



ORAU TEAM Dose Reconstruction Project for NIOSH

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New Total Rewrite Revision Page Change

FOR DOCUMENTS MARKED AS A TOTAL REWRITE, REVISION, OR PAGE CHANGE, REPLACE THE PRIOR REVISION AND DISCARD / DESTROY ALL COPIES OF THE PRIOR REVISION.

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EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
09/09/2004	00	First approved issue. New technical basis document for the Mound Site – Occupational Internal Dosimetry. Incorporates internal and NIOSH review comments. Incorporates additional internal review comments. Initiated by Jeffrey S. Vollmer.
03/13/2009	00 PC-1	<p>Approved page change initiated to remove dose reconstruction provisions applicable only to Monsanto Chemical Company. Some references to Monsanto Chemical Company necessarily remain, but dose reconstruction provisions retained are applicable only to the Mound Laboratory. These changes occurred on pages 9, 10, 11, 13, 14, 16 -19, 63 in Sections 5.1, 5.2, 5.3, and Attachment D, respectively. The year for the beginning time period changed from 1947 to 1948 in Table 5-4 on page 15 and in Table 5-20 on page 39. NIOSH required language was revised on page 7 in Section 5.1. References were updated on pages 42 and 45. Incorporates formal internal and NIOSH review comments. No sections were deleted. Training required: As determined by Task Manager. Initiated by Donald N. Stewart. Approval:</p> <p><u>Signature on File</u> <u>03/04/2009</u> Donald N. Stewart, Document Owner</p> <p><u>Signature on File</u> <u>03/04/2009</u> John M. Byrne, Task 3 Manager</p> <p><u>Signature on File</u> <u>03/04/2009</u> Edward F. Maher, Task 5 Manager</p> <p><u>Signature on File</u> <u>03/11/2009</u> Kate Kimpan, Project Director</p> <p><u>Signature on File</u> <u>03/13/2009</u> James W. Neton, Associate Director for Science</p>
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TABLE OF CONTENTS

<u>SECTION</u>	<u>TITLE</u>	<u>PAGE</u>
	Acronyms and Abbreviations	6
5.1	Introduction	8
	5.1.1 Purpose	9
	5.1.2 Scope	9
	5.1.3 Current Status of Special Exposure Cohort Petitions for Mound	9
	5.1.4 Radionuclides of Concern.....	10
5.2	Process Operations Involving Primary Radionuclides	11
	5.2.1 Polonium-210	11
	5.2.2 Plutonium	11
	5.2.2.1 Plutonium-238.....	12
	5.2.2.2 Plutonium-239.....	12
	5.2.3 Tritium	13
5.3	Process Operations Involving Secondary Radionuclides.....	13
	5.3.1 Radium, Thorium, and Actinium.....	13
	5.3.2 Thorium	14
	5.3.2.1 Th-232 Source Term	14
	5.3.2.2 Ionium	15
	5.3.3 Protactinium-231	15
	5.3.4 Uranium-233.....	15
	5.3.5 Reactor Fuels Projects	15
	5.3.6 Reactor Waste Project.....	16
	5.3.7 Americium	16
	5.3.8 Rare Radioactive Isotope Program	16
	5.3.9 Other Radionuclides	17
	5.3.10 Radon.....	17
5.4	Internal Dose Monitoring	19
	5.4.1 General Description.....	19
	5.4.2 <i>In Vitro</i> Monitoring	20
	5.4.3 Gross Alpha and Sequential Separation	21
	5.4.4 <i>In Vivo</i> Dose Monitoring.....	22
	5.4.5 Personnel Air Sampling Data.....	22
5.5	Bioassay Methodologies for Primary Radionuclides	23
	5.5.1 Polonium	23
	5.5.1.1 Bioassay Methods.....	23
	5.5.1.2 Polonium Internal Dose Monitoring Records	24
	5.5.2 Plutonium	25
	5.5.3 Tritium	26
5.6	Secondary Radionuclides Internal Dose Monitoring Programs	28
	5.6.1 Radium-Actinium	28
	5.6.2 Thorium	29
	5.6.2.1 Thorium-230.....	29
	5.6.3 Protactinium-231	30
	5.6.4 Uranium.....	30

5.6.5	Americium-241 and Curium-244 Records	31
5.6.6	Rare Isotopes	31
5.7	Internal Dose Reconstruction Guidance	31
5.7.1	General Considerations	31
5.7.1.1	Use of the Gross Alpha Method	31
5.7.1.2	Internal Dose Estimation Parameters	31
5.7.2	Equilibrium Assumptions	32
5.8	Reconstructing Dose from Primary Radionuclides Based on Urinalysis Data	34
5.8.1	Polonium-210	34
5.8.2	Plutonium	35
5.8.2.1	Special Considerations for Plutonium-238.....	36
5.8.2.2	Special Considerations for Plutonium-239.....	39
5.8.3	Tritium	39
5.9	Reconstructing Dose from Secondary Radionuclides Based on Urinalysis Data	40
5.9.1	Radium-Thorium-Actinium	40
5.9.2	Thorium	40
5.9.2.1	Thorium-232.....	40
5.9.2.2	Thorium-230.....	41
5.9.3	Protactinium-231	42
5.9.4	Uranium.....	43
5.9.5	Reactor Fuels Projects	43
5.9.6	Reactor Waste Project.....	43
5.9.7	Americium and Curium	44
5.9.8	Rare Isotopes	45
5.9.9	Radon.....	45
5.9.10	Summary of Correction Factors and Excretion Equations.....	45
5.9.11	Summary of Analytical Methods and Detection Limits	46
5.9.12	Use of Co-Worker Data for Internal Dose Reconstruction.....	47
5.10	Attributions and Annotations	47
	References	48
	Glossary	54
	ATTACHMENT A, CODING INSTRUCTIONS FOR URINALYSIS RECORDS	56
	ATTACHMENT B, UNCERTAINTY ASSESSMENT	57

LIST OF TABLES

<u>TABLE</u>	<u>TITLE</u>	<u>PAGE</u>
5-1	Radon isotope survey data by building and period	18
5-2	Bioassay programs and action levels	19
5-3	Summary of bioassay methodologies.....	20
5-4	Summary of historical plutonium analytical procedures	26
5-5	Reported analytical procedures and MDAs for tritium.....	27
5-6	Recommended equilibrium assumptions for secondary radionuclides.....	32
5-7	Radionuclide chemical recoveries for collected fecal samples	33
5-8	Retroactive MDAs for fecal samples	33
5-9	Reported background count rates and MDAs for ²¹⁰ Po.....	35
5-10	Heat source plutonium feed material characteristics	35
5-11	Weapons-grade plutonium feed material characteristics	35
5-12	Plutonium MDAs	36
5-13	Summary of limiting dissolution type versus the duration of a chronic inhalation intake of ²³⁸ Pu	37
5-14	Summary of limiting dissolution type versus the time after an acute intake a single urine sample is collected.....	38
5-15	IMBA input parameters for types M, S, and the nonmonotonic model L	39
5-16	Reported analytical procedures and detection limits for tritium	39
5-17	Natural thorium DLs	41
5-18	²³⁰ Th DLs	42
5-19	Protactinium DLs.....	42
5-20	Uranium DLs.....	43
5-21	²⁴¹ Am and ²⁴⁴ Cm DLs	44
5-22	²⁴¹ Am and ²⁴⁴ Cm detection limits	44
5-23	²²² Rn WLM assumptions for dose reconstruction	45
5-24	Summary of correction factors and excretion equations	46
5-25	Analytical methods and MDAs for primary radionuclides by period	47
A-1	Coding instructions for urinalysis records	56

ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
AMAD	activity median aerodynamic diameter
ANPP	Aircraft Nuclear Propulsion Project
Bq	becquerel
BZ	breathing zone
CFR	Code of Federal Regulations
Ci	curie
cm	centimeter
cph	counts per hour
cpm	counts per minute
d	day
D&D	decontamination and decommissioning
DCF	dose conversion factor
DE	dose equivalent
DL	detection limit
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
dpm	disintegrations per minute
DU	depleted uranium
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
F	fast (absorption type)
GSD	geometric standard deviation
hr	hour
HTO	tritiated water vapor
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
in.	inch
IREP	Interactive RadioEpidemiological Program
IRF	intake retention fractions
keV	kiloelectron-volt, 1,000 electron-volts
kg	kilogram
L	liter
LANL	Los Alamos National Laboratory
LS	liquid scintillation
LSC	liquid scintillation counter
M	moderate (absorption type)
mCi	millicurie
MDA	minimum detectable activity
min	minute
mL	milliliter
mo	month

MPC	maximum permissible concentration
mrem	millirem
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NU	natural uranium
ORAU	Oak Ridge Associated Universities
pCi	picocurie
PERM	passive environmental radon monitor
POC	probability of causation
PORECON	Polonium urinalysis bioassay results database
PURECON	Plutonium urinalysis bioassay results database
QF	quality factor
RTG	radioisotope thermoelectric generator
S	slow (absorption type)
SMT	stable metal tritide
SNAP	Systems for Nuclear Auxiliary Power
SRDB Ref ID	Site Research Database Reference Identification (number)
TTA	thenoyl trifluoroacetone
U.S.C.	United States Code
wk	week
WL	working level
WLM	working level month
yr	year
μCi	microcurie
μg	microgram
μm	micrometer
§	section or sections

5.1 INTRODUCTION

Technical basis documents and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

In this document the word “facility” is used as a general term for an area, building, or group of buildings that served a specific purpose at a site. It does not necessarily connote an “atomic weapons employer facility” or a “Department of Energy [DOE] facility” as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 7384l(5) and (12)]. EEOICPA defines a DOE facility as “any building, structure, or premise, including the grounds upon which such building, structure, or premise is located ... in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program)” [42 U.S.C. § 7384l(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled “Exposure in the Performance of Duty.” That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer “shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation¹] guidelines established under subsection (c) ...” [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) restrict the “performance of duty” referred to in 42 U.S.C. § 7384n(b) to nuclear weapons work (NIOSH 2010).

The statute also includes a definition of a DOE facility that excludes “buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program” [42 U.S.C. § 7384l(12)]. While this definition excludes Naval Nuclear Propulsion Facilities from being covered under the Act, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled “Exposure in the Performance of Duty”] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all occupationally-derived radiation exposures at covered facilities in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external occupational radiation exposures are considered valid for inclusion in a dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction. NIOSH, however, does not consider the following exposures to be occupationally derived (NIOSH 2010):

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

¹ The U.S. Department of Labor (DOL) is ultimately responsible under the EEOICPA for determining the POC.

5.1.1 **Purpose**

This technical basis document (TBD) contains an analysis of the internal doses received by Mound Laboratory workers from the period of the initial external dose monitoring on February 21, 1949, through the end of operations on the Mound site. The nature of the doses received by Mound workers is analyzed, and methods for reconstructing doses are given.

5.1.2 **Scope**

The information in this TBD facilitates the reconstruction of occupational internal dose to employees of the Mound facility. This TBD does not contain guidance to reconstruct doses at Dayton Laboratory facilities (other than the Mound Laboratory), or for dates earlier than February 1949.

Section 5.1.3 discusses classes of Mound employees added to the Special Exposure Cohort (SEC). Section 5.1.4 identifies the radionuclides of concern to internal dose. Section 5.2 describes process operations that involved the primary radionuclides, and Section 5.3 describes operations with secondary radionuclides. Section 5.4 discusses internal dose monitoring, Section 5.5 describes bioassay for the primary nuclides, and Section 5.6 discusses internal dose monitoring for the secondary nuclides. Sections 5.7 to 5.9 provide guidance for reconstruction of internal dose. Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 5.10.

5.1.3 **Current Status of Special Exposure Cohort Petitions for Mound**

Classes Added to the SEC

Employees of the Department of Energy (DOE), its predecessor agencies, and DOE contractors or subcontractors who worked in any areas at the Mound Plant site from October 1, 1949, through February 28, 1959, for a number of work days aggregating at least 250 work days or in combination with work days within the parameters established for one or more other classes of employees in the SEC (SEC-0090; Leavitt 2008).

All employees of the Department of Energy (DOE), its predecessor agencies, and its contractors and subcontractors who had at least one tritium bioassay sample and worked at the Mound Plant in Miamisburg, Ohio from March 1, 1959 through March 5, 1980, for a number of work days aggregating at least 250 work days, occurring either solely under this employment, or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort (SEC-00171; Sebelius 2010).

All employees of the Department of Energy (DOE), its predecessor agencies, and their contractors and subcontractors who worked at the Mound Plant in Miamisburg, Ohio, from September 1, 1972 through December 31, 1972, or from January 1, 1975 through December 31, 1976, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees included in the Special Exposure Cohort (SEC-00207; NIOSH 2012).

Dose reconstruction guidance in this document for periods: before March 1, 1959; September 1, 1972 through December 31, 1972; January 1, 1975 through December 31, 1976; or March 1, 1959 through March 5, 1980 for the SW (Semi-Works) Building, is presented to provide a technical basis for partial dose reconstructions for claims not covered within the SEC classes above.

Although NIOSH has found that it is not possible to completely reconstruct radiation doses for the SEC classes described in this section, NIOSH intends to use any internal and external monitoring data that might become available for an individual claim, and that can be interpreted using existing NIOSH dose reconstruction processes or procedures. Therefore, dose reconstructions for employees who worked in any areas of the Mound Plant during the periods from October 1, 1949 through February 28, 1959, or September 1, 1972 through December 31, 1972, or January 1, 1975, through December 31, 1976, or who worked in R or SW Buildings (as demonstrated by records of tritium bioassay sampling) during the period from March 1, 1959, through March 5, 1980, but who do not qualify for inclusion in the SEC, may be performed using these data as appropriate.

To summarize the classes added to the SEC:

- The first class of employees was added because the potential for unmonitored intakes of radium, actinium, and thorium was assumed to be present for all Mound employees; therefore, it was not possible to reconstruct doses or assign maximum doses to all Mound employees prior to 1959;
- Because it was not possible to bound radon doses to workers in the SW building, Mound workers who worked in R and/or SW buildings were added to the SEC. These workers are identified by at least one bioassay sample for tritium in their internal dose records; and
- Since it was found that during some periods that tritium records could not be found, all employees were included in the SEC during the periods September-December 1972, and the years 1975 and 1976.

For all of these classes of workers, internal doses are to be reconstructed using case-specific data, if present, when they may be estimated using current dose estimation processes and procedures.

5.1.4 Radionuclides of Concern

Radionuclides are categorized as either primary or secondary based on the potential extent of presumptive exposures to Mound employees. Primary radionuclides of concern are discussed in Mound internal dosimetry documentation including *History of Mound Bioassay Programs* (Meyer 1992) and *Mound Site Radionuclides by Location* (EG&G 1995). Primary radionuclides assessed for occupational exposure are

- Polonium (^{210}Po),
- Plutonium (^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Pu , ^{242}Pu), and
- Tritium (^3H).

Secondary radionuclides of concern include the following:

- Radium (^{226}Ra), thorium (^{228}Th), and actinium (^{227}Ac),
- Thorium (^{228}Th , ^{230}Th , and ^{232}Th),
- Protactinium (^{231}Pa),
- Uranium (^{238}U , ^{235}U , ^{234}U , and ^{233}U),
- Americium (^{241}Am) and curium (^{244}Cm),
- Rare isotopes (^{221}Fr , ^{217}As , ^{229}Th , ^{233}Th , ^{233}Pa , and ^{209}Po), and
- Radium (^{228}Ra , ^{226}Ra , ^{224}Ra , and ^{223}Ra).

5.2 PROCESS OPERATIONS INVOLVING PRIMARY RADIONUCLIDES

5.2.1 Polonium-210

Polonium-210 was used in early nuclear weapons as part of the triggering mechanism for the fission reaction. The devices, called initiators, were not themselves potent sources of neutrons until they were activated; but work with neutron sources was carried on concurrently with the weapons program. Mound's predecessor sites, the Dayton Laboratory facilities (Units III and IV), separated ^{210}Po from naturally occurring materials. By the time the Mound facility began polonium processing in 1949, the feed material was irradiated bismuth.

At the Dayton Laboratory, the separation and concentration steps of the polonium processing operation involved significant releases of airborne contaminants including the most dosimetrically significant, ^{210}Po . The polonium facility at the Mound site, T (Technical) Building, was purpose-built to entrain the material in process piping and vessels and inside specially designed gloveboxes so that the process could be done remotely. Operators controlled the activity from a shielded operations gallery (Anthony et al. 1956, BWXT 2002). This had the effect of restricting or eliminating significant releases of radioactive materials under routine processing conditions.

Polonium-210 was present in the HH (Hydrolysis House) Building from 1949 to 1959, which generated sludge materials with 0.02 Ci/L total alpha activity. Extensive use of ^{210}Po in the R Building involved research and electrodeposition studies from 1948 to 1951 and the manufacturing of Po-Be neutron source from 1956 to 1961.

The SW Building was used for counting ^{210}Po sources, neutron source measurement, source strength verification, and $^{209,210}\text{Po}$ retrieval. The primary processing and separation of ^{210}Po took place in T Building from 1949 to 1971. Facilities and activities included neutron source calibration, decontamination laboratories, a storage pool for irradiated bismuth slugs, neutron source processing, and analytical laboratories. Aqueous wastes that contain ^{208}Po , ^{209}Po , and ^{210}Po have been present in the WD and WDA waste disposal buildings from 1949 to the present. Chemical forms included polonium nitrate and polonium chloride. There was no cited ^{210}Po activity in the PP (Plutonium Processing) or SM (Special Metallurgical) Buildings. A single ^{210}Po incident was reported in the SW (Semi-Works) Building in 1961, and 11 incidents are associated with the T Building from 1954 to 1968, where primary polonium separation and processing took place.

