



# ORAU TEAM Dose Reconstruction Project for NIOSH

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<b>EFFECTIVE DATE</b>	<b>REVISION NUMBER</b>	<b>DESCRIPTION</b>
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.

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**ACRONYMS AND ABBREVIATIONS**

AEC	U.S. Atomic Energy Commission
AL	Action Limit
ALI	annual limit of intake
AMAD	activity median aerodynamic diameter
Bq	Becquerel
Ci	curie
cm	centimeter
cph	counts per hour
cpm	counts per minute
d	day
DOE	U.S. Department of Energy
DL	Detection Limit
dpm	disintegrations per minute
EDTA	ethylene diamine tetraacetic acid
hr	hour
HTO	tritiated water vapor
GSD	Geometric Standard Deviation
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
in	inch
keV	kilovolt-electron, 1,000 electron volts
kg	kilogram
kV	kilovolt
L	liter
mCi	millicurie
MDA	minimum detectable activity
min	minute
ml	milliliter
mo	month
MPBB	maximum permissible body burden
MPC	maximum permissible concentration
mrem	millirem
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NR	Not Reported
pCi	picocurie
PERM	Passive Environmental Radon Monitor
SNAP	System for Nuclear Auxiliary Power

wk        week  
WL        working level  
WLM      working level month

yr        year

$\mu$ Ci      microcurie  
 $\mu$ g        microgram  
 $\mu$ m        micrometer

## 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 historic background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

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

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled “Exposure in the Performance of Duty.” That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer “shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation<sup>1</sup>] guidelines established under subsection (c) ...” [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) define “performance of duty” for DOE employees with a covered cancer or restrict the “duty” to nuclear weapons work (NIOSH 2007).

The statute also includes a definition of a DOE facility that excludes “buildings, structures, premises, grounds, or operations covered by Executive Order No. 12344, dated February 1, 1982 (42 U.S.C. 7158 note), pertaining to the Naval Nuclear Propulsion Program” [42 U.S.C. § 7384l(12)]. While this definition excludes Naval Nuclear Propulsion Facilities from being covered under the Act, the section of EEOICPA that deals with the compensation decision for covered employees with cancer [i.e., 42 U.S.C. § 7384n(b), entitled “Exposure in the Performance of Duty”] does not contain such an exclusion. Therefore, the statute requires NIOSH to include all occupationally-derived radiation exposures at covered facilities in its dose reconstructions for employees at DOE facilities, including radiation exposures related to the Naval Nuclear Propulsion Program. As a result, all internal and external occupational radiation exposures are considered valid for inclusion in a dose reconstruction. No efforts are made to determine the eligibility of any fraction of total measured exposure for inclusion in dose reconstruction. NIOSH, however, does not consider the following exposures to be occupationally derived (NIOSH 2007):

- 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

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

### 5.1.1 Mound Feed Materials

The following process information for Mound feed materials was extracted from Oak Ridge Associated Universities (ORAU) files, *Processes and Characteristics of Major Isotopes Handled at Mound* (ORAU 2003a):

- Airport Cake (Cotter Cake): Primary feed material for extraction of Thorium-230 ( $^{230}\text{Th}$ ). The feedstock was raffinate produced by extraction of uranium from a hydrofluoric or nitric acid solution using diethyl ether followed by treatment with caustic or lime. This feed source was obtained from Mallinckrodt and the Cotter Corporation.
- Brazilian Monazite Sludge: Primary feed material for the thorium refinery project, treated with caustic soda to create insoluble oxides or hydroxides and soluble sodium phosphate. The materials were then leached with hydrochloric acid to extract various rare earth elements. The feed material was sometimes acidic and sometimes basic, depending on pretreatment for a particular batch received.
- Sperry Cake: One of several feed materials derived from uranium milling for extraction of Protactinium-231 ( $^{231}\text{Pa}$ ). Protactinium-231 extraction and purification was a complex process that is poorly described. There is no information to estimate the radioactive compounds likely to have been present.

### 5.1.2 Radionuclides of Concern

Radionuclides were categorized as either primary or secondary by the authors based on the potential extent of use and the amount of bioassay data available. Primary radionuclides of concern are discussed in Mound internal dosimetry documentation, *History of Mound Bioassay Programs* (Meyer 1992) and *Mound Site Radionuclides by Location* (King 1995). Primary radionuclides assessed for occupational exposure included:

- Polonium-210 ( $^{210}\text{Po}$ )
- Plutonium Isotopes: Plutonium-238 ( $^{238}\text{Pu}$ ), Plutonium-239 ( $^{239}\text{Pu}$ ), Plutonium-240 ( $^{240}\text{Pu}$ ), Plutonium-241 ( $^{241}\text{Pu}$ ), Plutonium-242 ( $^{242}\text{Pu}$ ),
- Protactinium-231 ( $^{231}\text{Pa}$ )
- Tritium ( $^3\text{H}$ )

These radionuclides were identified as primary because they can deliver relatively high doses to human receptors, had widespread use at the site in multiple buildings and facility processes, and had sufficient site documentation to evaluate internal dosimetry. Tritium was included due to potential absorption via hydrogen exchange across the skin barrier. Primary onsite operations used all of these radionuclides in conducting key mission activities. Polonium-210 was referred to as Postum, Thorium-230 was referred to as Ionium, and Tritium was referred to as Hot Gas in Mound documentation.

Secondary radionuclides of concern were assessed with limited internal dosimetry data and included the following in addition to daughter products from the uranium and thorium decay chains:

- Americium-241 ( $^{241}\text{Am}$ )
- Curium-244 ( $^{244}\text{Cm}$ )

- Uranium Isotopes: Uranium-238 ( $^{238}\text{U}$ ), Uranium-235 ( $^{235}\text{U}$ ), Uranium-234 ( $^{234}\text{U}$ ), Uranium-233 ( $^{233}\text{U}$ )
- Thorium Isotopes: Thorium-232 ( $^{232}\text{Th}$ ), Thorium-230 ( $^{230}\text{Th}$ ), Thorium-228 ( $^{228}\text{Th}$ )
- Actinium-227 ( $^{227}\text{Ac}$ )
- Radium Isotopes: Radium-228 ( $^{228}\text{Ra}$ ), Radium-226 ( $^{226}\text{Ra}$ ), Radium-224 ( $^{224}\text{Ra}$ ), Radium-223 ( $^{223}\text{Ra}$ )
- Radon Isotopes: Radon-222 ( $^{222}\text{Rn}$ ), Radon-220 (Thoron,  $^{220}\text{Rn}$ ),
- Radon-219 (Actinon,  $^{219}\text{Rn}$ )

These radionuclides are secondary due to limited use at the site or limited worker exposures. Americium-241 should be considered as a separate radionuclide resulting from alpha source production, neutron source production, analysis and vaporization of plutonium compounds,  $^{231}\text{Pa}$  separation, and reactor fuel research programs. Americium-241 was also a minor constituent of weapons-grade plutonium materials used at Mound. Short-lived radioactive gases and their respective daughter products were present at Mound from activities involving uranium and thorium chain extraction and separation activities or the manufacturing of sealed sources. However, worker exposures to these gases were minimized by a ventilation system installed in 1980 and were largely restricted to the SW building where radium and actinium processing took place. Xenon and krypton were present in H Building as a part of the Isotope Separation Program and xenon isotope separation activities. However, the International Commission on Radiological Protection (ICRP) stated in Publication 30 that external irradiation dominates worker dose to the extent that internal dose should be disregarded for most noble gases, including xenon and krypton (ICRP 1979). Radon and thoron are noble gases, but exposure could result in significant internal doses. Curium-244 ( $^{244}\text{Cm}$ ) and Neptunium-237 ( $^{237}\text{Np}$ ) were present at the site in minor concentrations in special nuclear heat source material from the Savannah River Site and present in aqueous wastes derived from plutonium recovery operations. Curium-244 and  $^{237}\text{Np}$  were also used in reactor fuel research and support to the heat source program. Reactor fuel research used  $^{237}\text{Np}$  and  $^{244}\text{Cm}$  in metallic or oxide form. Curium-244 has a relatively high dose conversion factor less than a factor of two below  $^{239}\text{Pu}$  and  $^{232}\text{Th}$  and therefore the potential to induce relatively high doses via the inhalation pathway should be taken into account by the dose reconstructor. However, both  $^{237}\text{Np}$  and  $^{244}\text{Cm}$  were used in minor quantities and were of secondary radiological concern.

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

### 5.1.3 Internal Dose Control Program

Bioassay included primarily *in vitro* urine data and limited fecal sample results. *In vitro* urine data comprises the primary source of information to support dose reconstruction. Nose swipe data were considered to be secondary information and were not evaluated. Whole body counting was not assessed for tritium,  $^{210}\text{Po}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{231}\text{Pa}$  because these nuclides are nearly pure alpha emitters or have weak gamma emissions. Air-sampling data was also considered secondary information contingent upon demonstration that measured concentrations were representative of breathing zone concentrations. Various bioassay programs were conducted at Monsanto Chemical

Company facilities and at Mound from 1945 to the present using routinely collected urine samples from workers (Meyer 1992). The polonium bioassay program described below was also used in Units I, III, and IV of the Monsanto Chemical Company; however, this document provides dose reconstruction provisions only for the Mound Laboratory. Table 5-1 lists primary bioassay programs with reported radionuclide action levels in counts per minute for a urine sample, workers who were monitored, and frequency of monitoring.

Table 5-1. Bioassay programs and action levels.

Bioassay program	Period	Action levels <sup>a</sup>	Monitored workers	Frequency
<b>In vitro monitoring</b>				
Po-210 (Postum) recovery	1945–1974	10 cpm 50 ml <sup>-1</sup>	Operations personnel	Weekly spots Weekly 24 hr
Ac-227, Ra-226, Th-228	1951–1960	8.3 cpm 24 hr <sup>-1</sup>	Research personnel	Weekly 24 hr
Th-232 sludge, re-drumming	1955–1975	2.8 cpm 24 hr <sup>-1</sup>	Th refinery personnel	Monthly 24 hr
Th-230 (Ionium) research	1956–1959	3.5 cpm 24 hr <sup>-1</sup>	Research personnel	Bi-weekly 24 hr
Pa-231 extraction	1956–1960	2.2 cpm 24 hr <sup>-1</sup>	Research personnel	Monthly 24 hr
U-233 research	1958–1960	20 cpm 1,500 ml <sup>-1</sup>	10 Research personnel	Weekly 24 hr
Tritium (hot gas) production	1957–present	10 µCi L <sup>-1</sup>	Recovery personnel	Weekly spots Weekly 24 hr
Pu-239 neutron source	1957–present	3.5 cpm 24 hr <sup>-1</sup>	Processing personnel	Weekly spots Weekly 24 hr
Pu-238 heat source	1960–present	3.5 cpm 24 hr <sup>-1</sup>	Processing personnel	Weekly spots Weekly 24 hr
Am-241 & Am-243	1983–present	0.0 pCi/d <sup>b</sup>	Potentially exposed personnel	Daily
Cm-244	1983–present	0.0 pCi/d <sup>b</sup>	Potentially exposed personnel	Daily
<b>In vivo monitoring</b>				
Whole body counting	1960–1970	Not available	Operations personnel	Annual
Chest counting	1970–present	Variable <sup>c</sup>	Operations personnel	Annual

- AL = action levels; observed counts per minute for 50 ml or 24-hour samples that result in a worker dose of 300 mrem wk<sup>-1</sup> to the target organ.
- Smallest reported activity in database.
- Chest counting MDAs reported in the Mound TBD, Table 5-19 for Am, Pu, Th, and U isotopes. Current day MDAs range from 0.1 nCi for Am-241 to 130 nCi for Pu-239.

