**ORAU TEAM**
**Dose Reconstruction**
**Project for NIOSH**

Oak Ridge Associated Universities | Dade Moeller | MJW Technical Services

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☐ New ☒ Total Rewrite ☐ Revision ☐ Page Change

## PUBLICATION RECORD

| 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 | 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:<br>Signature on File 03/04/2009<br>Donald N. Stewart, Document Owner<br>Signature on File 03/04/2009<br>John M. Byrne, Task 3 Manager<br>Signature on File 03/04/2009<br>Edward F. Maher, Task 5 Manager<br>Signature on File 03/11/2009<br>Kate Kimpan, Project Director<br>Signature on File 03/13/2009<br>James W. Neton, Associate Director for Science |
| 12/13/2010 | 01 | Revised to incorporate SEC-00090 and SEC-00171. Revised dose reconstruction guidance for Ra-Ac-Th in Section 5.3.2.4, radon dose reconstruction guidance for Building SW-19 in Section 5.3.2.1, and updated Table 5-19. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Donald N. Stewart. |
| 04/18/2013 | 02 | Revised to incorporate SEC-00207, as well as comments made on the original version by Sanford Cohen & Associates in 2006. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: as determined by the Objective Manager. Initiated by Donald N. Stewart. |
| 03/01/2017 | 03 | Revision initiated to incorporate updated guidance in DCAS-RPRT-005, Rev 00, *Alternative Dissolution Models for Insoluble Pu-238*, on highly insoluble $^{238}$Pu compounds. Minimum detectable amounts updated to reflect the latest available values for the period before Mound closure in 2006. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Donald N. Stewart. |

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## ACRONYMS AND ABBREVIATIONS

| | |
|---|---|
| AEC | U.S. Atomic Energy Commission |
| AMAD | activity median aerodynamic diameter |
| ANPP | Aircraft Nuclear Propulsion Project |
| AWE | atomic weapons employer |
| Bq | becquerel |
| BZ | breathing zone |
| CFR | Code of Federal Regulations |
| Ci | curie |
| cm | centimeter |
| cph | counts per hour |
| cpm | counts per minute |
| d | day |
| DCF | dose conversion factor |
| DL | detection limit |
| DOE | U.S. Department of Energy |
| DOL | U.S. Department of Labor |
| dpm | disintegrations per minute |
| DU | depleted uranium |
| EDE | effective dose equivalent |
| EEOICPA | Energy Employees Occupational Illness Compensation Program Act of 2000 |
| EU | enriched uranium |
| F | fast (absorption type) |
| g | gram |
| gal | gallon |
| GSD | geometric standard deviation |
| hr | hour |
| HTO | tritiated water vapor |
| ICRP | International Commission on Radiological Protection |
| IMBA | Integrated Modules for Bioassay Analysis |
| in. | inch |
| kg | kilogram |
| L | liter |
| LANL | Los Alamos National Laboratory |
| LSC | liquid scintillation counting |
| M | moderate (absorption type) |
| mCi | millicurie |
| MDA | minimum detectable activity |
| min | minute |
| mg | milligram |
| 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 |
| PuCd | plutonium-cadmium (alloy) |
| PuCu | plutonium-curium (alloy) |
| PuFe | plutonium-iron (alloy) |
| PuNi | plutonium-nickel (alloy) |
| PuVa | plutonium-vanadium (alloy) |
| PuYt | plutonium-yttrium (alloy) |
| QF | quality factor |
| RTG | radioisotopic thermoelectric generator |
| S | slow (absorption type) |
| SEC | Special Exposure Cohort |
| SMT | stable metal tritide |
| SRDB Ref ID | Site Research Database Reference Identification (number) |
| SRS | Savannah River Site |
| SS | super slow (absorption type) |
| TBD | technical basis document |
| 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 Department of Energy (DOE) or Atomic Weapons Employer (AWE) facilities or categories of DOE or AWE facilities. They will be revised in the event additional relevant information is obtained about the affected DOE or AWE facility(ies). These documents may be used to assist NIOSH staff in the evaluation of Special Exposure Cohort (SEC) petitions and the completion of the individual work required for each dose reconstruction.

In this document the word “facility” is used to refer to an area, building, or group of buildings that served a specific purpose at a DOE or AWE facility. It does not mean nor should it be equated to an “AWE facility” or a “DOE facility.” The terms AWE and DOE facility are defined in sections 7384l(5) and (12) of the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA), respectively. An AWE facility means “a facility, owned by an atomic weapons employer, that is or was used to process or produce, for use by the United States, material that emitted radiation and was used in the production of an atomic weapon, excluding uranium mining or milling.” 42 U.S.C. § 7384l(5). On the other hand, a DOE facility is defined 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 [DOE] (except for buildings, structures, premises, grounds, or operations … pertaining to the Naval Nuclear Propulsion Program);” and with regard to which DOE has or had a proprietary interest, or “entered into a contract with an entity to provide management and operation, management and integration, environmental remediation services, construction, or maintenance services.” 42 U.S.C. § 7384l(12). The Department of Energy (DOE) determines whether a site meets the statutory definition of an AWE facility and the Department of Labor (DOL) determines if a site is a DOE facility and, if it is, designates it as such.

Accordingly, a Part B claim for benefits must be based on an energy employee’s eligible employment and occupational radiation exposure at a DOE or AWE facility during the facility’s designated time period and location (i.e., covered employee). After DOL determines that a claim meets the eligibility requirements under EEOICPA, DOL transmits the claim to NIOSH for a dose reconstruction. EEOICPA provides, among other things, guidance on eligible employment and the types of radiation exposure to be included in an individual dose reconstruction. Under EEOICPA, eligible employment at a DOE facility includes individuals who are or were employed by DOE and its predecessor agencies, as well as their contractors and subcontractors at the facility. Unlike the abovementioned statutory provisions on DOE facility definitions that contain specific descriptions or exclusions on facility designation, the statutory provision governing types of exposure to be included in dose reconstructions for DOE covered employees only requires that such exposures be incurred in the performance of duty. As such, NIOSH broadly construes radiation exposures incurred in the performance of duty to include all radiation exposures received as a condition of employment at covered DOE facilities in its dose reconstructions for covered employees. For covered employees at DOE facilities, individual dose reconstructions may also include radiation exposures related to the Naval Nuclear Propulsion Program at DOE facilities, if applicable. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction.

NIOSH does not consider the following types of exposure as those incurred in the performance of duty as a condition of employment at a DOE facility. Therefore these exposures are not included in dose reconstructions for covered employees (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

### 5.1.1 Purpose

This technical basis document (TBD) contains an analysis of the internal doses Mound Laboratory workers received from the period of the initial external dose monitoring on February 21, 1949, through the end of operations at 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 in the 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 Special Exposure Cohort

**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* (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* (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* (Sebelius 2012).