The second phase of the polonium project consisted of manufacturing polonium heat sources for radioisotope thermoelectric generators (RTGs). The Systems for Nuclear Auxiliary Power (SNAP)-3A polonium-powered RTG was built in 1958. However, because the short half-life of ^{210}Po limited the useful life of the RTGs, this program was halted after 1969 and cancelled in 1971. Cleanup of polonium areas continued through approximately 1974.

5.2.2 Plutonium

Plutonium has been associated with Mound site research activities conducted for the U.S. Atomic Energy Commission (AEC) beginning as early as 1949. In addition to ongoing research activities, neutron sources that were fabricated using ^{239}Pu and ^{238}Pu for research projects were transferred from the Lawrence Livermore National Laboratory to Mound in 1959. As polonium production tailed off, ^{238}Pu activities transitioned into a large-scale effort to fabricate initiators, neutron sources, and fuel for RTGs and other devices using heat from the alpha decay of the fuel. Some AEC research programs carried out at Mound used ^{239}Pu mixtures that are referred to in this document as "weapons-grade plutonium." The plutonium for RTGs and other heat sources was primarily composed of ^{238}Pu , which is typically called "heat-source" plutonium in Mound historical documents and in this TBD. The

radionuclides and proportions of these mixtures are listed in Tables 5-10 and 5-11 later in this document.

5.2.2.1 Plutonium-238

Feed material from the Savannah River Plant as PuO₂ powder was enriched in ²³⁸Pu and processed at Mound to manufacture heat sources. Early ²³⁸Pu activities took place primarily in the SM Building. The PP Building was built to supplement and replace the ²³⁸Pu processing areas in SM.

SM Building activities included production of plutonium microspheres, fabrication of sealed sources, and plutonium recovery from high-risk aqueous waste. The microsphere manufacturing process in SM Building produced very fine PuO₂ particles in the submicrometer to 20- μ m range (EG&G 1995).

The microspheres would have been quite stable, and their initial particle size distribution would only be affected by depositional processes that remove particles above 10 μ m from worker breathing zones (BZs). Deposition would therefore tend to increase the inhalation of submicrometer particles. Reported solubility varied widely from type S in Room SM-60 to type M in Rooms SM-27 and SM-28. Use of a plasma torch process (primarily in SM-39) resulted in PuO₂ fumes.

Encapsulation, X-ray inspection, and calibration of Pu-Be neutron sources were conducted in T Building from 1955 to 1985. Counting and verification of sources was conducted in various laboratories of T Building. Two ²³⁸Pu exposure incidents were reported in 1986.

All SM processes were transferred to the PP Building in December 1967. The primary mission in this building was to process PuO₂ powder from the Savannah River Plant. A number of processes were developed to manufacture plutonium microspheres in the PP Building and involved the following:

- PuMo cermet process to coat plutonium particles from 43 to 250 μ m with molybdenum from 1969 to 1973,
- Pressed PuO₂ process to fabricate plutonium spheres for encapsulation as heat sources from 1970 to 1979,
- Milliwatt Generator Program process to seal 105- to 500- μ m plutonium shards with yttrium,
- Solid solution process from 1969 to 1974,
- Solution gel process from 1969 to 1971, and
- Microsphere process to produce 50- to 250- μ m ²³⁹PuO₂ ceramic spheres from 1967 to 1970.

The Mound site supplied RTGs for a number of space exploration missions, including Pioneer, Voyager, and Apollo. In 1977, production of the PuO₂ fuel itself was moved to the Savannah River Site, and activities at Mound were oriented around assembly of the RTG capsules (DOE 1993). Mound produced more than 500 RTGs from 1961 to 1992. In another ²³⁸Pu program, ²³⁴U was separated from aged ²³⁸Pu from 1970 to 1980. The heat source plutonium isotopic mixture listed by EG&G (1995) is given in Table 5-10 later in this document.

5.2.2.2 Plutonium-239

Plutonium-239 was separated from neutron-irradiated uranium metal from 1949 to 1952. Reactor fuel research was conducted with plutonium from 1956 to 1959. The Neutron Source Group started production of heat sources in the R Building using ²³⁹Pu in 1956. To date, NIOSH has not determined

the exact mixture of plutonium that made up the weapons-grade material, though one source describes it as “more than 95% pure ^{239}Pu with trace amounts of ^{240}Pu and ^{241}Pu ” (EG&G 1995). For the purposes of dose reconstruction, the mixture resulting in the greatest ingrowth of ^{241}Am was selected as a favorable-to-the-claimant assumption, in lieu of specific data. This mixture is listed in Table 5-11 based on Hanford plutonium (Battelle 2003).

5.2.3 Tritium

EG&G (1995) reported that tritium (^3H) recovery, purification, and use for solid tritiated metal compounds took place in 1948 in R building and that tritium, limited to testing and research activities, was used in T building as early as 1947 (sic², EG&G 1995). However, this is the only reference that provides information about the presence of tritium on the Mound site before the research program described by Meyer (1992) started in 1957. Because no other sources show that tritium was processed at Mound before this time, and there is no indication of a bioassay program, for this revision of the Mound site profile, no provisions are made to estimate dose from tritium before 1957.

In 1969, the retrieval of tritium from retired weapons systems began, which involved the analysis, recycling, separation, recovery, purification, and storage of tritium. In addition, weapons component testing and some research activities were conducted with metal tritides in SW Building and with tritiated organics in T Building. Tritium was consequently used in all buildings except the PP Building and the SM Building. Extensive use of tritium occurred in R, SW, and T Buildings. These activities resulted in tritium-contaminated soils under SW Building (EG&G 1995). Eight tritium incidents reported by EG&G (1995) occurred in SW Building from 1964 to 1986. Tritium exposures ended in 1995 as reported by EG&G (1995).

5.3 PROCESS OPERATIONS INVOLVING SECONDARY RADIONUCLIDES

5.3.1 Radium, Thorium, and Actinium

Because of the short half-life of ^{210}Po , the initiators required frequent remanufacture. For this reason, the Substitutes program was initiated in the late 1940s to identify longer-lived alpha emitters that could be used instead. Through research at Mound and the Los Alamos National Laboratory (LANL), ^{227}Ac was identified as the most promising replacement for ^{210}Po (even though ^{227}Ac is primarily a beta emitter, its short-lived progeny are potent alpha sources), and research was conducted on refinement and purification. The feed material was neutron-irradiated ^{226}Ra activated as shown below.



Process chemical research projects were carried out at Mound in the late 1940s and early 1950s using nonradioactive barium and strontium as chemical surrogates of the radium and actinium. For some activities, they were labeled with ^{140}Ba and ^{90}Sr radiotracers.

Once production-scale processing began, the actinium processing operation proved to result in unprecedented personnel exposures from airborne alpha emitters and high external dose rates in the operation, which was conducted in a containment facility called the “Old Cave” in SW-1 and SW-2. In July 1953, the program was deemphasized; large-scale actinium processing was discontinued in early 1954. Decontamination and decommissioning (D&D) of the actinium facility took place through early 1959 and ended in February. This activity resulted in large internal doses from radioactive gases and airborne particulates associated with the feed material and the process itself. Bioassay was performed for the involved individuals, who are identified in Mound records (Meyer 1994), but

² T Building was not occupied before February 1948; polonium operations began in September of that year (BWXT 2002a).

uncertainties associated with the method, and the likelihood of exposure of individuals who were not monitored under the internal dose monitoring program, led to the addition of a class of Mound employees to the SEC.

5.3.2 Thorium

Major processes at Mound that involved thorium were the Monex refinery and the Ionium project. Monex was the name of a Mound Laboratory proprietary process to refine natural thorium, ^{232}Th , for possible use as a reactor fuel. Ionium activities at Mound were part of a complex-wide project to stockpile ^{230}Th (ionium), also for potential uses in reactors.

5.3.2.1 Th-232 Source Term

The principal ^{232}Th feed material was monazite sludge. This was residue left after monazite sand milling. The milling process is believed to have involved treatment with a caustic to solubilize the phosphates, decanting the soluble phosphates, and leaching with acids to extract the rare earth elements. The caustic treatment was likely to have been a high-temperature process that would have resulted in such refractory and highly insoluble thorium compounds as ThO_2 . The material that was received by Mound was sometimes basic and sometimes acidic.

Mound's activities with ^{232}Th began with a preliminary proposal for a thorium refinery that was prepared in 1954 (Monsanto 1954). Mound was authorized to construct the refinery on March 11, 1955 (Wende 1955). The refinery was anticipated to process approximately 128 kg of thorium per hour (Huddleston 1955). In anticipation of this project, Mound received a large quantity of thorium sludges (approximately 6,000 55-gallon drums) from United Lead Company of Middlesex, New Jersey, between December 10 and 27, 1954 (Meyer 1979). This material came to Mound via rail in 64 boxcars (McCarthy 1955a). However, doubts about the entire breeder reactor program were being expressed in private by AEC as early as April 1955 (McCarthy 1955b), and the Mound refinery project was cancelled on May 6, 1955 (Meyer 1955).

This left Mound with the task of disposing of the thorium residues. On arrival at Mound, the drums were briefly stored in a warehouse and a garage, but increases in short-lived particulate progeny were observed. This prompted the decision to move the drums to an unfrequented warehouse. With the exception of 650 drums that had to be protected from the weather, which were briefly stored in a Quonset hut (Bradley 1954), most of the drums were eventually stored in an open field in the southern part of the plant site (Meyer 1979). Due to the exposed storage conditions and the corrosive nature of the sludges, drum deterioration and failure were a recurring problem over the next 12 years and required redrumming of the entire lot three times. The redrumming efforts were conducted in the outside storage areas during the summer months, and this resulted in contamination of the storage fields.

A two-bay storage basin known as Building 21 was completed in 1966, and the drums were emptied into this basin. The oxalate sludges (2.23×10^5 kg) occupied a smaller bay that occupied about 25% of the storage basin, and hydroxide sludges (1.2×10^6 kg) occupied the larger bay (about 75% of the basin).

The thorium residues were sold to General Atomics in 1974 and removed from the site by July 15, 1975 (Meyer 1979). Removal of the sludge was done under contract, and the contractor provided its own health physics oversight and dose monitoring. The final phase of the Building 21 decontamination was complete in September 1975. The completion of this effort also marked the end of the most significant exposure potential to ^{232}Th at the Mound site. Only incidental exposure potential to residual contamination existed after this date.

5.3.2.2 Ionium

A limited ionium research program was begun in early 1956. The program was to research separation of ^{230}Th from source material. The program was completed in 1958, with the last bioassay sample submitted in September of that year.

Primary feed material for extraction of ^{230}Th was the so-called Airport Cake (Cotter Cake). The feedstock was raffinate from extraction of uranium from a hydrofluoric or nitric acid solution using diethyl ether followed by treatment with caustic or lime. This feed source was obtained from Mallinckrodt and the Cotter Corporation.

5.3.3 Protactinium-231

An AEC program was undertaken in 1955 in the R Building to recover and purify a small amount of protactinium from natural sources. This limited research investigated ^{231}Pa for possible use in initiators or as source material for RTGs. The separation and processing of ^{231}Pa from Sperry Cake raffinates was conducted in the HH Building, Rooms HH-5 and HH-6, from 1955 to 1956. Separation of ^{231}Pa from Sperry Cake was also reported in R Building (R-148, R-149, R-167) from 1954 to 1959. The program began to wind down in 1958, and was concluded in 1959. Only limited laboratory use was reported from 1956 to 1987.

Minor quantities of ^{231}Pa were present in samples that were subjected to spectrographic analysis in R-140 and R-142 from 1956 to 1969. Minor concentrations of ^{231}Pa were found in WD/WDA Building, which received aqueous wastes from R and SW Buildings. No ^{231}Pa use was reported for SM, SW, T, or PP Buildings.

Protactinium-231 extraction and purification was a complex process that is poorly described. There is no information to estimate the radioactive compounds likely to have been present. Use of minimal quantities of ^{231}Pa in laboratory isotopic studies and during spectrographic analyses of samples after 1959 is assumed to be insignificant. Dose reconstruction should be conducted using available bioassay data from 1954 through 1959.

5.3.4 Uranium-233

A limited research project involving ten researchers took place in 1958 and 1959. Urine sampling started July 25, 1958, and ended July 1, 1959.

5.3.5 Reactor Fuels Projects

This program performed basic research on chemical and physical properties of proposed reactor fuels for several different types of reactors and reactor projects:

- Aircraft Nuclear Propulsion Project (ANPP). In 1953, Mound started a project to support the ANPP. The liquid cycle reactor was a fused salt design.
- Homogenous-Type Reactor. Begun in mid-1950s, this effort worked to identify satisfactory fuel systems. Mound measured the physical properties of uranium and thorium solutions and slurries at elevated temperatures and pressures. The program started in 1952 and used many measurement techniques from the ANPP program.
- Civilian Power Reactor Program. This program, started in 1957, was concerned with development of reactors for power and heat for civilian use; it examined the homogenous-type reactor using ^{233}U , ^{235}U , and ^{239}Pu as fuel.

The activities listed above investigated physical attributes and, other than sample preparation, involved only the required physical measurements:

- Plutonium fuel for fast breeder reactors. Mound was responsible for acquiring data on proposed fuel systems by determining density, viscosity, thermal capacity, thermal conductivity, and phase equilibriums of plutonium and plutonium alloys. These were Pu-Fe, Pu-Ni, Pu-Cu, Pu-Cd, Pu-Yt, and Pu-Va.
- In 1961 Mound participated in a project to experiment with the fabrication of plutonium-bearing glass fibers as a reactor fuel system.

These activities involved chemical processing of plutonium.

5.3.6 Reactor Waste Project

In January 1949, Mound began a study of processes for decontamination of reactor wastes for recovery of ^{239}Pu . From 1949 to 1952, three different waste streams were researched: bismuth phosphate and tributyl phosphate from the reduction-oxidation process from Hanford, and the wastes from the plutonium-uranium extraction process pilot plant at Oak Ridge. The bench-scale research was conducted in R-167 using milliliter aliquots of the wastes. Based on the results, a pilot plant was constructed in early 1952 in M Building, where cold (nonradioactive) runs were made. When these were complete, the process equipment was set up in HH building and became operational by November 1, 1952. A volume of approximately 150 to 200 gallons was processed. Some bioassay samples for 1951 and 1952 were analyzed for plutonium and likely were the ones taken for this program. The program was terminated by February 1953. The pilot plant was dismantled, decontaminated, and stored in March 1953 (Meyer 1992, p 378).

5.3.7 Americium

Americium-241 is a constituent of weapons-grade plutonium (^{239}Pu); however, the percent of gross alpha activity in heat source plutonium attributable to ^{241}Am is negligible. Where intakes of ^{241}Am occurred as a result of intake of weapons-grade plutonium, the ^{241}Am constituent should be estimated using the composition of weapons-grade plutonium in Table 5-11.

When not a constituent of weapons-grade or heat source plutonium, potential doses from ^{241}Am should be reconstructed separately; this is expected to be necessary for individuals working with Americium as part of research projects. This is due to the possibility that it posed a specific intake risk from alpha source production, neutron source production, analysis, and vaporization of plutonium compounds, ^{231}Pa separation, and the reactor fuel research program.

Americium-241 alpha sources were electroplated in Room R-120 between 1956 and 1962, and it was the major isotope of concern in that room. Exposure to ^{241}Am in R-120 could also have occurred coincident with intakes of weapons-grade plutonium.

5.3.8 Rare Radioactive Isotope Program

Mound conducted separation and purification of special heavy-element isotopes. The work was on a gram- or milligram-scale, and the isotopes were shipped to the Oak Ridge isotope pool. The radionuclides included:

- ^{231}Pa .
- ^{234}U obtained from "milking" ^{238}Pu .

- ^{229}Th obtained from “milking” ^{233}U . This program was to create ^{229}Th to use as a source material for ^{221}Fr , ^{217}As , and other members of the $4n + 1$ series. Thorium-229 is itself a natural product of ^{233}U and was produced in the thorium fuel cycle by neutron irradiation of ^{233}Th . The ^{229}Th project at Mound started in 1966. Work was performed in the hot cell in the SW Building. Mound used 1.5 kg of ^{233}U to produce 30 mg of ^{229}Th . The program concluded in early 1970s (Meyer 1992, p. 382).
- ^{233}U from an aged source of irradiated ^{232}Th to obtain ^{233}Pa , which decays to ^{233}U .
- ^{228}Th , ^{227}Ac , ^{209}Po , and ^{226}Ra . Began in the mid-1950s and continued until 1985. Separations carried out in SW, SM, and R Buildings.

Meyer (1992) states that routine bioassay programs covered all of these activities, but the bioassay method is likely to be based on the parent or more abundant radionuclide, such as ^{238}Pu rather than the ^{234}U harvested from it.

5.3.9 Other Radionuclides

Xenon and krypton were present in H Building as a part of the Isotope Separation Program and xenon isotope separation activities. For the purpose of dose reconstruction, external dose is the exposure of concern and internal dose is considered negligible.

Curium-244 and ^{237}Np were present at the site in minor concentrations in special nuclear heat source material from the Savannah River Site. They were also present in aqueous wastes derived from the research-scale plutonium recovery operations. Curium-244 and ^{237}Np were also used in metallic or oxide form in reactor fuel research and to support the heat source program. Curium-244 has a relatively high dose conversion factor (DCF) (less than a factor of 2 below ^{239}Pu and ^{232}Th) and therefore the potential to induce relatively high doses via the inhalation pathway should be taken into account by the dose reconstructor. However, both ^{237}Np and ^{244}Cm were used in minor quantities and were of secondary radiological concern.