Routine bioassay monitoring included collection of spot and 24-hr urine samples identified in Table 5-1. For example, only those workers conducting research to separate and purify protactinium were monitored monthly for <sup>231</sup>Pa. Twenty-four hour samples were also requested for <sup>210</sup>Po and the plutonium isotopes when requested. Monitoring frequencies were surmised from actual performance data and personnel sampling records.

Individuals identified as being involved in an internal exposure incident or had urinalysis results in excess of the reporting limits listed in Table 5-1 were required to submit additional urine 24-hr samples for analysis. Administrative personnel were not monitored and monitoring was discontinued for operational personnel if their work on specific projects was no longer needed. Operational personnel were monitored annually by whole body or chest counting or if there was a suspected exposure incident.

Mound used the action levels in Table 5-1 to evaluate and control worker doses to a 300 mrem/week target organ dose limit. Results in excess of action levels triggered re-sampling 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 re-sampling. Typical dosimetry controls were in place by 1949 when Mound operations were moved from the Dayton facility. These controls followed the limits stated in Handbook 52 published in 1953 (NBS 1953).

#### 5.1.4 Gross Alpha Analysis

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. Section 5.3.2.4 describes one of these procedures. Claimant-favorable assumptions have therefore been made when isotopic data were in question or unavailable. Assignment was also based on workplace information.

#### 5.1.5 Detection Limit and Minimum Detectable Amount Quantification

The decision levels and minimum detectable activities (MDAs) are calculated using the following relations for counting of urine (Currie 1968):

$$D_L = \frac{4.65}{2} \frac{\sqrt{C_{BACKGROUND}} + 3}{TREVA} \quad (5-1)$$

where

- $D_L$  = decision level in dpm L<sup>-1</sup>
- $C_{BACKGROUND}$  = background counts in the region of interest
- $T$  = count time
- $R$  = recovery
- $E$  = average detector efficiency
- $V$  = sample volume (usually 1.5 L)
- $A$  = the alpha abundance for the radionuclide in question

The decision level is the level at which activity is considered present in a sample with a 95% confidence level.

$$MDA = \frac{4.65}{2} \frac{\sqrt{C_{BACKGROUND}} + 3}{TREVA} \quad (5-2)$$

The reporting level is different from the MDA; it is the minimally acceptable decision level that the bioassay laboratory was required to achieve. MDAs are typically below the reporting levels. Mound technical documentation provided no quantitative relationship between the reporting level and MDAs. In general, detection limit data for urine bioassay was sparse.

## 5.2 BUILDING USE AND SITE PROCESSING ACTIVITIES

Table 5-2 summarizes radionuclide use by building based on information from King (1995). This information should be considered when reconstructing occupational doses. The numbers of rooms where radionuclides had primary use and were cited as a major dosimetric concern are shown below. Many radionuclides were not used or had limited use in buildings. For example, <sup>227</sup>Ac was not used in HH, PP, SM, and T Buildings. Dose assessments should not be conducted for radionuclides that had no demonstrated use in buildings where exposure occurred.

## 5.2.1 Particle Size Distributions

The Mound Laboratory conducted a wide variety of research and weapons production activities using primary and secondary radionuclides. Particle size distributions ranged from submicrometer to large metal-clad materials. In addition, Mound used or investigated a complex array of chemical compounds, which resulted in a full range of reported lung clearance types. Attachment A, Table A-1 lists recommended lung clearance class information for radionuclides used in primary building activities and processes. A claimant-favorable solubility should be used if a specific solubility type

Table 5-2. Radionuclide use by building.

Nuclide	Building <sup>a</sup>						
	HH	PP	R	SM	SW	T	WD/WDA
<b>Primary radionuclides</b>							
H-3	8/8/13	None	6/15/52	None	19/28/38	24/32/96	1/8/14
Po-210	4/4/13	None	30/29/52	None	4/5/38	49/69/96	4/5/14
Pu-239	None	39/39/39	5/28/52	19/19/19	6/7/38	3/15/96	1/11/14
Pu-238	None	39/39/39	25/28/52	19/19/19	7/7/38	4/21/96	10/11/14
Ac-227	None	None	24/29/52	None	2/29/38	None	0/6/14
Pa-231	2/2/13	None	5/13/52	None	8/13/38	None	0/2/14
<b>Secondary radionuclides</b>							
Cm-244	None	0/7/39	0/6/52	None	None	None	None
Am-241	None	0/7/39	6/13/52	0/5/19	0/1/38	0/6/96	0/10/14
Np-237	None	0/7/39	0/4/52	0/3/19	None	None	0/10/14
Uranium	0/2/13	None	6/27/52	0/2/19	0/29/38	0/13/96	0/11/14
U-233	0/1/13	None	2/2/52	None	1/1/38	0/2/96	None
Ra-228	None	None	None	None	None	None	0/2/14
Ra-226	None	None	0/27/52	None	3/9/38	0/3/96	0/3/14
Rn-222	None	None	0/27/52	None	3/9/38	0/3/96	0/2/14
Th-232	0/2/13	0/7/13	0/10/52	0/4/19	6/15/38	None	0/5/14
Th-230	2/1/13	None	5/30/52	None	7/11/38	None	0/5/14
Th-228	0/2/13	0/7/13	0/26/52	0/4/19	2/19/38	None	0/5/14

a. nn/nn/nn = number of rooms where a radionuclide was of major concern / total number of rooms using a radionuclide / total number of rooms where radioactive materials were used, stored, or present.

cannot be determined. In addition, solubility can be determined based on claim-specific intake or bioassay data for a given cancer type and might differ from solubility recommendations stated in table A-1. Particle size expressed in activity median aerodynamic diameter (AMAD) and lung clearance types were evaluated based on site and process information from Meyer (1992) and King (1995). In the absence of specific site data, AMAD particle size and lung clearance types were based on recommendations in ICRP Publications 66 and 68 (ICRP 1994, 1995).

Particle sizes at Mound reportedly ranged from submicrometer to 10s of micrometers (King 1995). Submicrometer particles were reported to be abundant, and significant quantities of plutonium dioxide were vaporized during plasma torch operations (King 1995). Such plutonium particles from plasma torch operations would be defined as fumes by industrial hygiene and toxicology terminology (Soule 1991). In addition, microsphere processing was conducted at Mound with a range of product particle sizes from 50 to 250  $\mu\text{m}$  (King 1995). Processes that likely produced submicrometer particle size distributions included:

- Plasma torch operations with vaporization of plutonium in the PP building
- Microsphere production using plutonium in the PP and SM buildings
- Polonium heat source manufacturing and research in the R building
- Plutonium metal production in the SM building

However, much of the reported particle size distribution information from the site was qualitative in nature and could not be verified by evaluation of quantitative particle size study results. In addition, it's not clear whether actual breathing zone particle sizes were based on reported estimates of process particle size distributions.

Due to these limitations, the default particle size distribution AMAD from ICRP Publication 66 (ICRP 1994) of 5  $\mu\text{m}$  should be used for all processes, as indicated in Table A-1. The default AMAD of 5  $\mu\text{m}$  should be used to estimate intake and dose with an associated geometric standard deviation of 2.5 or the default value given in IMBA.

## **5.2.2 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 following radium extraction by coprecipitation to quantify  $^{223}\text{Ra}$ ,  $^{224}\text{Ra}$ , and  $^{226}\text{Ra}$  activity. Evaluation of alpha activity with decay time allowed the activities of individual radium isotopes to be estimated. Table 5-3 lists claimant-favorable equilibrium assumptions to infer primary radionuclide activity in urine based on detected alpha activity.

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

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

## **5.3 PRIMARY BIOASSAY DETECTION DATA**

This section presents primary bioassay information to support intake and dose reconstruction. Primary bioassay data resulted from routinely collected *in vitro* urine samples followed by various analyses for radionuclide content. Analysis data were available for the primary radionuclides. Recommendations are provided to facilitate dose reconstruction.

### **5.3.1 Primary Radionuclides**

This section discusses specific information for the primary radionuclides, including *in vitro* bioassay data, history of MDAs, detection limits, reporting limits, percent recoveries, and analytical procedures. Recommendations are provided to facilitate dose reconstruction.

### 5.3.1.1 Tritium

There was reported tritium ( $^3\text{H}$ ) recovery, purification, and use for solid tritiated metal compounds in 1948 in R building and tritium use limited to testing and research activities in T building as early as 1947 (*sic*, King 1995)<sup>2</sup>. Tritium was used extensively at Mound since the start of weapons component production in 1954. 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,

Table 5-3. Recommended equilibrium assumptions for secondary radionuclides.

Radionuclide	Method of inference	Recommendations
Ac-227	Consecutive gross alpha counts of extracted radium. Ra-223 with 11.4-d half-life detected in urine sample.	Assume Ac-227 and Ra-223 are in equilibrium and activity intake rates are equal. <sup>a</sup>
Th-228	Consecutive gross alpha counts of extracted radium. Ra-224 with 3.6-d half-life detected in urine sample.	Assume Th-228 and Ra-224 are in equilibrium and activity intake rates are equal. <sup>a</sup>
Ra-226	Consecutive gross alpha counts of extracted radium. Ra-226 with 1,602-yr half-life detected in urine sample.	No assumption necessary.
Th-230	Gross alpha counting of extracted thorium in urine starting with sample 6507-11 on August 1954. Gross alpha counting of extracted radium in urine beginning with sample 6507-10I on September 1954-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 <sup>b</sup>	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. <sup>a</sup> 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. <sup>a</sup> Assume Th-228 and Th-232 are in equilibrium for doses based on gross alpha bioassay data.
Uranium isotopes: U-233, EU <sup>c</sup> , 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 enriched uranium.
Po-210	Electrodeposition followed by low background proportional counting.	Assume equilibrium with Pb-210 to determine worker intakes.
Uranium isotopes: natural U <sup>d</sup> , depleted U <sup>e</sup>	Fluorimetric or gross alpha determination.	Assume natural uranium ratios for U-238, U-234, and U-235 or use IMBA default ratios for U-nat and DU.

a IMBA Expert OCAS Edition 3.0.63 (Birchall et al. 2003) does not directly calculate excretion of progeny due to intake of parent radionuclides with progeny.

b Thorium-232 is also identified as natural thorium and Thorium-230 as ionium.

c Uranium enriched in  $^{234}\text{U}$  and  $^{235}\text{U}$ . The degree of enrichment was not reported. These uranium isotopes would not be present as isotopically pure materials.

<sup>2</sup> In fact, the construction of T Building was not complete until December 20, 1948, the first occupation of the building was March 15, 1948, and formal acceptance of the building was February 28, 1949 (SRDB Ref ID 25491: T Building Structural History and Process History Summary Background Document).

- d Uranium with naturally occurring mass and activity ratios.
- e Uranium depleted in <sup>234</sup>U and <sup>235</sup>U.

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 at all buildings except PP (Plutonium Processing) and SM (Special Metallurgical). Extensive use of tritium occurred in R (Research), SW, and T Buildings. These activities resulted in tritium-contaminated soils under SW Building (King 1995). Eight tritium incidents reported by King (1995) occurred in SW Building from 1964 to 1986. Tritium exposures ended in 1995 as reported by King (1995).

### ***In Vitro* Bioassay for Tritium**

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

Liquid scintillation counting began in 1961, with an approved procedure in 1968. This methodology used a 3-ml urine sample placed in an Ultimate Gold cocktail solution (Butler 1961). A quench curve was used to correct results for counting efficiency affected by chemical and physical factors that inhibit photon detection. In addition, many liquid scintillation detectors used external Barium-133 (<sup>133</sup>Ba) or <sup>226</sup>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  $\mu\text{Ci L}^{-1}$  of HTO (Mound 1981). Tritium results were always reported in units of activity per liter of urine. Table 5-4 lists the reported analytical procedures and detection limits for tritium.