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, partial dose reconstructions for employees who worked in any Mound area during the above 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 employees who worked in SW (Semi-Works) or R (Research) 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 class, may be performed using these data as appropriate.

To summarize the SEC classes:

- 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 for October 1, 1949, through February 28, 1959;

- The second class was added because it is not possible to bound radon doses to workers in SW and R Buildings from March 1, 1959, through March 5, 1980. These workers are identified by at least one bioassay sample for tritium in their internal dose records; and

- The third class was added because tritium records for some periods could not be found. This includes all employees in all locations from September 1 through December 31, 1972, and from January 1, 1975, through December 31, 1976.

For all of these classes of workers, internal doses are to be reconstructed using case-specific data, if available, when those doses 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 extent of potential 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 (Section 5.2):

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

Secondary radionuclides of concern include (Section 5.3):

- Radium ($^{226}$Ra) and actinium ($^{227}$Ac);
- Thorium ($^{230}$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)
- Radium ($^{226}$Ra, $^{224}$Ra, and $^{223}$Ra); and
- Radon ($^{222}$Rn, $^{219}$Rn, and $^{220}$Rn).

## 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 concurrent 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 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 R Building involved research and electrodeposition studies from 1948 to 1951 and manufacture of Po-Be neutron source from 1956 to 1961.

SW Building was used for counting $^{210}$Po sources, neutron source measurement, source strength verification, and $^{209,210}$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 WD (Waste Disposal) and WDA Buildings from 1949 to the present. Chemical forms included polonium nitrate and polonium chloride. There was no cited $^{210}$Po activity in PP (Plutonium Processing) or SM (Special Metallurgical) Buildings. One $^{210}$Po incident was reported in SW Building in 1961, and 11 incidents are associated with T Building where primary polonium separation and processing took place from 1954 to 1968.

The second phase of the polonium project consisted of manufacturing polonium heat sources for radioisotopic thermoelectric generators (RTGs). The Systems for Nuclear Auxiliary Power-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 $^{239}$Pu and $^{238}$Pu for research projects were transferred from the Lawrence Livermore National Laboratory to Mound in 1959. As polonium production tapered 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 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 consisted primarily of $^{238}$Pu, which is typically called “heat-source” plutonium in Mound historical documents and in this TBD. Tables 5-10 and 5-11 later in this document list the radionuclides and proportions of these mixtures.

#### 5.2.2.1 Plutonium-238

Feed material from the Savannah River Site (SRS) as $PuO_2$ powder was enriched in $^{238}$Pu and processed at Mound to manufacture heat sources. Early $^{238}$Pu activities took place primarily in SM Building. PP Building was built to supplement and replace the $^{238}$Pu processing areas in SM Building.

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 $PuO_2$ particles in the submicrometer to 20-μm range (EG&G 1995).

The microspheres would have been quite stable, and their initial particle size distribution would only have been affected by depositional processes that removed particles above 10 μm from worker breathing zones (BZs). Deposition would therefore have tended 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 $PuO_2$ 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 $^{238}$Pu exposure incidents were reported in 1986.

All SM processes were transferred to PP Building in December 1967. The primary mission in this building was to process $PuO_2$ powder from the SRS. A number of processes were developed to manufacture plutonium microspheres in PP Building and involved the following:

- PuMo cermet process to coat plutonium particles from 43 to 250 μm with molybdenum from 1969 to 1973,
- Pressed $PuO_2$ process to fabricate plutonium spheres for encapsulation as heat sources from 1970 to 1979,
- Milliwatt Generator Program process to seal 105- to 500-μm plutonium shards in yttrium,
- Solid solution process from 1969 to 1974,
- Solution gel process from 1969 to 1971, and
- Microsphere process to produce 50- to 250-μm $^{238}PuO_2$ ceramic spheres from 1967 to 1970.

The Mound site supplied RTGs for space exploration missions including Pioneer, Voyager, and Apollo. In 1977, production of the $PuO_2$ fuel itself was moved to the SRS, 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 $^{238}$Pu program, $^{234}$U was separated from aged $^{238}$Pu from 1970 to 1980. Table 5-10 later in this document lists the heat source plutonium isotopic mixture (EG&G (1995).

### 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 R Building using $^{239}$Pu in 1956. To date, NIOSH has not determined the exact mixture of plutonium that made up the weapons-grade material, although one source describes it as "more than 95% pure $^{239}$Pu with trace amounts of $^{240}$Pu and $^{241}$Pu" (EG&G 1995). For dose reconstruction, the mixture that results in the greatest ingrowth of $^{241}$Am was selected as favorable to claimants in lieu of specific data. Table 5-11 lists this mixture based on Hanford plutonium (BMI 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” [sic[1]] (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, this document contains no provisions 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 PP Building and 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 (EG&G 1995).

## 5.3 PROCESS OPERATIONS INVOLVING SECONDARY RADIONUCLIDES

### 5.3.1 Radium 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:

$$^{226}_{88}\text{Ra} + \eta \rightarrow {}^{227}_{88}\text{Ra} \rightarrow {}^{227}_{89}\text{Ac} + \beta^-$$

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 resulted 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 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 from the feed material and the process itself. Bioassay was performed for the involved individuals, who are identified in Mound records (Meyer 1992), but the 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

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

potential 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 Thorium-232

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 activities with $^{232}$Th began with a preliminary proposal for a thorium refinery 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-gal 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 by AEC as early as April 1955 (McCarthy 1955b), and the Mound refinery project was cancelled in May 1955 (McCarthy, 1955c).

This left Mound with the task of disposing of the thorium residues. On arrival, 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 ($2.23 \times 10^5$ kg) occupied a smaller bay that occupied about 25% of the storage basin, and the hydroxide sludges ($1.2 \times 10^6$ 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 Thorium-230

A limited ionium ($^{230}$Th) 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 came from Mallinckrodt Chemical Works and the Cotter Corporation.

### 5.3.3 Protactinium-231

An AEC program was undertaken in 1955 in 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 HH Building (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 ended 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 and WDA Buildings, which received aqueous wastes from R and SW Buildings. No $^{231}$Pa use was reported for SM, SW, T, or PP Buildings.

Extraction and purification of $^{231}$Pa 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 10 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 1952, 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 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 PuFe, PuNi, PuCu, PuCd, PuYt, and PuVa.
- Plutonium-bearing glass fibers. In 1961 Mound participated in a project to experiment with fabrication of these 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 gal 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), but 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 who worked with americium in research projects. This is due to the possibility that it posed a specific intake risk from $^{231}$Pa separation, the reactor fuel research program, and alpha source production, neutron source production, analysis, and vaporization of plutonium compounds.