Mound engaged in biomedical research with ^{45}Ca , ^{59}Fe , ^{60}Co , ^{65}Zn and other radionuclides. These were present in relatively minor quantities in comparison to the primary and secondary radionuclides and were used in very few buildings for limited periods. Most of these radionuclides were present as sealed sources. Worker exposure was less likely, and bioassay data was not collected for these radionuclides. Exposure to these radionuclides can therefore be considered insignificant.

5.3.10 Radon

Radon is a naturally occurring radioactive gas that emanates from materials in the earth's crust; however, when natural levels are enhanced by processing or accumulation of radioactive materials that were used, handled, or stored at DOE facilities, the dose must be considered along with other potential intakes. Radon is itself a noble gas, but its particulate progeny that are deposited in respiratory tract organs result in significant doses.

Few data are available to use for Mound dose reconstruction before the landmark study that was conducted in 1989 (UNC Geotech 1990). However, the SW Building experienced high levels of radon from the decay of radioactive materials generated during the radium-actinium processing effort in the Old Cave, despite serial remediation activities. A study occurred in the late 1970s to identify the proportions of radon, actinon, and thoron, (alternative names for ^{222}Rn , ^{219}Rn , and ^{220}Rn , each deriving from the predecessor radionuclide), and an attempt was made to measure the concentration. It has been determined that these data do not allow the calculation of potential doses with sufficient accuracy to estimate a maximum dose. For this reason, NIOSH is unable to determine an upper

bound for the radon exposures in SW-19 until the point at which a ventilation system was installed in early 1980 (before March 5, 1980) to vent SW Building air. The inability to bound the concentration in SW-19 extends to the contiguous air spaces in the SW and R Buildings. The lack of sufficient information to bound the dose from radon and its progeny formed the bases for the SEC class definitions in SEC-00171 (Sebelius 2010) and SEC-00207 (NIOSH 2012).

Thorium sludge from ²³²Th extraction and purification operations was stored outside in drums from 1954 to 1966. In 1966, thorium sludge was stored in 4,914 drums in Building 21. All thorium sludge drums were removed from Mound in 1975. In 1976, 1,258 drums of Cotter concentrate were stored to support purification of ²³¹Pa and ²³⁰Th. Table 5-1 shows the results of various radon surveys at Mound.

Table 5-1. Radon isotope survey data by building and period.

Building	Survey dates	Gas concentration (pCi/L)			WL	WLM/yr ^a
		Radon	Thoron	Actinon		
SW Tunnel ^b	10/12/1979	88,000	28,000	640,000		
SW19 after vent	03/1980–04/1980	7.7–13.4			0.03	0.4
	12/1989	2.6	Not detected			
	12/1999	1.0–5.^c				
	04/1990	0.7–7.2				
	1998–2000	5.8–12				
Building 21	05/1983	17.5–52.8			0.3–0.7	0.4–1.3 0.85 median 10.0 median
	12/1989	34–118				
	04/1990–06/1990	117–161				
		125				
Building 48	12/1989	3.2				
Old SD	12/1989	1.5–2.4				
Fire station	12/1989	1.2–1.4				
Paint shop	12/1989	0.9–1.2				
A	10/1988–02/1989	0.1–1.1				
All other buildings	12/1989	0.4–0.9				
Outside facilities	05/1983	0.24–0.59				
Reported background		0.1 to 2.1 Average = 0.5				

- WLM/yr values were determined as WL (2,000 hr/yr) (1/170 hr/WLM).
- The tunnel under SW was not an occupied workspace; concentration provided for information only.
- Bolded concentrations were derived from charcoal canister measurements.

Areas in the SW Building experienced high concentrations of radon, thoron, and actinon that emanated from residual radium, thorium, and actinium from the remediated SW-1 and SW-2 Rooms; some materials had been disposed in place and a new floor built above them. The area was renamed SW-17, -18, and -19. In addition to contaminated components and fill material, indications were that concentrations of radium, thorium, and actinium were high in the soil underneath the concrete flooring. Noble gases were transported through an underground tunnel into SW-17, -18, and -19. A ventilation system was installed in early 1980 to vent SW-17 tunnel air to an SW-18 exhaust.

The installed ventilation system reduced ²²²Rn concentrations in SW-19 by a factor of 10 or more, to 8.2 pCi/L in March 1980. This result was confirmed with a passive environmental radon monitor (PERM) measurement of 15.4 pCi/L and RDT310A continuous measurements from 7.7 pCi/L to 13.4 pCi/L. A working level (WL) measurement by the employee's desk was 0.03 WL or 0.03 (12 mo/yr) = 0.4 working level month (WLM) in comparison with an occupational limit of 4 WLM/yr.

Radon surveys in Building 21 were conducted with PERMs in 1983, and the results ranged from 17.5 to 52.8 pCi/L with WLs from 0.40 to 1.3. WLMs exceeded the occupational limit of 4 WLM with calculated values from 4.8 to 15.6 WLM (Author unknown, 1983a, 1983b, 1983c). Radon that was

measured during the radon study with charcoal canisters ranged from 34 to 118 pCi/L in 1989 (UNC Geotech 1990). Building 21 was an unoccupied silo for storage of thorium feed material.

A DOE radon study was conducted from December 12 to 15, 1989, to measure radon in various Mound buildings (UNC Geotech 1990). The majority of buildings had radon concentrations below 1.0 pCi/L ²²²Rn, except SW and Old SD buildings. In June 2000 a radon study summary report was issued based on 1990 and 1999 measurements (Doody 2000). Mound site radon background was reported to be 0.5 pCi/L with a range of 0.1 to 2.1 pCi/L. The SW Building, specifically room SW-19, was identified as the only area of potential occupational exposure to ²²²Rn and ²²⁰Rn.

Thoron and actinon measurement results were very limited despite the high concentrations that were observed in the underground tunnel. The only actinon measurements were made in Building 21 at 0.3 to 0.7 pCi/L, but the holding time before analysis was excessive in comparison with the half-life of ²¹⁹Rn (3.96 seconds). The only thoron measurement in SW-19 was below detection.

5.4 INTERNAL DOSE MONITORING

5.4.1 General Description

Mound site bioassay results consist of *in vitro* urine data and limited fecal sample results. The primary source of sampling data for Mound dose reconstruction is *in vitro* results in employee records. Nose swipe data were considered to be secondary information. Whole-body counting is not an efficient monitoring method for Mound's primary radionuclides (tritium, ²¹⁰Po, and ²³⁸Pu) or for many secondary radionuclides such as ²³⁹Pu, ²³¹Pa, Ra, or Ac. Table 5-2 summarizes Mound urinalysis bioassay programs with reported radionuclide action levels, monitored worker populations, and monitoring frequency.

Table 5-2. Bioassay programs and action levels.

Bioassay program	Period	Action levels ^a	Monitored workers	Frequency
<i>In vitro</i> monitoring				
Po-210 processing	1945–1974	10 cpm/50 mL	Operations personnel	Weekly spots Weekly 24 hr
Ac-227, Ra-226, Th-228	1951–1960	8.3 cpm/24 hr	Research personnel	Weekly 24 hr
Th-232 sludge, redrumming	1955–1975	2.8 cpm/24 hr	Th refinery personnel	Monthly 24 hr
Th-230 (ionium) research	1956–1959	3.5 cpm/24 hr	Research personnel	Bi-weekly 24 hr
Pa-231 extraction	1956–1960	2.2 cpm/24 hr	Research personnel	Monthly 24 hr
U-233 research	1958–1960	20 cpm/1,500 mL	10 research personnel	Weekly 24 hr
Tritium (hot gas) production	1957–present	10 µCi/L	Recovery personnel	Weekly spots Weekly 24 hr
Pu-239 neutron source	1957–present	3.5 cpm/24 hr	Processing personnel	Weekly spots Weekly 24 hr
Pu-238 heat source	1960–present	3.5 cpm/24 hr	Processing personnel	Weekly spots Weekly 24 hr
Am-241	7/1989–present	0.023 pCi/Sample ^b	Potentially exposed personnel	Daily
Cm-244	7/1989–present	0.009pCi/Sample ^b	Potentially exposed personnel	Daily
<i>In vivo</i> monitoring				
Whole-body counting	1960–1970	Not available	Operations personnel	Annual
Chest counting	1970–present	Variable ^c	Operations personnel	Annual

a. Observed cpm for 50 mL or 24-hour samples that result in a worker dose of 300 mrem/wk to the target organ.

b. Barton (1995); for results prior to this date, dose reconstructors should base MDA assumptions on actual data or refer to the 'Table of Other Radionuclides' (Author unknown 1955-1988).

c. Late MDAs at the Mound site ranged from 0.1 nCi for Am-241 to 130 nCi for Pu-239.

Routine bioassay monitoring included collection of spot and 24-hour urine samples depending on the radionuclide to which specific employees were assigned. Monitoring frequencies were described by Meyer (1992) and inferred from personnel sampling records. For programs that used spot sampling, such as ²¹⁰Po and ²³⁸Pu, individuals suspected of exposure during incidents, or who had urinalysis results in excess of the reporting limits, were required to submit additional urine 24-hour samples.

Administrative personnel were not monitored and monitoring was discontinued for operational personnel if their work on specific projects was no longer needed.

Mound used the action levels in Table 5-1 to evaluate and control worker doses to a 300-mrem/wk target organ dose limit. Results in excess of action levels triggered resampling and placement of the worker on the "hot roster" to control further exposures if necessary. Reporting limits for americium and curium were 0.0, and any detectable activity was reported and triggered resampling. The dosimetry controls in place by 1949 when Mound operations were moved from the Dayton Laboratory facilities were consistent with the limits stated in Handbook 52 published in 1953 (NBS 1953).

5.4.2 In Vitro Monitoring

In vitro monitoring programs were established for each activity at the Mound site as described in Meyer (1992) and summarized in Table 5-3. Although Mound maintained the ability to analyze samples for specific radionuclides, many analyses were conducted using the gross alpha procedure. Monitoring protocols for each program are listed below in the appropriate paragraphs; because the gross alpha procedure applied to many Mound bioassay programs, it is described, in general, in the section below.

Table 5-3. Summary of bioassay methodologies.

Nuclide	Method/description	Period	MDA	
H-3 (hot gas)	Vibrating reed electrometer	1957–1961	1.0 µCi/L	
	Liquid scintillation (LS)	1962–1995	0.01 µCi/L	
	Spontaneous electrodeposition from raw urine, low-background proportional counting	1944–1963	Spot samples 6.0 dpm 168 dpm/d	
Po-210	Spontaneous electrodeposition from acidified urine, low-background proportional counting.	1964–1973	Spot samples 0.7 dpm 20 dpm/d	24-hr samples 1.3 dpm/24 hr
	Radium extraction and activity inferred from ²²⁷ Th or ²²³ Ra dpm assuming secular equilibrium	08/17/1955–1956	0.3 dpm	24-hr samples 0.15 dpm/24 hr
Pa-231	Alkaline earth phosphate coprecipitation, Pu separation with cerium carrier gross alpha proportional counting	1957–1958	0.02 dpm	
		1959–12/07/1959	0.08 dpm	
		Before 1957	(0.85) pCi/sample	
Pu-238, Pu-239	Above with sample acidification	1957	(0.94) pCi/sample	
		1958	(0.6) pCi/sample	
		1959	(0.5) pCi/sample	
		1960	(0.5) pCi/sample	
		1961–1966	0.05 pCi/sample	
	Above with anion exchange, electrodeposition, gross alpha proportional counting	1967–05/1981	0.11 pCi/sample	
Above with alpha spectroscopy for Pu-238 and Pu-239/240, Pu-242 tracer, blanks counted, DLs quantified	06–12/1981	0.066, 0.044 pCi/sample		

Nuclide	Method/description	Period	MDA
	Radium separation, intakes and excreted Ra-226 and Th-230 assumed to be in equilibrium.	1982	0.063, 0.042 pCi/sample
		1983	0.064, 0.043 pCi/sample
		1984	0.066, 0.045 pCi/sample
		1985	0.047, 0.031 pCi/sample
		1986	0.042, 0.028 pCi/sample
		1987	0.022, 0.011 pCi/sample
		1988	0.017, 0.01 pCi/sample
		01–07/1989	0.019, 0.008 pCi/sample
		08/1989–12/1991	0.018, 0.008 pCi/sample
		1992–present	0.018, 0.009 pCi/sample
		Before 01/1994	0.3 dpm
Th-230 (Ionium)	Th-230 extraction, alpha spectroscopy.	01/1994–1995	0.17 dpm
	Thorium separation, proportional counting.	Before 12/1987	0.3 dpm
Th-natural	Thorium separation, alpha spectroscopy.	12/1987–1994	0.02 (Th-232) dpm
		01/1994–1995	0.08 (Th-228) dpm
	LiF fusion, fluoroscopy.	1950s–1985	0.4 µg/L
Uranium	Uranium separation, proportional counting.	Before 1985	0.3 dpm
	Uranium separation, alpha spectroscopy.	05/1985–1995	0.07 (U-234) dpm 0.03 (U-235) dpm 0.09 (U-238) dpm
	Americium or curium separation, proportional counting.	1950–1989	0.2 pCi
Am-241, Cm-244	Americium or curium separation, alpha spectroscopy.	Before 07/1989	0.3 pCi
		07/1989–1995	0.05 Ci

5.4.3 Gross Alpha and Sequential Separation

A procedure was developed at the Mound Laboratory for extraction of alpha emitters from human urine to assess the concentrations of radium, actinium, and thorium in MLM-1003 (Kirby and Brodbeck 1954). This procedure was later used for or adapted to a number of radionuclides over the operating period of the internal dose monitoring program at Mound.

The gross alpha procedure method involved initial concentration of a 24-hour urine sample by coprecipitation with alkaline earth phosphates and the addition of ammonium hydroxide; alkaline earth phosphates were separated with a cerium phosphate carrier. Alpha counts of the mounted cerium phosphate fraction resulted in a gross alpha count that could be assumed to be composed of the radionuclide an individual worked with, and this is typically what occurred over the long history of the program.

As an alternative after the cerium carrier step, sequential chemical separations could isolate plutonium, americium, neptunium, and uranium, respectively, in subsequent steps. Thorium, curium, transplutonic radionuclides, strontium, and polonium were not absorbed on the ion exchange column and would be present in the rinse operation that was conducted as step 2 of the plutonium separation.

The procedure itself was simple and provided consistent results; processing through the cerium carrier step gave acceptable results for plutonium or uranium as long as other radionuclides were not present. The procedure specific to plutonium and isotopes of thorium, uranium, americium, and others was published in a formal analytical procedures manual, MD-20736, in 1968 (Meyer 1992, p. 335). The gross alpha method was preferable due to its simplicity, and it was typically used for individuals who worked only with ²³⁸Pu, as all alpha activity could be assumed to result from ²³⁸Pu.

5.4.4 In Vivo Dose Monitoring

Whole-body counting has most likely been performed since 1960 when the Whole Body Counting Facility was completed. However, whole-body counting to assess accidental lung deposition of ^{238}Pu , ^{239}Pu , and ^{241}Am began in 1969. Chest counting began in the early 1970s using phosphor sandwich (phoswich) detectors.

Extensive modifications were made in the counting room to reduce natural background radiation and prevent influx of airborne contaminants using activated charcoal filtration. By 1982, dual 5-in.-diameter thallium-activated phoswich scintillation detectors were employed to detect radionuclides in the lungs. In addition, a single 4- by 8-in. sodium iodide detector was mounted under the counting bed in 1982 for *in vivo* counting of high-energy photon sources. All monitored persons were decontaminated before counting for a 2,000-second period. Detectors were placed directly on the upper chest while the person was lying flat on the counting bed. Chest wall thickness was determined with an ultrasonic transducer and used to quantify lung burdens using phantom calibration curves, but the documentation does not state when chest wall thickness measurements began. *In vivo* count records are not kept in a central electronic database; each count is maintained as a single hard-copy record. A whole-body count record and a chest count record on the same day are listed on a single hard-copy record.

Reported minimum detectable activities (MDAs) in 1990 ranged from 69.6 to 73.7 nCi for plutonium and 0.18 nCi for americium based on four female and four male subjects (Author unknown 1990). Personnel records from 1994 reported MDAs associated with chest counting of selected individuals and 171 counts (Author unknown 1993–1994).

MDAs were relatively high for the plutonium isotopes due to weak X-ray emissions and a half-value layer in tissue of only 0.6 cm for ^{239}Pu (Langham 1964). *In vivo* chest counting was therefore conducted as a secondary indication of worker contamination in conjunction with primary routine bioassay urinalysis. MDA data have been found to be limited or unavailable for periods before 1990. Dose reconstruction should therefore be based on urine bioassay results to assess missed worker doses. Chest-counting data should only be used to assess intake when counting was conducted as a follow-up for a known worker intake; it is not expected, based on experience with Mound claims, that this will be a common occurrence. Dose reconstructors needing to reconcile chest count results with urinalysis results should contact the lead dose reconstructor for the Mound site when MDA data are not listed with the in-vivo count results.

5.4.5 Personnel Air Sampling Data

A complex array of particulates, aerosols, dusts, and fumes was present during Mound operations. Representative area or personnel air sampling of these airborne materials would require sophisticated sampling equipment, procedures, and calibration efforts. In addition, area air sampling cannot be used effectively in some workplace scenarios and can greatly underestimate actual worker inhalation intakes. Obtaining air sample data would probably entail a time-consuming search of archived site records.

Personnel BZ air sample data should be used in preference to area air sampling data in instances where intakes cannot be estimated from bioassay data and no other means of estimating dose is possible. Area air sampling data should only be used to assign dose if it can be demonstrated to be representative of BZ concentrations. In addition, the DOE claims data do not generally provide air concentration data for nuclides other than tritium.