Table 5-4. Reported analytical procedures and detection limits for tritium.

Period	Analytical method	Detection limit ( $\mu\text{Ci L}^{-1}$ )
1948–1956	No urinalysis conducted	Use 95% dose <sup>a</sup>
1957–1961	Vibrating reed electrometer	1.0
1962–1995	Liquid scintillation	0.01

a. The upper 95 percentile dose of 340 mrem should be used to reconstruct doses.

### **Tritium Dose Calculations**

A method is presented in this section to convert recorded tritium doses based on LANL dosimetry used at Mound to current ICRP 67 and ICRP 68 dosimetry. Correction from ICRP 30 to ICRP 77 is not needed due to an overall correction factor of 1.0 as shown in equation 5-5 below.

A dosimetry method developed at Los Alamos National Laboratory (LANL) was used for acute tritium dose calculations from 1957 through 1988 (Lawrence 1957). This methodology assumed a maximum permissible body burden (MPBB) of 3.7 mCi, a relative biological equivalent of 1.7, an effective half-life of 12 days, and a body water compartment of 43.4 kg. This results in a maximum permissible concentration (MPC) of 85  $\mu\text{Ci L}^{-1}$  for a target dose limit of 300 mrem  $\text{wk}^{-1}$  under the LANL system of dosimetry (Lawrence 1957, DOE 1988). The target dose limit of 300 mrem  $\text{wk}^{-1}$  was based on an organ dose limit of 15 rem (FRC 1960). The LANL dose relationship used at Mound is shown in equation 5-3.

$$D(t) = 0.00873 B \tag{5-3}$$

where

$B$  = measured acute body concentration in urine or body water  
(uCi/L)

$D(t)$  = the dose commitment at time  $t$  following exposure (rem)

Effective  $T1/2 = 12$  days

Reported doses on the Radioactive Gas Exposure Evaluation forms confirm that Mound used equation 5-3 for tritium dose calculation and that  $6 \mu\text{Ci L}^{-1}$  corresponds to 52 mrem as entered on the forms. However, this equation should not be used to reconstruct dose and only provides the confirmed basis to correct doses reported by Mound using equation 5-4.

Current dosimetry for tritium uses a QF = 1.0 compared to an assumed LANL RBE = 1.7, a two compartment elimination model, and a target mass of 68.8 kg compared to 43.4 kg used in the LANL calculations (ICRP 67; ICRP 68). Equation 5-4 provides a ratio of ICRP 68 dose compared to LANL dose, which results in a ratio of 0.34.

$$\text{Ratio (ICRP68 / LANL)} = \frac{43.4}{68.8 \times 1.7} \times \frac{\int_0^{\infty} (0.97 \times e^{-0.693 \times t / 10} + 0.03 \times e^{-0.693 \times t / 40}) dt}{\int_0^{\infty} e^{-0.693 \times t / 12} dt} = 0.34 \quad (5-4)$$

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

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

### Tritium Reporting Limits

Mound reporting limits for tritium impacted what doses were reported for monitored workers. Doses in rem or microcuries were reported as blanks on Mound Tritium Exposure Reports when urine concentrations were less than  $6 \mu\text{Ci L}^{-1}$  (Meyer 1992). Reporting limits of 3 uCi/L as discussed by James (1997) or  $6 \mu\text{Ci L}^{-1}$  were both reported on the Radioactive Gas Exposure Evaluation Forms as actual bioassay results. The 6 uCi/L should be assumed when tritium doses have been reported as blanks, otherwise all actual reported bioassay results should be used.

Values of 0.0 were used for non-detection results, which biased the doses below the true estimate of the average dose. MDAs in Table 5-4 should therefore be used to estimate intakes at or below detection.

### Unmonitored Tritium Exposures

This section provides a methodology to reconstruct tritium doses by assuming that doses of tritium from 1948 through 1956 were similar to reported doses from 1959 to 1962. Personnel dose data from Bigler's Annual Monitoring Reports from 1959 to 1962 reported numbers of workers by category of annual doses as shown in Table 5-5 (Meyer 1992). These data clearly demonstrate that 94% to 98% of all workers received doses below one rem from 1959 to 1962. The dose in all years was therefore assumed to be the upper end of the 0-1 rem range. The value of 1000 mrem was assumed to be the approximate upper 95 percentile and claimant-favorable dose for most workers due to data limitations.

Table 5-5. Numbers of workers in reported dose ranges by year.

Dose rem/year	Number of workers (percent of total)			
	1959	1960	1961	1962
0-1	117 (94)	249 (96)	286 (98)	303 (98)
1-2	6 (5)	5 (2)	6 (2)	4 (1)
2-3	1 (1)	3 (1)	0	3 (1)
3-4	0	1	0	0
4-5	0	0	0	0
5-10	0	2 (1)	0	0
Max dose (rem)	2.299	8.353	1.693	2.974
Corrected max dose (rem)	0.782	2.840	0.576	1.011
Total # workers	124	260	292	310

All tritium doses reported in Table 5-5 were derived by the LANL dosimetry methodology given in equation 5-3. Correction of these doses using equation 5-4 to ICRP68 dosimetry results in an upper 95 percentile dose of  $1000 (0.34) = 340$  mrem. This ICRP68 equivalent annual upper 95 percentile dose should be assumed for all missing bioassay data from 1947 through 1956.

Reported annual uncorrected and corrected maximum doses in rem are also shown in Table 5-5 and ranged from 0.576 to a high of 2.840 corrected (Meyer 1992).

### Metal Tritides

Limited information is available on the metal tritides 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 U.S. Department of Energy (DOE) report notes that lithium tritide reacts with water vapor to produce mostly HT (DOE 1994). Therefore, the dose from intake of lithium tritide is bounded by the assumption that it reacts to produce HTO.

Insoluble tritides were also used at Mound primarily in SW, R, and T buildings. A lung clearance class of S should be assumed for all metal tritides other than lithium. A lung clearance class of S should also be assumed for insoluble tritides.

### Tritium Recommendations

The corrected upper 95 percentile dose of 340 mrem should be assumed in assigning annual doses to unmonitored radiological workers from 1948 to 1956. When Mound records from 1957 to 1995 only include dose without any concentrations, equations 5-4 should be used to correct reported Mound doses to current ICRP68 doses. Tritium doses reported as blanks from 1957 to 1995 should be reconstructed by assuming a  $6 \mu\text{Ci}$  per liter urine concentration reporting limit. Otherwise, all actual recorded bioassay results should be used. Doses reported at or below detection or as zeros should be corrected using the applicable MDAs given in Table 5-4. Doses would not be assigned to any administrative or other non-radiological workers.

Dose reconstructions should assume that the tritium source term was constant because reliable air data was not available from 1947 to 1957 and effluent records started in 1959, which precluded any evaluation of the tritium source terms during this period of missing urinalysis.

Intake of lithium tritides should be estimated by assuming exposure to HTO. Exposure to all other tritides should assume class S clearance.

#### 5.3.1.2 Polonium-210

Polonium work transferred to Mound in 1949 (Silverman 1962) from the Monsanto Chemical Company facilities (Units III and IV). Polonium-210 was primarily produced by neutron irradiation of

naturally occurring Bismuth-209 ( $^{209}\text{Bi}$ ) slugs to produce Bismuth-210 ( $^{210}\text{Bi}$ ) via a (n,  $\gamma$ ) reaction followed by beta decay to  $^{210}\text{Po}$ . The first documented polonium exposure occurred on August 28, 1944, via a probable skin absorption (Silverman 1962; Spitz 1989a; Runkle 1995). Polonium-beryllium (PoBe) neutron source work ( $^{210}\text{Po}$ ) started at Monsanto Chemical Company facilities in September 1945. Mound's first primary mission was to manufacture initiators for nuclear weapons. This included the extraction of  $^{210}\text{Po}$  from neutron-irradiated bismuth slugs and beryllium machining operations. In addition, Mound conducted polonium research and started the manufacturing of radioisotope thermal generators using polonium in 1954. Non-weapons polonium work at Mound ended in 1972.

Polonium-210 was present in Building HH (Hydrolysis House) from 1949 to 1959 generating sludge materials with  $0.02 \text{ Ci L}^{-1}$  total alpha activity. Extensive use of  $^{210}\text{Po}$  in the R Building involved research and electrodeposition studies from 1948 to 1951 and the manufacturing of PoBe neutron source from 1956 to 1961. PoBe sources were first manufactured at Mound as neutron source initiators to support weapons programs. Polonium was later used to produce  $^{210}\text{Po}$  heat source generators.

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

### **Polonium-210 Records**

Polonium-210 bioassay data were documented on several different types of records at Mound as discussed by Meyer (1992) and Spitz (1989). Weekly Reports of urinalysis data were kept for workers grouped by location with entries in cpm per specified volume and 24-hr samples identified. 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 collected on a single day to a volume weighted average 24-hr excretion in cpm. Finally, Special and Unscheduled Sample Forms were kept when any additional urine samples were required to document possible worker exposures or to confirm previous results with all entries in cpm and duplicate indicated. Bioassay results in activity per day are reported as Bq/d in the PURECON database under the heading of BQ\_DAY. Sample results have also been reported in cpm and dpm in this database.

### ***In Vitro* Bioassay of Polonium-210**

Summary data for  $^{210}\text{Po}$  are provided in Tables 5-6 and 5-19. Bioassay data for  $^{210}\text{Po}$  have been placed in the PORECON database by MJW by evaluating individual dosimetry files from original logbooks (MJW 2002). This database contains individual sample results from 1944 to 1973, encompassing polonium operations at both Monsanto Chemical Company facilities and the Mound Laboratory. Possible exposures to  $^{210}\text{Po}$  in 1943 are assumed to be insignificant based on limited work in a single laboratory and reported administrative activities prior to initiation of bioassay in 1944 (Silverman 1962). Some bioassay data from 1944 through the first half of 1945 was reported missing (Spitz 1989a).

Table 5-6. Reported background count rates and derived minimum detectable activities (MDAs) for  $^{210}\text{Po}$ .

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

- Polonium-210 spot and 24-hr sample MDAs were first normalized assuming 85% extraction efficiency and then corrected for a 10% chemical recovery from 1944-1963 and 63% from 1964-1973. No aliquot size corrections were made for polonium.
- Default sample volumes are 50 ml prior to 7-13-64 and 100 ml after 7-13-64. However, sample volumes as reported should always be used when available. Default sample volumes should only be used if no volume is reported in primary or secondary records.
- This includes the operational period of the Monsanto Chemical Company facilities at Units III and IV.

All Mound employees working in polonium operations in T, HH, WD, H, B, 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. Two samples were usually collected on Monday and Wednesday or three samples were collected on Monday, Wednesday, and Friday. Samples were initially treated without  $\text{HNO}_3$  digestion. Polonium in raw urine samples was spontaneously deposited on nickel disks and counted using a low background proportional counter.

Using an improved method with acid digestion, spot urine samples of 50 ml were collected each week and preserved with 50 mg of sulfamic acid before analysis as reported in MLM-460 (Spoerl 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-hr electrodeposition plating time. Electrodeposition was conducted primarily on 1-in. copper disks within a few hours of collection. Recovery was reported in 1950 as  $86.1 \pm 11.9\%$  for 50-ml samples in Spoerl (1950) and 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 non-normality. Spoerl (1950) also reported that samples with activities in excess of  $200 \text{ cpm } 50 \text{ ml}^{-1}$  had reduced recoveries from 56% to 64%. However, urine sample activity above  $200 \text{ cpm } 50 \text{ ml}^{-1}$  is limited to only 0.5% of the total polonium bioassay records in the PORECON database. The procedure stated that  $^{210}\text{Po}$  losses might occur during the time between sample collection and sample assay (Spoerl 1950). No specific detection limit was reported for this method of analysis in *Recommended Bioassay for Polonium* (Meyer 1992).