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 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 separation began in the mid-1950s and continued until 1985 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 dose reconstruction, external dose is the exposure of concern; 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 SRS. They were also present in aqueous wastes 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). Therefore, the potential to induce relatively high doses via the inhalation pathway should be taken into account by dose reconstructors. 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 very few buildings for limited periods, in relatively minor quantities in comparison with the primary and secondary radionuclides. Most of these radionuclides were in sealed sources. Worker exposure was less likely, and bioassay data was not collected for these radionuclides. Exposure to these radionuclides is therefore 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, which deposit 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, SW Building had high levels of radon from the decay of radioactive materials from 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 $^{222}$Rn, actinon ($^{219}$Rn), and thoron ($^{220}$Rn), and an attempt was made to measure the concentration. 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 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 Sebelius (2010) and Sebelius (2012).

Thorium sludge from $^{232}$Th 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 $^{231}$Pa and $^{230}$Th. Table 5-1 shows the results of various radon surveys at

Mound. For comparison, the reported background concentration of $^{222}$Rn ranged from 0.1 to 2.1 pCi/L with an average of 0.5 pCi/L.

Table 5-1. Radon isotope survey data gas concentrations and exposure levels by building and period.

| Building | Survey dates | Rn-222 (pCi/L) | Thoron (Rn-220) (pCi/L) | Actinon (Rn-219) (pCi/L) | WL | WLM/yr[a] |
|---|---|---|---|---|---|---|
| SW Tunnel[b] | 10/12/1979 | 88,000 | 28,000 | 640,000 | Not applicable | Not applicable |
| SW19 after vent | 03/1980–04/1980 | 7.7–13.4 | No data | No data | 0.03 | 0.4 |
| SW19 after vent | 12/1989 | 2.6 | Not detected | No data | No data | No data |
| SW19 after vent | 12/1999 | 1.0–5[c] | No data | No data | No data | No data |
| SW19 after vent | 04/1990 | 0.7–7.2[c] | No data | No data | No data | No data |
| SW19 after vent | 1998–2000 | 5.8–12 | No data | No data | No data | No data |
| Building 21 | 05/1983 | 17.5–52.8 | No data | No data | 0.4–1.3<br>0.85 median | 4.8–15.6<br>10.0 median |
| Building 21 | 12/1989 | 34–118[c] | No data | No data | No data | No data |
| Building 21 | 04/1990–06/1990 | 117–161 | No data | 0.3–0.7 | No data | No data |
| | | 125[c] | No data | No data | No data | No data |
| Building 48 | 12/1989 | 3.2 | No data | No data | No data | No data |
| Old SD | 12/1989 | 1.5–2.4 | No data | No data | No data | No data |
| Fire station | 12/1989 | 1.2–1.4 | No data | No data | No data | No data |
| Paint shop | 12/1989 | 0.9–1.2 | No data | No data | No data | No data |
| A | 10/1988–02/1989 | 0.1–1.1[c] | No data | No data | No data | No data |
| All other buildings | 12/1989 | 0.4–0.9 | No data | No data | No data | No data |
| Outside facilities | 05/1983 | 0.24–0.59 | No data | No data | No data | No data |
| Reported background | Not applicable | 0.1 to 2.1<br>Average = 0.5 | Not applicable | Not applicable | Not applicable | Not applicable |

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

Areas in SW Building had high concentrations of radon, thoron, and actinon 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 divided into and renamed Rooms 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 $^{222}$Rn 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 (Monsanto 1983a, 1983b, ca. 1983). 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 $^{222}$Rn, 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. SW Building, specifically Room SW-19, was identified as the only area of potential occupational exposure to $^{222}$Rn and $^{220}$Rn.

Thoron and actinon measurement results were very limited despite the high concentrations 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 $^{219}$Rn (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 are considered to be secondary information. Whole-body counting is not an efficient monitoring method for Mound's primary radionuclides ($^{210}$Po, $^{238}$Pu, and $^{3}$H) or for many secondary radionuclides such as $^{239}$Pu, $^{231}$Pa, those of radium, and those of actinium. 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.

**In vitro monitoring**

| Bioassay program | Period | Action levels[a] | Monitored workers | Frequency |
|---|---|---|---|---|
| Po-210 processing | 1945–1974 | 10 cpm/50 mL | Operations personnel | Weekly spots,<br>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,<br>Weekly 24 hr |
| Pu-239 neutron source | 1957–present | 3.5 cpm/24 hr | Processing personnel | Weekly spots,<br>Weekly 24 hr |
| Pu-238 heat source | 1960–present | 3.5 cpm/24 hr | Processing personnel | Weekly spots,<br>Weekly 24 hr |
| Am-241 | 07/1989–present | 0.023 pCi/sample[b] | Potentially exposed personnel | Daily |
| Cm-244 | 07/1989–present | 0.009 pCi/sample[b] | Potentially exposed personnel | Daily |

**In vivo monitoring**

| Bioassay program | Period | Action levels[a] | Monitored workers | Frequency |
|---|---|---|---|---|
| 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 before this date, dose reconstructors should base MDA assumptions on actual data or refer to the "Table of Other Radionuclides" (Author unknown 1955–1988).
c. Later 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 exposed. Monitoring frequencies were described by Meyer (1992) and inferred from personnel sampling records. For programs that used spot sampling, such as $^{210}$Po and $^{238}$Pu, individuals who were 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 5-2 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 National Bureau of Standards Handbook 52 (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 methods.

**Primary radionuclides of concern**

| Nuclide | Method/description | Period and MDA | |
|---|---|---|---|
| H-3 (hot gas) | Vibrating reed electrometer | 1957–1961, | 1.0 µCi/L |
| H-3 (hot gas) | Liquid scintillation counting (LSC) | 1962–1995, | 0.01 µCi/L |
| Po-210 (postum) | Spontaneous electrodeposition from raw urine, low-background proportional counting | 1944–1963, | Spot samples,6.0 dpm,[a]<br>168 dpm/d<br>24-hr samples, 1.3 dpm/24 hr[a] |
| Po-210 | Spontaneous electrodeposition from acidified urine, low-background proportional counting. | 1964–1973, | Spot samples, 0.72 dpm,[a]<br>20 dpm/d,<br>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,<br>1957,<br>1958,<br>1959–1960, | (0.85) pCi/sample,[b]<br>(0.94) pCi/sample,[b]<br>(0.6) pCi/sample,[b]<br>(0.5) pCi/sample,[b] |
| Pu-238, Pu-239 | Above with sample acidification | 1961–1966, | 0.05 pCi/sample |
| Pu-238, Pu-239 | Above with anion exchange, electrodeposition, gross alpha proportional counting | 1967–05/1981, | 0.11 pCi/sample[c] |