5.5 BIOASSAY METHODOLOGIES FOR PRIMARY RADIONUCLIDES

5.5.1 Polonium

5.5.1.1 Bioassay Methods

All Mound employees who worked in polonium operations in T, HH, WD, H, B (Biological), and R Buildings were required to submit weekly spot urine samples (Meyer 1992). Monitoring reports indicated that the number of monitored workers increased steadily from 1959 to 1963. Samples were usually collected on Monday and Wednesday, or three samples were collected on Monday, Wednesday, and Friday. Samples were initially treated without HNO₃ digestion. Polonium in raw urine samples was spontaneously deposited on nickel disks and counted using a low-background proportional counter.

Using an improved method with acid digestion, spot urine samples of 50 mL were collected each week and preserved with 50 mg of sulfamic acid before analysis as reported in MLM-460 (Spoerl 1950a). Preserved polonium samples were digested in the plating cell with nitric acid or hydrochloric acid to produce a 1 N final solution before a 2-hour electrodeposition plating time. Electrodeposition was conducted primarily on 1-in. copper disks within a few hours of collection. Recovery was reported in 1950 as 86.1 ±11.9% for 50-mL samples in Spoerl (1950a) and Meyer (1992). Spoerl (1950a) reported that 95% of all recoveries would fall between 62% and 110% based on 310 samples and the assumption of normality. However, three outliers were discarded from the original dataset and the complete dataset distribution exhibited nonnormality. Spoerl (1950a) also reported that samples with activities in excess of 200 cpm/50 mL had reduced recoveries from 56% to 64%. However, urine sample activity above 200 cpm/50 mL is limited to only 0.5% of the total polonium bioassay records in the PORECON database. The procedure stated that ²¹⁰Po losses might occur during the time between sample collection and sample assay (Spoerl 1950a). No specific detection limit (DL) was reported for this method of analysis in Meyer (1992).

All urine bioassay results were reported as cpm in 50 mL of urine and were converted to dpm per 50 mL using an assumption of 2π.

Chemical Recovery Correction

A study conducted in 1989 determined that significant recovery losses from raw undigested urine were observed for metabolized ²¹⁰Po due to plating and/or formation of precipitates (Fellman et al. 1989; Meyer 1992). This study was conducted using urine samples from primates by investigators at the New York Medical Center and by Spitz and Robinson at Mound and published in *Health Physics* (Fellman et al. 1989). Metabolized polonium was shown to have a low average chemical recovery of 10% without complete acid digestion or oxidation by ashing the sample. An average ratio of 10.5 was reported for ashed to unashed urine sample results (Fellman et al. 1989). Recovery of the added unmetabolized polonium tracer typically ranged from 75 to 85% (Fellman et al. 1989) as previously observed at Mound (Spoerl 1950a). Mound had originally assumed that metabolized polonium was plated with the same efficiency as the unmetabolized tracer. Actual chemical recoveries at Mound were therefore potentially only 10% rather than the assumed 86%. Reported bioassay urinalysis results would have to be corrected by a factor of 10 up until 1964 when complete acid digestion of urine samples was required (Meyer 1992).

Dr. Spitz and Dr. Skrable accepted the 10% chemical recovery and applied this correction to evaluate occupational doses from ²¹⁰Po in a retrospective study in 1989 (Spitz 1989a; Skrable 1989; Themalis 1989). In addition, DOE requested that Mound use the 10% chemical recovery to conduct ²¹⁰Po dosimetry (Spitz 1989b). A peer review committee was established in 1987 to study polonium dose methodology. This committee agreed in 1991 that the 10% recovery should be adopted for intake assessment (Spitz 1991).

MJW Corporation conducted a subsequent evaluation of the chemical recovery of polonium (MJW 2002a, Appendix B). The position paper concluded that the 10% recovery observed by Fellman et al. (1989) was not appropriate for correction of Mound worker bioassay results due to differing sample storage times after collection but before analysis. The majority of Mound sample analyses were thought to have occurred within a 4-hour timeframe after collection (Spoerl 1950a). In addition, MJW stated that the 10% chemical recovery for polonium was based solely on primate data and possibly not representative of recoveries observed for human urine samples. MJW therefore decided to use the original recovery that was reported by Spoerl (1950a) of 86%.

However, the data that was reported by Fellman et al. (1989) corroborated early recoveries that were reported by Mound in 1950 for undigested urine samples and demonstrated similar estimates of urinary ^{210}Po excretion for primate and human subjects using undigested urine sample procedures. Differences in sample storage times between the original Mound analyses and the Fellman study did not have any significant effect on reported recoveries. Fellman et al. reported recoveries that ranged from 79% to 85% for all human and primate undigested urine samples in comparison to Spoerl (1950a) at Mound with an average recovery of $86 \pm 11.9\%$. In addition, the Fellman et al. study is in good agreement with a previous Argonne National Laboratory study that was published in 1971 about seven individuals who were exposed to inhalation of ^{210}Po (Sedlet and Robinson 1971). This Argonne study also observed that in contrast to the added ^{209}Po tracer, "only a small fraction of the metabolized and excreted polonium could be deposited directly from acidified urine without preliminary oxidation."

Dose reconstruction is performed using the favorable-to-the-claimant assumptions described in Section 5.8.1.

Reported Half-Life

The effective half-life of ^{210}Po was investigated in 1950 and reported by Spoerl (1950b) to be 36 days. Dose calculations that were used at Mound assumed this effective half-life from 1950 to the end of polonium monitoring. Various quality factors (QFs) that ranged from 10 in the 1950s to 20 in the 1980s were used to determine dose after polonium exposure. International Commission on Radiological Protection (ICRP) Publication 30, Part 1, reported a 50-day biological half-life (ICRP 1979). However, Sheehan (ca. 1951) reported a much shorter effective half-life of only 24 days for exposure that resulted from a puncture wound involving a platinum wire contaminated with ^{210}Po at Mound.

5.5.1.2 Polonium Internal Dose Monitoring Records

Polonium-210 bioassay data were documented on several different types of records at Mound as discussed by Meyer (1992). Weekly Reports of urinalysis data were kept for workers who were grouped by location with entries in cpm per specified volume and 24-hour samples identified. Weekly results were transcribed onto Polonium Cards for individual workers with entries in cpm per 50 mL unless otherwise stated. Twenty-Four Hour Reports were also kept and used to convert individual 50-mL spot samples that were collected on a single day to a volume-weighted average 24-hour excretion in cpm. Finally, Special and Unscheduled Sample Forms were kept when any additional urine samples were required to document possible worker exposures or to confirm previous results with all entries in cpm and duplicate indicated. Bioassay results in activity per day are reported as becquerels per day in the PURECON database under the heading of BQ_DAY. Sample results are also reported in cpm and dpm in this database.

Bioassay data for ^{210}Po have been placed in the PORECON database by MJW by evaluating individual dosimetry files from original logbooks (MJW 2002b). This database contains individual

sample results from 1944 to 1973, which encompasses polonium operations at the Dayton Laboratory³ facilities as well as the Mound Laboratory.⁴

Primary and secondary records exist in most Mound claim files for polonium bioassay. The dose reconstructor should typically use the bioassay results listed in the PORECON database as most convenient. However, it is important to review all claim records to ensure the database reflects all listed bioassay results in the primary data.

5.5.2 Plutonium

In 1952, an attempt was made to electroplate plutonium onto copper plates by following the procedure for ²¹⁰Po. This method proved to be inconsistent and resulted in poor recoveries due to the inability to extract plutonium. An improved method was developed in 1954 and documented in MLM-1003 (Kirby and Brodbeck 1954). This newer method used an alkaline earth phosphate coprecipitation process followed by separation of plutonium with a cerium carrier and counting with a low-background proportional detector (Kirby and Brodbeck 1954; Meyer 1992). Recoveries for this method were reported to be 90% or greater. However, erratic recoveries from 6% to 85% were observed for metabolized plutonium in urine due to plating. This method continued from 1954 to 1960. A chemical recovery of 10% (based on the analysis of reduced polonium recoveries) should be assumed for plutonium bioassay data that was collected from 1954 to 1960.

Starting in 1961, urine samples were acidified to a pH of 2 with nitric acid, which resulted in recoveries for metabolized plutonium of 90% ±3% (Sheehan, Wood, and Kirby 1963). Sample preservation was modified to treat raw urine samples with alkaline phosphates to coprecipitate plutonium and prevent plating of metabolized plutonium present as organic complexes or colloids (Sheehan, Wood, and Kirby 1963). This method was largely continued until 1980 when anion exchange was permanently introduced to separate plutonium from the other alpha emitters. Anion exchange followed by electrodeposition was first introduced in 1966 with the MD-20736 procedure (Monsanto 1968). This procedure was modified in 1978 as MD-80030 to include use of a ²⁴²Pu tracer, counting of blanks to quantify DLs, and alpha spectroscopy (Phillips 1988). However, gross alpha results for bioassay were reported as plutonium for all ²³⁸Pu workers from July 1966 to 1971. Specific isotopic results for plutonium were not reported from 1971 to 1979 until the introduction of alpha spectrometry. Detected alpha activity from 1952 to 1980 was therefore assumed to be plutonium. These results might be overestimates of the true plutonium urine results due to the possible presence of uranium.

Table 5-4 summarizes the MDAs for plutonium isotopes in 24-hour bioassay urine samples over time (MJW 2002b). The lower limit of detection recommended by the Health Physics Society (HPS 1985) for accreditation of bioassay laboratories was 0.06 pCi/L for plutonium as reported in the Quarterly Environmental Science Reports in Meyer (1992). This standard was met in all years of analysis for ²³⁸Pu and ^{239,240}Pu except where MDAs in parentheses were corrected for a 10% chemical recovery.

From 1981 to 1983 average recovery of 85% and counting efficiency of 30% were reported for 3,000-minute counts, and a lower recovery of well below 85% was reported for 1,000-minute counts (MCP 2004). Approximately 90% of all samples were counted for 1,000 minutes.

³ The polonium bioassay program described below was also used in Units I, III, and IV of the Monsanto Chemical Company. Although this document provides dose reconstruction provisions only for the Mound Laboratory, polonium dose may be calculated for the earlier facilities by using the assumptions listed below for the period before 1964.

⁴ Possible exposures to ²¹⁰Po in 1943 are assumed to be insignificant based on limited work in a single laboratory and reported administrative activities before initiation of bioassay in 1944 (Silverman 1962). Some bioassay data from 1944 through the first half of 1945 was reported missing (Spitz 1989a).

Table 5-4. Summary of historical plutonium analytical procedures.

Period	Reporting level ^a	Analytical method
1954–1960	Kirby and Brodbeck 1954 0.05 dpm/1.5 L 0.5 dpm/1.5 L (corrected DL) ^b	Alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; gross alpha for plutonium; proportional counting; recoveries 6%–85% due to colloidal plating of metabolized Pu.
1961–1967	Kirby and Brodbeck 1954 0.1 dpm/1.5 L	Sample acidification; alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; gross alpha for plutonium; proportional counting; recoveries 90% ±3%. Uranium and plutonium results reported separately only in 1966 using anion exchange methods.
1968–1977	MD-20736 (Monsanto 1968) 0.1 dpm/1.5 L	Alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; anion exchange; electrodeposition; gross alpha reported for plutonium; proportional counting.
1978–1979	MD-80030 (Phillips 1988) 0.1 dpm/1.5 L	Alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; anion exchange; electrodeposition; alpha spectroscopy for Pu-238 and Pu-239/240; Pu-242 tracer.
1980–present	MD-80030 (Phillips 1988) 0.07 dpm/1.5 L for routine samples	Alpha spectroscopy for Pu-238 and Pu-239/240; anion exchange; Pu-242 tracer; blanks counted for decision level determination.

- a. The 24-hour reporting levels are based on a standard sample volume of 1.5 L. Standard Man in ICRP Publication 2 (ICRP 1959) and Reference Man in ICRP Publication 23 (ICRP 1975) both have a urine volume of 1.4 L. Actual collected urine volumes varied.
- b. DL was corrected for a low chemical recovery of 10% due to colloidal plating of plutonium.

Plutonium Internal Dose Monitoring Records

Primary and secondary records exist in many Mound claim files for plutonium bioassay. The dose reconstructor should typically use the listed bioassay results in the PURECON database as most convenient. However, it is important to ensure that the database reflects all bioassay results that are listed in the primary data. The dose reconstructor will also need to ensure that PURECON reflects earlier measurements using the gross alpha methodology and ensure that the gross alpha samples were taken as a plutonium intake monitoring program rather than another radionuclide that used the gross alpha bioassay method.

5.5.3 Tritium

No urinalysis data were available from 1948 to 1956; according to Meyer (1992), tritium urinalysis for workers started in 1957. A vibrating Reed Electrometer method of analysis was officially adopted from a Savannah River Plant procedure in 1958 and continued into the early 1960s. This method used 20 mL of urine mixed in a calcium chloride solution to liberate tritium gas, then analyzed the gas by passing an acetylene carrier gas from the urine sample into a 1-L metal ionization chamber. The measured current was therefore proportional to the tritium-acetylene mixture. The reported MDA for this method was 1 µCi/L in urine in Recommended Procedures for Tritium (Meyer 1992). Estimates of accuracy were 1 to 2 µCi/L at 1 µCi/L, 9.5 to 10.5 µCi/L at 10 µCi/L, and 99 to 101 µCi/L at 100 µCi/L (Meyer 1992).

Liquid scintillation (LS) counting began in 1961 followed by an approved procedure in 1968. This methodology used a 3-mL urine sample placed in an Ultimate Gold cocktail solution (Butler 1961). A quench curve was used to correct results for counting efficiency, which was affected by chemical and physical factors that inhibit photon detection. In addition, many liquid scintillation counters (LSCs) used external ¹³³Ba or ²²⁶Ra sources to automatically determine the degree of quench for each sample. Duplicate analyses were conducted to support quality evaluation. The reported MDA for this method was improved from early ionization detection methods to 0.01 µCi/L of tritiated water vapor (HTO) (Phillips 1988). Tritium results were always reported in units of activity per liter of urine. Table 5-5 lists the reported analytical procedures and DLs for tritium.

Table 5-5. Reported analytical procedures and MDAs for tritium.

Period	Analytical method	MDA ($\mu\text{Ci/L}$)
1957–1961	Vibrating reed electrometer	1.0
1962–1995	LS	0.01

Tritium Dose Calculations

A method is presented in this section to convert recorded tritium doses based on LANL dosimetry that was used at Mound to current ICRP Publication 67 and 68 dosimetry (ICRP 1994a, 1995). Correction from ICRP Publication 30 (ICRP 1979) to Publication 67 is not needed due to an overall correction factor of 1.0 as shown in Equation 5-3 below.

A dosimetry method developed at LANL was used for acute tritium dose calculations from 1957 through 1988 (Lawrence 1957). This methodology assumed a maximum permissible body burden of 3.7 mCi, a relative biological equivalent of 1.7, an effective half-life of 12 days, and a body water compartment of 43.4 kg. This results in a maximum permissible concentration (MPC) of 85 $\mu\text{Ci/L}$ for a target dose limit of 300 mrem/wk under the LANL system of dosimetry (Lawrence 1957; DOE 1988). The target dose limit of 300 mrem/wk was based on an organ dose limit of 15 rem (FRC 1960). The LANL dose relationship used at Mound is shown in Equation 5-1:

$$D(t) = 0.00873B \quad (5-1)$$

where

$$\begin{aligned} B &= \text{measured acute body concentration in urine or body water } (\mu\text{Ci/L}) \\ D(t) &= \text{the dose commitment at time } t \text{ after exposure (rem)} \\ \text{Effective } T_{1/2} &= 12 \text{ days} \end{aligned}$$

Reported doses on the Radioactive Gas Exposure Evaluation forms confirm that Mound used Equation 5-1 for tritium dose calculation and that 6 $\mu\text{Ci/L}$ corresponds to 52 mrem as entered on the forms. However, this equation should not be used to reconstruct dose and only provides the confirmed basis to correct doses reported by Mound using Equation 5-2.

Current dosimetry for tritium uses a quality factor (QF) of 1.0 in comparison to an assumed LANL relative biological effectiveness of 1.7, a two-compartment elimination model, and a target mass of 68.8 kg in comparison with 43.4 kg used in the LANL calculations [ICRP Publication 67 (1994a); Publication 68 (1995)]. Equation 5-2 provides a ratio of Publication 68 dose in comparison with LANL dose, which results in a ratio of 0.34:

$$\text{Ratio (ICRP 68/LANL)} = \frac{43.4}{68.8 \times 1.7} \times \frac{\int_0^{\infty} (0.97 \times e^{-0.693 \times t/10} + 0.03 \times e^{-0.693 \times t/40}) dt}{\int_0^{\infty} e^{-0.693 \times t/12} dt} = 0.34 \quad (5-2)$$

Beginning in 1989, tritium doses were reported in terms of effective dose equivalent (DE) under ICRP Publication 30 methodology (ICRP 1979). Equation 5-3 determines the ratio of Publication 68 (ICRP 1995) dose in comparison Publication 30 using the Publication 67 target mass of 68.8 kg (ICRP 1994a). The ratio of 1.0 confirms that no dose corrections would be required for doses reported from 1989 to 1995.

$$\text{Ratio (ICRP 68 / ICRP 30)} = \frac{63}{68.8} \times \frac{\int_0^{\infty} (0.97 \times e^{-0.693 \times t/10} + 0.03 \times e^{-0.693 \times t/40}) dt}{\int_0^{\infty} e^{-0.693 \times t/10} dt} = 1.0 \quad (5-3)$$

Tritium Reporting Limits

Mound reporting limits for tritium affected what doses were reported for monitored workers. Doses in rem or microcuries were reported as blanks on Mound Tritium Exposure Reports when urine concentrations were less than 6 $\mu\text{Ci/L}$ (Meyer 1992). Reporting limits of 3 $\mu\text{Ci/L}$ as discussed by James (1997) or 6 $\mu\text{Ci/L}$ were both reported on the Radioactive Gas Exposure Evaluation Forms as actual bioassay results.

