE Oak Ridge Operations documents, some DOE environmental reports, and some Manhattan Engineering District and AEC documents.

#### Personnel Interviews

Interviews have been conducted with approximately 15 active employees with various levels of experience at the Y-12 Plant. The names of initial interviewees were provided by facility management for key functional areas identified by ChemRisk. Each interviewee identified

additional points of contact. Candidates for interviews were also identified from association with key historical documents. Some desired interviewees have not yet been located, and not all desired interviews were completed. No one declined an interview during the Y-12 portion of the investigation.

Notes taken in the course of the interviews have been reviewed by appropriate classification reviewers. The information obtained in the interviews is being summarized and entered into the project repository and the associated database. In the course of interviews, active and retired workers were also asked about the existence of historical records that would be relevant to the study. In some cases, records were identified that might not have been received or indefinitely retained by the identified document repositories.

#### K-25 Plant Classified Records Vault

Miscellaneous Y-12 inventory reports for enriched uranium, beryllium, thorium, and normal uranium are stored at the K-25 Plant Records Vault. Samples of this information included Y-12 material inventories and K-25 shipments of material received by Y-12. Most of this information covered the time period 1950 to 1964 and is most likely duplicated in Y-12's accountability records. This data may be of potential use in reconstructing material usage rates for important materials used at Y-12.

It is estimated that the ChemRisk project team searched approximately 30% of the Y-12 classified documents that had unique titles or Y-12 report numbers. Approximately 400 classified documents were reviewed, and about 100 were found to be relevant to the Health Studies. It is estimated that the ChemRisk project team searched about 2.5% of the available unclassified records at Y-12.

### **4.3 EVALUATION OF ACTIVITIES FOR OFF-SITE SIGNIFICANCE**

Based upon the information reviewed during this feasibility study portion of Phase I Health Studies, the following areas at the Y-12 Site appear to be those that warrant highest priority in any further investigations to quantify their historical off-site releases, exposures, and potential health risks to affected populations. Following brief discussion of the potential focus areas here, summaries of information that is available to support dose reconstruction is presented in Section 4.4.

- **Electromagnetic Separation and Enrichment Operations**

Y-12's first mission was the electromagnetic separation of uranium-235 to supply highly enriched uranium to Los Alamos for construction of the first atomic bombs. Uranium enrichment operations involved large quantities of natural uranium to enrich sufficient quantities of uranium-235 for weapons development. Some of the largest releases of uranium from Y-12 to the off-site environment may have been from these processes. During the first year, 1944, potentially significant liquid releases of low to moderate enriched uranium occurred through drains and runoff into East Fork Poplar Creek. From 1945 to 1947, airborne and waterborne releases occurred which involved material that had a wide range of uranium enrichments.

- **Lithium Separation and Enrichment Operations**

Pilot and production-scale operations for the separation and enrichment of lithium-6 for use in thermonuclear weapons was a major effort at Y-12 from roughly 1950 to 1963. Millions of pounds of mercury were required in the Colex process to separate the naturally-occurring lithium-6 isotope from lithium-7. It has been reported in several documents that large quantities of mercury were released to surface waters and into the air from the Colex operations. Some additional releases did occur from the Elex pilot plant that operated from 1953 to 1956.

- **Uranium Weapon Component Manufacturing Operations**

Starting in the late 1940s, Y-12 began a gradual development into the largest enriched uranium weapon-components manufacturer's plant in the United States. Industrial-scale processing of enriched and depleted uranium and, to a lesser extent, thorium, in various compounds have made up Y-12's primary production streams. Process controls were used to minimize releases of highly enriched uranium. Process controls for other materials were much less effective, and processing of these materials led to routine and accidental releases from process exhaust stacks and storm sewers dating back to the late 1940s and the early 1950s. The magnitude of these operations and information gathered during our study indicate that there was a reasonably high potential for off-site release.

- **Beryllium Operations**

Starting in the early 1950s, Y-12 constructed large production facilities to handle beryllium compounds for use in nuclear weapons. The control of airborne beryllium in the work place has been a real challenge for process control engineers and industrial hygiene personnel at Y-12. While routine stack releases have been filtered to minimize releases to the off-site environment, some releases have occurred during Y-12's extensive

history of processing beryllium. Most of the processes and work areas have been routinely monitored to quantify the effectiveness of their material control measures, e.g. filtration of exhaust stacks. It is believed that unknown quantities may have been transported off-site on workers' clothes and shoes and may warrant further investigation (Googin, 1993). Limited unclassified data was found that described beryllium operations releases. A large amount of workplace and exhaust stack air sampling data located in Health Physics retired records stored in the Y-12 Records Center vault suggests that beryllium operations may have been a significant contributor to off-site releases.

- **Waste Disposal Operations**

Y-12's primary mission of large-scale production of weapons has resulted in the generation of large quantities of chemical and radioactive waste. These wastes have been in liquid, solid, and gas forms and have been disposed of in several areas in and around the Y-12 Plant site and to the air. Buildup of liquid waste in the uranium production areas forced Y-12 to construct large seepage ponds known as the S-3 ponds which are believed to have been a source of off-site releases of organic solvents and radionuclides. These ponds, constructed in 1951, were designed to collect waste-bearing nitric acid and other nitrate waste, and covered a three-acre area at the west end of the Y-12 plant.

New Hope Pond was added in 1963 in an attempt to reduce off-site releases from steam plant operations, mercury processes, and allow for pH adjustment to the East Fork Poplar Creek. New Hope Pond is known to have been a significant pathway for off-site migration of hazardous materials from the Y-12 site.

- **Classified Materials**

The presence of some potentially important materials at Y-12 is classified. Although review of their uses at Y-12 was limited during this investigation, it appears that some may have been used in quantities significant enough to warrant further evaluation.

#### **4.4 INFORMATION AVAILABILITY FOR FURTHER QUANTITATIVE EVALUATION**

In order to support the direction of any future efforts to quantify doses and health risks associated with contaminants released from the Y-12 site, information that is available concerning the identified areas for likely off-site significance has been reviewed. This section describes the information that has been identified during the Phase I Feasibility Study historical investigation.



Production summary reports and log books, health physics reports, and analytical data for effluent measurements are the types of information that would be useful in a further evaluation of off-site releases of uranium and other materials such as mercury and beryllium. Much of this data is classified but could be reported in an unclassified format. Monthly and quarterly health physics and industrial hygiene reports document the release quantities for several production related exhaust stacks. Beginning around 1950, the plant's Analytical Laboratory located in Building 9995 counted workplace and exhaust stack air samples for various materials and reported the results to the Health Physics Department. These results are summarized in health physics reports and raw data are reported on data sheets generated by the analytical laboratory, and are located in the Y-12 Central Files and the Y-12 Records Center vaults.

Enriched uranium, and to a lesser extent depleted uranium, have been closely controlled and monitored at Y-12 since the beginning of plant operations. Classified release and discard reports were identified during the investigation which describe the quantities of uranium released from chemical, metallurgical, and salvage operations. Much of the information is categorized into airborne and waterborne releases, solid waste and liquid storage or disposal. Most of the historical discard records for enriched uranium can be obtained within Y-12's Nuclear Material Control and Accountability Department (Mabe, 1993). Historical unclassified summary reports have been published which list the quantities and activity levels for radioactive releases for these operations.

In summary, the following records could be useful for further quantitative evaluation:

- Production data for material usages (classified and unclassified),
- Health Physics, Industrial Hygiene, and Safety reports (1947-1992),
- Y-12 Nuclear Control Accountability Program records (1943 to 1992),
- Y-12 Plant Quarterly and Annual Reports (1950s to 1992),
- Y-12 Plant Analytical Laboratory data records (Building 9995),
- Purchasing and storage records, and
- Y-12 Safety Analysis Reports and Operational Safety Reports.

The following sections briefly discuss available information for further quantitative evaluations of each of the focus areas described in Section 4.3 of this report.

## Electromagnetic Separation and Enrichment Operations

The Y-12 Plant's original mission to electromagnetically separate and enrich uranium-235 involved two main functional operations which processed large quantities of uranium compounds and other materials such as carbon tetrachloride and nitric acid. The first operation was the chemical operations which included the preparation of uranium tetrachloride feed material for the electromagnetic separation units (calutrons) and the chemical recovery of various uranium compounds from the calutrons. Uranium compounds recovered from the calutrons included tetrachloride, carbides, oxides, and metal. Most of the early releases of uranium and carbon tetrachloride are believed to have been from these operations. The second operation was electromagnetic separation of uranium in the alpha or beta calutrons which were normally operated under vacuum conditions. Releases from these operations are believed to be small in comparison with the uranium chemical operations (Griffith, 1957; Googin, 1993).

During the first year of uranium enrichment operations, liquid samples were collected for determining concentration and enrichment levels of uranium from the uranium preparation and recovery operations. Most of the liquid samples taken of the uranium product and waste streams were used for evaluating the efficiencies of uranium recovery processes and for maintaining a uranium accountability system. Monitoring liquid effluents from the alpha chemical processes (normal or depleted uranium) was not performed as often or was as quantitative as for the effluents from the beta chemical processes (enriched uranium-235) due to the fact that the beta material was much more valuable to Y-12's mission. Specific liquid or waterborne monitoring data compiled during these first few years of Y-12 operations were not located during this investigation. It is believed that most of this monitoring data for 1943 to 1947 was kept in material accountability records (Googin, 1993). Monthly Health Physics Progress reports starting in early 1948 began to include uranium waterborne monitoring results for potable water, sanitary sewers, and storm sewers (East Fork Poplar Creek).

During these early process operations, most of the airborne effluents were not routinely monitored for determining releases, although periodic air samples (grab samples) were collected in the workplace and analyzed for uranium. Measurement techniques used in obtaining grab samples were crude by today's standards, however, these did provide some quantitative information about the relative amounts of uranium (particularly enriched uranium) that were present in production areas that may have been lost through general building ventilation. As early as 1945, air sampling equipment was used at Y-12 to monitor airborne radioactive contaminants in the work area or exhaust points on the process equipment. One type of sampling equipment that was developed at the University of Chicago and used at Y-12 came with a high-efficiency asbestos-based filter paper through which the air was drawn and then counted for gross alpha activity. These were portable air samplers set up to monitor various processes in the alpha, beta, and recovery buildings. The filter paper was manufactured by Hollingsworth

and Vose Co., and was known as H.V. No. 9081. The H.V. No. 8912 was considered a substitute for the 9081 variety (Berggren, 1947). The paper was formed into a cylinder, supported by a special "Bird Cage," and placed in the sample apparatus, commonly referred to as filter tubes. Flowmeters were used to set the proper sampling flowrate for the filter collection tube. The rest of the sampling equipment consisted of a holder and plugs for the filter tube, a vapor removing canister, a source of suction (Filter Queen Vacuum Cleaner), and a bleed valve for controlling air flow. Drawings of this old equipment were located in the documents reviewed during our investigation (Berggren, 1947).

The sample tubes were generally analyzed by counting with an alpha counter located in Room 8 of Building 9203. This alpha counter was specially built to receive and count the filter tubes without further preparation. It was reported that accurate measurements could be obtained for samples as high as 100,000 counts per minute. A background (blank) filter tube was also counted and subtracted from the sample count (Berggren, 1947). Uranium releases were reported by converting the counts to a mass (kilograms) by applying the specific alpha activity of the material in a sample. Most samples had the same specific alpha activity as the uranium that was being processed at Y-12. When the isotopic concentration was different, Y-12 then collected a sample of the material and performed a specific alpha activity analysis. This was obtained by performing a fluorometric analysis and activity measurement of the specific production material (Berggren, 1947). Monitoring data collected from these samplers were found in some of the early Health Physics Progress reports although these only date back to 1947. Earlier air monitoring data collected in the alpha and beta buildings and in the chemical process areas such as 9202 and 9203 were not identified during this investigation. Some of this earlier data may have been compiled in old accountability records but was not identified during this investigation.

Estimates of early uranium releases from Y-12's uranium enrichment operations have been based upon available inventory and production records, known releases, and limited effluent monitoring. Table 4-8 summarizes the total production output from the Y-12 uranium enrichment program starting with the alpha feed material, between 1944 and 1947. However, information that describes the total amount of uranium received from outside suppliers during the Manhattan Project was not identified during this investigation. Production and accountability data dating back to 1943 for the early alpha and beta buildings were not identified during this investigation, but if located, could be potentially useful in developing material balances and reconstructing historical releases for uranium and, to a lesser extent, chemical releases. One interviewee mentioned that this old accountability information may be located at the Y-12 Records Center (Griffith, 1993). Earlier production and accountability records have been "retired" and are located in the Y-12 Records Center. Specific locations of relevant inventory data can be provided either by the Nuclear Material Control and Accountability (NMC&A) Department at Y-12 or through personal interviews with active and retired employees.

TABLE 4-8

**CALUTRON OPERATIONS**  
**JUNE 24, 1944 - MAY 4, 1947**

Beta Cycle Production Period	Alpha Cycle Product		UF <sub>6</sub> from K-25		UCL <sub>4</sub> Vaporized	Salvage to K-25		R Pocket Enriched Product		Q Pocket Depleted Tails	
	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U
06/24/44 - 08/04/44	27.69				75.08			1.12		4.96	
08/04/44 - 08/14/44	4.33				26.55			0.34		1.52	
08/14/44 - 08/28/44	7.70				24.24			0.28		1.26	
08/28/44 - 09/11/44	10.32				41.87			0.14		2.59	
09/11/44 - 09/25/44	8.64				57.85			0.46		3.26	
09/25/44 - 10/23/44	21.35				144.09			1.19		8.33	
10/23/44 - 11/06/44	14.94				104.12			1.00		7.39	
11/06/44 - 11/20/44	17.38				135.72			1.27		10.00	
11/20/44 - 12/04/44	30.18				159.38			1.60		12.36	
12/04/44 - 12/16/44	23.70				164.66			1.75		13.76	
12/16/44 - 12/31/44	34.11	10.98			220.81			3.26	73.04	16.16	0.58
12/31/44 - 01/14/45	32.70	10.75			212.30			3.26	79.37	22.84	0.37
01/14/45 - 01/27/45	35.31	10.17			229.44			3.29	80.63	23.53	0.30
01/27/45 - 02/11/45	34.86	10.30			217.82			3.41	81.14	24.67	0.30
02/11/45 - 02/25/45	34.69	9.94			210.30			3.22	82.59	24.61	0.30

TABLE 4-8  
(Continued)

CALUTRON OPERATIONS  
JUNE 24, 1944 - MAY 4, 1947

Beta Cycle Production Period	Alpha Cycle Product		UF <sub>6</sub> from K-25		UCL <sub>4</sub> Vaporized	Salvage to K-25		R Pocket Enriched Product		Q Pocket Depleted Tails	
	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U
02/25/45 - 03/11/45	33.99	10.02			223.51			3.67	82.10	28.19	0.30
03/11/45 - 03/25/45	33.38	9.82			225.32			3.58	82.75	26.89	0.30
03/25/45 - 04/07/45	33.22	9.72			219.55			3.26	82.51	24.41	0.30
04/07/45 - 04/21/45	33.35	10.06			208.46			3.12	83.53	23.96	0.29
04/21/45 - 05/19/45	70.80	10.47			410.32			6.02	86.53	47.73	0.22
05/19/45 - 06/02/45	39.17	11.78			202.10			3.11	88.38	24.07	0.17
06/02/45 - 06/16/45	32.60	12.09			265.23			4.05	87.32	31.72	0.16
06/16/45 - 07/28/45	114.41	11.92	259.95	18.05	1727.65	117.39	7.75	22.28	85.01	183.63	0.17
07/28/45 - 08/11/45	32.76	13.04	66.80	22.73	506.52	17.69	9.11	7.74	85.69	56.40	0.19
08/11/45 - 09/08/45	62.89	13.62	304.31	23.57	583.14	138.70	12.02	22.11	91.56	92.74	0.32
09/08/45 - 09/22/45	24.14	12.19	180.46	23.90	523.78	71.45	13.46	14.81	93.00	50.28	0.37
09/22/45 - 10/06/45	6.81	16.15	189.71	23.76	514.92	21.97	17.64	16.07	92.39	55.33	0.37
10/06/45 - 10/21/45			144.47	23.69	529.45	38.66	22.69	14.49	93.55	48.07	0.38
10/21/45 - 11/04/45			98.28	25.85	584.32	2.68	15.51	16.52	93.26	54.35	0.38

TABLE 4-8  
(Continued)

CALUTRON OPERATIONS  
JUNE 24, 1944 - MAY 4, 1947

Beta Cycle Production Period	Alpha Cycle Product		UF <sub>6</sub> from K-25		UCL <sub>4</sub> Vaporized	Salvage to K-25		R Pocket Enriched Product		Q Pocket Depleted Tails	
	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U
11/04/45 - 11/18/45	0.11		98.99	29.34	556.80	0.89	13.71	18.26	94.05	57.47	0.38
11/18/45 - 12/02/45			103.44	29.81	644.40			21.23	94.42	63.05	0.38
12/02/45 - 12/16/45			114.40	28.15	699.43			24.20	94.44	71.34	0.38
12/16/45 - 12/30/45			115.89	27.59	754.08			28.25	94.67	70.54	0.45
12/30/45 - 01/13/46			111.92	28.65	753.92			29.52	95.03	70.50	0.45
01/13/46 - 01/27/46			110.57	27.95	743.63			31.00	95.20	78.58	0.45
01/27/46 - 02/10/46			78.84	28.17	751.23	0.04	35.71	32.08	95.13	80.56	0.45
02/10/46 - 02/24/46			128.81	27.25	790.61			32.29	95.19	84.03	0.55
02/24/46 - 03/10/46			173.63	27.58	783.04			32.71	95.21	84.63	0.55
03/10/46 - 03/24/46			162.73	27.31	774.90	61.09	29.84	32.98	95.19	86.00	0.55
03/24/46 - 04/07/46			196.21	26.97	771.84	41.59	22.49	32.92	95.21	86.12	0.55
04/07/46 - 04/21/46			148.43	29.44	777.14	13.00	13.16	33.33	95.41	87.97	0.54
04/21/46 - 05/05/46			164.57	29.77	767.83	29.01	27.34	33.01	95.00	87.44	0.54
05/05/46 - 05/19/46			167.84	29.87	747.35	58.36	28.66	33.49	95.20	85.15	0.50

TABLE 4-8  
(Continued)

CALUTRON OPERATIONS  
JUNE 24, 1944 - MAY 4, 1947

Beta Cycle Production Period	Alpha Cycle Product		UF <sub>6</sub> from K-25		UCL <sub>4</sub> Vaporized	Salvage to K-25		R Pocket Enriched Product		Q Pocket Depleted Tails	
	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U
05/19/46 - 06/02/46			147.78	30.07	760.65	9.57	22.66	34.40	95.40	84.76	0.50
06/02/46 - 06/16/46			122.62	29.93	758.59	3.85	12.69	34.67	95.21	82.90	0.50
06/16/46 - 06/30/46			105.43	30.02	765.30			34.53	94.97	79.59	0.60
06/30/46 - 07/14/46			62.65	29.88	750.79			34.27	94.91	77.92	0.60
07/14/46 - 07/28/46			76.12	29.81	753.50			34.59	94.90	78.49	0.65
07/28/46 - 08/11/46			128.76	29.82	758.01			34.75	94.74	78.64	0.69
08/11/46 - 08/25/46			129.78	29.71	773.58			36.63	94.71	82.97	0.70
08/25/46 - 09/08/46			131.39	29.89	768.33			37.46	95.03	82.87	0.70
09/08/46 - 09/22/46			130.73	29.90	760.32			37.34	94.82	82.75	0.64
09/22/46 - 10/06/46			128.66	29.76	761.04			37.54	94.72	83.03	0.69
10/06/46 - 10/20/46			120.01	29.64	764.23			38.29	94.88	84.81	0.67
10/20/46 - 11/03/46			64.19	29.97	601.79			30.30	94.87	67.39	0.67
11/03/46 - 11/17/46			0.15	29.80	459.44			22.98	94.58	51.06	0.59
11/17/46 - 12/01/46					511.53			24.91	95.13	54.41	0.62

TABLE 4-8  
(Continued)

CALUTRON OPERATIONS  
JUNE 24, 1944 - MAY 4, 1947

Beta Cycle Production Period	Alpha Cycle Product		UF <sub>6</sub> from K-25		UCL <sub>4</sub> Vaporized	Salvage to K-25		R Pocket Enriched Product		Q Pocket Depleted Tails	
	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U	kg U	% <sup>235</sup> U
12/01/46 - 12/15/46			89.73	29.97	519.00			26.19	94.82	57.31	0.61
12/15/46 - 12/31/46			72.28	29.89	543.60			28.60	94.61	72.38	0.56
12/31/46 - 01/12/47					294.43			15.94	94.05	35.77	0.61
01/12/47 - 01/26/47					94.96			5.15	93.55	11.51	0.61
01/26/47 - 02/09/47					92.17			4.94	93.40	11.47	0.61
02/09/47 - 02/23/47					94.62			5.21	93.92	12.04	0.67
02/23/47 - 03/09/47					95.62			5.45	93.03	12.54	0.69
03/09/47 - 03/23/47					92.49			5.33	93.32	12.42	0.69
03/23/47 - 04/06/47			9.66	30.24	92.41			5.48	92.67	12.70	0.69
04/06/47 - 04/20/47			22.09	30.54	88.40			5.16	93.04	11.75	0.69
04/20/47 - 05/04/47			15.42	30.32	91.02			5.27	93.13	11.74	0.80

Source: Compere, 1991.



Estimates of early uranium releases were shown in Table 4-3 and again in Tables 4-8 and 4-9. These tables were prepared based on various references. Some of the data in these tables do not agree with one another and the actual reasons for these differences were not determined during this investigation. Table 4-3 contains estimates that were reconstructed around 1957 by Y-12 personnel and Tables 4-9 and 4-10 contain estimates that were reconstructed by Y-12 accounting and environmental personnel in the mid 1980's. These tables depict the types of available summary information that could be potentially useful in providing guidance for further investigations and for the development of reasonable uranium and other radionuclide release estimates. Attempts to resolve some of these differences and develop the best possible source term would likely be part of further investigations.

Carbon tetrachloride was used in large quantities to produce uranium tetrachloride during the Manhattan Project. Information for reconstructing usages and off-site releases of this material is available through early purchasing records, production and inventory reports, releases reports, and environmental data (Butterini, 1992). No evidence was identified during this investigation suggesting that Y-12 monitored early chemical releases. Also, a partial historic chemical release report was written in 1986 for important chemicals used at Y-12. Carbon tetrachloride usage and release estimates from this report were the following:

Year	Quantity Used (kg)	End Product (kg)	Airborne Release (kg)	Liquid Effluent (kg)
1943	261,800	75,394	2,618	172,788
1944	717,500	236,775	7,175	473,550
1945	668,500	220,275	6,685	441,540
1946	57,300	18,909	573	37,456

(Fee, 1986)

Other chemicals reportedly used and released to the off-site environment during the first few years of Y-12 operations included nitric acid, aluminum nitrate, cupric nitrate, hydrofluoric acid, and other waste or raffinate compounds. Many of these materials reportedly were released as liquid waste products to the process drains and storm sewers and ultimately drained into East Fork Poplar Creek (Googin, 1993). Monitoring data of these effluents were not identified during this investigation. Further evaluations in any future studies would need to determine the potential significance of these material releases.

#### **Weapons Component Manufacturing Operations (1947 - 1992)**

Due to the lack of monitoring, many of the estimates of early uranium losses were based upon material accountability information from production operations. Processes which handled normal or depleted uranium had much looser accountability requirements, and greater uncertainty is likely

TABLE 4-9

## SUMMARY OF ESTIMATED Y-12 PLANT RELEASES OF RADIOACTIVITY

Year	AIRBORNE		WATERBORNE			
	Uranium (Ci)	Uranium* (kg)	Uranium (Ci)	Uranium* (kg)	Thorium (Ci)	Thorium (kg)
1944	0.04	55	22.30	33,000	ND	ND
1945	0.07	102	4.70	7,000	ND	ND
1946	0.07	102	ND	ND	ND	ND
1947	0.04	55	0	0	ND	ND
1948	ND	ND	0.10	155	ND	ND
1949	ND	ND	0.30	454	ND	ND
1950	ND	ND	0.10	144	ND	ND
1951	ND	ND	0.06	98	ND	ND
1952	ND	ND	0.002	3	ND	ND
1953	0.01	30	0.651	953	ND	ND
1954	0.14	32	0.71	1,118	0.001	11
1955	0.14	32	0.62	1,058	0.003	26
1956	0.83	43	2.26	4,987	0.005	44
1957	0.71	41	5.65	8,448	0.005	49
1958	0.71	41	5.85	10,019	0.008	70
1959	1.93	120	5.15	10,410	0.367	3,363
1960	0.60	99	4.55	10,067	0.031	283
1961	0.61	109	2.00	3,064	0.101	927
1962	0.66	100	0.86	1,333	0	0
1963	0.85	103	0.82	1,248	0.002	20
1964	0.76	170	4.42	6,605	0.001	7
1965	0.48	281	5.91	8,852	ND	ND
1966	0.51	212	5.34	7,985	ND	ND
1967	0.51	212	10.20	15,217	ND	ND
1968	0.45	211	11.75	17,525	ND	ND
1969	0.46	223	2.80	4,189	ND	ND
1970	0.47	259	5.88	8,775	ND	ND

TABLE 4-9  
(Continued)

SUMMARY OF ESTIMATED Y-12 PLANT RELEASES OF RADIOACTIVITY

Year	AIRBORNE		WATERBORNE			
	Uranium (Ci)	Uranium* (kg)	Uranium (Ci)	Uranium* (kg)	Thorium (Ci)	Thorium (kg)
1971	0.16	290	2.37	3,546	ND	ND
1972	0.08	222	2.03	3,042	ND	ND
1973	0.07	206	0.74	1,119	ND	ND
1974	0.13	207	1.04	1,561	0.007	65
1975	0.21	209	1.09	1,638	0.021	195
1976	0.20	207	0.91	1,368	0.020	203
1977	0.13	206	0.50	755	0.019	176
1978	0.07	205	0.27	410	0.013	120
1979	0.13	206	0.24	366	0.010	93
1980	0.28	218	0.10	158	0.009	80
1981	0.20	207	0.45	687	0.009	85
1982	0.20	207	0.56	846	0.006	52
1983	0.20	208	0.14	222	0.005	49
1984	0.25	329	1.20	1,799	0.010	90
1985	0.18	210	0.72	783	0.017	153
1986	0.19	211	0.67	652	0.007	64
1987	0.14	116	0.57	715	0.003	27
TOTAL	13.87	6,296	116.58	182,374	0.680	6,253

\* Ratio of Ci/kg varies due to different isotopic enrichment

ND Data for these periods were not located in the Phase I investigation.

Note: These numbers are estimates, and do in some cases disagree with estimate reported in other reports.

Reference: USDOE, 1988.

TABLE 4-10

**ESTIMATED QUANTITIES OF RADIONUCLIDES  
CONTAINED IN Y-12 SOLID WASTE BURIED ONSITE\***

Year	Uranium (Ci)	Uranium (kg)**	Thorium (Ci)	Neptunium (Ci)***	Technetium (Ci)***
1944	2.09	33			
1945	16.14	255			
1946	13.23	209			
1947	0.93	371	0.0001		
1948	4.46	203	0		
1949	1.22	156	0		
1950	0.74	256	0.0001		
1951	0.76	662	0		
1952	3.05	1,466	0.0002		
1953	1.30	624	0	0.05	0.07
1954	1.53	2,293	0.0005	0.05	0.21
1955	9.04	21,806	0.0004	0.05	0.29
1956	9.92	22,957	0.001	0.05	0.29
1957	420.78	38,253	0.0007	0.05	1.50
1958	42.32	3,763	0.001	0.05	1.50
1959	116.63	21,931	0.062	0.05	1.50
1960	213.36	206,768	0.017	0.05	1.50
1961	558.89	1,491,895	0.103	0.05	1.50
1962	85.71	199,744	0.342	0.05	1.50
1963	111.81	325,843	0.560	0.05	1.50
1964	243.43	676,988	1.562	0.05	1.50
1965	135.73	375,841	2.076	0.05	1.50
1966	481.43	1,297,260	0.607	0.05	1.50
1967	358.80 #	979,909	0.645	0.05	1.50
1968	99.90	237,837	0.152	0.05	1.50
1969	141.31	390,073	0.173	0.05	1.50
1970	237.19	645,940	1.050	0.05	1.50
1971	199.87	556,242	0.953	0.05	1.50
1972	370.75	988,349	1.052	0.05	1.50
1973	276.65	761,729	0.822	0.05	1.50

TABLE 4-10  
(Continued)

ESTIMATED QUANTITIES OF RADIONUCLIDES  
CONTAINED IN Y-12 SOLID WASTE BURIED ONSITE\*

Year	Uranium (Ci)	Uranium (kg)**	Thorium (Ci)	Neptunium (Ci)***	Technetium (Ci)***
1974	221.87	614,406	0.012	0.05	1.50
1975	196.74	540,689	0.434	0.05	1.50
1976	168.27	457,290	0.388 ##	0.05	1.50
1977	15.10	34,562	0.194	0.05	3.29
1978	368.65	843,276	0.014	0.05	3.29
1979	51.04	12,324	0.056	0.05	3.29
1980	198.94	529,517	0.056	0.05	3.29
1981	267.33	703,601	0.023	0.05	3.29
1982	439.44	1,169,765	0.023	0.05	3.29
1983	295.11	809,790	7.001	0.05	1.50
1984	342.51	943,387	0.011	0.05	1.50
1985	266.29	730,298	0	0.05	1.50
1986	214.25	458,840	0	0.05	1.50
1987	92.20	263,070	0.196	0.05	1.50
TOTAL	7,097	17,290,523	18,588	1.75	58.10

\* All digits carried through to avoid rounding errors. Only first two digits are significant.

\*\* Ratio of Ci/kg varies due to different isotopic enrichment

\*\*\* Discharges of neptunium and technetium were discarded to the S-3 Ponds through 1983 as solution, but were recorded as burial.

# Values for 1967 and 1968 include uranium-233 in salvage material resulting from research and development work in fabrication of U-233 parts.

## The quantity shown for 1976 does not include 276 kg thorium placed in the Y-12 burial ground at the request of the State of Tennessee as a result of cleanup of Nuclear Chemicals and Metals Corporation at Huntsville, Tennessee.

Source: USDOE, 1988.

to be associated with those numbers. In the early days, effluent monitoring was rarely done for depleted uranium operations, although there was material accountability information maintained by production personnel (Griffith, 1957). Some of the early Monthly Health Physics Progress Reports (1947 to 1950) contain airborne uranium concentrations for buildings 9206 and 9212 which were determined from air samples taken in the general indoor air or worker breathing zones with portable air sampling equipment (Struxness, 1949). Y-12 began routine stack monitoring for enriched uranium in the early 1950s. Stack monitoring data and release quantities were published in monthly Health Physics Progress Reports.

In the 1950s, both permanently installed sampler heads and portable equipment were used in sampling exhaust stack gases. Four permanent samplers were installed in the main filter house on the exhaust or stack side of the filters. This filter house has serviced several enriched and depleted uranium process streams and is located in Building 9828. In addition to this, there were three other permanent samplers installed in other stacks or tributary ducts. Most of these samplers were operated continuously and air samples were counted for gross alpha activity. Once each month, portable high volume air samplers were used to sample exhaust systems in which the presence of acid fumes had prevented the use of permanent samplers. This was the case for many of the uranium chemistry areas, where large quantities of nitric acid and uranyl nitrate were processed. Again, the results of the stack sampling programs were reported to plant management at least once a month in Health Physics reports (Patterson et al., 1957; Sanders, 1992).

By 1969, Y-12 had nine airborne release points that were monitored for enriched and depleted uranium. Thorium airborne effluents were also monitored in Buildings 9201-5 and 9204-4. There were roughly 54 different operational exhaust systems that released process effluents through these nine stacks. Most of the stacks were monitored with permanently-installed isokinetic sampling probes. The various process airborne effluents were filtered upstream from the sampling probes with one or more filtration systems such as scrubbers, roughing filters, chemical traps, and HEPA filters. Detailed annual radioactive airborne effluent reports for these monitored stacks were compiled first in 1969 and 1970 and are shown Tables 4-11 and 4-12. These two reports listed the building, the exact release point for the airborne contaminant, sampling technique, release quantities, and other measurement parameters such as total volume of exhaust air (UCC-ND, 1971a; UCC-ND, 1971b). Some information has been eliminated from these tables because of security and classification issues. Table 4-11 often lists "0.00" as an airborne release quantity although it is not known if this is an absolute value or some approximation of the detection limit. Further investigation is warranted during future studies to better define what "0.00" means in the context of these earlier airborne releases of uranium and thorium. Additional monitoring results of this type were not identified during this investigation and suggests that these reports were discontinued after 1970. However, the data

TABLE 4-11

## 1969 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Volume (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	E-Wing Dust Collector	Casting	5	3.68 x 10 <sup>10</sup>	1.89
Uranium	Highly	9212	C-Wing Stack Exhaust	Fuel Element Fab.	4	6.83 x 10 <sup>9</sup>	0.19
Uranium	Highly	9212	D-Wing Stack	Chemical Conversion	6 or 1,4	2.15 x 10 <sup>10</sup>	0.32
Uranium	Highly	9212	West Head House Stack	Product Recovery	1,4	1.47 x 10 <sup>10</sup>	0.37
Uranium	Highly	9212	Reduction Area Exhaust	Metal Reduction	1,4	5.26 x 10 <sup>8</sup>	0.00
Uranium	Highly	9212	Room 1010 Sintering Furnace Exhaust	UO <sub>2</sub> Production	4	5.26 x 10 <sup>8</sup>	0.00
Uranium	Highly	9212	Room 1010 Exhaust	UO <sub>2</sub> Production	7	5.26 x 10 <sup>8</sup>	0.00
Uranium	Highly	9212	Dry Chemistry Reactor Hood Exhaust	Chemical Conversion	1,4	5.26 x 10 <sup>8</sup>	0.00
Uranium	Highly	9212	C-1 Wing PVC Stack	Product Preparation	6 or 1,4	3.68 x 10 <sup>9</sup>	0.31
Uranium	Highly	9212	C-1 Wing Room Exhaust	Product Preparation	7	7.88 x 10 <sup>9</sup>	0.03
Uranium	Highly	9212	Process Exhaust (Dissolver Calciner)	Product Preparation	6 or 1,5	5.26 x 10 <sup>8</sup>	0.01
Uranium	Highly	9212	B-1 Wing 3rd and 4th Floors Room Exhaust	Chemical Salvage	7	7.88 x 10 <sup>9</sup>	0.10
Uranium	Highly	9212	B-1 Wing Denitrator Room and Hood Exhaust	Chemical Conversion	1,5	2.10 x 10 <sup>9</sup>	0.03
Uranium	Highly	9212	B-1 Wing 2nd Floor Calciner and Dissolver	Product Preparation	1,5	2.10 x 10 <sup>9</sup>	0.00
Uranium	Highly	9212	B-1 Wing 2nd Floor Exhaust	Chemical Salvage	7	5.26 x 10 <sup>8</sup>	0.03
Uranium	Highly	9212	B-1 Wing 2nd Floor Exhaust	Chemical Salvage	7	5.26 x 10 <sup>8</sup>	0.02
Uranium	Highly	9212	B-1 Wing Conversion Area Exhaust	Chemical Salvage	7	5.26 x 10 <sup>8</sup>	0.02

TABLE 4-11  
(Continued)

1969 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Volume (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	B-1 Wing 1st Floor Dissolver Area Exhaust	Chemical Salvage	7	5.26 x 10 <sup>9</sup>	0.00
Uranium	Highly	9212	B-1 Wing 1st Floor Conversion Area Exhaust	Chemical Salvage	1,4,5	5.26 x 10 <sup>9</sup>	0.01
Uranium	Highly	9212	B-1 Wing Feed Preparation Dry Filter Exhaust	Chemical Salvage	1,5	2.10 x 10 <sup>9</sup>	0.04
Uranium	Highly	9212	C-Wing Rover Exhaust	Fuel Element Fab.	4	2.10 x 10 <sup>10</sup>	0.01
Uranium	Highly	9212	E-Wing Machine Shop	Machining	5	2.10 x 10 <sup>10</sup>	0.03
Uranium	Highly	9212	E-Wing Reduction Exhaust Plenum Chamber	Metal Conversion	1,4,5	7.35 x 10 <sup>9</sup>	0.01
Uranium	Highly	9212	Room 1022 E-Wing Lab Stack Exhaust	Chemical Lab	7	7.36 x 10 <sup>9</sup>	0.05
Uranium	Highly	9212	Room 1021 (South) E-Wing Lab Stack Exhaust	Chemical Batch	4	7.88 x 10 <sup>9</sup>	0.00
Uranium	Highly	9212	Room 1021 (West) E-Wing Lab Stack Exhaust	Chemical Lab	7	2.63 x 10 <sup>7</sup>	0.04
Uranium	Highly	9212	Room 1021 (East) E-Wing Lab Stack Exhaust	Chemical Lab	7	2.63 x 10 <sup>9</sup>	0.12
Uranium	Highly	9215	O-Wing Mill & Room Exhaust	Metal Rolling	1,5	2.94 x 10 <sup>10</sup>	0.01
Uranium	Highly	9215	M-Wing Exhaust	Machining	1,5	2.31 x 10 <sup>10</sup>	0.03
Uranium	Depleted	9998	Dust Collector, South	Foundry	1,5	5.89 x 10 <sup>10</sup>	13.83
Uranium	Depleted	9998	Dust Collector, North	Foundry	1,5	5.89 x 10 <sup>10</sup>	10.33



TABLE 4-11  
(Continued)

1969 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Volume (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	Rover Machining	Machining	4	5.26 x 10 <sup>9</sup>	0.00
Thorium	Natural	9201-5	Delta	Arc Melting & Forming	1,4	4.89 x 10 <sup>10</sup>	0.00
Uranium	Depleted	9204-4	Press Area	Grinding	1,2	2.73 x 10 <sup>9</sup>	36.00
Uranium	Depleted	9202	Foundry	Casting & Forming	2	1.07 x 10 <sup>10</sup>	1.01
Uranium	Depleted	9202	1st Floor Lab	Chemical Labs	1,2	2.25 x 10 <sup>10</sup>	0.53
Uranium	Depleted	9202	2nd Floor Lab	R&D	1,2,4	1.15 x 10 <sup>10</sup>	0.51
Uranium	Depleted	9202	3rd Floor Lab	R&D	1,2,4	1.52 x 10 <sup>10</sup>	1.59

- 1 Prefilter  
Efficiency: 20-35% (NBS Test or ASHRAE Standard 52-68)
- 2 Particulate Air Filter  
Efficiency: 90-95% (NBS Test or ASHRAE Standard 52-68)
- 3 Particulate Air Filter  
Efficiency: 60-65% (NBS Test or ASHRAE Standard 52-68)
- 4 High Efficiency Particulate Air Filter (HEPA)  
Efficiency: 99.97-99.99% (DOP Penetration Test)
- 5 Bag (Wool Felt)  
Efficiency: >99% (One micron and larger dust particles)
- 6 Wet scrubber
- 7 General Room Exhaust. No air cleaning facilities provided.

Source: UCC-ND, 1971a

TABLE 4-12

## 1970 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Vol (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	E-Wing Duct Collector	Casting	5	3.68 x 10 <sup>0</sup>	2.08
Uranium	Highly	9212	C-Wing Stack Exhaust	Fuel Element Fab.	4	6.83 x 10 <sup>0</sup>	0.12
Uranium	Highly	9212	D-Wing Stack	Chemical Conversion	6 or 1,4	2.15 x 10 <sup>0</sup>	0.02
Uranium	Highly	9212	West Head House Stack	Product Recovery	1,4	1.47 x 10 <sup>0</sup>	0.23
Uranium	Highly	9212	Reduction Area Exhaust	Metal Reduction	1,4	5.26 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	Room 1010 Sintering Furnace Exhaust	UO <sub>2</sub> Production	4	5.26 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	Room 1010 Exhaust	UO <sub>2</sub> Production	7	5.26 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	Dry Chemistry Reactor Hood Exhaust	Chemical Conversion	1,4	5.26 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	C-1 Wing PVC Stack	Product Preparation	6 or 1,4	3.68 x 10 <sup>0</sup>	0.33
Uranium	Highly	9212	C-1 Wing Room Exhaust	Product Preparation	7	7.88 x 10 <sup>0</sup>	0.06
Uranium	Highly	9212	Process Exhaust (Dissolver Calciner)	Product Preparation	6 or 1,5	5.26 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	B-1 Wing 3rd and 4th Floors Room Exhaust	Chemical Salvage	7	7.88 x 10 <sup>0</sup>	0.09
Uranium	Highly	9212	B-1 Wing Denitrator Room and Hood Exhaust	Chemical Conversion	1,5	2.10 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	B-1 Wing 2nd Floor Calciner and Dissolver	Product Preparation	1,5	2.10 x 10 <sup>0</sup>	<0.01
Uranium	Highly	9212	B-1 Wing 2nd Floor Exhaust	Chemical Salvage	7	5.26 x 10 <sup>0</sup>	0.02
Uranium	Highly	9212	B-1 Wing 2nd Floor Exhaust	Chemical Salvage	7	5.26 x 10 <sup>0</sup>	0.02
Uranium	Highly	9212	B-1 Wing Conversion Area Exhaust	Chemical Salvage	7	5.26 x 10 <sup>0</sup>	0.02

TABLE 4-12  
(Continued)

1970 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Vol (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	B-1 Wing 1st Floor Dissolver Area Exhaust	Chemical Salvage	7	5.26 x 10 <sup>9</sup>	<0.01
Uranium	Highly	9212	B-1 Wing 1st Floor Conversion Area Exhaust	Chemical Salvage	1,4,5	5.26 x 10 <sup>9</sup>	0.01
Uranium	Highly	9212	B-1 Wing Feed Preparation Dry Filter Exhaust	Chemical Salvage	1,5	2.10 x 10 <sup>9</sup>	0.02
Uranium	Highly	9212	C-Wing Rover Exhaust	Fuel Element Fab.	4	2.10 x 10 <sup>10</sup>	0.01
Uranium	Highly	9212	E-Wing Machine Shop	Machining	5	2.10 x 10 <sup>10</sup>	0.01
Uranium	Highly	9212	E-Wing Reduction Exhaust Plenum Chamber	Metal Conversion	1,4,5	7.35 x 10 <sup>9</sup>	0.01
Uranium	Highly	9212	Room 1022 E-Wing Lab Stack Exhaust	Chemical Lab	7	7.36 x 10 <sup>9</sup>	0.07
Uranium	Highly	9212	Room 1021 (South) E-Wing Lab Stack Exhaust	Chemical Batch	4	7.88 x 10 <sup>9</sup>	<0.01
Uranium	Highly	9212	Room 1021 (West) E-Wing Lab Stack Exhaust	Chemical Lab	7	2.63 x 10 <sup>9</sup>	0.02
Uranium	Highly	9212	Room 1021 (East) E-Wing Lab Stack Exhaust	Chemical Lab	7	2.63 x 10 <sup>9</sup>	0.17
Uranium	Highly	9215	O-Wing Mill & Room Exhaust	Metal Rolling	1,5	2.94 x 10 <sup>10</sup>	0.01
Uranium	Highly	9215	M-Wing Exhaust	Machining	1,5	2.31 x 10 <sup>10</sup>	0.03
Uranium	Depleted	9998	Dust Collector, South	Foundry	1,5	4.70 x 10 <sup>10</sup>	2.81
Uranium	Depleted	9998	Dust Collector, North (no longer in service)		3,2		

TABLE 4-12  
(Continued)

1970 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Vol (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	Rover Machining	Machining	4	5.26 x 10 <sup>9</sup>	<0.01
Thorium	Natural	9201-5	Delta	Arc Melting & Forming	1,4	4.89 x 10 <sup>10</sup>	<0.01
Uranium	Depleted	9204-4	Press Area	Grinding	1,2	2.73 x 10 <sup>9</sup>	33.30
Thorium Uranium	Natural Depleted	9202	Foundry	Electro Chemistry	1,2	4.26 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	1st Floor Lab	Graphite and Plastics	1,2	7.46 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	2nd Floor Lab	Chemical and Corrosion	1,2	1.52 x 10 <sup>10</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	3rd Floor Lab	Ceramics	1,4	8.94 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Ceramics	1,4	4.11 x 10 <sup>10</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Organic Chemistry	1	3.99 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Metallurgy (Inactive)	1,2	3.15 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Ceramics	1,4	2.63 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Applications and Ceramics	1,4	2.63 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Ceiling Exhaust, Ceramics	7	6.31 x 10 <sup>9</sup>	<0.01

TABLE 4-12  
(Continued)

1970 MONITORED AIRBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Area Exhausted	Type Operation	Type Filter System	Total Release Point Air Vol (ft <sup>3</sup> )	Quantity Radioactive Material Released (kg)
Thorium Uranium	Natural Depleted	9202	Research and Development	Ceramics	1,4	1.89 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Foundry	1,2	1.64 x 10 <sup>10</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Hood (Furnace)	1,2	2.10 x 10 <sup>9</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Foundry	1,2	1.06 x 10 <sup>10</sup>	<0.01
Thorium Uranium	Natural Depleted	9202	Research and Development	Foundry	1,2	1.07 x 10 <sup>10</sup>	<0.01

- 1 Prefilter  
Efficiency: 20-35% (NBS Test or ASHRAE Standard 52-68)
- 2 Particulate Air Filter  
Efficiency: 90-95% (NBS Test or ASHRAE Standard 52-68)
- 3 Particulate Air Filter (Not Applicable This Reporting Period)  
Efficiency: 60-65% (NBS Test or ASHRAE Standard 52-68)
- 4 High Efficiency Particulate Air Filter (HEPA)  
Efficiency: 99.97-99.99% (DOP Penetration Test)
- 5 Bag (Wool Felt)  
Efficiency: >99% (One micron and larger dust particles)
- 6 Wet scrubber
- 7 General Room Exhaust. No air cleaning facilities provided.

Source: UCC-ND, 1971b.

in Tables 4-11 and 4-12 are presented in this report to provide useful guidance in identifying those relevant release points or outfalls which were significant for off-site releases.

After 1970, health physics and environmental reports describe Y-12 radioactive effluents for the release points at the plants, but do not contain as much sampling information, e.g., stack number or total volume of exhausted air for each stack, as reported in the UCC-ND, 1971a and UCC-ND, 1971b reports mentioned in the above paragraph. Starting in 1984, Y-12 began to report radioactive effluents quantities in a classified annual "Y-12 Plant Radioactive Effluent Report." Y-12 has also provided an unclassified version of the same report. Each report only lists release quantities by individual exhaust stack. This newest report is required by DOE Order 5484.1 and identifies total quantities for both airborne and liquid effluent monitored at the Y-12 Plant. Around 1987, Y-12 significantly upgraded their stack monitoring program and added state-of-the-art air sampling and counting equipment to the production-related exhaust stacks. Of approximately 350 process exhaust stacks at Y-12, over 100 serve operations with a potential for generating airborne radioactive uranium. Today, there are 60 or more monitored stacks or exhaust lines for depleted uranium operations and 50 or more monitored stacks for enriched uranium operations. Air samples are counted, and the raw count data are compiled by the plant laboratory in 9995. More recent release estimates from these measurements can be found in Y-12 environmental reports or the previously mentioned annual discharge reports, the Y-12 Plant Radioactive Effluent Report.

Other effluents at Y-12 have been routinely filtered and monitored. For example, the Plutonium Laboratory located in Building 9995, which was under construction 1950-51, required extensive ventilation facilities, including a highly efficient air cleaner (Patterson et al., 1957; Struxness, 1951a). The lab was primarily established to perform analytical evaluation of small quantities of plutonium produced in the remaining calutron facilities in Building 9204-3 administered and operated by the Oak Ridge National Laboratory at Y-12. The lab also has been used to analyze samples taken from incoming Savannah River and Idaho Falls materials. Apparently, it was decided to monitor the effluent air from the cleaner for radioactive particulate contamination by installing a fixed, isokinetic sampler in the discharge ducts. Results for this monitoring were reported in Health Physics or Industrial Hygiene reports and can be located in the Y-12 Records Center vault.

Y-12 also decided to monitor the surrounding air outside the production buildings to determine if process controls were effective at maintaining uranium air releases at safe levels. Starting in the early 1950s, the Y-12 Health Physics Department maintained two continuous outdoor air samplers, one of which was a continuous sampler, and one of which collected samples twenty-four hours a day for only five days a week. The continuous sampler, which collected air samples seven days a week, was located a short distance east of building 9723-12. A smaller unit, which sampled five days a week, was located a short distance south of building 9983

(Patterson et al., 1957). For the twenty-four-hour, five-day samplers, air samples were exchanged at the beginning and at the end of each day shift, so that each twenty-four hour sample is actually a composite of two samples, one of approximately eight hours duration and one of approximately sixteen. This was done to lessen the negative bias of alpha count determinations which would be caused by the masking effect or self-absorption of some of the alpha particles by non-radioactive dusts collected on the filter. This masking effect was noticed on samples that were collected over the weekend (Patterson et al., 1957). The samples collected by these ambient outdoor samplers were counted for both alpha and beta-gamma activity. The average airborne contamination in the atmosphere as indicated by these samples were reported to plant supervision and others in the monthly General Plant Air Contamination Report (Patterson et al., 1957). The continuous air sample was used to evaluate trends in air concentrations outside the production building. More samplers have been added through the years. In the late 1970s and early 1980s, some of the additional samplers were added based on studies that determined actual wind patterns in and around the Y-12 plant. Most of the samplers are located centrally in the Y-12 valley and surrounding ridges, generally downwind from most of the uranium processing areas (Hougland et al., 1982; Hunt, 1993). Starting in 1971, monitoring results for these samples have been published in the annual AEC, ERDA, or DOE environmental reports for the Oak Ridge Reservation.

Starting in the mid 1970s, isotopic alpha spectrometry of uranium-238, uranium-235, and uranium-234 and gross alpha/beta counting of perimeter air filters have been performed by the Oak Ridge Y-12 Plant Laboratory (Building 9995) in support of the Environmental Monitoring Section of the Radiation Safety Department. Weekly samples have been gross alpha/beta counted and an isotopic analysis performed on quarterly composites. Calculations of air concentrations and data management are done in the plant lab mentioned above (Hinton et al., 1984).

Release estimates from monitored liquid effluents from Y-12's main production and support facilities have been historically reported in monthly or quarterly Health Physics, Industrial Hygiene, and Y-12 Plant reports. Much of the uranium releases were from depleted uranium operations. Limited information regarding the number and types of samples taken prior to 1960 was identified during the investigation. By 1969, Y-12 had significantly increased the number of samples for both depleted and enriched uranium effluents. Detailed annual radioactive liquid effluent reports were published for 1969 and 1970, and are shown in Tables 4-13 and 4-14 (UCC-ND, 1971a; UCC-ND, 1971b). Some information has been eliminated from these tables because of security and classification issues. Similar reports after 1970 were not identified during this investigation. However, the data in Tables 4-13 and 4-14 are presented in this report to provide useful guidance in identifying those relevant and release points or outfalls which were significant for off-site releases.

TABLE 4-13

## 1969 MONITORED WATERBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Type Operation	Type In-Plant Waste Treatment	Type In-Plant Release Point	Final Release Point To Environment	Total Vol Waste Stream (gals)	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	Uranium Recovery	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	44,670	1.5
Uranium	Highly	9212	Uranium Recovery	Acid Ponds	Pipeline	Ground	433,300	1.8
Uranium	Highly	9212	Uranium Recovery	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	35,700	1.2
Uranium	Highly	9212	Uranium Recovery	Acid Ponds	Pipeline	Ground	800	2.1
Uranium	Highly	9212	Uranium Recovery	Acid Ponds	Pipeline	Ground	404,000	1.3
Uranium	Highly	Area 5	Uranium Recovery	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	1,248,000	3.7
Uranium	Highly	Area 5	Uranium Recovery	City of Oak Ridge West Sewage Treatment Plant	Sanitary Sewer	East Fork Poplar Creek	-	1.9
Uranium	Highly	Area 5	Uranium Recovery	None	-	-	-	1.9
Uranium	Depleted	9995	Plant Laboratory	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	1,220	39
Uranium	Depleted	9998	Pickling	Acid Ponds	Pipeline	Ground	2,800	3,500 <sup>2</sup>
Uranium	Depleted	9204-4	Plating	Acid Ponds	Trucked to Acid Pond	Ground	850	310 <sup>2</sup>
Uranium	Depleted	9204-4	Plating	Acid Ponds	Trucked to Acid Pond	Ground	2,900	280 <sup>2</sup>
Thorium	Normal	9201-5	Pickling	Acid Ponds	Trucked to Acid Pond	Ground	12,000	4,400 <sup>4</sup>



TABLE 4-13  
(Continued)

1969 MONITORED WATERBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Type Operation	Type In-Plant Waste Treatment	Type In-Plant Release Point	Final Release Point To Environment	Total Vol Waste Stream (gals)	Quantity Radioactive Material Released (kg)
Uranium	Highly	New Hope Pond	Settling Basin	Settling Equalization	None	East Fork Poplar Creek	$3.6 \times 10^9$	11 <sup>2</sup>
Uranium	Depleted	New Hope Pond	Settling Basin	Settling Equalization	None	East Fork Poplar Creek	$3.6 \times 10^9$	39 <sup>2</sup>
Uranium	Depleted	Acid Ponds	Percolation Ponds	Percolation	None	Ground to Bear Creek	$9 \times 10^{(3)}$	535 <sup>2</sup>
Thorium	Normal	Acid Ponds	Percolation Ponds	Percolation	None	Ground to Bear Creek	$9 \times 10^{(3)}$	95 <sup>2</sup>

- 1 Two monitoring stations (one in each of two storm sewers which serve Area 5) take continuous samples which are composited and analyzed weekly for radioactive materials.  
 2 These are estimated quantities.  
 3 Bear Creek flow at sample point.

Source: UCC-ND, 1971a.

TABLE 4-14

## 1970 MONITORED WATERBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Type Operation	Type In-Plant Waste Treatment	Type In-Plant Release Point	Final Release Point To Environment	Total Vol Waste Stream (gals)	Quantity Radioactive Material Released (kg)
Uranium	Highly	9212	Uranium Recovery	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	43,000	1.3
Uranium	Highly	9212	Uranium Recovery	Acid Ponds	Pipeline	Ground	446,000	2.2
Uranium	Highly	9212	Uranium Recovery	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	29,000	0.9
Uranium	Highly	9212	Uranium Recovery	Acid Ponds	Pipeline	Ground	500	1.0
Uranium	Highly	9212	Uranium Recovery	Acid Ponds	Pipeline	Ground	331,000	2.0
Uranium	Highly	Area 5	Uranium Recovery	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	1,078,000	3.5
Uranium	Highly	Area 5	Uranium Recovery	City of Oak Ridge West Sewage Treatment Plant	Sanitary Sewer	East Fork Poplar Creek	-	1.5
Uranium	Highly	Area 5	Uranium Recovery	None	-	-	-	1.5
Uranium	Depleted	9995	Plant Laboratory	New Hope Pond	Storm Sewer <sup>1</sup>	East Fork Poplar Creek	925	20
Uranium	Depleted	9998	Pickling	Acid Ponds	Pipeline	Ground	2,800	2,800
Uranium	Depleted	9204-4	Plating	Acid Ponds	Trucked to Acid Pond	Ground	430	160 <sup>(2)</sup>
Uranium	Depleted	9204-4	Plating	Acid Ponds	Trucked to Acid Pond	Ground	1,450	140 <sup>(2)</sup>
Thorium	Normal	9201-5	Pickling	Acid Ponds	Trucked to Acid Pond	Ground	7,200	720 <sup>(2)</sup>
Uranium	Highly	New Hope Pond	Settling Basin	Settling Equalization	None	East Fork Poplar Creek	3.2 x 10 <sup>9</sup>	9 <sup>(2)</sup>

TABLE 4-14  
(Continued)

1970 MONITORED WATERBORNE RELEASES FOR THE Y-12 PLANT

Type Radioactive Material	Enrichment	Bldg. No.	Type Operation	Type In-Plant Waste Treatment	Type In-Plant Release Point	Final Release Point To Environment	Total Vol Waste Stream (gals)	Quantity Radioactive Material Released (kg)
Uranium	Depleted	New Hope Pond	Settling Basin	Settling Equalization	None	East Fork Poplar Creek	$3.2 \times 10^9$	20 <sup>(2)</sup>
Uranium	Depleted	Acid Ponds	Percolation Ponds	Percolation	None	Ground to Bear Creek	$9 \times 10^{8(3)}$	594 <sup>(2)</sup>
Thorium	Normal	Acid Ponds	Percolation Ponds	Percolation	None	Ground to Bear Creek	$9 \times 10^{8(3)}$	105 <sup>(2)</sup>

- 1 Two monitoring stations (one in each of two storm sewers which serve Area 5) take continuous samples which are composited and analyzed weekly for radioactive materials.  
 2 These are estimated quantities.  
 3 Bear Creek flow at sample point.

Source: UCC-ND, 1971b

Y-12 operations, which used chlorinated solvents such as PCBs, perchloroethylene, and carbon tetrachloride and other materials such as benzene, were monitored on a regular basis dating back to the late 1940s. Most of the data pertains to general work area air monitoring, most likely performed in order to insure safe working conditions inside Y-12 process buildings. These monitoring results can be found in health physics and industrial hygiene reports. The information reviewed during the investigation suggests that effluent monitoring for these and other chemicals as well as metals were not routinely performed at Y-12. Historical chemical release data was not identified during this investigation. Many of the old chemical inventory, storage, or purchasing records have been destroyed, however, some of this information was identified during this investigation. Retired production and limited inventory records are available at the Y-12 Records Center. These records characterize the types and quantities of chemicals and the time period they were used at Y-12. These records may be of potential value in a further quantitative evaluation of chemical releases from the Y-12 Plant. And finally, a historical chemical release summary report was compiled by Y-12 back in 1986 and may be of some value in future studies (Fee, 1986). This report contains quantities of specific chemicals used and released from the Y-12 Plant for those periods of history where information was found.