All urine bioassay results were reported as counts per minute in 50 ml of urine and were converted to disintegrations per 50 ml using an assumption of  $2\pi$  geometry and the following relation:

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

### Chemical Recovery Corrections

A study conducted in 1989 determined that significant recovery losses from raw undigested urine were observed for metabolized  $^{210}\text{Po}$  due to plating and/or formation of precipitates (Fellman et al. 1989; Meyer 1992). This study was conducted using urine samples from primates by investigators at the New York Medical Center and by Spitz and Robinson at Mound and published in *Health Physics* (Fellman et al. 1989). Metabolized polonium was shown to have a low average chemical recovery of 10% without complete acid digestion or oxidation by ashing the sample. An average ratio of 10.5 was reported for ashed to unashed urine sample results (Fellman et al. 1989). Recovery of the added unmetabolized polonium tracer typically ranged from 75 to 85% (Fellman et al. 1989) as previously

observed at Mound (Spoerl 1950). Mound assumed that metabolized polonium is plated with the same efficiency as the unmetabolized tracer. Actual chemical recoveries at Mound were therefore potentially only 10% rather than the assumed 86%. Reported bioassay urinalysis results would have to be corrected by a factor of 10 up until 1964 when complete acid digestion of urine samples was required (Meyer 1992).

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

MJW conducted a subsequent evaluation of the chemical recovery of polonium (MJW 2001). The position paper concluded that the 10% recovery observed by Fellman (1989) was not appropriate for correction of Mound worker bioassay results due to differing sample storage times following collection but prior to analysis. The majority of Mound sample analyses were thought to have occurred within a 4-hr timeframe post-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 reported by Spoerl (1950) of 86%.

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

A polonium chemical recovery of 10% should therefore be used to correct  $^{210}\text{Po}$  data collected before 1964. Database entries prior to 1964 should be normalized using 85% and then corrected assuming 10% recovery as shown equation 5-7. This lower recommended recovery has been selected because the fundamental assumption of recovery based on deposition of tracer activity has been shown to be invalid for metabolized polonium present in raw undigested or unoxidized urine.

After 1964 to 1973, a correction factor of 63% should be utilized based on ashed sample results duplicating methods used previously at Mound with a 2-hr spontaneous plating time conducted at room temperature (Fellman et al. 1989). Corrections to database entries after 1964 should be normalized using 85% and then corrected assuming 63% recovery as shown equation 5-8.

### Detection Limits and MDAs

Reductions in MPBBs at Mound resulted in development of improved analytical methods for detection of  $^{210}\text{Po}$ . The MPBB in  $\mu\text{Ci}$  and MPC in  $\text{cpm } 50 \text{ ml}^{-1}$  or  $\text{dpm } 50 \text{ ml}^{-1}$  of urine resulting in a target dose of  $300 \text{ mrem wk}^{-1}$ . An MPBB of  $2.0 \mu\text{Ci}$  was used in the early years of operations at Mound from 1944 to 1946. This MPBB was then reduced by a factor of 10 based on recommendations from the National Bureau of Standards (NBS 1953, NBS 1959). The U.S. Atomic Energy Commission (AEC) rescinded use of the  $0.2 \mu\text{Ci}$  MPBB in 1964 and directed Mound to use  $0.04 \mu\text{Ci}$  as recommended by NCRP (Myers 1992).

Polonium-210 detection limits were improved after analytical modifications resulted in a four-fold increase in counting sensitivity and a detection limit of 0.2 dpm for a 24-hr sample. Modifications to the standard sample preparation procedure in 1964 included use of a larger Lucite plating cell to accommodate 100-ml urine samples and electrodeposition onto a single side of a larger stainless-steel disk (Sheehan, 1964). If warranted, 24-hr urine samples were collected to determine potential dose to target organs. The MD-20736 procedure was vigorously investigated and was the primary method of analysis from 1964 to 1973 (Mound 1968).

MDAs were assessed by evaluating reported background counting rates during the early years from 1944 to 1963 and from 1964 to 1973 when detection limits were improved. Table 5-6 presents MDAs calculated using the Currie equation as shown in equation 5-2 (Currie 1968). Background was determined to be 0.2 cpm/24-hr urine sample in 1958 in Annual Overviews (Meyer 1992). This background counting rate was assumed for the early period of <sup>210</sup>Po detection from 1944 to 1963. A lower background counting rate of 0.1 cpm was reported in 1964 to 1966, and from 1971 to 1973 in Annual Monitoring Reports, Consultations, and Annual Overviews (Meyer 1992). This counting rate was assumed for the later period from 1964 to 1973. A two-fold reduction in the background counting rate is consistent with improved analytical sensitivity resulting from a two-fold increase in sample volume used after 1963 (Sheehan 1964).

MDAs were derived for 50-ml spot samples using a 60 minute count time specified in procedure MD-20736 and an assumed 1000 minute count time for 24-hr samples (Mound Laboratory 1968). The background value of 0.1 cpm/24-hr sample for the period from 1964 to 1973 results in a derived MDA of 0.16 dpm using equation 5-2, a detector efficiency of 50%, and recovery of 63%. Estimated MDAs for spot samples were considerably higher due to a much shorter count time of one hour. MDAs derived for 1944 to 1963 were higher primarily due to the assumed chemical recovery of 10% prior to proper acidification in 1964 and a higher background count rate of 0.2 cpm. MDAs in terms of daily excretion are also reported in dpm/d for spot samples.

### Daily Excretion Equations

Sample results in activity per day are reported as Bq/d in the PURECON database under the heading of BQ\_DAY. All Bq/d database values can be corrected by multiplying by 85% and dividing by 10% from 1944 to 1963 and 63% from 1964 to 1973 as shown Table 5-19. All Bq/d database values have already been corrected for a daily sample size of 1400 ml and counting efficiency of 50%. Urine samples identified as 24-hr samples would only require correction for chemical extraction.

Daily excretion in dpm/d for <sup>210</sup>Po spot samples using the PORECON database reported in cpm should be calculated using equations 5-7 and 5-8 as follows:

$$\text{Daily Excretion Po-210} = \text{Database cpm} \div 50\% \times 85\% \div 10\% \times 1400 \text{ ml} / \text{Aliquot ml} \quad (1944 - 1963) \quad (5-7)$$

$$\text{Daily Excretion Po-210} = \text{Database cpm} \div 50\% \times 85\% \div 63\% \times 1400 \text{ ml} / \text{Aliquot ml} \quad (1964 - 1973) \quad (5-8)$$

### Reported Half-Life

The effective half-life was investigated in 1950 and reported by Spoerl et al. (1950) to be 36 d. Dose calculations used at Mound assumed this effective half-life from 1950 to the present. Various quality factors ranging from 10 in the 1950s to 20 in the 1980s were used to determine dose following polonium exposure. ICRP (1979) reported a 50-d biological half-life. However, Sheehan (1951) reported a much shorter effective half-life of only 24 d for exposure resulting from a puncture wound involving a platinum wire contaminated with <sup>210</sup>Po at Mound.

Mound bioassay data seems to favor a 36-day effective half-time rather than the 50-day half-time used by the ICRP in Publication 67. When DRs use the default systemic parameters in IMBA, they should be aware that changes to systemic parameters that effectively decrease retention time might produce better fits to an individual worker's urinalysis data.

### Polonium-210 Recommendations

PURECON database entries in Bq/d should be used when possible. All urinalysis data for  $^{210}\text{Po}$  collected before 1964 should be normalized assuming 85% extraction efficiency and then corrected for a 10% chemical recovery due to plating of metabolized polonium. Polonium-210 data reported after 1963 should be normalized assuming 86% extraction efficiency and then corrected for a 63% chemical recovery based on an analysis by Fellman et al. (1989). Bioassay results at or below detection from 1944 through 1963 should be estimated using derived MDAs in Table 5-6. Daily excretion should be determined using equations 5-7 and 5-8.

#### 5.3.1.3 Protactinium-231

Mound conducted limited research to investigate  $^{231}\text{Pa}$  as a long-lived alpha emitter for possible use as initiators or radiothermal generator source material. ICRP lists  $^{231}\text{Pa}$  with a 32,700-yr half-life (ICRP 1995). The AEC investigated the chemistry and potential applications of  $^{231}\text{Pa}$ . The separation and processing of  $^{231}\text{Pa}$  from Sperry Cake raffinates was conducted in the HH Building, rooms HH5 and HH6, from 1955 to 1956. Separation of  $^{231}\text{Pa}$  from Sperry Cake was also reported in R Building (R148, R149, R167) from 1954 to 1959, and limited laboratory use was reported from 1956 to 1987. In addition, minor quantities of  $^{231}\text{Pa}$  were present in samples subjected to spectrographic analysis conducted in R140 and R142 from 1956 to 1969. Minor concentrations of  $^{231}\text{Pa}$  were found in WD/WDA Building, which received aqueous wastes from R and SW Buildings. No  $^{231}\text{Pa}$  use was reported for SM, SW, T, and PP Buildings.

Use of minimal quantities of  $^{231}\text{Pa}$  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.

#### *In Vitro* Bioassay of 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  $^{231}\text{Pa}$  concluded on December 7, 1959 (Meyer 1992). Bioassay was limited to collection of 24-hr samples. A significant volume of urine was required to detect an MPC target dose of  $300 \text{ mrem wk}^{-1}$  due to the very slow elimination of  $^{231}\text{Pa}$ .

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 Protactinium-231 from natural sources was discussed by Kirby and Figgins (1959), Kirby (1959), and Salutsky et al. (1956). Residues were initially filtered from aqueous raffinate solutions following a diethyl ether extraction of uranium. Filtrates were generally dissolved in HCL saturated with NaCl, precipitated with a silicate or Ce carrier and NaOH or  $\text{NH}_4\text{OH}$ , digested in 9 N HCl or  $\text{HNO}_3$  solutions, and either count the precipitate directly or isolate by anion exchange.

The Thorium Fraction results in cph were converted to cpm, corrected for a 50% counting efficiency, and entered directly into the ORAU database as dpm (ORAU 2003e). Thorium Fraction activity results were then assumed to be  $^{231}\text{Pa}$  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 1959). Analyses were later conducted in accordance with procedure MD-20736 (Mound 1968).

Table 5-7 lists reported background count rates given in Section 4, Bigler's Annual Monitoring Reports by time period and MDAs derived using Currie's equation 5-2 (Meyer 1992).

Table 5-7. Protactinium detection limits (dpm).<sup>a</sup>

Period	Background count rate (cpm)	Detection limit (dpm)
1954–8/17/55	No bioassay	0.3 <sup>b</sup>
8/17/55–1956	0.3	0.3
1957–1958	0.2	0.02
1959–12/7/59	0.15	0.08

- a. Correction for aliquot size should be made for the DL if available.  
b. A detection limit 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 reported on primary records. An evaluation of the <sup>231</sup>Pa ORAU database determined that some sample volumes were not reported and the modal aliquot was 120 ml. If aliquot or sample volumes are not reported in the ORAU database a modal aliquot of 120 ml and a standard man sample volume of 1400 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 (Th) ORAU database dpm as shown in equation 5-9 using actual sample and aliquot volumes or by assuming a standard man sample volume of 1400 ml and a modal aliquot volume of 120 ml (ORAU 2003e):

$$\text{Daily Excretion Pa-231} = \text{Th Fraction Database dpm} \times \frac{\text{Sample or 1400 ml}}{\text{Aliquot or 120 ml}} \quad (5-9)$$

### Protactinium Recommendations

Protactinium-231 use did not occur in Buildings PP, SM, and T. Intakes to workers should be estimated only for exposures in Buildings HH, R, and SW. Activity data in dpm should only be used as taken from the ORAU database for the Thorium Fraction (ORAU 2003e). MDAs in Table 5-7 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 8/17/1955. Daily excretion should be estimated using equation 5-9.