| Nuclide | Method/description | Period and MDA | |
|---|---|---|---|
| Pu-238,<br>Pu-239 | Above with alpha spectroscopy for Pu-238 and Pu-239/240, Pu-242 tracer, blanks counted, detection limits (MDAs) quantified | 06–12/1981, | 0.066 pCi/sample Pu-238,<br>0.044 pCi/sample Pu-239, |
| | | 1982, | 0.063 pCi/sample Pu-238,<br>0.042 pCi/sample Pu-239, |
| | | 1983, | 0.064 pCi/sample Pu-238,<br>0.043 pCi/sample Pu-239, |
| | | 1984, | 0.066 pCi/sample Pu-238,<br>0.045 pCi/sample Pu-239, |
| | | 1985, | 0.047 pCi/sample Pu-238,<br>0.031 pCi/sample Pu-239, |
| | | 1986, | 0.042 pCi/sample Pu-238,<br>0.028 pCi/sample Pu-239, |
| | | 1987, | 0.022 pCi/sample Pu-238,<br>0.011 pCi/sample Pu-239, |
| | | 1988, | 0.017 pCi/sample Pu-238,<br>0.01 pCi/sample Pu-239, |
| | | 01–07/1989, | 0.019 pCi/sample Pu-238,<br>0.008 pCi/sample Pu-239, |
| | | 08/1989–12/1991, | 0.018 pCi/sample Pu-238,<br>0.008 pCi/sample Pu-239, |
| | | 1992–present, | 0.018 pCi/sample Pu-238,<br>0.009 pCi/sample Pu-239 |

**Secondary radionuclides of concern**

| Nuclide | Method/description | Period and MDA | |
|---|---|---|---|
| Pa-231 | Radium extraction and activity inferred from Th-227 or Ra-223 dpm assuming secular equilibrium | 08/17/1955–1956,<br>1957–1958,<br>1959–12/07/1959 | 0.3 dpm,<br>0.02 dpm,<br>0.08 dpm |
| Th-230 (ionium) | Radium separation, intakes and excreted Ra-226 and Th-230 assumed to be in equilibrium. | Before 01/1994, | 0.3 dpm |
| Th-230 (ionium) | Th-230 extraction, alpha spectroscopy. | 01/1994–1995, | 0.17 dpm |
| Th-natural | Thorium separation, proportional counting. | Before 12/1987,<br>12/1987–1994, | 0.3 dpm,<br>0.02 dpm Th-232, |
| Th-natural | Thorium separation, alpha spectroscopy. | 01/1994–1995, | 0.08 dpm Th-228 |
| Uranium | LiF fusion, fluoroscopy. | 1950s–1985, | 0.4 µg/L, |
| Uranium | Uranium separation, proportional counting. | Before 1985, | 0.3 dpm |
| Uranium | Uranium separation, alpha spectroscopy. | 05/1985–1995, | 0.07 dpm U-234,<br>0.03 dpm U-235,<br>0.09 dpm U-238 |
| Am-241,<br>Cm-244 | Americium or curium separation, proportional counting. | 1950–1989, | 0.2 pCi |
| Am-241,<br>Cm-244 | Americium or curium separation, alpha spectroscopy. | Before 07/1989,<br>07/1989–1995, | 0.3 pCi,<br>0.05 Ci |

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

In general, the MDAs in Table 5-3 should be used only when no value accompanies the result. MESH records, available starting in 1991, typically list the MDA for each result, which are to be used for results after the dates in the table. The Mound bioassay program collected samples as late as 2006.

### 5.4.3 Gross Alpha and Sequential Separation

A procedure was developed at 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 consist 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, which was 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 $^{238}$Pu, as all alpha activity could be assumed to result from $^{238}$Pu.

### 5.4.4 In Vivo Dose Monitoring

An effort to upgrade the Mound internal dose monitoring program with an in vivo counting facility began in 1964, and the “reasonably standard“ Mound Body Counting Facility was completed with a Packard Instrument Company steel room in 1969. Detectors installed at that time were two standard sodium iodide detectors, 4 inches in diameter (Tomlinson et al. 1974a). Once personnel counting began, problems were identified, and later in that year, the system became operational with a single phoswich detector (ORAUT 2007b); in December, a second phoswich detector was added, and the modified system became fully operational in January 1970 (ORAUT 2008). Another source states that the dual-phoswich counter was operational in May of 1970 (Tomlinson et al. 1974a). A switch was made to dual five-inch phoswich detectors in October of 1972 (Tomlinson et al. 1974a). During this period, in accordance with guidance in the National Bureau of Standards Handbook 80, the MDA was determined to be the 3-sigma value of the background count (Tomlinson et al. 1974b). Table 5-4 summarizes the in vivo counting systems and associated MDAs for specified time periods.

Table 5-4. In vivo Counting Systems and MDAs.

| Dates | Detector Types | MDAs |
|---|---|---|
| 1969 | Dual NaI(Tl) detectors, 10.2 cm | 11 nCi Pu-238 (claimed)[a] |
| July 1969 | Single Phoswich, 10.2 cm | Not listed |
| January or May 1970 | Dual Phoswich, 10.2 cm | 5.1 nCi Pu-238 (claimed)[a] |
| October 1972 | Dual Phoswich 12.7 cm | 4 nCi Pu-238 (claimed)[a] |
| 1990 | Dual Phoswich 12.7 cm | 69.6 to 73.7 nCi Pu-238, 0.18 nCi Am-241[b] |

a. Tomlinson et al. (1974a).
b. Tomlinson (1990).

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 (Barton, 1995, pp 325 – 327). 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.

Tomlinson et al. (1974b) discuss chest wall thickness measurements at the Mound site. Measurements were made using a Hewlett-Packard ultrasonic sounding instrument. Two methods for ultrasonic measurements were investigated, along with a chest prediction method based on various body measurements. Though the ultrasonic methods proved to be most accurate, Mound limited the measurements to counts to four to six per week to conserve time spent (Tomlinson et al. 1974b). In vivo count records are not kept in a central electronic database; each count is maintained as a 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 (Tomlinson 1990). Some records observed of those available (extant logbooks from 1969–1993) report MDAs for each count; however many do not. An example of a lung count record from 1976 shows that measurement-specific MDAs were to be reported with the counting result based on ultrasound chest-thickness measurements (Tomlinson 1976). 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.