Values of 0.0 were used for nondetection results, which biased the doses below the true estimate of the average dose. MDAs in Table 5-5 should therefore be used to estimate intakes at or below detection.

Stable Metal Tritides

Limited open information is available on stable metal tritides (SMTs) to which workers could have been exposed. Uranium and lithium were most likely used to trap tritium gas in closed tritium handling systems. In general, these materials are reactive with air and water, especially when they are in a finely divided state. Uranium tritide reacts with air to produce HTO. A DOE report notes that lithium tritide reacts with water vapor to produce mostly HT (DOE 1994).

5.6 SECONDARY RADIONUCLIDES INTERNAL DOSE MONITORING PROGRAMS

5.6.1 Radium-Actinium

Section 2 of Meyer (1992) provides a description of bioassay procedures used for ^{226}Ra , ^{227}Ac , and ^{228}Th . A detailed description of the initial procedure is entitled *Indirect Radiochemical Urinalysis for Actinium, Thorium and Radium Isotopes* (Meyer 1992). Additional detail was found in MLM-773 (Kirby 1952), MLM-1003 (Kirby and Brodbeck 1954), and MD-20736 (Monsanto 1968). Radium was extracted from collected 24-hour urine bioassay samples. Intakes of ^{227}Ac and ^{228}Th were thought to be approximately in equilibrium with their radium progeny, subject to the caution that the irradiated ^{226}Ra was allowed to age for several weeks before being processed in the SW Building.

Radium extracted from urine samples was subjected to a series of gross alpha counts for various periods of up to 30 or 35 days. The resultant count data for a single sample was manipulated to form a series of simultaneous linear equations that could be solved for individual count rates due to ^{226}Ra , ^{224}Ra , and ^{223}Ra . Each bioassay urine sample of interest was apparently evaluated in this manner. Interpretation of these bioassay data requires some rather involved calculations.

Due to the large uncertainties implicit in the reconstruction of dose based on these procedures, NIOSH has determined that it cannot estimate internal exposures to radium, actinium, and thorium from the arrival of K-65 sludge in October 1949 through February 28, 1959, when the related area of work was decontaminated and decommissioned and sufficient monitoring was in place. The lack of sufficient information to bound the dose from these exposures, even for workers with specific bioassay monitoring results in their records, formed the basis for the SEC class definition in SEC-00090 (Leavitt 2008).

Radium-226, Actinium-227, and Thorium-228 Records

Although dose from this bioassay method may not be calculated for partial dose reconstructions, the records are described to allow the dose reconstructor to discriminate between these records and records for other bioassay protocols.

Primary bioassay data for these radionuclides were found in four brown spiral notebooks labeled RA#1a and RA#1c and on forms O-318. Manila folders also contained primary bioassay data with collected counts, assumptions, equations, theoretical curve computations, and a written analysis of the observed data (Meyer 1992).

Secondary urinalysis results were written on hand-drawn forms and placed in the "J. B. Black binder." Each form had an employee name, sample identification number, and the ^{223}Ra , ^{224}Ra , ^{226}Ra , and ^{227}Ac results. Secondary data also appeared in various forms such as weekly reports, and on forms O-756 and O-634. The Site Research Database (SRDB) contains available secondary bioassay records for the Radium Irradiation project, which are typically included in the DOE claim files. As stated above, these must be reviewed to separate results from the radium-actinium program, which may not be reconstructed, to other thorium results.

5.6.2 Thorium

5.6.2.1 Thorium-230

Twenty-four hour samples were taken for the urinalysis program for the ionium activities. A thenoyl trifluoroacetone (TTA) extraction procedure was used to identify the ^{230}Th . There is some evidence that the MLM-1003 procedure was used as well, and thorium separation was performed (Meyer 1992, p. 270).

Thorium-230 bioassay procedures are described briefly in MLM-MV-93-93-0003, *History of Mound Bioassay Programs*, Sections 1 and 2 (Meyer 1992). Thorium-230 bioassay was based on 24-hour urine specimens. The objective of the urine bioassay sampling was to detect an MPC that corresponded to a 300-mrem/wk radiation exposure to the critical organ per the metabolic model in use at the time. A count rate of 3.5 cpm for a 24-hour urine sample corresponded to a DE of 300 mrem/wk. Based on Meyer (1992, Section 2), three different urine bioassay procedures were tried for thorium. A TTA urine extraction was the initial procedure (Kirby and Brodbeck 1954). Starting with sample 6507-11, nine samples were analyzed by the "Kirby process-thorium fraction." This apparently refers to the thorium separation and isolation procedure in MLM-1003 from August 1954 (Kirby and Brodbeck 1954, p. 14).

Urine bioassay was subsequently changed to the strategy of radium separation in September 1954 (Meyer 1992). This approach was based on a pair of speculative assumptions. First, Mound appeared to have assumed that intakes of ^{230}Th were in equilibrium with ^{226}Ra . The average ratio of the ^{230}Th to ^{226}Ra in the materials that were handled by Mound workers is not known. However, it is well known that sulfuric acid leaching of uranium ore materials results in a raffinate solution that is enriched in thorium and depleted in radium, and in tails that are enriched in radium and depleted in thorium. The assumption of equilibrium might not be correct. The assumption of secular equilibrium should therefore not be made as discussed and recommended by Meyer (1992). Second, Mound appears to have assumed that the excreted ^{226}Ra was in equilibrium with ^{230}Th . This assumption can be compensated for by using ratios of the incremental urine excretion curves from Potter (2002). Excretion curves for acute and chronic exposures can also be readily calculated using IMBA. ICRP Publication 68 assigns radium to absorption type M, but thorium can be either type M or S (ICRP 1995).

Thorium-232

Monitoring for ^{232}Th began in 1955 to support thorium refining; preliminary research began in the R Building. Exposures to ^{232}Th and progeny could have continued until thorium bulk storage areas were cleaned out (by July 15, 1975). Several rooms in Buildings 38, PP, WD/WDA, HH, SW, and SM had activities that could have resulted in exposure to ^{232}Th .

Thorium-232 Records

Thorium-232 records are diverse due to programs that were conducted over many years. Primary ^{232}Th bioassay data were entered into a small brown spiral notebook marked "Radium-Thorium" and "Radium-Thorium Separation from 8/15/1955 to 2/2/1959" (Meyer 1992). Additional primary ^{232}Th bioassay data were recorded in a large hardcover record book. However, the first 38 pages from this

record book, for July 6, 1959, to January 9, 1961; July 19, 1964, to November 15, 1964; and May 30, 1965, to June 6, 1965, were removed. Secondary records in weekly reports contained ^{232}Th results as cpm per 24-hour sample beginning March 17, 1958. In August 1959, secondary results were reported on form O-756. The available data is a table of secondary ^{232}Th bioassay data extracted from other primary records (Author unknown 1955–1988). Meyer (1992) states that bioassay results for this operation will always have both radium and thorium entries. The corresponding primary ^{232}Th records from the 1950s carry a sample number that ends in “T.” This analysis assumed the “T” result is from the thorium (which includes ^{232}Th and its progeny ^{228}Th) fraction and the “R” indicates that results are from the radium fraction for a given sample.

5.6.3 Protactinium-231

Urinalysis was started for workers to monitor separation and purification program activities in R Building on August 17, 1955 (Meyer 1992). Bioassay data for ^{231}Pa concluded on December 7, 1959 (Meyer 1992). Bioassay was limited to collection of 24-hour samples. A significant volume of urine was required to detect an MPC target dose of 300 mrem/wk due to the very slow elimination of ^{231}Pa .

Samples were subjected to various precipitation and dissolution procedures to extract radium, thorium, and protactinium fractions (Kirby 1952; Kirby and Brodbeck 1954; Kirby 1959). Purification of ^{231}Pa from natural sources was discussed by Kirby and Figgins (1964), Kirby (1959), and Salutsky et al. (1956). Residues were initially filtered from aqueous raffinate solutions after a diethyl ether extraction of uranium. Filtrates were generally dissolved in HCl saturated with NaCl, precipitated with a silicate or cerium carrier and NaOH or NH_4OH , digested in 9 N HCl or HNO_3 solutions, and either counted the precipitate directly or isolated by anion exchange.

The thorium fraction results in cph were converted to cpm, corrected for a 50% counting efficiency, and entered directly into dpm in the database Author unknown (1955–1988). Thorium fraction activity results were then assumed to be ^{231}Pa with a small contribution from ingrowth of decay products. Decay products present in average-age composite solutions at Mound were reported to contribute only 0.05% of the total alpha count (Kirby and Figgins 1964). Analyses were later conducted in accordance with procedure MD-20736 (Monsanto 1968).

5.6.4 Uranium

For uranium, samples were processed using the MLM-1003 procedure (Kirby and Brodbeck 1954). Although Meyer's statement is not definitive, it appears that most routine samples were spot samples that were processed through the cerium-carrier stage. For investigative samples, 24-hour samples were collected and uranium was extracted using the sequential separation protocol (Meyer 1992, p. 321).

Both spot and 24-hour samples were collected from workers for bioassay. The maximum permitted concentration of uranium in urine bioassay samples was 20 cpm in a 1,500-mL urine sample. According to Section 2 of Meyer (1992), the uranium gross count rate for spot samples was scaled up to 1,500 mL on the primary record to determine a 24-hour sample result.

Site-specific values for background uranium dietary intake at Mound have not been listed for Mound workers. Typical intakes of uranium attributable to background should be on the order of 4.9 Bq/yr ^{238}U , 4.9 Bq/yr ^{234}U , 0.2 Bq/yr ^{235}U (UNSCEAR 1993), and 12 Bq/yr for total uranium (NCRP 1992). ICRP Publication 23 (ICRP 1975) reports that uranium background urinary excretion rates for Reference Man range from 0.07 to 0.7 dpm/d in the urine and 2.1 to 2.7 dpm/d in the feces. A single reference (Author unknown undated a) lists a detection level in mass units for natural uranium (NU). This is listed in a 488-page file that is a collection of references with the title of “Historical MDAs at the

Mound site,” and the page in question has “Unknown Document 1” hand-written at the top and is titled “Appendix A, Detectable Amounts for Urinalysis.” There is no evidence that this is a Mound document and, because the bioassay methods listed ^{90}Sr and ^{147}Pr , neither of which was ever performed at Mound, it is not likely that it refers to Mound specifically.

5.6.5 Americium-241 and Curium-244 Records

Documentation for ^{241}Am and ^{244}Cm is very limited. Primary records for ^{241}Am and ^{244}Cm are unknown. Secondary data for ^{241}Am and ^{244}Cm are found in Author unknown (1955–1988), which indicate that ^{244}Cm results are related to the plutonium logbooks. All ^{241}Am and ^{244}Cm urinalysis data are in units of activity (picocuries). MJW (2002b) assumed that ^{241}Am and ^{244}Cm data in Mound dosimetry records such as forms O-756 are in units of activity excreted per 24-hour period. There are no aliquot data in the database.

5.6.6 Rare Isotopes

For most rare isotopes, the parent or more abundant radionuclide is likely to present the greatest dose consequence based on the small amounts of the radionuclides processed. For example, in the production of small amounts of ^{234}U from a ^{238}Pu “cow,” it is the plutonium that represents the highest potential for dose.

5.7 INTERNAL DOSE RECONSTRUCTION GUIDANCE

Most estimates of internal dose are based solely on urinalysis data. For urinalysis, standard parameters are used rather than site-specific values.

5.7.1 General Considerations

5.7.1.1 Use of the Gross Alpha Method

Gross alpha analyses were conducted at Mound and, until 1980, activity was assigned to specific isotopes of plutonium and other alpha emitters based on sequential gross alpha counting or chemical separation followed by gross alpha counting. Alpha spectrometry capable of identifying specific alpha emitting isotopes was not introduced until 1980.

5.7.1.2 Internal Dose Estimation Parameters

Particle Size

This project assumes a particle size of 5 micrometers activity median aerodynamic diameter (AMAD).

Absorption Type

Rather than identifying the most likely absorption type based on the form of the chemical compounds of the radionuclides worked with, the dose reconstruction project makes a determination of the appropriate absorption type based on a fit to the data in the case or based on efficiency measures that assign the absorption type that results in the higher or highest dose for overestimates or the lower or lowest dose when underestimates of internal dose are appropriate. Note that an absorption type specific to Mound has been identified based on historical internal dose measurements at Mound and at LANL. Guidance has been developed to estimate doses to high-fired forms of ^{238}Pu , which can under some circumstances exhibit nonmonotonic clearance rates (ORAUT 2013a). The type L model developed from Mound site data is unlikely to be typical of Mound site exposures to ^{238}Pu , but each potential exposure should be evaluated for applicability of this model. Type L is modeled using specific IMBA input parameters in Table 5-15 later in this document.

Exposure Dates

The assumptions described in ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2007a), are used to establish appropriate estimated intake dates when case data do not suggest an appropriate date to use.

5.7.2 Equilibrium Assumptions

Gross alpha counting was conducted on bioassay urine samples using a procedure of thorium or radium chemical extraction. Individual radium bioassay samples were counted three times in a low-background proportional counter over a period of weeks. Then the individual radium isotope activities were estimated by solving a system of simultaneous equations. This procedure requires equilibrium assumptions to properly infer activity associated with individual decay chain radionuclides. For example, sequential gross alpha counting was conducted on bioassay urine samples after radium extraction by coprecipitation to quantify ^{223}Ra , ^{224}Ra , and ^{226}Ra activity. Evaluation of alpha activity with decay time allowed the activities of individual radium isotopes to be estimated. Table 5-6 lists equilibrium assumptions that are favorable to claimants to infer primary radionuclide activity in urine based on detected alpha activity.

Table 5-6. Recommended equilibrium assumptions for secondary radionuclides.

Radionuclide	Method of inference	Recommendations
Th-230	Gross alpha counting of extracted thorium in urine starting with sample 6507-1I in August 1954. Gross alpha counting of extracted radium in urine beginning with sample 6507-10I from September 1954 to September 9, 1958.	Assume Th-232 and Th-228 have minimal radiological impact to workers from uranium feed material. Assume equilibrium and equal intakes for Th-230 and Ra-226 for exposure to feed materials. Assume MDA for negative bioassay radium result for demonstrated exposure to only thorium compounds.
Th-232 ^b	Gross alpha counting of extracted thorium and radium in urine for monazite refinery workers.	Assume Th-228, Th-232, and Ra-224 are all in equilibrium for exposure to feed materials. ^a Assume minimum possible ratio of 0.42 for Th-228/Th-232 and equilibrium between Ra-224 and Th-228 for exposure to only thorium compounds for doses based on Ra-224 bioassay data. ^a Assume Th-228 and Th-232 are in equilibrium for doses based on gross alpha bioassay data.
Uranium isotopes: U-233, EU ^c , U-234	Gross alpha counting of specific radionuclides.	No assumptions required. Assume all activity is attributable to U-233 in absence of specific radionuclide data. Use U-234 for EU.
Po-210	Electrodeposition followed by low-background proportional counting.	Assume equilibrium with Pb-210 to determine worker intakes.
Uranium isotopes: NU ^d , DU ^e	Fluorometric or gross alpha determination.	Assume NU ratios for U-238, U-234, and U-235 or use IMBA default ratios for NU and DU.

- IMBA does not directly calculate excretion of progeny due to intake of parent radionuclides with progeny.
- Th-232 is also identified as natural thorium and Th-230 as ionium.
- Uranium enriched in U-234 and U-235. The degree of enrichment was not reported. These uranium isotopes would not be present as isotopically pure materials.
- Uranium with naturally occurring mass and activity ratios.
- Uranium depleted in U-234 and U-235.

Mound did bench-scale reworking of K-65 uranium extraction residue that was used at Mallinckrodt, Weldon Springs, and Fernald as a potential source of radium. There was also an initial attempt to produce ^{210}Po initiators by reworking radium and uranium tails. This process was quickly abandoned after the first year. All pre-1946 ^{210}Po intake estimates (at Monsanto Chemical Company facilities) should therefore assume equilibrium with ^{210}Pb .

Extraction operations using Mound uranium cake involved possible exposure to all thorium isotopes from this particular source term. However, ^{232}Th and ^{228}Th had very low activity percentages (0.4%) in comparison with ^{230}Th . Therefore, these thorium isotopes would have resulted in a minimal radiological impact to workers. Because of the relatively low activity abundance of ^{232}Th and ^{228}Th in this source term, it is reasonable to assume for dose reconstruction purposes that all of the activity is from ^{230}Th . Depending on solubility type, this could result in a slight underestimate of dose of less than 2%. Both ^{232}Th and ^{228}Th can therefore be ignored when estimating the intake and doses associated with ^{230}Th .

In vivo data

In vivo records for Mound employees are sparse; the site lagged behind in the use of chest counts for alpha emitters, and whole-body counts were performed only in the later history of the internal dose monitoring program. Some claims do contain whole-body count records, but these often carry a notation saying "Practice Exam" and may not be considered valid records for this reason. Due to low gamma emitter use at the Mound site, valid whole-body count records should only be used to estimate a missed dose in cases where case data imply a clear exposure to beta-emitting radionuclides.

Chest count results may only be used to verify urinalysis results.

Fecal sample data

Fecal samples were also infrequent during most of the history of the Mound site's internal dose monitoring program. When fecal data are present in claim records, they should be reconciled with urinalysis data in estimating the internal dose. Guidance for this is given in ORAUT-OTIB-0060 (ORAUT 2007a). MDAs for fecal data are listed in the table below.