#### **Lithium Separation and Enrichment Operations**

From 1950 to 1963, the lithium separation process resulted in significant airborne and waterborne releases of mercury. Most of the waterborne effluents occurred through the East Fork Poplar Creek (EFPC). Historical monitoring results for mercury concentrations in liquid effluents (e.g., storm drains) have been reported in Health Physics or Industrial Hygiene reports since the 1950s. Since 1971, these results have been published in annual AEC, ERDA, and DOE environmental reports, which include other areas of the Oak Ridge Reservation (K-25 and X-10).

Routine airborne monitoring of the workplace for mercury was performed in the lithium isotope separation buildings starting back in the early 1950s. Monitoring results were reported in monthly health physics and industrial hygiene reports, special project reports or memorandums, and quarterly Y-12 plant reports. Many of these earlier reports can be located at the Y-12 Records Center. These include air sampling results for the lithium separation pilot production buildings (9201-4, 9201-5, 81-10, 9202, 9201-2, and 9204-4). One report discussed in detail mercury concentrations and how these were reduced with the installation of large exhaust fans located at the end of the process building. Specific concentrations of mercury and building air exchange rates are described in this report. The air exchange rate is the number of times, in a given area or building, that the volume of air contained within the building is exchanged with outside air, and usually expressed as the number of building air exchanges per hour or per day.

An extensive program was used to determine the concentrations of non-radioactive toxic contaminants in the plant atmosphere. The program was administered by the Industrial Hygiene Department. The Health Physics Department provided the manpower to do the sample collection and reported the data to the Industrial Hygiene group for interpretation and corrective action. For example, a routine mercury vapor sampling program was maintained in Buildings 9202, 9201-2, 9201-4, 9201-5, 81-10, and 9204-4; buildings in which a potentially serious mercury vapor problem was believed to have existed. Samples were collected at locations other than these at the request of the Industrial Hygiene group or area supervision. Results for these samples were reported in Health Physics and Industrial Hygiene reports.

The information gathered during this investigation suggests that the Classified Mercury File will be potentially very useful in performing a further quantitative evaluation of mercury uses at Y-12. These files contain inventory and production records and air/water monitoring results which are likely to be useful in any future study. Certainly, at a minimum, these files will help highlight important activities related to mercury use at Y-12 and provide guidance for further investigations of source term development. The Mercury Files are located at the Y-12 Records Center.

### **Beryllium Operations**

An air program for the detection of beryllium in the atmosphere was maintained for Building 9766 until the 1960s, at which time the beryllium machining operations were shut down and moved to Building 9201-5. Since the beryllium contamination typically has been in a particulate rather than a vapor form, continuous samplers using filtering collectors were used to detect the presence of beryllium in the workplace. A system of permanently installed sampling heads was used to collect samples for determining the level of general air contamination. In the early days, portable equipment was used to collect non-routine samples in areas which were considered general air, breathing zones, or operational areas. Permanent samplers for determining general air beryllium concentration were located in the machine shop and in the storage and changehouse facilities. In addition to those for general air sampling, two permanent sampling heads were installed in the filter house and exhaust stack of the shop ventilation system. These gave a rough indication of the beryllium being exhausted to the outside atmosphere.

Each morning, the 24-hour air samples collected during the previous day were removed from the samplers and analyzed for beryllium. Whatman No. 44 filter paper discs were used to collect these beryllium samples. The filter papers were placed in individual porcelain crucibles and transported to the laboratory, where they were ashed and counted. The data necessary for the calculation of beryllium concentration in  $\mu\text{g}/\text{m}^3$  of air was recorded on the "Airborne Beryllium Record" data card. The concentrations were then reported in monthly or quarterly Health Physics and Industrial Hygiene reports. This same monitoring protocol was continued in Building 9201-5 when the beryllium operations were moved there around the mid 1960s.

Much of this later monitoring data was found in Industrial Hygiene and Health Physics reports and can be located in the Y-12 Records Center vault.

### Waste Disposal Operations

Starting in the early 1950s, Y-12 collected surface water samples in and around the plant which were known to be impacted by Y-12 operations or waste disposal activities. There were essentially two main sampling points for evaluating surface waters. One sampling point was at the east end of the plant in East Fork Poplar Creek. The other was located at the west end of the plant in Bear Creek. Routine water monitoring results that cover most of Y-12's history can be found in Y-12 Plant quarterly reports and Health Physics reports. The locations and types of radioactive and chemical analyses generally have increased over the years and are reflected in these types of reports (ChemRisk, 1993).

Other useful reports include the U.S. Atomic Energy Commission's Annual Environmental Monitoring Reports for the Oak Ridge Reservation and surrounding areas, which were first published in 1971. These reports provide environmental monitoring data for air, water from surface streams, soil, plants and wildlife and address releases of radioactive and non-radioactive materials from Oak Ridge National Laboratory (ORNL), the Oak Ridge Gaseous Diffusion Plant (ORGDP), and the Y-12 Plant. For example, Y-12 monitoring results for the New Hope Pond and Bear Creek can be found in some of these reports and may be useful in any future studies. Over the years, these reports have undergone significant changes in the scope and volume of information pertaining to environmental monitoring and research data.

The Y-12 site contractors and the Department of Energy have generated partial release reports which describe estimates of historical off-site releases from the Y-12 Plant for various contaminants (Owings, 1986; DOE, 1988). Tables 4-9 and 4-10 are examples of the types of material and estimated quantities released from the Y-12 Plant. The validity of the data presented in these tables was not verified during this investigation and are only presented to show the types of information that is available that might be useful in reconstruction of historical activities and hazardous material releases associated with the Y-12 Plant. There is reportedly serious debate concerning the accuracy of many of the environmental/annual reports that have been published by DOE and its predecessors (Mobley, 1993). Extensive validation of these data and other supporting information is warranted in any future dose reconstruction studies.

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## 5.0 ENVIRONMENTAL MONITORING AND RESEARCH DATA

This section of the report addresses Task 2 of the project. Project Task 2 focusses on developing an understanding of environmental monitoring and research data for off-site areas potentially affected by releases from the DOE Oak Ridge Operations facilities. These data are reviewed to describe the extent of information which is available and to establish the potential usefulness of these data for future investigations aimed at quantifying historical exposures and health risks to off-site populations.

Available environmental monitoring and research data were identified and evaluated based on information gathered by way of personnel interviews and document reviews. Key information sources included active and former investigators, plant libraries and archives at the K-25, X-10, and Y-12 Sites, and various state and federal agencies. For each relevant document identified during the records review process, a brief abstract was prepared. The abstract and other relevant information were logged into the project document database.

Overall information availability is summarized in the following section in the form of a series of tables which outline the time periods, locations, and analytes addressed in over 100 studies of historical environmental monitoring on- and off-site of the Oak Ridge Reservation (ORR). Brief descriptions of each of these studies are provided in the text. In addition, brief discussions of several areas of off-site contamination in the vicinity of the ORR that are in some way tied to past DOE activities are included. It is important to note that while this section summarizes a large number of studies of historical environmental monitoring on- and off-site of the ORR, it should not be construed as a complete historical record of every study which has been conducted to date. It is expected, however, that this section addresses the majority of the most comprehensive and potentially useful environmental data sources for future dose reconstruction work.

### 5.1 AREAS OF INVESTIGATION

The Task 2 investigation of available environmental monitoring and research data focussed on the primary environmental media likely to have been associated with ORR releases and contaminant exposures to off-site populations. The following section describes the geographical areas of interest during the investigation. These areas were selected based on knowledge of the key release sources at the ORR facilities, an understanding of the surface water bodies that have received ORR wastes, knowledge of reported areas of contaminant accumulation in surface water sediments, and our preliminary review of environmental data availability.

### 5.1.1 Surface Water Data

Task 2 efforts to identify environmental monitoring and research data associated with surface waters have focussed on the availability of sampling and monitoring data for water, sediments, and biota in the creeks, rivers, lakes, and reservoirs that have been potentially affected by historical ORR releases.

The areas of concern for Task 2 investigations of available surface water environmental data include:

- White Oak Creek
- White Oak Lake
- White Oak Creek Embayment
- Clinch River
- Melton Hill Reservoir
- Watts Bar Reservoir
- East Fork Poplar Creek
- Poplar Creek
- Bear Creek

While White Oak Creek, White Oak Lake, White Oak Creek Embayment, and Bear Creek are not off-site of the ORR, these surface water bodies are included in this report because of the potential usefulness of the extensive volume of data collected in these areas for estimating or trending releases from the X-10 site.

Short descriptions of the surface water areas for which environmental data were sought during Task 2 investigations are provided below. The locations of these surface water areas of interest is shown in Figure 5-1.

#### White Oak Creek

White Oak Creek (WOC) is the primary surface water drainage for the X-10 site. The WOC watershed drainage area is approximately 6.5 square miles. It is a natural system that has carried discharges from ORNL facilities to off-site areas since 1943. The WOC basin includes Bethel Valley, where most of the ORNL facilities are located, and Melton Valley, site of additional facilities and radioactive waste disposal areas (MMES, 1991). WOC receives Melton Valley drainage through Melton Branch, which drains approximately 1.5 square miles in Melton Valley and enters WOC 1.56 miles above the Clinch River (Blaylock et al., 1993). Both Melton Branch and WOC receive liquid effluents from ORNL operations and leachates from radioactive waste disposal areas in the drainage basin.

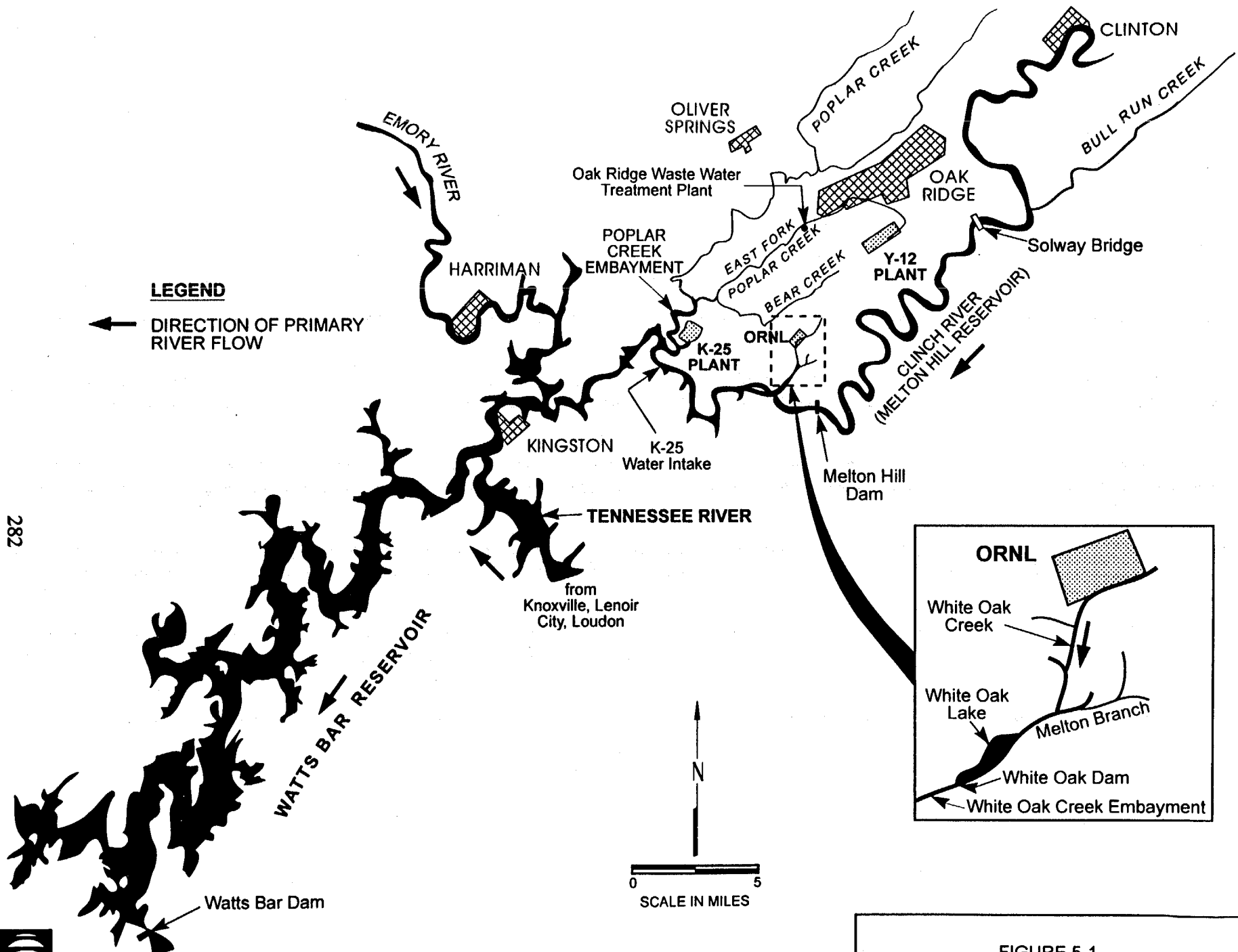


FIGURE 5-1  
SURFACE WATERS AND LANDMARKS  
OAK RIDGE RESERVATION AREA

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### White Oak Lake

White Oak Lake (WOL) is a shallow impoundment extending approximately 2300 ft above White Oak Dam (WOD). The lake has served as the final settling basin for low-level radioactive effluent from ORNL since 1943 (Blaylock et al., 1993). In the fall of 1941, the TVA placed a culvert and an earthen fill at White Oak Creek Mile (WOCM) 0.6 to be used as a highway crossing (Blaylock et al., 1993). In the spring of 1943, in order to provide a dilution and settling basin for Laboratory waste, a cofferdam was placed in the inlet side of the existing culvert. The dam was designed to hold water at an elevation of 750.0 ft (Oakes et al., 1982a). On September 29, 1944, floodwaters raised the lake elevation to 753.6 ft, one foot over the top of the roadway (Morgan and Western, 1947). The lake level remained at 746.5 ft until June, 1948, when it was lowered to 745.3 ft to facilitate mud sampling. Normal operation from 1948 to 1955 was between elevations 747 ft and 749 ft, while during flood stages the level was over 751 ft (Oakes et al., 1982a).

By 1954, WOL had reached equilibrium with WOC in terms of its ability to dilute radioactive materials (Lee and Auerbach, 1959). To provide ORNL with more volume for holdup in case of large releases and to reduce the probability of ducks residing in the contaminated water, the fish population was poisoned with rotenone and removed, and in October 1955 the lake was partially drained. The lake was drained slowly so as not to disturb the contaminated silt. Revegetation of the lake bed occurred rapidly. By June 1956, most of the lake bed was covered. Flooding in 1956 and later caused the stream channels to be eroded to preimpoundment depth (Blaylock et al., 1993). In 1961, an investigation was made to determine the deposition and/or losses of sediment in the lake bed since the draining in 1955 (Oakes et al., 1982a). Comparison of measurements of sediment depth in the lake bed and stream channel with measurements made by TVA in June 1953 showed that approximately 150,000 ft<sup>3</sup> of sediment had been transported from the lake bed due to stream channel and lake bed erosion. The observed losses could also be due to the processes of compaction and oxidation which commonly occur when a lake is drawn down and left dry for an extended period of time (Leming, 1993).

After the draining of the lake in 1955, the lake was allowed to gradually refill. From 1955 to 1969, the size of the lake gradually increased due to changes in gate elevation (Blaylock et al., 1993). In 1960, the surface area of the lake was increased to 8 acres. In 1963, the gate was reworked due to construction of Melton Hill Dam, and the lake size increased to 15.0 acres. By 1969, the lake encompassed a surface area of 26.3 acres, and remained at approximately this size until the elevation was reduced to 742 ft in 1979 for improvements in the structure of the dam and construction of a new weir (Blaylock et al., 1993). To further improve the system, a new discharge system on the northwest side that would accommodate a 100-year flood was built in 1983 (Blaylock et al., 1993). During this period, the lake surface area was 13 acres. Currently, the lake is maintained at elevation 745 ft with a surface area of approximately 17 acres (Blaylock et al., 1993).



WOD is the final control point on WOC. Low levels of radioactivity are released over the dam and are either deposited in the embayment sediments or transported to the Clinch River (Blaylock et al., 1993). Estimates of radioactive releases over WOD from 1944 to the present have been reported by ORNL and DOE, and are discussed in Section 2 of this report. Current releases are much lower than those of earlier years.

Flood flows in the WOC basin have been extensively analyzed due to concern with releases of radionuclides, especially through sediment transport (Edgar, 1978).

#### White Oak Creek Embayment

White Oak Creek Embayment (WOCE) is the name given to the body of water between WOD and the Clinch River. WOCE extends 0.6 miles downstream from WOD and converges with the Clinch River at Clinch River Mile (CRM) 20.8. Currently, the water level in WOCE is controlled by Melton Hill Dam and Watts Bar Dam in the Clinch River (Blaylock et al., 1993). The summer water elevation at the mouth of WOC in the Clinch River is 741 ft (Blaylock et al., 1993). In winter, the elevation is 733 ft. During the winter, WOC below WOD is a small stream that meanders through mud flats that are covered by water at the summer pool elevation. During the winter, the lower mud flats are occasionally covered by water due to high precipitation or the operation of Melton Hill Dam (Blaylock et al., 1993). The embayment is currently surrounded by a security fence, and only authorized personnel are permitted entry (Blaylock et al., 1992). Walkover radiation surveys conducted inside the fenced area at summer-pool (741 ft MSL) and at winter-pool (733 ft MSL) level indicated a maximum exposure rate of 3 mR/hr one meter above the soil surface (Blaylock et al., 1993).

Melton Hill Dam was completed in 1963 (Blaylock et al., 1993). The dam is used as a peaking unit in the Tennessee Valley Authority (TVA) power production grid, usually generating electricity twice a day. When the generators are operating, release of water from the dam reverses flow at WOCE and increases the depth of water at the mouth of the embayment by more than two feet within a few minutes. When the generators cease to operate, current in WOCE reverses direction and excess water is rapidly discharged into the Clinch River. This phenomenon increases the potential for sediment erosion in the lower portion of the embayment and sediment transport from the embayment to the Clinch River (Blaylock et al., 1993). Variations in flow releases from Melton Hill Dam also affect the diffusion of radioactive releases from WOD into the Clinch River (Struxness et al., 1967). High flow releases from Melton Hill Dam cause the level of water in the Clinch River to rise rapidly and thus block the outflow of water from WOCE. The effect of changes in flows over Melton Hill Dam on transport of contaminated waters from WOCE into the Clinch River were evaluated in 1963 as part of the Clinch River Study (Struxness et al., 1967).

As the result of the discovery of relatively high levels of radioactivity in the surface sediments in the lower portion of WOCE in 1990 and 1991 (Blaylock et al., 1993) and subsequent site characterization efforts, DOE acting through Martin Marietta Energy Systems conducted a time-critical removal action pursuant to the Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA), as amended by the Superfund Amendments and Reauthorization Act and provisions of the latest version of the National Contingency Plan, 40 CFR Part 300, March 1980 (Blaylock et al., 1993). This action consisted of the design and construction of a sediment-retention structure across the mouth of WOCE to prevent off-site migration of contaminated sediments into the Clinch River.

### Clinch River

The portion of the Clinch River included in the Task 2 evaluations was that between Melton Hill Dam and the confluence of the Clinch River with the Tennessee River at Tennessee River Mile (TRM) 567.6. All of the surface water systems on the ORR eventually drain into the Clinch River.

Flow in the Clinch River is regulated by releases from Norris Dam and Melton Hill Dam (MMES, 1991). At the mouth of WOC, the Clinch River is primarily influenced by discharges at Melton Hill Dam, although the actual water level at this point is regulated by both Watts Bar Dam and discharges from Melton Hill. The average discharge from Melton Hill Dam from 1963 through 1979 was 5300 ft<sup>3</sup>/s. Days of no discharge are not uncommon. Under certain operating conditions at Watts Bar, Fort Loudoun, and Melton Hill dams, flow reversals can occur in the Clinch River (MMES, 1991). TVA performed a special study in August 1991 in which they measured velocities at CRM 14.5. Measurements suggest flow reversals are of short duration and low magnitude (MMES, 1991).

### Melton Hill Reservoir

Melton Hill Reservoir is defined for the purposes of this study as that part of the Clinch River from the Solway bridge to Melton Hill Dam, at CRM 23.1. Melton Hill Dam was completed by the TVA in 1963. Discharges from Melton Hill Dam have the primary influence on the flow of the Clinch River near the ORR. Melton Hill Reservoir is located upstream of WOC's confluence with the Clinch River and thus is upstream of discharge points from ORNL. However, it has received leachate from ash disposal areas located on the ORR near the McCoy Branch and at the Bull Run Steam Plant (Suter, 1991; Leming, 1993).

While the area of Melton Hill Reservoir of primary focus for this study begins at the Solway bridge, areas above the bridge may have contributed contamination to the Clinch River directly or indirectly as a result of operations with materials of DOE origin. These include the Elza

Gate, Dupont-Smith, and CSX Railroad sites, which are described in Section 5.4, and the American Nuclear Corporation (ANC) site on the Braydon Branch. The ANC site housed a private operation that received radioactive material from ORNL and prepared sealed radiation sources for commercial distribution. Environmental contamination of radionuclides, including cobalt-60, is reported to have occurred at the ANC site.

#### Watts Bar Reservoir

Watts Bar Reservoir is the name given to the widening of the Tennessee River from the confluence of the Clinch River to Watts Bar Dam. Watts Bar Reservoir was created in 1942 when the TVA built Watts Bar Dam 38 miles downstream from the mouth of the Clinch River at TRM 529.8 (MMES, 1991). Watts Bar Reservoir is the first impoundment downstream of the ORR. Its sediments hold an accumulation of materials released from ORR facilities. Screening studies indicate that Watts Bar Reservoir is a highly efficient trap for particle-bound contaminants (Olsen et al., 1990).

#### East Fork Poplar Creek

East Fork Poplar Creek (EFPC) is a surface water drainage for much of the Y-12 facility. EFPC originates from a spring beneath the Y-12 complex in Bear Creek Valley. The stream naturally flows northeasterly along the axis of the valley. Until around 1988, the stream received discharges from the Y-12 Plant through New Hope Pond, which was constructed in 1963 to neutralize the effluent from Y-12 (Bailey and Lee, 1991). Above New Hope Pond, EFPC creek was channelized and received discharges from more than 200 individual outfalls from Y-12 (Loar et al., 1989). Beyond the former New Hope Pond, the stream channel turns 90 degrees, flows through a gap in Pine Ridge, and through the city of Oak Ridge. Beyond Oak Ridge, EFPC flows west back onto the ORR, where it drains into Poplar Creek at Poplar Creek Mile (PCM) 5.5. The total drainage area for EFPC is 29.8 square miles. Flow in EFPC is maintained year-round by effluent from the Y-12 Plant, which contributes as much as 20 ft<sup>3</sup>/s. The City of Oak Ridge municipal sewage treatment plant adds as much as 10 ft<sup>3</sup>/s. Discharges of EFPC at the United States Geological Survey (USGS) continuous record gage have been recorded since at least 1960 (Bailey and Lee, 1991).

#### Bear Creek

Bear Creek is a surface water drainage for the Y-12 Plant, with its headwaters near the former site of the S-3 Ponds. Bear Creek flows in a southwesterly direction along the axis of Bear Creek Valley and then flows northwest through Pine Ridge to drain into EFPC at approximately EFPC Mile (EFPCM) 1.5 (Bailey and Lee, 1991). The Bear Creek watershed drainage area is approximately 7.13 square miles (Bolye et al., 1982). The creek does not serve as a watershed for the main site of the Y-12 Plant; however, drainage from waste disposal and refuse areas

servicing the Y-12 Plant, ORNL, and K-25 Plant are collected in this basin (Boyle et al., 1982). Intermittent measurements of streamflow in Bear Creek at State Highway 95 (BCM 2.8) were made from April 1959 to June 1964. The average discharge from March 1985 through September 1990 was 6.38 ft<sup>3</sup>/s, with a maximum instantaneous discharge of 364 ft<sup>3</sup>/s on January 20, 1988 (Flohr et al., 1991).

### Poplar Creek

Poplar Creek originates in the Cumberland Mountains, where it drains several strip mining areas (MMES, 1990). The creek enters the ORR just north of the K-25 Site, flows through the plant area, and enters the Clinch River near CRM 12.0 (Loar et al., 1981a). Poplar Creek joins EFPC just north of the K-25 Plant area at Poplar Creek Mile (PCM) 5.5. Below the confluence of the East Fork, the creek is known as Poplar Creek Embayment. Poplar Creek Embayment is the primary surface water drainage for the K-25 Plant. The total drainage area of Poplar Creek is approximately 912 square miles (Boyle et al., 1982).

Historical surface water and sediment sampling studies performed on or near the ORR are summarized in Tables 5-1 and 5-2, respectively. For each study, the following information is provided: dates of sample collection, locations of sample collection, analytes, site or sites to which the data might be relevant, an approximation of the quantity of data compiled in the study, and a text reference, or "Study Number," which refers the reader to a specific study description that can be found under that number in Section 5.3.1 if further information is desired. The "Quantity of Data" designation is assigned based on a rough estimation of the total number of data points available in given study. In general, a study in which a very small number of samples was collected (e.g., less than 10) at a limited number of sampling locations was given a quantity designation of "Low." A study in which a large number of samples was collected (e.g., greater than 100) at a number of sampling locations and/or over a number of sampling periods was given designation of "High". A study in which an intermediate number of samples was collected was designated as "Medium".

#### **5.1.2 Atmospheric Data**

Investigation of available environmental monitoring and research data pertaining to atmospheric releases focussed on routine monitoring at ambient air monitoring stations operated by ORNL staff. Data were collected in the vicinity of airborne release points on the ORR and at a number of remote locations. Stations were located at distances of up to 120 miles from the ORR. In addition, meteorological data have been collected on the ORR as part of several different programs since the beginning of facility operations.

Table 5-1: Historical Surface Water Sampling Performed On or Near the Oak Ridge Reservation

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)		
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low				
█												White Oak Dam	Radioactivity	✓				✓				2
████████████████████												White Oak Dam	Gross Alpha and Gross Beta Radioactivities, Cs-137, Ru-106, Sr-89, Sr-90, Ce-144, Zr-95, Nb-95, I-131, Co-60, Tritium, Total Rare Earths, Plutonium	✓				✓				13
●												White Oak Creek between ORNL Monitoring Stations 2 and 3	Sr-90	✓						✓		32
●												White Oak Creek Mile 0.40	Metals, Volatile Organics, PCBs, Gross Alpha and Gross Beta Radioactivities, Tritium, I-131, Pa-234m, Sr-90, U-234, U-235, Cs-137, Co-60, Np-237, Th-228	✓				✓			38	
●												White Oak Lake	Cs-137, Co-60, Be-7, Pu-239, Pu-240, Pu-238, Mercury	✓				✓			47	
████												First Creek, Fifth Creek, White Oak Creek	Mercury	✓				✓			48	
●												Clinch River Near Harriman, TN	Radioactivity	✓	✓	✓				✓	4	
█												Clinch River Near K-25 Site, Above Elza Gate	Fluorine, Uranium, Gross Alpha and Gross Beta Radioactivities		✓	✓		✓			5	
●												Clinch Below Poplar Creek	Mercury, Manganese		✓	✓				✓	9	
█												Clinch Below Poplar Creek	Mercury		✓	✓		✓			10	
████████████████████												Clinch Miles 1.2 to 41.5	Gross Alpha and Gross Beta Radioactivities, Tritium, Co-60, Sr-90, Ru-106, Cs-137, Plutonium	✓	✓	✓		✓			13	
████												Clinch Miles 1.2 to 22.9	Sr-90, Ru-106, Co-60, Cs-137, Organics	✓	✓	✓		✓			14	
●												Melton Hill Reservoir	Mercury	✓						✓	21	
████████████████████												Clinch Below Poplar Creek	Uranium, Metals	✓	✓	✓		✓			13	

Table 5-1: Historical Surface Water Sampling Performed On or Near the Oak Ridge Reservation (Continued)

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low		
												Clinch River Near Clinch River Breeder Reactor Site, Melton Hill Dam	Metals, Water Quality Parameters	✓				✓		30
												Clinch Miles 6.8 to 24.0	Metals, Volatile Organics, Gross Alpha and Gross Beta Radioactivities, Tritium, I-131, Pa-234m, Sr-90, U-234, U-235, Cs-137, Co-60, Np-237, Th-228, Tc-99, Pu-238, Pu-239, Pb-212, U-238	✓	✓	✓		✓		38
												Watts Bar Reservoir	Cs-137	✓	✓	✓	✓			46
												Clinch River from Norris Reservoir to Watts Bar Reservoir	Metals, Organics, Tritium, Co-60, Cs-137, Sr-90	✓	✓	✓	✓			49
												Watts Bar, Melton Hill, and Norris Reservoirs	Metals, Organics, PCBs, tritium, Co-60, Cs-137, Eu-152/154, Pa-234, Sr-89, Sr-90, U-234, U-235, U-238, Pu-242, Pu-238, Pu-239/240, Pu-241, Am-241, Cm-242, Cm-243/244	✓	✓	✓		✓		50
												East Fork Poplar Creek near Y-12 Plant	Mercury, Gross Alpha and Gross Beta Radioactivities			✓	✓			6
												East Fork Poplar Creek	Mercury, Manganese			✓			✓	9
												East Fork Poplar Creek	Mercury			✓			✓	21
												East Fork Poplar Creek at New Hope Pond	Uranium, Metals			✓	✓			13
												East Fork Poplar Creek Miles 0.03 to 14.36	Metals, Volatile Organics, PCBs, Gross Alpha and Gross Beta Radioactivities, Tritium, Bi-214, Pb-214, I-131, Pa-234m, Sr-90, U-234, U-235, Cs-137, Co-60, Np-237, Th-228			✓		✓		38
												West End Wastewater Treatment Facility Discharge	Metals, Radionuclides			✓			✓	39

**Table 5-1: Historical Surface Water Sampling Performed On or Near the Oak Ridge Reservation (Continued)**

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low		
										■	East Fork Poplar Creek Near Poplar Creek	Gross Alpha and Beta Radioactivities, Thorium, U-235			✓	✓			13	
										■	East Fork Poplar Creek at New Hope Pond	Am-241, Cs-137, Co-60, Np-237, Nb-95, Pu-238, Pu-239/240, Ra-226, Ru-106, Sr-90, Tc-99, Th-228, Th-230, Th-232, U-234, U-235, U-238, Zr-95, Tritium			✓	✓			13	
		■									Poplar Creek Near K-25 Site	Fluorine, Uranium, Gross Alpha and Gross Beta Radioactivities		✓	✓		✓		5	
			●								Poplar Creek Above and Below East Fork	Mercury, Manganese		✓	✓			✓	9	
				■							Poplar Creek Mile 5.5, K-25 Site Discharge Points	Mercury		✓	✓	✓			10	
					■						Poplar Creek	Organics, Sr-90, Ru-106, Co-60, Cs-137		✓	✓		✓		14	
										■	Poplar Creek Above and Below K-25	Uranium, Metals		✓	✓	✓			13	
										●	Poplar Creek Near K-25 Site	Nitrates, Uranium		✓	✓			✓	24	
										●	Poplar Creek Mile 13.8	Metals		✓			✓		38	
											Poplar Creek Below East Fork and at Hartland Bridge	Metals, Volatile Organics, Semivolatile Organics, Pesticides, PCBs, Tritium, Co-60, Cs-137, Sr-90		✓	✓		✓		49	
											Bear Creek	Mercury, Gross Alpha and Gross Beta Radioactivities			✓	✓			6	
										●	Bear Creek	Mercury			✓			✓	21	
											Bear Creek	Uranium, Metals			✓	✓			13	
											Bear Creek Miles 2.83, 7.74	Radionuclides, PCBs			✓	✓			22	
										●	Bear Creek Near S-3 Ponds	Nitrates, Uranium			✓			✓	24	

**Table 5-1: Historical Surface Water Sampling Performed On or Near the Oak Ridge Reservation (Continued)**

Period of Sampling											Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992			X-10	K-25	Y-12	High	Medium	Low	
									●		Bear Creek Above and Below West Stream, Above and below East Stream	Organics, Metals			✓		✓		36
									●		Bear Creek	Volatile Organics, PCBs, Metals			✓		✓		37
										●	Bear Creek Mile 0.55	Metals, Volatile Organics, PCBs, Gross Alpha and Gross Beta Radioactivities, Tritium, I-131, Pa-234m, Sr-90, U-234, U-235, Cs-137, Co-60, Np-237, Th-228			✓			✓	38
										—	Bear Creek Miles 2.02 to 7.68	Metals, Volatile Organics, PCBs, Gross Alpha and Gross Beta Radioactivities, Am-241, Cm-242/243, Cm-244, Cs-137, K-40, Np-237, Pu-238, Pu-239/240, Tc-99, Th-232, Th-230, U-233/234, U-235, U-238			✓	✓			41
										—	Bear Creek	Gross Alpha and Beta Radioactivities, Thorium, U-235			✓	✓			13
										●	Bear Creek Miles 0.87 to 7.68	Herbicides, Pesticides, PCBs			✓	✓			45



**Table 5-2: Historical Sediment Sampling Performed On or Near the Oak Ridge Reservation**

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)		
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low				
█												White Oak Creek, White Oak Lake	Gross Beta Radioactivity	✓						✓		1
█												White Oak Lake	Radioactivity	✓					✓		3	
█												White Oak Lake Bank	Gross Beta Radioactivity	✓						✓	7	
█												White Oak Lake Bed	Sr-90, Cs-137, Co-60, Ce-144/Pr-144, Ru-106/Rh-106, Rare Earths	✓						✓	11	
●												White Oak Lake Bed	Surface Radiation	✓						✓	12	
●												White Oak Lake Bed	Gross Gamma Radioactivity	✓						✓	15	
●												White Oak Lake Bed	Ru-106	✓						✓	17	
●												White Oak Lake Bed	Co-60, Y-90, Sr-90	✓						✓	18	
●												White Oak Lake, White Oak Creek Miles 1.12, 2.68	Ru-106, Cs-137, Co-60	✓						✓	19	
●												White Oak Creek Mile 0.0, White Oak Lake	Sr-90, Cs-137	✓						✓	20	
●												White Oak Creek, White Oak Lake	Cs-137, Ruthenium	✓						✓	23	
█												White Oak Creek Mile 0.3	PCBs	✓						✓	26	
█												White Oak Creek	Sr-90, Co-60, Cs-137	✓					✓	33		
●												White Oak Creek Mile 0.18 to 0.55	PCBs, Metals, Organics, Gross Alpha and Gross Beta Radioactivities, Uranium, Sr-89, Sr-90, Co-60, Cs-134, Cs-137, K-40, Ra-226, Th-234, Eu-152, Eu-154, Am-241, Ac-228, Pa-234, Pu-238, Pu-239, Cm-244	✓					✓		40	
█												First Creek, Fifth Creek, White Oak Creek	Mercury	✓						✓	48	
█												White Oak Creek Embayment	Metals, Organics, Gamma Radioactivity, Co-60, Cs-137	✓					✓		51	

Table 5-2: Historical Sediment Sampling Performed On or Near the Oak Ridge Reservation (Continued)

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)												
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low														
██████████												Clinch River Miles 0.0 - 27.5, Tennessee River Miles 354 - 571	Surface Gamma Radiation, Gross Beta Radioactivity, Cs-137, Sr-90, Ce-144, Rare Earths, Ru-106, Co-60	✓	✓	✓	✓												8			
██████████												Clinch Miles 5.5, 14.4, 41.5; Tennessee Miles 471, 530, 591	Metals, Sr-90, Ru-106, Co-60, Cs-137	✓	✓	✓	✓													14		
●												Clinch Miles 0 to 15	Radionuclides	✓	✓	✓													16			
●												Melton Hill Reservoir	Mercury	✓	✓	✓														21		
██████████												Clinch River near Grubb Island, Melton Hill Reservoir	PCBs	✓	✓	✓														26		
●												Watts Bar and Melton Hill Reservoirs	Metals, Water Quality Parameters, PCBs	✓	✓	✓														27		
██████████												Clinch Above and Below Poplar Creek	Metals	✓	✓	✓														29		
●												Clinch Miles 0.0 to 20.8; Tennessee Above and Below Clinch	Cs-137, Co-60, Sr-90, Pu-239, Pu-240, Am-241, Cm-244	✓	✓	✓	✓													31		
●												Clinch Above and Below Poplar Creek	PCBs	✓	✓	✓														34		
●												Melton Hill Reservoir, Tennessee River from Watts Bar Reservoir to Guntersville Dam	Mercury, PCBs, Chromium, Gross Alpha and Gross Beta Radioactivities, Uranium, Sr-89, Sr-90, Co-60, Cs-137, K-40, Ra-226, Th-234, Eu-152, Eu-154, Am-241, Ac-228, Pa-234, Pu-238, Pu-239, Cm-244	✓	✓	✓	✓														40	
●												Clinch River from near K-25 Site to Watts Bar Reservoir	Metals, Organics, Cs-137, Co-60, Be-7, U-235, U-238	✓	✓	✓	✓														42	
●												Watts Bar Reservoir	Cs-137	✓	✓	✓	✓														46	
██████████												Clinch River from Norris Reservoir to Watts Bar Reservoir	Metals, Volatile Organics, Semivolatile Organics, Pesticides, PCBs, Tritium, Am-241, Cm-243/244, Cm-245/246, Cm-248, Co-60, Cs-137, Pu-238, Pu-239/240, Sr-90, U-234, U-235, U-238	✓	✓	✓	✓															49

Table 5-2: Historical Sediment Sampling Performed On or Near the Oak Ridge Reservation (Continued)

Period of Sampling											Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992			X-10	K-25	Y-12	High	Medium	Low	
										●	Melton Hill, Watts Bar, and Norris Reservoirs	Metals, Organics, PCBs, Tritium, Co-60, Cs-137, Eu-152,154, Pa-234, Sr-89, Sr-90, U-234, U-235, U-238, Pu-242, Pu-238, Pu-239/240, Pu-241, Am-241, Cm-242, Cm-243/244	✓	✓	✓	✓			50
						●					East Fork Poplar Creek at New Hope Pond	Mercury			✓		✓	21	
						■					East Fork Poplar Creek	Mercury			✓		✓	25	
						■					East Fork Poplar Creek below New Hope Pond to Near Blair Road	PCBs			✓		✓	26	
						■					East Fork Poplar Creek	Mercury			✓	✓		28	
										■	East Fork Poplar Creek	Metals			✓		✓	29	
										●	East Fork Poplar Creek	PCBs			✓		✓	34	
										■	East Fork Poplar Creek Miles 1.3 to 14.2	Metals			✓	✓		35	
										●	East Fork Poplar Creek Miles 1.2 to 14.4	PCBs, Metals, Organics, Gross Alpha and Gross Beta Radioactivities, Uranium, Sr-89, Sr-90, Co-60, Cs-134, Cs-137, K-40, Ra-226, Th-234, Eu-152, Eu-154, Am-241, Ac-228, Pa-234, Pu-238, Pu-239, Cm-244			✓	✓		40	
										●	East Fork Poplar Creek near K-25	Metals, Organics, Cs-137, Co-60, Be-7, U-235, U-238			✓	✓		42	
						■					Poplar Creek	Mercury		✓	✓		✓	25	
						■					Poplar Creek at K-25 Discharge Points	PCBs		✓	✓		✓	26	
										■	Poplar Creek Miles 0.0 to 5.5	Metals		✓	✓	✓		29	
										●	Poplar Creek Above K-25 Site to Mouth	PCBs		✓	✓	✓		34	
										●	Poplar Creek Miles 0.8 to 7.1	Metals, Organics, Cs-137, Co-60, Be-7, U-235, U-238		✓	✓	✓		42	
										●	Poplar Creek Near Blair Road Bridge	Metals, Organics, Cs-137, U-238, K-40		✓	✓		✓	44	

Table 5-2: Historical Sediment Sampling Performed On or Near the Oak Ridge Reservation (Continued)

Period of Sampling											Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)	
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992			X-10	K-25	Y-12	High	Medium	Low		
										■	Poplar Creek	Metals, Volatile Organics, Semivolatile Organics, Pesticides, PCBs, Tritium, Am-241, Cm-243/244, Cm-245/246, Cm-248, Co-60, Cs-137, Pu-238, Pu-239/240, Sr-90, U-234, U-235, U-238		✓	✓	✓				49
						●					Bear Creek	Mercury			✓			✓	21	
										■	Bear Creek	Mercury			✓			✓	25	
										■	Bear Creek Miles 0.4 to 7.6	Metals			✓		✓		35	
										●	Bear Creek Upstream & Downstream of Burial Grounds	Volatile Organics, PCBs, Metals			✓		✓		37	
										●	Bear Creek Miles 0.55 to 7.40	PCBs, Metals, Organics, Gross Alpha and Gross Beta Radioactivities, Uranium, Sr-89, Sr-90, Co-60, Cs-134, Cs-137, K-40, Ra-226, Th-234, Eu-152, Eu-154, Am-241, Ac-228, Pu-234, Pu-238, Pu-239, Cm-244			✓	✓			40	
										■	Bear Creek	Metals, Organics, PCBs, Gross Alpha and Gross Beta Radioactivities, Am-241, Cm-242/243, Cm-244, Cs-137 K-40, Np-237, Pu-238, Pu-239/240, Tc-99, Th-232, Th-230, U-233/234, U-235, U-238			✓	✓			41	
										●	Bear Creek Miles 0.64 to 7.5	Metals, PCBs, Volatile Organics, Cs-137, K-40, U-235, U-238			✓	✓			43	
										●	Bear Creek Mile 0.87 to 7.68	Herbicides, Pesticides, PCBs			✓		✓		45	

Historical monitoring of airborne releases performed on or near the Oak Ridge Reservation is summarized in Table 5-3. Each text reference in the table identifies the specific study description in Section 5.3.2 of this report that can be consulted for further details.

### **5.1.3 Biological Monitoring Data**

Investigation of biological monitoring data focussed on the availability of data for aquatic and terrestrial foodstuff potentially impacted by releases from the ORR, including fish and other aquatic biota, milk, waterfowl, deer, beef, and vegetation. Investigations of aquatic biota emphasized those studies in which data were collected from the previously mentioned surface water systems of concern. Beginning around 1961, milk in off-site areas was monitored routinely by ORNL staff. In addition, beginning in the late 1980s, levels of contaminants in deer and waterfowl captured from on-site locations were routinely measured to evaluate the potential hazards to hunters. Special short-term studies evaluating the presence of contaminants in waterfowl, beef, deer, turtles, and vegetation in on- and off-site areas have also been evaluated.

Historical sampling of aquatic biota performed on or near the Oak Ridge Reservation is summarized in Table 5-4. Sampling programs that have addressed milk, waterfowl, deer, vegetation, and beef on or near the Reservation are summarized in Table 5-5. Each text reference in these tables identifies the specific study description in Section 5.3.3 that can be consulted for further details.

### **5.1.4 Soil Data**

The investigation of available environmental monitoring data pertaining to contamination of soil in off-site areas of the ORR identified routine soil sampling and surface radiation measurements at the perimeter and remote air monitoring network stations. Special studies of floodplain soils and areas where ORR soils have been used as fill have also been identified.

Historical soil sampling and surface radiation level monitoring performed on or near the Oak Ridge Reservation are summarized in Table 5-6. Each text reference in the table identifies the specific study description in Section 5.3.4 of this report that can be consulted for further details.

### **5.1.5 Drinking Water/ Groundwater Data**

The investigation of available data on contaminant concentrations in drinking water focussed on routine monitoring of drinking water intakes for the K-25 site and the cities of Oak Ridge and Kingston and monitoring of wells and other water supplies in areas beyond the ORR boundary. Groundwater on the ORR has also been routinely monitored by K-25, X-10, and Y-12 staff since the mid-1980s.



**Table 5-4: Historical Aquatic Biota Sampling Performed On or Near the Oak Ridge Reservation  
(Fish Unless Otherwise Noted)**

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)			
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low					
■												White Oak Creek/White Oak Lake	Radioactivity	✓						✓			53
■												White Oak Lake	Sr-89, Sr-90, Cs-137, Y-90	✓				✓				54	
●												White Oak Lake	Cs-137, Co-60, Sr-90	✓						✓		55	
■												White Oak Lake	Cs-137	✓						✓		56	
●												White Oak Lake	Tritium	✓						✓		61	
●												White Oak Creek/White Oak Lake	Metals	✓					✓			66	
●												White Oak Creek Mile 0.2, White Oak Lake	Metals, Organics, PCBs, Bi-214, Tc-99, Ac-228, Cs-134, Cs-137, Co-60, Sr-89, Sr-90, Pb-214	✓					✓			71	
■												Northwest Tributary of WOC, Melton Branch, Fifth Creek, WOC	Metals, Organics	✓					✓			72	
■												Clinch Miles 13, 18, 21	Sr-89, Sr-90, Cs-137, Y-90	✓	✓	✓	✓				54		
●												Clinch Mile 21	Sr-90	✓						✓		55	
■												Clinch River Downstream of WOC to Watts Bar Reservoir	Gamma Emitters and Sr-90	✓					✓			56	
■												Clinch River	Gamma Emitters, Sr-90, Pu-239, Pu-238, U-235, U-238, U-234, Cs-137, Co-60, K-40, Mercury, PCBs	✓	✓	✓	✓	✓				59	
■												Clinch Miles 4.5 - 13.5, Melton Hill Reservoir	Mercury		✓	✓	✓					62	
■												Clinch River	Mercury	✓	✓	✓	✓					63	
●												Clinch Miles 10.5, 11.5, 15.0	Metals, PCBs		✓	✓	✓					64	
●												Clinch River	Uranium		✓					✓		65	
●												Clinch Miles 19, 22, Melton Hill Reservoir	Metals	✓	✓	✓			✓			66	

**Table 5-4: Historical Aquatic Biota Sampling Performed On or Near the Oak Ridge Reservation (Continued)  
(Fish Unless Otherwise Noted)**

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low		
										●		Clinch Miles 2, 6, 11; Melton Hill Dam	Metals, Organics, PCBs, Bi-214, Tc-99, Ac-228, Cs-134, Cs-137, Co-60, Sr-89, Sr-90, Pb-214	✓	✓	✓	✓			71
										■		Clinch River near X-10	Metals, Organics	✓					✓	72
										■		Watts Bar and Melton Hill Reservoirs	Metals, Pesticides, PCBs	✓	✓	✓		✓		74
										■		Watts Bar and Melton Hill Reservoirs	PCBs	✓	✓	✓		✓		75
										●		Clinch Miles 0 - 51, Tennessee Miles 518 -557	Metals, PCBs, Pesticides, Semivolatile Organics, Co-60, Cs-137, Sr-90	✓	✓	✓	✓			76
						●						East Fork Poplar Creek	Mercury			✓			✓	60
										●		East Fork Poplar Creek Miles 1.3 to 14.2	Mercury			✓	✓			67
										●		East Fork Poplar Creek from New Hope Pond to Bear Creek Bridge	Mercury (frogs and crayfish)			✓			✓	69
										●		Confluence of EFPC with Poplar Creek, and EFPC	Mercury (turtles)			✓			✓	70
										●		East Fork Poplar Creek Miles 4.0, 8.8, 13.8	Metals, Organics, PCBs, Bi-214, Tc-99, Ac-228, Cs-134, Cs-137, Co-60, Sr-89, Sr-90, Pb-214			✓	✓			71
										■		East Fork Poplar Creek	Metals, Organics			✓		✓		72
										●		East Fork Poplar Creek Miles 0 to 14.4	Mercury			✓			✓	73
										■		Poplar Creek Miles 0.0 to 5.5	Mercury		✓	✓	✓			62
										■		Poplar Creek	Mercury		✓	✓	✓			63
										●		Poplar Creek Miles 0.5, 5.5, 11.0	Metals, PCBs		✓	✓	✓			64
										●		Poplar Creek	Uranium		✓				✓	65
										●		Poplar Creek Above & Below EFPC, near Mile 0.0	Mercury		✓	✓		✓		68
										■		Mitchell Branch of Poplar Creek	Metals, Organics		✓	✓		✓		72



**Table 5-4: Historical Aquatic Biota Sampling Performed On or Near the Oak Ridge Reservation (Continued)  
(Fish Unless Otherwise Noted)**

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low		
										●		Poplar Creek Miles 0.5 to 6.0	Metals, PCBs, Pesticides, Semivolatile Organics, Co-60, Cs-137, Sr-90		✓	✓		✓		76
						●						Bear Creek	Mercury			✓			✓	60
										●		Bear Creek Miles 0.4 to 7.6	Mercury			✓	✓			67
										●		Bear Creek	Mercury (frogs, crayfish)			✓			✓	69
										●		Bear Creek Mile 0.4	Metals, Organics, PCBs, Bi-214, Tc-99, Ac-228, Cs-134, Cs-137, Co-60, Sr-89, Sr-90, Pb-214			✓	✓			71

Table 5-5: Historical Biological Monitoring Performed On or Near the Oak Ridge Reservation

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)					
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low							
<b>MILK</b>																									
[Redacted]												Within 50 Miles of ORNL	I-131 and Sr-90 in Cows' Milk	✓	✓	✓	✓								77
<b>WATERFOWL</b>																									
●												White Oak Lake	Radioactivity (Coots & Ducks)	✓								✓			78
[Redacted]												White Oak Lake	Cs-137, Sr-90 (Ducks)	✓							✓			79	
[Redacted]												ORNL Settling Ponds	Cs-137 (Geese)	✓							✓			80	
<b>DEER</b>																									
[Redacted]												Oak Ridge Reservation	Cs-137, Gamma Emitters	✓	✓	✓				✓				82	
●												ORNL Solid Waste Storage Areas	Sr-90	✓							✓			83	
[Redacted]												East Fork Poplar Creek Floodplain	Metals			✓					✓			84	
<b>VEGETATION</b>																									
[Redacted]												Perimeter and Remote Air Monitoring Stations	Fluoride, Uranium, Plutonium, Tc-99, Cs-137, Be-7, Sr-90, Nb-95, Zr-95, Ru-103, La-140, Ce-144 (grass)	✓	✓	✓	✓							85	
[Redacted]												Perimeter of K-25	Tc-99		✓					✓				86	
●												East Fork Poplar Creek Floodplain	Mercury (pasture grass)			✓				✓				87	
[Redacted]												East Fork Poplar Creek Floodplain	Mercury (native vegetation and garden vegetables)			✓				✓			88		
<b>BEEF</b>																									
[Redacted]												Cattle Thyroids Collected from Within 100 miles of ORR	I-131	✓						✓				77	
●												East Fork Poplar Creek Floodplain	Mercury (cow tissue)			✓					✓			89	

**Table 5-6: Historical Soil and Surface Radiation Monitoring Performed On or Near the Oak Ridge Reservation**

Period of Sampling												Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)						
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992	X-10			K-25	Y-12	High	Medium	Low								
																		Perimeter and Remote Air Monitoring Stations	Plutonium, Uranium, K-40, Sr-90, Cs-137, Ra-226 (soil)	✓	✓	✓	✓			90
																		Perimeter of K-25	Tc-99		✓			✓		91
																		East Fork Poplar Creek Floodplain	Metals, PCBs, Radionuclides (soil)			✓		✓		92
																		Oak Ridge Sewer Beltway	Mercury, Uranium (soil)			✓		✓		93
																		Standard Monitoring Locations, Perimeter and Remote Monitoring Stations	External Gamma Radiation	✓	✓	✓	✓			94
																		Oak Ridge Reservation	Aerial Background Radiation Surveys	✓	✓	✓		✓		97
																		Oak Ridge Reservation	Aerial Gamma Survey; Gamma Radiation, Cs-137, Co-60, Pa-234m, Tl-208, U-235, Bi-214, K-40	✓	✓	✓	✓			96
																		Oak Ridge Reservation	Surface Gamma Radiation	✓	✓	✓			✓	95

Historical drinking water monitoring performed near the Oak Ridge Reservation is summarized in Table 5-7. Each text reference in the table identifies the specific study description in Section 5.3.5 of this report that can be consulted for further details.

## **5.2 THE INVESTIGATIVE PROCESS USED FOR ENVIRONMENTAL DATA**

The investigation of the availability of historical off-site environmental monitoring data involved an extensive review of historical records and published documents. This section describes information sources that proved most useful and summarizes the approaches used to locate information relevant to the study. Of the over 1150 documents summarized to-date for this study, at least 550 address environmental monitoring data.

### **5.2.1 Key Information Sources: Document Centers**

The majority of environmental monitoring data identified to be of interest were located in reports and correspondence on file at the X-10, K-25, and Y-12 document centers, including:

- X-10 Lab Records,
- The X-10 Tiger Team Records Center,
- The K-25 Site Document Response Center,
- The K-25 Environmental Management Document Center,
- The K-25 Compliance and Environmental Policy Document Center, and
- The K-25 and Y-12 Environmental Restoration Document Centers.

Each of these document centers is described in detail in Sections 2, 3, or 4, depending on the document center's location. In addition, a large number of documents of interest were identified at the DOE Information Resources Center for the Environmental Restoration Program.

### **5.2.2 Key Information Sources: Agencies**

The availability of environmental monitoring data from local, state, and national agencies which have conducted work in the vicinity of the ORR is discussed below.

**Table 5-7: Historical Drinking Water Sampling Performed On or Near the Oak Ridge Reservation**

Period of Sampling											Location	Analyte	Relevant to Evaluating Emissions From			Quantity of Data			Text Reference (Study Number)	
1943	1945	1950	1955	1960	1965	1970	1975	1980	1985	1992			X-10	K-25	Y-12	High	Medium	Low		
—————											Kingston Water Plant, K-25 Site Water Intake, City of Oak Ridge Water Intake	Sr-90, Ce-144, Cs-137, Ru-103/106, Co-60, Zr-95/Nb-95	✓	✓	✓	✓				98
									● ●		Communities to the North of K-25	Radionuclides	✓	✓	✓			✓	99	
										●	Bradbury and Poplar Springs Communities South of K-25 Site	Gross Alpha and Gross Beta Radioactivities, Gamma Activity, Tritium	✓	✓	✓			✓	101	
										●	Oliver Springs, Harriman, Knoxville, Lenoir City, Kingston	Water Quality Parameters, Cs-137, Gross Alpha and Gross Beta Radioactivities, Strontium, Tritium	✓	✓	✓			✓	102	
										●	Kingston, Harriman	Sr-90, Mercury, Tritium, Gross Alpha and Gross Beta Radioactivities, PCBs, Metals	✓	✓	✓			✓	103	
										—	Kingston, Oliver Springs	Volatile Organics, Metals, Fluorides, Sulfate, Nitrate, Uranium, Gross Alpha and Gross Beta Radioactivities, Tritium, Strontium, Tc-99, Co-60	✓	✓	✓		✓		104	

### The United States Environmental Protection Agency (USEPA)

The USEPA maintains two major computer database networks for environmental monitoring data: the Aerometric Information Retrieval System (AIRS) and the data storage and retrieval system (STORET). Both database systems are based in North Carolina at the National Computer Center (NCC). These databases contain monitoring data collected nationwide by state and federal agencies. AIRS includes air quality compliance data while STORET contains ambient environmental monitoring data, including fish tissue and surface water data. Data are available for metals, organic compounds, and radionuclides. Data can be extracted by sample location or parameter type.

### The United States Department of Public Health

Beginning in at least the mid-1960s, the United State Department of Public Health conducted a milk monitoring problem at locations concentrated around the ORR facilities. The program was recently curtailed due to lack of funding. Analyses were for radioactive iodine and strontium-90. These data were not located. However, it is expected that the data are on file with the Tennessee Division of Radiological Health (TDRH) (Mobley, 1993).

### The United States Geological Survey (USGS)

The USGS has collected water resource data in the vicinity of the ORR for at least 40 years. Data are primarily hydrological data (e.g., flow and general chemistry) for tributary systems to the Tennessee and Cumberland river systems. In more recent years, these data have been compiled in annual reports for the water year. Data specific to the ORR are available since the mid-1980s. The Knoxville office of the USGS has a collection of documents related to hydrological data for river systems in the Knoxville area. This database was queried for the surface water systems of interest potentially affected by releases from the ORR. Documents determined to be of interest are referenced in Section 5.6.

### The Tennessee Valley Authority (TVA)

The TVA collects surface water, groundwater, air, and aquatic biota data for the entire Tennessee Valley region. Information relevant to the ORR includes studies of contaminant levels in sediment and fish in TVA reservoirs, including Watts Bar and Melton Hill Reservoirs, collection of surface water and fish tissue screening data from Oak Ridge area rivers and streams, and collection of air quality monitoring data from TVA power plants in the vicinity of the ORR. In addition, the TVA has worked closely with DOE, TDHE, and the three Oak Ridge facilities in a number of large monitoring programs, including an ecological study of White Oak Creek during the 1950s and a comprehensive instream contaminant study of the East Fork Poplar Creek floodplain during the mid-1980s.

A water quality monitoring network is maintained by the TVA for collection of water samples throughout the Tennessee Valley (TVA, 1974). Data are collected to provide up-to-date information on existing water quality conditions for long-range planning of resource development programs and for the power program, and to determine long-term trends in water quality conditions in the Tennessee Valley. The TVA has conducted water quality management studies in the Tennessee Valley since 1936. Monthly sampling for water quality parameters has been conducted at 33 TVA and Alcoa hydroelectric plants and dams since January 1974. These stations include Melton Hill Dam and Watts Bar Dam. Water samples are also collected quarterly for more complete analysis at 45 locations in the Tennessee Valley and eight locations outside the Valley. These stations are located at several of the hydroelectric and steam plants and at municipal water treatment plants and stream gaging stations. These stations include Watts Bar Dam and four locations in the Clinch River including Melton Hill Dam and Bull Run Steam Plant at CRM 48.6. Parameters monitored quarterly include water quality parameters and metals, including arsenic, beryllium, chromium, lead, and mercury. These data are all available on the USEPA STORET database. In addition, hydrological data (e.g., stream flow and precipitation) have been collected by the TVA since the 1930s and are available from the TVA Water Quality Group.

Fish community sampling and water quality data have been collected by the TVA in the Clinch River and TVA reservoirs, including Watts Bar and Melton Hill Reservoirs, since the mid-1970s. These studies focus primarily on water quality parameters, mercury, and PCBs. Since the mid-1980s, these data have been compiled in published reports. Earlier data are on file at the TVA Water Quality office in Chattanooga. All TVA fish tissue data are in the USEPA STORET database.