#### 5.3.1.4 Plutonium-238 and Plutonium-239

This document uses the terms *plutonium-238* and *plutonium-239* as synonyms for *heat source plutonium* and *weapons-grade plutonium*. Both heat source plutonium and weapons-grade plutonium were actually mixtures of several plutonium isotopes with smaller amounts of <sup>241</sup>Am and other isotopes. Plutonium-239 was separated from neutron-irradiated uranium metal from 1949 to 1952. In addition, plutonium was used to conduct reactor fuel research from 1956 to 1959. The Neutron Source Group started production of heat sources in the R Building using <sup>239</sup>Pu in 1956. The Metallurgical Research Group used plutonium to conduct research on plutonium metal compounds from 1956 to 1959.

In 1959, the Mound facility initiated use of <sup>238</sup>Pu. Plutonium metal production was used to manufacture heat sources from 1961 to 1965 in support of the SNAP Program in the SM Building. Uranium-234 was separated from aged <sup>238</sup>Pu from 1970 to 1980.

Plutonium use at Mound was primarily conducted in the R, SM, T, and PP Buildings in support of metallurgical and chemical research, heat source production, and microsphere fabrication. Feed material received from the Savannah River Plant as plutonium-dioxide powder was enriched in <sup>238</sup>Pu and processed at Mound to manufacture heat sources. Table 5-8 lists the percentage isotopic mixture by weight and activity for this material.

Table 5-8. Heat source plutonium feed material characteristics.<sup>a</sup>

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

a. Source: King (1995).

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

However, <sup>238</sup>Pu and <sup>239</sup>Pu were both used at Mound, including research work confined to the use of weapons-grade <sup>239</sup>Pu and the manufacture of initiators. Weapons-grade <sup>239</sup>Pu used in various projects was reported to be over 95% pure with trace amounts of <sup>240</sup>Pu and <sup>241</sup>Pu (King 1995). Table 5-9 lists the percentage mixture by weight and activity for this material.

Table 5-9. Weapons-grade plutonium feed material characteristics.<sup>a</sup>

Nuclide	% Mass	% Activity by effective age <sup>b</sup> (years)		
		1.0 yr	3.2 yr	10 yr
Pu-236	0.0001	0.0	0.0	0.0
Pu-238	0.029	1.2	1.3	1.6
Pu-239	93.2	14.0	15.1	19.3
Pu-240	5.71	3.1	3.4	4.3
Pu-241	0.33	81.6	79.7	73.3
Pu-242	0.1	0.0	0.0	0.0
Am-241	0.02	0.1	0.4	1.5

a. Source: HPS (1985).

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

In general, <sup>238</sup>Pu compounds are more soluble than <sup>239</sup>Pu due to greater specific activity and therefore a greater energetic alpha recoil for <sup>238</sup>Pu.

A particle size of 5 μm should be assumed for plutonium and all other alpha emitters in the R Building at Mound in the absence of specific information on actual particle size distributions (King 1995). The R Building included process laboratories, research laboratories, analytical laboratories, and vault storage facilities that supported plutonium work. After 1969, heat source manufacturing and plutonium separation were associated with R Building, and <sup>238</sup>Pu was the major radionuclide of concern. Plutonium compounds were subjected to spectrographic analysis in R140 and R142 with possible inhalation of fumes. Various incidents were reported in R Building from 1960 to 1974 involving exposure to <sup>238</sup>Pu. A single plutonium–beryllium exposure incident occurred in 1960.

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<sub>2</sub> particles in the submicrometer to 20 μm range (King 1995). Microspheres would be quite stable and their initial particle size distribution would only be affected by depositional processes that remove particles above 10 μm from worker breathing zones. Deposition would therefore tend to increase the inhalation of submicrometer particles. Reported solubility varied widely from type S in room SM60 to types M and F in SM27 and SM28. Use of a plasma torch process primarily in SM39 resulted in PuO<sub>2</sub> fumes.

Encapsulation, X-ray inspection, and calibration of plutonium–beryllium neutron sources was conducted in T Building from 1955 to 1985. Counting and verification of sources was conducted in various laboratories of T Building. Two  $^{238}\text{Pu}$  exposure incidents were reported in 1986.

All SM processes were transferred to the PP Building in December 1967. The primary mission in this building was to process  $\text{PuO}_2$  powder from the Savannah River Plant. In addition, a number of processes were developed to manufacture plutonium microspheres, which occurred primarily in the PP Building. These processes involved the following:

- Plutonium–molybdenum Cermet process to coat plutonium particles from 43 to 250  $\mu\text{m}$  with molybdenum from 1969 to 1973
- Pressed plutonium-oxide process to fabricate plutonium spheres for encapsulation as heat sources from 1970 to 1979
- Milliwatt Generator Program process to seal 105- to 500- $\mu\text{m}$  plutonium shards with yttrium
- Solid solution process from 1979 to 1974
- Solution gel process from 1969 to 1971
- Microsphere process to produce 50- to 250- $\mu\text{m}$   $^{239}\text{Pu}$ -dioxide ceramic spheres from 1967 to 1970

The primary radiological concern at the PP Building was the possible inhalation of respirable plutonium particles. Plutonium microsphere processing in rooms PP162 and PP163 was reported to produce a product that had a solubility type S due to plasma processing and high-temperature fusing into oxides (King 1995). However, between 1967 and 1970, plutonium processing in rooms PP144, PP147, and PP148 was reported to produce class D or W materials (King 1995). Classes D and W were assumed to correspond to current ICRP lung clearance types F and M (ICRP 1995). A number of plutonium compounds were used in PP Building. Table 5-10 summarizes the number of rooms where plutonium compounds were used.

Table 5-10. Plutonium compound usage in the PP Building.

Plutonium compound	# Rooms	Use period
Plutonium dioxide– $\text{PuO}_2$	26	1967–present
Plutonium nitrate– $\text{Pu}(\text{NO}_3)_4$	15	1967–present
Plutonium oxalate– $\text{Pu}(\text{C}_2\text{O}_4)_4$	13	1967–present
Plutonium hydroxide– $\text{Pu}(\text{OH})_4$	14	1967–present
Plutonium fluoride– $\text{PuF}_4$	5	1967–1980
Plutonium carbide– $\text{PuC}$	4	1967–1980

Many of the reported plutonium compounds are highly soluble and are ICRP type M. Plutonium nitrate is considered type M, and plutonium ethylene diamine tetraacetic acid (EDTA) complexes should be considered type F due to rapid assimilation in the body (ICRP 1986). Plutonium-238 bioassay data was evaluated in 1996 and determined that the likely lung solubility was a mixture of 50% class W and 50% class Y (Siebert 1996). This mixture was assumed to correspond to ICRP Publication 68 solubility classes 50% M and 50% S (ICRP 1995).

#### ***In Vitro* Bioassay of Plutonium-238/239**

Bioassay of the plutonium isotopes at Mound underwent considerable change and improvement with the development of specific methodologies to isolate and count  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$ . In 1952, an attempt

was made to electroplate plutonium onto copper plates by following the procedure for  $^{210}\text{Po}$ . 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% should be assumed for plutonium bioassay data collected from 1954 to 1960, based on the analysis of reduced polonium recoveries discussed in Section 5.3.1.2.

Starting in 1961, urine samples were acidified to a pH of 2 with nitric acid, resulting in recoveries for metabolized plutonium of  $90\% \pm 3\%$  (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 (Mound 1968). This procedure was modified in 1978 as MD-80030 to include use of a  $^{242}\text{Pu}$  tracer, counting of blanks to quantify detection limits, and alpha spectroscopy (Mound 1981). However, gross alpha results for bioassay were reported as plutonium for all  $^{238}\text{Pu}$  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 and thorium alpha activity. The gross alpha results were reported as  $^{239}\text{Pu}$  for workers in R Building and  $^{238}\text{Pu}$  for SM and PP Building workers until 1980. Table 5-11 summarizes the bioassay history for plutonium.

Table 5-11. Summary of historical plutonium analytical procedures.

Period	Reporting level <sup>a</sup>	Analytical method
1954–1960	Kirby and Brodbeck 1954 0.05 dpm $1.5 \text{ L}^{-1}$ 0.5 dpm $1.5 \text{ L}^{-1}$ (Corrected DL) <sup>b</sup>	Alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; gross alpha for plutonium; proportional counting; recoveries 6-85% due to colloidal plating of metabolized Pu.
1961–1967	Kirby and Brodbeck 1954 0.1 dpm $1.5 \text{ L}^{-1}$	Sample acidification; alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; gross alpha for plutonium; proportional counting; recoveries $90 \pm 3\%$ . Uranium and plutonium results reported separately only in 1966 using anion exchange methods.
1968–1977	MD-20736 (Mound 1963) 0.1 dpm $1.5 \text{ L}^{-1}$	Alkaline earth phosphate coprecipitation; plutonium separation with cerium carrier; anion exchange; electrodeposition; gross alpha reported for plutonium; proportional counting.
1978–1979	MD-80030 (Mound 1981) 0.1 dpm $1.5 \text{ L}^{-1}$	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 (Mound 1981) 0.07 dpm $1.5 \text{ L}^{-1}$ 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-hr 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 will vary.

b. Detection limit was corrected for a low chemical recovery of 10% due to colloidal plating of plutonium.

The precision of  $^{239}\text{Pu}$  analysis was reported in 1981 as 6.7% at a concentration of  $3.2 \text{ dpm kg}^{-1}$  in urine with a large counting error of 73% in Section 3, Bioassay Data Quality Assessment/Control

(Meyer 1992). The ratio of measured to standard results was  $1.06 \pm 0.08$ , which indicates high accuracy.

Table 5-12 summarizes the MDAs for plutonium isotopes in 24-hr bioassay urine samples over time (MJW 2002). The lower limit of detection recommended by the Health Physics Society (HPS 1985) for accreditation of bioassay laboratories was  $0.06 \text{ pCi L}^{-1}$  for plutonium as reported in the Quarterly Environmental Science Reports in Meyer (1992). This standard was met in all years of analysis for  $^{238}\text{Pu}$  and  $^{239,240}\text{Pu}$  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-min counts, and a lower recovery of well below 85% was reported for 1,000-min counts (Mound 1996). Approximately 91% of all samples were counted for 1,000 min.

Daily excretion of the plutonium isotopes should be estimated using the following equations 5-10 and 5-11.

$$\text{Daily Excretion Pu-238,239} = \text{Pu Database dpm} \times 85\% \div 10\% \text{ (1957 - 1960)} \quad (5-10)$$

$$\text{Daily Excretion Pu-238,239} = \text{Pu Database dpm} \text{ (1961 - Present)} \quad (5-11)$$

Table 5-12. Plutonium for reported periods (pCi/sample of urine).<sup>a,b</sup>

Period	Pu-238 MDAs	Pu-239/240 MDAs
Before 1957	(0.85)	(0.85)
1957	(0.94)	(0.94)
1958	(0.6)	(0.6)
1959	(0.5)	(0.5)
1960	(0.5)	(0.5)
1961-1966	0.05	0.05
1967-May 1981	0.11	0.11
Jun-Dec 1981	0.066, 0.027	0.044, 0.020
1982	0.063, 0.023	0.042, 0.017
1983	0.064, 0.024	0.043, 0.018
1984	0.066, 0.026	0.045, 0.019
1985	0.047, 0.017	0.031, 0.012
1986	0.042, 0.015	0.028, 0.011
1987	0.022	0.011
1988	0.017	0.01
Jan-Jul 1989	0.019	0.008
Aug 1989-Dec 1991	0.018	0.008
1992-1995	0.018 <sup>a</sup>	0.009 <sup>a</sup>

a. Plutonium MDAs from before 1957 to 1960 were normalized assuming 85% extraction efficiency and then corrected for 10% chemical recovery due to plating. Based on Mound (1996).

b. All table values based on 1000-minute counts followed by 3000-minute counts from 1981-1986.