Due to the very high MDAs, missed dose should be based on urine bioassay results for dose reconstruction. Chest-counting data would best be used to assess intake when counting was conducted as a follow-up for a known worker intake and records are available to the dose reconstructor. It is not expected, based on experience with Mound claims, that chest counts results will be commonly available. 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

Mound operations produced a complex array of particulates, aerosols, dusts, and fumes. 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 is unlikely to accurately estimate actual worker inhalation intakes in most situations

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 METHODS FOR PRIMARY RADIONUCLIDES

### 5.5.1 Polonium-210

#### 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 $HNO_3$ digestion. Polonium-210 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 1950). 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 (Spoerl 1950; Meyer 1992). Spoerl (1950) 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 (1950) 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 $^{210}$Po losses might occur during the time between sample collection and sample assay (Spoerl 1950). 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 $2\pi$.

**Chemical Recovery Correction**

A 1989 study determined that significant recovery losses from raw undigested urine were observed for metabolized $^{210}$Po due to plating or formation of precipitates (Fellman et al. 1989; Meyer 1992). This study used urine samples from primates by investigators at the New York Medical Center and by Spitz and Robinson at Mound (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 1950). 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 through 1963, after which complete acid digestion of urine samples was required (Meyer 1992).

The 10% chemical recovery correction was applied to evaluate occupational doses from $^{210}$Po in a retrospective study in 1989 (Spitz 1989a; Themelis 1989). In addition, DOE requested that Mound use the 10% chemical recovery to conduct $^{210}$Po dosimetry (Spitz 1989b). A peer review committee was established in 1987 to study polonium dose method.

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 1950). 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 (1950) of 86%.

However, the data in 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 a 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 (1950) at Mound with an average recovery of 86 ±11.9%. In addition, the Fellman et al. study is in good agreement with a previous Argonne National Laboratory study 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 should be performed using the assumptions in Section 5.8.1, which are favorable to claimants.

**Reported Half-Life**

The effective half-life of $^{210}$Po was investigated in 1950 and reported by Spoerl (1951) 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 per 24-hour sample. Weekly results were transcribed onto Polonium Cards for individual workers with entries in cpm/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 additional urine samples were required to document possible worker exposures or to confirm previous results with all entries in cpm and duplicates 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 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 Laboratory[2] facilities as well as the Mound Laboratory.[3]

Most Mound claim files contain primary and secondary records for polonium bioassay. As a general procedure, conduct dose reconstruction using the primary records contained in the claimant files. The dose reconstructor may also use the databases as convenient listings of the claimant information but

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[2] 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 listed assumptions for the period before 1964.

[3] Possible exposures to $^{210}$Po 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).

reconcile their use with the primary records. If variations from the general procedure occur, validate – to the extent practical, all information used in dose reconstruction calculations.

### 5.5.2 Plutonium

In 1952, an attempt was made to electroplate plutonium onto copper plates by following the procedure for $^{210}$Po. 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 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 $^{242}$Pu tracer, counting of blanks to quantify MDAs, and alpha spectroscopy (Phillips 1988). However, gross alpha results for bioassay were reported as plutonium for all $^{238}$Pu 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 5-5 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 $^{238}$Pu 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.

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

| Period | Reporting level[a] | Analytical method |
|---|---|---|
| 1954–1960 | Kirby and Brodbeck 1954<br>0.05 dpm/1.5<br>0.5 dpm/1.5 L (corrected MDA)[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<br>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)<br>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)<br>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)<br>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. MDA was corrected for a low chemical recovery of 10% due to colloidal plating of plutonium.

**Plutonium Internal Dose Monitoring Records**

Most Mound claim files contain primary and secondary records for plutonium bioassay. As a general procedure, conduct dose reconstruction using the primary records contained in the claimant files. The dose reconstructor may also use the databases as convenient listings of the claimant information but reconcile their use with the primary records. If variations from the general procedure occur, validate – to the extent practical, all information used in dose reconstruction calculations.

The dose reconstructor also needs to ensure that PURECON reflects earlier measurements using the gross alpha method and that the gross alpha samples were taken for 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 an SRS 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 counting (LSC) began in 1961 followed by an approved procedure in 1968. This method 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 scintillation counters used external $^{133}$Ba or $^{226}$Ra 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 5-6 lists the reported analytical procedures and MDAs for tritium.

Table 5-6. Reported analytical procedures and MDAs for tritium.

| Period | Analytical method | MDA (µCi/L) |
|---|---|---|
| 1957–1961 | Vibrating reed electrometer | 1.0 |
| 1962–2006[a] | LSC | 0.01 |

a. No conclusive data have been identified supporting tritium MDAs past 1995, so this value is assumed through the end of Mound operations.

### Tritium Dose Calculations

This section presents a method 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 necessary due to an overall correction factor of 1.0 as shown in Equation 5-3 below.

A dosimetry method from LANL was used for acute tritium dose calculations from 1957 through 1988 (Lawrence 1957). This method assumed a maximum permissible body burden of 3.7 mCi, a relative biological effectiveness (RBE) 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 µCi/L for a target dose limit of 300 mrem/wk under the LANL system (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:

$$D(t) = 0.00873B \tag{5-1}$$

where

$D(t)$ = the dose commitment at time $t$ after exposure (rem)
$B$ = measured acute body concentration in urine or body water (µCi/L)

Reported doses on Radioactive Gas Exposure Evaluation forms confirm that Mound used Equation 5-1 for tritium dose calculation and that 6 µCi/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 reported doses using Equation 5-2.

Current dosimetry for tritium uses a quality factor (QF) of 1.0 in comparison with 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 in the LANL calculations (ICRP Publications 67 and 68; ICRP 1994a, 1995). Equation 5-2 provides a ratio of Publication 68 dose in comparison with LANL dose, which results in a ratio of 0.34:

$$Ratio\left(\frac{ICRP68}{LANL}\right) = \frac{43.4}{68.8 \times 1.7} \times \frac{\int_0^\infty \left(0.97 \times e^{-0.693 \times t/10} + 0.03 \times e^{-0.693 \times t/40}\right) dt}{\int_0^\infty e^{-0.693 \times t/12} dt} = 0.34 \tag{5-2}$$

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

$$Ratio\left(\frac{ICRP68}{ICRP30}\right)=\frac{63}{68.8}\times\frac{\int_0^\infty\left(0.97\times e^{-0.693\times t/10}+0.03\times e^{-0.693\times t/40}\right)dt}{\int_0^\infty e^{-0.693\times t/10}dt}=1.0 \qquad (5\text{-}3)$$

**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 µCi/L (Meyer 1992). Reporting limits of 3 µCi/L as discussed by James (1997) or 6 µCi/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. The MDAs in Table 5-6 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-226, Actinium-227, and Thorium-228

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 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 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-230 and -232

### 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 an EDE 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 6507-1I, 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).