Average percent recoveries for plutonium, uranium, thorium, and americium for fecal samples were determined from an analysis of fecal sample data collected from July to October 1989 Oak Ridge Associated Universities (ORAU) Team files (Author unknown undated b). Table 5-7 summarizes these chemical recoveries as determined using ^{242}Pu , ^{232}U , ^{229}Th , and ^{243}Am as tracers. Specific MDAs for these fecal analyses could not be quantified without reported background sample counts. However, the MDAs appeared to range from 0.01 to 0.04 pCi/sample for ^{238}Pu and from 0.001 to 0.008 for ^{239}Pu , assuming that reported error terms associated with the sample activities were 1 sigma.

Table 5-7. Radionuclide chemical recoveries for collected fecal samples.

Nuclide	Samples	Average	Minimum	Maximum
Pu-242	27	0.77	0.32	1.02
U-232	9	0.69	0.13	1.05
Th-229	2	0.89	0.74	1.03
Am-243	2	0.77	0.71	0.82

For dose reconstruction, resolution of fecal samples may be performed using the MDAs listed in Table 5-8 below. These MDAs were reported as part of a retrospective project to establish MDAs for Mound bioassay procedures conducted in 1995 (Author unknown undated).

Table 5-8. Retroactive MDAs for fecal samples.

Radionuclide	MDA (pCi/sample)	Earliest date
Pu-238	0.012	December 1989
Pu-239	0.010	December 1989
Am-241	0.023	July 1989
U-234	0.27	January 1990

5.8 RECONSTRUCTING DOSE FROM PRIMARY RADIONUCLIDES BASED ON URINALYSIS DATA

This section provides specific information to dose reconstructors for estimating internal dose to Mound workers from the primary radionuclides.

The goal of the internal dose monitoring program at Mound was to conduct bioassay for all programs that could result in internal deposition of radioactive material (Meyer 1992). For this reason, for most claims urinalysis data are abundant for primary radionuclides and the chances that an individual experienced significant intakes in the absence of bioassay data are remote. However, for individuals who, based on case-specific information, can be presumed to have experienced intakes that cannot be based on bioassay data, coworker intakes have been developed as documented in ORAUT-OTIB-0061, *Internal Coworker Doses for the Mound Site* (ORAUT 2012a). For those claims that dose reconstructors determine that the use of coworker dose is necessary, justification should be included in the dose reconstruction report.

5.8.1 Polonium-210

Polonium-210 sample results in the PORECON database are typically complete, but primary data are also present in claim files and should be reviewed to ensure the accuracy and completeness of the PORECON results. Note that PORECON includes results as far back as 1944; results from before 1949 would be for the Dayton Laboratory facilities. Although dose estimation for Dayton Laboratory facilities is outside the scope of this TBD, doses from potential intakes of polonium may be calculated using the dose estimation recommendations in this section.

Urinalysis data for ²¹⁰Po collected before 1964 should be normalized assuming 85% extraction efficiency, then corrected for a 10% chemical recovery. Polonium-210 data that was reported after 1963 should be normalized assuming 85% extraction efficiency and then corrected for a 63% chemical recovery based on the analysis by Fellman et al. (1989). Polonium intakes were controlled using cpm per sample, and were not corrected for aliquot sizes. Sample volumes as reported should always be used when available. Default sample volumes are 50 mL before July 13, 1964, and 100 mL on and after July 13, 1964. Spot samples and 24-hour samples were analyzed using the same analysis procedure. The required corrections are summarized in Equations 5-4 and 5-5:

$$1944-1963 \quad {}^{210}\text{Po Daily Excretion} = \frac{\text{Database cpm}}{0.50} \times \frac{0.85}{0.10} \times \frac{1400 \text{ mL}}{\text{Aliquot mL}} \quad (5-4)$$

$$1964 \text{ onward} \quad {}^{210}\text{Po Daily Excretion} = \frac{\text{Database cpm}}{0.50} \times \frac{0.85}{0.63} \times \frac{1400 \text{ mL}}{\text{Aliquot mL}} \quad (5-5)$$

All urine bioassay results were reported as cpm in 50 mL of urine and were converted to dpm per 50 mL using an assumption of 2π geometry and Equation 5-6:

$$\frac{\text{dpm}}{\text{mL}} = 2 \times \frac{\text{cpm}}{\text{mL}} \quad (5-6)$$

Missed doses should be estimated using MDAs recorded with the results; however, when MDAs are not listed with results, dose reconstructors should use the derived MDAs in Table 5-9.

Table 5-9. Reported background count rates and MDAs for ²¹⁰Po.

Period	Default sample volume (mL)	Detector efficiency (%)	Recovery efficiency (%)	Reported background (cpm/24 hr)	MDAs	
					Spot samples (dpm/50 mL)	24-hr samples (dpm/24 hr)
1944–1963	50	50	10	0.2	6.0 dpm, 168 dpm/d	1.3
1964–1973	100	50	63	0.1	0.72 dpm, 20 dpm/d	0.16

5.8.2 Plutonium

In most cases, presumptive exposures of Mound employees to plutonium can be assumed to result from heat source plutonium. However, claim data must be reviewed to ensure that this is indeed the case, and that the worker was not also (or instead) exposed to potential intakes of weapons-grade material.

The dose reconstructor should keep in mind that both gross alpha and alpha spectrometric programs were used to detect ²³⁸Pu, and that the PURECON database results should be verified against the primary data in claim files, even though PURECON results are typically listed as gross alpha results when this is the case. Isotopic results that list both ²³⁸Pu and ²³⁹Pu begin to appear in 1984. This should not typically be presumed to represent exposures to both types of plutonium. Rather, this indicates that the spectrographic analysis described in Section 5.5.2 above was used.

The radionuclides associated with both plutonium source terms are listed in Tables 5-10 and 5-11 for heat-source plutonium and weapons-grade material, respectively.

Table 5-10. Heat source plutonium feed material characteristics.^a

Nuclide	% Mass	% Activity by effective age ^b		
		1.0 yr	3.2 yr	10 yr
Pu-236	0.0001	0.0	0.0	0.0
Pu-238	80.2	95.6	95.9	96.8
Pu-239	15.9	0.1	0.1	0.1
Pu-240	3.0	0.0	0.0	0.1
Pu-241	0.6	4.3	4.0	3.0
Pu-242	0.1	0.0	0.0	0.0
Am-241	0.0	0.0	0.0	0.1

a. Source: EG&G (1995).

b. Effective age is the age when ²⁴¹Am activity is zero.

Plutonium urinalysis data from 1954 to 1960 should be normalized assuming 85% extraction efficiency and then corrected for a 10% chemical recovery due to plating of organic complexes or colloidal plutonium before sample analysis (Sheehan, Wood, and Kirby 1963; Spoerl 1950a). This correction is not required for later samples. These corrections are summarized in Equations 5-7 and 5-8:

Table 5-11. Weapons-grade plutonium feed material characteristics.^a

Nuclide	wt%	% Activity by effective age ^b (Ci/g)			
		Fresh (0 yr)	5 yr	10 yr	15 yr
Pu-238	0.05	8.56E-03	8.23E-03	7.91E-03	7.6E-03
Pu-239	93.1	5.77E-02	5.77E-02	5.77E-02	5.77E-02
Pu-240	6	1.36E-02	1.36E-02	1.36E-02	1.36E-02
Pu-241	0.8	8.24E-01	6.48E-01	5.09E-01	4.00E-01
Pu-242	0.05	1.97E-06	1.97E-06	1.97E-06	1.97E-06
Am-241	0.00	0	5.83E-03	1.04E-02	1.39E-02

a. Source: PNNL (2003).

b. Effective age is the age when ²⁴¹Am activity is zero.

$$1957-1960 \quad {}^{238,239}\text{Pu Daily Excretion} = \frac{\text{Pu database dpm} \times 0.85}{0.10} \quad (5-7)$$

$$1960-2006 \quad {}^{238,239}\text{Pu Daily Excretion} = \text{Pu database dpm} \quad (5-8)$$

From 1981 to 1983, average recovery of 85% and counting efficiency of 30% were reported for 3,000-minute counts, and a lower recovery of well below 85% was reported for 1,000-minute counts (MCP 2004). Approximately 90% of all samples were counted for 1,000 minutes.

The MDAs in Table 5-12 should be used to estimate missed dose. If the sample count time is unknown, then the 1000-minute MDAs should be used to reconstruct doses. Daily excretion should be determined using Equations 5-7 and 5-8 as appropriate.

Table 5-12. Plutonium MDAs (pCi/sample of urine).^a

Period	Pu-238 MDAs		Pu-239/240 MDAs	
	$t_c = 1,000$ min	$t_c = 3,000$ min	$t_c = 1,000$ min	$t_c = 3,000$ min
Before 1957	(0.85)		(0.85)	
1957	(0.94)		(0.94)	
1958	(0.6)		(0.6)	
1959	(0.5)		(0.5)	
1960	(0.5)		(0.5)	
1961-1966	0.05		0.05	
1967-05/1981	0.11		0.11	
06-12/1981	0.066	0.027	0.044	0.020
1982	0.063	0.023	0.042	0.017
1983	0.064	0.024	0.043	0.018
1984	0.066	0.026	0.045	0.019
1985	0.047	0.017	0.031	0.012
1986	0.042	0.015	0.028	0.011
1987	0.022		0.011	
1988	0.017		0.01	
01-07/1989	0.019		0.008	
08/1989-12/1991	0.018		0.008	
1992-1995	0.018 ^a		0.009 ^a	

a. t_c = count time; plutonium MDAs from before 1957 to 1960 were normalized assuming 85% extraction efficiency and then corrected for 10% chemical recovery due to plating of organic complexes or colloidal plutonium before sample analysis. Based on Mound (1996).

Table 5-12 summarizes the MDAs for plutonium isotopes in 24-hour bioassay urine samples over time (MJW 2002b). The lower limit of detection recommended by American National Standards Institute (HPS 1985) for accreditation of bioassay laboratories was 0.06 pCi/L for plutonium as reported in the Quarterly Environmental Science Reports in Meyer (1992). This standard was met in all years of analysis for ²³⁸Pu and ^{239,240}Pu except where MDAs in parentheses were corrected for a 10% chemical recovery.

5.8.2.1 Special Considerations for Plutonium-238

Plutonium-238 has a higher specific activity than ²³⁹Pu; for this reason its solubility is typically higher than weapons-grade plutonium mixtures. However, based on data from an incident at Los Alamos National Laboratory (LANL), and on an analysis of several results at Mound, some exposures to heat

source material can exhibit nonmonotonic excretion curves. The dose implications of the complex excretion pattern are described by ORAUT (2013a).

A dissolution model that better describes the unusual ²³⁸Pu urinary excretion patterns that have been observed at Mound was derived from five Mound intake cases reported by Guilmette, Griffith, and Hickman (1995), and described in ORAUT (2013a). The Mound-specific dissolution type is called type L to distinguish it from type J, a LANL-specific model. The type L dissolution model uses custom dissolution functions within the framework of the standard ICRP Publication 66 respiratory tract model (ICRP 1994b). The use of such custom dissolution functions is the approach recommended by the ICRP and allows the models to be implemented in IMBA.

There are certain intake scenarios for which the assumption of standard types M and S plutonium gives higher organ dose estimates than the assumption of type L plutonium, and there are scenarios where the assumption of type L plutonium gives higher dose estimates. The dissolution types that give the highest organ doses for typical dose reconstruction scenarios are identified in Tables 5-13 and 5-14. For Table 5-13, the urine sample is assumed to be collected at the end of the listed chronic intake period.

Table 5-13. Summary of limiting dissolution type versus the duration of a chronic inhalation intake of ²³⁸Pu.

Organ or tissue	Chronic intake period (d)					
	10,000	1,000	400	100	40	10
Adrenals	S	S	S	S	L	L
Urinary bladder	S	S	S	S	L	L
Brain	S	S	S	S	L	L
Breast	S	S	S	S	L	L
Gall bladder	S	S	S	S	L	L
Heart wall	S	S	S	S	L	L
Kidneys	S	S	S	S	L	L
Liver	S	S	S	S	L	L
Muscle	S	S	S	S	L	L
Ovaries	S	S	S	S	L	L
Pancreas	S	S	S	S	L	L
Testes	S	S	S	S	L	L
Thyroid	S	S	S	S	L	L
R.B.M.	S	S	S	S	L	L
Bone surface	S	S	S	S	L	L
Stomach	S	S	S	S	L	L
S.I.	S	S	S	S	L	L
U.L.I.	S	S	S	S	L	L
L.L.I.	S	S	S	S	L	L
Skin	S	S	S	S	L	L
Spleen	S	S	S	S	L	L
Thymus	S	S	S	S	L	L
Uterus	S	S	S	S	L	L
ET	S	S	S	S	S	S
Lung	S	S	S	S	S	S
Colon	S	S	S	S	L	L
ET1	S	S	S	S	S	L
ET2	S	S	S	S	S	S
LN(ET)	S	S	S	S	S	S
BBsec	S	S	S	S	S	L
BBbas	S	S	S	S	S	L
bb	S	S	S	S	S	S

Organ or tissue	Chronic intake period (d)					
	10,000	1,000	400	100	40	10
AI	S	S	S	S	S	S
LN(TH)	S	S	S	S	S	S
Esophagus	S	S	S	S	L	L

Table 5-14. Summary of limiting dissolution type versus the time after an acute intake a single urine sample is collected.

Organ or tissue	Day sample collected after acute intake (d)							
	10,000	1,000	400	100	40	15	10	2
Adrenals	M	S	S	S	S	S	L	L
Urinary bladder	M	S	S	S	S	S	L	L
Brain	M	S	S	S	S	S	L	L
Breast	M	S	S	S	S	S	L	L
Gall bladder	M	S	S	S	S	S	L	L
Heart wall	M	S	S	S	S	S	L	L
Kidneys	M	S	S	S	S	S	L	L
Liver	M	S	S	S	S	S	L	L
Muscle	M	S	S	S	S	S	L	L
Ovaries	M	S	S	S	S	S	L	L
Pancreas	M	S	S	S	S	S	L	L
Testes	M	S	S	S	S	S	L	L
Thyroid	M	S	S	S	S	S	L	L
R.B.M.	M	S	S	S	S	S	L	L
Bone surface	M	S	S	S	S	S	L	L
Stomach	M	S	S	S	S	S	L	L
S.I.	M	S	S	S	S	S	L	L
U.L.I.	M	S	S	S	S	S	L	L
L.L.I.	M	S	S	S	S	S	L	L
Skin	M	S	S	S	S	S	L	L
Spleen	M	S	S	S	S	S	L	L
Thymus	M	S	S	S	S	S	L	L
Uterus	M	S	S	S	S	S	L	L
ET	S	S	S	S	S	S	S	L
Lung	S	S	S	S	S	S	S	L
Colon	M	S	S	S	S	S	L	L
ET1	S	S	S	S	S	S	S	L
ET2	S	S	S	S	S	S	S	L
LN(ET)	S	S	S	S	S	S	S	S
BBsec	S	S	S	S	S	S	S	L
BBbas	S	S	S	S	S	S	S	L
bb	S	S	S	S	S	S	S	L
AI	S	S	S	S	S	S	S	S
LN(TH)	S	S	S	S	S	S	S	S
Esophagus	M	S	S	S	S	S	L	L

The overall conclusion is that standard plutonium dissolution types (M and S) should be assumed for cases where urine samples are collected more than two weeks after an acute intake and for chronic intakes in excess of three months in duration. Under these conditions types M and S will give the highest organ doses for all organs (i.e., type M or S plutonium is limiting). The type L dissolution type should be used for cases that do not meet these criteria. Therefore, in practice, type L should be added to the evaluation regimen and applied in the same fashion as types M and S, selecting whichever gives the largest dose for the particular case.

Parameters that relate to the type L model are reproduced in Table 5-15 along with those for types M and S.

Table 5-15. IMBA input parameters for types M, S, and the nonmonotonic model L.

Rate constant	Absorption type		
	M	S	L
$k_{p,t}$	90	100	0.02
$k_{t,blood}$	0.005	0.0001	0.003341
$k_{p,blood}$	10	0.1	1.00 E-06
k_r	100	100.1	0.02
k_s	0.005	0.0001	0.003341
f_r	0.09995	0.001	-0.20048

5.8.2.2 Special Considerations for Plutonium-239

It might be necessary for the dose reconstructor to infer exposure to one or the other of the two plutonium source terms based on claim-specific information. For the most part, ^{239}Pu exposures would be to workers in laboratory settings. During part of the time, R Building results were noted as such in bioassay logs and represented presumptive exposures to weapons-grade materials (Meyer 1994); SM Building results were assumed to represent potential exposures to heat-source material.

In general for the dose reconstruction project, when an exposure to weapons-grade plutonium is presumed, the chance of exposure to highly insoluble plutonium is a possibility that cannot be ignored, and the Mound site is no different in this respect. Dose reconstructions for Mound should evaluate the potential for exposure to type super S plutonium in accordance with ORAUT-OTIB-0049, *Estimating Doses for Plutonium Strongly Retained in the Lung* (ORAUT 2010). As stated above, such exposures were likely to be uncommon at the Mound site because work with the ^{239}Pu isotope was a small part of the work that was conducted. If presumptive exposures to weapons-grade material are assumed, the basis should be documented in the dose reconstruction report.

5.8.3 Tritium

Meyer describes the initiation of tritium research as beginning in 1957, and bioassay records begin in that year. Information in EG&G (1995) implies that research began in 1947 in R Building, but that was before construction of R or T Buildings and there are no specific records that suggest that the material was actually used, handled, or stored at Mound during this period. If claim data suggest tritium exposure (positive tritium doses in MESH), dose may be assigned before this time. Zero tritium doses in the MESH database for years before the beginning of tritium bioassay (before 1957) are artifacts from the recordkeeping system and are not to be assumed to be presumptive exposures for which missed dose is required to be assigned.