TVA air monitoring stations in the vicinity of the ORR were established primarily to monitor air quality in the vicinity of TVA power plants, including Bull Run Steamplant and the Kingston Power Plant. Data are collected for criteria air pollutants, including sulfur dioxide (SO<sub>2</sub>) and particulates. These data have been entered into the USEPA AIRS database.

#### The Tennessee Department of Health and Environment (TDHE)

The environmental laboratory of the TDHE analyzes environmental protection monitoring data collected by other state agencies, including the Tennessee Division of Radiological Health, the Tennessee Air Pollution Control Division, and the Tennessee Division of Water Pollution Control. The laboratory maintains records of all analytical results.

The Tennessee Division of Radiological Health (TDRH)

The TDRH has collected water data from approximately 10 locations around the ORR since 1986. One location, on the Clinch River upstream of the DOE facilities, was eliminated from the network in 1991. Two locations, on the Clinch River downstream of the K-25 site and at WOD, are monitored monthly. The remaining locations, which include Melton Hill Dam and EFPC, were monitored monthly until approximately 1989, when the frequency of sampling was reduced to quarterly. Samples are analyzed for gross alpha, gross beta, and gamma radioactivity. In addition, analyses are conducted for tritium and strontium at some locations. The data are available in raw form from the TDRH and are entered into a database maintained on the TDHE Prime mini-computer. This database will be moved to a Data General System.

The TDRH has routinely collected air monitoring data in the vicinity of three nuclear plants, including the Watts Barr Nuclear Plant on the Watts Bar Reservoir, since at least the mid-1980s. Samples are collected at least once a week and analyzed for radioactive materials. The data are compiled on a database.

The Tennessee Division of Water Pollution Control (TDWPC)

The TDWPC primarily directs its efforts to areas other than those in the immediate vicinity of ORR, since this area is heavily monitored by a number of other programs. However, the TDWPC reviews and evaluates the procedures for collection and analysis of data used by other researchers. All of the TDWPC data are either in STORET or on file at the TDWPC's Knoxville office. In addition, the TDWPC has been delegated the authority by the USEPA to issue NPDES permits to DOE for the ORR. As such, the TDWPC is responsible for enforcement of these permits.

The Tennessee Department of Environment and Conservation, Department of Energy Oversight Division (TDEC/DOE-O)

The TDEC/DOE-O, located in Oak Ridge, is a multidisciplinary group which oversees ongoing waste management, monitoring, and environmental restoration activities on the ORR and the immediate vicinity. This division interfaces with its aforementioned sister divisions (e.g., TDHE, TDRH, TDWPC) on a daily basis. The TDEC/DOE-O was created in May, 1991 when DOE and the State of Tennessee entered into the Tennessee Oversight Agreement, under which the State of Tennessee is responsible for reviewing DOE environmental programs and monitoring of surface and groundwater, fish and wildlife, and air quality both on and off the ORR (DOE, 1992).

The agreement requires the state to assess the current DOE program for environmental



monitoring to determine the need for improvement. In addition, DOE must provide monthly monitoring data to the state and develop a database of monitoring information to be shared with the state. The state must provide annual reports to DOE for public distribution on its monitoring results and its evaluation of DOE's environmental monitoring and surveillance program (DOE, 1991).

### 5.2.3 Key Information Sources: Major Multi-Media Studies

The following section briefly describes key multi-media environmental monitoring programs. Following these general overviews, specific monitoring details and selected results are described in Section 5.3.

#### ORR Routine Annual Environmental Monitoring

A program of periodic environmental monitoring of the ORR has been conducted by the Applied Health Physics Division of ORNL since at least 1953. The results of this monitoring have been summarized in two annual report series. Copies of reports from the first series, the ORNL Applied Health Physics Annual Reports, have been located for the time period from 1956 to 1983. After 1983, these reports became known as the Environmental and Occupational Safety Division Annual Progress Reports. This series summarizes the activities of the ORNL Applied Health Physics Division, including radiation monitoring, safety surveys, and industrial safety, as well as environmental surveillance. Environmental surveillance activities include surface water monitoring for radionuclides in WOC, the Clinch and Tennessee Rivers, Poplar Creek, EFPC, and Bear Creek; sediment monitoring for radionuclides in the Clinch and Tennessee Rivers; monitoring for airborne radionuclides at stations within the Local, Perimeter, and Remote Area Monitoring networks (LAMs, PAMs, and RAMs); fish monitoring for radionuclides in the Clinch River; milk monitoring for strontium-90 and iodine-131 at off-site sampling locations; grass and soil monitoring for radionuclides at the perimeter and remote monitoring stations; and background radiation surveys.

The other series of annual reports is the reservation-wide annual environmental monitoring reports. These reports have been published under a number of different names (e.g., Environmental Monitoring Report United States Department of Energy Oak Ridge Facilities Calendar Year XXXX; Environmental Surveillance of the U.S. Department of Energy Oak Ridge Reservation and Surrounding Environs During XXXX; or Oak Ridge Reservation Environmental Report for XXXX), and are available for the time period from 1971 to the present. These reports provide summary data from effluent monitoring and off-site environmental surveillance, and evaluate potential exposure pathways to humans. The numbers and locations of samples of off-site media varied, but included surface water monitoring for radionuclides on the Clinch and Tennessee Rivers, Poplar Creek, EFPC, and Bear Creek; sediment monitoring for radionuclides in the Clinch and Tennessee Rivers and for metals in the

Clinch River, EFPC, and Poplar Creek; air, rainwater, and fallout monitoring at stations in the LAM, PAM, and RAM networks; fish monitoring for radionuclides, mercury, and polychlorinated biphenyls (PCBs) in the Clinch River; milk monitoring for strontium-90 and iodine-131 at off-site locations; deer and waterfowl tissue monitoring for radionuclides on the ORR; soil and grass monitoring for radionuclides at PAM and RAM stations; background radiation measurements; and groundwater and drinking water monitoring. During the period of overlap for the two series of reports (1971 to 1983), data from monitoring by the ORNL Applied Health Physics Division are presented in both.

Additional information provided in the annual environmental monitoring reports for the ORR which may be useful for future dose reconstruction studies includes descriptions of site geology and hydrogeology, compliance summaries for discharges from each of the three ORR facilities, discussions of waste generation and waste management activities, summaries of meteorological data, and discussions of populations within the vicinity of ORR according to direction and distance. In addition, estimated potential radiation and chemical doses to the public, based upon measured radionuclide and chemical releases and on-site meteorological data, are presented. Exposure estimates are based upon calculated doses via several exposure pathways, including air, water, the terrestrial food chain, and direct radiation/ direct chemical exposure, for populations within an 80-km (50 mile) radius of the ORR.

Only summary data are presented in the annual environmental monitoring reports. Availability of historical raw data used to prepare these report varies, and a systematic approach to archiving these data was not instituted until the mid-1980s. Raw data may be obtained through agreement with the DOE (Kornegay, 1993).

### The Clinch River Study

An interagency evaluation of radionuclide levels in the Clinch River was conducted from 1960 to 1964 to assess the dilutant capacity of the Clinch River and the long-term impact of releases of low-activity wastes from the Oak Ridge facilities (Struxness et al., 1967). Initiation of the study coincided with a period of significantly elevated ruthenium-106 releases to the Clinch River resulting from seepage from ORNL waste pits. Numerous federal and state agencies, as well as ORNL, took part in the study. Involved agencies included the U.S. Atomic Energy Commission (USAEC), the U.S. Geological Survey (USGS), the U.S. Public Health Service (USPHS), the Tennessee Valley Authority (TVA), the Tennessee Department of Public Health (TDPH), the Tennessee Stream Pollution Control Board (TSPCB), the Tennessee Game and Fish Commission (TGFC), and the ORNL Health Physics Division. The distribution and concentration of radionuclides of primary importance, selected based on quantities released, radioactive half-lives, and recommended maximum permissible concentrations in water, were evaluated by collection and analysis of water, bottom sediments, and fish. Radionuclides of

primary importance were identified as cobalt-60, strontium-90, ruthenium-106, and cesium-137. In addition, most samples were analyzed for stable environmental constituents, including metals.

Results of the Clinch River Study are summarized in a series of annual reports of study progress (Morton, 1961; 1962a; 1962b; 1963; 1965; Churchill et al., 1965; Cowser and Snyder, 1966; Carrigan et al., 1967; Carrigan and Pickering, 1967).

### The Mercury Task Force

Following the publication of the declassified version of the 1977 Mercury Inventory Report (Case, 1977) on May 17, 1983, the Mercury Task Force was established to address concerns regarding the use and impacts of use of mercury at the Y-12 Plant (UCC, 1983). The Task Force was organized to collect the available historical information on mercury accountability, to study the various programs for mercury salvage and recovery instituted through the years, and to summarize the several studies of mercury impacts on worker health and the environment (UCC, 1983). All data which were collected through 1984 to evaluate the mercury contamination of the environment in the vicinity of the ORR resulting from operations at the Y-12 Plant were compiled by the 1983 Mercury Task Force and are described in "Mercury at Y-12: A Study of Mercury Use at the Y-12 Plant, Accountability, and Impacts on Y-12 Workers and the Environment- 1950 to 1983" (UCC, 1983; Turner, 1993).

### The Oak Ridge Task Force

From 1984 through 1988, the Oak Ridge Task Force (ORTF) conducted a series of studies to investigate the possible health hazards associated with contamination of EFPC (Travis et al., 1989). Agencies involved in the Task Force included the TVA, which performed an instream contaminant study to determine the extent of contamination of surface water, sediment, fish, and floodplains; the USGS, which investigated the extent of groundwater contamination; Oak Ridge Associated Universities (ORAU), which investigated the extent of contamination of the EFPC floodplain and the terrestrial foodchain; and the United States Department of the Interior (USDI) which collected stream flow data. The focus of the evaluations was on contamination by mercury. Other contaminants evaluated included other metals, organics, and radionuclides. Numerous reports were produced in conjunction with the work of the ORTF. Reports which were identified during Project Task 2 are listed in Table 5-8.

### The TVA Instream Contaminant Study

The TVA Instream Contaminant Study was conducted to quantify the presence of contaminants in sediment and aquatic biota downstream of Oak Ridge facilities, with a particular emphasis on assessing the extent and transport of mercury contaminated sediments. A total of 1,526 samples of water, sediment, and biota were collected between April 16, 1984 and April 7, 1985.

TABLE 5-8

REPORTS PRODUCED BY THE OAK RIDGE TASK FORCE (ORTF)

TVA
TVA. 1985. Instream Contaminant Study- Task 1, Water Sampling and Analysis.
TVA. 1985. Instream Contaminant Study- Task 2, Sediment Characterization, V. 1.
TVA. 1985. Instream Contaminant Study- Task 2, Sediment Characterization, V. 2.
TVA. 1985. Instream Contaminant Study- Task 3, Sediment Transport.
TVA. 1985. Instream Contaminant Study- Task 4, Fish Sampling and Analysis.
TVA. 1986. Instream Contaminant Study- Task 5, Summary Report.
Travis, C.C., F.O. Hoffman, B.G. Blaylock, K.L. Daniels, C.S. Gist, C.W. Weber. 1986. Preliminary Review of TVA Fish Sampling and Analysis Report, Report of Task Group Five.
USGS
USGS. 1985. Open-File Report 85-165, Water-quality Data for 34 Sites, April and June, 1984, Near the Y-12 Plant, the Oak Ridge Reservation, Tennessee.
USGS. 1985. Open-File Report 85-553, Water-quality Data for 35 Sites, September 1984, Near the Y-12 Plant, the Oak Ridge Reservation, Tennessee.
USGS. 1986. Open-File Report 85-68, Streamflow and Specific-conductance Data for Bear Creek, August 13, 1985, the Oak Ridge Reservation, Tennessee.
USGS. 1986. Water-Resources Investigations Report 86-4165, Reconnaissance of Surficial Geology, Regolith Thickness, and Configuration of the Bedrock Surface in Bear Creek and Union Valleys, near Oak Ridge, TN.
USGS. 1988. Water-Resources Investigations Report 88-4010, Preliminary Evaluation of Ground-water Flow in Bear Creek Valley, the Oak Ridge Reservation, Tennessee.
USGS. 1988. Water-Resources Investigations Report 88-4068, Well Construction, Lithology, and Geophysical Logs for Boreholes in Bear Creek Valley Near Oak Ridge, Tennessee.
USDI
USDI. 1984. Open-File Report 84-625, Streamflow and Specific-conductance Data for Selected Sites, February 15 through April 9, 1984, Near the Y-12 Plant, the Oak Ridge Reservation, Tennessee.

Systems included in the evaluation were EFPC, Bear Creek, Poplar Creek, WOC, the Clinch River including Melton Hill Reservoir, and the Tennessee River, including Watts Bar Reservoir.

Samples were analyzed to determine the presence of metals, radionuclides, and organics. The Instream Contaminant Study consisted of five tasks:

- Task 1: Water quantity and quality data for predicting sediment transport
- Task 2: Sediment volume and contaminant characterization
- Task 3: Transport and fate of sediment in EFPC
- Task 4: Contaminant concentrations in fish
- Task 5: Summary of the previous reports and the management implications of the results

These tasks are described in greater detail in Section 5.3 according to the media investigated.

#### Environmental Monitoring and Surveillance of the Oak Ridge Community

Following the discovery of mercury contamination of EFPC, the DOE in 1983 requested Oak Ridge Associated Universities (ORAU) to assist in monitoring of the Oak Ridge community. A program of environmental monitoring and surveillance was initiated in response to citizens' requests for an investigation of soils, sediments, vegetables, and well water for mercury contamination. Areas included in the study were the EFPC floodplain, the City of Oak Ridge sewerline beltway, and private properties where floodplain soils may have been used as fill. In addition to mercury, analyses were conducted for radionuclides, other metals, and PCBs.

Sampling data from the ORAU studies were reported monthly to DOE and distributed to federal, state, and local government agencies. Data are summarized in the annual environmental monitoring reports (MMES, 1984; 1985; 1986; 1987; 1988). Raw data are available (TDHE, 1983; Hibbitts, 1984; Hibbitts, 1986). The program was terminated in September 1987.

#### The TDHE and CDC Biomonitoring Study for Mercury

In June through July 1984, the TDHE and Centers for Disease Control (CDC) conducted a pilot study to document human body levels of mercury, to determine whether exposure to mercury-contaminated soils or consumption of fish presumed to be contaminated with mercury constituted an immediate health risk to the Oak Ridge population (Rowley et al., 1985). The study evaluated exposure histories of 2,627 residents and city workers to mercury-contaminated soil and/or fish. Mercury concentrations in urine and hair were measured for representative subsamples of the population with high and low levels of exposure. Urine and hair mercury concentrations were reportedly not at levels associated with known health risks.

### NPDES Biological Monitoring and Abatement Programs

National Pollution Discharge Elimination System (NPDES) permits were originally issued in 1974, and were reissued to each of the three Oak Ridge facilities beginning in 1985. The reissued permits required implementation of Biological Monitoring and Abatement Programs (BMAPs) at each facility. The BMAP for each facility was developed by ORNL's Environmental Sciences Division (ESD). The BMAPs establish a system of biological monitoring designed to demonstrate that the interim effluent limits established for each facility protect the classified uses of the receiving stream (for example, growth and propagation of fish and aquatic life). Information gathered under the BMAPs is used to characterize the ecological impacts of operations (present and past) and to evaluate the potential ecological consequences of various alternatives for remedial action (developed under RCRA, CERCLA, etc.), including construction and operation of major new pollution abatement facilities.

Each BMAP encompasses four major tasks: 1) ambient toxicity testing; 2) bioaccumulation of nonradiological contaminants in aquatic biota studies; 3) biological indicator studies (including measurement of selected biochemical parameters and histopathological analyses); and 4) benthic invertebrate and fish community surveys, instream ecological monitoring, assessment of contaminants in terrestrial environment, and assessment of contaminant, transport, distribution, and fate (Loar et al., 1992).

Data collected in the BMAPs are summarized in annual reports (Smith et al., 1988; Adams et al., 1992). In addition, a computerized database which allows for retrieval of records and data from all NPDES Permit sampling and monitoring activities is maintained on the Environmental Monitoring and Compliance Section's VAX computer (Taylor, 1990). The system permits tracking of all sampling sites and includes date and time of collection, identity of individuals collecting the sample, and a description of how and under what conditions each sample was collected. Analytical data are transferred to the database from entries verified by the laboratory supervisor in the Analytical Chemistry Division's computer.

### The Clinch River Remedial Investigation

In 1989, the Clinch River Remedial Investigation (CRRI) was initiated by the DOE to address the transport, fate, and distribution of waterborne contaminants released from the ORR to the Clinch and Tennessee Rivers and to gather information to evaluate the potential risks to human health and the environment associated with these contaminants (Cook et al., 1992). Phase 1 of the investigation involved preliminary sampling and analysis of water, sediment, and aquatic organisms in 10 reaches, six of which are potentially affected by releases from the ORR, and four which serve as reference, or background, areas. Areas evaluated included Melton Hill Reservoir (CRM 23.1 to 52.0), Clinch River from Melton Hill Dam to its confluence with the

Tennessee River (CRM 0.0 to 23.0), Poplar Creek (PCM 0.0 to 6.8), and Watts Bar Reservoir (TRM 530.0 to 567.5).

Data collected during Phase I were analyzed using screening human health and ecological risk assessment methodologies to identify contaminants, exposure pathways, and river and reservoir reaches of concern. Results of these analyses will be used to help guide the design of the CRR Phase 2 sampling plan.

### 5.3 SUMMARIES OF STUDIES OF INTEREST

Available environmental monitoring and research data describing the historical presence and behavior of contaminants in off-site areas of the ORR are summarized below. Media addressed include surface water, sediment, ambient air, aquatic and terrestrial foodstuffs, soil, drinking water, and groundwater. In addition, the availability of hydrologic data and meteorologic data is discussed following the discussions of surface water monitoring and air monitoring, respectively. Study numbers provided in this section correspond with the Section 5.1 summary tables. Tables and figures excerpted from some of these studies are included in Appendices A and B. These tables and figures have been included to show the types of data which have been collected in some of the major studies. However, since these tables and figures were excerpted directly from the cited reports, the quality of data presented has not been verified.

#### 5.3.1 Historical Surface Water/ Sediment Data

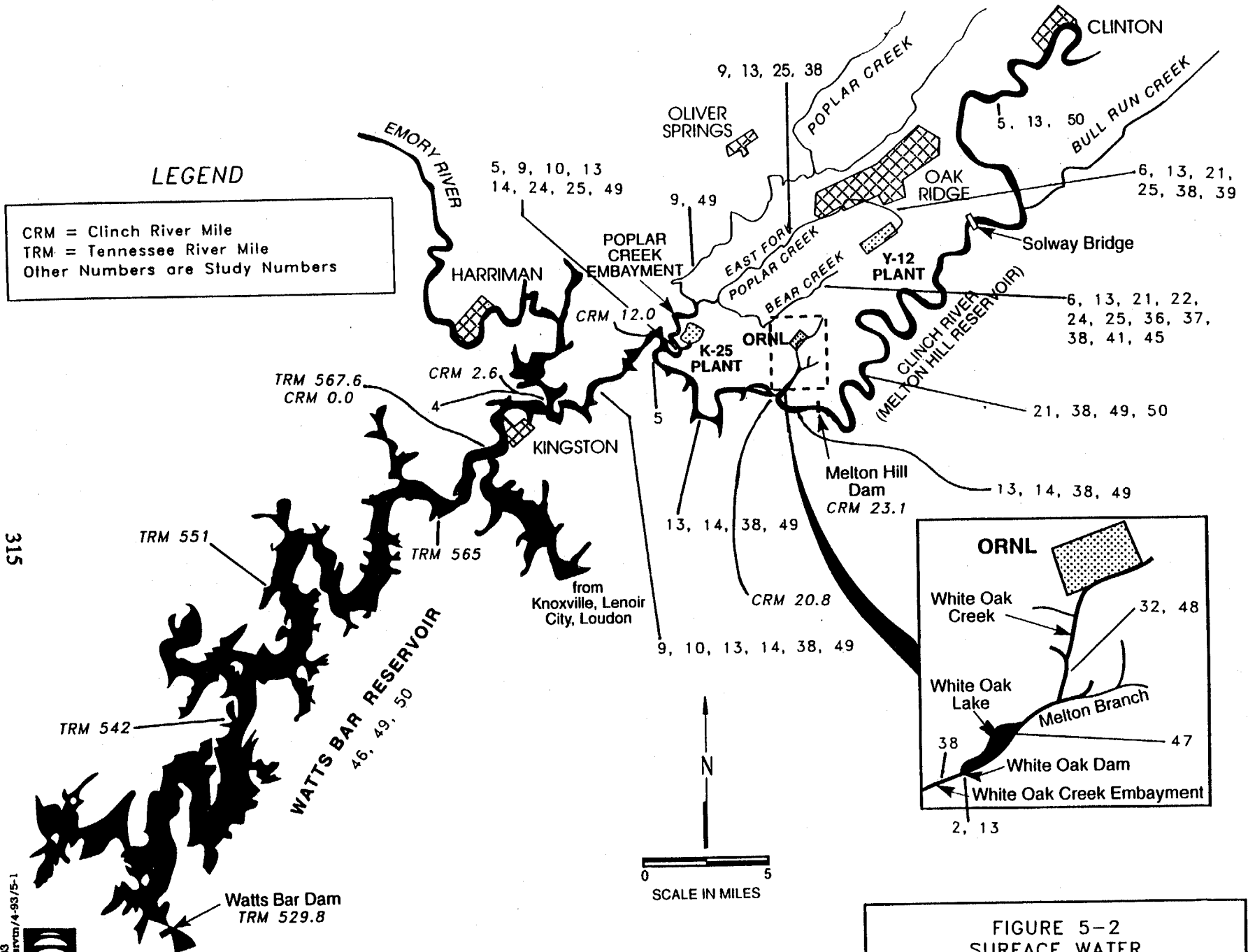
Available surface water and sediment data collected for the areas of concern identified in Section 5.1 are described below. Summaries of these studies were presented earlier in Tables 5-1 and 5-2, respectively. Locations of the studies described are depicted in Figures 5-2 and 5-3. Examples of reported data excerpted from the original reports are presented in Appendices A and B.

##### Study 1: Radioactivity in WOC/WOL Surface Mud (1944-1946)

The earliest located data on radioactivity in sediments of surface water systems surrounding the ORR were reported by Cheka and Morgan (1947). Measurements were made by the ORNL Health Physics Division from 1944 to 1946 at locations above, in, and below WOL, including three locations in the Clinch River near the mouth of WOC. Samples were collected to evaluate the effect of changes in plant processes on the total activity and distribution of radioisotopes in the basin. Surface mud samples of the top 0.5 cm of sediment from the creek bed, lake bottom, and mud banks were measured for gross beta. In addition, selected samples were analyzed for Ba, Sr, and Cs. The report summarizes samples collected by other researchers as well, including measurements of total radioactivity in sediments by L.H. Weeks in March and April, 1945 and by H.R. Crafts in October, 1946, and chemical analyses by Overstreet and Jacobson

**LEGEND**

CRM = Clinch River Mile  
 TRM = Tennessee River Mile  
 Other Numbers are Study Numbers



**FIGURE 5-2  
 SURFACE WATER  
 STUDY LOCATIONS**

315



### LEGEND

CRM = Clinch River Mile  
 TRM = Tennessee River Mile  
 Other Numbers are Study Numbers

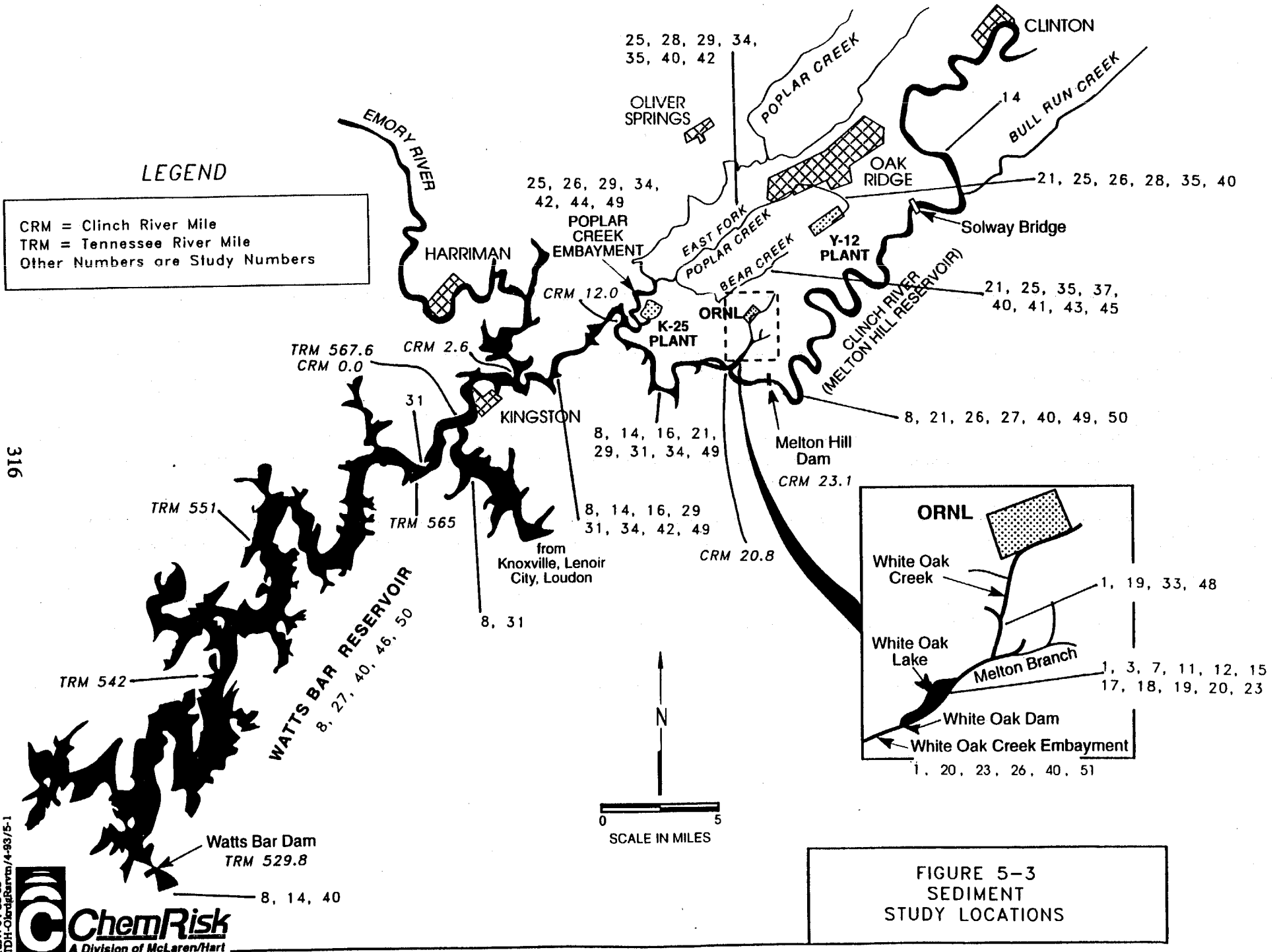


FIGURE 5-3  
 SEDIMENT  
 STUDY LOCATIONS

in April, 1944, J.G. Hamilton in June, 1944, D.M. Black in April and August, 1945, and J.E. Hudgens in February, 1946. Average measurements of total activity in the White Oak drainage system in 1945-46, as reported by Cheka and Morgan (1947), are presented in Table A-1 (Appendix A).

#### Study 2: Radioactivity in WOL Discharge Water (1944-1946)

Data on the average distribution of fission products in the water leaving WOL from September 1944 to February 1945 were compiled by Morgan and Western (1947). The principal long life fission products were chemically separated and measured for the beta activities of the separated elements. In addition, water leaving WOL during unusual conditions, including periods of flooding and elevated activity, was collected for analysis during September 1944, January 1946, and March 1946. Data are tabulated in the report.

#### Study 3: Radioactivity in WOL Sediments (1945-1952)

Annual surveys of the radioactivity content of WOL sediments were conducted by ORNL beginning in 1945. The results of the 1950, 1951, and 1952 surveys are reported by Abee (1953). Samples were collected at approximately 100-foot intervals along transects of the lake using a hollow metal pipe. Radioactivity was measured in the top 3 inches of the sediment. Additionally, a stratification sample was collected at a random point along each transect for the measurement of activity below the 3-inch depth. Activity in these samples was measured at 1-inch intervals. Radioactivity data for each sample collected during 1950, 1951, and 1952 are presented and compared with data on the total radioactive content of WOL for 1945, 1946, and 1948.

#### Study 4: Radioactivity in Clinch River Water (1947)

A radioactivity survey of the Clinch and Emory Rivers within a radius of several miles of the town of Harriman was conducted by TVA and the ORNL Health Physics Division in June 1947 (Morgan, 1947). Background samples were collected upstream on the Clinch River in the vicinity of Norris Dam. Mud samples and 5-gallon water samples were collected. Each water sample was evaporated to 1 liter, filtered, and evaporated to dryness. Activity was measured in the filtered silt, evaporated filtrate, and mud. Results are not tabulated; however, average values are discussed.

#### Study 5: Radioactivity in Poplar Creek and Clinch River Water (1947-1948)

Water samples from Poplar Creek and the Clinch River were collected by K-25 plant personnel beginning in late October 1947 (Ketcham, 1947a; Ketcham, 1947b; Visner, 1947; Visner,

1948a; Visner, 1948b). These data are presented in monthly progress reports of Health Physics activities at the K-25 Plant and in monthly reports of Industrial Hygiene Laboratory analyses. Samples were collected to monitor drinking, drainage, and river water for possible contamination by fluorine, uranium, and gross alpha and beta radioactivity. The program covered "all possible sources of contamination on the K-25 Area," including in the Clinch River below the S-50 Sewer Outfall and several discharge points into Poplar Creek. Samples were also collected in the Clinch River at two points upstream of the ORR facilities: west of the Jones Scrap Yard, and one-half mile above Elza Gate. Sample collection consisted of two daily samples, 12 weekly samples, and 13 monthly samples. These reports present data for October through December 1947, and October through December 1948. Survey results for samples collected at the "Clinch River, effluent from K-25" in October and November 1947 are shown in Appendix A (Table A-2).

#### Study 6: Radioactivity and Mercury in EFPC and Bear Creek Water (early 1950s-present)

Composite water samples from EFPC and Bear Creek have been collected for measurement of gross alpha and gross beta radioactivity since the early 1950s (UCC, 1983). Samples from EFPC were also analyzed for mercury. Samples were collected by the Y-12 Health Physics Department to monitor product losses. Until 1977, all mercury analyses were done on unpreserved samples. Since 1977, grab samples for mercury have been collected and preserved by acidification in the laboratory, and since 1982, grab samples for mercury have been acid-preserved in the field at the time of sample collection. Data are reported in Y-12 Health Physics Department monthly reports. Exact locations of sample collection were not identified.

#### Study 7: Radionuclides in WOC/WOL Sediments (1950-1953)

Samples of sediment and vegetation in the WOC drainage basin were collected from June 1950 through June 1953 as part of a cooperative ecological study conducted by the TVA and the Atomic Energy Commission (Krumholz, 1954a; 1954b; 1954c, 1954d). The program was conducted to investigate the physical and biological effects of radioactive materials and wastes in the environment surrounding the ORR. Early in the course of the investigation, extensive construction was underway on the ORNL site. As a result, a considerable amount of silt was carried into WOL such that the waters of WOC were almost continuously muddy. Following seeding of the denuded areas, the waters cleared toward the latter part of the study. Samples were collected from the top three inches of sediment at five stations on the north bank of WOL. Since previous surveys had focused only on radioactivity present in the lake bottom, all five stations were established within the zone of water level fluctuation of the lake bank. Sediment samples were analyzed by direct radioassay for gross beta. To determine the accumulation of fission products by vegetation near WOC, vegetation samples, including willows, ragweed, sweet clover, and elm, were collected and analyzed by direct radioassay and radiochemical

analyses for determination of gross beta and specific radionuclides, including strontium, cesium, ruthenium, rare earths, and zirconium. Data are presented in the reports for individual samples.

#### Study 8: Radionuclides in Clinch and Tennessee River Sediments (1951-1966)

Annual sediment surveys were conducted in the Clinch and Tennessee Rivers beginning in 1951. The results of the 1951-1953 surveys were reported by Garner and Kochtitzky (1956). From 1954 to 1958, the surveys were conducted by the Area Monitoring Group of the Health Physics Division; these data were reported by Cottrell (1960). Beginning in 1959, samples were collected by the ORNL Applied Health Physics Division and reported in the Applied Health Physics Annual Reports. Annual sediment surveys of the Clinch River by the Applied Health Physics Division were discontinued following 1966. Results of the 1959-1966 surveys are summarized in the ORNL Applied Health Physics Annual Reports.

Sample collection locations varied somewhat each year; however, in general, data were collected on the Clinch River from CRM 27.5, above the mouth of WOC, to the confluence of the Clinch River with the Tennessee River (TRM 567.6), and on the Tennessee River from TRM 570.8, upstream of the mouth of the Clinch River, to TRM 475.1, just north of Chattanooga. During the 1951, 1957, and 1958 surveys, measurements extended as far downstream as Guntersville Reservoir, at approximately TRM 354.4. In 1952, measurements were taken along the extent of the Tennessee River from Oak Ridge to the Tennessee River mouth, and in the Ohio River above and below the mouth of the Tennessee River. In addition, in 1952 and in 1954 through 1958, measurements were taken at background locations in several reservoirs on tributaries to the Tennessee River, including Norris, Fontana, and Hiwassee Reservoirs. In 1966, the sediment sampling program extended downstream through the Kentucky Reservoir.

Surface sediment gamma radiation measurements were taken on river cross sections at predetermined intervals using a submersible Geiger-Mueller counting system called a "flounder". From 1954 on, a composite silt sample was collected on each cross section and counted for gross beta activity and analyzed for radionuclides, including cerium-137, strontium-90, cerium-144, trivalent rare earths, ruthenium-106, and cobalt-60. Data are tabulated in the reports by radionuclide, sample location, and sample date. Plots showing the average reported surface sediment gamma counts measured in 1954-1958 in the Clinch River downstream of the confluence with WOC and in the Tennessee River downstream of the confluence with the Clinch River are shown in Appendix B (Figures B-1 and B-2, respectively).

Tables A-3 through A-6 in Appendix A presents summaries of radioactivity measurements in silt from the Clinch and Tennessee Rivers over the period from 1959 through 1965. These tables were excerpted from the Applied Health Physics Annual Reports. Values are presented for approximately eleven locations in each river, with Fort Loudoun Lake silt data presented in some

cases as an indicator of background levels. Figure B-3 (Appendix B) presents a summary of gamma radiation levels measured at the surface of Clinch River bottom silt as a function of downstream distance for 1952, 1960, and 1961. Figure B-4 reflects average gamma count rates at the surfaces of Clinch and Tennessee River silts over the period from 1951 through 1963.

Study 9: Mercury and Manganese in Clinch River, EFPC, and Poplar Creek Water (1955)

A survey of mercury and manganese levels in Clinch River, EFPC, and Poplar Creek water was conducted for a period of nine days in July 1955 (Kwasnoski, 1955). The survey was conducted by K-25 staff to determine trends and daily variations in concentrations. Samples were collected at seven locations in Poplar Creek. These locations were above and below the confluence of Poplar Creek with EFPC and at effluent release points on the K-25 site. Samples were also collected at one location in EFPC and one location in the Clinch River below the Poplar Creek junction. Exact sample locations are not given. Raw data are presented in the report.

Study 10: Mercury in Clinch River and Poplar Creek Water (1955-1957)

Following the above study of mercury and manganese in the Clinch River, EFPC, and Poplar Creek (Study 9), surface water samples were collected semi-weekly near the K-25 Site by K-25 Staff and analyzed for mercury (Kwasnoski and Whitson, 1955, 1956, 1957). Monthly data are reported by the K-25 Technical Division for October 1955 through September 1957. Samples were collected in Poplar Creek at its junction with EFPC, at the K-1513 pumphouse, at the point of entry of effluent from the water purification plant, and at the source of K-891 supply water. Samples were also collected in the Clinch River one mile downstream of the confluence with Poplar Creek.

Study 11: Radionuclides in WOL Bed Soils (1956-1958)

Six-inch core samples of lake bed soils from the upper and lower beds of the drained WOL were collected in 1956, 1957, and 1958 by the ORNL Health Physics Division and analyzed for radionuclides, including total strontium-90, cesium-137, cobalt-60, cerium-144/praseodymium-144, ruthenium-106/rhodium-106, and trivalent rare earths (Auerbach et al., 1958; 1959). To determine the inventory of strontium-90 contained in lake bed soil, 68 soil samples were collected and analyzed for strontium-90 during this time period. Changes in the strontium-90 content of the lake bed over the 3-year period were calculated. Data are discussed and summarized in the Applied Health Physics Annual Progress Reports for the periods ending July 31, 1958 and July 31, 1959.

Study 12: Radioation Above WOL Bed (1959)

An evaluation of the radiation field above the drained WOL was conducted in 1959 by the ORNL Health Physics Division (Lee and Auerbach, 1959). Measurements of the horizontal and vertical distribution of air dose rates over the bed were collected to be used in investigations of the effects of chronic low-level radiation upon mammals and other organisms inhabiting the area. The radiation was predominately gamma (90%). A significant difference in the exposure rate over the upper and lower lake bed was revealed: regions near the inlet end of the lake bed were associated with higher exposure rates (50 mR/hr) while regions along the shore line were associated with lower exposure rates (10 mR/hr).

Study 13: Radionuclides and Metals in WOC, Clinch River, Poplar Creek, East Fork Poplar Creek, and Bear Creek Water (1944-Present)

Surface water samples were routinely collected by the ORNL Applied Health Physics Division to evaluate the gross concentration of radioactivity entering the Clinch River as a result of discharges of low-level radioactive liquid wastes from ORNL operations to WOC. Radiological analyses of waters discharged over WOD have been reportedly made since at least 1944 (Ohnesorge, 1986). Data from 1944-1948 are for gross-beta activity only; data are not available for individual radionuclides until 1949. Individual radionuclides included in the analyses at WOD were cesium-137, ruthenium-106, strontium-89, strontium-90, cerium-144, zirconium-95, niobium-95, iodine-131, cobalt-60, tritium, and total rare earths minus cerium. The earliest data located as part of Project Task 2 for waters discharged over WOD were gross beta and plutonium measurements reported in the Applied Health Physics Quarterly Report for the second quarter of 1954 (ORNL, 1954). These data together with measurements of flow in the Clinch River were used to estimate radionuclide concentrations in the Clinch River at its the confluence with WOCE (CRM 20.8).

Beginning in at least 1959, water samples were also collected in the Clinch River at a number of locations ranging from above Melton Hill Dam to just upstream of its confluence with the Tennessee River. Samples were collected at several water intake locations on the Clinch River. Water samples were collected at the Kingston Water Plant at CRM 4.5 beginning in at least 1959. Beginning in 1962, samples were collected at the City of Oak Ridge water intake at CRM 41.5 and beginning in at least 1971, samples were collected at the K-25 Site water intake at CRM 14.5. Water samples were analyzed for radionuclides, including strontium-90, cerium-144, cesium-137, ruthenium-103/106, cobalt-60, and zirconium-95/niobium-95. These data are summarized in the Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports. Table A-8 (Appendix A) presents water monitoring results for 1965, as presented in the Applied Health Physics Annual Report for that year. Table A-9, excerpted from Ohnesorge (1986) summarizes water monitoring results at the Kingston Water Plant (CRM 4.5) from 1961-1984.

Beginning in at least 1971, water samples were also collected at five locations and analyzed for water quality parameters and metals including uranium, cadmium, chromium, mercury, lead, and zinc. Sample collection points were located on Poplar Creek upstream and downstream of the K-25 Site, the Clinch River downstream of its confluence with Poplar Creek, EFPC at the outlet of New Hope Pond, and Bear Creek. Samples were collected weekly until 1976 and continuously thereafter. These data are summarized in the reservation-wide annual environmental monitoring reports. Table A-10 (Appendix A) summarizes the data for EFPC for 1972.

Beginning in at least 1985, water samples were collected from EFPC near its confluence with Poplar Creek and in Bear Creek, and analyzed for gross alpha and gross beta radioactivities, thorium, and uranium-235. Analyses in Bear Creek also included neptunium-237, technetium-99, and plutonium-239/240. Beginning in at least 1987, samples were collected in EFPC at the outlet of New Hope Pond and analyzed for specific radionuclides, including americium-241, cesium-137, cobalt-60, neptunium-237, niobium-95, plutonium-238, plutonium-239/240, radium-226, ruthenium-106, strontium-90, technetium-99, thorium-228, thorium-230, thorium-232, uranium-234, uranium-235, uranium-238, zirconium-95, and tritium. These data are summarized in the reservation-wide annual environmental monitoring reports.

#### Study 14: Radionuclides and Chemicals in WOC, Clinch River, and Tennessee River Water and Clinch River and Poplar Creek Sediments (1960-1964)

Water samples were collected, as part of the Clinch River Study, beginning in November 1960 (Struxness et al., 1967). Samples were collected from seven locations: one on WOC at WOD, three on the Clinch River at CRM 5.5, 14.4, and 41.5, and three on the Tennessee River, at Chickamauga Dam (TRM 471.0), Watts Bar Dam (TRM 529.9), and TRM 591.4. Two of the seven stations served as background locations since they were located upstream of the sources of contamination: Station 1 was located on the Clinch River upstream of WOC (CRM 41.5) and Station 4 was located on the Tennessee River upstream of the Clinch River (TRM 591.4). Samples were collected from each station at least once daily through November 1962. Samples were collected in volumes proportional to the river flow, composited, and analyzed weekly or monthly for radionuclides (strontium-90, ruthenium-106, cobalt-60, and cesium-137) and stable chemicals. Supplemental sampling was also conducted in November 1962 and November 1963 at Station 2 (WOD) for major, minor, and trace-element constituents. Radionuclide water concentrations measured during this study are summarized in Table A-11 (Appendix A).

Samples of bottom sediment from the Clinch River were also collected as part of the Clinch River Study (Carrigan et al., 1967; Carrigan and Pickering, 1967; Struxness et al., 1967). Two-foot core samples were collected during the summers of 1960 through 1964 between CRM 1.2 and CRM 22.9. During 1962, additional samples were collected at two locations in the Emory River and two locations in Poplar Creek. Sample locations and numbers of samples

collected varied from year to year. Samples were analyzed for organics, radioactivity, and specific radionuclides (strontium-90, ruthenium-106, cobalt-60, and cesium-137). Core samples were collected to determine the longitudinal and vertical distribution of radioactivity in the sediment. In addition, grab samples were collected to determine the potential for radionuclides to desorb from the sediment. Sample results are presented in these reports in summary tables and graphs.

Study 15: Radioactivity in WOL Bed Sediments (1961)

During April and May, 1961, the vertical and lateral distribution of radioactivity in sediments of WOL was evaluated by the ORNL Health Physics Division (Lomenick et al., 1961). A total of 15 six-foot deep sediment cores were collected along three silt ranges at the upper, middle, and lower end of the lake. For the first six-inches of each core, each inch was analyzed; at greater depths, larger increments were used. Samples were analyzed for gross gamma activity. Data are tabulated in the report by sample location and core depth.

Study 16: Radionuclides in Clinch River Sediments (1962)

An inventory of radioactive materials in the bottom sediment of a 21-mile stretch of the Clinch River was performed during July 1962 (Carrigan and Pickering, 1967). A total of 95% of the radionuclides in sediment were determined to be in the section of channel between CRM 0 and CRM 15. Data are not presented in the report by Carrigan and Pickering (1967) and have not been located.

Study 17: Ruthenium in WOL Bed Sediments (1962)

WOL sediments were sampled by the ORNL Health Physics Division in 1962 and analyzed for ruthenium-106 (Lomenick et al., 1962). Cores varied in depth from 24 to 60 inches and were collected approximately 50 feet apart along transects at right angles to the surface flow of waste over the lake bed. Results indicate that the highest concentrations of ruthenium-106 were found in the upper two inches of soil, and approximately 75% of the ruthenium-106 was found in the first two feet of sediment. Further, although transported through the lake bed soil by groundwater, it was determined that very little ruthenium-106 reached WOC by this route. Data are not presented in the report. A cross section of WOL showing ruthenium concentrations is presented.

Study 18: Radionuclides in WOL Bed Sediments (1963)

To determine the quantity, type, and distribution of radionuclides in the WOL bed and to aid in defining the migration of these products, 250 two-foot deep sediment core samples were



collected in the lake bed by the ORNL Health Physics Division during 1963 and analyzed for radionuclide content (Lomenick et al., 1963). Samples were segmented into six-inch increments and scanned using a Packard automatic gamma counting system, which consists of a spectrometer with well-type scintillation detector, and analyzed for ruthenium-106, cesium-137, and cobalt-60 using a IBM 7090 computer program for stripping the gamma spectrum. Standard radiochemical procedures were used for strontium and total-rare-earth analyses. The total quantity and distribution of radionuclides in the lake bed is tabulated by depth below the surface. The results show that ruthenium-106 and cesium-137 account for more than 90% of the total activity, and cobalt-60, the rare earths exclusive of any yttrium-90 contamination, and strontium-90 comprise the remainder. Most of the activity was determined to occur in the upper six-inches of soil.

#### Study 19: Radionuclides in WOC/ WOL Sediments (1964)

During 1964, samples of the upper 0.2 feet of sediment were collected by the ORNL Health Physics Division at ten locations in the WOC basin above and below ORNL at WOCM 1.12 to WOCM 2.68 (McMaster and Richardson, 1964). Samples were collected to determine the distribution of ruthenium-106, cesium-137, and cobalt-60 in the silt and smaller size fractions at various locations in the drainage basin. Analytical methods are not discussed. Data are presented in the report in a bar graph of radionuclide concentrations for each sampling station.

#### Study 20: Radionuclides in WOC/ WOL Sediments (1970)

In 1970, sediment samples were collected to test the tentative conclusion presented by Pickering et al. (1966) that most cesium-137 is associated with clays and that a significant portion of strontium-90 is associated with limestone (Tamura et al., 1970). One sample was collected from the mouth of WOC where it enters the Clinch River, and one sample was collected from WOL. Samples were analyzed for strontium-90 and cesium-137. No description of sampling methods is provided, although analytical techniques are discussed. Results are tabulated. cesium-137 was found primarily in the clay mineral band, while strontium-90 was found in most bands, including bands containing calcium carbonate.

#### Study 21: Mercury in EFPC, Bear Creek, and Melton Hill Reservoir Water and Sediments (1970)

In 1970, a survey was initiated by M. Sanders, the Y-12 Environmental Coordinator, to determine the mercury content in water, sediment, and fish samples from various parts of the Oak Ridge area. Results were reported to J.D. McLendon in an internal memorandum dated August 6, 1970. This memorandum was not located; however, the results are summarized in UCC (1983). A total of 12 water samples and 10 mud samples from New Hope Pond, EFPC, Bear Creek, and Melton Hill Reservoir were collected and analyzed for mercury. Exact sample locations were not given.

Study 22: Radionuclides and PCBs in Bear Creek Water (1971-present)

A monitoring station was established at BCM 2.83 in 1971 (Turner et al., 1988). This station became an NPDES monitoring station in 1974. Data are collected at this station by Y-12 Plant Staff to monitor stream discharge and selected constituents. Additional contaminants not on the NPDES permit, including radionuclides and PCBs, have been analyzed from this location as part of Y-12 Plant radiological and PCB monitoring plans. Following signing of the Memorandum of Understanding (MOU) on May 26, 1983 by DOE, EPA, and TDHE, an additional monitoring station was established at BCM 7.74 in July 1983. This location is monitored weekly for water quality parameters.

Study 23: Radionuclides in WOC/WOL Sediments (1972)

An update of the 1965 and 1970 assessments of bottom sediments of WOL and WOC by the Environmental Sciences Division was conducted in 1972 (Blaylock et al., 1972a). Surface sediment and core samples were analyzed for radionuclide concentrations. An attempt was made to include the sampling locations used in the previous two assessments. As in the earlier studies, cesium-137 was the most abundant radionuclide, accounting for 62 to 85% of the total gamma at most sites. However, while in the previous evaluations the majority of the activity was associated with the top 15-cm (6-in) of sediment, in this study the majority of activity was between 16- and 34-cm (6.3- and 13-in). This change was attributed to a decrease in yearly discharges of radionuclides to WOL. Additionally, ruthenium concentrations were reported to be considerably lower due to radioactive decay. Sampling and analytical methods are not discussed. Raw data are tabulated by location.

Study 24: Nitrates and Uranium in Bear Creek and Poplar Creek Water (1972)

From June through August 1972, water samples were collected from Poplar Creek near the Building 9720-5 storm sewer outfall and from Bear Creek near the S-3 disposal ponds as part of a Y-12 Development Division project (Francke, 1972). Samples were analyzed for nitrates and soluble uranium. Nitrate and uranium concentrations are compared to rainfall totals. Raw data are tabulated. The letter reports describe the experimental application of raw water from upper Bear Creek to 12 plots of hardwood forest land located on a ridge on the north side of Bear Creek Road. The experiment was conducted to evaluate the effect of nitrates on forest ecosystems.

Study 25: Mercury in Bear Creek, EFPC, and Poplar Creek Sediment (1972-1974)

Preliminary surveys of water and sediment in EFPC, Poplar Creek, and Bear Creek were conducted during 1972, 1973, and 1974 (Reece, 1974). The surveys were conducted to identify

possible areas of concern and to determine the continuance or abatement of problems. Water samples were not analyzed for mercury; however, it is not known what other contaminants were evaluated. Sediment samples showed mercury levels ranging from less than 0.05 ppm to 72 ppm. This report was not located. Results are discussed in UCC (1983).

Study 26: PCBs in Clinch River, EFPC, Poplar Creek, and WOC Sediments (1973-1974)

A special sampling program of bottom sediments in selected streams and ponds on the ORR was conducted in 1973 and 1974, to determine the baseline concentration of PCBs (Jordan, 1978). Samples locations included the Clinch River near Grubb Island, Melton Hill Reservoir, New Hope Pond, EFPC from below the New Hope Pond outfall to near Blair Road, Poplar Creek at K-25 Site effluent discharge points, K-25 Site lagoons and ponds, WOCE, and the Scarboro Embayment. The program was conducted to comply with recommendations made during the 1973 Environmental Management Appraisals of UCC-ND-operated facilities. Sample results are tabulated in the report by location, and are presented in Appendix A of this report (Table A-12).

Study 27: Metals and PCBs in Watts Bar and Melton Hill Reservoir Sediments (1973 and 1982)

During 1973 and again during 1982, sediment samples were collected by the TVA from mainstream and tributary reservoirs of the Tennessee River, including Watts Bar and Melton Hill Reservoirs, to determine the accumulation of trace contaminants in sediments in the reservoirs (TVA, 1986a). In 1973, samples were analyzed for metals and water quality parameters. In 1982, samples were also analyzed for PCBs. Samples were collected in Watts Bar Reservoir on August 1, 1973 and June 8, 1982, and in Melton Hill Reservoir on June 7, 1973 and June 29, 1982. During the latter study, approximately nine sediment cores were collected from each site. The top 3 centimeters (1.2 in) of each core were removed and composited. Results are tabulated by location, date of collection, and analytical parameter.

Study 28: Mercury in EFPC Sediments and Poplar Creek Fish (1974-1975)

From August 1974 through March 1975, the ORNL Environmental Sciences Division conducted a special short-term aquatic surveillance study to supplement available data used to describe the aquatic systems on the ORR identified as possible areas of impact from the Oak Ridge facilities (ERDA, 1975). The study involved analysis of sediment from EFPC for mercury. Fish from Poplar Creek were also analyzed. These data were used along with routine monitoring data to prepare the "Preliminary Draft Environmental Analysis, Oak Ridge Operations" (ERDA, 1975).

Study 29: Metals in Clinch River, EFPC, and Poplar Creek Sediments (1975-present)

In 1975, a sediment sampling program was initiated by the ORNL Applied Health Physics Division to determine concentrations of metallic ions in the sediment of Poplar Creek. Samples

were collected twice a year at locations on Poplar Creek ranging from above plant discharge points downstream to the Clinch River. Locations and numbers of samples varied. Samples were analyzed for uranium, mercury, lead, cadmium, copper, nickel, chromium, zinc, manganese, aluminum, and thorium. The sampling program was subsequently expanded and by 1977 included two additional sampling locations on the Clinch River and one on EFPC. Raw data for 1975 to 1981 have been compiled (ORGDP, 1981). Data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide environmental monitoring reports. The sediment sampling locations for 1978 are shown in Figure 5-4. Sampling results for that year are shown in Table A-13 (Appendix A).

#### Study 30: Metals in Clinch River Water (1975-1978)

An assessment of nonradiological water quality parameters and nonfisheries biological communities in the Clinch River prior to construction of the Clinch River Breeder Reactor Plant was conducted by TVA for the period March 1975 through October 1978 (TVA, 1979). Surface water samples were collected at three stations in the Clinch River in the vicinity of the proposed plant and at Melton Hill Dam, and analyzed for metals and other water quality parameters. Results are tabulated in the report by location, date of sample collection, and analyte. These data are also available on the USEPA STORET system. In addition, groundwater samples from seven wells near the site were collected and analyzed for metals.

#### Study 31: Radionuclides in Clinch and Tennessee River Sediments (1977)

From June to November, 1977, sediment cores were collected to evaluate changes in the radionuclide distribution patterns since the completion of the Clinch River Study in 1964 and to evaluate the effect of the alteration of the Clinch River flow regime following construction of Melton Hill Dam in 1963 (Oakes et al., 1982b). When the Clinch River Study was initiated in 1960, the Clinch River had unidirectional flow. However, due to the interactive effects between Melton Hill Dam and Watts Bar Dam about 61 river miles downstream, the flow pattern has been estuarine since 1963. The study was also conducted to establish baseline levels of contamination in the Clinch River in the vicinity of the proposed Clinch River Breeder Reactor (CRBR) prior to its construction and operation.

Core samples were collected by a private consultant under contract to ORNL using a Swedish foil sampler. Cores were collected along the full length of the Clinch River from the WOC outfall (CRM 20.8) to locations in the Tennessee River on either side of the mouth of the Clinch River, with emphasis on locations near the proposed Clinch River Breeder Reactor (CRBR) site (CRM 14.6 to 18.6) and the WOC outfall. Sample locations were selected to give a representative distribution sufficient to determine the spatial distribution of radioactivity in the bottom sediment of the river. Samples were analyzed for cesium-137 and cobalt-60 by gamma

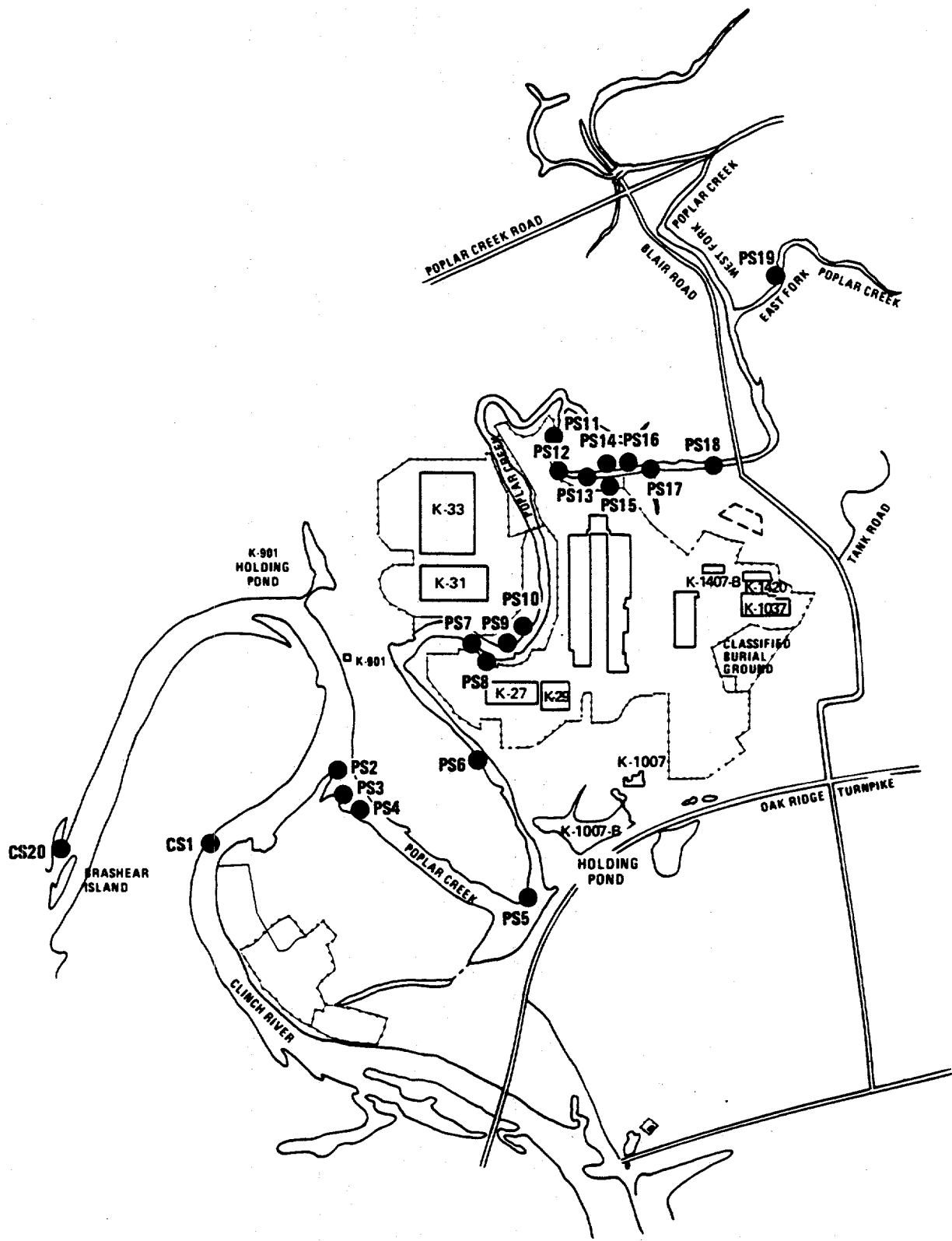


Figure 5-4

OAK RIDGE GASEOUS DIFFUSION PLANT SEDIMENT SAMPLING LOCATIONS

spectroscopy and for strontium-90, plutonium-239, plutonium-240, americium-241, and curium-244 by specific radiochemical determination. Prior to collection of the core samples, an attempt was made to gain an *in situ* estimate of the relative gamma-beta activity at various locations on the surface of the bottom sediments using a flounder. However, this procedure did not prove useful since radioactivity measurements of interest in the sediment could not be distinguished from background radioactivity. Sample results are tabulated in the report.