### Plutonium Recommendations

A default AMAD of  $5 \mu\text{m}$  should be used for the plutonium isotopes. However, possible exposure to submicrometer materials during plasma torch operations, microsphere processing, and plutonium metal production should be considered. Plutonium nitrate and plutonium EDTA complexes should be considered soluble type M (ICRP 1986). Exposures during final microsphere production should be considered type S. All other plutonium compounds should be considered a mixture of 50% class M and 50% class S.

All 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 prior to sample analysis (Sheehan, Kirby, and Wood 1963; Spoerl 1950).

MDAs in Table 5-12 should be used to estimate missing or undetected bioassay results unless reporting levels in Table 5-11 are greater than the MDA. 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 5-10 and 5-11 as appropriate.

### 5.3.2 Secondary Radionuclides

Several hundred isotopes were present in Mound facilities because the compositions of irradiated materials are complex. Of these, the radionuclides of secondary interest at Mound are  $^{230}\text{Th}$ ,  $^{232}\text{Th}$ ,  $^{226}\text{Ra}$ ,  $^{227}\text{Ac}$ ,  $^{241}\text{Am}$ , and  $^{233}\text{U}$  through  $^{238}\text{U}$ . With the exception of  $^{228}\text{Ra}$  and  $^{226}\text{Ra}$ , most of these isotopes have a low abundance of low-energy gamma emissions. As a group, these isotopes have restrictive annual limits of intake, and with the exception of soluble forms of uranium, have slow biological clearance by urinary excretion. These factors make it difficult to attain useful *in vitro* detection limits by urine bioassay.

Attachment A, Table A-1, lists lung clearance type assumptions for radionuclides used in site process activities, and Table 5-3 lists assumptions about the degree of equilibrium. Attachment B, Table B-1, lists the room numbers where secondary radionuclides were used.

#### 5.3.2.1 Radon Isotopes

Source terms for radon ( $^{222}\text{Rn}$ ), thoron ( $^{220}\text{Rn}$ ), and actinon ( $^{219}\text{Rn}$ ) were present at Mound due to radium and thorium processing. Exposure to these radioactive gases should be included in dose reconstruction.

Source terms for radon ( $^{222}\text{Rn}$ ), thoron ( $^{220}\text{Rn}$ ), and actinon ( $^{219}\text{Rn}$ ) were present at Mound due to radium and thorium processing and separation of  $^{231}\text{Pa}$  and  $^{227}\text{Ac}$ . Thorium sludge from  $^{232}\text{Th}$  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 cotten concentrate were stored to support purification of  $^{231}\text{Pa}$  and  $^{230}\text{Th}$ . Table 5-13 shows the results of various radon surveys at Mound.

Table 5-13. Radon isotope survey data by building and period.

Building	Survey dates	Gas concentration (pCi/l)			WL	WLM yr <sup>-1</sup> (a)
		Radon	Thoron	Actinon		
SW Tunnel	10/12/79	88,000	28,000	640,000		
SW19 before vent	6/79–10/79	67–160				
SW19 after vent	3/80–4/80	7.7–13.4			0.03	0.4
	12/89	2.6	Not detected			
	12/99	<b>1.0–5.8<sup>b</sup></b>				
	4/00	<b>0.7–7.2</b>				
	1998–2000	5.8–12				
SW21	5/83	17.5–52.8			0.4–1.3 0.85 median	4.7–15.3 10.0 median
	12/89	<b>34–118</b>		0.3–0.7		
	4/90–6/90	117–161				
		<b>125</b>				
SW22	6/90	<b>125</b>				
SW48	12/89	3.2				
Old SD	12/89	1.5–2.4				

Fire station	12/89	1.2–1.4			
Paint shop	12/89	0.9–1.2			
A	10/88–2/89	<b>0.1–1.1</b>			
All other buildings	12/89	0.4–0.9			
Outside facilities	5/83	0.24–0.59			
Reported background		0.1 to 2.1 Average = 0.5			

- Working level months per year (WLM yr<sup>-1</sup>) were determined as WL (2000 hr yr<sup>-1</sup>) (1/ 170 hr WLM<sup>-1</sup>).
- Bolded concentrations were derived from charcoal canister measurements.

The SW19 Building had elevated radon concentrations emanating from radium and thorium processing wastes in the “Old Cave.” High concentrations of radon, thoron, and actinon were transported from an underground tunnel into SW19 work areas. Measurements of <sup>222</sup>Rn near an employee’s desk ranged from 67 to 160 pCi L<sup>-1</sup> in 1979. On October 12, 1979, inert gas concentrations in the tunnel were measured at 88,000 pCi L<sup>-1</sup> <sup>222</sup>Rn, 28,000 pCi L<sup>-1</sup> <sup>220</sup>Rn, and 640,000 pCi L<sup>-1</sup> <sup>219</sup>Rn. A <sup>222</sup>Rn concentration of 88,000 pCi L<sup>-1</sup> would correspond to 352 WL, assuming an ICRP 50 indoor mean radon daughter equilibrium factor of 0.45 (ICRP 1987). A ventilation system was installed in early 1980 to vent SW17 tunnel air to an SW18 exhaust.

This system reduced <sup>222</sup>Rn concentrations in SW19 by a factor of 10 or more to 8.2 pCi L<sup>-1</sup> in March 1980. This result was confirmed with a Passive Environmental Radon Measurement (PERM) measurement of 15.4 pCi L<sup>-1</sup> and RDT310A continuous measurements from 7.7 pCi L<sup>-1</sup> to 13.4 pCi L<sup>-1</sup>. A working level (WL) measurement by the employee’s desk was 0.03 WL or 0.03 (12 mo yr<sup>-1</sup>)=0.4 working level month (WLM) compared with an occupational limit of 4 WLM yr<sup>-1</sup>.

Radon surveys in Building 21 were conducted with PERMs in 1983 and ranged from 17.5 to 52.8 pCi L<sup>-1</sup> with WLs from 0.40 to 1.3. WLMs exceeded occupational limits of 4 WLM with calculated values from 4.8 to 15.6 WLM. Radon measured during the radon study with charcoal canisters ranged from 34 to 118 pCi L<sup>-1</sup> in 1989.

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<sup>-1</sup> <sup>222</sup>Rn, except SW and Old SD buildings. In June 2000 a radon study summary report was issued based on 1990 and 1999 measurements (BWXT 2000). Mound site radon background was reported to be 0.5 pCi L<sup>-1</sup> with a range of 0.1 to 2.1 pCi L<sup>-1</sup>. SW-19 was the only building at Mound identified as an area of potential occupational exposure to <sup>222</sup>Rn and <sup>220</sup>Rn.

Thoron and actinon measurement results were very limited despite the high concentrations observed in the underground tunnel. The only actinon measurements were made in Building 21 at 0.3 to 0.7 pCi L<sup>-1</sup>, but the holding time before analysis was excessive compared to the half-life of <sup>219</sup>Rn in seconds. The only thoron measurement in SW19 was below detection. Thoron and actinon exposures would not be detectable in excreta by bioassay due to very short half-lives.

### Radon Isotope Recommendations

The recommended WLM values in Table 5-14 should be used to reconstruct doses following radon exposures. All WLMs should be assumed to be median values of lognormal distributions with a GSD of 3.0.

Table 5-14. Radon-222 WLM assumptions for dose reconstruction.

Building conditions	Period	Radon (pCi/L)	Radon (WL)	Radon (WLM)
SW Tunnel prior to ventilation	1949–1979	88,000	352	4,100
SW-19 Ra-Ac pilot project	1949–1952	160	0.72	8.5
SW-19 Ra-Ac Old Cave production	1952–1960	160	0.72	8.5

SW-19 post production, prior to ventilation	1961–1979	160	0.72	8.5
SW-19 post production, post to ventilation	1980–1995	0.7–13.4	0.03	0.4
SW-21	1952–1995	17.5–125	0.85	10.0
SW-22	1966–1995	125	0.56	6.6
SW-48	1980–1995	3.2	0.01	0.2
R Building Ra-Ac processing	1949–1955	2.1	0.009	0.1
All other buildings	1949–1995	0.9	0.004	0.05
Background–indoors	1949–1995	0.5	0.002	0.03

Exposure to radioactive gases in the tunnel itself should assume concentrations of 88,000 pCi L<sup>-1</sup> <sup>222</sup>Rn, 28,000 pCi L<sup>-1</sup> <sup>220</sup>Rn, and 640,000 pCi L<sup>-1</sup> <sup>219</sup>Rn. A WL of 352 and WLM per year of 4100 should be used to estimate dose to workers only during documented exposure inside the tunnel.

To reconstruct missed doses a background radon concentration of 0.5 pCi L<sup>-1</sup> should be used, which converts to 0.03 WLM indoors assuming 100% occupancy and 0.45 (ICRP 1987).

### 5.3.2.2 Thorium-230

Attachment B, Table B-1, lists the buildings and rooms where <sup>230</sup>Th exposures could have occurred. Most of these exposures would have occurred in conjunction with exposures to other thorium isotopes or <sup>231</sup>Pa. Small research activities involving the extraction of <sup>230</sup>Th (ionium) from raffinate materials occurred in R, HH, and SW Buildings. The program began in early 1956 and ended in 1958, with the last bioassay sample processed on September 9, 1958. This activity was evidently related to an interest in <sup>230</sup>Th as a tracer and as a target material for the production of the isotopes <sup>231</sup>Pa and <sup>232,233</sup>U by neutron irradiation of <sup>230,232</sup>Th (Figgins and Kirby 1966). Exposure to <sup>230</sup>Th waste materials also would have occurred in Building WD/WDA.

Important feed materials included raffinates and their precipitates such as Cotter Cake, which is also known as Airport Cake. Cotter Cake appears to be derived from a raffinate that had been neutralized by caustic or lime after ore material was dissolved in nitric or hydrofluoric acid and extracted with diethyl ether.

#### Thorium-230 Records

Primary <sup>230</sup>Th bioassay records consisted of a logbook, and apparently duplicate records in a brown notebook. Count data were typically recorded on Form O-318 followed by an "I" or "Io". Secondary <sup>230</sup>Th results started as weekly reports on March 17, 1958. Weekly reports included Name, Isotope, and Result. Prior to 1958, secondary <sup>230</sup>Th results were reported on Form O-634 including Name, Badge Number, Date, Type of Analysis, Isotope, and Result. However, some secondary documents have problems with reporting units. Some results are reported to be cph per 24-hour sample when primary records indicate that they are actually cpm per aliquot. The ORAU database should therefore be considered a secondary record extracted from primary records.

#### Bioassay Procedures and Detection Limits

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-hr urine specimens. The objective of the urine bioassay sampling was to detect an MPC that corresponded to a 300-mrem wk<sup>-1</sup> 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-hr urine sample corresponded to a dose equivalent of 300 mrem wk<sup>-1</sup>. Based on Meyer (1992, Section 2), three different urine bioassay procedures were tried for thorium. A thenoyl trifluoro acetone (TTA) urine extraction was the initial procedure (Kirby and Brodbeck 1954). Starting with sample 6507-11, nine samples were analyzed by the "Kirby process-thorium fraction." This apparently refers to the thorium separation and isolation procedure given in MLM-1003 dated 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  $^{230}\text{Th}$  were in equilibrium with  $^{226}\text{Ra}$ . The average ratio of the  $^{230}\text{Th}$  to  $^{226}\text{Ra}$  in the materials 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  $^{226}\text{Ra}$  was in equilibrium with  $^{230}\text{Th}$ . This assumption can be compensated for by using ratios of the incremental urine excretion curves provided by Potter (2002). Excretion curves for acute and chronic exposures can also be readily calculated using the IMBA software (Birchall et al. 2003). ICRP 68 assigns radium to absorption type M; however, thorium can be either type M or S.