For 1957 onward the listed and zero doses may be used, with equation 5-2, to assign a dose only when dose overestimates are appropriate. If they are not appropriate, it is necessary for the dose reconstructor to review tritium logbooks for the employee's name for the years in question and to assign dose from tritium bioassay, or missed dose based on these records instead.

Bioassay results that are reported at or below detection or as zeros should be corrected using the applicable MDAs in Table 5-16. Although a class of Mound employees has been added to the SEC to include 1972, 1975 and 1976, tritium logbook data are available for portions of the years 1972 and 1976.

Table 5-16. Reported analytical procedures and detection limits for tritium.

Period	Analytical method	Detection limit ($\mu\text{Ci/L}$)
1948–1956	No urinalysis conducted	Use 95% dose ^a
1957–1961	Vibrating reed electrometer	1.0

1962–1995	Liquid scintillation	0.01
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- a. The upper 95 percentile dose of 340 mrem should be used to reconstruct doses.

Stable Metal Tritides

Work was performed with SMTs at Mound, as well as with elemental tritium and HTO. Due to the concern that urinalysis bioassay samples for tritium, together with the metabolic model that was used, are unlikely to generate accurate dose estimates for the less soluble tritides, an evaluation was made of other data sources.

For reconstruction of doses, data from urinalysis bioassay are to be used for the small number of personnel performing actual “hands-on” work with SMT. These individuals can be identified by name and, in this case, doses should be reconstructed using the methodology described in ORAUT (2007b). Dose reconstructors should contact the lead dose reconstructor for Mound when determining whether specific individuals worked with special tritium compounds.

For other workers, doses are likely to be small for all organs, as described in ORAUT-RPRT-0057, *A Method for Estimating Stable Metal Tritide Exposures to Tritium Workers Based on Swipe Data in Rooms SW-9, SW-13, SW-150, and R-108 at the Mound Laboratory, 1968 to 1989* (ORAUT 2013b). This report describes a method for estimating doses to incidentally exposed workers.

5.9 RECONSTRUCTING DOSE FROM SECONDARY RADIONUCLIDES BASED ON URINALYSIS DATA

5.9.1 Radium-Thorium-Actinium

For the actinium refinery project in the Old Cave, it has been determined that it is not possible to estimate dose or a maximum dose with sufficient accuracy. Therefore, these doses may not be reconstructed, and only partial dose reconstructions are possible, in accordance with the wording of the designation of the class listed in (Leavitt 2008).

5.9.2 Thorium

5.9.2.1 Thorium-232

Bioassay for ^{232}Th was based on 24-hour urine samples and followed Mound bioassay procedures MLM-460 (Spoerl 1950a) and MD-20736 (Monsanto 1968). Mound conducted alpha counting of extracted thorium and radium in urine for monazite refinery workers. This was the gross alpha procedure, with specific analysis for radium and thorium. Meyer (1992) states that for these workers, bioassay results will always contain both thorium and radium results. It is assumed that ^{228}Th , ^{232}Th , and ^{224}Ra are in equilibrium in feed materials; however, an examination of the ratios of radium to thorium in these results shows that the proportions vary. To estimate doses from these results, dose reconstructors should:

- Calculate dose from ^{224}Ra in the mixture based on the radium result;
- Assume the thorium result is composed of ^{228}Th and ^{232}Th in equal proportions.

Meyer (1992, Section 1) indicates that 2.8 cpm on a 24-hour urine sample corresponded to an intake that would result in a 300-mrem/wk DE to the critical organ under the dosimetry system in effect when the sample was collected. Where no aliquot volume is given in the claimant data, assume that the dpm value has been corrected for background, detection efficiency, and represents a 24-hour urine excretion. If an aliquot volume is given, then assume that the bioassay result is in dpm per aliquot. No other corrections are required for urinalysis results in dpm. If urinalysis results are in cpm, then a

correction for 50% detector efficiency and 92% extraction efficiency should be made to determine dpm results unless it is clear from primary records that these corrections have already been made.

Table 5-17 lists ²³²Th DLs by period. These DLs have units of dpm excreted per 24-hour when no aliquot size is given. However, DL units should be dpm per aliquot, if an aliquot volume was reported.

Table 5-17. Natural thorium DLs (dpm).

Period	DL	Reference
Before 12/1987	0.3	MDA (MJW 2002b) ^a
12/1987–1994	0.02 (Th-232)	MDA (Barton 1995)
01/1994–present	0.08 (Th-228)	MDA (Barton 1995)

a. Correction for aliquot size should be made if available. Most database entries do not have reported aliquot volumes.

Thorium-232 bioassay results in Author unknown (1955–1988) contain results for radium, thorium, or both. If a radium result is provided for a ²³²Th urine bioassay, the primary record sample identification number usually ends in “R” and refers to ²²⁴Ra. From these results, the daily excretion of ²²⁴Ra can be obtained using Equation 5-9:

$$\text{Daily Excretion Ra-224} = \text{Ra Database dpm} \div 90\% \quad (5-9)$$

A primary record identification number that ends in “T” corresponds to a natural thorium result that includes ²³²Th and ²²⁸Th. The daily excretion of natural thorium can be calculated using Equation 5-10:

$$\text{Daily Excretion Th} - 228 + \text{Th} - 232 = \text{Th Database dpm} \div 92\% \quad (5-10)$$

Equations 5-9 and 5-10 include correction factors of 90% and 92%, respectively, for radiochemical yield because database results for radium and thorium have not been corrected for chemical extraction.

Local background thorium urinary excretion rate data is not available. However, ICRP Publication 23 (ICRP 1975) reports that background excretion rates for natural thorium range from 0.05 to 1.0 dpm/d, and that Reference Man excretes 0.05 dpm/d. For dose reconstruction, assume that background natural thorium in the diet of monitored workers resulted in urinary excretion of 0.05 pCi/d (ICRP 1975).

5.9.2.2 Thorium-230

Gross alpha counting of extracted thorium in urine started with sample 6507-11 in August 1954. Gross alpha counting of extracted radium in urine began with sample 6507-10I in September 1954 and continued until September 9, 1958. Dose reconstructors should assume:

- ²³²Th and ²²⁸Th from uranium feed material have minimal radiological impact to workers.
- The MDA for negative bioassay radium result for demonstrated exposure to thorium compounds only.

When reconstructing a particular dose history, careful attention must be paid to the reporting basis that Mound used because several different bioassay strategies were attempted for ²³⁰Th. If the reporting basis for an ionium bioassay result is ambiguous, assume that ²³⁰Th was extracted and counted. Excretion rates in dpm/d should be calculated using Equation 5-11 and entries from the

Author unknown (1955–1988) that contain information on volume, aliquot size, and dpm results, unless contradicted by another bioassay record:

$$\text{Daily Excretion Th-230} = \text{Database dpm} \times \frac{\text{Sample mL}}{\text{Aliquot mL}} \quad (5-11)$$

If no aliquot volume is given and no definitive primary record is available, then assume that the sample volume is equal to the aliquot volume. No other corrections should be made for urinalysis results in dpm. If urinalysis results are in cpm, then a correction for 50% detector efficiency and 90% extraction efficiency should be made to determine dpm results.

Table 5-18 lists available ²³⁰Th DLs. Where no aliquot size is given in Author unknown (1955–1988) assume the DL is in dpm excreted per 24-hour sample.

Table 5-18. ²³⁰Th DLs (dpm).^a

Period	DL	Reference
Before 01/1994	0.3	MDA (MJW 2002b) ^a
01/1994–present	0.17	MDA (Barton 1995)

a. DL recommended for all secondary radionuclides (MJW 2002b).
Correction for aliquot size should be made if available.

5.9.3 Protactinium-231

The primary program for ²³¹Pa was completed in 1959. For partial dose reconstructions, doses from potential intakes of ²³¹Pa may be included only when records in the case show that bioassay was conducted.

Intakes to workers should be estimated only for exposures in HH, R, and SW Buildings. Activity data in dpm should only be used as taken from Author unknown (1955–1988) for the thorium fraction. MDAs in Table 5-19 should be used to reconstruct missing results at or below detection. An MDA of 0.3 dpm should be assumed for all exposures that occurred from 1954 to August 17, 1955. Daily excretion should be estimated using Equation 5-12. If no aliquot size is listed, the default aliquot size is 120 mL.

Table 5-19. Protactinium DLs (dpm).^a

Period	Background count rate (cpm)	DL (dpm)
1954–08/17/1955	No bioassay	0.3 ^b
08/17/1955–1956	0.3	0.3
1957–1958	0.2	0.02
1959–12/07/59	0.15	0.08

a. Correction for aliquot size should be made for the DL if available.
b. A DL of 0.3 dpm should be used when there are no bioassay data available.

Reported bioassay results in cph were generally corrected for a 78% extraction efficiency, 50% detector efficiency, and background counting rate. However, no corrections were made for aliquot volumes in primary records. An evaluation of the ²³¹Pa in Author unknown (1955–1988) determined that some sample volumes were not reported and the modal aliquot was 120 mL. If aliquot or sample volumes are not reported, a modal aliquot of 120 mL and a Standard Man sample volume of 1,400 mL should be assumed to correct reported results in terms of daily excretion. Protactinium-231 daily excretion should only be inferred from the thorium fraction dpm from Author unknown (1955–1988), as shown in Equation 5-12, using actual sample and aliquot volumes or by assuming a standard man sample volume of 1400 mL and a modal aliquot volume of 120 mL:

$$\text{Daily Excretion Pa-231} = \text{Th Fraction Database dpm} \times \frac{\text{Sample or 1400 mL}}{\text{Aliquot or 120 mL}} \quad (5-12)$$

5.9.4 Uranium

The screening procedure for uranium is likely to be simply the gross alpha procedure through the cerium carrier step, and the results were assumed to be uranium based on the worker's job assignment. Sequential separation would isolate uranium only, so the original results should be checked in the logbook if an exposure to uranium is suspected. Dose reconstruction assumptions are the same for both contingencies:

- All activity is attributable to ²³³U in absence of specific radionuclide data.
- Use ²³⁴U for EU. For NU or depleted uranium (DU), fluorometric or gross alpha determination.
- Assume NU ratios for ²³⁸U, ²³⁴U, and ²³⁵U or use IMBA default ratios for NU and DU.

Table 5-20 lists uranium DLs. Equation 5-13 provides correction for aliquot volume, if necessary:

Table 5-20. Uranium DLs (dpm).

Period	DL	Reference
Early 1950s–1985	0.4 (NU) µg/L	Author unknown undated a ^a
Before 05/1985	0.3	MDA (MJW 2002b) ^b
05/1985–present	0.07 (U-234) 0.03 (U-235) 0.09 (U-238)	MDA (Barton 1995)

- a. Use for fluorometric results if reported. Statistical basis for this MDA in urine is unknown.
- b. Correction for aliquot size should be made for the DL if available.

$$U \text{ Daily Excretion} = \text{Database dpm} \times \frac{1400 \text{ mL}}{\text{Aliquot mL}} \quad (5-13)$$

In addition to gross alpha and specific analysis for uranium, one information source lists an MDA of 0.4 µg/L for uranium fluorometry (Author unknown undated a). However, no evidence could be found that this bioassay method was ever used at Mound. Dose reconstructors should, if they find records of fluorometry (using mass instead of activity units), assume the value above as the DL. The source lists this DL for NU, and a specific activity of 683 pCi/mg for NU should be used to convert the value to activity for use in the IMBA code.

5.9.5 Reactor Fuels Projects

Most projects involved physical testing of fuel forms and compounds, which did not involve the possibility of significant intakes. For activities that did involve potential intakes, bioassay records will be present in claim information for internal dose monitoring for plutonium, uranium, or thorium. Dose may not be calculated for presumptive exposures without specific claim information.

5.9.6 Reactor Waste Project

Processing of milliliter aliquots of reactor waste in laboratory conditions is unlikely to have resulted in significant intakes of radioactive material. However, bioassay samples for uranium and plutonium may be included in claim files. Dose may not be calculated for presumptive exposures without specific claim information.

5.9.7 Americium and Curium

Mound had the ability to selectively monitor for exposure to americium using the gross alpha and sequential separation protocols described above. If a specific exposure to americium is determined to be likely, as opposed to a potential intake of americium as a constituent of a plutonium mixture, it is important to keep in mind that a specific bioassay for americium might have been coded in PURECON as a plutonium result; an analysis of the bioassay log for that year should give insight into the actual result.

Americium present as part of a mixture of plutonium is included in the intake of ²³⁹Pu. For heat source plutonium the mixture is assumed to be composed of 100% ²³⁸Pu.

Americium-241 alpha sources were electroplated in Room R-120 between 1956 and 1962, and it was the major isotope of concern in that room. Exposure to ²⁴¹Am in R-120 could also have occurred coincident with intakes of weapons-grade plutonium.

Bioassay Procedures and DLs

Dose reconstructors should assume that all data in the ORAU database are in units of pCi excreted per 24 hours and that no additional corrections are required. The DL of 0.3 recommended by MJW (2002b) is consistent with the DL of 0.2 for the period from the 1950s to 1989. The 0.3 DL should therefore be used as the estimate of the DL from the 1950s to July 1989 as more favorable to claimants. Table 5-21 lists DLs by period for ²⁴¹Am and ²⁴⁴Cm.

Table 5-21. ²⁴¹Am and ²⁴⁴Cm DLs (dpm excreted per 24 hours).

Period	DL (dpm/d)	Reference
Early 1950s–1989	0.2	Author unknown undated a ^a
Before 07/1989	0.3	MDA (MJW 2002b) ^b
07/1989–present	0.05	MDA (Barton 1995)

- a. Statistical basis is unknown and therefore recommend using 0.3 dpm/d as reported by MJW (2002).
- b. Correction for aliquot size should be made for the DL if available.

Daily excretion should be determined using Equation 5-14.

$$\text{Daily Excretion Am-241 or Cm-244} = \text{Database pCi} \quad (5-14)$$

Americium-241 and Curium-244 Recommendations

Exposure to ²⁴¹Am in heat source plutonium should be considered negligible. Intakes of ²⁴¹Am from exposure to weapons-grade plutonium should be estimated using the percent activities in Table 5-12. Dose reconstructors should assume the bioassay data in the ORAU database are for personnel who were working with ²⁴¹Am and ²⁴⁴Cm source materials (Author unknown 1955–1988).

All data in the ORAU database are in units of pCi excreted per 24 hours, and no additional corrections are required. The 0.3 DL should be used as the estimate of the DL from the 1950s to July 1989 as more favorable to claimants than a lower value reported anecdotally for this period.

Table 5-22. ²⁴¹Am and ²⁴⁴Cm detection limits (dpm/24 hours).

Period	Detection limit	Reference
Before 7/1989	0.3	MDA (MJW 2002) ^a
July 1989–present	0.05	MDA (Barton 1995)

- a. Correction for aliquot size should be made for the DL if available.

5.9.8 Rare Isotopes

Very few workers were involved in this program, which yielded small quantities of the rare isotopes of interest over the years of its operation. Internal dose monitoring was likely to reflect the progenitor radionuclides of ^{238}Pu or ^{233}U . For ^{238}Pu -derived radionuclides, the standard assumption that the exposure was to 100% ^{238}Pu (neglecting the presence of the constituent radionuclides resulting in lower doses) will overestimate dose from the rare radionuclide of interest. The same is true when the standard assumption that exposure to uranium mixtures were composed of 100% ^{234}U . Dose may not be calculated for presumptive exposures without specific claim information.

5.9.9 Radon

NIOSH is unable to put an upper bound on the radon exposures in SW-19 until the point at which a ventilation system was installed in early 1980 (before March 5, 1980) to vent SW Building tunnel air. The inability to bound the concentration in SW-19 extends to the contiguous air spaces in the SW and R Buildings. The lack of sufficient information to bound the dose from radon and its progeny formed the bases for the SEC class definitions in SEC-00171 (Sebelius 2010) and SEC-00207 (NIOSH 2012). The recommended WLM values in Table 5-23 should be used to reconstruct doses from radon exposures for the buildings and periods shown.

Table 5-23. ^{222}Rn WLM assumptions for dose reconstruction.

Building conditions	Period	Radon (pCi/L)	Radon (WL)	Radon (WLM/y)
SW-19 postproduction and after addition of ventilation	03/06/1980–1995	0.7–13.4	0.03	0.4
Building 21	1966–1995	17.5–125	0.85	10.0
Building 48	1980–1995	3.2	0.01	0.2
R Building Ra-Ac processing	1949–1955	2.1	0.009	0.1
All other buildings	1949–1995	0.9	0.004	0.05
Background–indoors	1949–1995	0.5	0.002	0.03

All WLMs should be assumed to be median values of lognormal distributions with a geometric standard deviation (GSD) of 3.0.

5.9.10 Summary of Correction Factors and Excretion Equations

Extraction efficiencies should be used to correct data that are reported in cpm or cph. Protactinium-231, ^{230}Th , and uranium data have been corrected for chemical recovery in Author unknown (1955–1988). However, if any data for these radionuclides are reported in cpm or cph, extraction efficiencies of 78%, 90%, and 80% should be used for ^{231}Pa , ^{230}Th , and uranium, respectively. PORECON database entries for ^{210}Po must first be normalized assuming 86% extraction efficiency and then corrected for a chemical recovery 10% before 1964 and 63% for 1964 and after.

A summary of corrections for all radionuclides is provided below in Table 5-24. The primary data types are listed with corrections necessary to estimate intake. In addition, equations are provided to calculate daily excretion. Most radionuclides are in units of dpm and do not require detector or extraction efficiency corrections except the radium-actinium data in cph. The radium-actinium and ^{241}Am - ^{244}Cm data do not require aliquot corrections. The uranium and ^{230}Th data require aliquot corrections when an aliquot volume is available, and MDAs should also be corrected when necessary. If no aliquot volume is reported, MDAs should be assumed to be dpm/24-hour sample.