#### Study 32: Radionuclides in WOC Water (1977)

Water samples were collected in WOC to investigate sources of strontium-90 that contribute significantly to instream concentrations in WOC between ORNL Monitoring Stations 2 and 3 (Stueber et al., 1978). Data previously collected from the five monitoring stations on WOC suggest that the bulk of the strontium-90 released to the Clinch River is acquired in WOC between Stations 2 and 3. To investigate possible sources of strontium-90 to WOC, groundwater and surface water samples were collected three times per week during October 1977 in the vicinity of Solid Waste Disposal Area (SWDA) 4. In addition, during December 1977, seven surface water sample profiles were collected in WOC between monitoring stations 2 and 3. Surface water samples were also collected from the floodplain areas. Based on these data, it was determined that SWDA 4, Waste Ponds 3539 and 3540, and the Sewage Treatment Plant were the primary sources of strontium-90 discharge to WOC between monitoring stations 2 and 3. Sampling data are tabulated in the report by sampling date and location.

#### Study 33: Radionuclides in WOC Gravel (1978-1985)

A number of studies were conducted to investigate the bedload transport of contaminated gravel in the WOC drainage basin, including all major streams and tributaries (Spalding and Cerling, 1979; Cerling and Spalding, 1981, 1982; Cerling and Turner, 1982; Cerling, 1985). The purpose of the studies was to evaluate the distribution of contaminants in different size classes of sediment and to evaluate the potential use of streambed sediments for locating and monitoring sources of contamination in WOC. Gravels were collected from the upper 10-cm (4-in) of sediment and wet sieved in the streamwater. Samples were collected at approximately 35 m (115 ft) intervals.

The first three studies focussed on the distribution of strontium-90, cobalt-60, and cesium-137. Data gathered during the studies showed that the concentration of strontium-90 in water can be back-calculated from the concentration in gravel using the appropriate distribution coefficient. cesium-137 was shown to bind irreversibly to gravel, and as such could be used to provide information about the time and duration of releases. cobalt-60 behaves in a manner intermediate between these two; that is, it is highly retained by gravel in oxidizing portions of the stream, and released in reducing portions of the stream. Levels of contaminants in streambed gravels

were compared with releases from various sources to determine the sources of strontium-90 in the watershed and their contribution to total discharge. In addition to radionuclides, the 1985 study also evaluated the behavior of metals, including arsenic, cadmium, copper, and lead, and organic contaminants.

Study 34: PCBs in Clinch River, EFPC, and Poplar Creek Sediments (1979)

On March 20, 1979, twelve bottom sediment samples were collected in the Clinch River, Poplar Creek, and EFPC to determine the concentrations of PCBs in surface sediments (Long, 1979). Samples were collected at two locations in the Clinch River above and below the Poplar Creek outfall, nine locations in Poplar Creek ranging from above the K-25 Site downstream to the Clinch River, and one location in EFPC. Results are tabulated in the letter report by sample number and location.

Study 35: Metals in Bear Creek and EFPC Sediments and Fish (1981-1982)

Samples of moss, liverwort, and sycamore roots along Bear Creek and EFPC were collected in December 1981 by S.B. Gough, an ORNL biologist, and L. Gough, a scientist with USGS. This study is discussed by UCC (1983). Data were gathered to justify a joint DOE-USGS research project on the presence of heavy metals reported to be in local environment. Samples were originally analyzed by the USGS Geochemistry Laboratory in Denver, and were reanalyzed at the Y-12 Plant.

Because of differences in the analytical results between the two laboratories, additional samples were collected by the ORNL Environmental Sciences Division and analyzed at Y-12 in May 1982. The 1982 samples included multiple samples at each location. At the request of Y-12 Plant management, the Environmental Sciences Division at ORNL conducted a study to determine the concentration of mercury in sediment, fish, moss, and pasture grass in EFPC and Bear Creek drainages and to ascertain whether mercury is still being released from Y-12 (Van Winkle et al., 1982). Surface sediment, fish, moss and liverwort, and pasture grass samples were collected along the length of EFPC (EFPCM 1.3 to 14.2) and Bear Creek (BCM 0.4 to 7.6) in April 1982. In addition, a sediment core was collected from New Hope Pond on May 5, 1982 to determine the historical record of mercury contamination of the pond. Results are tabulated in Van Winkle et al. (1982).

Study 36: Organics, Metals, and Priority Pollutants in Bear Creek Water (1983)

In September and October, 1983, water samples were collected by Bechtel National, Inc. at four locations on Bear Creek, upstream and downstream of the West Stream discharge and upstream and downstream of the East Stream discharge (1984). Samples were analyzed for organics, metals, and priority pollutants. The study was conducted under the MOU between the DOE,

USEPA, and TDHE which requires a description of runoff from the Oil Landfarm. Data are tabulated in the report.

Study 37: Volatile Organics, PCBs, and Metals in Bear Creek Sediments and Water (1983)

An investigation of the Burial Grounds in Bear Creek Valley was initiated in 1983 by Bechtel National, Inc., in response to a request by USEPA and TDHE for characterization of the oil retention ponds (McCauley, 1984). Sediment and water samples were collected in September, 1983 at several stations in Bear Creek, upstream and downstream of the confluences of the burial ground drainage streams with Bear Creek. Samples were analyzed for volatile organic priority pollutants, PCBs, and metals. Data are tabulated in the report.

Study 38: Mercury, Organics, and Radionuclides in Bear Creek, Clinch River, EFPC, and WOC Surface Water (1984)

Surface water samples were collected by the TVA in 1984 during Task 1 of the Instream Contaminant Study (TVA, 1985a). Task 1 focussed on evaluation of surface waters in EFPC, Bear Creek, lower WOC, Mill Branch, and the Clinch River, including Melton Hill Reservoir and Watts Bar Reservoir. The emphasis was on determination of hydrologic characteristics and mercury concentrations in EFPC and Bear Creek; these data were used in the prediction of sediment transport. Sampling for additional contaminants was conducted to determine the need to add contaminants to ongoing monitoring programs. Analyses were conducted under both baseflow and storm conditions. During the baseflow survey conducted on May 30-31, 1984, samples were collected at nine stations: five in the Clinch River (CRM 6.80 to 24.00), one in EFPC (EFPCM 14.36), one in WOC (WOCM 0.40), and one in Poplar Creek (PCM 13.80). Baseflow measurements included water quality parameters and analysis for selected metals, nutrients, priority pollutants, including volatile organics and PCBs, and radiological parameters, including gross alpha and gross beta, tritium, iodine-131, protactinium-234, strontium-90, uranium-234, uranium-235, cesium-137, cobalt-60, neptunium-237, and thorium-228. During the two stormflow surveys, samples were collected at five locations in EFPC (EFPCM 0.03 to 14.36), one in Mill Branch, and one in Bear Creek (BCM 0.55). Measurements were for water quality parameters, mercury, and radionuclides. Samples were collected from May through November, 1984. Analytical results are presented as appendices to the TVA report. Data collected during the baseflow and stormflow surveys are summarized in Table A-14 and A-15, respectively (Appendix A).

Study 39: Metals and Radionuclides in EFPC Water (1984)

To evaluate the contribution of Y-12 sewage discharges to the Oak Ridge Sewage Treatment Facility, ORAU began monitoring sewage sludge from the West End Wastewater Treatment



Facility for radionuclides and metals in 1984. Data were used to determine whether Y-12 sewage discharges to the Oak Ridge Sewage Treatment Facility contribute significant amounts of radioactivity and metals to the EFPC watershed (MMES, 1985).

Study 40: Mercury and Radionuclides in Bear Creek, Clinch River, EFPC, and WOC Sediments (1984)

Mercury contamination in sediments and floodplain deposits along EFPC, Bear Creek, and lower WOC was evaluated by the TVA during Tasks 2 and 3 of the Instream Contaminant Study (TVA, 1985b; 1985c; 1985d). Task 2 focussed on the evaluation of the extent of sediment contamination. Samples were collected from June through November 1984. Sediment core samples were collected at 122 locations in the floodplain of EFPC and four locations in the floodplain of Bear Creek. Instream samples for mercury and radiological analyses were collected at 19 locations in EFPC and four locations in lower WOC. To measure the concentration of other contaminants in surface layer sediments downstream of DOE facilities, surface layer samples for mercury, priority pollutants (metals and organics), cyanide, phenols, PCBs, and radiological parameters were collected at 34 locations: 16 on EFPC (EFPCM 1.2 to 14.36), three on Bear Creek (BCM 0.55 to 7.40), four on lower WOC (WOCM 0.18 to 0.55), three on Poplar Creek (PCM 1.00 to 6.80), four on the Clinch River in Watts Bar Reservoir (CRM 3.70 to 18.30), one in Melton Hill Reservoir (CRM 24.00), and three in Norris Reservoir (for background). Finally, to obtain information on the presence of mercury, radionuclides, PCBs, and chromium in sediment in the Clinch and Tennessee Rivers, sediment cores were collected at eight locations in the Clinch River below Melton Hill Dam and analyzed for mercury and radionuclides, and seven locations in the Tennessee River from Watts Bar Reservoir to Gunter'sville Dam and analyzed for mercury, PCBs, and chromium. Radiological parameters analyzed in Task 2 included gross alpha and gross beta, uranium, strontium-89, strontium-90, Co-60, cesium-134, cesium-137, potassium-40, radium-226, thorium-234, europium-152, europium-154, americium-241, actinium-228, protactinium-234, plutonium-238, plutonium-239, and curium-244. Raw data from sampling and analysis of sediment are presented in the appendices to the TVA report.

Measurements of metal and radionuclide concentrations in surface sediments are summarized in Tables A-16 and A-17, respectively (Appendix A). Mercury concentrations in core samples collected in the Clinch and Tennessee Rivers are presented in Tables A-18 and A-19, respectively.

Task 3 of the Instream Contaminant Study focussed on evaluation of the annual net transport of mercury in contaminated sediment in the EFPC and Bear Creek watersheds during stormflow events (TVA, 1985d). Three storms were sampled: October 22-23, 1984; November 10-11, 1984; and April 5-6, 1985. Storm event data were used to develop sediment and mercury rating functions, which were applied to streamflow duration distributions available from USGS stream

gages at EFPCM 3.3 and BCM 0.8. These data were used to determine the average annual sediment and mercury loadings from the EFPC and Bear Creek watersheds. A flow model was used to predict waterflow velocities in overbank sections of the EFPC floodplain; these data were used to predict the potential for future floods to scour mercury-contaminated sediment from floodplain areas.

#### Study 41: Metals, Organics, and Radionuclides in Bear Creek Sediments and Water (1984-1986)

A number of analyses of Bear Creek surface waters and sediments have been conducted since neutralization of the S-3 Ponds in 1983. These studies are summarized by Turner et al. (1988). Studies were conducted by Bechtel National, Inc. (HSEAD 1985), Roy F. Weston, Inc. (R.R. Turner, 1987, personal communication to G.R. Southworth, ORNL/ESD), the Y-12 Plant (R.R. Turner, 1987, personal communication to G.R. Southworth, ORNL/ESD), USGS (Pulliam 1985), and ORNL (Southworth et al., 1992). Analyses were for metals, organics, including volatile organics and PCBs, gross alpha and gross beta radioactivity, and radionuclides, including americium-241, curium-242/243, curium-244, cesium-137, potassium-40, neptunium-237, plutonium-238, plutonium-239/240, technetium-99, thorium-232, thorium-230, uranium-233/234, uranium-235, and uranium-238. During the ORNL studies, water samples were collected in August 1984, January 1986, May 1986, and September 1986 at 7 locations in Bear Creek ranging from BCM 2.02 to BCM 7.68. Data from these studies are compiled in the appendices to Turner et al. (1988).

#### Study 42: Metals, Organics, and Radionuclides in Clinch River, EFPC, and Poplar Creek Sediments (1985)

From January 10 through February 28, 1985, a survey of sediments in streams surrounding the K-25 Plant was conducted to identify points where pollutants have historically or are currently entering the surface waters (Ashwood et al., 1986). Approximately 180 surface-sediment grab samples were collected in the Clinch River, in Poplar Creek at its confluence with the Clinch River to about 1.6 miles upstream of the mouth of EFPC, in EFPC, and in tributaries draining K-25. Three sediment cores were also collected: one in EFPC near its the confluence with Poplar Creek, one in lower Poplar Creek near its mouth on the Clinch River, and one in a sediment accumulation zone where the Clinch River widens into Watts Bar Lake. Cores were collected to gain an understanding of the historical deposition of contaminated sediment. Samples were analyzed for metals, organics, and radionuclides, including beryllium-7, cesium-137, cobalt-60, uranium-235, and uranium-238. The parameters selected for evaluation were those contaminants identified by Hoffman et al. (1984) as warranting further study on the ORR. Sample results are tabulated by sample number, location, and analyte (Ashwood et al., 1986). Based on the results of the analyses, the authors concluded that mercury, cesium-137, and cobalt-60 originated from sources outside K-25, and that external sources also contributed

to uranium and miscellaneous organic contamination. The major sources of contamination within K-25 were concluded to be the K-1700 stream on the Mitchell Branch, the K-901A chromate pond, the K-710A powerhouse, and K-1007B pond. These sources are associated with uranium, chromium, nickel, copper, silver, and PCB contamination.

Study 43: Metals, Volatile Organics, PCBs, and Radionuclides in Bear Creek Sediments (1985)

During April 1985, a study of the depth distribution of contaminants, including metals, volatile organics, PCBs, gross alpha and gross beta radioactivity, and radionuclides, including cesium-137, potassium-40, uranium-235, and uranium-238, in Bear Creek floodplain soils was conducted. Two or three core samples up to 27 inches in length were collected at five locations along Bear Creek from BCM 0.64 to BCM 7.5. Analyses were conducted by the ORNL Analytical Chemistry Division. The data are discussed and presented by Turner et al. (1988). However, the source of these data are not discussed. A second sampling of soils near BCM 2.92 was conducted in October 1986 based on the results of the 1985 study, to better define the areal and depth distribution of PCBs and uranium, and in November 1985 near BCM 5.85 (Turner et al., 1988).

Study 44: Metals, Organics, and Radionuclides in Poplar Creek Sediments (1985)

On June 25, 1985, one floodplain soil core and one creekbed sediment core were collected at the proposed construction site for the new Blair Road Bridge over Poplar Creek, to determine the vertical distribution of contaminants (Olsen and Cutshall, 1985). Samples were collected by the Environmental Sciences Division of ORNL and were analyzed for radionuclides, including cesium-137, uranium-238, and potassium-40, metals, and organics. Raw data are available in this report.

Study 45: Herbicides, Pesticides, and PCBs in Bear Creek Sediments and Water (1985)

In August 1985, surface water data from a number of locations in Bear Creek ranging from BCM 0.87 to BCM 7.68 were collected by R.F. Weston, Inc. (Kimbrough, 1986). Data were collected as part of the remedial investigation plans for Bear Creek Valley Waste Disposal Areas developed in response to requirements of TDHE and USEPA. Water samples were analyzed for herbicides, pesticides, and PCBs. Groundwater, soil, and sediment samples were also collected. Raw data are presented in the report.

Study 46: Metals, PCBs, and Radionuclides in Clinch River Sediments and Water (1986)

In 1986, during the first phase of the Clinch River RCRA Facility Investigation, sediment and water samples were collected in Watts Bar Reservoir and analyzed for cesium-137 (Olsen et al., 1992). Approximately 190 surface-sediment grab samples and more than 60 sediment core

samples were collected in the reservoir. The study was conducted to identify contaminant accumulation patterns and potential problem or "hot spot" areas, and to estimate the total accumulation of cesium-137 in Watts Bar Reservoir sediments. The study focussed on cesium-137 since it can be used as a cost-effective tracer for identifying the transport and accumulation patterns of many other particle-reactive contaminants, including mercury, lead, Pu, and PCBs. Cs-137 concentrations measured in sediments are tabulated in the report by sample number and sample depth.

Reservoirs are efficient traps for riverborne particles, nutrients, and contaminants and are sites of rapid sediment and contaminant accumulation (Olsen et al., 1992). Thus, Watts Bar Reservoir serves as the major zone of contaminant accumulation in the Clinch River (Olsen et al., 1992). Based on the results of the 1986 study, 290 Ci of cesium-137 resided in Watts Bar Reservoir sediments at that time. Records of discharge indicate that a decay-corrected total of approximately 335 Ci of cesium-137 were released into the river system between 1949 and 1986. The study estimated that 75 metric tons of mercury have also accumulated in Watts Bar Reservoir. Vertical distributions of cesium-137 and mercury in dated sediment cores were used to estimate levels in the water column during the past 40 years. Measurements of the distribution of contaminants between aqueous and particulate phases are presented in Table A-20 (Appendix A). Sediment core data for TRM 567.5 and estimates of sediment deposition are presented in Table A-21.

#### Study 47: Mercury and Radionuclides in Clinch River and Watts Bar Reservoir Water (1986)

Abnormally high cobalt-60 concentrations were measured in WOL on November 25 and 26, 1986. As part of the Clinch River RCRA Facility Investigation, water samples were collected in Watts Bar Reservoir and at the mouth of the Clinch River to determine whether the elevated cobalt-60 levels could be traced into Watts Bar Reservoir (Olsen et al., 1992). In addition to cobalt-60, water samples were analyzed for mercury, cesium-137, cobalt-60, beryllium-7, plutonium-239 and plutonium-240, and plutonium-238. Data from the study indicated that it takes approximately two to three weeks for the cobalt-60 released into WOC to be transported via the Clinch River into Watts Bar Reservoir. Sediment data were also collected as part of this program.

#### Study 48: Mercury in WOC Sediments and Water (1988-present)

In 1988, a program was initiated in compliance with the Clean Water Act and ORNL's National Pollutant Discharge Elimination System (NPDES) permit to identify, locate, and minimize all sources of mercury contamination in ORNL discharges to the aquatic environment. Under this program, a network of water and sediment monitoring and sampling stations was established using available information on mercury deposits in receiving streams, knowledge of mercury

discharges from pipes to streams, and a review of chemical data from previous surveys. Monitoring stations are located on First Creek, Fifth Creek, and White Oak Creek. The data collected for 1988 and 1989 are presented by Taylor (1990).

Study 49: Metals, Organics, and Radionuclides in Clinch River and Poplar Creek Sediments and Water (1989-1990)

Surface water samples were collected as part of the Clinch River Remedial Investigation from December 1989 to February 1990 (Cook et al., 1992). The study was conducted to confirm suspected low dissolved concentrations of contaminants identified in previous scoping studies (Suter, 1991; Hoffman et al., 1991; Olsen et al., 1992). Surface water samples were collected at Norris Reservoir, upper Melton Hill Reservoir, Solway, lower Melton Hill Reservoir, Jones Island, the mouth of Grassy Creek embayment, Poplar Creek at the Hartland Bridge, four sites within Poplar Creek below the mouth of East Fork, Brashear Island, Kingston City park, and upper, mid, and lower Watts Bar Reservoir. Samples were analyzed for metals, organics, including volatile organics, semivolatile organics, pesticides, PCBs, and radionuclides, including tritium, cobalt-60, cesium-137, and strontium-90. Raw data are tabulated in the report by location and analyte.

Sediment samples were collected from December 1989 through July 1990 as part of the Clinch River Remedial Investigation (Cook et al., 1992). The study was conducted to evaluate contaminant release histories as shown by the depositional history of particle-associated contaminants. Both sediment core samples and surface sediment grab samples were collected. Sediment core samples were collected at Norris Reservoir, Solway, lower Melton Hill Reservoir, Jones Island, the mouth of Grassy Creek embayment, Poplar Creek at the mouth of Mitchell Branch, lower Poplar Creek, Brashear Island, Kingston Steam Plant, Kingston City park, and upper, mid and lower Watts Bar Reservoir. Surface sediment grab samples were collected from Melton Hill Park, Walker Branch Embayment, Grubb Island, the K-770 area, Campbell Bend, Sugar Grove, the Emory River, and Webster Bluff. Samples were analyzed for metals, organics, including volatile organics, semivolatile organics, pesticides, and PCBs, and radionuclides, including tritium, americium-241, curium-243/244, curium-245/246, curium-248, cobalt-60, cesium-137, plutonium-238, plutonium-239/240, strontium-90, uranium-234, uranium-235, and uranium-238. In addition to the above program, surface sediment samples were collected from near-shore sediment in Watts Bar Reservoir to provide information on the risks posed to human health from surface sediments in shallow waters of near-shore areas. These sediments are exposed during winter when water levels are lowered as part of the water management activities of the TVA. Raw data are presented in the report by location and analyte.

Figures B-5 through B-8 in Appendix B graph average concentrations of metals, mercury, Cs-137, and cobalt-60, respectively, measured in sediment cores at each of the sampling locations.

Study 50: Metals, Organics, and Radionuclides in Melton Hill, Norris, and Watts Bar Reservoir Sediments and Water (1990)

During May and June 1990, the TVA collected five 12-inch core sediment samples and one water sample (grab) from each of 12 recreational areas on Watts Bar Reservoir, three recreational areas on Melton Hill Reservoir, and three recreational areas on Norris Reservoir (for background) (TVA, 1991b). Data were collected to address concerns regarding the health and safety of persons who use beaches in the area for swimming or other water-body-contact sports. The five sediment samples from each recreational area were collected from equally spaced locations in the swimming area and used to create one composite sample. In addition, one core sample and one water sample were collected from each of 11 water intakes located throughout Norris, Melton Hill, and Watts Bar Reservoirs. Sediment samples were analyzed for metals, organics, including PCBs, and radionuclides, including tritium, cobalt-60, cesium-137, europium-152/154, protactinium-234, strontium-89, strontium-90, uranium-234, uranium-235, uranium-238, plutonium-242, plutonium-238, plutonium-239/240, plutonium-241, americium-241, curium-242, and curium-243/244. Water samples were analyzed for tritium. Sample results for most contaminants are tabulated in the report by location and analyte. Sampling results for cobalt-60, europium-152/154, and protactinium-234 were not located.

Study 51: Metals, Organics, and Radionuclides in White Oak Creek Embayment Sediment (1990-1991)

Sediment samples were collected from WOCE during 1990 and 1991 to provide a more thorough characterization of contamination in WOCE than was previously available, and to provide data to be used in a screening analysis to identify contaminants of concern from a human health standpoint (Blaylock et al., 1993). The screening analysis was conducted as a follow-up to the preliminary screening of contaminants in off-site surface waters by Hoffman et al. (1990), which relied upon historical sampling data. Hoffman et al. (1990) concluded that the embayment is an area of concern because of potential external exposure to cesium-137.

A total of 31 sediment grab samples were collected near the mouth of WOCE on August 30, 1990, and 41 grab samples were collected from the upper portion of the embayment on September 18, 1990 (Blaylock et al., 1993). Samples were analyzed for gamma-emitting radioactivity, cobalt-60, and cesium-137. These data, together with historical and recent sediment core data, were used to estimate inventories for radioactive contaminants in embayment sediment. Historical data were obtained from studies by Cerling and Spalding (1981), Oakes

et al. (1982b), and TVA (1985b), as well as the Clinch River Remedial Investigation (Cook et al., 1990). In addition, two sediment cores were collected in WOCE in January 1991, sectioned into 4-cm lengths, and analyzed for 91 organic contaminants, including PCBs. Three cores collected after July 1990 were analyzed for metals. All data collected during this study, as well as all historical data included in the screening analysis, are included in appendices to the report.

The results of this study and subsequent site characterization efforts at WOCE led to a CERCLA time-critical removal action, led and funded by DOE. This removal action consisted of the design and construction of a sediment-retention structure across the mouth of WOCE to prevent off-site migration of contaminated sediments into the Clinch River (Blaylock et al., 1993).

### 5.3.2 Historical Hydrologic Data

The USGS collects surface water discharge and precipitation data at a number of stations in the WOC watershed and in the Clinch River, Bear Creek, EFPC, and Poplar Creek (Borders et al., 1991; Bailey and Lee, 1991). These data are available in USGS Water Data Reports for the Water Year (Flohr et al., 1991; Mercer et al., 1992). USGS surface water discharge data and precipitation data can be accessed by remote connection to the USGS computer system in Nashville, TN; data are available in unit values (five minutes to hourly) at selected stations and daily values at all stations. Near real-time data (discharge, precipitation, etc.) are available from a number of stations connected to the USGS computer system by satellite telemetry (Borders et al., 1991). Flow in the WOC watershed has been monitored at three locations by the USGS since the early 1950s. These data are summarized by Webster (1976). Early hydrologic data for Bear Creek are summarized by McMaster (1967).

Hydrologic data are currently collected by a number of divisions within ORNL. The Environmental Sciences Division's (ESD) Watershed Hydrology Group collects and processes discharge data at a number of stations in the WOC watershed and vicinity for modeling studies, independent research, and remedial action activities. These data are available in raw stage data format, hourly or daily discharge, and in hardcopy or electronic formats. Available information on the hydrogeology and ecologic characteristics of the WOC flow system prior to 1985 are summarized by Sherwood and Loar (1986). Annual summaries of hydrologic data for the WOC watershed have been produced by the Remedial Action Program (RAP) of ORNL since 1986 (Sherwood and Borders, 1987). The summaries document hydrologic data collected in ongoing ORNL environmental studies and monitoring programs. Most data are maintained in the RAP database management system (Voorhees et al., 1986).

### 5.3.3 Historical Air Monitoring Data

Historical ambient air monitoring performed on or near the Oak Ridge Reservation was summarized earlier in Table 5-3. Descriptions of key programs are provided in this section.

Tables and figures showing selected examples of reported data excerpted from the original reports are presented in Appendix A and Appendix B, respectively.

#### Study 52: Routine Ambient Air Monitoring

Three networks for routine monitoring of airborne radioactivity were established in eastern Tennessee by the Applied Health Physics Division: the Local Area Monitoring Stations (LAMs) close to ORNL operational activities, the Perimeter Area Monitoring Stations (PAMs) on the perimeter of the DOE-controlled area, and the Remote Area Monitoring Stations (RAMs) outside of the DOE-controlled area at distances of 12 to 120 miles from ORNL. The number of stations in each monitoring network varied somewhat from year to year. Data were collected at the ORNL perimeter stations to evaluate the specific impact of the facility upon local air quality. Data were collected at the DOE perimeter stations to evaluate the impact of the entire ORR on air quality, and at the remote stations to evaluate background for eastern Tennessee.

Two types of air monitoring data were collected continuously at the LAMs, PAMs, and RAMs. Airborne radioactivity was measured using air filtration techniques, and radioparticulate fallout material was collected by impingement on gummed paper trays. Rainwater was also collected for measurement of fallout occurring as rain out. Beginning around 1963, samples were collected using charcoal cartridges and analyzed for radioiodine and, beginning around 1980, samples were collected at selected LAMs using silica gel and analyzed for tritium. The methods used by the Department of Environmental Management staff to collect samples as part of the ORNL air monitoring network are described by Oakes et al. (1981). Figures 5-5 through 5-8 show the locations of routine air monitoring stations that made up the Local, Perimeter, and Remote Air Monitoring networks.

Tables A-22 through A-27 and Figures B-9 through B-11 in Appendices A and B, respectively, present sample summary data of routine measurements associated with atmospheric releases from the ORR taken directly from Applied Health Physics Annual Reports. Tables A-22 and A-23 present results of average long-lived activity measurements based on filter paper analyses from 1954 and 1965, respectively. Along with average long-lived activity results, Table A-23 lists the average number of particles collected per cubic foot of air and the number of particles falling into several radioactivity intensity ranges. Figure B-9 reflects the trends of weekly average airborne radioactivity concentrations based on local, perimeter, and remote air station measurements during 1965. Table A-24 presents an example of isotopic analysis results from air filter samples during 1979.

Table A-25 shows an example of radioactive fallout measurements taken by use of gummed paper at the routine air monitoring stations during 1965. Figure B-10 reflects the trend of gummed-paper fallout measurements on and near the ORR for the same year.



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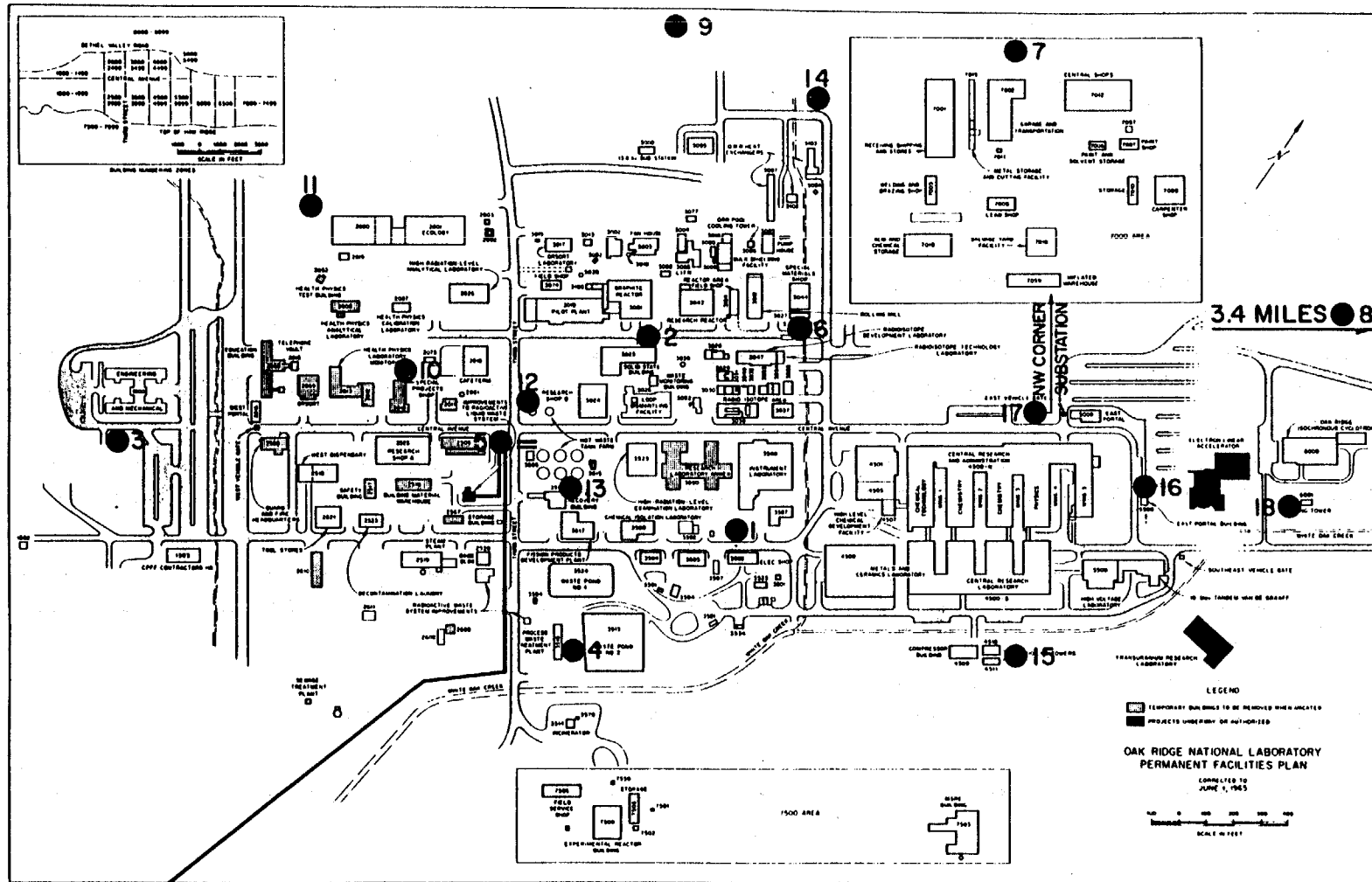
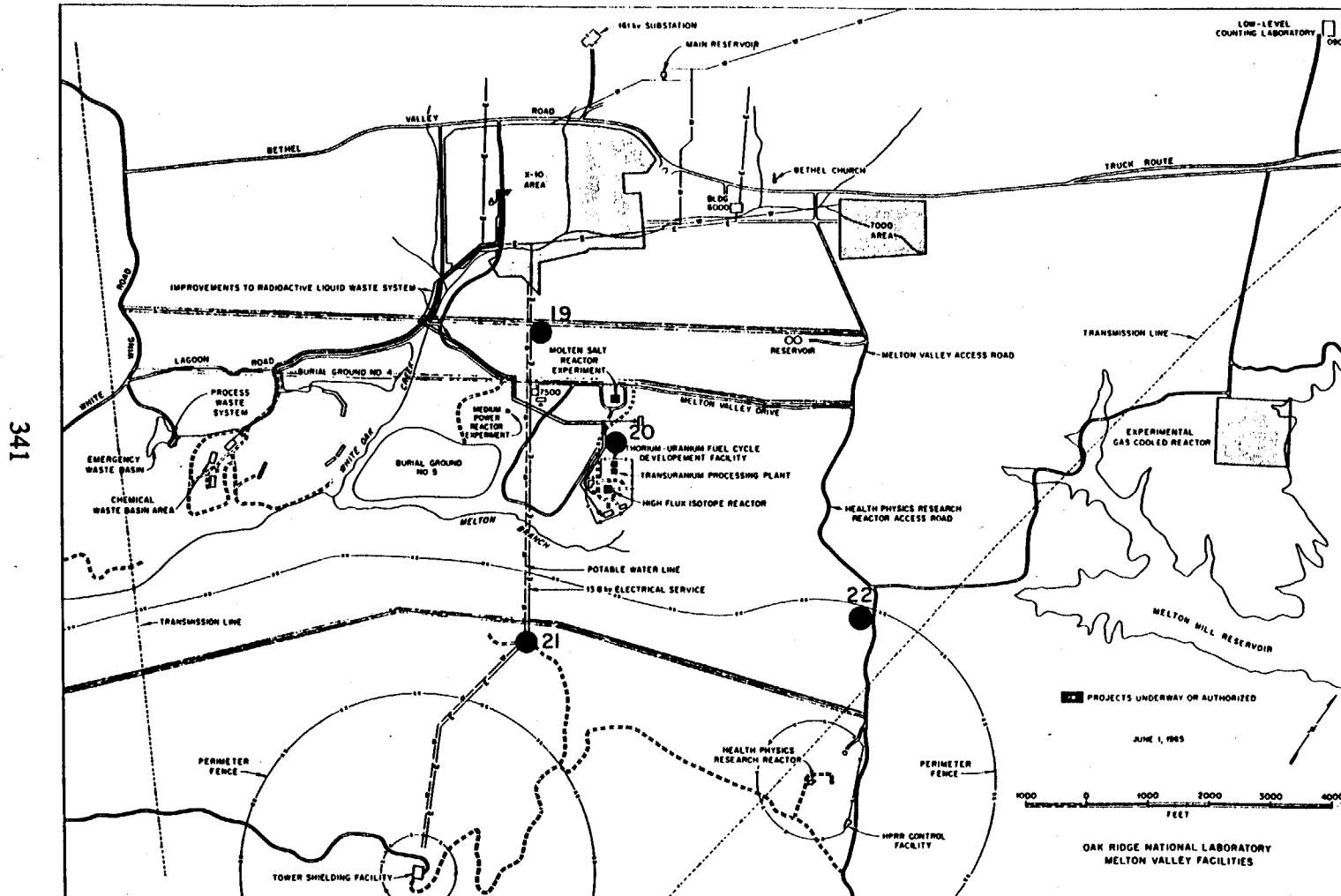
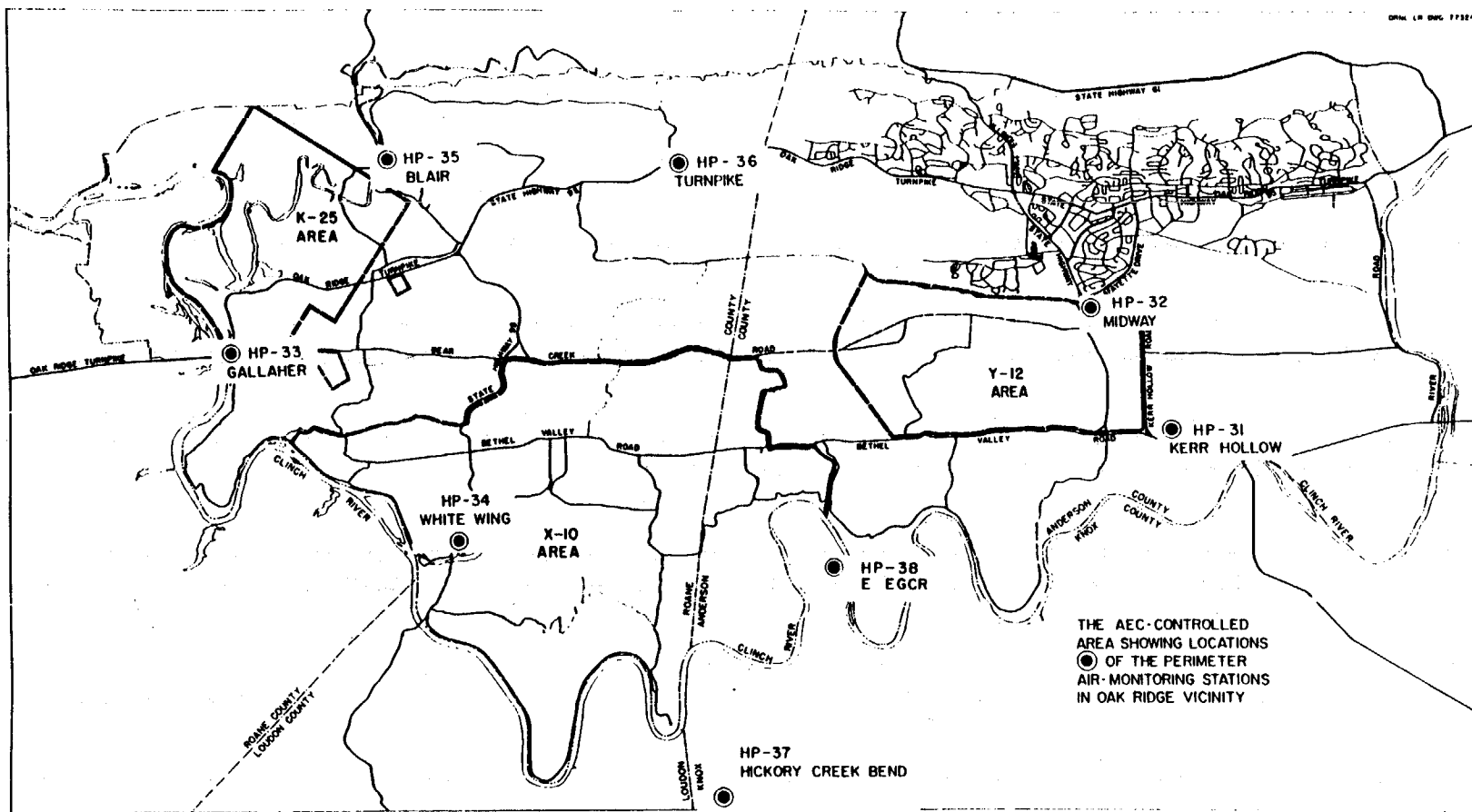


Figure 5-5 Map of X-10 Area Showing the Approximate Location of 18 of 22 of the Local Monitoring Stations Constituting the LAM Network.

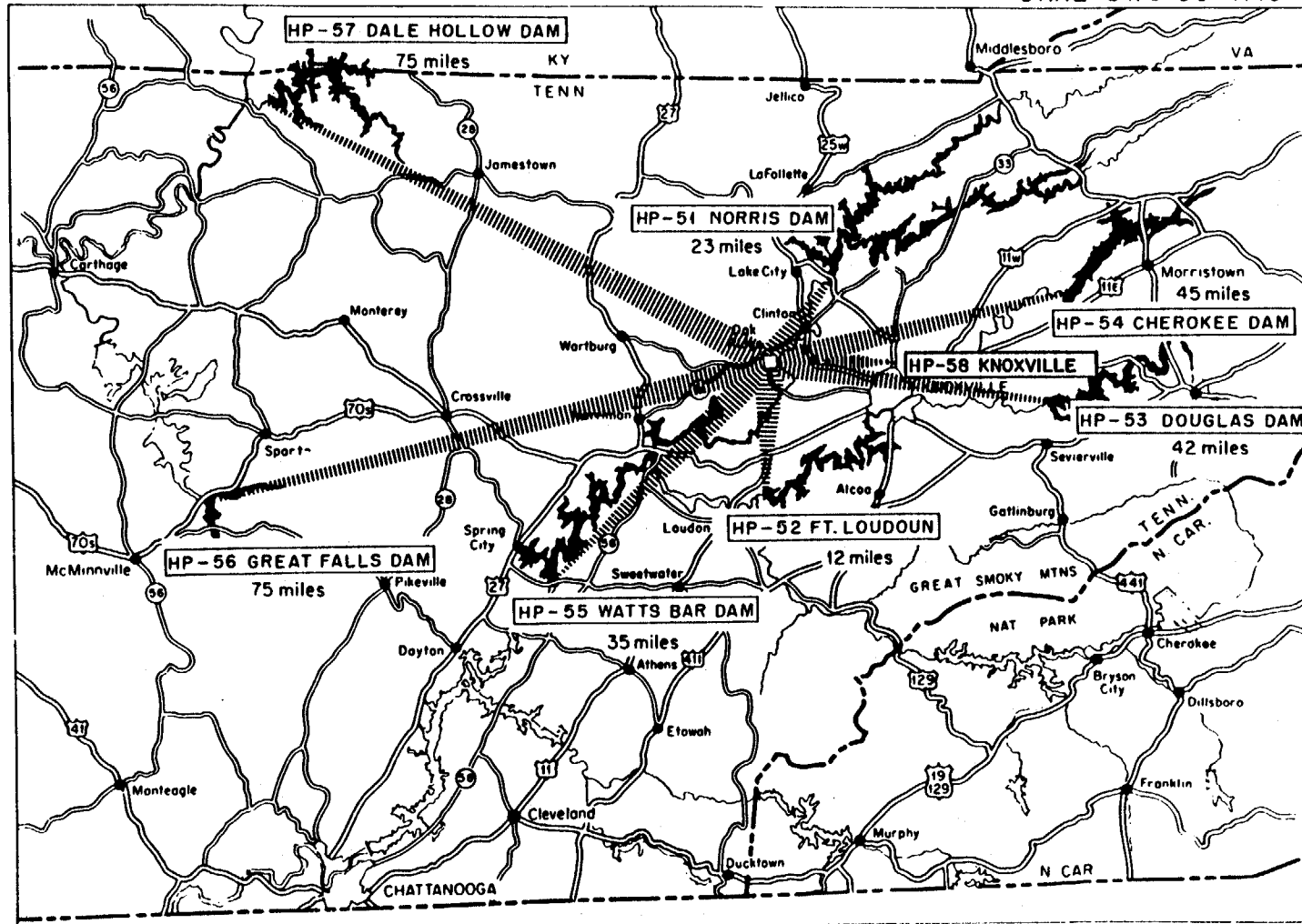


**Figure 5-6** Map of Laboratory Area Showing the Approximate Location of 4 of the 22 Local Monitoring Stations Constituting the LAM Network.



**Figure 5-7** Map of the AEC Controlled Area and Vicinity Showing the Approximate Location of the Perimeter Air Monitoring Stations Constituting the PAM Network.

Source: UCC (1966)



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**Figure 5-8** . Map of a Section of the East Tennessee Area Showing TVA and U.S. Corps of Engineering Dam Sites at Which Are Located the Remote Air Monitoring Stations Constituting the RAM Network.

Source: UCC (1966)

Figure B-11 shows the reported trend of iodine-131 measurements in the air at perimeter air stations during 1964 along with reported iodine-131 discharges from ORNL stacks for the same time period. An accidental release of about 20 curies of iodine-131 noted during the 12th week appears to have been detected at the perimeter air samplers. Table A-26 summarizes gross alpha, gross beta, and iodine-131 measurements at air monitoring stations from 1961-1984.

Tables A-27 is an example of the results of measurements of "rainout" radioactivity captured in precipitation samples collected at selected air monitoring stations in 1965.

Ambient air monitoring data for the K-25 Site have been located for at least as early as 1955. Data were collected for uranium and fluorides. These data are located in the K-25 Plant Records Center Box 6-A, 8, 48 Site Records vault. By the mid-1980s the program included five ambient air monitoring stations located in the predominant wind direction. Samples were collected for uranium, nickel, lead, chromium, and total suspended particulates (TSP) at all five stations, and particulate matter in the less than 10  $\mu\text{m}$  fraction ( $\text{PM}_{10}$ ) was collected at one station. Two additional stations were added near the K-25 site in 1988 to measure potential releases of PCBs, furans, dioxins, hexachlorobenzene, and uranium from the K-1435 TSCA Incinerator (MMES, 1991). Data from the mid-1980's are summarized in the site-wide annual environmental monitoring reports.

Air monitoring data for uranium were collected at the Y-12 Plant as early as 1958 (Patterson, 1958). However, the measurements were made to evaluate workplace air within the enriched uranium processing areas of the Y-12 Plant. Four ambient mercury monitoring stations were established at the Y-12 Plant in 1986 (one on the east end of the plant, one on the west end of the plant, and two stations near Building 9201-4) (MMES, 1991). An additional station was established at New Hope Pond in August 1987. Following closure of New Hope Pond in December 1988, this station was moved to a new location approximately 700 feet east of the original site. Since no established method had previously existed, the Environmental Sciences Division of ORNL developed a method for measurement of mercury in ambient air using a Teflon filter and iodated charcoal sampling tube. In 1990, an ambient air monitoring network of 12 stations was established on the perimeter of the Y-12 Plant. At the Y-12 perimeter stations, samples were collected for suspended uranium particulates and fluoride, as well as TSP and  $\text{PM}_{10}$ .

#### 5.3.4 Historical Meteorological Data

Until 1985, no system for continuous collection of meteorological data on the ORR was established. While data are available for various locations on the ORR since as early as 1943, periods of data collections were variable (e.g., one year, several years), as was the length of time collection equipment was operated per day (Kornegay, 1993).

The first meteorological station on the ORR was installed in 1943 to assist in monitoring of airborne radioactivity at ORNL in conjunction with the startup of the "Clinton pile" (now known as the Graphite Reactor) (DOE, 1979). This station was installed on a 120 ft water tank, with an added 60 ft mast to record temperatures continuously at six levels from 4 ft to 183 ft, wind direction and speed at approximately 130 ft, and wet-bulb temperature at 4 ft (U.S. Weather Bureau, 1953). Hourly, daily, and monthly records from the instruments were kept by the Health Physics Division of Clinton Laboratories (now ORNL). In addition, daily measurements of rainfall, barometric pressure, and cloudiness were recorded (U.S. Weather Bureau, 1953). Data were collected at this station until 1972.

Meteorological data on the Y-12 and K-25 sites were collected shortly following the initiation of operations at these facilities. At K-25, hourly records of temperature and wet-bulb temperature were maintained by the Process Utilities Department at two cooling tower installations. In addition, partial records of barometric pressure were kept by the K-25 Research and Development Laboratory during this period (U.S. Weather Bureau, 1953). Beginning in 1948, this lab also maintained a recording anemometer and wind vane. In 1947, the three ORR stations were designated as official U.S. Weather Bureau cooperative offices (DOE, 1979).

In addition to the on-site meteorological stations at each of the three plant sites, two off-site meteorological stations were historically operated in the vicinity of the ORR: a National Oceanic and Atmospheric Administration/ Atmospheric Turbulence and Diffusion Laboratory (NOAA/ATDL) station in the city of Oak Ridge, with records dating from 1947, and a U.S. Weather Bureau office at the McGhee-Tyson Airport in Knoxville, approximately 21 miles ESE of ORNL, with records dating from 1942 (U.S. Weather Bureau, 1953). Measurements made at the city of Oak Ridge include continuous temperature, humidity, wind direction and wind speed, and daily records of precipitation. Measurements at the airport include hourly temperature, relative humidity, wind direction, wind speed, and precipitation, 6-hourly to hourly visual observations of sky conditions, weather phenomena, and barometric pressure, and daily maximum and minimum temperature, total precipitation, prevailing wind direction, and fraction of possible sunshine (U.S. Weather Bureau, 1953).

Upper level meteorological observations have been recorded at several stations in the southeastern United States within a several-hundred mile radius of the ORR since the 1920s. Upper-air wind observations were recorded based on visual tracking of a free balloon at a station in Knoxville. These observations were recorded four times per day since before the initiation of operations on the ORR. Upper-air wind observations based on radio tracking of a balloon were recorded twice a day at Nashville, 130 miles west of the ORR, since January 1947. In addition, observations of upper-air temperature, humidity, and pressure were recorded twice a day beginning in 1943 at Nashville, Atlanta, 140 mi south of the ORR, and Greensboro, NC, 250 miles east of the ORR, since 1939 (U.S. Weather Bureau, 1953).

In 1956, a meteorological recording facility was established at the Tower Shielding Facility (TSF), located near Melton Hill Dam approximately two miles south of the ORNL tower site. Measurements for temperature, wind speed and direction, and dew point were made through 1972. Temperature gradient measurements are available through March 1965 (Strand and Miller, 1978). In 1977, the ATDL established a station at the Walker Branch Watershed. Measurements at this station include continuous measurements of diffuse and direct solar radiation, horizontal wind speed, wind direction, and air temperature (Strand and Miller, 1978).

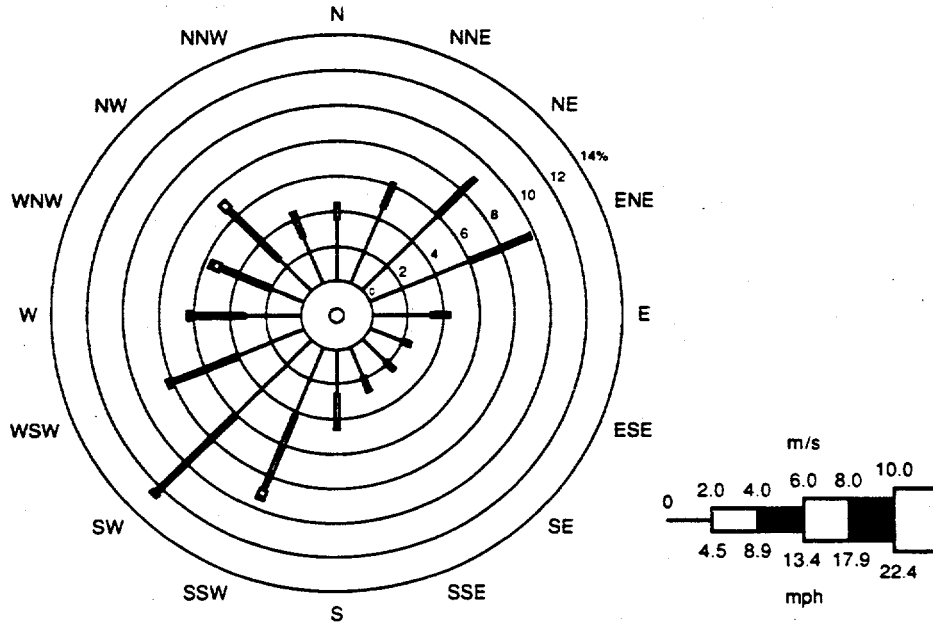
Detailed analysis of meteorological parameters for Oak Ridge area weather from 1948 to 1952 have been compiled by the U.S. Weather Bureau (U.S. Weather Bureau, 1953). A supplement to this report was prepared in 1963 (Hilsmeier, 1963). Meteorological data for the period from 1951 to 1971 have been compiled by NOAA (NOAA, 1972). The availability of meteorological data in computer database systems accessed through the ORNL Environmental Sciences Data Center, and the time spans for which data are available, are described by Strand and Miller (1978).

In 1985, a network of meteorological observation towers on the ORR was put in operation (MMES, 1987). This network consists of one 60-m (197-ft) tower at K-25 (Tower 1); one 100-m tower (328-ft) (Tower 2) and two 30-m (98-ft) towers (Towers 3 and 4) on the ORNL site; one 100-m tower (Tower 5) and one 60-m tower (Tower 6) on the Y-12 site; one 100-m tower (Tower 7) on the Walker Branch watershed; and one 110-m tower (361-ft) (Tower 8) on the Clinch River Breeder Reactor Project (CRBRP) site. Towers 7 and 8 are not commonly used for routine modeling or emergency response activities (MMES, 1988). Data are collected on the towers at multiple levels to determine the vertical structure of the atmosphere. Data are collected on all towers at 10 m and at the top of the tower. In addition, on the 100-m towers, data are collected at intermediate levels (30-or 60-m) (MMES, 1991). At each level that data are collected, measurements are made for temperature, wind speed, and wind direction. Atmospheric stability data are also measured at each tower. Precipitation, humidity, and solar radiation is measured at tower 2 at ORNL. Tower data are collected by a dedicated control computer at each site (15-minute and hourly values) (MMES, 1991), and are summarized in the Annual Environmental Monitoring Reports for the Oak Ridge Reservation beginning in 1986.

ORNL's Environmental Sciences Division (ESD) Watershed Hydrology Group has collected precipitation data at a number of stations in the WOC watershed since 1980 (Borders et al., 1991). These data are available in the RAP database, and are summarized in a series of annual reports on the hydrology of the WOC watershed beginning in 1988 (Borders et al., 1991).

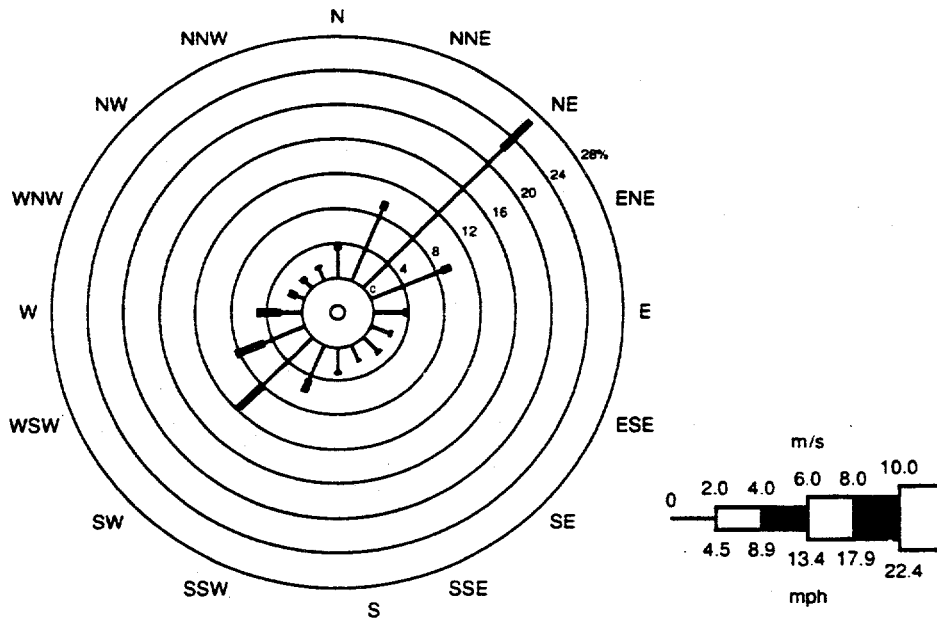
Wind roses for 1991 from the K-25 site (Tower 1), the X-10 site (Tower 3), and the Y-12 Site (MTE or Tower 5), using data collected at the 10 m (33 ft) level are shown in Figures 5-9, 5-10, and 5-11, respectively. Prevailing winds are generally up-valley from the southwest and west-southwest, or down-valley from the northeast and east-northeast (MMES, 1991).

Figure 5-9



1991 wind rose for K-25 tower MT1 (10-m level), with 91.4% of possible data.

Figure 5-10

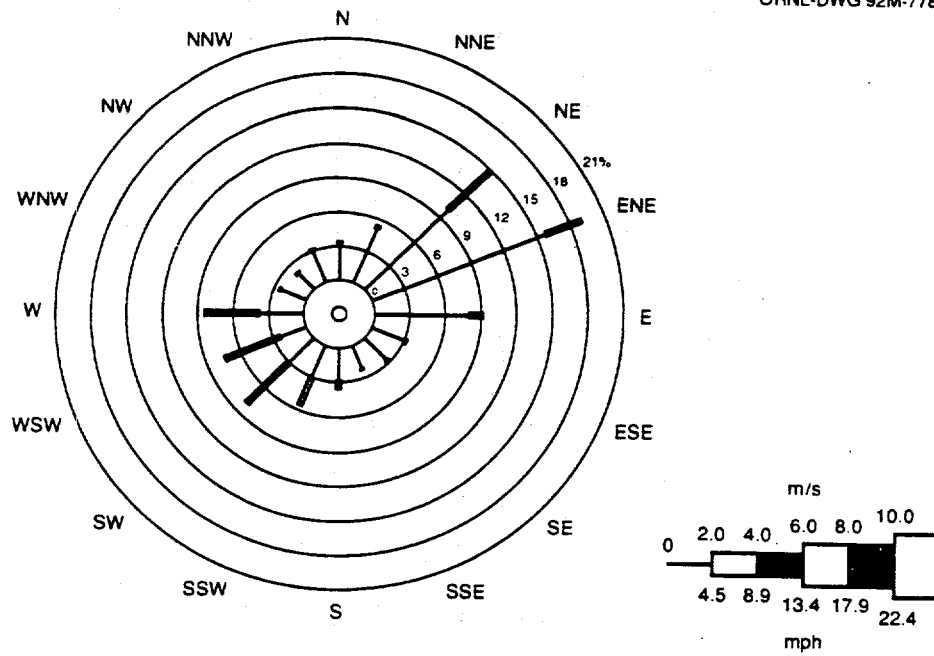


1991 wind rose for ORNL tower MT3 (10-m level), with 84.0% of possible data.



Figure 5-11

ORNL-DWG 92M-7785



1991 wind rose for Y-12 Plant tower MTE (east) (10-m level), with 99.3% of possible data.