### 5.6.2.2 Thorium-232

Monitoring for $^{232}$Th began in 1955 to support thorium refining; preliminary research began in 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. Only one document (Author unknown undated a, p. 2) lists a DL in mass units for natural uranium (NU). 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

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 by NIOSH (2016). Guidance has been developed to estimate doses to high-fired forms of $^{238}$Pu, which can under some circumstances exhibit nonmonotonic clearance rates (NIOSH 2016). A Mound-specific alternative dissolution 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. The alternative dissolution is modeled using specific IMBA input parameters in Table 5-13 later in this document.

**Exposure Dates**
The assumptions described in ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2014), 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 5-7 lists equilibrium assumptions that are favorable to claimants to infer primary radionuclide activity in urine based on detected alpha activity.

Table 5-7. 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. |

a. IMBA does not directly calculate excretion of progeny due to intake of parent radionuclides with progeny.
b. Th-232 is also identified as natural thorium and Th-230 as ionium.
c. 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.
d. Uranium with naturally occurring mass and activity ratios.
e. 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 2014). 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 (Barton 1995, p. 4).

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 |
| U-238 | 0.28 | 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 claims for which 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 $^{210}$Po 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

$$\text{Po-210 Daily Excretion} = \frac{\text{Database cpm}}{0.50} \times \frac{0.85}{0.10} \times \frac{1{,}400\ \text{mL}}{\text{Aliquot mL}} \tag{5-4}$$

1964 onward

$$\text{Po-210 Daily Excretion} = \frac{\text{Database cpm}}{0.50} \times \frac{0.85}{0.63} \times \frac{1{,}400\ \text{mL}}{\text{Aliquot mL}} \tag{5-5}$$

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:

$$\frac{dpm}{mL} = 2 \times \frac{cpm}{mL} \tag{5-6}$$

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 Po-210.

| Period | Default sample volume (mL) | Detector efficiency (%) | Recovery efficiency (%) | Reported background (cpm/24 hr) | Spot sample MDA (dpm/50 mL) | 24-hr sample MDA (dpm/24 hr) |
|---|---|---|---|---|---|---|
| 1944–1963 | 50 | 50 | 10 | 0.2 | 6.0 dpm,<br>(168 dpm/d) | 1.3 |
| 1964–1973 | 100 | 50 | 63 | 0.1 | 0.72 dpm,<br>(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 $^{238}$Pu, and that the PURECON database results are to 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 $^{238}$Pu and $^{239}$Pu 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 percent composition and percent activity by effective age.[a,b]

| Nuclide | % mass | % activity at 1.0 yr (pCi/g) | % activity at 3.2 yr (pCi/g) | % activity at 10 yr (pCi/g) |
|---|---|---|---|---|
| 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 Am-241 activity is zero.

A review was performed of well-documented weapons-grade plutonium mixtures for DOE and its predecessor agencies. It was determined that the Hanford mixture was bounding with respect to the proportion of $^{241}$Am, and this mixture is to be used to estimate dose to Mound workers (BMI 2003).

Table 5-11. Weapons-grade plutonium feed material weight percent composition and percent activity by effective age.[a,b]

| Nuclide | wt% | % activity at 0 yr (fresh) (Ci/g) | % activity at 5 yr (Ci/g) | % activity at 10 yr (Ci/g) | % activity at 15 yr (Ci/g) |
|---|---|---|---|---|---|
| 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: BMI (2003).
b. Effective age is the age when Am-241 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 1950). This correction is not required for later samples. These corrections are summarized in Equations 5-7 and 5-8:

1957–1960
$$\textit{Pu-238,239 Daily Excretion} = \frac{\textit{Pu database dpm} \times 0.85}{0.10} \tag{5-7}$$

1960–2006
$$\textit{Pu-239,239 Daily Excretion} = \textit{Pu database dpm} \tag{5-8}$$

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 when the MDA is not listed with the result (MESH results, from 1991, typically list result-specific MDAs). 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, $t_c$ = 1,000 min | Pu-238, $t_c$ = 3,000 min | Pu-239/240, $t_c$ = 1,000 min | Pu-239/240, $t_c$ = 3,000 min |
|---|---|---|---|---|
| Before 1957 | (0.85)[b] | Not applicable | (0.85)[b] | Not applicable |
| 1957 | (0.94)[b] | Not applicable | (0.94)[b] | Not applicable |
| 1958 | (0.6)[b] | Not applicable | (0.6)[b] | Not applicable |
| 1959 | (0.5)[b] | Not applicable | (0.5)[b] | Not applicable |
| 1960 | (0.5)[b] | Not applicable | (0.5)[b] | Not applicable |
| 1961–1966 | 0.05 | Not applicable | 0.05 | Not applicable |
| 1967–05/1981 | 0.11 | Not applicable | 0.11 | Not applicable |
| 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 | Not applicable | 0.011 | Not applicable |
| 1988 | 0.017 | Not applicable | 0.01 | Not applicable |
| 01–07/1989 | 0.019 | Not applicable | 0.008 | Not applicable |
| 08/1989–12/1991 | 0.018 | Not applicable | 0.008 | Not applicable |
| 1992–1995 | 0.018[a] | Not applicable | 0.009[a] | Not applicable |

a. $t_c$ = count time; plutonium MDAs from before 1957 to 1960 were 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. Based on Mound (1996).

b. MDAs in parentheses were corrected for a 10% chemical recovery.

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).

### 5.8.2.1 Use of the Mound Alternate Dissolution Model for Plutonium-238

Workers who are exposed to airborne intakes of $^{238}$Pu may exhibit urinary excretion patterns indicative of a variety of dissolution rates of plutonium in the lung. Guilmette et al. (1994) described the urinary excretion resulting from exposures to $^{238}$Pu at three facilities, including the Mound site, and the patterns were observed to be very different. This is of concern from a dose reconstruction point of view, as the standard dissolution model of ICRP Publication 66 (ICRP 1994b) may, in some situations, result in underestimates of dose.

The complete dataset for Mound urine bioassay was reviewed, as described in *Alternate Dissolution Models for Insoluble Pu-238* (NIOSH 2016). The review included results from 2,082 workers. Of these, records of nine workers were shown both to exhibit non-monotonic excretion rates characterized in Guilmette et al. (1994) and to be suitable for modeling.

A model was formulated for an alternate dissolution model for Mound, and specific parameters for use in IMBA are listed in Table 5-13. Mound site dose reconstructions assigning dose from $^{238}$Pu must evaluate this alternate model, the Mound highly-insoluble (MHI) plutonium-238 model, along with the standard absorption types (NIOSH 2016).