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  $^{230}\text{Th}$ . If the reporting basis for an Ionium bioassay result is ambiguous, assume that  $^{230}\text{Th}$  was extracted and counted. Excretion rates in dpm/day should be calculated using equation 5-12 and entries from the ORAU database that contain information on volume, aliquot size, and dpm results, unless contradicted by another bioassay record (ORAU 2003e):

$$\text{Daily Excretion Th-230} = \text{Database dpm} \times \frac{\text{Sample ml}}{\text{Aliquot ml}} \quad (5-12)$$

If no aliquot volume is given in the ORAU database 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  $^{230}\text{Th}$  detection limits. Where no aliquot size is given in the ORAU database, assume the detection limit is in dpm excreted per 24-hr sample (ORAU 2003e).

Table 5-15. Thorium-230 detection limits (dpm).

Period	Detection limit	Reference
Prior to 1/1994	0.3	MDA (MJW 2002) <sup>a</sup>
January 1994–present	0.17	MDA (Barton 1995)

a. Detection limit recommended for all secondary radionuclides (MJW 2002). Correction for aliquot size should be conducted if available.

### 5.3.2.3 Natural Thorium

Monitoring for natural thorium began in 1955 to support a thorium refining; preliminary research began in the R Building. Exposures to  $^{232}\text{Th}$  and daughters 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  $^{232}\text{Th}$ .

The principal  $^{232}\text{Th}$  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 thorium dioxide. Material received by Mound was sometimes basic and sometimes acidic.

### Thorium-232 Records

Thorium-232 records are diverse due to programs conducted for many years. Primary <sup>232</sup>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 <sup>232</sup>Th bioassay data were recorded in a large hardcover record book. However, the first 38 pages from this record book were removed from 7/6/59 to 1/9/61, 7/13/64 to 11/15/64 and 5/30/65 to 6/6/65. These record books apparently do not contain true primary data, but calculated results such as cpm excreted per day. Secondary records in weekly reports contained <sup>232</sup>Th results as cpm/24-hr samples beginning March 17, 1958. In August 1959, secondary results were reported on form O-756. The ORAU database is a record of secondary <sup>232</sup>Th bioassay data extracted from other primary records (ORAU 2003e). Most <sup>232</sup>Th records in the ORAU database have both a thorium and radium entry in dpm (ORAU 2003e). The corresponding primary <sup>232</sup>Th records from the 1950s carry a sample number that ends in "T". Assume that the "T" result is from the natural thorium fraction and "R" indicates that results are from the radium fraction for a given sample.

### Bioassay Procedures and Detection Limits

Bioassay for <sup>232</sup>Th was based on 24-hr urine samples and followed Mound bioassay procedures MLM-460 (Spoerl 1950) and MD-20736 (Mound 1968). Meyer (1992, Section 1) indicates that 2.8 cpm on a 24-hr urine sample corresponded to an intake that would result in a 300-mrem wk<sup>-1</sup> dose equivalent to the critical organ under the dosimetry system in effect when the sample was collected. Meyer (1992), Section 2 recommends correcting bioassay data by converting to counts per minute and then dividing by 1.11 to obtain units of picocuries per 24-hr period.

Where no aliquot volume is given in the ORAU database, assume that the dpm value has been corrected for background, detection efficiency, and represents a 24-hr urine excretion. If an aliquot volume is given in the ORAU database, then assume that the bioassay result is in units of dpm per aliquot. No other corrections are required for urinalysis results in dpm. If urinalysis results are in cpm then a correction for 50% detector efficiency and 92% extraction efficiency should be made to determine dpm results unless it is clear from primary records that these corrections have already been made.

Table 5-16 lists thorium-232 detection limits by period. Detection limits in Table 5-14 have units of dpm excreted per 24-hr when no aliquot size is given in the ORAU database. However, detection limit units should be dpm per aliquot, if an aliquot volume was reported in the database. When interpreting gross alpha thorium bioassay data for thorium sludge activities, dose reconstructors should use the degree of equilibrium assumptions in Table 5-3 and the lung solubility assumptions in Attachment A, Table A-1, unless additional information is available.

Table 5-16. Natural thorium detection limits (dpm).

Period	Detection limit	Reference
Prior to 12/1987	0.3	MDA (MJW 2002) <sup>a</sup>
December 1987–1994	0.02 (Th-232)	MDA (Barton 1995)
January 1994–Present	0.08 (Th-228)	MDA (Barton 1995)

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

Thorium-232 bioassay results in the ORAU database contain a result for radium (Ra) and/or for thorium (Th) (ORAU 2003e). If a radium result is provided for a <sup>232</sup>Th urine bioassay, the primary record sample ID usually ends in "R" and refers to Ra-224. From these results, the daily excretion of Ra-224 can be obtained using equation 5-13 as follows:

$$\text{Daily Excretion Ra-224} = \text{Ra Database dpm} \div 90\% \quad (5-13)$$

A primary record ID that ends in "T" corresponds to a natural thorium (Th) result that includes  $^{232}\text{Th}$  and  $^{228}\text{Th}$ . The daily excretion of natural thorium in the ORAU database can be calculated using equation 5-14 as follows:

$$\text{Daily Excretion Natural Thorium} = \text{Th Database dpm} \div 92 \% \quad (5-14)$$

Equations 5-13 and 5-14 include correction factors of 90% and 92% for radiochemical yield respectively, because database results for radium and thorium have not corrected for chemical extraction.

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

### 5.3.2.4 Radium-226, Actinium-227, and Thorium-228

Mound completed a facility in the SW Building in 1952 for processing neutron-irradiated  $^{226}\text{Ra}$ . The principal activation products of interest were  $^{227}\text{Ac}$  and  $^{228}\text{Th}$ . Actinium-227 and  $^{228}\text{Th}$  decay to  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$ , respectively. Work continued on irradiated  $^{226}\text{Ra}$  until 1960. Attachment A, Table A-1, and Table 5-2 contain additional details of when and where these radionuclides were used.

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

Primary bioassay data for these radionuclides were found in four brown spiral notebooks and on forms O-318. Notebooks are labeled RA#1a and RA#1c. Secondary urinalysis results were written on hand drawn forms and placed in the "J. B. Black binder." Each form had an employee name, sample ID, and the  $^{223}\text{Ra}$ ,  $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$ , and  $^{227}\text{Ac}$  results.

In addition, manila folders contained primary bioassay data with counts collected, assumptions made, equations used, theoretical curve computations, and a written analysis of the observed data (Meyers 1992). Much of this information has already been included in the DOE response to claims.

Secondary data appeared in various forms such as weekly reports, and on forms O-756 and O-634. The ORAU database contains available secondary bioassay records for the Radium Irradiation project, and excerpts are typically found in the DOE response to claims.

The ORAU radium-actinium database provides results for  $^{223}\text{Ra}$ ,  $^{224}\text{Ra}$ ,  $^{226}\text{Ra}$ , and  $^{227}\text{Ac}$  in units of cph for 24-hour urine samples (ORAU 2003d). No aliquot volumes were given in this database.

#### Bioassay Procedures and Detection Limits

Section 2 of Meyer (1992) provides a description of bioassay procedures used for  $^{226}\text{Ra}$ ,  $^{227}\text{Ac}$ , and  $^{228}\text{Th}$ . 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 (Mound 1968).

Radium was extracted from collected 24-hr urine bioassay samples. Intakes of  $^{227}\text{Ac}$  and  $^{228}\text{Th}$  should be approximately in equilibrium with their radium progeny, subject to the caveat that the irradiated  $^{226}\text{Ra}$  was allowed to age for several weeks before being processed in the SW Building.

Radium extracted from urine samples was then subjected to a series of gross alpha counts for various periods of up to 30 or 35 d. 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 <sup>226</sup>Ra, <sup>224</sup>Ra, and <sup>223</sup>Ra. Each bioassay urine sample of interest was apparently evaluated in this manner.

Interpretation of these bioassay data requires some rather involved calculations. After the <sup>223</sup>Ra and <sup>224</sup>Ra excretion rates are determined, it is necessary to estimate the corresponding excretion rates that would have occurred from equal intakes of <sup>227</sup>Ac and <sup>228</sup>Th.

To convert cph urinalysis data a counting efficiency of 50% and 90% extraction efficiency must be applied as documented by Meyer (1992). Assume that all secondary cph records are based on 24-hr samples.

MDAs were not evaluated for <sup>223</sup>Ra, <sup>224</sup>Ra, and <sup>226</sup>Ra because observed gross alpha counts were partitioned into activities attributable to individual radionuclides using complex equations, assumptions, and corrections for ingrowth would be required. However, evaluation of selected records indicated that a reasonable gross alpha background count rate is 4 cph. The background gross alpha count rate can then be used to derive an MDA of 0.09 dpm for missed worker doses by assuming a 50% detector efficiency, 1000 minute count time, and a 90% extraction efficiency in equation 5-2. The 0.09 dpm of total alpha activity should be claimant-favorably assigned to <sup>226</sup>Ra.

The ORAU radium-actinium database resulted from the radium irradiation project (ORAU 2003d). Assume that intakes of Ac-227 and Th-228 were in equilibrium with their radium progeny. Urine bioassay results are in units of cph for Ra-223, Ra-224, and Ra-226. Actinium-227 intakes must be derived from Ra-223 bioassay result, <sup>228</sup>Th intakes must be surmised from Ra-224 bioassay results, and Ra-226 daily excretion is calculated directly using equations 5-15, 5-16, and 5-17 as follows:

$$\text{Daily Excretion of Ac-227} = \text{Ra-223 Database cph} \div 90\% \times \frac{2 \text{ dpm}}{60 \text{ cph}} \quad (5-15)$$

$$\text{Daily Excretion of Th-228} = \text{Ra-224 Database cph} \div 90\% \times \frac{2 \text{ dpm}}{60 \text{ cph}} \quad (5-16)$$

$$\text{Daily Excretion of Ra-226} = \text{Ra-226 Database cph} \div 90\% \times \frac{2 \text{ dpm}}{60 \text{ cph}} \quad (5-17)$$

### 5.3.2.5 Uranium Isotopes

A small-scale research project involving <sup>233</sup>U was conducted from 1958 to 1960. Uranium-233 and other uranium isotopes were analyzed by chemical separation and gross alpha counting using Mound procedure MD-20736 (Mound 1968). It appears that <sup>233</sup>U activities stopped in 1960, but this procedure continued to be used for other uranium isotopes.

#### Uranium Records

Primary records for the uranium isotopes were found in individual claims on forms such as O-318. Information from these records appears to be summarized in the ORAU database (ORAU 2003e). Results for spot uranium samples are in units of dpm per sample aliquot and have been corrected for background, carrier count, extraction efficiency of 80%, and 50% detector efficiency.

#### Bioassay Procedures and Detection Limits

Both spot and 24-hr 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-hr sample result.

Sample results from the ORAU database in cph need to be corrected for aliquot size. The following equation should be used to obtain excretion rates in units of dpm for 24-hr samples using equation 5-18 and from data presented in the ORAU database (ORAU 2003e):

$$\text{Daily Excretion Uranium Isotopes} = \text{Database dpm} \times \frac{1400 \text{ ml}}{\text{Aliquot ml}} \quad (5-18)$$

Site-specific values for background uranium dietary intake at Mound were not available for this analysis. Typical intakes of uranium attributable to background should be on the order of 4.9 Bq yr<sup>-1</sup> <sup>238</sup>U, 4.9 Bq yr<sup>-1</sup> <sup>234</sup>U, and 0.2 Bq yr<sup>-1</sup> <sup>235</sup>U (United Nations 1993) and 12 Bq yr<sup>-1</sup> 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/day in the urine and 2.1 to 2.7 dpm/day in the feces. For dose reconstruction, assume that uranium in the diet of monitored workers resulted in the excretion of 0.07 dpm/day (urine) and 2.1 dpm/day (feces).