### 5.3.5 Historical Biological Monitoring Data

Available data from monitoring of aquatic biota, beef, milk, deer, waterfowl, and vegetation were summarized earlier in Table 5-4 and Table 5-5. Summaries of key studies of these media are presented in this section. Tables and figures showing selected examples of reported data excerpted from the original reports are presented in Appendix A and Appendix B, respectively.

#### 5.3.5.1 Fish Monitoring Data

##### Study 53: Radioactivity in WOC/WOL Fish (1944-1945)

Studies of radioactivity in fish from WOC and WOL were conducted by the ORNL Biology Section from May 3, 1944 to April 3, 1945 (Morgan and Western, 1947). A correlation was reported between activity in the mud of WOL and beta activity in the tissues of the fish. Results are discussed, but no data are presented.

##### Study 54: Radioactivity/Radionuclides in Clinch River and WOC/ WOL Fish (1948-1949)

Surveys of radioactivity in fish in the WOC drainage system and the Clinch River were conducted in 1948 and 1949 by the Health Physics Division of ORNL in cooperation with the Fish and Game Branch of the TVA (Knobf, 1951). The surveys were conducted to determine whether consumption of fish from these systems would pose a human health risk. Samples were collected on October 4 and October 26, 1948, and May 2 and 3, 1949, from CRM 13.3, CRM 18-19, the confluence of the Clinch River and WOC, and WOL. A total of 198 fish were processed for measurement of radioactivity. Analyses included gross beta, strontium-89, strontium-90, Cs-37, and yttrium-90. Summary data are presented in the report. Table A-28 (Appendix A) presents average gross beta measurements for fish collected during 1948 from the Clinch River, WOC, and WOL.

##### Study 55: Strontium-90 in Clinch River Fish (1962)

Flesh and bone from white crappies collected in the Clinch River in the vicinity of WOC were analyzed for strontium-90 content in 1962 (Nelson and Griffith, 1962). The study was conducted to evaluate the distribution of strontium-90 and stable strontium between fish and water. Results are discussed in the Health Physics Annual Progress Report.

##### Study 56: Radionuclides in Clinch River and WOC Fish (1962-1963)

As part of the Clinch River Study, fish were collected from the Clinch River for a radiation safety evaluation in May and June 1962 and in May 1963, using hoop, gill, and trammel nets

and by electric shocking (Struxness et al., 1967). Fish were collected in the Clinch River downstream of the mouth of WOC, and analyzed by ORNL and USPHS laboratories. The fish were analyzed for gamma-emitting radionuclides by gamma spectroscopy, and for strontium-90 by standard wet-chemical procedures and low-background beta counting. Data are summarized in Status Report No. 5 on Clinch River Study (Morton, 1965). Average concentrations of radionuclides measured in Clinch River and Tennessee River fish are shown in Table A-29 (Appendix A).

During the tagging operations, fish were caught in hoop nets, identified, measured, weighed, tagged, and released. Tagging was conducted from CRM 16.5 to 21.7 during July to September 1960 and from CRM 14.0 to 20.8 during April to July 1961.

Study 57: Radionuclides in WOL Fish (1966)

To determine if the trophic level of fish is correlated with its concentration of a radionuclide, five species of fish were collected from WOL in 1966 by the ORNL Health Physics Division and analyzed for cesium-137, cobalt-60, and strontium-90 (Kevern and Griffith, 1966). Results are summarized in the report.

Study 58: Cesium-137 in WOL Fish (1967-1969)

Concentrations of cesium-137, stable cesium, and potassium and specific activities of cesium-137 were determined for seven species of fish in WOL from June 1967 to January 1969 (Kolehmainen and Nelson, 1969). The study was conducted to determine the intake of cesium-137, which was used to calculate feeding rates of bluegill at different times of year. Summary results are tabulated in the report.

Study 59: Mercury, PCBs, and Radionuclides in Clinch River Fish (1967-present)

Fish samples were collected routinely from the Clinch River by the ORNL Applied Health Physics Division beginning in 1967. Various species were collected and fish from each species were composited and analyzed for gamma and specific radionuclides, including strontium-90, plutonium-239, plutonium-238, uranium-238, uranium-235, uranium-234, cesium-137, cobalt-60, and potassium-40. Analyses and specific locations varied. Beginning in 1978, fish were also routinely analyzed for mercury, and beginning in 1984, fish were monitored for PCBs. These data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports. Table A-30 in Appendix A presents the concentrations of radionuclides and mercury measured in Clinch River fish, as presented in the 1978 Environmental Monitoring Report.

Study 60: Mercury in EFPC and Bear Creek Fish (1970)

Fish samples were collected in EFPC and Bear Creek in 1970 by M. Sanders, the Y-12 Environmental Coordinator, and analyzed for mercury. Results were reported to J.P. McLendon in an internal memorandum dated August 6, 1970. This memorandum was not located; however, the results are summarized in UCC (1983). Exact sample locations were not given.

Study 61: Tritium in WOL Fish (1972)

To determine the distribution and biological turnover of tritium in fish in a chronically contaminated lake, a study was conducted by the ORNL Environmental Sciences Division in which bluegills from an uncontaminated lake were placed in WOL during 1972 and assayed for tritium over a 36-day period (Blaylock et al., 1972b). Concentration ratios of tritium in fish to tritium in water and turnover of tissue-bound and body-water tritium were determined.

Study 62: Mercury in Clinch River and Poplar Creek Fish and Sediments (1974-1976)

An evaluation of mercury contamination in the EFPC- Poplar Creek- Clinch River drainage was conducted from 1974 through 1976 by the Environmental Sciences Division of ORNL (Elwood, 1984). The study was conducted to verify the apparent mercury contamination in the drainage and to establish the extent of contamination in fish from these environs. Fish were collected in May, June, and October 1976 from Poplar Creek and the Clinch River and analyzed for mercury. Collection locations included the Clinch River from CRM 4.5 to 13.5, Melton Hill Reservoir, and Poplar Creek from PCM 0.0 to the confluence with EFPC. During 1976, a total of 11 samples were collected in Melton Hill Reservoir, 86 in Poplar Creek, and 186 in the Clinch River upstream and downstream of the confluence with Poplar Creek. Raw data are tabulated in the report by Elwood (1976).

Study 63: Mercury in Clinch River and Poplar Creek Fish (1976-1977)

During 1976 and 1977, an evaluation of mercury levels in fish from Poplar Creek and the Clinch River was conducted by the ORNL Applied Health Physics Division following the measurement of mercury concentrations above background in creek sediments during the routine sediment monitoring program. These data were not located.

Study 64: Metals and PCBs in Clinch River and Poplar Creek Fish (1977)

From April to September 1977, a comprehensive biological sampling program was conducted by the ORNL Environmental Sciences Division to characterize the aquatic communities in Poplar Creek and the Clinch River in the vicinity of the K-25 Site (Loar et al., 1981a). A total of 362

fish of 15 species were analyzed for metals, including mercury, lead, cadmium, zinc, copper, chromium, and nickel, and PCBs. Samples were collected at three locations in Poplar Creek (at PCM 0.5, 5.5, and 11.0) and three locations in the Clinch River (at CRM 10.5, 11.5, and 15.0). Data are tabulated in the report by sample number and date.

Study 65: Uranium in Clinch River and Poplar Creek Fish (1977)

In summarizing historical environmental monitoring data available for the K-25 Site area, Long and Rogers (1985) report that fish in Poplar Creek and the Clinch River were analyzed in 1977 for uranium. Reported concentrations of uranium in fish from Poplar Creek ranged from less than 0.005 ppm to 0.061 ppm, while concentrations in fish from the Clinch River ranged from 0.005 ppm to 0.072 ppm. The source of these data was not referenced and could not be located.

Study 66: Metals in Clinch River, WOCE, and WOL Fish (1979)

Fish samples were collected and analyzed for seven trace elements in 1979 as part of a biological sampling program to evaluate the effects of ORNL operations on aquatic biota in the WOC watershed (Loar et al., 1981b). Field samples were collected by the ORNL Environmental Sciences Division. Data were to be used in the Environmental and Safety Report for ORNL. Muscle tissue of several fish species from five stations in the Clinch River, including CRM 19.0, CRM 22.0, WOL, WOCE, and Melton Hill Reservoir, were analyzed for cadmium, chromium, copper, lead, mercury, nickel, and zinc. Muscle tissue samples from each fish were analyzed for total mercury by flameless atomic absorption spectroscopy, and for other metals by graphite furnace atomic absorption spectroscopy. Summary data (e.g., means) are presented in the report. These data are shown in Table A-31 (Appendix A).

Study 67: Mercury in Bear Creek and EFPC Fish and Sediments (1982)

At the request of Y-12 Plant management, the Environmental Sciences Division at ORNL conducted a study to determine the concentration of mercury in sediment, fish, moss, and pasture grass in EFPC and Bear Creek drainages and to ascertain whether mercury is still being released from Y-12 (Van Winkle et al., 1982). Surface sediment, fish, moss and liverwort, and pasture grass samples were collected along the length of EFPC (EFPCM 1.3 to 14.2) and Bear Creek (BCM 0.4 to 7.6) in April, 1982. In addition, a sediment core was collected from New Hope Pond on May 5, 1982 to determine the historical record of mercury contamination of the pond. Results for each fish sampled are tabulated in the report. A table summarizing fish concentrations at each sampling location is presented in Appendix A (Table A-32).

Study 68: Mercury in EFPC and Poplar Creek Fish (1982)

During 1982, 96 fish of 14 species were collected in Poplar Creek from three locations near the K-25 Site (Stiff, 1982). The locations were upstream and downstream of the confluence with EFPC, and near the mouth of Poplar Creek. Samples were analyzed for organic mercury. Results are tabulated in the report.

Study 69: Mercury in Bear Creek and EFPC Frogs and Crayfish (1983)

Frogs and crayfish were collected from EFPC between New Hope Pond and the Bear Creek Road bridge at the entrance to the Y-12 Plant area, and in Bear Creek (Blaylock et al. 1983). Samples were analyzed for mercury. This report was not located; data are discussed by Richmond and Auerbach (1983). The date of sample collection is not given. The mean concentration of mercury in the muscle tissue of frog legs and crayfish tails was 0.60 and 2.50  $\mu\text{g/g}$  fresh weight, respectively.

Study 70: Mercury in EFPC and Poplar Creek Turtles (1984)

As part of the ORAU environmental monitoring and surveillance studies of the Oak Ridge community, turtles were collected from Poplar Creek and EFPC during 1984 and analyzed for mercury (Hibbitts, 1984). Samples were collected along the length of EFPC from Y-12 to its confluence with Poplar Creek. Raw data are tabulated in letter reports. Floodplain soils, stream sediments, vegetables, and well water were also monitoring as part of these studies.

Study 71: Metals, PCBs, and Radionuclides in Bear Creek, EFPC, WOC/ WOL, Melton Hill Reservoir and Watts Bar Reservoir Fish, Frogs, Turtles, and Crayfish (1984)

Fish and aquatic animals were collected for analyses by the TVA during Task 4 of the Instream Contaminant Study (TVA, 1985e). Fish samples were collected in the Clinch River (CRM 2.0, 6.0, 11.0), EFPC (EFPCM 4.0, 8.8, and 13.8), Bear Creek (BCM 0.4), lower WOC (WOCM 0.2), WOL, and at Melton Hill Dam. Samples were collected from mid-May through June, 1984. Analyses varied but included priority pollutant metals and organics, PCBs, and radiological parameters, including gross alpha and gross beta radioactivities, bismuth-214, technetium-99, actinium-228, cesium-134, cesium-137, potassium-40, cobalt-60, strontium-89, strontium-90, and lead-214. In addition to fish, samples of aquatic animals, including frogs, snapping turtles, and crayfish in EFPC (EFPCM 4.0, 8.8, and 13.8), and frogs and crayfish in Bear Creek (BCM 0.4), were collected. Raw data are presented in the appendix to the TVA report. Mercury concentrations measured in fish are presented in Table A-33 (Appendix A). Maximum radionuclide concentrations are shown in Table A-34.

Study 72: Biological Monitoring and Abatement Plans for EFPC, Mitchell Branch, Clinch River, WOC, Northwest Tributary, Melton Branch, First Creek, and Fifth Creek (1985-present)

Under the modified NPDES permit for Y-12, issued on May 24, 1985, a program for biological monitoring of EFPC was required (Loar et al., 1989). Six permanent BMAP sampling sites were established on EFPC. Criteria used in the selection of these locations included: 1) previous sampling locations, 2) known or suspected sources of discharge, 3) proximity to DOE ORR boundaries, 4) concentration of mercury in adjacent floodplain, and 5) accessibility. Since the Y-12 facility is located at the headwaters of EFPC, no upstream reference site was selected. Benthic invertebrates and fishes are routinely sampled at the identified locations. Samples are analyzed for nonradiological contaminants.

Under the revised NPDES permit for the K-25 Site, revised on September 11, 1986, a program for biological monitoring of the Mitchell Branch was required (Loar et al., 1992). This permit was subsequently modified on October 1, 1992 (Shoemaker, 1993). Mitchell Branch is a second-order tributary of Poplar Creek near the northeast boundary of the K-25 Site; it receives effluent from the K-1407B holding pond and leachate from the waste disposal sites. Flow in lower Mitchell Branch is augmented by as much as 30% by discharges from the K-25 Site (Smith et al., 1988). Seven permanent sampling sites have been established on Mitchell Branch, above and below three discharge locations. Benthic invertebrates and fishes are routinely sampled for nonradiological contaminants at these locations.

Under the modified NPDES permit for X-10, issued on April 1, 1986, a program for biological monitoring of the Clinch River, WOC, Northwest Tributary of WOC, Melton Branch, Fifth Creek, and First Creek was required (Loar et al., 1991). Emphasis of this program was on evaluating the radioecology of WOC and WOL and determining the transport, distribution, and fate of contaminants in the WOCE-Clinch River-Watts Bar Reservoir system. Fifteen sampling locations for routine sampling of benthic invertebrates and fishes were established on WOC and its tributaries. These locations were: 1) point-source discharge locations, 2) area sources with confirmed seepage to surface waters, 3) known areas of impact, and 4) previous sampling locations. Reference samples were also to be collected at upstream areas and on streams outside ORR boundaries. Samples are analyzed for nonradiological contaminants.

Fish are collected and analyzed for non-radiological contaminants (e.g., PCBs) at least annually from the routine sampling locations. Data collected as part of the BMAPs are presented in annual reports for each facility and are summarized in the reservation-wide annual monitoring reports.

Study 73: Mercury in EFPC Fish, Sediments, and Water (1986)

In 1986, the TVA as part of the ORTF conducted an evaluation to predict mercury and PCB uptake by fish in EFPC (TVA, 1987). Sediment and water data collected as part of the ORTF Instream Contaminant Study were used in the evaluation. These data were collected from EFPCM 0.0 to 14.36. Raw data are tabulated in this report.

Study 74: Metals, Pesticides, and PCBs in Melton Hill and Watts Bar Reservoir Fish (1986-1989)

From 1986 to 1989, the TVA conducted tissue screening studies from fish collected at a number of locations in reservoirs on the Tennessee and Cumberland Rivers (TVA 1988; 1989; 1990; 1991a). Composite samples of fish collected from each location were analyzed for priority pollutant metals, pesticides, and PCBs. The program was conducted to monitor the general, overall level of contamination of fish and to identify problem areas where contamination levels may warrant limiting fish consumption to protect human health. Two distinct groups of studies were conducted. One program focussed on the collection of annual samples at inflow points from eight of the major tributaries into the Tennessee River, including the Clinch River. This study was conducted to identify year-to-year trends of contaminants entering the reservoir system from major watersheds. The second program focussed on the collection of samples from reservoirs throughout the Tennessee Valley, including Watts Bar and Melton Hill. Study results are tabulated in the reports by sample location, data, and analyte. All data are also stored in the USEPA STORET database system.

Study 75: PCBs in Melton Hill and Watts Bar Reservoir Fish (1987-1988)

Studies of PCB contamination of fish from east Tennessee reservoirs were conducted by the TVA for several years (Dycus, 1989; 1990). The studies were conducted to define the geographical boundaries where contamination in fish ceases to be a problem and to determine the temporal trend in PCB concentrations in fish from reservoirs where the geographical extent of contamination has been defined. Studies during 1987 and 1988 focussed on Watts Bar and Melton Hill Reservoirs. Composite tissue samples were analyzed by gas chromatography. Data are tabulated in the reports by sample date and location. Summary data for catfish collected at sampling locations in Watts Bar and Melton Hill Reservoirs during 1986, 1987, and 1988 are show in Table A-35 (Appendix A).



Study 76: Metals, PCBs, Pesticides, Semivolatiles, and Radionuclides in Clinch and Tennessee River Fish (1989)

Fish samples were collected during 1989 as part of the Clinch River Remedial Investigation to determine the range of contaminant concentrations in Clinch River/Watts Bar Reservoir fish and the spatial distribution of these concentrations in relation to the ORR; to collect data for human health and ecological risk assessments, and to establish reference (background) concentrations (Cook et al., 1992). A total of 288 fish representing three species from six sites on the Clinch River from CRM 0.0 to 51, three sites on the Tennessee River downstream of its confluence with the Clinch River from TRM 518.0 to 557.0 (Watts Bar Reservoir), five sites on tributaries of the Clinch River, including Poplar Creek from PCM 0.5 to 6.0, and one reference site (Norris Reservoir) on the Clinch River upstream of any influence of ORR, was collected. Samples were analyzed for metals, PCBs, pesticides, semivolatiles, and radionuclides, including cobalt-60, cesium-137, and strontium-90. Raw data are tabulated in the report by location and analyte. Surface water and sediment data were also collected in this study.

**5.3.5.2 Milk Monitoring Data**

Study 77: Routine Milk Sampling by ORNL Applied Health Physics Division (1961-present)

Beginning around 1961, raw milk samples were collected by the ORNL Applied Health Physics Division and analyzed for strontium-90 and iodine-131. The number of samples collected varied somewhat from year-to-year, beginning with six in 1961 and expanding to a maximum of 14 sites in 1979. One group of samples was collected outside the AEC controlled area but within a 12-mile radius of ORNL; these samples were collected to evaluate the possible effects of effluent from ORNL operations on radionuclide content in milk. A second group of samples was collected outside of the 12-mile radius but within a radius of 50-miles; these samples were collected to establish an index of background data to which the impact of releases of radionuclides from Oak Ridge could be compared. These data are summarized in the ORNL Applied Health Physics Division Annual Reports and the reservation-wide annual environmental monitoring reports.

Figures 5-12 and 5-13 indicate the local and remote milk sampling locations used in the ORNL program. Figure B-12 reflects weekly average iodine-131 concentrations in raw milk sampled from the immediate environs of the ORR during 1965 along with reported iodine-131 discharges from ORNL stacks from the same period. Figure B-13 shows the trend of strontium-90 found in milk samples for the same time period. Table A-36, excerpted from Ohnesorge (1986), summarizes strontium-90 and iodine-131 concentrations in milk measured within 25 miles of ORNL from 1961-1984.

In addition to the monitoring of raw milk, Applied Health Physics personnel also sampled and analyzed cattle thyroids collected within 100 miles of the Reservation. Counting of cattle thyroids was reported to provide a more sensitive iodine-131 detection limit than milk analyses, and the method required little in the way of laboratory preparation. Weekly analyses began around 1962, and in 1967 the program included analysis of six thyroids per week. Figure B-14 shows the trend of iodine-131 concentrations in milk and in cattle thyroids collected during 1962. Figure B-15 shows the trend of iodine-131 concentrations measured in cattle thyroids during 1965. The peak observed around June can be compared to the trends of iodine-131 discharged and measured at perimeter air stations during the same period, as shown in Figure B-16.

### 5.3.5.3 Waterfowl Monitoring Data

#### Study 78: Radioactivity in Waterfowl on WOL (1951)

A preliminary study of the accumulation and distribution of radioactivity in waterfowl was initiated by the ORNL Health Physics Division in 1951 (Eastwood et al., 1952). Coots and ducks were collected from WOL. These data were not located.

#### Study 79: Radionuclides in Waterfowl on WOL (1986-1988)

Since 1986, studies of radionuclides in waterfowl on the ORR have been conducted as part of the BMAPs. In 1986, a study of cesium-137 and strontium-90 in Canada geese residing near contaminated ponds on ORNL showed a possibility of off-site transport of radioactive materials by these birds (MMES, 1987). In 1987, a study of waterfowl use of waste disposal ponds and settling basins near ORNL was initiated as part of the ORNL BMAP (MMES, 1988). The study was initiated to characterize resident and migratory waterfowl populations that inhabit these areas and to determine the potential exposure to humans from consuming these waterfowl. Concentrations of cesium-137 and cobalt-60 were measured in seven mallard ducks and three American coots collected from WOL during 1987 and 1988. The mallards were considered to be migrants since they were observed on the lake for only a few days. The coots were considered residents since they were observed on the lake for more than three months. Summary data are presented in the reservation-wide annual environmental monitoring reports. Average concentrations of cesium-137 and cobalt-60 measured in ducks and coots during 1988 are shown in Table A-37.

#### Study 80: Radioactivity in Geese from Settling Ponds (1987-present)

Annual live whole-body counting of geese captured at ORNL in the vicinity of the 3500 area settling ponds was conducted beginning in 1987 (MMES, 1988; 1989; 1990). In 1989, breast

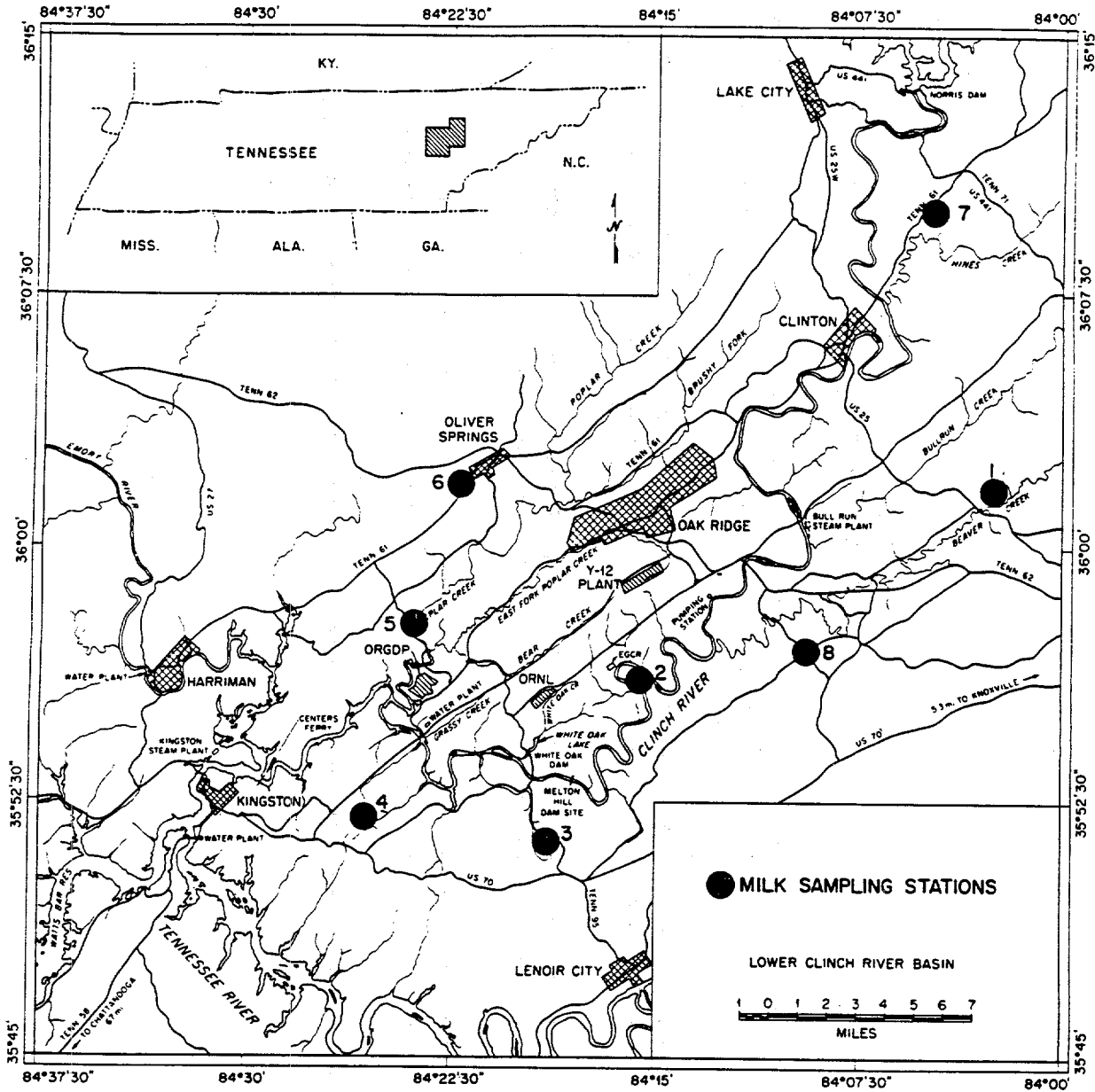
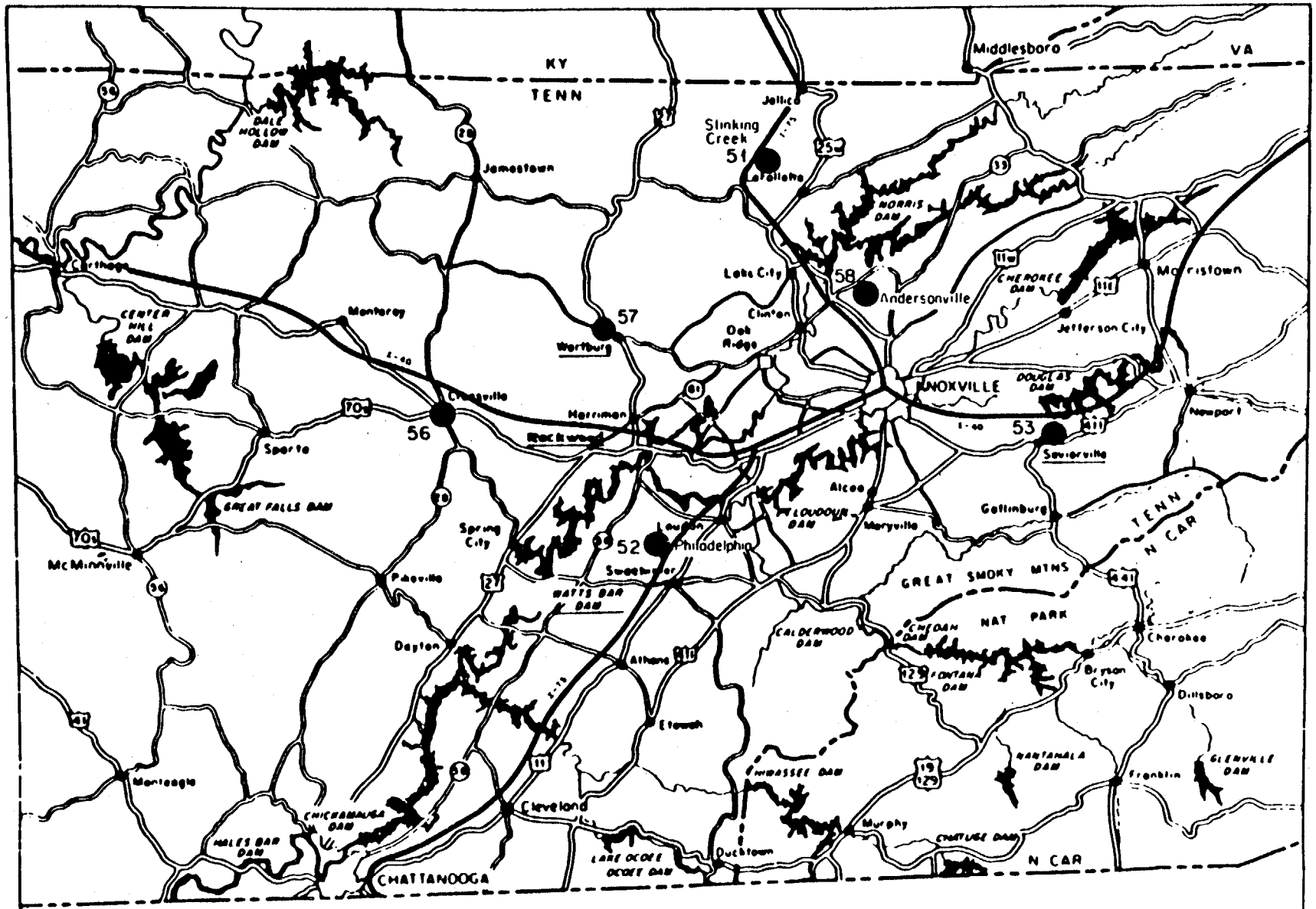


Figure 5-12 Map Showing Milk Sampling Stations in the East Tennessee.



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Figure 5-13 Remote Environs Milk Sampling Locations

tissue concentrations of cesium-137 in two geese were 150,000 and 106,000 Bq/kg ( $4.1 \times 10^{-6}$  and  $2.9 \times 10^{-6}$  Ci/kg) (MMES, 1990). Results are summarized in the reservation-wide annual environmental monitoring reports.

Study 81: Migration of Geese on Waste Disposal Ponds (1988)

A banding study of Canada geese on ORR was initiated in 1988 in cooperation with the Tennessee Wildlife Resources Agency (TWRA) (MMES, 1988). The study was conducted to determine the probability that geese inhabiting the waste disposal ponds and settling basins would be harvested by hunters.

**5.3.5.4 Deer Monitoring Data**

Study 82: Radionuclides in Deer from the ORR (1977-present)

Beginning in 1977, deer killed by automobiles on ORR were analyzed, when possible, for gamma emitters. Beginning in 1985, organized public deer hunts were conducted to reduce the size of the deer population on ORR. All animals killed during the hunts were included in a radiological survey, during which a rapid screening technique was used to measure the concentration of cesium-137 and other gamma emitting radionuclides. These data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports.

Study 83: Accumulation of Strontium-90 in Deer Consuming Browse Vegetation in Solid Waste Storage Areas (1986)

In 1986, a preliminary study was conducted by the Environmental Sciences Division of ORNL to characterize strontium-90 levels in browse vegetation in the vicinity of seeps in ORNL solid waste storage areas, where deer were suspected to accumulate strontium through ingestion of contaminated vegetation (Garten and Lomax, 1987). Strontium-90 concentrations in honeysuckle and blackberry shoots were determined. The results of the study were modeled, based on metabolic data for mule deer, to estimate the theoretical accumulation of strontium-90 in bone of whitetail deer following ingestion of contaminated vegetation. The results were extrapolated to other areas of the ORR (e.g., the WOC watershed).

Study 84: Metals in Deer from the EFPC Floodplain (mid-1980s)

As part of the ORTF studies of the contamination of the EFPC floodplain, samples of white tail deer tissue were collected to evaluate the importance of the soil-plant-animal pathway (Gist, 1987 as cited in Travis, 1989). The animals included in the study were killed in vehicle collisions on the Oak Ridge Turnpike paralleling the floodplain. Deer liver and muscle were

analyzed for metals, including arsenic, mercury, beryllium, chromium, thorium, and uranium. Data collected during this study were not located.

### 5.3.5.5 Vegetation Monitoring Data

#### Study 85: Fluoride and Radionuclides in Grass from Perimeter and Remote Monitoring Stations (1977-present)

Beginning in 1977, grass samples were collected routinely by the ORNL Applied Health Physics Division from the ORNL perimeter and remote air monitoring stations. Samples were collected either annually or semiannually. Samples were composited and analyzed by gamma spectroscopy for fluoride, uranium, plutonium, and other isotopes, including technetium-99, cesium-137, beryllium-7, strontium-90, niobium-95, zirconium-95, ruthenium-103, lanthanum-140, and cerium-144. Specific analyses varied from year to year. These data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports. Results for 1978 are summarized in Table A-38 (Appendix A).

#### Study 86: Technetium-99 in Vegetation near K-25 (1978-1979)

Vegetation and soil samples were collected in 1978 and 1979 at two locations just outside of the fenceline perimeter of the K-25 site, and analyzed for technetium-99 (Hoffman et al., 1980; Hoffman, 1982). Samples were collected to evaluate the uptake of technetium by plants and the behavior of technetium compounds in soil. Samples were also collected at the perimeter of two other DOE gaseous diffusion plants: the Paducah Gaseous Diffusion Plant in Paducah, Kentucky and the Portsmouth Gaseous Diffusion Plant in Portsmouth, Ohio. Samples were collected in the approximate predominant wind direction from estimated sources. Above-ground vegetation was collected by hand and analyzed primarily using radiochemical separations and low-level beta counting spectroscopy. Summary sampling results are tabulated in these reports. Soil sampling performed as a part of this study is described in Section 5.3.6.1 as Study 91.

#### Study 87: Mercury in Sediment, Fish, Moss, and Pasture Grass in EFPC and Bear Creek Drainages (1982)

At the request of Y-12 Plant management, the Environmental Sciences Division at ORNL conducted a study to determine the concentration of mercury in sediment, fish, moss, and pasture grass in EFPC and Bear Creek drainages and to ascertain whether mercury is still being released from Y-12 (Van Winkle et al., 1982). Surface sediment, fish, moss and liverwort, and pasture grass samples were collected along the length of EFPC (EFPCM 1.3 to 14.2) and Bear Creek (BCM 0.4 to 7.6) in April, 1982. Levels of mercury measured in pasture grass collected

from the EFPC floodplain are shown in Table A-39 (Appendix A). In addition, a sediment core was collected from New Hope Pond on May 5, 1982 to determine the historical record of mercury contamination of the pond. Results are tabulated in the report by Van Winkle et al. (1982).

Study 88: Mercury in Native Vegetation and Garden Vegetables on the EFPC Floodplain (1983-1987)

To measure the transfer of mercury from soil to vegetation in the EFPC floodplain, both native vegetation and garden vegetable samples were collected in 1983 through 1987 and analyzed for mercury as part of the ORAU environmental monitoring and surveillance studies of the Oak Ridge community (Gist, 1987 as cited in Travis, 1989). Samples of native vegetation were selected from those species of plants consumed by deer and cattle which presently or have historically grazed or browsed on the floodplain. Vegetables were planted in enclosed plots and included beets, carrots, radishes, and spinach. In addition, vegetable samples were collected from several additional gardens during the course of the community sampling by ORAU. These data are summarized in letter reports prepared by ORAU. Floodplain soil, stream sediment, turtle, and well-water samples were also collected as part of these studies.

**5.3.5.6 Beef Monitoring Data**

Study 89: Mercury in Tissues from a Cow and a Horse Grazing on EFPC Floodplain (1982)

Hair samples from a cow and a horse grazing on the EFPC floodplain and drinking out of the creek were collected in August 1982 and analyzed for mercury. These data are summarized in UCC (1983). Samples were also collected at the Comparative Animal Research Laboratory (CARL) from animals not exposed to mercury-contaminated grasses or waters. In November 1982, kidney, liver, brain, and muscle tissue samples from one of the cows grazing on the EFPC floodplain were analyzed for mercury. These data were not located.

**5.3.6 Historical Soil and Surface Radiation Monitoring Data**

Available data from analyses of contaminants in soils and from measurements of surface radiation levels are summarized in Table 5-6 which was presented earlier. Summaries of key studies of these types are presented in this section. Tables and figures showing selected examples of reported data excerpted from the original reports are presented in Appendix A and Appendix B, respectively.

### 5.3.6.1 Soil Monitoring

#### Study 90: Radionuclides in Soil at Perimeter and Remote Monitoring Stations (1971-present)

Beginning in 1971, soils were collected routinely by the ORNL Applied Health Physics Division from locations near the perimeter and remote air monitoring network stations and analyzed for plutonium, uranium, and specific radionuclides, including potassium-40, strontium-90, cesium-137, and radium-226. Specific analyses varied from year-to-year. Samples were collected annually through 1977 and semiannually thereafter. These data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports. Results for 1978 are summarized in Table A-40 (Appendix A).

#### Study 91: Technetium-99 in Soils near K-25 (1978-1979)

Vegetation and soil samples were collected in 1978 and 1979 at two locations just outside of the fenceline perimeter of the K-25 Site, and analyzed for technetium-99 (Hoffman et al., 1980; Hoffman, 1982). Samples were collected to evaluate the uptake of technetium by plants and the behavior of technetium compounds in soil. Samples were also collected at the perimeter of two other DOE gaseous diffusion plants: the Paducah Gaseous Diffusion Plant in Paducah, Kentucky and the Portsmouth Gaseous Diffusion Plant in Portsmouth, Ohio. Samples were collected in the approximate predominant wind direction from estimated sources. Large quantities of soil from 0 to 15 cm depth were collected using a shovel to obtain samples representative of the root zone. Subsequent sampling differentiated between samples collected at 0 - 2 cm and samples collected below 2 cm. Summary sampling results are tabulated.

#### Study 92: Metals, PCBs, and Radionuclides in EFPC Floodplain Soils (1983-1987)

During the ORAU environmental monitoring and surveillance studies of the Oak Ridge community, surface soil samples were collected in the EFPC floodplain and analyzed for mercury and other metals, radionuclides, and PCBs (MMES, 1985). Transects were established on the EFPC floodplain perpendicular to the creek at intervals of 100 meters (325 feet). During the initial sampling program, surface soil samples were collected every 50 meters (163 feet) along the transects. In areas with elevated mercury concentrations, more detailed sampling was conducted. Soil samples were collected in the EFPC floodplain from 1983 to 1987. Background soil samples from locations in Oklahoma, Colorado, Utah, Nevada, and New Mexico were collected in August 1984 and analyzed for mercury. Stream sediments, vegetables, turtles, and well water were also monitored as part of these studies.



### Study 93: Radiation Survey of the Oak Ridge Sewer Beltway (1984)

A radiation survey of the sewer beltway in Oak Ridge was conducted by ORAU during the spring of 1984, since contaminated soil from the EFPC floodplain had been used for fill in this area (MMES, 1985). Little documentation was available on the areas where EFPC floodplain soil had been used as fill; hence, the study was initiated to gain an understanding of the extent of the beltway contamination. The walkover survey covered the entire sewer beltway. In addition, surface soil samples were collected every 100 m (325 feet) along the beltway centerline and analyzed for mercury and uranium. During the survey, an area of elevated radioactivity was discovered on Emory Valley Road, and elevated mercury levels were measured in soil samples from the same area. The source of the radioactivity was believed to be a private business; as such, there was concern that the source was ongoing and that it was contributing to radioactivity in the sewer system.

#### 5.3.6.2 External Gamma Radiation Monitoring

*See Study 93 above, plus:*

### Study 94: External Gamma Radiation Measurements (1949-present)

Beginning around 1948, ORNL health physics personnel assigned to an "Area Monitoring" group performed periodic surveys of background radiation levels at a number of locations on and near the ORR. Figure B-17 shows the results of radiation measurements taken three feet above the ground surface at 16 standard locations on and near ORNL during 1962. Off-site readings averaged a factor of five lower than on-site ORNL measurements. Figure B-18 facilitates comparison of average radiation levels at ORNL monitoring stations with off-site locations over the period from 1950 through 1965.

Beginning in 1961, external gamma radiation background measurements were taken routinely by the ORNL Applied Health Physics Division during servicing visits to the RAM stations, using calibrated GM and scintillation-type detectors at a distance of three feet above the surface of the ground. Measurements were taken at a frequency of once every five weeks. Beginning around 1973, background measurements were taken at the PAM stations using thermoluminescent dosimeters (TLDs). Beginning in 1975, measurements were taken routinely along the bank of the Clinch River from the mouth of WOC to several hundred yards downstream, to evaluate gamma radiation levels from ORNL liquid effluent releases and "sky shine" from an experimental cesium-137 plot near the riverbank. Beginning in 1977, measurements were taken at the LAMs and at one station near Melton Hill Reservoir. Dosimeters were collected and analyzed monthly at the PAMs and Melton Hill monitoring station and semiannually at the LAM and RAM stations. These data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports.

### Study 95: Surface Radiation Exposures to Hunters on ORR

A theoretical evaluation of the impact of low level radiation exposure to a standard-person who may have hunted in one of two areas of the ORR that had previously been open for deer hunting was conducted by the Environmental Sciences Division and the Office of Environmental and Health Protection of ORNL (Lu et al., 1991). Public deer hunting began at designated areas on the ORR in 1985. Because of the potential for low level radiation exposure to the hunter, White Wing Scrap Yard and ER-11 (two radioisotope treatment plots on Chestnut Ridge) were closed to hunting in 1989 and 1990, respectively. Surface gamma measurements were taken at the two areas. Based on the calculated average exposure rate, an individual hunting in the White Wing Scrap Yard area could have received 4 mrem whole body dose equivalent, and in the ER-11, 0.2 mrem whole body dose equivalent over a six-day period. No other areas on the ORR open for hunting were identified as presenting the potential for radiation exposures to hunters above background. Results of the study are discussed in the report.

#### 5.3.6.3 Aerial Background Surveys

### Study 96: Airborne Gamma Radiation Surveys (1973-1974, 1980, 1986, 1989, and 1992)

Periodic airborne measurements of gamma radiation from natural and man-made radionuclides on or in surface terrain were conducted at the ORR and surrounding areas by EG&G Energy Measurements, as part of the nationwide Aerial Radiological Measuring System (ARMS). The ARMS is a continuing program initiated in 1958 by the AEC to monitor radiation levels in and around facilities producing, utilizing, or storing radioactive materials (Burson, 1976). Surveys at the ORR were conducted in September 1973 and November 1974 (Burson, 1976), June 1980 (Boyns, 1984), September and October 1986 (Fritzsche, 1987), September 1989 (Maurer, 1992), and April 1992. All but the 1986 survey were reservation-wide. The 1986 survey emphasized the WOC floodplain. The purpose of the surveys was to give a broad overview of the distribution of radioactivity and to identify the distribution of specific gamma-producing radioactive contaminants. Radionuclides identified included cesium-137, cobalt-60, protactinium-234, thallium-208, uranium-235, bismuth-214, and potassium-40. Results were plotted as radiation intensity isopleths superimposed on maps or photographs. The plots are presented in the reports.

### Study 97: Routine Aerial Background Surveys (1959-1968)

From 1959 through 1968, aerial background surveys were conducted by ORNL personnel over the Laboratory area and for several miles distant in the general direction of the prevailing winds, to quantify background levels of radioactivity in the event that a major release occurred. Surveys were conducted at least once each quarter, using a scintillation detector in an airplane

flying at an altitude of approximately 300 feet above ground level. These data are summarized in the ORNL Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports.

Figure 5-14 shows a typical flight pattern followed by ORNL personnel in their routine aerial background radiation surveys of the ORR area. Figure B-19 (Appendix B) presents radiation profiles of the ORNL area that were determined by aerial surveys during 1959, 1960, 1961, and 1962. While the graphs are in "arbitrary units", the most prominent areas of elevated radiation during these surveys were the ORNL stacks and burial grounds.

### 5.3.7 Historical Drinking Water/ Groundwater Data

Sources of data on drinking water and groundwater monitoring are summarized in Table 5-7 and are described in more detail in this section.

#### Study 98: Radionuclides in Water from Clinch River Water Intakes (1959-Present)

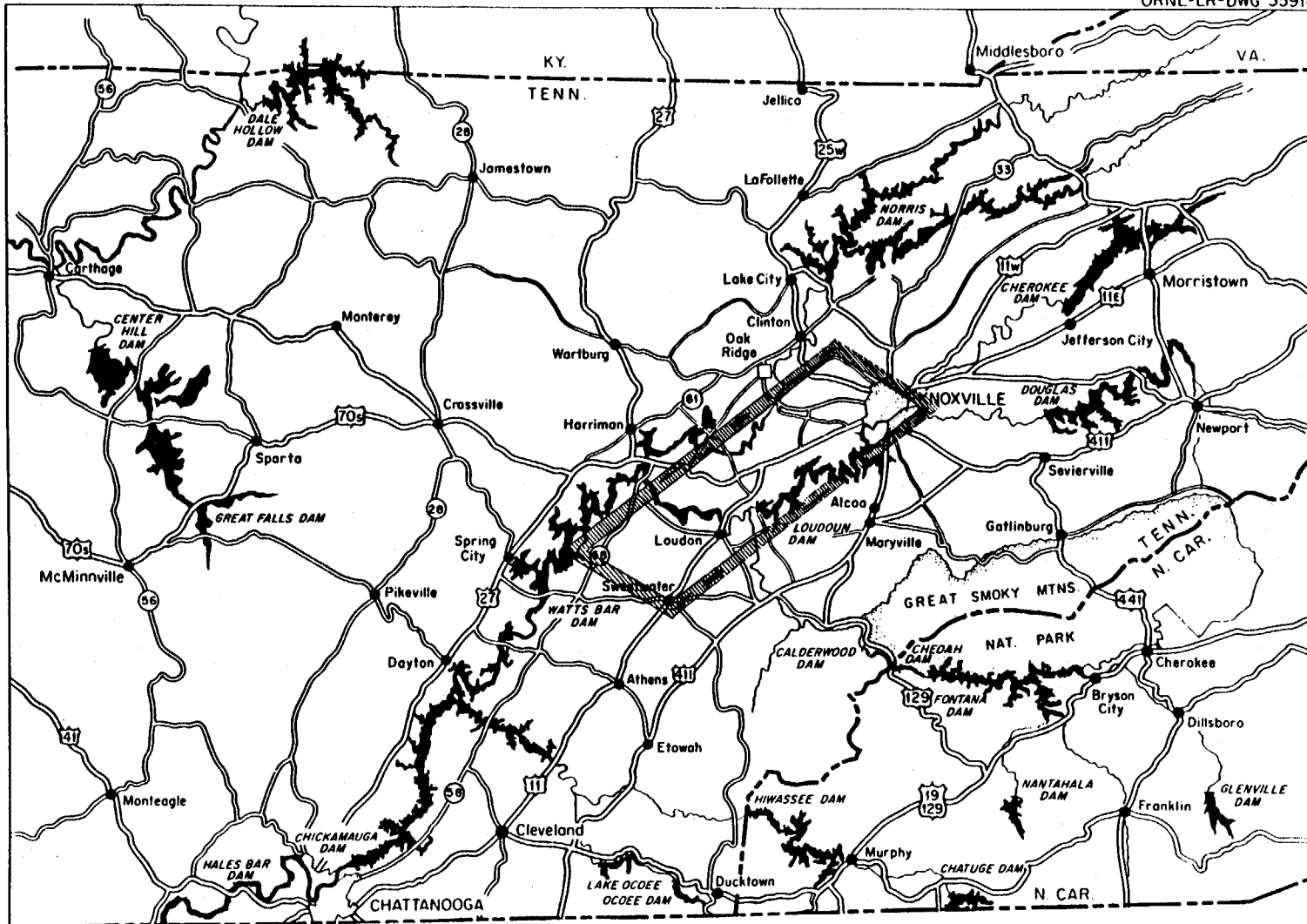
Beginning in at least 1960, water samples were collected by the ORNL Applied Health Physics Division at several water intake locations on the Clinch River. Water samples were collected at the Kingston Water Plant at CRM 4.5 beginning in at least 1959. Beginning in 1962, samples were collected at the City of Oak Ridge water intake at CRM 41.5 and beginning in at least 1964, samples were collected at the K-25 Site water intake at CRM 14.5. Water samples were analyzed for radionuclides, including strontium-90, cerium-144, cesium-137, ruthenium-103/106, cobalt-60, and zirconium-95/niobium-95. These data are summarized in the Applied Health Physics Annual Reports and the reservation-wide annual environmental monitoring reports.

#### Study 99: Radionuclides and Metals in Residential Drinking Water (1981, 1983)

Seven well water samples were collected in 1981 and two samples were collected in 1983 by ORNL's Analytical Chemistry Division (Golliher, 1988). The seven samples collected in 1981 were from the areas of Harriman, Oliver Springs, Blair, Sugar Grove, Dickey Springs, and Dyllis, to the north of the K-25 Site. Data were mostly for radionuclides, with some data on metals and physical parameters. Both sets of data are available from ORNL's Analytical Chemistry Division, (ORNL 1981; 1983) as cited in the Golliher (1988) letter.

#### Study 100: Clinch River Breeder Reactor Project Drinking Water Data

Site characterization data were collected as part of the Clinch River Breeder Reactor Project (CRBRP, 1981). This study is identified by Golliher (1988) as being a source of historical well water monitoring data; however, actual data have not been located.



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Figure 5-14 Pattern Flown with Light Aircraft During Aerial Background Survey.

Study 101: Radioactivity in Residential Drinking Water (1985)

Residential wells in the Bradbury and Poplar Springs communities, south of the K-25 Site across the Clinch River, were sampled by the TDHE in November 1985 (TDHE, 1986). Samples were analyzed for gross alpha, gross beta, and gamma activity, and tritium. Results are tabulated in the TDHE report.

Study 102: Radioactivity, Radionuclides, and Inorganics in Residential Drinking Water (1986)

An Off-Site Residential Drinking Water Sampling Program was conducted in August and September, 1986 by the Environmental Monitoring and Compliance Section of ORNL, to analyze drinking water wells at 16 residences (Murphy, 1990). Samples were collected in Oliver Springs, Harriman, Knoxville, Lenoir City, and Kingston. Samples were analyzed for inorganic water quality parameters (not including mercury) and radiological contaminants, including gross beta and gross alpha radioactivities, cesium-137, strontium-90, and tritium. Results are tabulated in the report.

Study 103: Metals, Organics, and Radionuclides in Residential Drinking Water (1986)

Well water in the communities of Kingston and Harriman was sampled by the ORAU in 1986 (Gist, 1986). Samples were analyzed primarily for strontium-90 and mercury, though some analyses were conducted for additional radiological parameters, including tritium, gross alpha, and gross beta, PCBs, and metals. Results are tabulated in the report.

Study 104: Metals, Organics, and Radionuclides in Drinking Water (1989-present)

Routine sampling of off-site drinking water was begun in 1989 by ORNL (MMES, 1991). Areas sampled include the K-25 Plant drinking water intake on the Clinch River, the city of Kingston drinking water intake on Watts Bar Lake, and the city Oliver Springs drinking water source from Bacon Springs. Samples were analyzed for volatile organics, metals, fluoride, sulfate, nitrate, nitrite, uranium, gross alpha, gross beta, tritium, strontium-90, technetium-99, and cobalt-60. Non-location specific summaries of the data collected are presented in the reservation-wide annual monitoring reports. The summary for 1990 is presented in Table A-41.

Study 105: Routine Groundwater Monitoring

Groundwater sampling is a relatively recent activity at the ORR. Most sampling activities date from the 1980s with only limited sampling identified prior to that time. The groundwater monitoring program at Y-12 was initiated in 1975 at 17 wells in Bear Creek Valley. The groundwater monitoring programs were expanded in 1983 in response to increased requirements of the Resource Conservation and Recovery Act (RCRA) regulations which is the same year that

sampling results were reported in the annual reports. The expanded monitoring at Y-12 included initiation of studies by outside contractors (Law Engineering and Bechtel), installation of new wells, and an increase in the number of parameters analyzed. Included in this expansion was the installation of wells to characterize subsurface mercury. In 1983, 29 wells were in use at Y-12. These wells were located in the vicinity of the disposal facilities. Samples were collected quarterly from all wells. Analyses included metals, gross radioactivity, organics, water quality parameters, and specific radionuclides. Additional wells were installed during 1985 in Bear Creek Valley and on Chestnut Ridge.

Groundwater monitoring wells were installed at the K-25 Site in 1985 (DOE, 1989). Monitoring was conducted at the classified burial ground and the K-1407-C holding pond. Twenty-one wells were monitored at the K-25 Site for inorganic and organic constituents as of 1983. However, as stated in the 1985 annual environmental monitoring report (MMES, 1986), these wells were of "questionable reliability" and their use was discontinued in 1985. A new monitoring program was developed at the K-25 Site under the K-25 Environmental Management Staff. This program became operational in 1987.

Twenty-two RCRA water quality wells were installed at ORNL surface impoundments in 1985 (MMES, 1986). These wells are used for site characterization and compliance purposes. Samples from these wells are monitored for metals, organics, and radionuclides. Older monitoring wells are located at SWSA 4, 5, 6, and the trench areas. The date of installation of these wells was not determined. These wells are monitored for radioactivity and radionuclides. Wells are selected from this group of approximately 100 wells for quarterly or semiannually monitoring, based on studies conducted by the Environmental Sciences Division and surface water flow patterns. These wells are monitored for radioactivity and radionuclides. In addition, samples from a portion of the wells are routinely analyzed for extensive chemical analyses.

Study 106: East Fork Poplar Creek - Sewer Line Beltway Remedial Investigation (1990-1992)

From 1990-1992, a remedial investigation (RI) of EFPC and the sewerline beltway was conducted (Science Applications International Corp, 1993). This RI integrated the requirements of CERCLA, Sect. 3004(v) of the Resource Conservation and Recovery Act of 1980 (RCRA) as amended by the Hazardous and Solid Waste Amendments of 1984, and the National Environmental Policy Act (NEPA). As part of the Remedial Investigation - RCRA facility investigation (RI-RFI), groundwater, surface water, creek sediment, and surface and subsurface soil samples were collected in EFPC and the EFPC floodplain. Samples were collected to determine the nature of contamination and identify chemicals of concern, including radionuclides, organic compounds, metals, PCBs, and pesticides.

In Phase Ia and Ib of the investigation, a total of 4014 field samples and 709 quality control (QC) samples were collected. In addition, as part of the RI-RFI, baseline human health and ecological risk assessments were completed, to determine whether current and future exposure potential presents an "imminent and substantial" endangerment to human health and the environment and to evaluate the need for site remediation (Science Applications International Corp., 1993). The results of these investigations are presented in the RI report.

Based on these investigations, among heavy metals, mercury contributed 85% of the total toxicity. Among the radionuclides, total uranium (uranium-234, uranium-235, and uranium-238) accounted for 98% of the total activity. Polycyclic aromatic hydrocarbons (PAHs) and PCBs were the primary organic compounds of interest.

## **5.4 AREAS OF OFF-SITE CONTAMINATION**

In addition to those areas contaminated by airborne or surface water releases associated with historical operations at the three ORR facilities, several sites located off the ORR have been discovered to be contaminated. These sites apparently became contaminated as a result of past operations involving handling or processing of materials contaminated with radioactivity and other hazardous materials, reported to have originated in part from various AEC/ERDA/DOE sites and from civilian and defense-related activities. Summaries of available information concerning these areas of off-site contamination near the ORR are provided in this section.

### **5.4.1 The Elza Gate Site**

The Elza Gate site is located at the eastern end of the City of Oak Ridge in what is currently known as the Melton Lake Industrial Park (Bechtel, 1991). Access to the site is unrestricted. None of the original Manhattan Engineer District (MED) warehouses remain, although the pads on which the warehouses were built are still in place.

In the early 1940's, the site was developed by the MED as a storage area for pitchblende, a high-grade uranium ore from Africa, and for ore processing residues (Bechtel, 1991). Radioactive materials were stored in three of the five warehouses on site. In 1946, the site was transferred to the Atomic Energy Commission (AEC) and, upon cessation of use of the warehouses for storage of pitchblende around 1950, was used to store equipment and materials from ORNL and Y-12. The site was used by the AEC until 1970, when it was vacated. Prior to transfer of the site to the city of Oak Ridge, a radiation survey and clean-up project was performed in 1971 to remove uranium contaminated soils and decontaminate the floors in the warehouses. Following decontamination, the site was determined to be usable with no radiological restrictions (Bechtel, 1991). In 1972, the site was transferred to the City of Oak Ridge. Subsequently, the property was sold to Jet Air, Inc., which operated a fabricating and metal plating facility on the site.

In 1987, a survey of the site was conducted by ORAU at the request of the TDHE, because of the possibility of contamination from the metal plating facility (Bechtel, 1991). Soil samples were collected and analyzed for uranium, metals, and PCBs. Elevated levels of PCBs were found during the investigations; these were attributed to the storage of PCB-contaminated electrical equipment on site, including electrical equipment from the stripping of the Calutron Electromagnetic Separation processes at Y-12, and other miscellaneous equipment. Electrical equipment from the Calutrons consisted of several hundred electrical "cubicles" containing sealed capacitors filled with pyranol, a dielectric fluid which contains PCBs. Spills and leaking may have occurred. The survey also identified elevated levels of uranium in the southern portion of the site (Bechtel, 1991).

In 1988, the site was sold to MECO, a development company. At the request of DOE, a preliminary radiological survey was conducted by ORNL in October and November, 1988 (Bechtel, 1991). Levels of residual radioactivity exceeding the DOE cleanup criteria were identified in each of the four parcels evaluated. On November 30, 1988, the entire industrial park was designated for inclusion in the DOE Formerly Utilized Sites Remedial Action Program (FUSRAP).

To support a remedial action alternative, a radiological characterization was conducted in 1989 (Bechtel, 1991). The characterization consisted of a near-surface walkover gamma scan, soil sampling and analysis, and gamma logging of boreholes. Data were collected for PCBs and radionuclides. In 1990, samples were collected and analyzed for PCBs, metals, and organics. A hydrogeologic investigation was also conducted. Data from these studies are tabulated (Bechtel, 1991).

#### **5.4.2 David Witherspoon, Inc. Scrap Metal Yards**

David Witherspoon, Inc. was established in 1948. The Witherspoon property consists of three parcels of land in South Knoxville: the Candora site, also known as Witherspoon Recycling, at 901 Maryville Pike; the Screen Art site (part of which is referred to as the "hot field") at 1630 Maryville Pike; and the David Witherspoon landfill to the rear of the Screen Arts site. All three sites were used for the processing of scrap metals.

During the 1950s, 1960s, and 1970s, tons of scrap metal contaminated with radionuclides and chemicals were handled at the sites. The facility was authorized to receive these materials under a license from the AEC. In 1966, the license from AEC was terminated and the State of Tennessee became the licensing authority. The license authorized the processing of 5000 tons of scrap metal with uranium surface contamination not to exceed 0.1% by weight. Much of the metal was from DOE facilities, including ORNL.



Sales of scrap metal from DOE contractors to David Witherspoon, Inc. continued until at least April, 1984 (according to 1990 DOE memo addressed to G. Wilson Horde, Jr., General Council of Martin Marietta Energy Systems, Inc.). During this time, the TDRH exercised oversight over operations at the facility, and mandated routine cleanup activities. In the early 1980s, a partial cleanup was conducted at the Screen Art site. Soil contaminated with PCBs and uranium was removed from the surface, placed in barrels, and moved to the Candora site. Following cleanup, the Screen Art site was covered with a clay cap. In 1986, the barrels and several hundred tons of scrap metal were removed from the Candora site back to the ORR.