Table 5-13. Parameters of the Mound alternate dissolution model (NIOSH 2016).

| **Parameter** | $f_r$ | $s_r$ | $s_s$ | $f_1$ |
|---|---|---|---|---|
| Value | -8.46818E-02 | 2.91697E-02 | 2.6468E-03 | 1E-08 |

The general steps to enter the alternate dissolution model into IMBA are listed below.

- Select $^{238}$Pu as indicator radionuclide.
- Select IRCP-68 Type S default parameters.
- Select “Absorption” button.
- Select “Alternative Representation” tab.
- Select “User Defined” button.
- Enter Table 5-13 values for $f_r$, $s_r$, $s_s$, and $f_1$.

### 5.8.2.2 Special Considerations for Type SS 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 1992); 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 SS 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 5-14. 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 5-14. Reported analytical procedures and MDAs for tritium (μCi/L).

| Period | Analytical method | MDA |
|---|---|---|
| 1948–1956 | No urinalysis conducted | Use 95% dose[a] |
| 1957–1961 | Vibrating reed electrometer | 1.0 |
| 1962–1995 | Liquid scintillation | 0.01 |

a. The upper 95th-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 method described in ORAUT (2007a). 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 2013). 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-226, Actinium-227

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-230 and -232

### 5.9.2.1 Thorium-230

Gross alpha counting of extracted thorium in urine started with sample 6507-1I 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:

- $^{232}$Th and $^{228}$Th 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 $^{230}$Th. If the reporting basis for an ionium bioassay result is ambiguous, assume that $^{230}$Th was extracted and counted. Excretion rates in dpm/d should be calculated using Equation 5-9 and entries from Author unknown (1955–1988) that contain information on volume, aliquot size, and dpm results, unless contradicted by another bioassay record:

$$\mathit{Daily\ Excretion\ Th\text{-}230} = \mathit{Database\ dpm} \times \frac{\mathit{Sample\ mL}}{\mathit{Aliquot\ mL}} \tag{5-9}$$

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 5-15 lists available $^{230}$Th MDAs. Where no aliquot size is given in Author unknown (1955–1988) assume the MDA is in dpm excreted per 24-hour sample.

Table 5-15. Th-230 MDAs (dpm).[a]

| Period | MDA | Reference |
|---|---|---|
| Before 01/1994 | 0.3 | MDA (MJW 2002b)[a] |
| 01/1994–present | 0.17 | MDA (Barton 1995) |

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

### 5.9.2.2 Thorium-232

Bioassay for $^{232}$Th was based on 24-hour urine samples and followed Mound bioassay procedures MLM-460 (Spoerl 1950) 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 always contain both thorium and radium results. It is assumed that $^{228}$Th, $^{232}$Th, and $^{224}$Ra are in equilibrium in feed materials, but 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 $^{228}$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 EDE 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 and detection efficiency and represents a 24-hour urine excretion. If an aliquot volume is given, 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, 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 5-16 lists $^{232}$Th MDAs by period. These MDAs have units of dpm excreted per 24-hour when no aliquot size is given. However, MDA units should be dpm per aliquot, if an aliquot volume was reported.

Table 5-16. Natural thorium MDAs (dpm).

| Period | MDA | 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 $^{232}$Th urine bioassay, the primary record sample identification number usually ends in “R” and refers to $^{224}$Ra. From these results, the daily excretion of $^{224}$Ra can be obtained using Equation 5-10:

$$\textit{Daily Excretion Ra-224} = \textit{Ra Database dpm} \div 90\% \tag{5-10}$$

A primary record identification number that ends in “T” corresponds to a natural thorium result that includes $^{232}$Th and $^{228}$Th. The daily excretion of natural thorium can be calculated using Equation 5-11:

$$\textit{Daily Excretion Th-228} + \textit{Th-232} = \textit{Th Database dpm} \div 92\% \tag{5-11}$$

Equations 5-10 and 5-11 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 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 (ICRP 1975). Dose reconstructors should assume that background natural thorium in the diet of monitored workers resulted in urinary excretion of 0.05 pCi/d (ICRP 1975).

### 5.9.3 Protactinium-231

The primary program for $^{231}$Pa was completed in 1959. For partial dose reconstructions, doses from potential intakes of $^{231}$Pa 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 5-17 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 5-17. Protactinium MDAs.[a]

| Period | Background count rate (cpm) | MDA (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 MDA if available.
b. A MDA 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 $^{231}$Pa data 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 1,400 mL and a modal aliquot volume of 120 mL:

$$\mathit{Daily\ Excretion\ Pa\text{-}231} = \mathit{Th\ Fraction\ Database\ dpm} \times \frac{\mathit{Sample\ mL}\ \text{or}\ 1{,}400\ \text{mL}}{\mathit{Aliquot\ mL}\ \text{or}\ 120\ \text{mL}} \tag{5-12}$$

### 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 only uranium, 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 $^{233}$U in absence of specific radionuclide data.
- Use $^{234}$U for EU. For NU or depleted uranium (DU), use fluorometric or gross alpha determination.
- Assume NU ratios for $^{238}$U, $^{234}$U, and $^{235}$U or use IMBA default ratios for NU and DU.

Table 5-18 lists uranium MDAs. Equation 5-13 provides correction for aliquot volume, if necessary:

Table 5-18. Uranium MDAs.

| Period | MDA | Reference |
|---|---|---|
| Before 05/1985 | 0.3 dpm | MDA (MJW 2002b)[b] |
| 05/1985–present | 0.07 dpm U-234<br>0.03 dpm U-235<br>0.09 dpm U-238 | MDA (Barton 1995) |

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

$$\mathit{U\ Daily\ Excretion} = \mathit{Database\ dpm} \times \frac{1400\ \text{mL}}{\mathit{Aliquot\ mL}} \tag{5-13}$$

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, p. 3). 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 IMBA.

### 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, claim information includes bioassay records 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 $^{239}$Pu. For heat source plutonium the mixture is assumed to be 100% $^{238}$Pu.

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.

**Bioassay Procedures and MDAs**

Dose reconstructors should assume that all data in the ORAU database are in pCi excreted per 24 hours and that no additional corrections are required. The MDA of 0.3 recommended by MJW (2002b) is consistent with the MDA of 0.2 for the period from the 1950s to 1989. The 0.3 MDA should therefore be used as the estimate of the MDA from the 1950s to July 1989 as it is more favorable to claimants. Table 5-19 lists MDAs by period for $^{241}$Am and $^{244}$Cm.