Table 5-24. Summary of correction factors and excretion equations.

Nuclide	Period	Primary data type	Aliquot correction	Detector efficiency ^a	Extraction efficiency ^b
Po-210 ^c	1944–1963	cpm or Bq/d	no	no	10
	1964–1973	cpm or Bq/d	no	no	63
	<i>Daily Excretion Po-210 = Po Database Bq/d × 86% ÷ 10% (1944 to 1963)</i> <i>Daily Excretion Po-210 = Po Database Bq/d × 86% ÷ 63% (1964 to 1973)</i>				
Pu-238 Pu-239	1957–1960	dpm	no	no	10
	1961–1966	dpm	no	no	no
	1967–5/81	dpm	no	no	no
	06/1981–1995	dpm	no	no	no
<i>Daily Excretion Pu = Pu Database dpm × 85% ÷ 10% (1957 to 1960)</i> <i>Daily Excretion Pu = Pu Database dpm (1961 to present)</i>					
Pa-231	08/17/1955–1956	dpm	yes	no	no
	1957–1958	dpm	yes	no	no
	1959–12/1959	dpm	yes	no	no
<i>Daily Excretion Pa-231 = Thorium Database dpm × $\frac{\text{Sample mL}}{\text{Aliquot mL}}$</i>					
Th-230	Before 01/1994	dpm	yes	no	no
	01/1994–1995				
<i>Daily Excretion Th-230 = Database dpm × $\frac{\text{Sample mL}}{\text{Aliquot mL}}$</i>					
Th-232	Prior 12/87	dpm	yes	no	92
	12/87–1994				
	1/94–1995				
<i>Daily Excretion Ra-224 = Ra Database dpm ÷ 90%</i> <i>Daily Excretion Natural Thorium = Th Database dpm ÷ 92%</i>					
Uranium	1950s–1985	dpm	yes	no	no
	Prior 1985				
	5/85–1995				
<i>U Daily Excretion = Database dpm × $\frac{1400 \text{ mL}}{\text{Aliquot mL}}$</i>					
Am-241 Cm-244	1950–1989	pCi	no	no	no
	Prior 7/89				
	7/89–1995				
<i>Daily Excretion Am-241 or Cm-244 = Database pCi</i>					

a. Detector efficiencies of 50% should be used to correct any data in cpm or cph.

b. Assume that intake of Ra-223 and Ra-224 were in equilibrium with Ac-227 and Th-228, respectively.

c. Po-210 database values must be normalized for existing 86% extraction and then corrected to 10% and 63%.

5.9.11 Summary of Analytical Methods and Detection Limits

This section presents information required to ensure proper evaluation of bioassay data as well as a means to assign missed dose. Table 5-25 summarizes radionuclide analytical methods and MDAs by period for urinalysis results for primary Mound radionuclides.

Table 5-25. Analytical methods and MDAs for primary radionuclides by period.

Nuclide	Method/description	Period	MDA	
H-3 (hot gas)	Vibrating reed electrometer	1957–1961	1.0 µCi/L	
	LS	1962–1995	0.01 µCi/L	
Po-210 (Postum)	Spontaneous electrodeposition from raw urine, low-background proportional counting.	1944–1963	Spot samples 6.0 ^a dpm 168 dpm/d	24-hr samples 1.3 dpm/24 hr ^a
	Spontaneous electrodeposition from acidified urine, low-background proportional counting.	1964–1973	Spot samples 0.72 ^a dpm 20 dpm/d	24-hr samples 0.15 dpm/24 hr ^a
Pu-238, Pu-239	Alkaline earth phosphate coprecipitation, Pu separation with cerium carrier gross alpha proportional counting.	Before 1957	(0.85) pCi/sample ^b	
		1957	(0.94) pCi/sample ^b	
		1958	(0.6) pCi/sample ^b	
		1959	(0.5) pCi/sample ^b	
		1960	(0.5) pCi/sample ^b	
	Above with sample acidification.	1961–1966	0.05 pCi/sample	
	Above with anion exchange, electrodeposition, gross alpha proportional counting.	1967–05/1981	0.11 pCi/sample ^c	
	Above with alpha spectroscopy for Pu-238 and Pu-239/240, Pu-242 tracer, blanks counted, DLs quantified. ^c	06–12/1981	Pu-238	Pu-239
			(pCi/sample)	
			0.066	0.044
		1982	0.063	0.042
		1983	0.064	0.043
		1984	0.066	0.045
1985		0.047	0.031	
1986		0.042	0.028	
1987	0.022	0.011		
1988	0.017	0.01		
01–07/1989	0.019	0.008		
08/1989–12/1991	0.018	0.008		

- Polonium-210 spot and 24-hour samples were first normalized assuming 85% extraction efficiency, then corrected for a 10% chemical recovery from 1944-1963. A chemical extraction efficiency of 64% was assumed for 1964-1973.
- Plutonium DL's from the period 1957-1960 were normalized assuming 85% extraction efficiency, then corrected for a 10% chemical recovery during electroplating.
- Plutonium MDA's from MCP (2004).

5.9.12 Use of Co-Worker Data for Internal Dose Reconstruction

For the most part, employees potentially exposed to intakes of radioactive material were monitored during their employment at the Mound site. Co-worker data are available to assign doses from assumed exposures to primary radionuclides at the Mound site, should case-specific information suggest an unmonitored exposure to potential intakes (ORAUT 2012a).

5.10 ATTRIBUTIONS AND ANNOTATIONS

All information requiring identification was addressed via references integrated into the reference section of this document.

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GLOSSARY

absorption type

Categories for materials according to their rate of absorption from the respiratory tract to the blood, which replaced the earlier inhalation clearance classes. Defined by the International Commission on Radiological Protection, the absorption types are F: deposited materials that are readily absorbed into blood from the respiratory tract (fast solubilization), M: deposited materials that have intermediate rates of absorption into blood from the respiratory tract (moderate rate of solubilization), and S: deposited materials that are relatively in the respiratory tract (slow solubilization). Also called solubility type.

alpha radiation

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

beta radiation

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

contamination

Radioactive material in an undesired location including air, soil, buildings, animals, and persons.

curie

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

dosimetry

Measurement and calculation of internal and external radiation doses.

fission

Splitting of the nucleus of an atom (usually of a heavy element) into at least two other nuclei and the release of a relatively large amount of energy. This transformation usually releases two or three neutrons.

gamma radiation

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

in vitro bioassay

Measurements to determine the presence of or to estimate the amount of radioactive material in the excreta or in other biological materials removed from the body.

in vivo bioassay

Measurements of radioactive material in the human body utilizing instrumentation that detects radiation emitted from the radioactive material in the body.

ionizing radiation

Radiation of high enough energy to remove an electron from a struck atom and leave behind a positively charged ion. High enough doses of ionizing radiation can cause cellular damage. Ionizing particles include alpha particles, beta particles, gamma rays, X-rays, neutrons, high-speed electrons, high-speed protons, photoelectrons, Compton electrons, positron/negatron pairs from photon radiation, and scattered nuclei from fast neutrons.

isotope

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

nuclide

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

radiation

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

radioactivity

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

radionuclide

Radioactive nuclide. See *radioactivity* and *nuclide*.

rem

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

U.S. Atomic Energy Commission (AEC)

Federal agency created in 1946 to assume the responsibilities of the Manhattan Engineer District (nuclear weapons) and to manage the development, use, and control of nuclear energy for military and civilian applications. The U.S. Energy Research and Development Administration and the U.S. Nuclear Regulatory Commission assumed separate duties from the AEC in 1974. The U.S. Department of Energy succeeded the U.S. Energy Research and Development Administration in 1979.

ATTACHMENT A CODING INSTRUCTIONS FOR URINALYSIS RECORDS

The following coding instructions are summarized for urinalysis records that were reported on Urinalysis Cards, Weekly Reports, and 24-hour reports at Mound.

Table A-1. Coding instructions for urinalysis records.

Code	Description
All dates	MM/DD/YY or MM/DD/YYYY
Am	Am-241 results
CC	Container believed to be contaminated
Cm	Cm-244 results
CSU	Contamination source unknown
DBS	Destroyed by survey
D/M	dpm
DM-24	dpm/24 hours
H	Highest result
H-24	Highest 24-hour sample result
HS	Home sample
I, Io	Ionium (Th-230) results
INS	Insufficient
LATE	Arrived too late to process
LOST	Lost in processing
NO	No sample
NS	New sample
OS	Old sample
R	Radium fraction results
R-24	Recount 24-hour sample
S&W	Soap and water
SPIL	Spills
T	Natural thorium results
U	Uranium results
V/I	Vacation, illness, out of town
24 HR	24-hour sample
1AS	First aid sample
?	? After cpm entry

**ATTACHMENT B
UNCERTAINTY ASSESSMENT**
Page 1 of 4

TABLE OF CONTENTS

<u>SECTION</u>	<u>PAGE</u>
B.1 Contamination of Samples	57
B.2 Assignment of Gross Alpha Results as Plutonium.....	57
B.3 Missing Bioassay Records	57
B.4 Assumption of Equilibrium.....	58
B.5 Particle Size Distributions.....	58
B.6 Solubility and Lung Clearance Type.....	58
B.7 Radionuclide Use by Location.....	59
B.8 Analytical Procedures	59

Uncertainties in estimating worker dose commitments from bioassay data result from uncertainty in monitoring measurements and assessment of intake (ICRP 1998). Uncertainties for Mound bioassay measurements were not stated in the records.

B.1 CONTAMINATION OF SAMPLES

Cross contamination of samples was not specifically discussed in the historical information, and verifiable quality control laboratory procedures were implemented at Mound. This possible source is therefore considered a minor contributor to overall uncertainty.

B.2 ASSIGNMENT OF GROSS ALPHA RESULTS AS PLUTONIUM

Before alpha spectrometry, gross alpha activity based on chemical extractions was administratively assigned to ²³⁸Pu or ²³⁹Pu based on the employee workplace. If the gross alpha result for bioassay urine samples was due to any other alpha emitter, then worker dose could have been inaccurately estimated and improperly allocated among the organs.

B.3 MISSING BIOASSAY RECORDS

The record of urinalysis data is generally complete for the Mound site. In addition, some data are available for the Monsanto Chemical Company Dayton Laboratory facilities before the beginning of operations at the Mound Laboratory. Reportable worker exposures in the early years could also have been undetected due to low recovery that was caused by plating on sample containers before sample analysis. Intakes for missing bioassay results should be estimated using the MDAs in Table 5-9 for spot and 24-hour samples.

Tritium bioassay data were not reported from 1948 to 1956. Dose reconstructors have been advised to use a value of 1,000 mrem corrected to 340 mrem ICRP Publication 68 dosimetry (ICRP 1995) for unmonitored workers. This value was assumed to be the approximate upper 95th-percentile dose

ATTACHMENT B UNCERTAINTY ASSESSMENT

Page 2 of 4

estimate and considered favorable to claimants for all categories of radiation workers. Uncertainty of extrapolating doses from the 1959- to-1962 period to the 1947-to-1956 period are considered minor and compensated by use of the upper 95th-percentile dose estimate. Doses should not be assigned to administrative or other nonradiological workers.

B.4 ASSUMPTION OF EQUILIBRIUM

Dose uncertainty in relation to degree of equilibrium is potentially important when assigning dose from bioassay for secondary radionuclides. For example, there are indications that ^{230}Th doses were assigned based on the presumption of equilibrium of ^{230}Th with ^{226}Ra in some instances. This could result in errors of up to a factor of 10 when acute intakes of unspecified feed materials such as raffinate, tailings, and various production residues occurred. For instance, raffinate from sulfuric acid leaching of ore would have a relatively low concentration of radium in comparison with thorium due to radium sulfate precipitation. Therefore, the inferred ^{230}Th intake could be greatly underestimated. The assumption of equilibrium is therefore largely dependent on the true nature of feed materials to which workers were exposed and the actual degree of observed equilibrium. In many instances, the origin and radiological and chemical composition of feed materials was unknown.

Errors associated with the assumption of equilibrium between ^{228}Th and ^{224}Ra , and between ^{227}Ac and ^{223}Ra , are less than the ^{230}Th case because (1) the ^{238}U decay chain and possible cases of disequilibria are not involved, and (2) both ^{223}Ra and ^{224}Ra have short half-lives in days and therefore quickly achieve equilibrium with their respective parent radionuclides.

B.5 PARTICLE SIZE DISTRIBUTIONS

Microsphere processing, plutonium metal production, and plasma torch operations at Mound all produced particle distributions of plutonium that potentially included submicrometer sizes dependent on the specific processes. These processes were used in buildings, and reconstruction necessitates deviation from the default ICRP Publications 66 and 68 assumption of an AMAD of 5 μm for occupational exposure (ICRP 1994b, 1995). Alpha emitters with the larger 5- μm particles and lung clearance types S or M tend to have smaller inhalation effective dose coefficients than do smaller 1- μm particles. Use of a smaller AMAD of 1 μm increases worker dose after inhalation of type M ^{239}Pu by a factor of 1.5.

Of the secondary radionuclides, the difference in DCFs for 1- μm versus 5- μm particles with clearance types S or M was greatest for ^{231}Pa by a factor of 2. The opposite trend was seen for type F alpha emitters, where 5- μm particles of uranium or ^{227}Ac have effective inhalation dose coefficients that are 20% larger than corresponding dose coefficients for 1- μm particles. Therefore, uncertainty in dose coefficients in relation to particle size distribution is dependent on lung clearance type and is comparatively small based on data in ICRP Publication 68 (ICRP 1995). Uncertainties in dose to a specific organ would likely be somewhat greater than the uncertainty in the effective dose.

B.6 SOLUBILITY AND LUNG CLEARANCE TYPE

Uncertainties in ICRP Publication 68 dose coefficients due to lung clearance kinetics are much greater than uncertainties that relate to particle size (ICRP 1995). Solubility of a radionuclide is one of the most important parameters that affects internal dose (NIOSH 2002). Lung clearance types should therefore be assigned on a case-by-case basis if there is sufficient bioassay data for radionuclides to surmise lung clearance behavior for acute exposures. Assigning a radionuclide to type S when it is

ATTACHMENT B UNCERTAINTY ASSESSMENT

Page 3 of 4

actually M, or the opposite, can lead to a potential factor of 3 error in the dose coefficient. Assigning a radionuclide to type F when it is actually M, or the reverse, can lead to similar error.

B.7 RADIONUCLIDE USE BY LOCATION

The uncertainty associated with radionuclide use by location was almost entirely documented by EG&G (1995) and was considered minor in comparison with other sources of uncertainty. Radionuclide use by location has been sufficiently documented for primary radionuclides (Table 5-1). There is little uncertainty associated with documentation for tritium, ^{210}Po , and the plutonium isotopes and their processes by building. Protactinium-231 had limited use, and there is therefore less documentation judged to be sufficient to evaluate use by location and potential worker exposures.

Secondary radionuclide documentation was generally less extensive, and some building production and research processes are unknown or poorly described. Isotopic uranium data were limited in comparison with those for NU. Isotopic radium and thorium data were also limited. Almost no information was available for ^{244}Cm and ^{237}Np other than as a contaminant in the Savannah River Plant source material.

Information on the location and use of radioactive gases was primarily only available for ^{222}Rn (radon) from 1979 to the present. Very limited information was available for ^{220}Rn (thoron) and ^{219}Rn (actinon), and the measurements were questionable due to excessive decay times before analysis. However, measurements in the underground tunnel in 1979 confirmed the presence of ^{222}Rn at 88,000 pCi/L, ^{220}Rn at 28,000 pCi/L, and ^{219}Rn at 640,000 pCi/L. These concentrations clearly indicate the potential for indoor worker exposure to these inert radioactive gases.

The lack of data for the presence of thoron and actinon inside buildings at Mound could cause a large degree of uncertainty in actual worker exposures. Radon, thoron, and actinon in particular could have caused substantial worker exposure inside buildings before implementation of proper ventilation controls in 1980. Lung dose commitments from actinon could have been significant due to a very short actinon half-life of less than 4.0 seconds and direct deposition of actinon progeny on respiratory tract tissues.

Uncertainty from lack of radioactive gas information could underestimate worker doses by a factor of at least 10. However, use of an assumed GSD of 3 compensates for this uncertainty.

B.8 ANALYTICAL PROCEDURES

Uncertainties associated with analytical procedures Mound used throughout the history of bioassay measurements are significant. Early analytical procedures resulted in large errors and MDAs in relation to required regulatory dose limits. Reductions in regulated body burdens for ^{210}Po required modifications to improve analytical sensitivities. Positive bioassay detections would therefore correspond to significant worker dose commitments at the lower end of detection in urine for some radionuclides.

Systematic errors occurred in analytical procedures in the early years due primarily to ineffective plating techniques and low recoveries. Low recoveries were due largely to colloidal plating of metabolized $^{238,239}\text{Pu}$ and ^{210}Po from raw urine onto sample containers. Polonium-210 recovery in the early years before sample acidification could have resulted in an order of magnitude correction for

ATTACHMENT B
UNCERTAINTY ASSESSMENT

Page 4 of 4

²¹⁰Po urinalysis results. Use of an alkaline earth phosphate coprecipitation and separation of plutonium with a cerium carrier appeared effective throughout the history of bioassay analyses.

Additional errors occurred in the early years as a result of the inability to accurately assign gross alpha activity determined with low-background proportional counting to specific alpha-emitting radionuclides. Gross alpha activity was therefore assigned to particular radionuclides based on known work locations. This situation was resolved at Mound and within the DOE complex with the advent of alpha spectrometry and anion exchange capabilities. Accuracy in early years was also affected by the lack of tracers to determine recoveries. In addition, historical documents indicate that blanks were not counted before 1978, which precluded rigorous quantification of DLs.