On September 27, 1989, an aerial radiological survey of the Witherspoon sites was conducted by EG&G Energy Measurements, Inc., as part of the Aerial Measuring System survey conducted that year for the Oak Ridge area (Maurer, 1992). A 2-square mile area in south Knoxville was surveyed at an altitude of 125 feet to maximize the AMS sensitivity for the low specific activity of the radioactive materials. Results were plotted as a terrestrial gamma exposure rate contour map overlaid on a USGS topographic map. The map is presented in a report by Maurer (1992).

Samples at all three sites in 1990 revealed high levels of PCBs and heavy metals in soils at all three sites and PAHs at high levels at the Screen Arts site. As a result of this sampling, all three sites were placed on the Tennessee Superfund list on Dec. 4, 1990. Following this date, TDRH's oversight authority at the sites was terminated.

Phase I Remedial Investigations, including collection of soil and groundwater data, were conducted at the three sites in 1991 and 1992. These data are currently being evaluated by the USEPA. Extensive data from investigations of all three sites are on file at the Knoxville office of the TDEC.

#### 5.4.3 Dupont-Smith

Dupont-Smith is a scrap-yard on Melton Lake Drive in Oak Ridge. In the late-1960s, Dupont-Smith was issued a license by the State of Tennessee to receive material with a specified amount of surface contamination, including radioactivity. Prior to issuing of the license, it is not believed that any contaminated materials were stored at the site (Mobley, 1993).

The facility operated as a recycling facility for scrap metals. As a consequence of these activities, site soils reportedly became contaminated with uranium and mercury (Mobley, 1993). Scrap metal with radioactive contamination is reported to be buried on the site. Currently, the junkyard is no longer active.

Remedial investigations are currently underway at the site. DOE has agreed to clean up the site since all of the materials on the site can be traced to DOE activities (Mobley, 1993). Samples have been collected to assess the level of contamination in soils. Future plans for site cleanup

by DOE include removal of scrap metal and other contamination from the surface, followed by soil remediation during Phase 2. (Andel, 1993)

Sampling data from the site are available at the Knoxville office of the TDEC (Andel, 1993).

#### **5.4.4 Cesium-137 Contamination Along the CSX Transportation Group Railroad Tracks**

A preliminary dose assessment of radiation exposure to members of the public due to cesium-137 contamination along the CSX Transportation Group railroad tracks in Oak Ridge was conducted in 1990 (Kocher, 1990). The evaluation used data collected in three previous studies by ORNL and ORAU. The primary exposure pathway was identified as direct exposure to gamma radiation. No significant risk to the public or railroad workers due to cesium-137 exposure was indicated.

The contaminated railroad beds are located east of Scarboro Road and across from the Y-12 Plant. The area is outside of fenced areas of the Oak Ridge Utility District and Y-12 Plant, and as of 1991, was unrestricted to the public (Kocher, 1990; Lingle et al., 1991). The source of the cesium-137 contamination along the railroad tracks is somewhat uncertain, although it is thought to have originated in the early 1960s when Oak Ridge was the eastern regional burial ground for low-level radioactive waste generated by civilian and defense activities (Kocher, 1990). Waste was routinely shipped to Oak Ridge by train along these tracks. The source of contamination has been tentatively identified as leaking concrete casks containing cesium-137-bearing animal carcasses.

A preliminary radiological survey of the railroad bed was conducted in June 1986 by the ORNL Radiological Survey Activities Group (Kocher, 1990). The survey focussed only on a portion of the CSX Transportation Group railroad tracks in Oak Ridge east of Scarboro Road and across from the Y-12 Plant. Gamma exposure rates at the surface of the railroad bed and cesium-137 concentrations in ballast materials of the bed were measured. Results indicated that localized portions of the railroad bed contained elevated levels of cesium-137.

A more comprehensive survey of the railroad beds on the east and west sides of Scarboro Road near Y-12 was conducted in November 1986 by the Radiological Site Assessment Program at ORAU (ORAU, 1987; Kocher, 1990). Analyses were the same as in the June 1986 study, with the addition of measurements of activity in water and sediments of the stream crossed by the railroad tracks. The results showed elevated levels of cesium-137 on both sides of Scarboro Road, while stream water and sediment samples were typical of background for the Oak Ridge area.

A survey by ORNL's Pollutant Assessment Group from Grand Junction, CO in March and June 1990 investigated the portion of the railroad bed between the northernmost point of the previous surveys and the east end of the City of Oak Ridge (Roemer, 1990; Kocher, 1990). Results showed elevated cesium-137 in portions of the railroad bed, primarily in the warehouse district of east Oak Ridge. Adjacent residential properties were also surveyed; these showed no elevated levels of cesium-137.

## 5.5 SUMMARY AND CONCLUSIONS

The following generalizations can be made regarding the availability of environmental monitoring data for the time periods and geographical areas of interest:

### Surface Water Monitoring

- Routine surface water monitoring data for radioactivity and specific radionuclides have been located for all of the identified surface water systems of interest since the mid-1950s. Prior to this time, data on radiological parameters are sporadic.
- Samples for mercury were routinely collected in EFPC and Bear Creek near the Y-12 facility beginning in the early 1950s. In Poplar Creek, EFPC, and the Clinch River, metals data were collected during several short-term studies beginning as early as 1955. Routine sampling data for metals in Poplar Creek and the Clinch River have been located beginning in 1971.
- PCBs were monitored in Bear Creek at the NPDES discharge point beginning in 1971. Additional surface water data for organics are generally limited to studies of short duration.

### Sediment Sampling

- Sediment sampling data for radiological parameters in the White Oak Creek drainage system and the Clinch and Tennessee Rivers have been located since the mid-1940s. With the exception of the annual surveys of the Clinch River from 1951 through 1966, these data were not routinely collected as part of any single program. However, a succession of periodic, short-term studies was conducted. Data on radiological contaminants in sediments for other surface water systems of interest, including Bear Creek, EFPC, and Poplar Creek, were not identified for periods prior to the 1980s.

- Data for metals in the sediments of the Clinch River are available beginning in the late 1950s. No routine studies were conducted until the mid-1980s. Mercury data were collected in EFPC, Poplar Creek, and Bear Creek beginning in the early 1970s. Beginning in 1975, metals data, including uranium, mercury, lead, nickel, and chromium, have been collected in Poplar Creek, the Clinch River, and EFPC near its confluence with Poplar Creek. Prior to the 1980s, no data have been located for metals other than mercury in Bear Creek and in EFPC near the Y-12 Plant.
- PCBs data have been located for the Clinch River, Poplar Creek, and EFPC since as early as 1973; no other organics data have been located until the 1980s.

#### Ambient Air Monitoring

- All off-site ambient air monitoring data which were located have been collected as part of the routine environmental monitoring program.
- Data have been collected for radiological parameters at stations in the perimeter and remote air monitoring network since at least the late 1950s. Data have been collected in the area in the immediate vicinity of ORNL since at least the late 1940s.
- Data for selected metals in ambient air in the immediate vicinity of the K-25 Site are available since at least the mid-1950s. Data for mercury in ambient air in the vicinity of the Y-12 Site are available since at least the mid-1980s.

#### Fish Monitoring

- A large number of studies have been conducted to evaluate radionuclide concentrations in fish in WOC and the Clinch River; these data are available since as early as the 1940s. No radiological data were identified for Bear Creek, EFPC, or Poplar Creek until the 1980s.
- Mercury data and PCBs data are available for Clinch River and Poplar Creek fish since the mid-1970s. No metals or organics data were identified for Bear Creek or EFPC until the 1980s.

#### Biological Monitoring (Other than Fish)

- Cow's milk was routinely analyzed for iodine-131 and strontium-90 since the mid-1960s. Cattle thyroids were routinely monitored for iodine-131 from the mid-1960s to the mid-1970s.

- Waterfowl have been monitored for radionuclides in a number of studies since the mid-1980s, although data were collected in one study as early as 1950.
- Samples of deer tissue have been measured for radioactivity since the late 1970s. Deer tissue was collected and analyzed for metals, including mercury, during the mid-1980s.
- Sampling of beef tissue is limited to analysis of mercury concentrations in a cow grazing on the EFPC floodplain during 1982.
- Grass samples were routinely collected at stations in the perimeter and remote air monitoring network analyzed for radionuclides since the mid-1970s.
- Pasture grass, native vegetation, and garden vegetables were collected from the EFPC floodplain and analyzed for mercury during the mid-1980s.

#### Soil Monitoring

- Soil monitoring data for off-site areas are limited. Routine soil monitoring data for radiological parameters were collected at the perimeter and remote air monitoring stations beginning in the early 1970s.
- No other off-site soil monitoring data were located with the exception of data for mercury, PCBs, and radionuclides collected in the EFPC floodplain and the Oak Ridge Sewer Beltway during a four-year period in the mid-1980s.

#### Surface Radiation Measurements

- Surface gamma radiation has been routinely measured at locations on or near the ORR since around 1948. In addition, a surface radiation survey of the Oak Ridge Sewer Beltway was conducted in 1984.
- Aerial surveys of background radiation were conducted periodically by ORNL during a ten-year period beginning in 1959.
- Aerial gamma radiation surveys of the ORR and surrounding areas have been conducted periodically since 1973.

### Drinking Water Data

- Water from drinking water intakes on the Clinch River, including the Kingston, K-25 Site, and City of Oak Ridge drinking water intakes, has been routinely monitored for radiological parameters since at least the early 1960s.
- Sampling of off-site drinking water wells is largely limited to short-term studies during the 1980s. Routine monitoring of off-site drinking water wells was apparently not initiated until the late 1980s.
- Biological monitoring data were collected for a number of media other than aquatic biota; however, these data are in general rather limited. Routine monitoring data include analyses of cows' milk for iodine-131 and strontium-90 since the mid-1960s and cattle thyroids for iodine-131 from the mid-1960s to the mid-1970s. Grass samples were routinely collected at stations in the perimeter and remote air monitoring network since the mid-1970s and analyzed for radionuclides. Waterfowl have been monitored for radionuclides in a number of studies since the mid-1980s, though data are available for one study as early as 1950. Deer have been measured for radioactivity since the late 1970s, and for metals, including mercury, during the mid-1980s. Pasture grass, native vegetation, and garden vegetables were collected from the EFPC floodplain and analyzed for mercury during the mid-1980s.

A large number of documents and information sources describing environmental monitoring data collected in off-site areas potentially affected by releases from Oak Ridge Reservation facilities have been reviewed. Those studies and information sources considered to be most useful for evaluation of off-site exposures have been summarized in this report. Although this has not been an exhaustive review, it is believed that the majority of relevant studies have been identified and information sources have been characterized in a manner that will support conduct of the feasibility study and focussing of any further quantitative evaluation of historical doses and health risks.

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**APPENDIX A**  
**ENVIRONMENTAL MONITORING DATA:**  
**EXAMPLE TABLES**

**IMPORTANT NOTICE**

Appendices A and B present examples of environmental monitoring data directly excerpted from the cited source documents. As such, the quality of the data summarized has not been verified, and no attempt has been made to alter the format in which the data are presented. In a number of cases, these sample tables and figures contain data that appear to be contrary to typical or expected distributions of radionuclides around nuclear facilities, expected relationships between specific radionuclides and/or gross radioactivity measurements, and customary reporting conventions for environmental data. These examples of environmental monitoring data serve to illustrate many of the aspects of environmental contaminant sampling, measurement, and reporting that will require much closer evaluation before historical environmental data can be used as a basis for estimation of off-site exposures or health risks. These tables and figures do not necessarily represent all of the data collected in a given study during a given time period. Study numbers, which refer to text summaries in Section 5.3 that can be consulted for more information, are provided for each table/figure in the attached list.

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Table A-1

Total activity of White Oak drainage system, 1945-46

Location	Distance below settling pond [m (ft)]	1945		1946	
		Average [MBq/m <sup>2</sup> (μCi/ft <sup>2</sup> )]	Total [TBq (Ci)]	Average [MBq/m <sup>2</sup> (μCi/ft <sup>2</sup> )]	Total [TBq (Ci)]
Marsh	646 ( 2,120)	36.4 (91.5 )	1.58 (42.7)	58.0 (145.5)	2.51 (67.8)
Intermediate pond	991 ( 3,250)	34.9 (87.6 )	0.17 ( 4.6)		
W.O. Lake mud flats	2103 ( 6,900)	8.8 (22.0 )	0.55 (14.9)	3.8 ( 9.6)	0.24 ( 6.5)
W. O. Lake	2621 ( 8,600)	3.4 ( 8.5 )	0.25 ( 6.8)	6.8 ( 17.1)	0.5 (13.6)
Area below spillway	3155 (10,350)	0.41 ( 1.04)	0.01 ( 0.3)	1.1 ( 2.7)	0.03 ( 0.9)
<b>Total</b>			<b>2.56 (69.3)</b>		<b>3.28 (88.8)</b>

Source: Cheka and Morgan, 1947.

Table A-2  
Water Survey Program  
October and November 1947

<u>Plot Plan Reference No.</u>	<u>Location</u>	<u>Date Sampled</u>	<u>Uranium Conc. P.P.P.</u>	<u>Fluoride Conc. P.P.M.</u>	<u>Activity Alpha Counts per Min. per 100 ml.</u>
13 D	Clinch River, effluent from K-25	10- 2-47	0	---	-
		10- 9-47	0	0.2	Less than 0.5
		10-10-47	0	0.1	1
		10-13-47	0	0.4	Less than 0.5
		10-14-47	0	0.1	1
		10-15-47	2	0.1	Less than 0.5
		10-16-47	0	No analysis	1
		10-17-47	0	No analysis	2
		10-20-47	0	0.5	2
		10-21-47	0	0.5	1
		10-22-47	0	Less than 0.2	1
		10-23-47	0	Less than 0.2	1
		10-24-47	0	0.2	1
		10-27-47	0	Less than 0.2	Less than 0.5
		10-28-47	0	Less than 0.2	Less than 0.5
		10-29-47	0	Less than 0.2	1
		10-30-47	0	Less than 0.2	2
		10-31-47	0	Less than 0.2	1
		11- 3-47	0	Less than 0.2	2
		11- 4-47	0	Less than 0.2	1
		11- 5-47	0	Less than 0.2	Less than 0.5
		11- 6-47	0	Less than 0.2	1
		11- 7-47	0	0.2	1
		11-10-47	0	0.3	1
		11-11-47	0	Less than 0.2	3
		11-12-47	0	Less than 0.2	2
		11-13-47	0	Less than 0.2	1
		11-14-47	0	Less than 0.2	2
		11-17-47	0	Less than 0.2	1

Source: Ketcham (1947a)



Table A-3

RADIONUCLIDES IN RIVER SILT - 1959

UNITS OF  $10^{-6}$   $\mu\text{c/g}$  OF DRIED MUD

Sample Location	Cs (as Cs-Ba <sup>137</sup> )	Ce (as Ce-Pr <sup>144</sup> )	Sr (as Sr <sup>90</sup> )	Co (as Co <sup>60</sup> )	Ru (as Ru-Rh <sup>106</sup> )	Nb (as Nb <sup>95</sup> )	Zr (as Zr <sup>95</sup> )	TRE*	
Clinch R. M	21.5	4.5	6.8	0.5	0.9	4.5	2.7	5.3	
	19.1	527.0	92.3	9.5	69.4	18.0	3.2	251.1	
	16.3	464.0	70.7	9.0	53.2	16.7	2.3	151.5	
	15.2	391.0	54.1	7.2	41.0	18.0	2.3	141.9	
	14.0	464.0	65.3	8.6	52.7	17.1	3.2	148.8	
	11.0	228.8	27.0	5.4	26.1	9.9	3.2	57.8	
	8.0	236.5	29.7	5.9	27.9	11.3	3.6	71.3	
	5.8	207.2	20.3	4.5	29.3	7.7	2.7	56.5	
	4.7	168.9	17.6	4.5	25.7	7.2	3.6	32.9	
	2.6	171.2	9.0	1.4	18.5	6.3	2.3	19.3	
	1.1	258.6	20.3	2.7	20.0	9.9	2.3	40.3	
Av.	283.8	37.5	5.4	33.2	11.5	2.9	2.1	88.8	
Tenn. R. M	570.8	1.8	2.3	0.5	0.9	3.6	2.7	1.4	1.7
	562.7	40.5	7.7	0.9	6.8	6.3	3.6	1.8	8.5
	552.7	40.5	5.0	1.4	8.1	5.0	2.7	0.9	7.1
	543.8	23.4	2.7	0.9	5.0	3.6	2.7	0.9	5.3
	532.0	25.7	5.4	1.4	3.6	3.1	4.5	0.9	6.7
	509.9	6.8	**	0.5	**	**	**	**	8.0
	491.9	13.5	3.2	0.9	3.6	3.6	3.2	1.8	2.9
	475.1	10.8	4.5	0.9	2.7	5.4	2.7	1.4	5.6
	434.1	12.6	9.0	0.9	2.7	8.6	3.6	0.9	7.1
	381.2	3.2	1.8	0.5	1.4	2.7	2.7	0.9	0.2
	354.4	3.6	3.6	0.5	1.8	4.1	3.6	0.9	2.5
Av.	16.6	4.5	0.8	3.7	4.6	3.2	1.1	5.1	

\* Tl<sup>204</sup> was used as a reference standard for tri-valent rare earth fraction.

\*\* Insufficient sample for complete analyses.

**Table A-4**

RADIONUCLIDES IN CLINCH RIVER SILT - 1962-1963  
(Units of  $10^{-6}$   $\mu\text{c/g}$  of Dried Mud)

Location	Cs <sup>137</sup>		Ce <sup>144</sup>		Sr <sup>90</sup>		Co <sup>60</sup>		Ru <sup>103-106</sup>		Zr <sup>95</sup> +Nb <sup>95</sup>		TRE <sup>a</sup> + Y <sup>90</sup> (as Y <sup>90</sup> )	
	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963
CRM 42.8		2.6		17		0.29		*		14		14		15
39.1		1.3		7.7		0.49		*		6.4		6.3		6.3
34.7		2.6		19		0.34		*		14		16		16
31.1		2.7		16		0.36		*		13		12		9
29.0		2.7		16		0.52		*		12		13		9
27.0		3.0		22		0.50		*		16		19		15
24.9		3.0		20		0.52		*		15		17		14
23.4		0.90		3.8		0.43		*		2.8		3.4		3.3
Average		2.4		14		0.43				12		13		11
CRM 21.5	3.2	2.7	11	0.43	0.36	0.63	*	*	11	0.45	16	0.14	10	0.89
19.1	5.2	2.9	3.8	0.90	0.41	0.74	0.72	1.9	6.1	4.4	6.2	0.90	3.5	5.2
16.3	58	218	5.2	4.2	0.72	3.1	8.1	16	50	17	3.9	2.2	14	34
15.2	55	16	5.2	3.5	0.90	0.81	7.3	2.8	46	13	4.2	3.8	13	5.6
14.0	237	150	6.2	5.0	1.8	1.7	20	12	43	29	3.6	3.4	31	23
11.0	63	75	6.9	8.8	1.0	1.2	8.6	8.0	68	35	5.4	7.4	16	22
8.0	59	62	8.5	8.8	1.0	0.90	8.6	8.9	70	48	5.4	6.9	18	16
5.8	94	67	8.4	12	1.6	1.3	12	9.5	68	49	6.5	10	22	19
4.7	86	53	9.5	11	1.2	1.4	14	8.5	86	44	6.0	9.7	22	17
2.6	73	63	7.7	6.7	0.72	0.81	10	7.7	77	26	5.6	5.4	16	17
1.1	56	68	13	17	0.72	1.4	9.0	8.5	76	50	11	17	16	23
Average	72	71	7.7	7.1	0.95	1.3	9.8	8.4	55	55	6.7	6.1	17	17

<sup>a</sup>Total Rare Earths minus cerium

\*None detected

Source: ORNL (1964)

Table A-5

RADIONUCLIDES IN TENNESSE RIVER SILT - 1962-1963  
(Units of  $10^{-6}$   $\mu\text{c/g}$  of Dried Mud)

Location	$\text{Cs}^{137}$		$\text{Ce}^{144}$		$\text{Sr}^{90}$		$\text{Co}^{60}$		$\text{Ru}^{103-106}$		$\text{Zr}^{95} + \text{Nb}^{95}$		$\text{TRE}^a + \text{Y}^{90}$ (as $\text{Y}^{90}$ )	
	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963	1962	1963
TRM 570.8	1.4	1.8	3.9	7.2	0.45	0.61	*	0.45	2.1	7.5	1.2	6.3	3.0	4.1
562.7	23	18	4.3	12	0.45	0.77	3.0	2.7	18	21	2.1	9.8	5.0	9.2
552.7	29	26	5.1	14	0.77	0.65	3.8	3.4	18	26	2.2	12	6.0	13
543.8	33	14	6.4	11	0.32	0.52	5.3	2.3	27	14	2.8	11	6.7	11
532.0	29	13	5.9	10	0.45	0.56	4.2	2.3	24	13	2.9	9.5	4.6	8.8
491.9	13	10	3.6	10	0.36	0.52	2.0	1.8	14	10	2.3	8.1	3.5	9.8
475.1	9.9	9.9	2.7	11	0.36	0.74	1.7	1.8	9.9	9.9	1.8	9.0	1.7	8.9
434.1	9.5	8.1	4.2	20	0.32	0.23	1.6	0.90	15	8.1	2.4	22	3.1	19
381.2	5.3	5.9	1.8	9.1	0.18	0.61	0.90	0.90	6.4	5.9	0.60	9.0	0.80	8.4
354.4	4.9	5.0	2.7	13	0.27	1.1	0.95	0.90	11	5.0	1.4	11	2.6	12
Average	16	10	4.1	12	0.39	0.63	2.3	1.7	14	12	2.0	11	3.7	10
Fort Loudoun Background Data														
TRM 604.4	1.8	2.2	4.8	10	*	0.61	*	*	5.1	9.4	3.0	6.8	2.7	5.9
615.8	--	2.1	--	9.5	--	0.54	--	*	--	8.5	--	5.5	--	9.5
Average	1.8	2.2	4.8	9.8		0.58			5.1	9.0	3.0	6.2	2.7	7.2

<sup>a</sup>TRE - total rare earths minus cerium

\* None detected

--No samples taken in 1962

**Table A-6**

**RADIONUCLIDES IN CLINCH AND TENNESSEE RIVER SILT - 1964-1965**  
(Units of  $10^{-8}$   $\mu\text{c/g}$  of Dried Silt)

Location	$^{137}\text{Cs}$		$^{144}\text{Ce}$		$^{90}\text{Sr}$		$^{60}\text{Co}$		$^{103-106}\text{Ru}$		$^{95}\text{Zr} + ^{95}\text{Nb}$		$\text{TRE}^{\text{a}} + ^{90}\text{Y}$ (as $^{90}\text{Y}$ )		
	1964	1965	1964	1965	1964	1965	1964	1965	1964	1965	1964	1965	1964	1965	
CRM	42.8	8.7	3.4	21	2.0	0.45	0.29	*	0.17	16	6.0	*	0.39	9.0	6.0
	34.7	7.5	5.4	19	3.4	0.72	0.25	*	0.17	16	8.1	*	0.25	7.2	9.5
	31.1	6.7	5.8	18	3.4	0.50	0.41	*	*	13	10	*	0.26	5.0	12
	24.9	5.1	5.0	14	2.6	0.45	0.23	*	*	9.9	5.6	*	0.08	4.5	8.3
	Average	7.0	4.9	18	2.9	0.53	0.30		0.08	13.7	7.5		0.25	6.4	8.9
CRM	21.5	0.68	0.62	1.4	0.57	0.29	0.38	0.36	0.23	0.99	0.91	*	0.15	*	*
	16.3	172	145	5.6	2.6	1.4	1.2	11	10	12	2.1	*	*	6.0	7.9
	14.0	62	49	6.8	1.2	0.50	0.41	6.2	6.4	20	11	*	0.10	3.2	3.1
	11.0	77	58	13	1.7	0.77	0.92	8.9	8.4	34	9.9	*	0.10	4.3	6.2
	8.0	105	95	14	2.1	0.88	0.70	12	12	43	22	*	0.16	7.9	12
	5.8	89	114	14	2.8	0.83	0.81	11	15	40	28	*	*	6.4	11
	4.7	100	96	15	3.1	0.86	0.79	11	14	41	25	*	0.26	8.2	9.9
	1.1	67	78	21	2.8	0.59	0.79	7.8	13	37	23	*	0.20	6.8	11
	Average	84	79	11	2.1	0.77	0.74	8.5	9.9	28	15		0.12	5.4	7.8
TRM	570.8	2.8	3.1	9.9	2.2	0.23	0.56	*	*	6.3	4.8	0.18	0.20	2.1	5.5
	562.7	19	20	15	2.2	0.32	0.32	2.1	2.6	15	9.2	0.36	0.25	2.2	6.5
	552.7	26	23	9.8	2.0	0.88	1.1	3.7	2.8	19	8.3	*	0.22	6.6	6.6
	543.8	20	16	9.6	1.9	0.54	0.50	3.2	2.4	17	8.0	*	0.18	6.6	4.8
	532.0	16	12	7.7	1.5	0.74	0.72	2.9	1.9	15	6.2	*	0.17	4.8	4.3
Average	17	15	10	2.0	0.54	0.65	2.4	1.9	14	7.3	0.11	0.20	4.5	5.5	
Fort Loudoun Background Data															
TRM	615.8	--	2.8	--	1.6	--	0.23	--	0.03	--	4.2	--	0.19	--	3.9
	604.4	3.3	3.1	8.1	1.5	0.47	0.34	*	*	5.8	5.0	0.59	0.14	2.4	4.8
Average		2.9		1.5		0.29		0.015		4.6		0.17		4.4	

<sup>a</sup>TRE - total rare earths minus cerium

\*None detected

--No samples taken

**Table A-7**

Annual Discharges of Radionuclides from White Oak Creek to the Clinch River, 1944 to 1985  
(Curies)

Year	Gross Beta	<sup>137</sup> Cs	<sup>106</sup> Ru	<sup>89</sup> Sr	<sup>90</sup> Sr	TRE(-Ce) <sup>a</sup>	<sup>144</sup> Ce	<sup>95</sup> Zr	<sup>95</sup> Nb	<sup>131</sup> I	<sup>60</sup> Co	<sup>3</sup> H	TRU
1944 <sup>b</sup>	600												
1945 <sup>b</sup>	500												
1946 <sup>b</sup>	900												
1947 <sup>b</sup>	200												
1948 <sup>b</sup>	494												
1949		77	110		150	77	18	180	22	77		NA <sup>c</sup>	0.04 <sup>d</sup>
1950		19	23		38	30	NA	15	42	19			0.04
1951		20	18		29	11	NA	5	2	18			0.08
1952		10	15		72	26	23	19	18	20			0.03
1953		6	26		130	110	7	8	4	2			0.08
1954		22	11		140	160	24	14	9	4	NA		0.07
1955		63	31		93	150	85	5	6	7	7		0.25
1956		170	29		100	140	59	12	15	4	46		0.28
1957		89	60		83	110	13	23	7	1	5		0.15
1958		55	42	NA	150	240	30	6	6	8	9		0.08
1959		76	520	0.3	60	94	48	27	30	1	77		0.68
1960		31	1,900	1.9	28	48	27	38	45	5	72		0.19
1961		15	2,000	2.0	22	24	4	20	70	4	31		0.07
1962		6	1,400	1.7	9	11	1	2	8	0.4	14		0.06
1963		4	430	1.0	8	9	2	0.3	0.7	0.4	14		0.17
1964		6	190	0.8	7	13	0.3	0.2	0.1	0.3	15	1,900	0.08
1965		2	69	0.6	3	6	0.1	0.3	0.3	0.2	12	1,200	0.50
1966		2	29	0.9	3	5	0.1	0.7	0.7	0.2	7	3,100	0.16
1967		3	17	0.7	5	9	0.2	0.5	0.5	0.9	3	13,300	1.03
1968		1	5	0.6	3	4	0.03	0.3	0.3	0.3	1	9,700	0.04
1969		1	2	0.3	3	5	0.02	0.2	0.2	0.5	1	12,200	0.20
1970		2	1	0.3	4	5	0.06	0.02	0.02	0.3	1	9,500	0.40
1971		1	0.5	0.2	3	3	0.05	0.01	0.01	0.2	1	8,900	0.05
1972		2	0.5	NA	6	5	0.03	0.01	0.01	0.3	1	10,600	0.07
1973		2	0.7		7	NA	0.02	0.05	0.05	0.5	1	15,000	0.08
1974		1	0.2		6		0.02	0.02	0.02	0.2	0.6	8,600	0.02
1975		0.6	0.3		7		NA	NA	NA	0.3	0.5	11,000	0.02

**Table A-7**

Annual Discharges of Radionuclides from White Oak Creek to the Clinch River, 1944 to 1985  
(Continued)

Year	Gross Beta	<sup>137</sup> Cs	<sup>106</sup> Ru	<sup>89</sup> Sr	<sup>90</sup> Sr	TRE(-Ce) <sup>a</sup>	<sup>144</sup> Ce	<sup>95</sup> Zr	<sup>95</sup> Nb	<sup>131</sup> I	<sup>60</sup> Co	<sup>3</sup> H	TRU
1976		0.2	0.2		5					0.03	0.9	7,400	0.01
1977		0.2	0.2		3					0.03	0.4	6,200	0.03
1978		0.3	0.2		2					0.04	0.4	6,300	0.03
1979		0.2	0.1		2.4					0.04	0.4	7,700	0.03
1980		0.6	0		1.5					0.04	0.4	4,600	0.04
1981		0.2	0.1		1.5					0.04	0.7	2,900	0.04
1982		1.5	0.2		2.7					0.06	1.0	5,400	0.03
1983		1.2	0.2		2.1					0.004	0.3	5,600	0.05
1984		0.6	0.2		2.6					0.05	0.2	6,400	0.03
1985		0.4	0.007		3.0						0.6	3,700	0.008

<sup>a</sup>Total rare earths minus cerium.

<sup>b</sup>Individual radionuclide data not available.

<sup>c</sup>No analysis performed.

<sup>d</sup>Estimated from measurements made during last quarter of 1949.

Table A-8

RADIOACTIVITY IN CLINCH RIVER - 1965

Location	Concentration of Radionuclides of Primary Concern in Units of $10^{-8}$ $\mu\text{c/ml}$						Average Concentration of Total Radioactivity	$(\text{MPC})_w^a$	% of
	$^{90}\text{Sr}$	$^{144}\text{Ce}$	$^{137}\text{Cs}$	$^{103-106}\text{Ru}$	$^{60}\text{Co}$	$^{95}\text{Zr-}^{95}\text{Nb}$	$10^{-8}$ $\mu\text{c/ml}$	$10^{-8}$ $\mu\text{c/ml}$	$(\text{MPC})_w$
CRM 41.5 <sup>b</sup>	0.06	0.03	0.03	0.24	*	*	0.36	1.6	0.23
CRM 20.8 <sup>c</sup>	0.06	0.01	0.03	0.79	0.17	< 0.01	2.8	4.6	0.61
CRM 4.5 <sup>b</sup>	0.15	0.04	0.17	1.2	0.23	0.01	1.7	2.8	0.60

<sup>a</sup>Weighted average  $(\text{MPC})_w$  calculated for the mixture, using  $(\text{MPC})_w$  values for specific radionuclides specified by AEC Manual, Chapter W0524, Appendix, Annex 1, Table II.

<sup>b</sup>Measured values.

<sup>c</sup>Values given for this location are calculated values based on the levels of waste released and the dilution afforded by the river; they do not include amounts of radioactive material (e.g., fallout) that may enter the river upstream from CRM 20.8.

\*None detected.

Table A-9

Water Monitoring Results  
Clinch River - Below ORNL, 1961 - 1984  
(pCi/l)

Year	Potable Water Sr-90	River Water (CRM 4.5, Center's Ferry)*					
		Sr-90	Ce-144	Cs-137	Ru-103/ Ru-106	Co-60	Zr-95 Nb-95 H-3
1961	0.3	4.3	1.1	0.5	390	3.9	
1962	0.5	2.3	1.0	0.7	160	3.2	
1963		2.9	2.7	2.3	80	4.7	4.0
1964	1.3	2.8	1.0	3.3	45	4.3	0.25
1965	0.64	1.5	0.4	1.7	12	2.3	0.1
1966	0.79	2.1	0.6	1.6	3.9	3.7	<0.1
1967	0.80	1.2	0.2	1.5	0.6	1.2	0.1
1968	0.35	1.5	0.4	1.4	0.7	2.7	0.2
1969	0.43	1.1	0.2	1.9	1.4	3.5	0.4
1970	0.51	1.1	0.3	1.9	0.5	1.0	0.2 2,200
1971	0.51	1.0	0.3	0.9	2.0	0.8	<0.1 1,850
1972	0.40	1.1		0.5	0.6		<1,620
1973	0.44	1.2		0.5	0.5		<1,530
1974	0.34	0.45		0.08	0.31		1,240
1975	0.60	0.31		0.05	0.19		1,100
1976	0.05	0.24		0.02	0.15		1,900
1977	0.12	0.27		0.5	0.19		1,000
1978	0.10	0.10		0.71	0.73		2,200
1979	0.46	0.33		0.02	0.11		1,590
1980	0.68	0.75		0.08	0.17		5,580
1981	1.5	1.4		0.12	0.03	0.1	1,640
1982	1.1	1.8		0.67		0.6	3,400
1983	0.51	2.0		0.28		0.2	3,800
1984	0.7	2.2		<0.54		<0.54	1,700

\*Sample point deleted in 1980 - Gallaher data used for 1980 and following.



Table A-10

NON-RADIOACTIVE WATER MONITORING DATA—EAST FORK POPLAR CREEK  
(Location E-1, Figure 3)

1972

Substance	Number of Samples	Concentration, mg/l				% STD.	
		Maximum	Minimum	Average	STD.		
Cd	12	< 0.01	< 0.01	< 0.01*		0.01 <sup>a</sup>	< 100
Cl <sup>-</sup>	12	18.5	8.7	11.7	± 1.6	250 <sup>a</sup>	5
Cr	12	0.34	0.012	0.16	± 0.08	0.05 <sup>a</sup>	320
CN	12	< 0.01	< 0.01	< 0.01*		0.01 <sup>a</sup>	< 100
DO	19	8.4	3.0	4.3	± 0.8	5.0 <sup>c</sup>	116
F <sup>-</sup>	12	1.3	0.4	1.0	± 0.2	1.2 <sup>a</sup>	83
Hg	12	0.0009	< 0.0005	< 0.0006	± 0.00006	0.005 <sup>b</sup>	< 12
NO <sub>3</sub> <sup>-</sup>	12	12.4	0.4	2.8	± 2.1	450 <sup>a</sup>	6
Pb	12	0.025	< 0.002	< 0.02	± 0.004	0.05 <sup>a</sup>	< 40
SO <sub>4</sub> <sup>=</sup>	12	72	40	56	± 5	250 <sup>a</sup>	22
T.D.S.	12	275	35	196	± 40	500 <sup>a</sup>	39
Zn	12	0.6	0.03	0.10	± 0.1	5.0 <sup>a</sup>	2

<sup>a</sup>U.S. Public Health Service Drinking Water Standards.

<sup>b</sup>Proposed EPA Standards. <sup>(a)</sup>

<sup>c</sup>Tennessee Water Quality Standard.

\* All values below limit of detection.

NOTE: Stream not a source of drinking water. Drinking water standards used for water quality comparison only.

**Table A-11**

Concentration of Radionuclides at Sampling Stations (curies/liter)

	<sup>90</sup> Sr			<sup>106</sup> Ru			<sup>60</sup> Co		<sup>137</sup> Cs	
	Flow Weighted Mean Concentration	Maximum Concentration	Mean Activity in Suspended Solids (%)	Flow Weighted Mean Concentration	Maximum Concentration	Mean Activity in Suspended Solids (%)	Maximum Concentration	Mean Activity in Suspended Solids (%)	Maximum Concentration	Mean Activity in Suspended Solids (%)
	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>	× 10 <sup>-12</sup>
Clinch River at Oak Ridge Water Plant (CRM 41.5)	0.71	5.0	24	23	223	44	5	5	6	82
White Oak Creek at White Oak Dam (WOCM 0.6)	1,349	17,450	2	109,800	294,412	6	4,095	19	6,409	69
Clinch River at ORGDP water Intake (CRM 14.4)	4.5	11.7	6	345	769	17	18	27	21	92
Clinch River above Centers Ferry (CRM 5.5)	4.2	42.6	9	317	2,633	16	52	30	35	86
Tennessee River at Loudon, Tennessee (TRM 591.4)		2.3			296		1		34	
Tennessee River at Watts Bar Dam (TRM 529.9)	1.6	16.4	9	63	192	7	33	2	18	30
Tennessee River at Chickamauga Dam (TRM 471.0)	1.6	14.1	10	51	269	8	3	3	6	19

Source: Struxness et al. (1967)

Table A-12

## Baseline Concentrations of PCB's

<u>Location</u>	<u>Concentration</u> <u>µg/g</u>
New Hope Pond (12 samples)	38 avg.
New Hope Pond Outfall (22 water samples)	< 0.0005 µg/l avg.
Sludge Pond Y-12 (3 samples)	137 avg.
East Fork Poplar Creek	
Below Outfall New Hope Pond	0.1
Near Bear Creek Road	0.1
Near Gamble Valley Road	0.2
Near Howard's	0.1
Near Tennis Courts (top layer)	0.6
Near Tennis Courts (bottom layer)	0.2
Turnpike West of Gallaher Cemetery	0.1
Halfway between Turnpike and Blair Road	0.3
Near Blair Road	0.2
Poplar Creek (East Fork near K-25)	8
Poplar Creek near K-33 Cooling Tower	12
Poplar Creek near Mouth	6
Poplar Creek near K-1203 Sewage Plant	14
Poplar Creek near K-1410	12
Poplar Creek near K-1131 Condensate Line	15
Poplar Creek near K-1407-B Outfall	9
K-901-A Lagoon near Weir	24
K-1407-C Sludge Lagoon near Middle	25
K-1007-B Pond	0.4
K-1001-B Pond	< 0.1
Spring (background)	0.1
Clinch River near Grubb Island	< 0.1
Clinch River, Right Side of Grubb Island	< 0.1
Grubb Island (submerged vegetation)	< 0.1
Melton Hill Dam Swimming Area	< 0.1
Melton Hill Dam near Danger Buoy	< 0.1
White Oak Creek, 0.3 mi. above mouth	2.0
White Oak Creek, 0.3 mi. above mouth (rocky)	< 0.1
Scarboro Embayment, 1A	0.1
Scarboro Embayment, 1B	0.2

Note: All samples are sediment samples except as noted.

Table A-13

STREAM SEDIMENT SAMPLES  
 July/November 1978  
 Average Concentration ( $\mu\text{g/g}$  dry weight basis)

STATION	U	Hg	Pb	Ni	Cu	Zn	Cr	Mn	Cd	Al	Th
CS1	16	< 0.2	30	30	20	60	43	1875	<5	34000	<40
PS2	15	7	63	86	114	124	100	546	<5	102000	<40
PS5	9	2	39	53	41	100	59	798	<5	56000	<40
PS6	14	8	42	94	53	159	129	511	<5	62000	<40
PS9	4	2	47	59	57	86	71	929	<5	52000	<40
PS10	21	17	37	187	58	149	60	562	<5	40000	<40
PS12	17	6	47	69	42	96	53	715	<5	46000	<40
PS15	95	21	65	388	165	169	237	520	<5	50000	<40
PS17	12	< 2	46	50	36	72	47	973	<5	46000	<40
PS18	6	5	45	37	36	74	48	539	<5	42000	<40
PS19	9	14	54	36	61	69	48	813	<5	46000	<40
PS21	13	6	55	82	100	125	79	765	<5	84000	<40
PS22	10	3	47	60	37	98	52	746	<5	50000	<40
CS20	8	< 0.2	35	50	40	90	57	1105	<5	59000	<40

NOTE: An evaluation of the Clinch River and Poplar Creek hydrology resulted in the addition of two new sampling points (PS21 and PS22) and the deletion of eight sampling points, which provided for a more representative sampling program.

Table A-14

INSTREAM CONTAMINANT STUDY - TASK 1  
 BASEFLOW SURVEY - MAXIMUM CONCENTRATIONS OF SIGNIFICANT RADIOISOTOPES IN  
 WATER SAMPLES AND APPLICABLE STANDARDS AND BACKGROUND LEVELS

Isotope	LLD <sup>1</sup> (pCi/L)	Standards and Background Levels (pCi/L)			Concentrations of Significant Isotopes - Baseflow Survey <sup>5</sup> (pCi/L)			
		Drinking Water Standard <sup>2</sup>	MPC <sup>3</sup>	Tenn. River <sup>4</sup>	White Oak Creek	Bear Creek	Clinch River	East Fork Poplar Creek
Gross Alpha	2.0	15	30	4.0	11 (36%)	31 (103%)	3 (10%)	8 (25%)
Gross Beta	2.4	-- <sup>6</sup>	3,000	9.6	690 (23%)	330 (11%)	4 (0.1%)	17 (0.6%)
Tritium	330.0	20,000	3,000,000	712.0	544,000 (18%)	500 (0.02%)	500 (0.02%)	400 (0.01%)
Cs-137	5.0	-- <sup>6</sup>	20,000	-- <sup>7</sup>	68 (0.34%)	-- <sup>7</sup>	-- <sup>7</sup>	-- <sup>7</sup>
Co-60	5.0	-- <sup>6</sup>	30,000	-- <sup>7</sup>	19 (0.06%)	-- <sup>7</sup>	-- <sup>7</sup>	-- <sup>7</sup>
Sr-90	--	-- <sup>6</sup>	300	-- <sup>8</sup>	-- <sup>8</sup>	-- <sup>8</sup>	0.6 (0.2%)	-- <sup>8</sup>

<sup>1</sup> Lower limit of detection as calculated by the method developed by Pasternack and Harley and described in HASL-300 and Nuclear Instruments Methods 91, 533-40 (1971).

<sup>2</sup> Interim Primary Drinking Water Regulations as outlined in 40 CFR 141.

<sup>3</sup> Maximum Permissible Concentrations (MPC) recommended by 10 CFR 20 for nonoccupational exposure.

<sup>4</sup> Maximum concentrations reported by TVA in the Tennessee River samples collected in 1981-83.

<sup>5</sup> The percentage of the MPC value is reported in parenthesis.

<sup>6</sup> No standard available.

<sup>7</sup> Isotope not identified in gamma spectral analyses.

<sup>8</sup> Analysis not performed.

Source: TVA (1985a)

**Table A-15**

INSTREAM CONTAMINANT STUDY - TASK 1  
 STORMFLOW SURVEYS - MAXIMUM CONCENTRATIONS OF SIGNIFICANT RADIOISOTOPES IN  
 WATER SAMPLES AND APPLICABLE STANDARDS AND BACKGROUND LEVELS

Isotope	LLD <sup>1</sup> (pCi/L)	Standards and Background Levels (pCi/L)			Concentrations of Significant Isotopes - Two Stormflow Surveys <sup>5</sup> (pCi/L)	
		Drinking Water Standard <sup>2</sup>	MPC <sup>3</sup>	Tenn. River <sup>4</sup>	Bear Creek	East Fork Poplar Creek
Gross Alpha	2.0	15	30	4.0	8 (27%)	15 (50%)
Gross Beta	2.4	-- <sup>6</sup>	3,000	9.6	36 (1%)	55 (2%)
I-131	8.0	-- <sup>6</sup>	300	-- <sup>7</sup>	-- <sup>7</sup>	14 (5%)
Pa-234m	--	-- <sup>6</sup>	--	-- <sup>7</sup>	-- <sup>7</sup>	268

<sup>1</sup> Lower limit of detection as calculated by the method developed by Pasternack and Harley and described in HASL-300 and Nuclear Instruments Method 91, 533-40 (1971).

<sup>2</sup> Interim Primary Drinking Water Regulations as outlined in 40 CFR 141.

<sup>3</sup> Maximum Permissible Concentrations (MPC) recommended by 10 CFR 20 for nonoccupational exposure.

<sup>4</sup> Maximum concentrations reported by TVA in the Tennessee River samples collected from 1981-83.

<sup>5</sup> The percentage of the MPC value is reported in parenthesis.

<sup>6</sup> No standard available.

<sup>7</sup> Isotope not identified in gamma spectral analyses.

Source: TVA (1985a)

**Table A-16**  
**INSTREAM CONTAMINANT STUDY - TASK 2**  
**SURFACE LAYER, FINE-PARTICLE SEDIMENT - SUMMARY OF METAL CONCENTRATIONS**

Parameter (ppm) <sup>1</sup>	East Fork Poplar Creek			Bear Creek			White Oak Creek			Poplar Creek		
	Max	Min (n=16)	Mean	Max	Min (n=3)	Mean	Max	Min (n=4)	Mean	Max	Min (n=3)	Mean
Mercury	165.0	11.0	40.0	0.7	<0.1	0.3	6.0	2.2	3.3	5.9	0.1	3.4
Arsenic	14.0	3.8	6.9	11.0	4.8	6.5	12.0	5.0	8.7	11.0	7.4	8.9
Cadmium	8.2	<0.5	1.6	8.6	<0.5	8.6	2.4	0.6	1.4	3.5	2.1	2.9
Chromium	58.0	24.0	37.0	35.0	16.0	22.0	290.0	66.0	163.0	38.0	19.0	27.0
Lead	170.0	36.0	80.0	85.0	35.0	52.0	51.0	33.0	40.0	38.0	23.0	32.0
Nickel	74.0	20.0	37.0	155.0	28.0	67.0	30.0	24.0	26.0	65.0	43.0	56.0
Silver	45.0	2.0	8.0	<1.0	<1.0	<1.0	10.0	2.0	6.0	2.0	<1.0	2.0
Zirconium	590.0	350.0	448.0	500.0	430.0	500.0	480.0	260.0	365.0	470.0	220.0	340.0

Parameter (ppm) <sup>1</sup>	Background Stations								
	Clinch River (Watts Bar)			Clinch River (Melton Hill)			Norris Reservoir <sup>2</sup>		
	Max	Min (n=4)	Mean	Max	Min (n=1)	Mean	Max	Min (n=3)	Mean
Mercury	2.8	0.3	0.8	<0.1	<0.1	<0.1	0.1	0.1	0.1
Arsenic	11.0	5.1	8.1	17.0	17.0	17.0	26.0	16.0	22.0
Cadmium	4.2	0.7	1.8	0.5	0.5	0.5	<0.5	<0.5	<0.5
Chromium	25.0	9.0	16.0	25.0	25.0	25.0	23.0	21.0	22.0
Lead	38.0	14.0	30.0	28.0	28.0	28.0	77.0	58.0	67.0
Nickel	38.0	14.0	21.0	36.0	36.0	36.0	28.0	24.0	26.0
Silver	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0	<1.0
Zirconium	890.0	400.0	650.0	230.0	230.0	230.0	270.0	180.0	220.0

<sup>1</sup> ppm is equivalent to mg/kg.

<sup>2</sup> Values for Norris Reservoir include Clinch River Miles 85.3 and 94.1 and Powell River Mile 6.0.

Table A-17

INSTREAM CONTAMINANT STUDY - TASK 2  
 SURFACE LAYER, FINE-PARTICLE SAMPLING - MAXIMUM CONCENTRATIONS REPORTED FOR SIGNIFICANT  
 RADIOISOTOPES IN SURFACE SEDIMENT SAMPLES (All concentrations are pCi/g, dry weight)<sup>1</sup>

ANALYSIS/ISOTOPE	Clinch River	East Fork Poplar Creek	Bear Creek	White Oak Creek	Poplar Creek	Norris Reservoir Control Station		Comparison Data	
						Clinch River Upstream	Powell River	Tennessee River <sup>2</sup>	Clinch River <sup>3</sup>
Gross Alpha	11	160	32	3	11	4	3	15	100
Gross Beta	398	110	150	6,600	60	42	37	65 <sup>4</sup>	91
Uranium	7	90	200	4.1	14	5.9	2.3	-	2.0
Sr-89	4	.7	.5	0	1	4	1	14	12.1
Sr-90	1.8	.1	.1	900	.2	.4	.5	.6	0.6
<b>GAMMA SPECTRAL ANALYSIS<sup>5</sup></b>									
Co-60	1.2	-	-	184	-	-	-	.6	2.7
Cs-134	-	-	-	1.2	-	-	-	.13	.01
Cs-137	167	8.6	.2	12,100	1.9	.9	.7	5.5	83
K-40	27	21	18	25	16	27	16	27	84
Ra-226	.7	-	-	-	-	-	-	2.3	1.4
Th-234	4	23	63	-	3.8	-	-	-	-
Eu-152	-	-	-	8	-	-	-	-	-
Eu-154	-	-	-	7	-	-	-	-	-
Am-241	-	-	-	4	-	-	-	-	-
Ac-228	2.2	2	1.8	4	1.1	2.2	1.8	2.7	2.1
Pa-234	-	-	-	-	-	-	-	4.6	4.8
<b>TRANSURANICS</b>									
Pu-239	.73	.10	.001	1.08	.02	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>
Pu-238	.03	.02	.001	.007	<.0002	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>
Am-241	.51	.05	.04	.46	.02	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>
Cm-244	.02	.01	.009	.01	<.0008	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>	- <sup>4</sup>

<sup>1</sup>Uranium reported in units of ug/g, dry weight.

<sup>2</sup>Maximum concentrations reported by TVA in surface sediment samples collected from the Tennessee River from 1981-83.

<sup>3</sup>Maximum concentrations reported by TVA in surface sediment samples collected from the Clinch River from 1974-1983.

<sup>4</sup>Analysis not performed.

<sup>5</sup>Dash indicates isotope not identified in gamma spectral analysis.

NOTE: The lower limits of detection for all isotopes, as determined by the method developed by Pasternack and Harley, and described in HASL-300 and Nucl. Inst. Methods 91, 533-40 (1971), are typically 1 pCi/g, dry weight, or less.

Source: TVA (1985b)



**Table A-18**  
 INSTREAM CONTAMINANT STUDY - TASK 2  
 CLINCH RIVER CORE SAMPLING - SUMMARY OF MERCURY RESULTS  
 AND COMPARISON TO PREVIOUS ORNL CORE DATA

Location	Surface Layer Concentration (mg/kg)	Highest Concentration (mg/kg)	Sediment <sup>1</sup> Covering Highest Concentration (inches)	Core Penetrated to Background <sup>2</sup>
CRM 15.6	0.1	0.5	5.5	Yes
(CRM 11.0) <sup>3</sup>		180.0		
CRM 10.0	0.7	0.8	4.0	Yes
(CRM 6.8) <sup>4</sup>	3.8	13.2	32.0	Yes
CRM 3.7	3.8	12.0	30.7	No
(CRM 1.0) <sup>4</sup>	3.0	46.0	39.0	No
Grubb Island (CRM 18.3)	<0.1	<0.1	0	Yes
Jones Island				
CRM 20.5	<0.1	<0.1	0	Yes
CRM 20.1	<0.1	<0.1	0	Yes
CRM 19.7	<0.1	<0.1	0	Yes

<sup>1</sup>Depth of sediment covering the layer of highest mercury concentration.

<sup>2</sup>Background concentration is 0.2 mg/kg based on Table 16 for mean concentrations of the Clinch River and tributaries to the Upper Tennessee River.

<sup>3</sup>Data collected by ORNL in 1977 (9).

<sup>4</sup>Data collected by ORNL in 1983 (9).

Source: TVA (1985b)

**Table A-19**  
**INSTREAM CONTAMINANT STUDY - TASK 2**  
**TENNESSEE RIVER CORE SAMPLING - SUMMARY OF MERCURY**  
**RESULTS AND COMPARISON TO PREVIOUS ORNL DATA**

Location	Surface Layer Concentration (mg/kg)	Highest Concentration (mg/kg)	Sediment <sup>1</sup> Covering Highest Concentration (inches)	Core Penetrated to Background <sup>2</sup>
WATTS BAR RESERVOIR				
TRM 574.4 Control Station	0.1	0.3	27.7	No
TRM 552.0	1.3	7.5	37.4	No
(TRM 550.0) <sup>3</sup>	<1.0	14.0	36.0	Yes
TRM 540.0	0.7	7.8	31.5	Yes
(TRM 538.0) <sup>3</sup>	<0.5	7.0	14.0	Yes
CHICKAMAUGA RESERVOIR				
(TRM 502.0) <sup>3</sup>	<0.35	0.5	30.0	No
TRM 509.0	0.3	0.7	9.8	Yes
TRM 487.9	0.5	2.6	13.8	Yes
TRM 475.0	0.6	3.0	13.8	No
(TRM 472.0) <sup>3</sup>	<0.7	3.5	14.0	Yes
GUNTERSVILLE RESERVOIR				
TRM 387.0	0.5	0.5	2.0	Yes

<sup>1</sup>Depth of sediment covering the layer of highest mercury concentration.

<sup>2</sup>Background concentration is 0.2 mg/kg based on Table 16 for mean concentrations of the Clinch River and tributaries to Upper Tennessee River.

<sup>3</sup>Data collected by ORNL in 1983 (9).

Table A-20  
Contaminant distributions between aqueous and particulate phases

Date	Nuclide	Suspended load (mg/L)	Dissolved (fCi/L)	Particulate (pCi/g)	Distribution <sup>a</sup> $K_d$
<i>City of Kingston—Mouth of the Clinch River</i>					
12/1/86	<sup>60</sup> Co	14	24	1.1	$5 \times 10^4$
	<sup>137</sup> Cs		35	6.8	$2 \times 10^5$
	<sup>7</sup> Be		92	5.4	$6 \times 10^4$
	<sup>239,240</sup> Pu		0.38	0.04	$1 \times 10^5$
	<sup>238</sup> Pu		0.11	0.005	$5 \times 10^4$
12/5/86	<sup>60</sup> Co	11	250	4.3	$2 \times 10^4$
	<sup>137</sup> Cs		49	14.6	$3 \times 10^5$
	<sup>7</sup> Be		65	5.3	$8 \times 10^4$
	Hg (ppb) <sup>b</sup>		0.005	2360	$5 \times 10^5$
12/17/86	<sup>60</sup> Co	7	215	7.3	$3 \times 10^4$
	<sup>137</sup> Cs		103	26.5	$3 \times 10^5$
	<sup>7</sup> Be		76	8.0	$1 \times 10^5$
<i>Thief Neck—Watts Bar Reservoir</i>					
12/22/86	<sup>60</sup> Cs	7	12	0.9	$8 \times 10^4$
	<sup>137</sup> Cs		17	5.1	$3 \times 10^5$
	<sup>7</sup> Be		78	8.7	$1 \times 10^5$
<i>Mouth of White Creek—Watts Bar Reservoir</i>					
3/9/89	Hg (ppb) <sup>b</sup>	21	0.004	510	$1 \times 10^5$

<sup>a</sup>Particle-to-water distribution  
 $K_d = \frac{\text{concentration per kilogram of particles}}{\text{concentration per liter of water}}$

<sup>b</sup>ppb = parts per billion or  $\mu\text{g/L}$ .

Table A-21

Watts Bar sediment core 567.5 (August 22, 1986)

Sediment depth (cm)	Dry weight (g)	<sup>137</sup> Cs sediment (pCi/g ± 1σ)	Hg sediment (μg/g)	Hg dissolved <sup>a</sup> (μg/L)	Historical time period <sup>b</sup> (years)
0-4	72.58	5.26 ± 0.06	1.52	0.015	1986
4-8	119.62	5.68 ± 0.07	1.36	0.014	1984-1985
8-12	127.51	5.06 ± 0.08	1.77	0.018	1983
12-16	140.85	6.05 ± 0.19	2.42	0.024	1981-1982
16-20	147.80	7.39 ± 0.07	3.16	0.032	1980
20-24	144.13	6.51 ± 0.06	2.32	0.023	1978-1979
24-28	138.34	6.61 ± 0.06	2.32	0.023	1977
28-32	109.97	7.85 ± 0.06	3.33	0.033	1975-1976
32-36	101.34	11.01 ± 0.09	3.44	0.034	1974
36-40	133.20	9.81 ± 0.06	2.98	0.030	1972-1973
40-48	144.17	12.03 ± 0.24	2.91	0.029	1969-1971
48-56	149.98	13.56 ± 0.11	4.12	0.041	1966-1968
56-64	161.15	19.02 ± 0.15	5.10	0.051	1963-1965
64-72	156.36	22.24 ± 0.11	6.80	0.068	1960-1962
72-80	145.92	49.08 ± 0.21	24.40	0.244	1957-1959
80-84	70.62	58.36 ± 0.17	19.00	0.190	1955-1956
84-88	78.68	28.66 ± 0.11	6.31	0.063	1952-1954
88-92	81.22	13.01 ± 0.07	1.24	0.012	1950-1951
92-96	79.15	18.59 ± 0.08	0.36	0.004	1948-1949
96-100	77.05	10.64 ± 0.07	0.35	0.003	1946-1947
100-104	76.68	13.11 ± 0.07	0.31	0.003	
104-108	75.71	5.84 ± 0.05	0.34	0.003	
108-112	81.48	0.87 ± 0.02	0.12	0.001	
112-116	118.11	0.25 ± 0.03	0.09	0.001	
116-120	119.10	0.27 ± 0.05	0.05	0.001	
120-122	125.07	0.00 ± 0.00			

<sup>a</sup>Estimated using a particle-to-water distribution ratio of  $1 \times 10^5$ .<sup>b</sup>Estimated using a sediment accumulation rate of 1.4 cm/year.