Table 5-19. Am-241 and Cm-244 MDAs (dpm/24 hours).

| Period | MDA (dpm/d) | Reference |
|---|---|---|
| Before 07/1989 | 0.3 | MDA (MJW 2002b)[a,b] |
| 07/1989–present | 0.05 | MDA (Barton 1995) |

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

Daily excretion should be determined using Equation 5-14:

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

**Americium-241 and Curium-244 Recommendations**

Exposure to $^{241}$Am in heat source plutonium should be considered negligible. Intakes of $^{241}$Am 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 $^{241}$Am and $^{244}$Cm 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. Table 5-18 provides the MDAs for $^{241}$Am and $^{244}$Cm. The 0.3 MDA should be used as the estimate of the MDA from the 1950s to July 1989 as more favorable to claimants than a lower value reported anecdotally for this period.

### 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 $^{238}$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) overestimates dose from the rare radionuclide of interest. The same is true under the standard assumption that uranium mixtures were 100% $^{234}$U. Dose may not be calculated for presumptive exposures without specific claim information.

### 5.9.9 Radon

NIOSH is unable to determine an upper bound for 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 Sebelius (2010) and Sebelius (2012). The recommended WLM values in Table 5-20 should be used to reconstruct doses from radon exposures for the buildings and periods shown.

Table 5-20. Rn-222 WLM assumptions for dose reconstruction.

| Building conditions | Period | Radon (pCi/L) | Radon (WL) | Radon (WLM/y) |
|---|---|---|---|---|
| SW-19 postproduction and after addition of ventilation | 03/06/1980–2006[a] | 0.7–13.4 | 0.03 | 0.4 |
| Building 21 | 1966–2006 | 17.5–125 | 0.85 | 10.0 |
| Building 48 | 1980–2006 | 3.2 | 0.01 | 0.2 |
| R Building Ra-Ac processing | 1949–1955 | 2.1 | 0.009 | 0.1 |
| All other buildings | 1949–2006 | 0.9 | 0.004 | 0.05 |
| Background–indoors | 1949–2006 | 0.5 | 0.002 | 0.03 |

a. End-date figures are currently unavailable; the year 2006 is assumed for the purpose of the table, but should be used through the end of covered employment past this date.

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 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 of 10% before 1964 and 63% for 1964 and after.

Table 5-21 summarizes corrections for all radionuclides. The primary data types are listed with corrections necessary to estimate intake. In addition, equations are provided to calculate daily excretion. In the records, most radionuclide results are in dpm and do not require detector or extraction efficiency corrections except the radium and actinium data in cph. The radium and actinium data and the $^{241}$Am-$^{244}$Cm data do not require aliquot corrections. The uranium and $^{230}$Th data require

Table 5-21. Summary of correction factors and excretion equations.

| Nuclide | Period[a] | Primary data type | Aliquot correction | Detector efficiency[b] | Extraction efficiency[c] | Equation |
|---|---|---|---|---|---|---|
| Po-210[d] | 1944–1963 | cpm | Yes | Yes | 10% | *Daily Excretion Po-210 = Po cpm/0.5 x 0.85/0.1 x 1400 mL/Aliquot mL* |
| Po-210[c] | 1964–1973 | cpm | Yes | Yes | 63% | *Daily Excretion Po-210 = Po cpm/0.5 x 0.85/0.1 x 1400 mL/Aliquot mL* |
| Pu-238\ Pu-239 | 1957–1960 | dpm | No | No | 10% | *Daily Excretion Pu = Pu Database dpm x 85% ÷ 10%* |
| Pu-238\ Pu-239 | 1961–2006 | dpm | No | No | No | *Daily Excretion Pu = Pu Database dpm* |
| Pa-231 | 08/17/1955–12/1959 | dpm | Yes | No | No | $\text{Daily Excretion Pa-231} = \text{Thorium Database dpm} \times \frac{\text{Sample mL}}{\text{Aliquot mL}}$ |
| Th-230 | 01/1994–2006 | dpm | Yes | No | No | $\text{Daily Excretion Th-230} = \text{Database dpm} \times \frac{\text{Sample mL}}{\text{Aliquot mL}}$ |
| Th-232 | Before 12/87-2006 | dpm | Yes | No | *92%* | *Daily Excretion Ra-224 = Ra Database dpm ÷ 90%*<br>*Daily Excretion Natural Thorium = Th Database dpm ÷ 92%* |
| Uranium | 1950s–2006 | dpm | Yes | No | No | $\text{Daily Excretion U} = \text{Database dpm} \times \frac{1{,}400\text{ mL}}{\text{Aliquot mL}}$ |
| Am-241/ Cm-244 | 1950–2006 | pCi | No | No | No | *Daily Excretion Am-241or Daily Excretion Cm-244 = Database pCi* |

a. Ending dates set to an assumed date of 2006.
b. Detector efficiencies of 50% should be used to correct data in cpm or cph
c. Assume intakes of Ra-223 and Ra-224 were in equilibrium with Ac-227 and Th-228, respectively.
d. Po-210 database values must be normalized for existing 86% extraction and then corrected to 10% and 63%.

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.

For the most part, employees with the potential for intakes of radioactive material were monitored during their employment at the Mound site. Coworker data are available to assign doses from assumed exposures to primary radionuclides at the Mound site, if case-specific information suggests an unmonitored exposure to potential intakes (ORAUT 2012).

## 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 insoluble in the respiratory tract (slow solubilization).

**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 undesired locations including air, soil, buildings, animals, and persons.

**curie (Ci)**

Traditional unit of radioactivity equal to 37 billion ($3.7 \times 10^{10}$) 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

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## ATTACHMENT B
## UNCERTAINTY ASSESSMENT

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 $^{238}$Pu or $^{239}$Pu 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 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 estimate and considered favorable to claimants for all categories of radiation workers. Uncertainty of extrapolating doses from the period from 1959 to1962 to that from 1947 to 1956 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 absorption 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 absorption 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 20% larger than corresponding dose coefficients for 1-μm particles. Therefore, uncertainty in dose coefficients in relation to particle size distribution is dependent on absorption 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). The absorption type of a radionuclide is one of the most important parameters that affects internal dose (NIOSH 2002). Lung absorption 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 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 the primary radionuclides of concern (Table 5-1). There is little uncertainty associated with documentation for $^{210}$Po, the plutonium isotopes, and tritium along with 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 SRS source material.

## ATTACHMENT B
## UNCERTAINTY ASSESSMENT

Information on the location and use of radioactive gases was primarily available only 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 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 Mound analytical procedures 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 $^{238,239}$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 $^{210}$Po 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 that was 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 MDAs.