**Table A-22**  
Air Monitoring Data From 1954

Part B. Statistical Data

1. Air Activity

a. Constant Air Monitors

<u>Station Number</u>	<u>Location</u>	Average Long Lived Activity	
		<u>Wkly Av. to Date This Year, Conc. x 10<sup>-13</sup> <math>\mu\text{c}/\text{cc}</math></u>	<u>Deviation from 1953 Weekly Average</u>
HP-1	N 3550	56.68	+63.3%
HP-2	S 3001	7865.40	+28294.9%
HP-3	S 1000	14.02	-37.8%
HP-4	W 3513	7.49	-37.5%
HP-5	E 2506	117.25	-59.6%
HP-6	SE 3012	1372.51	+11375.9%
HP-7	W 7001	8.27	-15.0%
HP-8	Rock Quarry	2.40	-79.6%
HP-9	A-10 Site	7.27	-35.8%
HP-10	E 2074	24.91	-22.2%

Average all stations 947.62

Deviation of this year's average long lived activity to date from last year's average +1942.3%

2. Particulate Studies

a. USPHS Filters

<u>Number</u>	<u>Location</u>	<u>Weekly Average to Date This Yr. Particles per 1000 ft<sup>3</sup></u>	<u>Deviation of Wkly Av. to Date This Year from Wkly Av. Last Year</u>
A-1	3026	1.51	-58.9%
A-2	3003	0.18	-74.3%
A-3	1000	0.31	-55.7%
A-4	7001	0.08	-73.3%

Average all stations 0.52

Deviation of this year's weekly average to date from weekly average last year -62.2%

**Table A-23**

CONCENTRATION OF RADIOACTIVE MATERIALS IN AIR - 1965  
(Filter Paper Data—Weekly Average)

Station Number	Location	Long-Lived Activity 10 <sup>-13</sup> uc/cc	No. of Particles by Activity Ranges				Total	Particles Per 1000 ft <sup>3</sup>
			< 10 <sup>5</sup> d/24 hr	10 <sup>5</sup> -10 <sup>6</sup> d/24 hr	10 <sup>6</sup> -10 <sup>7</sup> d/24 hr	> 10 <sup>7</sup> d/24 hr		
Laboratory Area								
HP-1	S 3587	2.6	5.3	0.00	0.00	0.00	5.3	0.22
HP-2	NE 3025	3.6	0.76	0.04	0.00	0.00	0.80	0.04
HP-3	SW 1000	2.6	0.81	0.00	0.00	0.00	0.81	0.04
HP-4	W Settling Basin	3.2	0.84	0.00	0.00	0.00	0.84	0.05
HP-5	E 2506	3.3	1.2	0.00	0.00	0.00	1.2	0.07
HP-6	SW 3027	2.7	0.56	0.02	0.00	0.00	0.58	0.03
HP-7	W 7001	1.9	0.02	0.00	0.00	0.00	0.02	0.00
HP-8	Rock Quarry	2.2	0.57	0.00	0.00	0.00	0.57	0.03
HP-9	N Bethel Valley Rd.	2.5	0.98	0.00	0.00	0.00	0.98	0.05
HP-10	W 2075	2.9	0.54	0.02	0.00	0.00	0.56	0.03
Average		2.8	1.2	0.01	0.00	0.00	1.2	0.06
Perimeter Area								
HP-31	Kerr Hollow Gate	1.7	1.1	0.00	0.00	0.00	1.1	0.02
HP-32	Midway Gate	1.9	1.2	0.02	0.00	0.00	1.2	0.02
HP-33	Gallaher Gate	1.5	0.75	0.00	0.00	0.00	0.75	0.02
HP-34	White Wing Gate	1.8	0.71	0.00	0.00	0.00	0.71	0.02
HP-35	Blair Gate	1.9	1.8	0.00	0.00	0.00	1.8	0.04
HP-36	Turnpike Gate	2.2	0.63	0.02	0.00	0.00	0.65	0.01
HP-37	Hickory Creek Bend	1.6	1.0	0.00	0.00	0.00	1.0	0.02
HP-38	E EGCR	2.2	1.4	0.00	0.00	0.00	1.4	0.04
HP-39	Townsite	3.5	1.7	0.00	0.00	0.00	0.0	0.05
Average		2.1	1.1	0.00	0.00	0.00	1.2	0.03
Remote Area								
HP-51	Norris Dam	2.1	1.2	0.04	0.00	0.00	1.2	0.02
HP-52	Loudoun Dam	1.7	0.98	0.02	0.00	0.00	1.0	0.02
HP-53	Douglas Dam	1.9	0.77	0.00	0.00	0.00	0.77	0.02
HP-54	Cherokee Dam	1.9	1.2	0.02	0.00	0.00	1.3	0.02
HP-55	Watts Bar Dam	1.9	1.5	0.02	0.00	0.00	1.5	0.03
HP-56	Great Falls Dam	1.8	1.4	0.02	0.00	0.00	1.4	0.03
HP-57	Dale Hollow Dam	1.8	1.3	0.02	0.00	0.00	1.3	0.03
*HP-58	Knoxville	2.0	1.8	0.05	0.00	0.00	1.9	0.04
Average		1.9	1.3	0.02	0.00	0.00	1.3	0.02

\* Installed March, 1965.

**Table A-24**  
**Continuous Air Monitoring Data Specific Radionuclides in Air - 1979**  
**(Composite Samples)**  
**Units of  $10^{-15}$   $\mu$ Ci/cc**

Radionuclides	Yearly Average		
	Local Stations	Perimeter Stations	Remote Stations
<sup>7</sup> Be	104	106	95
<sup>90</sup> Sr	0.23	0.15	0.19
<sup>106</sup> Ru	1.4	1.56	1.36
<sup>125</sup> Sb	0.32	0.37	0.33
<sup>137</sup> Cs	0.96	0.67	0.61
<sup>144</sup> Ce	1.52	1.85	1.10
<sup>228</sup> Th	0.01	0.01	0.002
<sup>230</sup> Th	0.01	0.02	0.01
<sup>232</sup> Th	0.02	0.01	0.01
<sup>234</sup> U	0.33	0.44	0.01
<sup>235</sup> U	0.01	0.03	0.01
<sup>238</sup> U	0.08	0.26	0.02
<sup>238</sup> Pu	0.0001	0.002	0.0007
<sup>239</sup> Pu	0.01	0.04	0.01

\*Not detectable.

**Table A-25**  
**RADIOPARTICULATE FALLOUT — 1965**  
 (Gummed Paper Data—Weekly Average)

Station Number	Location	Long-Lived Activity 10 <sup>-4</sup> µc/ft <sup>2</sup>	No. of Particles by Activity Ranges				Total Particles Per Sq. Ft.
			< 10 <sup>5</sup> d/24 hr	10 <sup>5</sup> -10 <sup>6</sup> d/24 hr	10 <sup>6</sup> -10 <sup>7</sup> d/24 hr	> 10 <sup>7</sup> d/24 hr	
Laboratory Area							
HP-1	S 3587	1.3	1.71	0.15	0.00	0.00	1.87
HP-2	NE 3025	1.1	1.61	0.06	0.04	0.02	1.73
HP-3	SW 1000	0.7	1.13	0.08	0.00	0.00	1.21
HP-4	W Settling Basin	1.4	0.67	0.04	0.00	0.00	0.71
HP-5	E 2506	0.6	1.23	0.19	0.00	0.00	1.42
HP-6	SW 3027	1.1	1.85	0.13	0.02	0.00	2.00
HP-7	W 7001	0.4	0.20	0.00	0.00	0.00	0.20
HP-8	Rock Quarry	0.5	0.83	0.06	0.00	0.02	0.91
HP-9	N Bethel Valley Rd.	0.4	1.00	0.06	0.00	0.00	1.06
HP-10	W 2075	2.1	2.29	0.17	0.00	0.04	2.50
Average		1.0	1.28	0.10	0.01	0.01	1.40
Perimeter Area							
HP-31	Kerr Hollow Gate	0.48	0.23	0.13	0.00	0.00	0.37
HP-32	Midway Gate	0.56	1.12	0.00	0.00	0.00	1.12
HP-33	Gallaher Gate	0.41	0.92	0.06	0.00	0.00	0.98
HP-34	White Wing Gate	0.44	0.88	0.02	0.00	0.00	0.90
HP-35	Blair Gate	0.44	0.90	0.02	0.00	0.00	0.92
HP-36	Turnpike Gate	0.48	0.92	0.04	0.00	0.00	0.96
HP-37	Hickory Creek Bend	0.42	0.79	0.04	0.00	0.00	0.83
HP-38	E EGCR	0.45	1.25	0.02	0.00	0.00	1.27
HP-39	Townsite	0.49	1.42	0.02	0.00	0.00	1.44
Average		0.46	0.94	0.04	0.00	0.00	0.98
Remote Area							
HP-51	Norris Dam	0.45	0.37	0.02	0.00	0.00	0.38
HP-52	Loudoun Dam	0.37	0.17	0.13	0.00	0.00	0.31
HP-53	Douglas Dam	0.36	0.40	0.04	0.00	0.00	0.44
HP-54	Cherokee Dam	0.35	0.46	0.02	0.00	0.00	0.48
HP-55	Watts Bar Dam	0.39	0.35	0.02	0.00	0.00	0.37
HP-56	Great Falls Dam	0.42	1.06	0.06	0.00	0.00	1.12
HP-57	Dale Hollow Dam	0.45	0.63	0.02	0.00	0.00	0.65
*HP-58	Knoxville	0.40	0.45	0.10	0.00	0.00	0.55
Average		0.40	0.49	0.05	0.00	0.00	0.54

\*Installed March, 1965.



**Table A-26**  
**Air Monitoring Data Summary**  
**Gross Alpha, Gross Beta, and I-131, 1961-1984**  
**(E-6 pCi/cc)**

Year	Gross Alpha		Gross Beta		Iodine-131	
	Local	Remote	Local	Remote	Local	Perimeter
1961			1.6	1.7		
1962			3.7	4.3		
1963			4.9	4.3		0.015
1964			1.3	1.1		0.03
1965			0.28	0.19		0.015
1966			0.17	0.11	0.23	0.014
1967			0.22	0.10	0.43	0.019
1968			0.29	0.16	0.13	0.013
1969			0.26	0.16	0.31	0.018
1970			0.33	0.23	0.03	0.01
1971			0.44	0.64	0.061	0.011
1972			0.13	0.084	0.037	0.011
1973			0.065	0.028	0.047	0.009
1974			0.10	0.084	0.029	0.008
1975			0.058	0.042	0.025	0.007
1976			0.046	0.026	0.026	0.008
1977	0.0019	0.0009	0.062	0.049	0.062	0.007
1978	0.0021	0.0011	0.089	0.076	0.031	0.008
1979	0.0021	0.0009	0.044	0.024	0.021	0.004
1980	0.0019	0.0011	0.052	0.029	0.003	0.001
1981	0.002	0.0011	0.089	0.070	0.003	0.001
1982	0.0022	0.0010	0.043	0.022	0.003	0.001
1983	0.0022	0.0010	0.051	0.024	0.005	0.001
1984	0.0026	0.0011	0.027	0.014	0.004	0.002

**Table A-27**

CONCENTRATION OF RADIOACTIVE MATERIALS IN RAINWATER - 1965  
(Weekly Average by Stations)

Station Number	Location	Activity in Collected Rainwater, $\mu\text{c/ml}$
Laboratory Area		
HP-7	West 7001	$0.35 \times 10^{-7} \mu\text{c/ml}$
Perimeter Area		
HP-31	Kerr Hollow Gate	$0.40 \times 10^{-7} \mu\text{c/ml}$
HP-32	Midway Gate	0.42
HP-33	Gallaher Gate	0.42
HP-34	White Wing Gate	0.38
HP-35	Blair Gate	0.48
HP-36	Turnpike Gate	0.37
HP-37	Hickory Creek Bend	0.40
HP-38	E EGCR	0.59
HP-39	Townsite	0.39
Average		$0.42 \times 10^{-7} \mu\text{c/ml}$
Remote Area		
HP-51	Norris Dam	$0.62 \times 10^{-7} \mu\text{c/ml}$
HP-52	Loudoun Dam	0.58
HP-53	Douglas Dam	0.49
HP-54	Cherokee Dam	0.57
HP-55	Watts Bar Dam	0.42
HP-56	Great Falls Dam	0.47
HP-57	Dale Hollow Dam	0.49
HP-58	Knoxville	0.40
Average		$0.50 \times 10^{-7} \mu\text{c/ml}$

**Table A-28** Radioactivity in fish taken in White Oak Lake and the Clinch River, 1948 (gross beta)

Site of collection	Number processed	Average counts per minute per gram of sample		
		Flesh	Bone	Scale
Clinch River mile 14.4	31	2	13	39
Clinch River mile 18-19	62	5	51	92
White Oak Creek	54	27	525	1204
Lower White Oak Lake	38	134	1264	1971
Upper White Oak Lake	13	423	2290	2901

Source: Knobf, 1951.

**Table A-29**

Concentration of Radionuclides in Clinch River Fish  
(picocuries per kg of fresh weight)

Fish Species	Sample Period	<sup>90</sup> Sr		<sup>137</sup> Cs		<sup>106</sup> Ru		<sup>60</sup> Co	
		Flesh	Total <sup>a</sup>	Flesh	Total <sup>a</sup>	Flesh	Total <sup>a</sup>	Flesh	Total <sup>a</sup>
Carp	1960-1962	(17) <sup>b</sup> 500 ± 140 <sup>c</sup>	(40) 5100 ± 1700	(71) 510 ± 57	(39) 560 ± 79	(69) 170 ± 18	(39) 290 ± 78	(67) 66 ± 6.1	(39) 49 ± 9.9
	1963	(20) 91 ± 22		(20) 320 ± 110					
Carp sucker	1960-1962	(18) 540 ± 190	(39) 940 ± 120 (39) 4800 <sup>d</sup>	(122) 1200 ± 460	(37) 640 ± 67	(22) 120 ± 30	(37) 56 ± 16	(22) 120 ± 19	(37) 32 ± 6.8
	1963	(20) 22 ± 4.4		(20) 460 ± 82					
Buffalo	1960-1962	(3) 240 ± 89	(30) 830 ± 110	(5) 480 ± 94	(30) 590 ± 92	(5) 110 ± 32	(30) 150 ± 38	(5) 78 ± 21	(30) 32 ± 6.8
	1963	(20) 43 ± 14		(21) 560 ± 84					
Sight feeders <sup>e</sup>	1960-1962	(109) 180 ± 83		(126) 680 ± 120		(127) 120 ± 32		(127) 22 ± 11	

<sup>a</sup>Total fish consists of flesh and bone.

<sup>b</sup>Parenthetical values are numbers of fish analyzed.

<sup>c</sup>± values represent 1 standard deviation.

<sup>d</sup>Includes four carpsuckers (composited) collected at CRM 19.6.

<sup>e</sup>Sight feeders include white crappie, bluegill, white bass, largemouth bass, sauger, drum, and catfish.

Source: Struxness et al. (1967)

Table A-30

RADIONUCLIDE CONTENT IN CLINCH RIVER FISH

1978

pCi/kg Wet Weight

Location	Species <sup>a</sup>	<sup>90</sup> Sr	<sup>239</sup> Pu	<sup>238</sup> Pu	<sup>238</sup> U	<sup>235</sup> U	<sup>234</sup> U	<sup>137</sup> Cs	<sup>60</sup> Co	<sup>40</sup> K	% MPI <sup>b</sup>	Hg(ng/g)	% of A.L. <sup>c</sup>
CRM 4.0	Bass	1.8	0.01	0.10	0.08	0.01	0.10	121	5.1	4117	0.03	3.5	0.7
	Blue Gill	7.8	0.07	0.08	0.16	0.16	0.34	608	10.7	4489	0.20	17.0	3.4
	Carp	4.5	0.01	0.01	0.27	0.08	0.59	76	0.1	3280	0.03	2.8	0.6
	Shad	3.7	0.04	0.02	1.90	0.26	2.23	106	13.6	3101	0.05	1.6	0.3
CRM 5.0	Bass	1.7	0.01	0.01	0.12	0.06	29.26	136	0.4	3830	0.15	7.2	1.4
	Blue Gill	3.2	0.02	0.01	0.39	0.16	0.39	122	12.0	4254	0.04	9.0	1.8
	Carp	4.6	0.01	0.01	0.23	0.17	0.41	348	0.4	2258	0.07	5.9	1.2
	Shad	5.9	0.01	0.01	0.29	0.15	0.21	181	0.5	4743	0.06	1.2	0.2
CRM 12.0	Bass	0.6	0.02	0.03	0.03	0.13	0.38	166	3.8	3891	0.18	1.9	0.4
	Blue Gill	4.9	0.03	0.02	2.63	0.42	2.77	94	6.3	3727	0.04	2.7	0.5
	Carp	2.9	0.01	0.01	1.20	0.24	1.20	71	2.9	3644	0.03	6.0	1.2
	Shad	5.5	0.16	0.22	4.20	0.47	3.89	23	11.6	5052	0.04	4.5	0.9
	Crappie	11.9	0.12	0.12	0.59	0.48	0.59	12	20.2	3590	0.09	7.6	1.5
CRM 20.8 <sup>d</sup>	Bass	42.2	0.03	0.01	0.16	0.07	0.24	10287	28.2	3925	0.51	3.1	0.6
	Blue Gill	128.0	0.18	0.75	0.39	0.14	0.53	3369	79.2	3912	1.25	3.2	0.6
	Carp	33.5	0.02	0.01	0.48	0.06	0.67	440	12.6	2044	0.28	3.1	0.6
	Shad	59.8	0.05	0.11	3.33	0.23	5.06	1208	30.7	2852	0.54	0.7	0.1
	Crappie	41.0	0.12	0.54	0.44	0.18	2.77	3293	16.6	4903	0.56	3.7	0.7
CRM 22.0	Bass	9.5	0.02	0.02	0.56	0.01	0.22	61	15.0	3890	0.07	4.3	0.8
	Blue Gill	19.3	0.03	0.02	0.23	0.30	0.68	175	23.2	3617	0.15	5.0	1.0
	Carp	2.6	0.01	0.01	0.06	0.06	0.23	164	3.8	3840	0.03	1.4	0.3
	Shad	4.8	0.01	0.01	1.86	0.04	2.70	300	14.5	3350	0.07	0.6	0.1
	Crappie	4.8	0.05	0.01	0.13	0.12	0.45	48	7.9	3168	0.01	0.8	0.2
CRM 24.0	Bass	1.3	0.01	0.01	0.21	0.04	0.28	96	4.7	3428	0.02	0.8	0.2
	Blue Gill	3.1	0.06	0.03	0.14	0.13	0.32	12	7.8	3744	0.02	3.4	0.7
	Carp	1.5	0.17	0.03	0.08	0.03	0.14	25	3.6	3648	0.01	1.5	0.3
	Shad	2.4	0.01	0.01	0.92	0.14	1.15	27	5.5	3288	0.02	0.1	0.03

<sup>a</sup>Composite of 10 fish in each species.

<sup>b</sup>Maximum Permissible Intake - Intake of radionuclide from eating fish is calculated to be equal to a daily intake of 2.2 liters of water, over a period of one year, containing the concentration guide of radionuclides in question. Consumption of fish is assumed to be 16.8 kg/yr of the species in question. Only man-made radionuclides were used in the calculation.

<sup>c</sup>Percent of proposed FDA action level of 500 ng/g.

<sup>d</sup>Average of quarterly samples.

Source: UCC (1979)

Table A-31

Comparison of the mean concentration ( $\pm 1$  standard error) of seven trace elements in axial muscle of bluegill collected at five sites (approximate locations given in parentheses)

n = 10 for all sites except WOCK 0.2 (n = 4)

Site	Mean weight (g)	Mean concentration, $\mu\text{g/g}$ wet wt						
		Cd <sup>a</sup>	Cr <sup>b</sup>	Cu	Hg <sup>c</sup>	Ni	Pb	Zn <sup>d</sup>
White Oak Lake (WOCK 1.1)	86.2 (4.8)	5.7 (1.8)	0.027 (0.003)	0.16 (0.01)	0.70 (0.07)	0.46 (0.14)	0.039 (0.006)	5.9 (0.2)
White Oak Creek embayment (WOCK 0.2)	48.0 (5.4)	3.9 (2.5)	0.042 (0.008)	0.31 (0.17)	0.57 (0.07)	0.22 (0.15)	0.040 (0.008)	10.1 (3.0)
Clinch River (CRK 30.6)	85.6 (8.4)	20.7 (6.6)	0.056 (0.012)	0.31 (0.10)	0.06 (0.01)	0.60 (0.16)	0.061 (0.026)	5.4 (0.5)
Clinch River (CRK 35.4)	77.2 (7.0)	9.7 (3.5)	0.038 (0.004)	0.19 (0.03)	0.21 (0.10)	0.88 (0.36)	0.044 (0.007)	5.8 (0.4)
Melton Hill Reservoir (CRK 84)	89.7 (7.5)	17.8 (5.0)	0.030 (0.002)	0.16 (0.02)	0.06 (<0.01)	0.40 (0.06)	0.027 (0.002)	5.4 (0.3)

<sup>a</sup> ng/g, wet wt. "Less than" values were ignored in the computation of mean concentration. In the order listed, n = 9, n = 2, n = 6, n = 8, n = 10, respectively. No statistical analysis performed.

<sup>b</sup> Mean concentration at CRK 30.6 was significantly different from that at sites WOCK 1.1 and CRK 84 (p < 0.05).

<sup>c</sup> ng/g, wet wt. Mean concentration at sites WOCK 1.1 and WOCK 0.2 was not significantly different (p > 0.05), but the concentration at both sites was significantly different from that at the other three stations (p < 0.05).

<sup>d</sup> Mean concentration at WOCK 0.2 was significantly different from that at the other four sites (p < 0.05).

Table A-32

Unadjusted and adjusted (normalized) mean concentration ( $\pm 2$  SE) of total mercury in axial muscle of bluegill from East Fork Poplar Creek (ERPC) and Bear Creek (BC). Values in parentheses are the minimum and maximum concentrations. Mean concentrations for EFPC sampling stations 7, 5, and 1 were adjusted by normalizing to the average concentration of a 63-g bluegill using the regression equation of total mercury concentration on fish weight calculated from data for each sampling station

Stream-sampling station	Mean weight (g)	Total mercury concentration ( $\mu\text{g/g}$ , fresh wt)	
		Unadjusted mean	Adjusted mean
EFPC-8	62.7	2.13 $\pm$ 0.61 (1.70 - 3.60) n=7	-- <sup>a</sup>
EFPC-7	61.1	1.56 $\pm$ 0.38 (0.66 - 2.5) n=11	1.66 $\pm$ 0.32 <sup>b</sup>
EFPC-5	54.6	1.39 $\pm$ 0.31 (0.73 - 2.20) n=11	1.45 $\pm$ 0.26 <sup>c</sup>
EFPC-1	32.5	0.56 $\pm$ 0.08 (0.32 - 0.72) n=11	0.66 $\pm$ 0.14 <sup>d</sup>
BC-1	32.4	0.41 $\pm$ 0.22 (0.33 - 0.51) n=3	--

<sup>a</sup>Regression coefficient not significantly different from zero ( $P > 0.90$ ).

<sup>b</sup>Total Hg concentration = 0.9180 + (0.0118) (fish weight in g);  
 $r^2 = 0.45$ .

<sup>c</sup>Total Hg concentration = 0.9824 + (0.0075) (fish weight in g);  
 $r^2 = 0.28$ .

<sup>d</sup>Total Hg concentration = 0.4418 + (0.0035) (fish weight in g);  
 $r^2 = 0.25$ .

Source: Van Winkle et al. (1984)

Table A-33

INSTREAM CONTAMINANT STUDY - TASK 4  
 MERCURY CONCENTRATIONS IN FISH AND OTHER AQUATIC ORGANISMS

Sampling Location Species	Number of Fish	Mercury Concentration (mg/kg)	
		Mean	Range
Scarboro Creek			
Channel catfish	10	U*	U
Largemouth bass	10	.11	U-.44
Bluegill	10	.06	U-.21
McCoy Branch			
Channel catfish	9	U	U
Yellow bullhead	1	U	U
Largemouth bass	10	.08	U-.17
Bluegill	10	.04	U-.14
Melton Hill Dam			
Largemouth bass	10	.11	U-.23
Bluegill	9	.02	U-.14
White Oak Lake			
Largemouth bass	2	.41	.24-.57
Bluegill	10	.23	U-.46
Yellow bass	7	.07	U-.23
White Oak Creek Mile 0.2			
Channel catfish	8	.06	U-.16
Black bullhead	2	.06	U-.11
Largemouth bass	5	.13	U-.36
Bluegill	10	.16	U-.56
Striped bass - hybrid	5	.11	U-.15
Clinch River Mile 11.0			
Smallmouth buffalo	10	.46	U-1.2
Largemouth bass	10	.34	.19-.58
Bluegill	10	.16	U-.40
Clinch River Mile 6.0			
Largemouth bass	10	.31	.20-.56
Bluegill	10	.19	.12-.33
Clinch River Mile 2.0			
Largemouth bass	10	.10	U-.26
Bluegill	10	.03	U-.13
Emory River Mile 1.0			
Largemouth bass	10	.12	U-.32
Bluegill	10	.05	U-.20



Table A-33

(CONTINUED)

INSTREAM CONTAMINANT STUDY - TASK 4  
MERCURY CONCENTRATIONS IN FISH AND OTHER AQUATIC ORGANISMS

Sampling Location Species	Number of Fish	Mercury Concentration (mg/kg)	
		Mean	Range
Tennessee River Mile 572			
Largemouth bass	10	.16	U-.45
Bluegill	10	.02	U-.17
Sauger	2	.30	.30-.30
Paddlefish	1	U	U
Tennessee River Mile 558			
Largemouth bass	10	.05	U-.14
Bluegill	10	.04	U-.18
East Fork Poplar Creek Mile 13.8			
Common carp	4	1.0	.57-1.3
Largemouth bass	8	1.3	.80-1.9
Bluegill (composite)	2	.68	.54-.82
Redbreast sunfish	10	1.7	.24-3.3
Frogs	10	1.6	U-3.0
Snapping turtles	5	.72	.43-1.2
Crayfish	2	.82	.43-1.2
East Fork Poplar Creek Mile 8.8			
Bluegill	6	.80	.51-1.0
Redbreast sunfish	10	.96	.64-1.4
Snapping turtle	5	.63	.16-1.0
Crayfish (composite)	4	.22	.24-.72
East Fork Poplar Creek Mile 4.0			
Bluegill	5	.89	.20-1.2
Redbreast sunfish	2	.67	.64-.70
Green sunfish	1	.52	.52
Warmouth	1	.96	.96
Rock bass	1	1.0	1.0
Yellow perch	1	.93	.93
White sucker	2	.97	.54-1.4
Black redhorse	1	.57	.57
Gizzard shad (composite)	1	.12	.12
Snapping turtle	4	.95	.41-1.4
Crayfish (composite)	1	.29	.29
Bear Creek Mile 0.4			
Bluegill	2	.59	.52-.66
Redbreast sunfish	2	U	U
Rock bass	10	.31	.17-.43
White sucker	4	.38	.24-.49
Northern hog sucker	3	.23	.17-.27
Frog	1	U	U
Crayfish (composite)	1	U	U

**Table A-33**  
(CONTINUED)

INSTREAM CONTAMINANT STUDY - TASK 4  
MERCURY CONCENTRATIONS IN FISH AND OTHER AQUATIC ORGANISMS

Sampling Location Species	Number of Fish	Mercury Concentrations (mg/kg)	
		Mean	Range
Poplar Creek Mile 0.2			
Channel catfish	10	.14	U-.42
Largemouth bass	10	.50	.24-1.3
Bluegill	10	.38	.18-.82
Smallmouth buffalo	7	.93	.13-1.7
Striped bass - hybrid	3	.04	U-.12
White bass	1	U	U
Sauger	2	.84	.37-1.3

\*U - Below minimum detection amount of 0.10 mg/kg

Source: TVA (1985e)

Table A-34

INSTREAM CONTAMINANT STUDY - TASK 4  
SIGNIFICANT MAXIMUM CONCENTRATIONS FOR RADIOISOTOPES IN FISH FLESH SAMPLES - pCi/g, DRY WEIGHT

Analysis/Isotope	NRC Reporting Level <sup>a</sup>	Tennessee River <sup>b</sup>	Lower Limit of Detection <sup>c</sup>	Clinch River	East Fork Poplar Creek	White Oak Lake	White Oak Embayment	Bear Creek
Gross Alpha	d	5	0.1	.07	.9	.05	.07	.02
Gross Beta	d	45	0.1	64	74	76	40	29
Sr-89	d	1.2	0.5	.1	.2	.2	-1.6 <sup>e</sup>	f
Sr-90	d	0.3	0.1	.01	.06	.25	1.3	f
Gamma Spectral Analysis <sup>g</sup>								
Co-60	40	.03	.01	-	.03	.12	.07	-
Cs-134	4	-	.08	-	.11	.04	-	-
Cs-137	8	.2	.02	18	2.6	26	9.7	.4
K-40	d	20	1.00	18	22	17	14	18

- a - Reporting levels for radioactivity concentrations in environmental samples as outlined in Draft NUREG-0472, Rev. 3, Standard Radiological Effluent Technical Specifications For Pressurized Water Reactors, January 1983.
- b - Maximum concentrations reported by TVA in fish samples collected from the Tennessee River from 1981-1983.
- c - Lower limit of detection as determined by the method developed by Pasternack and Harley and described in HASL-300 and Nucl. Instr. Methods 91, 533-40 (1971).
- d - No reporting level given.
- e - Negative value is an artifact of counting statistics and does not infer a negative activity.
- f - Analysis not performed.
- g - Dash indicates isotope not identified in gamma spectral analysis.

Source: TVA (1985e)

Table A-35

Summary (Minimum, Maximum, and Mean) for Lengths, Weights, Lipid Contents, and PCB Concentrations in Catfish from Watts Bar and Melton Hill Reservoirs During Each Year of Study

Location	Year	Species <sup>a</sup>	N <sup>b</sup>	Length (mm)			Weight (g)			Lipid content (%)			PCBs µg/g		
				Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean	Minimum	Maximum	Mean
<b>Watts Bar</b>															
TRM 532	1988	CHC	10	398	706	531	494	4210	1763	0.7	16.0	4.6	<0.1	4.3	1.4
TRM 565	1987	CHC	6	310	561	470	239	1786	1103	1.4	3.8	2.5	0.1	4.4	1.4
TRM 565	1988	CHC	10	390	657	492	411	2765	1124	0.9	13.0	5.5	1.3	7.5	2.7
TRM 573	1987	CHC	10	436	640	492	806	2814	1225	1.5	8.3	4.9	0.9	3.0	2.1
TRM 573	1988	CHC	10	346	615	450	264	2425	929	0.2	7.6	3.7	0.1	7.4	2.1
TRM 585	1987	CHC	5	383	556	460	327	1566	860	2.0	7.2	5.3	0.4	3.2	2.2
		BLC	5	445	535	480	718	1327	941	1.1	7.7	3.9	0.3	1.4	1.1
	1988	-	-	-	-	-	-	-	-	-	-	-	-	-	-
TRM 598	1986	CHC	3	360	523	457	336	1330	757	3.3	7.3	5.3	2.9	4.3	3.6
		BLC	1	411	411	-	555	555	-	1.2	1.2	-	0.6	0.6	-
	1987	CHC	10	395	609	477	521	2578	1098	1.9	10.0	4.6	0.4	3.1	1.5
	1988	CHC	10	452	659	504	829	2957	1289	2.1	8.5	5.2	0.8	4.4	2.4
TRM 602	1985	CHC	10	383	546	430	430	1860	771	6.0	12.7	8.8	0.2	2.0	1.4
	1986	CHC	7	356	493	446	400	1300	829	1.9	6.4	4.4	0.9	3.4	2.1
		BLC	3	327	474	415	365	890	557	0.9	1.4	1.1	0.3	0.8	0.5
	1987	-	-	-	-	-	-	-	-	-	-	-	-	-	-
	1988	-	-	-	-	-	-	-	-	-	-	-	-	-	-
CRM 2.0	1988	CHC	8	435	605	510	745	2262	1278	<0.1	11.0	5.3	<0.1	4.6	2.2
CRM 19	1988	CHC	8	309	495	407	340	1242	660	2.2	5.7	3.5	0.2	2.4	0.6
<b>Melton Hill</b>															
CRM 21	1988	CHC	10	370	790	513	406	6118	1774	1.0	11.5	3.8	<0.1	1.6	0.5
CRM 39 <sup>c</sup>	1988	CHC	8	553	690	620	1748	3906	2630	-	-	-	-	-	-
CRM 50 <sup>c</sup>	1988	CHC	8	462	640	531	1010	3470	1587	-	-	-	-	-	-

a. CHC = channel catfish; BLC = blue catfish

b. N = number of fish

c. Tissue information available for only one fish at each site due to technical problems in the laboratory; see pages 16 and 17 for further explanation and table A-4 (page 48) for PCB and lipid data.

Source: Dycus (1990)

Table A-36

Milk Results for Samples Collected Within 25 Miles  
of ORNL, 1961-1984  
(pCi/liter)

Year	Local		Remote	
	Sr-90	I-131	Sr-90	I-131
1961	11	8		
1962	33	96	20	30
1963	43	13		
1964	20	11	23	
1965	19.8	7.4	20.4	
1966	26.1	9.0	24.4	
1967	28	17	25	6.4
1968	20	5.7	19	
1969	17.8	5.9	15.7	
1970	11.4	<10	9.3	<10
1971	11.4	<10	9.4	<10
1972	11	<11.4	8.6	<13.3
1973	8.1	<10	6.1	<10
1974	4.1	<0.49	2.7	<.65
1975	3.7	<0.56	3.2	<.47
1976	2.8	3.0	2.8	5.4
1977	3.4	<1.9	2.5	<1.1
1978	3.1	<0.76	2.5	<0.59
1979	2.5	<0.52	1.9	<0.45
1980	1.7	<0.45	1.6	<0.45
1981	1.6	<0.45	1.3	<0.45
1982	1.4	<0.45	1.4	<0.45
1983	1.2	<0.45	1.1	<0.45
1984	<1.6	<0.8	<1.2	<0.8

Source: Ohnesorge (1986)

Table A-37

Average concentrations ( $\pm 1$  s.d.) of  $^{137}\text{Cs}$  and  $^{60}\text{Co}$   
in mallard ducks and American coots from White Oak Lake

Sample type	Number of samples	Concentration (pCi/g wet wt)	
		$^{137}\text{Cs}$	$^{60}\text{Co}$
<i>Mallards</i>			
Whole-body	7	$7.0 \pm 1.5$	$0.49 \pm 0.1$
Liver	7	$1.6 \pm 7.3$	<i>a</i>
Muscle	7	$3.8 \pm 1.9$	<i>a</i>
<i>Coots</i>			
Whole-body	3	$39.0 \pm 5.8$	$3.3 \pm 0.8$
Liver	3	$9.5 \pm 0.8$	<i>a</i>
Muscle	3	$16.9 \pm 3.1$	<i>a</i>

<sup>a</sup>Not detectable.

Source: MMES (1989)

Table A-38

**RADIOACTIVITY IN GRASS SAMPLES FROM PERIMETER AND REMOTE MONITORING STATIONS**  
**1978**  
 (Units of pCi/g—Dry Weight)

SAMPLING LOCATION <sup>a</sup>	<sup>7</sup> Be	<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>144</sup> Ce	<sup>239</sup> Pu	<sup>238</sup> Pu	<sup>238</sup> U	<sup>235</sup> U	<sup>234</sup> U	<sup>232</sup> Th	<sup>230</sup> Th	<sup>228</sup> Th
						<u>Perimeter<sup>b</sup></u>						
HP-31	17	.7	.4	3	.003	.001	.06	.007	.11	.04	.03	.05
HP-32	19	.7	.3	2	.002	.001	.07	.018	.19	.02	.02	.06
HP-33	17	.5	.4	3	.004	.002	.05	.005	.06	.02	.02	.03
HP-34	13	.5	.4	3	.003	.001	.02	.002	.02	.02	.02	.051
HP-35	18	.4	.3	3	.003	.001	.07	.007	.11	.03	.03	.07
HP-36	15	.6	.3	2	.004	.001	.07	.005	.08	.04	.04	.04
HP-37	17	.7	.3	2	.003	.001	.04	.004	.04	.02	.02	.02
HP-38	16	.5	.5	4	.002	.001	.10	.004	.04	.01	.01	.04
HP-39	18	.4	.3	3	.004	.001	.05	.005	.11	.01	.02	.04
Average	17	.6	.4	3	.003	.001	.06	.005	.08	.02	.02	.04
						<u>Remote<sup>c</sup></u>						
HP-51	16	.3	.1	1	<.002	<.0013	.02	.002	.004			
HP-52	17	.6	.2	1	.001	<.0002	.02	.002	.01			
HP-53	8	.4	.1	1	.001	<.0001	.01	.001	.01			
HP-54	11	.5	.6	4	.003	.0003	.07	.006	.08			
HP-55	16	.2	.2	1	.002	.0014	.03	.006	.03			
HP-56	15	.7	.1	1	.001	<.0005	.02	.008	.02			
HP-57	11	.8	.2	2	.003	<.0005	.02	.004	.02			
HP-58	12	.8	.2	1	.018	<.0005	.003	.008	.03			
Average	13	.5	.2	2	<.004	<.0006	.02	.005	.03			

<sup>a</sup>See Figures 1 and 2

<sup>b</sup>Average of two samples

<sup>c</sup>One sample

Source: UCC (1979)

Table A-39

Total mercury concentration in live and dead foliage of pasture grass along East Fork Poplar Creek, May 1982

Station number	Distance <sup>a</sup> (m)	Live foliage		Dead foliage	
		n	(Hg) <sup>b</sup>	n	(Hg) <sup>b</sup>
5	5	3	0.23 ± 0.12	3	4.43 ± 1.30
5	30	2 <sup>c</sup>	0.18 ± 0.06	3	2.13 ± 0.66
5	100	3	NA <sup>d</sup>	3	0.37 ± 0.44
3	5	1 <sup>e</sup>	0.11	3	6.97 ± 0.88
3	30	3	< 0.10	3	0.42 ± 0.26
3	100	3	NA	3	NA
Control	-	1 <sup>e</sup>	0.10	3 <sup>c</sup>	0.12 ± 0.02

<sup>a</sup>Distance from the edge of the creek.

<sup>b</sup>Mercury concentration (µg Hg/g fresh wt ± 2 SE).

<sup>c</sup>1 additional sample was < 0.10.

<sup>d</sup>NA = not analyzed.

<sup>e</sup>2 additional samples were < 0.10.

Source: Van Winkle et al. (1982)



**Table A-40**  
**RADIOACTIVITY IN SOIL SAMPLES FROM PERIMETER AND REMOTE MONITORING STATIONS**  
**1978**  
 (Units of pCi/g—Dry Weight)

SAMPLING LOCATION <sup>a</sup>	<sup>90</sup> Sr	<sup>137</sup> Cs	<sup>226</sup> Ra	<sup>234</sup> U	<sup>235</sup> U	<sup>238</sup> U	<sup>238</sup> Pu	<sup>239</sup> Pu	<sup>232</sup> Th	<sup>230</sup> Th	<sup>228</sup> Th
<u>Perimeter<sup>b</sup></u>											
HP-31	.5	2	1.0	.52	.02	.30	.001	.03	.24	.17	.27
HP-32	.9	2	0.8	.99	.04	.60	.002	.03	.33	.20	.37
HP-33	.4	3	0.8	.44	.01	.30	.002	.04	.19	.16	.21
HP-34	.5	3	1.0	.35	.02	.24	.001	.04	.37	.20	.47
HP-35	.5	2	0.8	.48	.03	.36	.003	.04	.26	.20	.24
HP-36	.6	2	1.0	.52	.02	.38	.002	.04	.29	.26	.34
HP-37	.5	1	1.0	.26	.02	.21	.0004	.02	.36	.31	.42
HP-38	.4	1	1.0	.28	.01	.21	.001	.02	.35	.22	.45
HP-39	.4	3	1.2	.67	.03	.26	.001	.03	.26	.34	.30
Average	.5	2	1.0	.50	.02	.32	.002	.03	.29	.23	.34
<u>Remote<sup>c</sup></u>											
HP-51	.3	1	1.2	.70	.02	.60	.0005	.03			
HP-52	.4	3	0.8	.50	.01	.39	.0009	.02			
HP-53	.3	2	1.8	.90	.14	.80	.0018	.03			
HP-54	.2	.3	<1.0	.22	.01	.17	.0005	.004			
HP-55	.4	2	1.3	.42	.02	.35	.0009	.02			
HP-56	.5	2	1.1	.39	.01	.35	.0009	.03			
HP-57	.8	3	1.1	.39	.01	.37	.0009	.05			
HP-58	.3	2	1.3	.50	.02	.44	.0070	.02			
Average	.4	2	<1.2	.50	.03	.43	.0017	.03			

<sup>a</sup>See Figures 1 and 2

<sup>b</sup>Average of two samples

<sup>c</sup>One sample

Source: UCC (1979)

Table A-41

## Constituents in off-site groundwater during 1990

Analyte	Number detected	Number of samples	Values above detection limit			Reference value <sup>c</sup>	Number of values exceeding reference [ref] <sup>d</sup>
			Max <sup>a</sup>	Min <sup>a</sup>	Av <sup>b</sup>		
<b>Anions (mg/L)</b>							
Chloride	21	21	62	1.0	6.1*	250	0[3]
Fluoride	13	21	6.0	0.10	0.68	4.0	1[2]
Nitrate	11	21	17	2.0	4.3*	10	1[2]
Sulfate as SO <sub>4</sub>	21	21	51	2.0	13*	200	0[3]
<b>Field measurements</b>							
Conductivity (mS/cm)	21	21	1.2	0.11	0.29*	<i>e</i>	[ <i>e</i> ]
pH (standard units)	21	21	9.2	6.9	7.5*	(6.5, 8.5)	2[3]
Temperature (°C)	21	21	24	14	19*	31	0[1]
<b>Metals</b>							
Arsenic, total (mg/L)	1	21	0.0050	0.0050	0.0050	0.050	0[2]
Barium, total (mg/L)	16	21	0.32	0.0097	0.093*	1.0	0[2]
Calcium, total (mg/L)	21	21	93	1.8	43*	<i>e</i>	[ <i>e</i> ]
Cadmium, total (mg/L)	7	21	0.022	0.0033	0.0096*	0.010	2[1]
Cobalt, total (mg/L)	1	21	0.0081	0.0081	0.0081	<i>e</i>	[ <i>e</i> ]
Copper, total (mg/L)	20	21	0.21	0.0042	0.029*	1.0	0[1]
Iron, total (mg/L)	13	21	10	0.0047	1.7	0.30	5[3]
Magnesium, total (mg/L)	21	21	29	0.69	14*	<i>e</i>	[ <i>e</i> ]
Manganese, total (mg/L)	6	21	0.73	0.0090	0.19	0.050	5[3]
Sodium, total (mg/L)	21	21	340	0.49	31*	<i>e</i>	[ <i>e</i> ]
Nickel, total (mg/L)	1	21	0.010	0.010	0.010	0.10	0[1]
Lead, total (mg/L)	9	21	0.027	0.0041	0.011*	0.050	0[1]
Uranium, total (mg/L)	4	21	0.0010	0.0010	0.0010	<i>e</i>	[ <i>e</i> ]
Uranium, total <sup>f</sup> (pCi/L)	4	21	0.66	0.66	0.66	<i>e</i>	[ <i>e</i> ]
Zinc, total (mg/L)	20	21	1.1	0.0034	0.15*	5.0	0[1]
<b>Radionuclides (pCi/L)</b>							
<sup>60</sup> Co	1	21	7.0	7.0	7.0	200	0[4]
Gross alpha	20	21	4.6	1.1	3.1*	15	0[2]
Gross beta	19	21	51	4.9	10*	50	1[2]
<sup>99</sup> Tc	5	21	2.3	0.95	1.6*	4,000	0[4]
<sup>89</sup> Sr + <sup>90</sup> Sr	11	21	3.5	0.73	1.5*	40	0[4]
<b>Tentatively identified compounds (µg/L)</b>							
Cyclohexane	8	8	6.0	JB 5.0	5.6*	<i>e</i>	[ <i>e</i> ]

<sup>a</sup>Prefixes J and B mean that the value was estimated or found in the laboratory blank, respectively.

<sup>b</sup>An asterisk (\*) follows a mean that is significantly greater than zero.

<sup>c</sup>If a reference limit exists, the source is coded as:

1. Rules of Tennessee Department of Health and Environment, Bureau of Environment, Division of Water Pollution Control, Chapter 1200-4-3, General Water Quality Criteria, February 1987.
2. 40 CFR (7-1-1989 Edition) Part 141—National Primary Drinking Water Regulations, Subpart B—Maximum Contaminant Levels.
3. 40 CFR (7-1-1989 Edition) Part 143—National Secondary Drinking Water Regulations.
4. DOE Order 5400.5, February 8, 1990, Chapter III, Derived Concentration Guides for Air and Water.

<sup>d</sup>The source of the reference limit is enclosed within brackets.

<sup>e</sup>Not applicable.

<sup>f</sup>Activity is calculated from mass assuming natural abundance.

Source: MMES (1991)

**APPENDIX B**  
**ENVIRONMENTAL MONITORING DATA:**  
**EXAMPLE FIGURES**

**IMPORTANT NOTICE**

Appendices A and B present examples of environmental monitoring data directly excerpted from the cited source documents. As such, the quality of the data summarized has not been verified, and no attempt has been made to alter the format in which the data are presented. In a number of cases, these sample tables and figures contain data that appear to be contrary to typical or expected distributions of radionuclides around nuclear facilities, expected relationships between specific radionuclides and/or gross radioactivity measurements, and customary reporting conventions for environmental data. These examples of environmental monitoring data serve to illustrate many of the aspects of environmental contaminant sampling, measurement, and reporting that will require much closer evaluation before historical environmental data can be used as a basis for estimation of off-site exposures or health risks. These tables and figures do not necessarily represent all of the data collected in a given study during a given time period. Study numbers, which refer to text summaries in Section 5.3 that can be consulted for more information, are provided for each table/figure in the attached list.

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- B-6 Average Concentrations of Mercury for Cores Sampling Sites (Study 49: Cook et al., 1992)
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- B-12 Weekly Average Concentration of  $^{131}\text{I}$  in Raw Milk in the Immediate Environs of Oak Ridge Compared with  $^{131}\text{I}$  Discharges from ORNL Stacks - 1965 (Study 77: ORNL, 1966)
- B-13 Weekly Average Concentration of  $^{90}\text{Sr}$  in Raw Milk in the Immediate Environs of Oak Ridge - 1965 (Study 77: ORNL, 1966)
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- B-19 Radiation Background Profile of the ORNL Area as Determined by Aerial Survey Techniques, 1959-1962 (Study 97: ORNL, 1963)

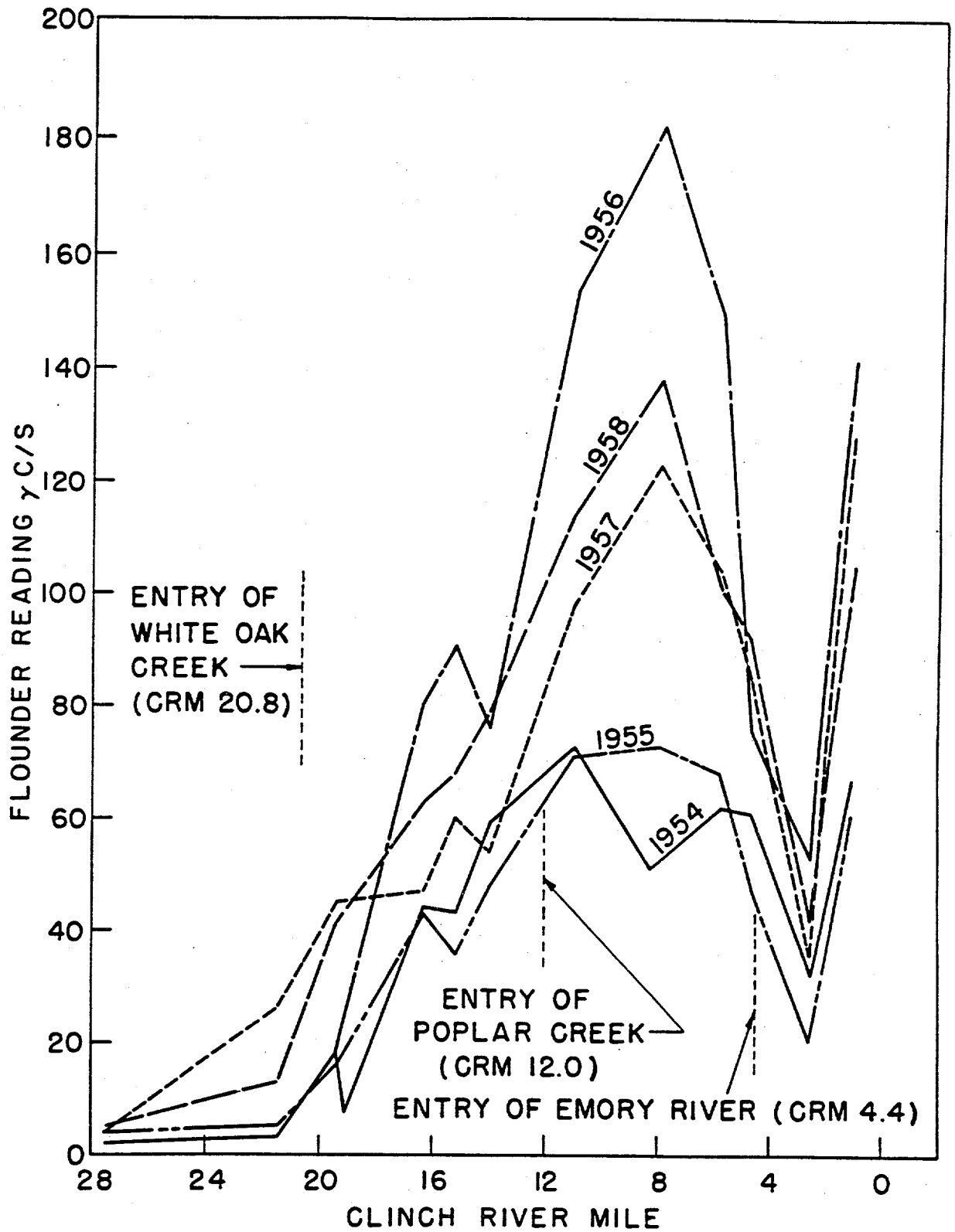


Figure B-1 Gamma Count at Surface of Clinch River Sediment.  
Source: Cottrell (1960)

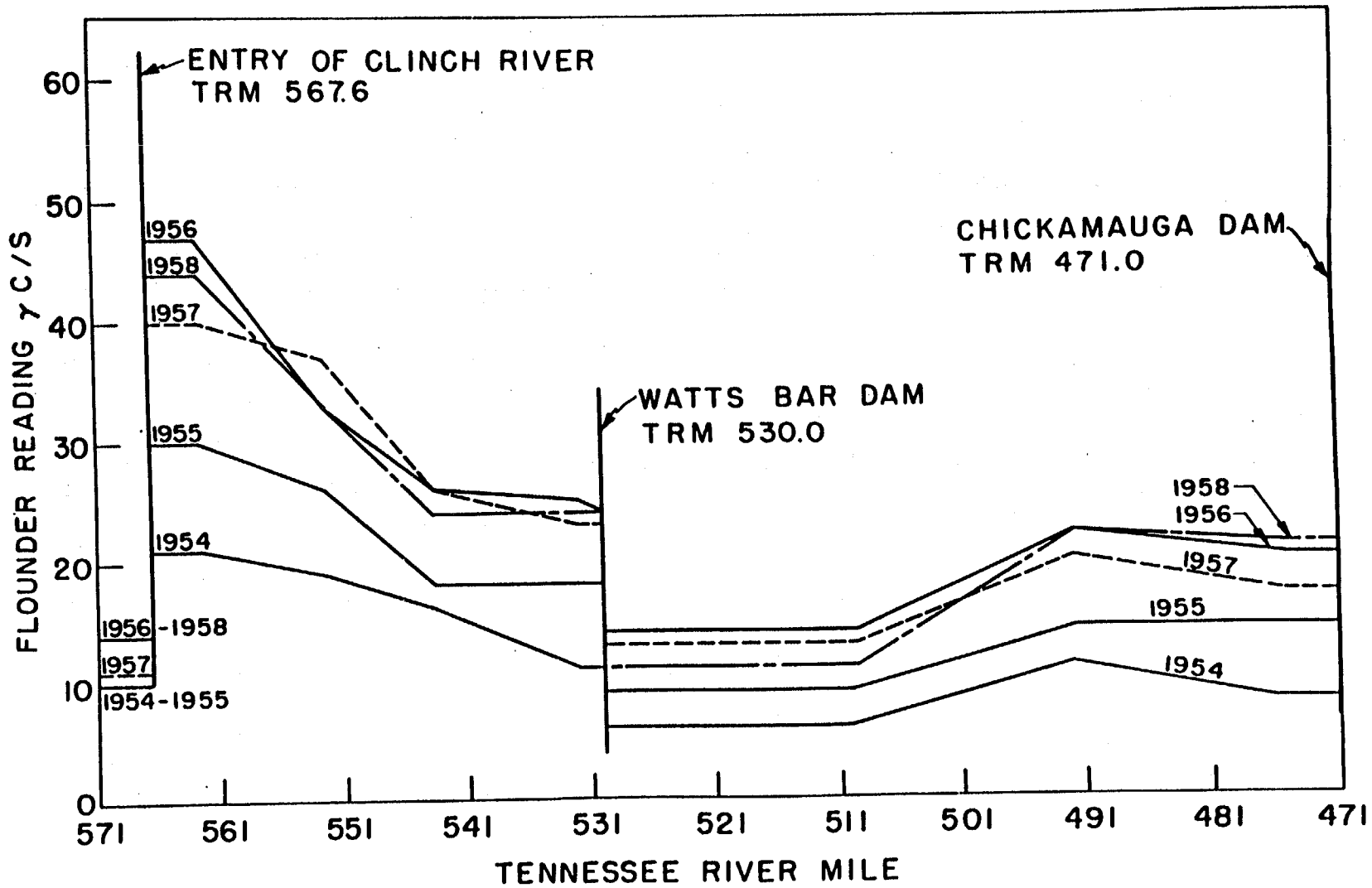
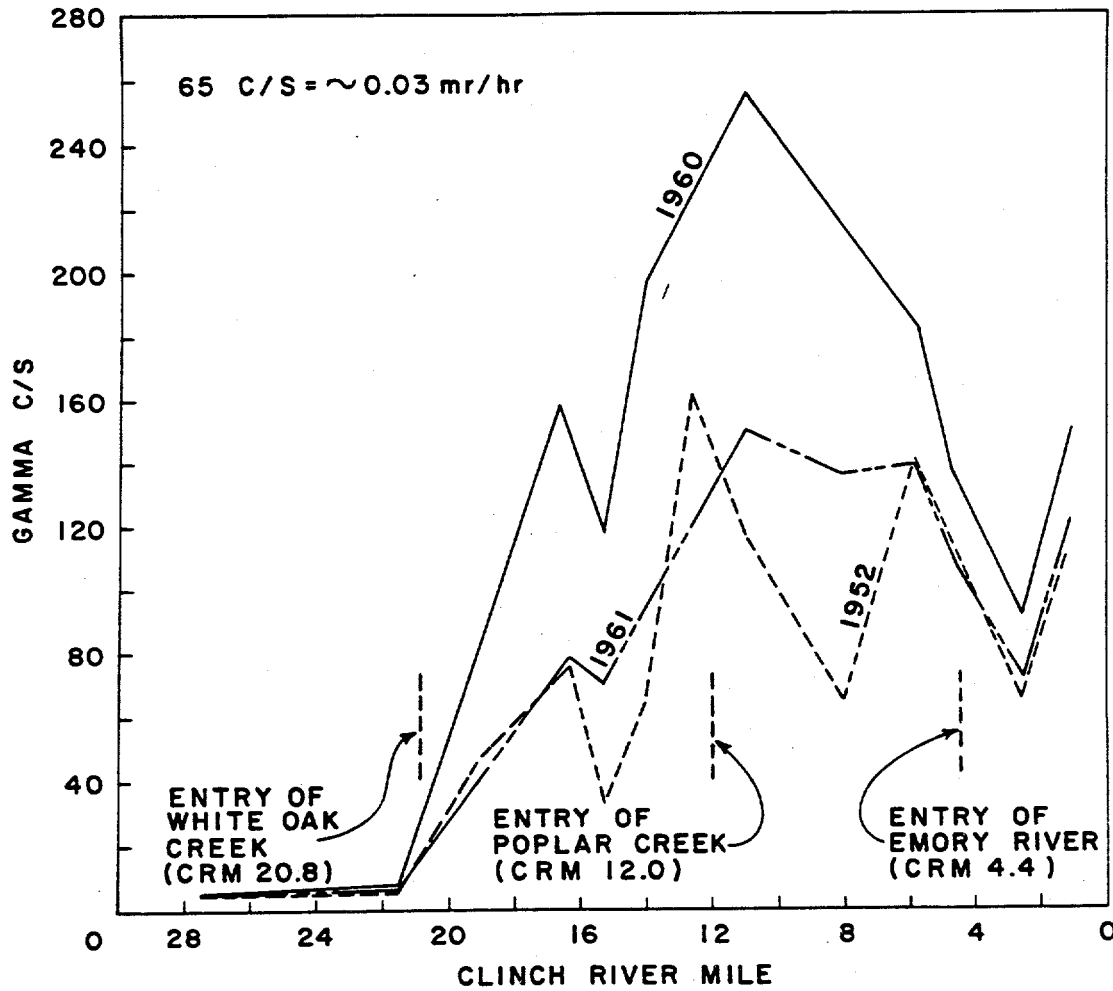


Figure B-2 Gamma Count at Surface of Tennessee River Sediment.

Source: Cottrell (1960)



**Figure B-3** Gamma Radiation Count at Surface of Clinch River Bottom Silt for the Years 1952, 1960, and 1961.

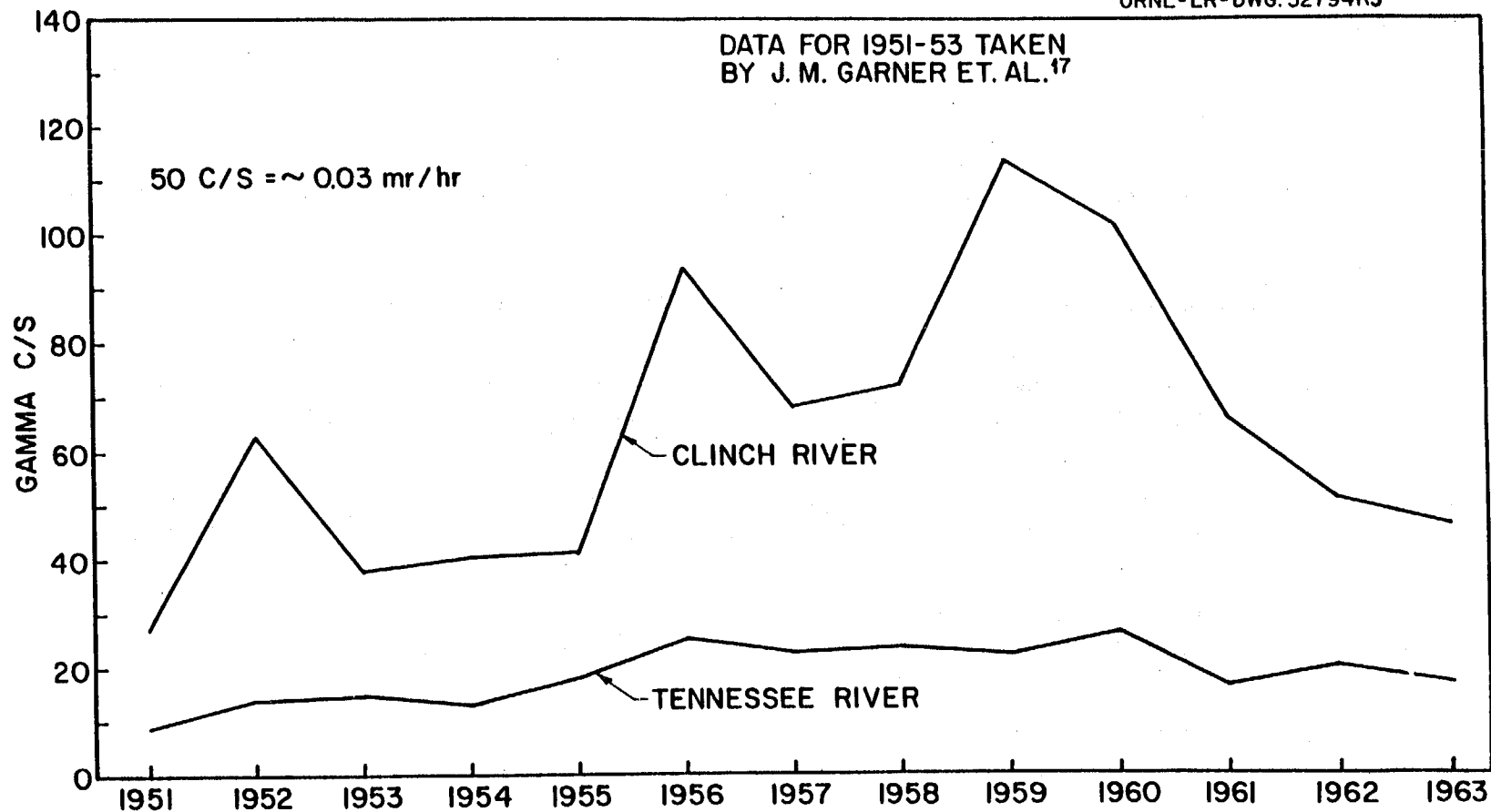


Figure B-4 Average Gamma Count at Surface of Silt Clinch and Tennessee Rivers, 1951-63.



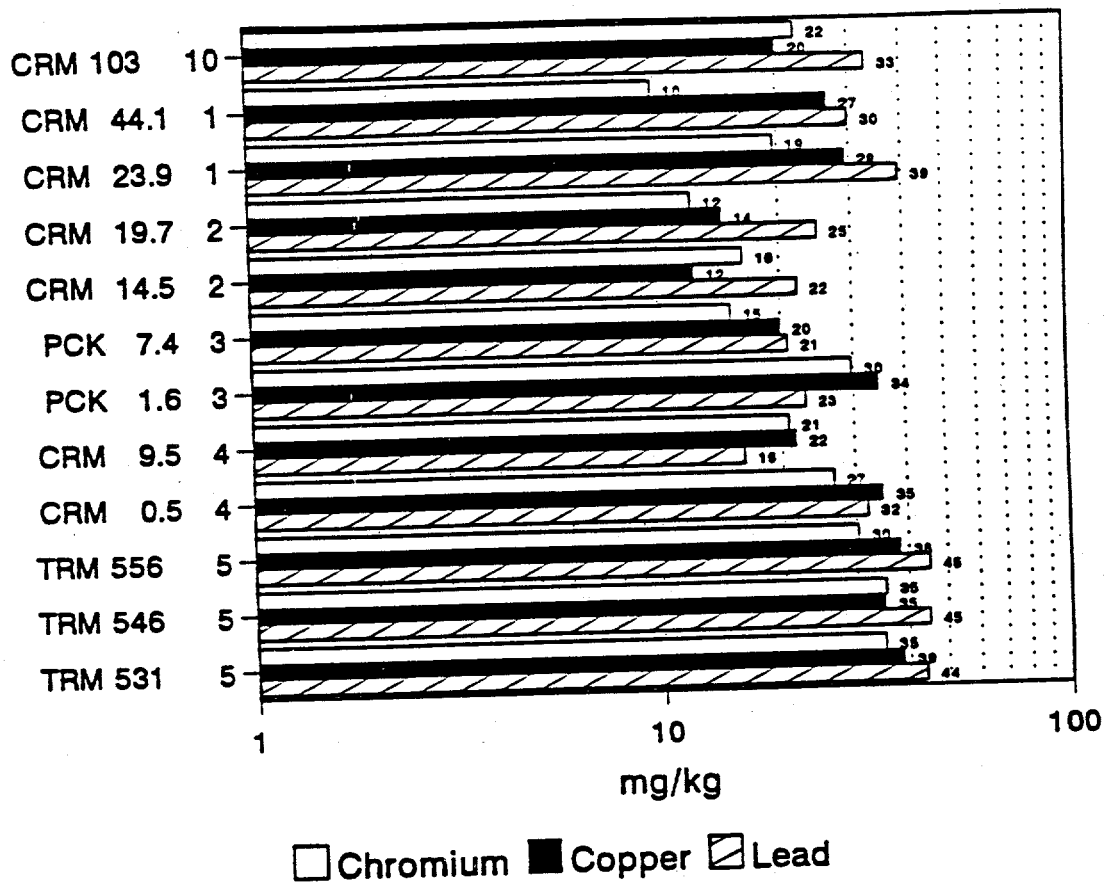


Figure B-5 Average concentrations of chromium, copper, and lead for cores sampling sites. Reaches and cores within reaches are arranged from upstream on the top to downstream on the bottom.

Source: Cook et al. (1992)

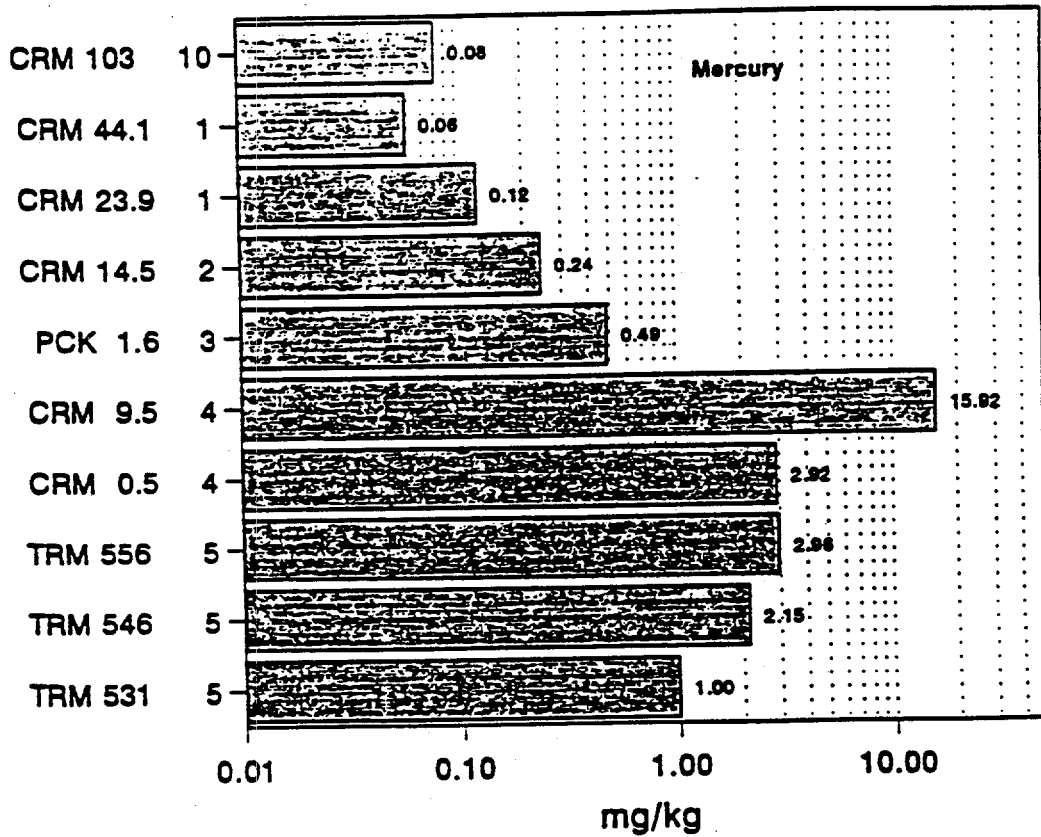
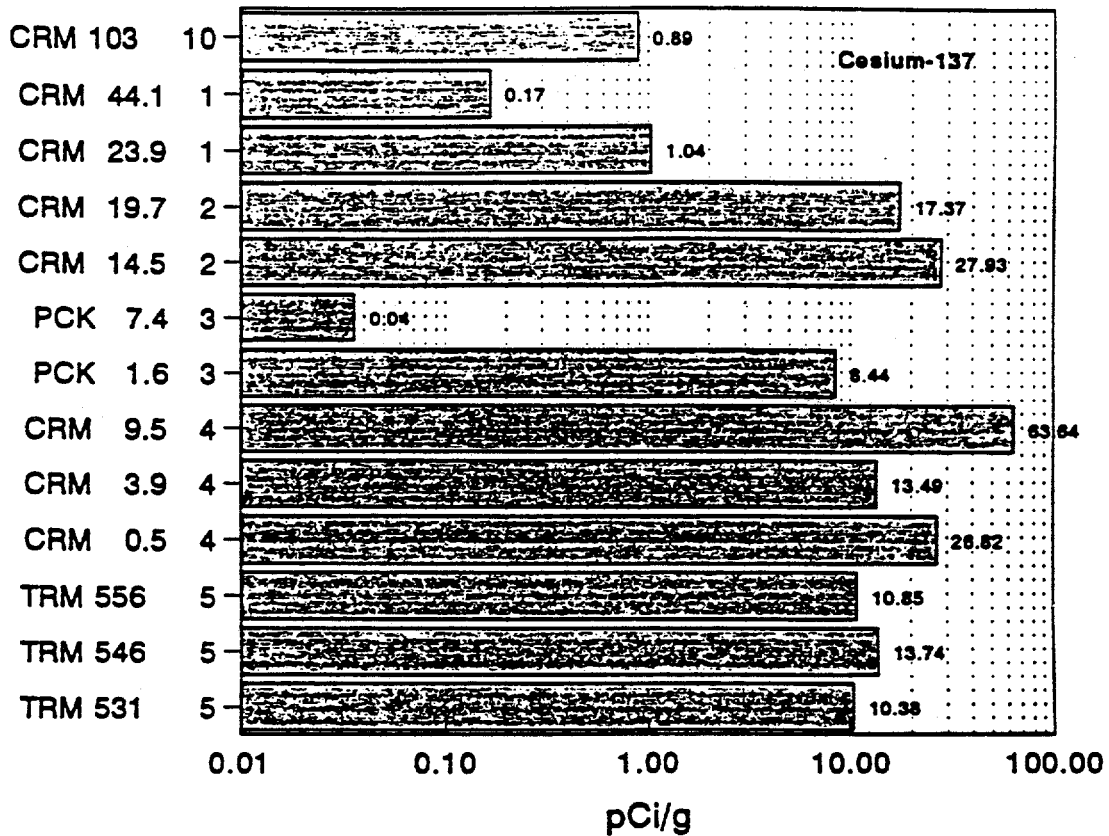


Figure B-6 Average concentrations of mercury for cores sampling sites. Reaches and cores within reaches are arranged from upstream on the top to downstream on the bottom.

Source: Cook et. al. (1992)



**Figure B-7** Average concentrations of <sup>137</sup>Cs for cores sampling sites. Reaches and cores within reaches are arranged from upstream on the top to downstream on the bottom.

Source: Cook et al. (1992)

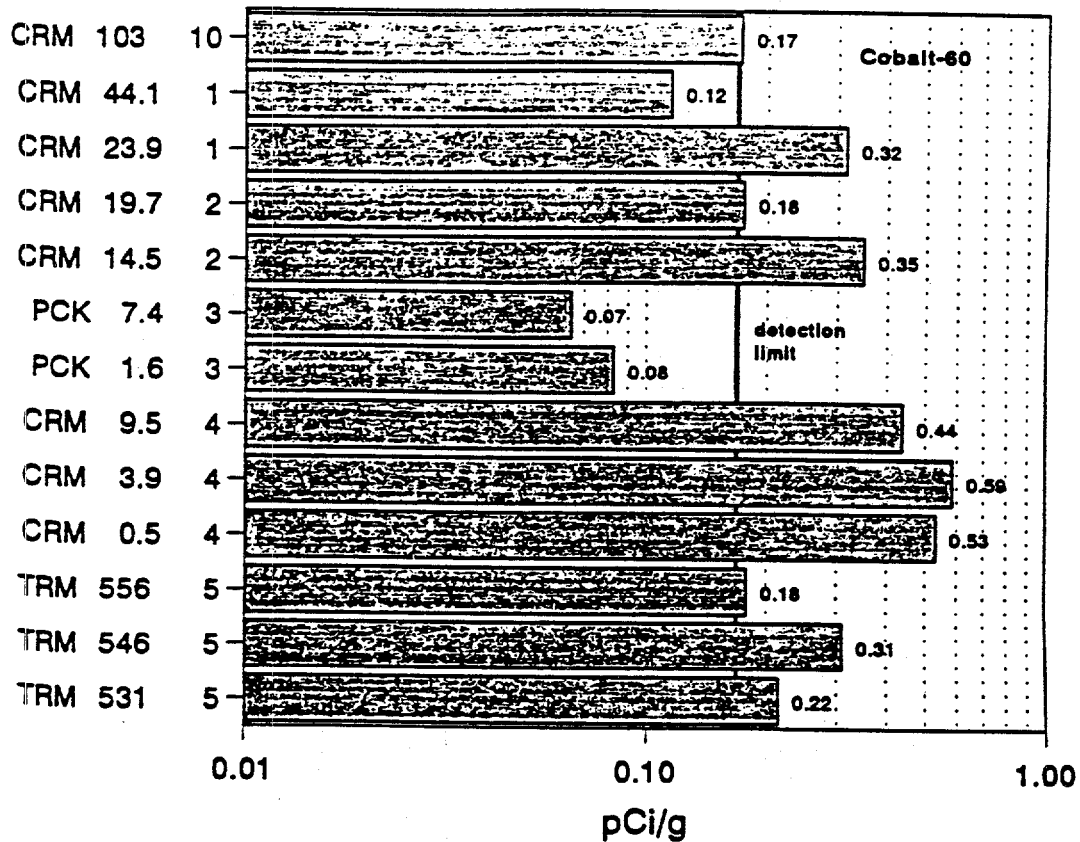


Figure B-8 Average concentrations of <sup>60</sup>Co for core sampling sites. Reaches and cores within reaches are arranged from upstream on the top to downstream on the bottom.

Source: Cook et al. (1992)

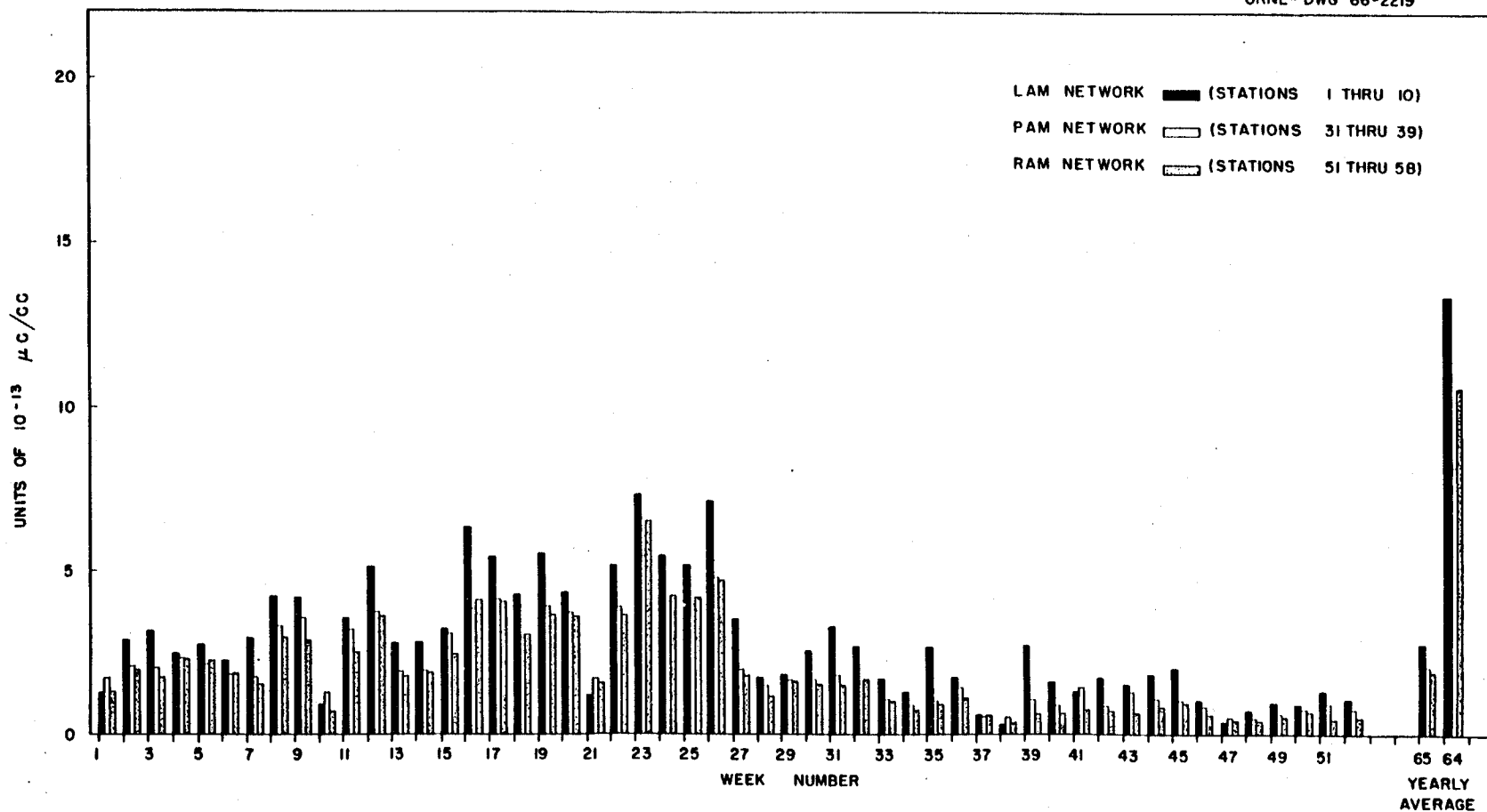
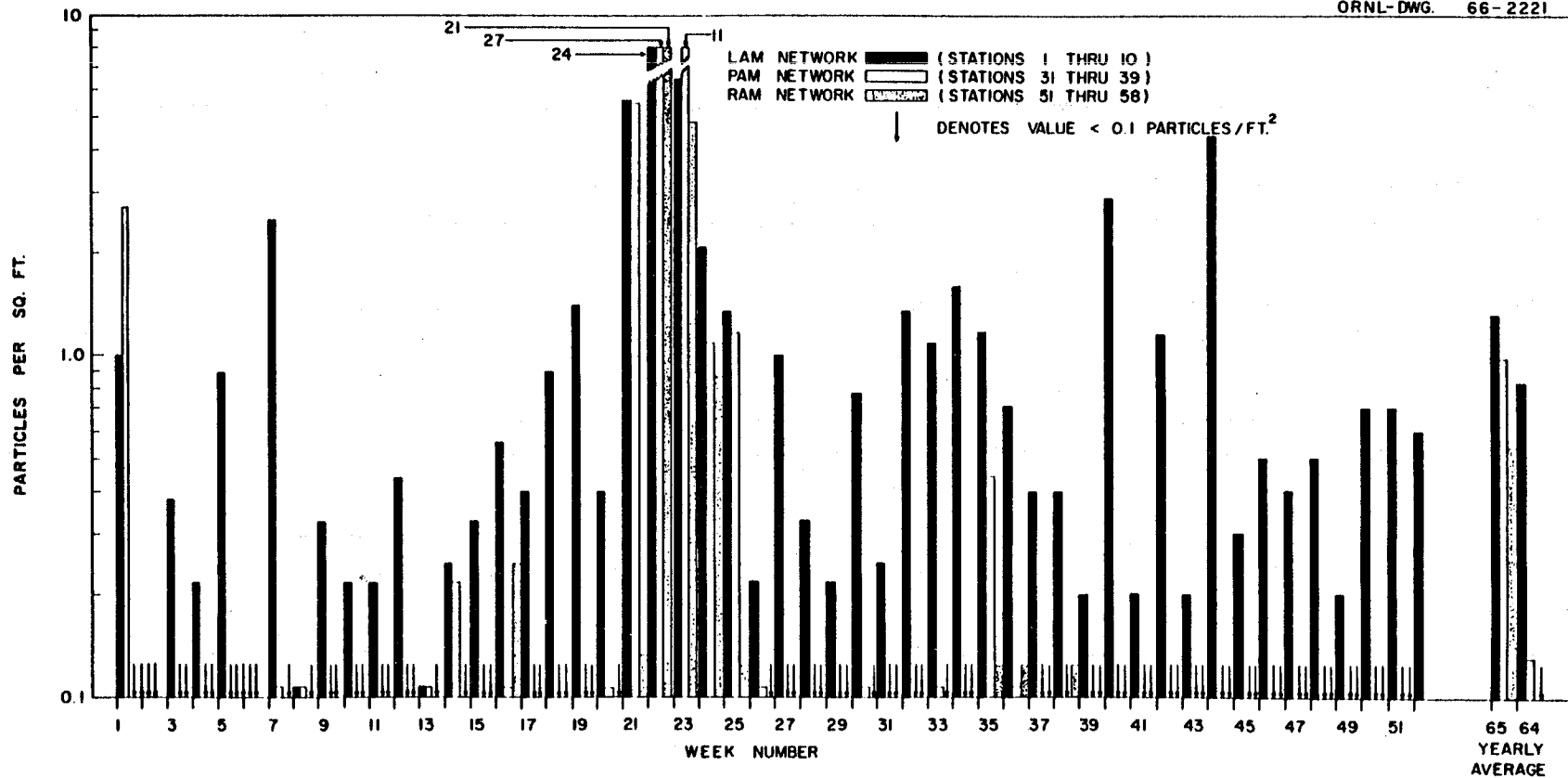


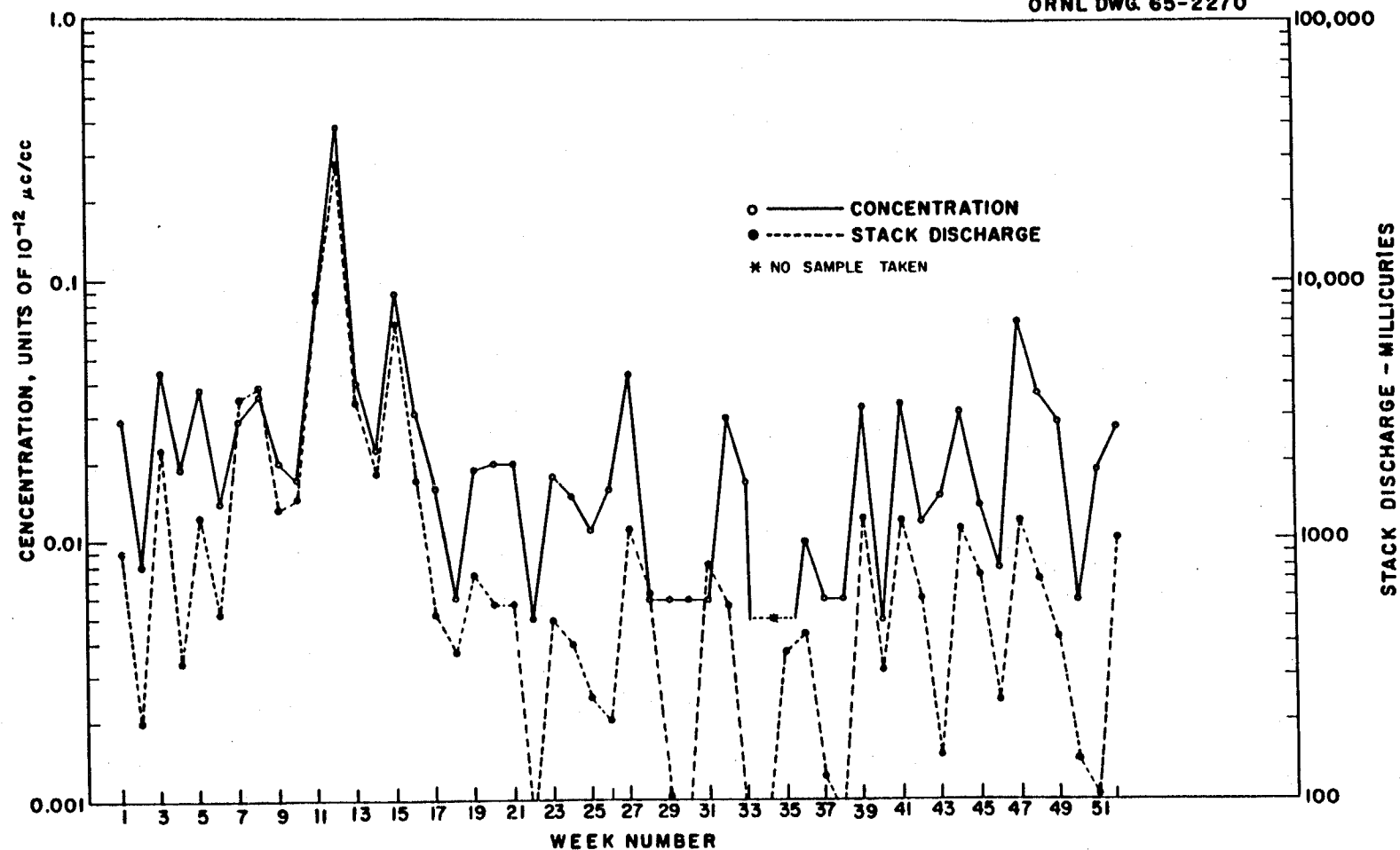
Figure B-9 Concentration of Radioactive Materials in Air as Determined from Filter Paper Data - 1965.

Source: ORNL (1966)



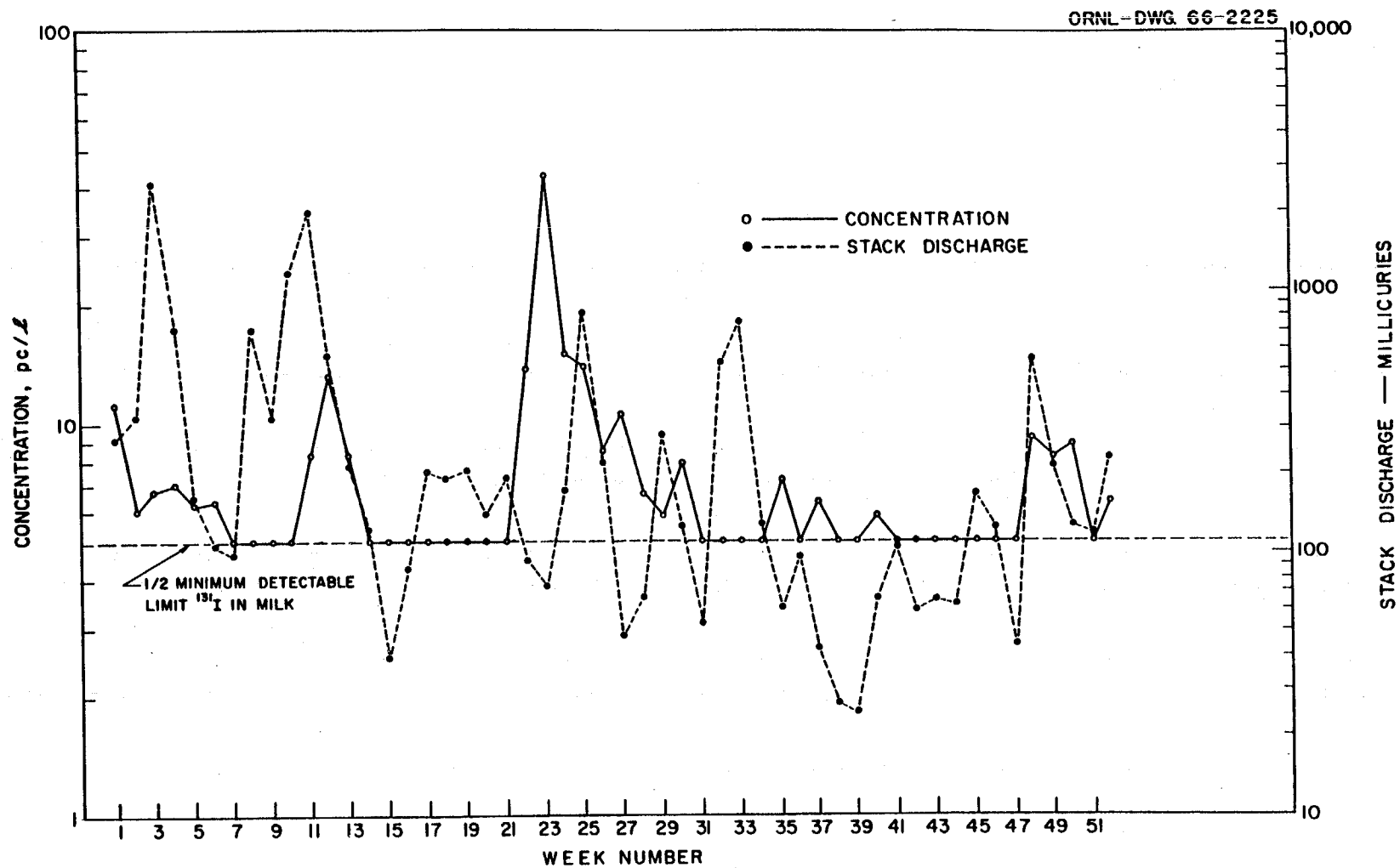
**Figure B-10** Radioparticulate Fallout Measurements as Determined by Autoradiographic Techniques Using Gummed Paper Collectors - 1965.

Source: ORNL (1966)



**Figure B-11** Weekly Average Concentration of  $^{131}\text{I}$  in Air at the Perimeter of the Controlled Area Compared with  $^{131}\text{I}$  Discharges from ORNL Stacks - 1964 (Peak during 12th week was due to accidental release of about 20 curies of  $^{131}\text{I}$ .)

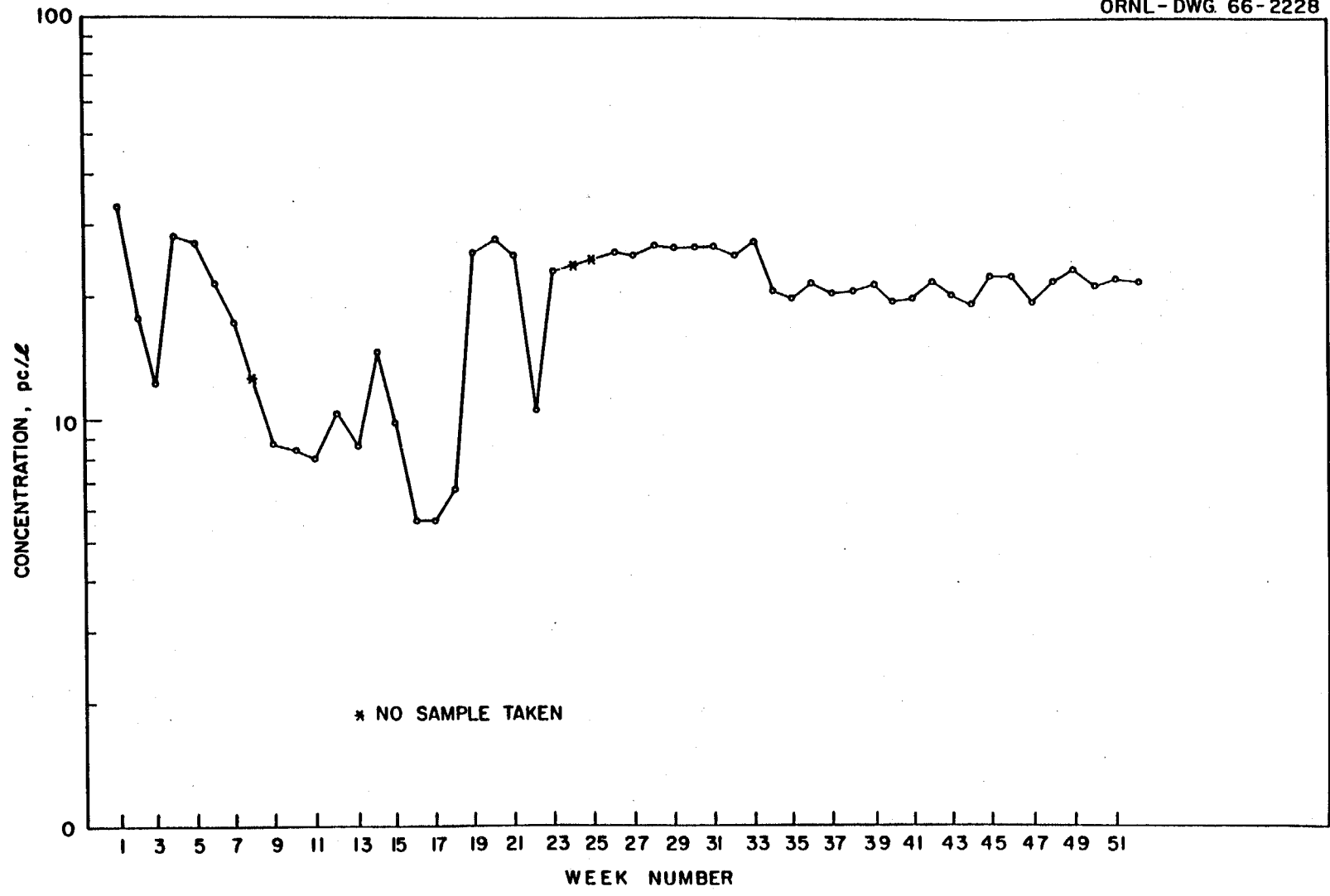
Source: ORNL (1965)



**Figure B-12** Weekly Average Concentration of <sup>131</sup>I in Raw Milk in the Immediate Environs of Oak Ridge Compared with <sup>131</sup>I Discharges from ORNL Stacks - 1965.

Source: ORNL (1966)





**Figure B-13** Weekly Average Concentration of <sup>90</sup>Sr in Raw Milk in the Immediate Environs of Oak Ridge - 1965.

Source: ORNL (1966)

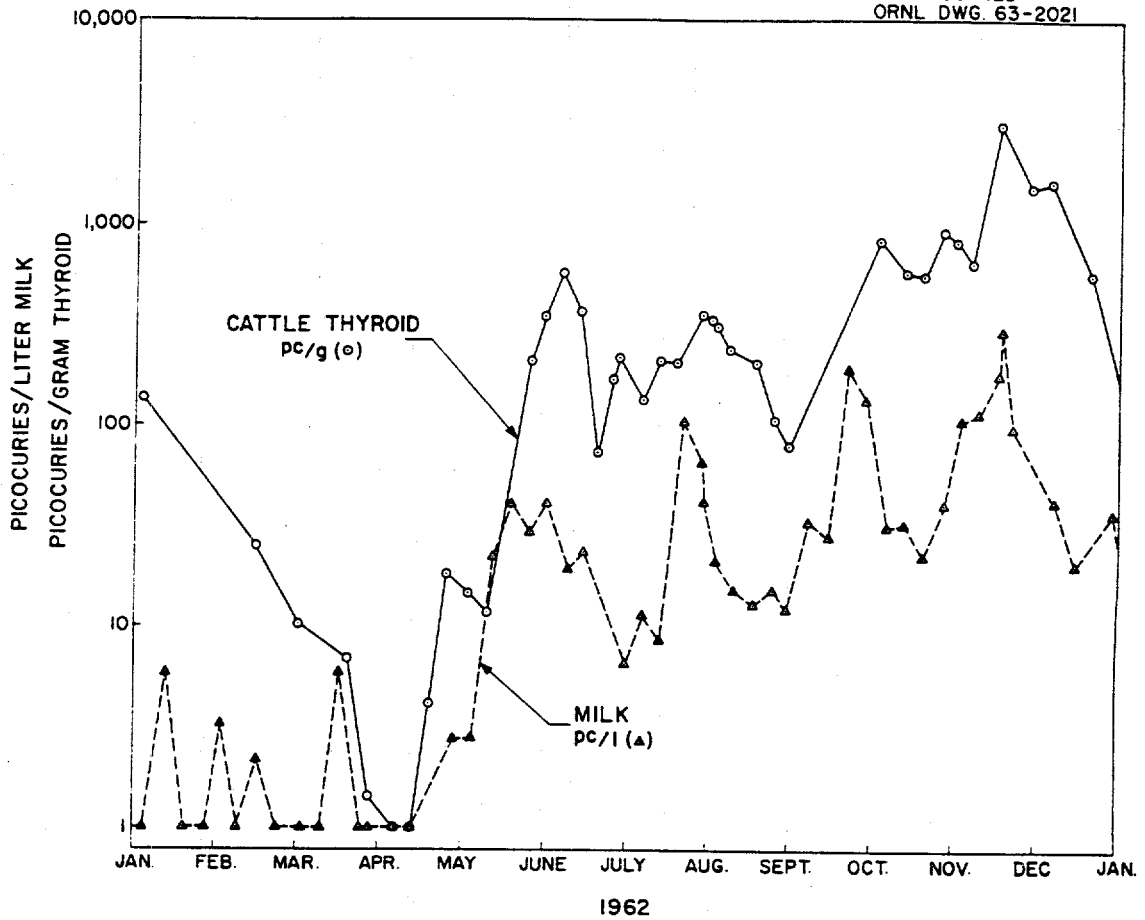


Figure B-14 Concentration of  $I^{131}$  in Milk and Cattle Thyroids, 1962

Source: ORNL (1963)

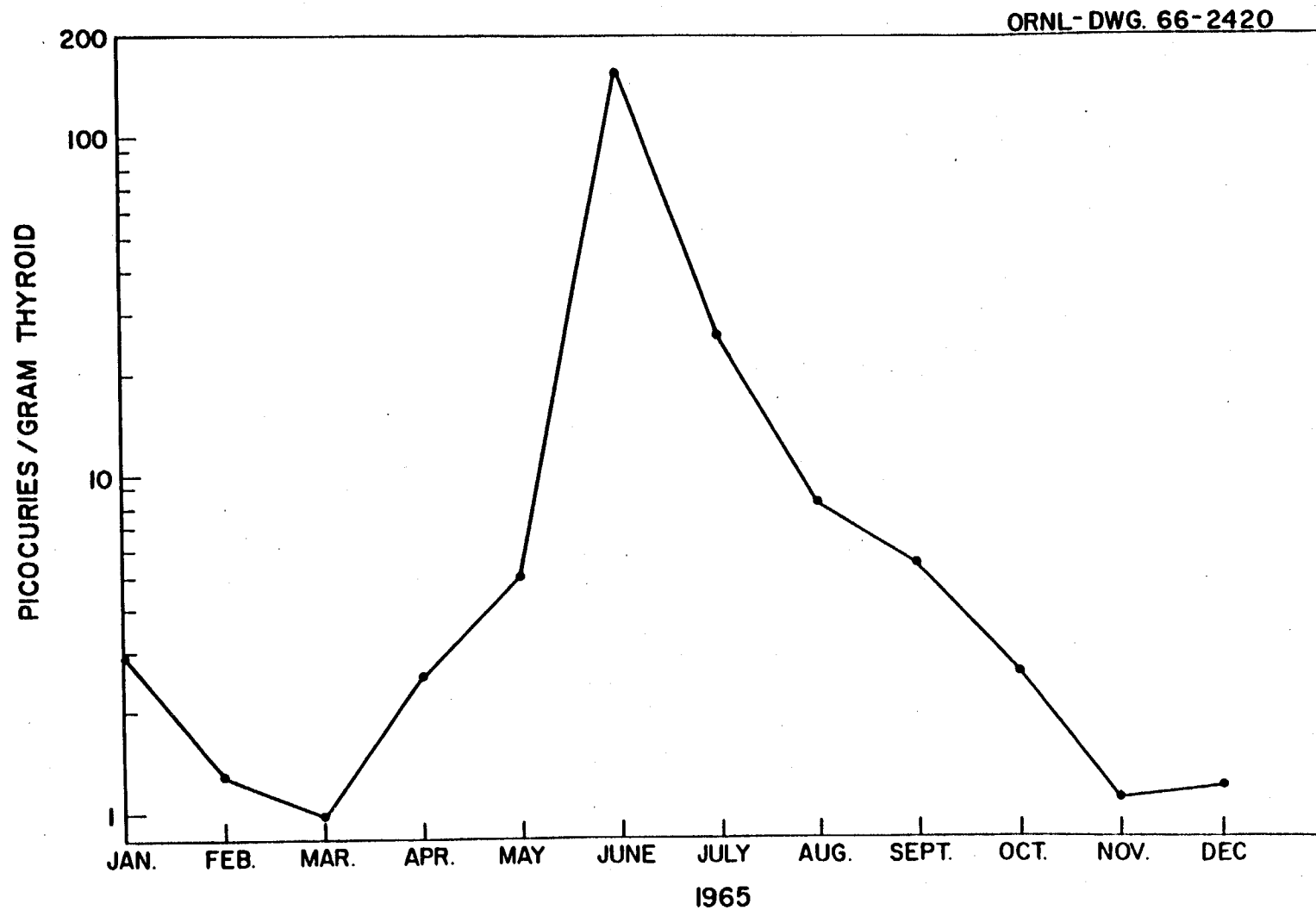
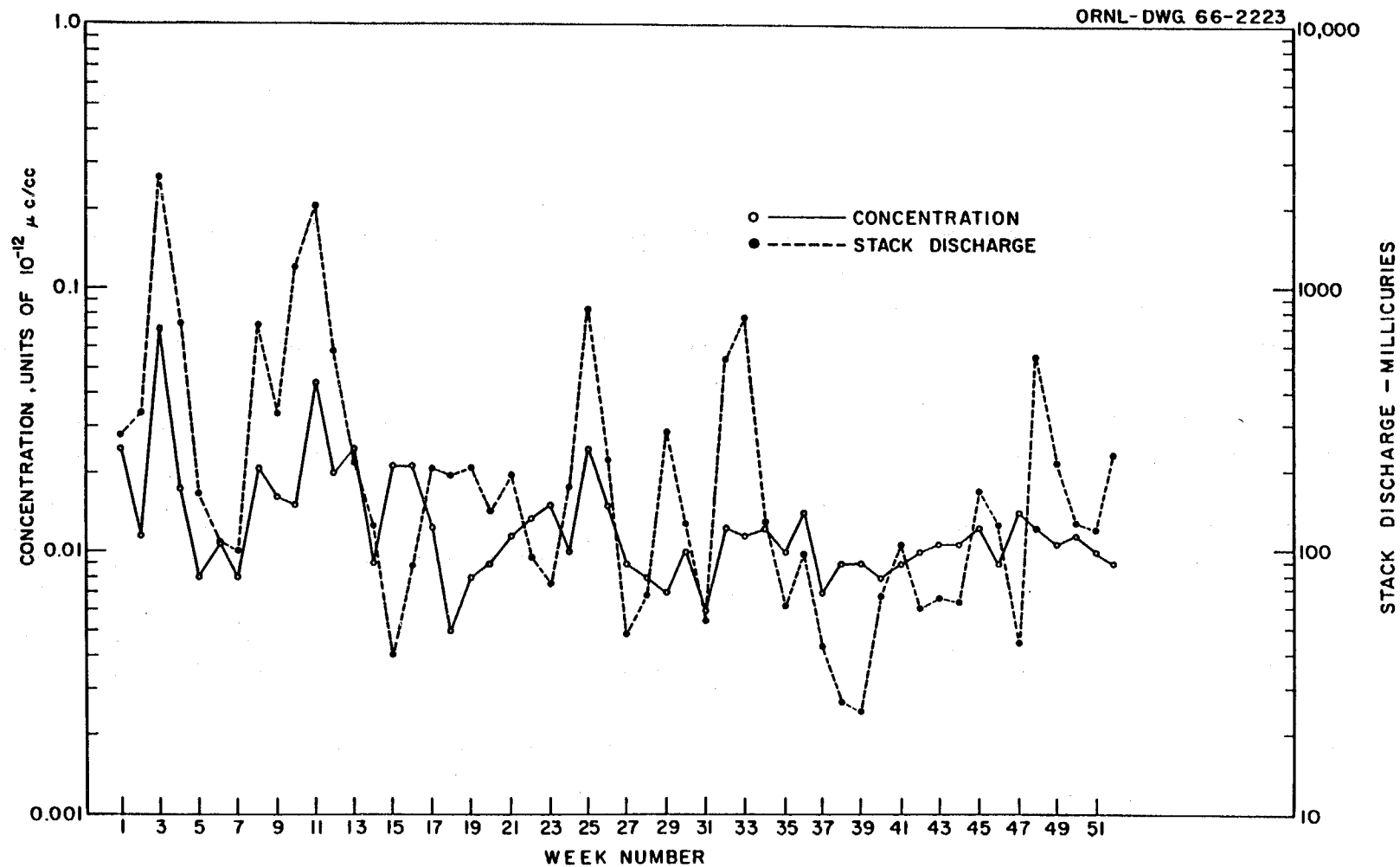


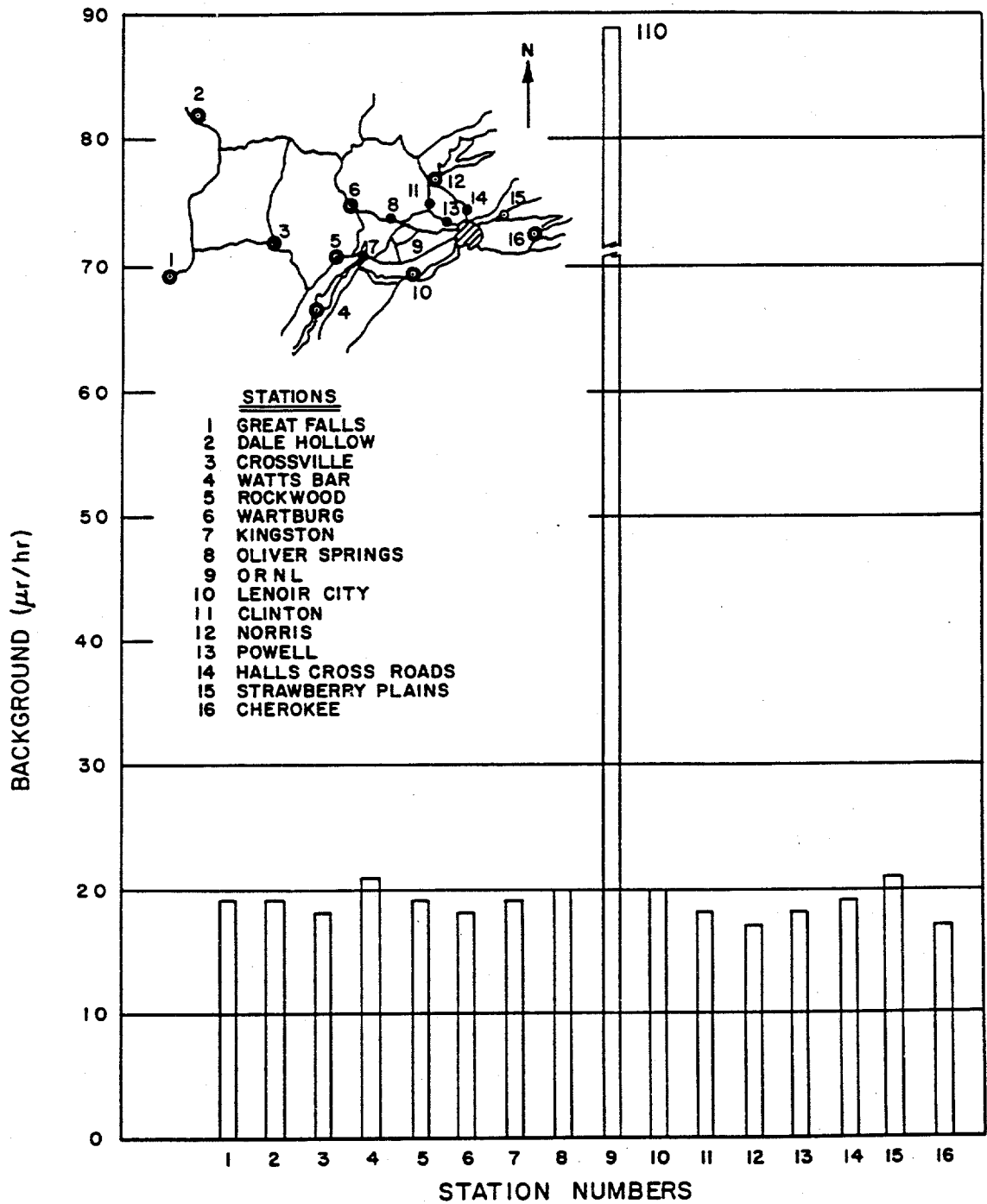
Figure B-15 Concentration of  $^{131}\text{I}$  in Cattle Thyroids - 1965.

Source: ORNL (1966)



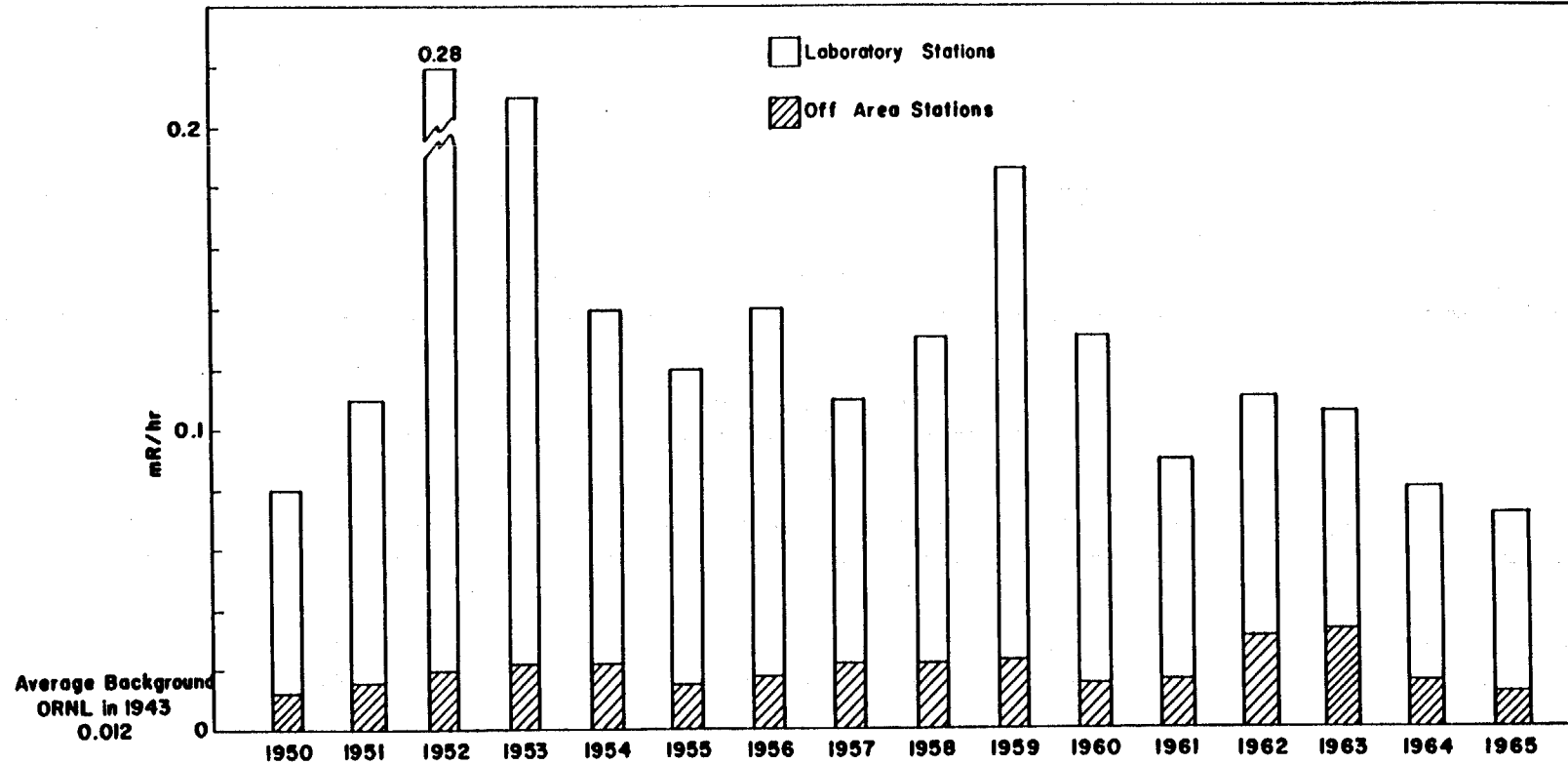
**Figure B-16** Weekly Average Concentration of  $^{131}\text{I}$  in Air at the Perimeter of the Controlled Area Compared with  $^{131}\text{I}$  Discharges from ORNL Stacks - 1965.

Source: ORNL (1966)



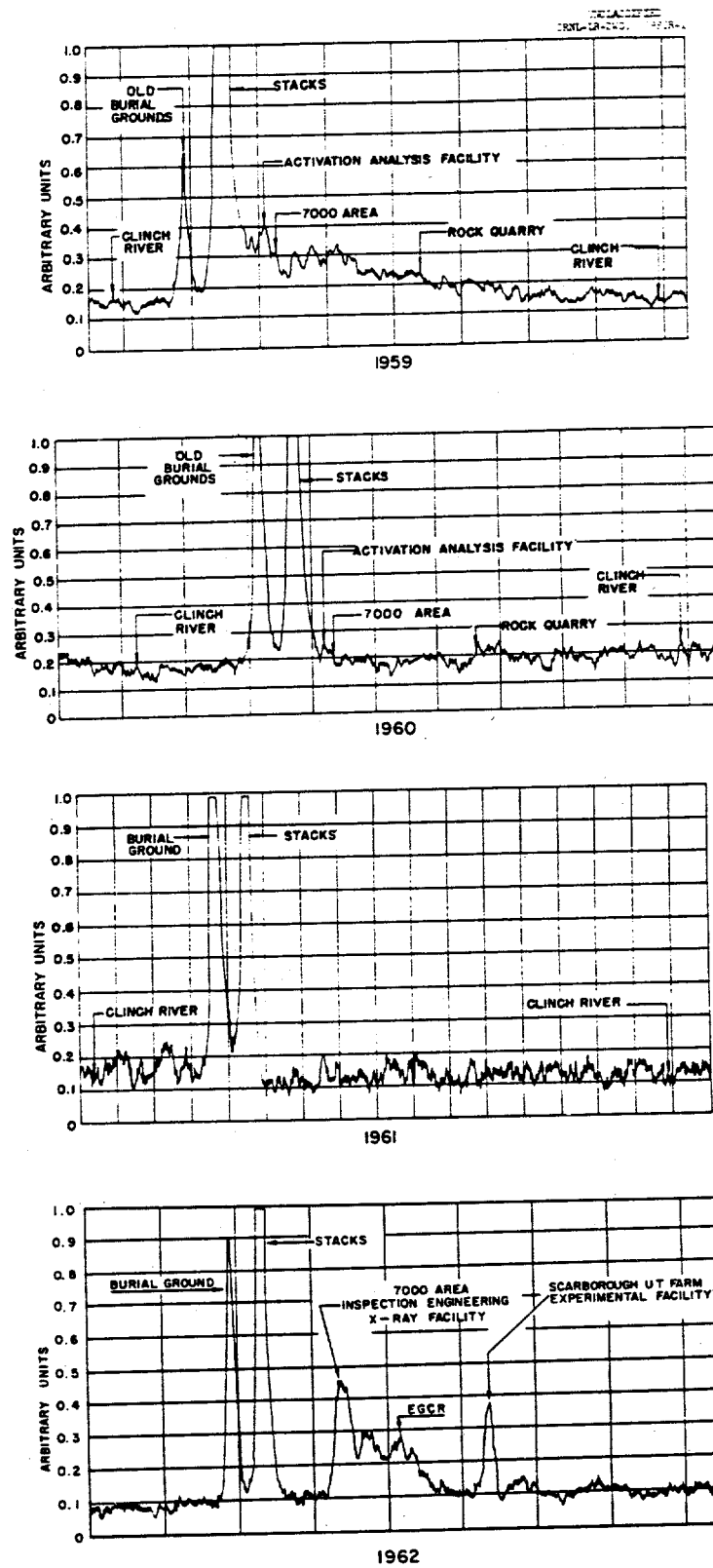
**Figure B-17** Radiation Measurements Taken During 1962, 3 ft above the Ground Surface out to Distances of 75 Miles from ORNL.

Source: ORNL (1963)



**Figure B-18** Radiation Measurements Taken 3 ft Above the Ground Surfaces at ORNL Compared with Like Measurements Taken Elsewhere within the AEC Controlled Area for the Years 1950-1965.

Source: ORNL (1966)



**Figure B-19** Radiation Background Profile of the ORNL Area as Determined by Aerial Survey Techniques, 1959-1962.

Source: ORNL (1963)



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