Table 5-17 lists detection limits by period for the uranium isotopes, including a DL for fluorometric results if available.

Table 5-17. Uranium isotope detection limits (dpm).

Period	Detection limit	Reference
Early 1950s–1985	0.4 (U-nat) µg L <sup>-1</sup>	ORAU 2003b <sup>a</sup>
Prior to 5/1985	0.3	MDA (MJW 2002) <sup>b</sup>
May 1985–present	0.07 (U-234) 0.03 (U-235) 0.09 (U-238)	MDA (Barton 1995)

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

Attachment A, Table A-1, lists assumptions for lung clearance properties, and Table 5-3 lists isotopic equilibrium assumptions.

### 5.3.2.6 Americium-241 and Curium-244

Americium-241 is a ubiquitous constituent of heat source plutonium (<sup>238</sup>Pu) and weapons-grade plutonium (<sup>239</sup>Pu). The percent of gross alpha activity in heat source plutonium attributable to <sup>241</sup>Am is negligible. Where intakes of <sup>241</sup>Am occurred as a result of intake of weapons-grade plutonium, the <sup>241</sup>Am constituent should be estimated using the composition of weapons-grade plutonium in Table 5-8.

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 <sup>241</sup>Am in R-120 could also have occurred coincident with intakes of weapons-grade plutonium.

#### Americium-241 and Curium-244 Records

Documentation for <sup>241</sup>Am and <sup>244</sup>Cm is very limited. Primary records for <sup>241</sup>Am and <sup>244</sup>Cm are unknown. Secondary data for <sup>241</sup>Am and <sup>244</sup>Cm are found in the ORAU database (ORAU 2003e). The ORAU database indicates that <sup>244</sup>Cm results are related to the plutonium logbooks. All <sup>241</sup>Am and <sup>244</sup>Cm urinalysis data are in units of activity (pCi). The MJW Report (MJW 2002) assumed that <sup>241</sup>Am and <sup>244</sup>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 presented in the database.

### Bioassay Procedures and Detection Limits

Assume that all data in the ORAU database are in units of pCi excreted per 24 hours and that no additional corrections are required. The DL of 0.3 recommended by MJW (2002) is consistent with the DL of 0.2 for the period from the 1950s to 1989. The 0.3 DL should therefore be used as the more claimant-favorable estimate of detection from the 1950s to 7/1989. Table 5-18 lists detection limits by period for <sup>241</sup>Am and <sup>244</sup>Cm.

Table 5-18. Americium-241 and curium-244 detection limits (dpm excreted per 24 hours).

Period	Detection limit (dpm/d)	Reference
Early 1950s–1989	0.2	ORAU 2003b <sup>a</sup>
Prior to 7/1989	0.3	MDA (MJW 2002) <sup>b</sup>
July 1989–present	0.05	MDA (Barton 1995)

a. Statistical basis is unknown and therefore recommend using 0.3 dpm/d as reported by MJW (2002).

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

Daily excretion should be determined using equation 5-19.

$$\text{Daily Excretion Am-241 or Cm-244} = \text{Database pCi} \quad (5-19)$$

#### 5.3.2.7 Americium-241 and Curium-244 Recommendations

Exposure to <sup>241</sup>Am in heat source plutonium should be considered negligible. Intakes of <sup>241</sup>Am resulting from exposure to weapons-grade plutonium should be estimated using the percent activities given in Table 5-8 of the TBD as 0.1% at one year, 0.4% at 3.2 years, and 1.5% at 10 years of in-growth. Assume that the bioassay data provided in the ORAU database were for personnel who were working with <sup>241</sup>Am and <sup>244</sup>Cm source materials (ORAU 2003e).

All data in the ORAU database are in units of pCi excreted per 24 hours and no additional corrections are required. The 0.3 DL should be used as the more claimant-favorable estimate of detection from the 1950s to 7/1989.

#### 5.3.3 Summary of Correction Factors and Excretion Equations

A summary of corrections for all radionuclides is provided below in Table 5-19. The primary data types are listed and those corrections required to estimate intake. In addition, equations are provided to calculate daily excretion. Most radionuclides are in units of dpm and do not require detector or extraction efficiency corrections except the radium-actinium data in cph. The radium-actinium and <sup>241</sup>Am-<sup>244</sup>Cm data do not require any aliquot corrections. The uranium and <sup>230</sup>Th data will require aliquot corrections when an aliquot volume is available and MDAs should also be corrected when necessary. If no aliquot volume is reported, MDAs should be assumed to be dpm/24-hr sample.

Thorium-232 will require aliquot corrections in only a few instances. If <sup>230</sup>Th, <sup>232</sup>Th, or uranium urinalysis data are ever reported in cpm or cph, rather than the standard dpm units, correction factors for detector and extraction efficiencies are presented below in Table 5-19.

Extraction efficiencies should be used to correct data reported in cpm or cph. Protactinium-231, <sup>230</sup>Th, and uranium data have been corrected for chemical recovery in the ORAU database (ORAU 2003e). However, if any data for these radionuclides are reported in cpm or cph, extraction efficiencies of 78%, 90% and 80% should be used for <sup>231</sup>Pa, <sup>230</sup>Th, and uranium respectively. PORECON database entries for <sup>210</sup>Po must first be normalized assuming 86% extraction efficiency and then corrected for a chemical recovery 10% prior to 1964 and 63% after 1964.

### 5.3.4 Summary of Analytical Methods and Detection Limits

This section presents information required to ensure proper evaluation of bioassay data as well as a means to assign missed dose. Table 5-20 summarizes radionuclide analytical methods and MDAs by time period for urinalysis results.

### 5.4 SECONDARY BIOASSAY DETECTION DATA

This section discusses secondary bioassay data to support estimation of radionuclide intakes and dose calculations, including fecal analyses, *in vivo* chest counting, and air sampling. These sources of bioassay information were considered secondary due to limited availability and likely uncertainty. Air-sampling data must be demonstrated to be representative of breathing zone concentrations to be used effectively in dose reconstruction.

Table 5-19. Summary of correction factors and excretion equations.

Nuclide	Period	Primary data type	Aliquot correction	Detector efficiency <sup>a</sup>	Extraction efficiency <sup>b</sup>
Po-210 <sup>c</sup>	1944–1963	cpm or Bq/d	no	no	10
	1964–1973	cpm or Bq/d	no	no	63
	<i>Daily Excretion Po-210 = Po Database Bq/d × 86 % ÷ 10 % (1944 – 1963)</i> <i>Daily Excretion Po-210 = Po Database Bq/d × 86 % ÷ 63 % (1964 – 1973)</i>				
Pu-238 Pu-239	1957–1960	dpm	no	no	10
	1961–1966	dpm	no	no	no
	1967–5/81	dpm	no	no	no
	6/81–1995	dpm	no	no	no
	<i>Daily Excretion Pu = Pu Database dpm × 85 % ÷ 10 % (1957 – 1960 )</i> <i>Daily Excretion Pu = Pu Database dpm (1961 – Present )</i>				
Pa-231	8/17/55–1956	dpm	yes	no	no
	1957–1958	dpm	yes	no	no
	1959–12/759	dpm	yes	no	no
	<i>Daily Excretion Pa-231 = Thorium Database dpm × <math>\frac{\text{Sample ml}}{\text{Aliquot ml}}</math></i>				
Th-230	Prior 1/94	dpm	yes	no	no
	1/94–1995				
<i>Daily Excretion Th-230 = Database dpm × <math>\frac{\text{Sample ml}}{\text{Aliquot ml}}</math></i>					
Th-Natural	Prior 12/87	dpm	yes	no	92
	12/87–1994				
	1/94–1995				
	<i>Daily Excretion Ra-224 = Ra Database dpm ÷ 90 %</i> <i>Daily Excretion Natural Thorium = Th Database dpm ÷ 92 %</i>				
Ra-226 Ac-227 Th-228	1952–1960	cph	no	50	90
	<i>Daily Excretion of Ra-226 = Ra-226 Database cph ÷ 90 % × <math>\frac{2 \text{ dpm}}{60 \text{ cph}}</math></i>				
	<i>Daily Excretion of Ac-227 = Ra-223 Database cph ÷ 90 % × <math>\frac{2 \text{ dpm}}{60 \text{ cph}}</math></i>				
	<i>Daily Excretion of Th-228 = Ra-224 Database cph ÷ 90 % × <math>\frac{2 \text{ dpm}}{60 \text{ cph}}</math></i>				

Nuclide	Period	Primary data type	Aliquot correction	Detector efficiency <sup>a</sup>	Extraction efficiency <sup>b</sup>
Uranium	1950s–1985	dpm	yes	no	no
	Prior 1985				
	5/85–1995				
<i>Daily Excretion Uranium Isotopes = Database dpm × <math>\frac{1400 \text{ ml}}{\text{Aliquot ml}}</math></i>					
Am-241 Cm-244	1950–1989	pCi	no	no	no
	Prior 7/89				
	7/89–1995				
<i>Daily Excretion Am-241 or Cm-244 = Database pCi</i>					

- Detector efficiencies of 50% should be used to correct any data in cpm or cph.
- Assume that intake of radium-223 and radium-224 were in equilibrium with actinium-227 and thorium-228 respectively.
- Polonium-210 database values must be normalized for existing 86% extraction and then corrected to 10% and 63%.

Table 5-20. Analytical methods and detection limits by time period.

Nuclide	Method/description	Period	MDA
H-3 (hot gas)	No urinalysis conducted	1948–1956	Use 95% Dose 340 mrem
	Vibrating reed electrometer	1957–1961	1.0 $\mu\text{Ci L}^{-1}$
	Liquid scintillation	1962–1995	0.01 $\mu\text{Ci L}^{-1}$
Po-210 (Postum)	Spontaneous electrodeposition from raw urine, low background proportional counting.	1944–1963	Spot samples 6.0 <sup>a</sup> dpm 168 dpm/d 24-hr samples 1.3 <sup>a</sup> dpm-24 hr
	Spontaneous electrodeposition from acidified urine, low background proportional counting.	1964–1973	Spot samples 0.72 <sup>a</sup> dpm 20 dpm/d 24-hr samples 0.15 <sup>a</sup> dpm-24 hr
Pa-231	Radium extraction and activity inferred from <sup>227</sup> Th or <sup>223</sup> Ra dpm assuming secular equilibrium.	8/17/55–1956	0.3 dpm
		1957–1958	0.02 dpm
		1959–12/7/59	0.08 dpm
Pu-238, Pu-239	Alkaline earth phosphate coprecipitation, Pu separation with cerium carrier gross alpha proportional counting.	Before 1957	(0.85) pCi/sample <sup>b</sup>
		1957	(0.94) pCi/sample <sup>b</sup>
		1958	(0.6) pCi/sample <sup>b</sup>
		1959	(0.5) pCi/sample <sup>b</sup>
		1960	(0.5) pCi/sample <sup>b</sup>
	Above with sample acidification.	1961–1966	0.05 pCi/sample
	Above with anion exchange, electrodeposition, gross alpha proportional counting.	1967–May 1981	0.11 pCi/sample
	Above with alpha spectroscopy for Pu-238 and Pu-239/240, Pu-242 tracer, blanks counted, detection limits quantified.	Jun–Dec 1981	0.066, 0.044 pCi/sample
		1982	0.063, 0.042 pCi/sample
		1983	0.064, 0.043 pCi/sample
		1984	0.066, 0.045 pCi/sample
		1985	0.047, 0.031 pCi/sample
		1986	0.042, 0.028 pCi/sample
		1987	0.022, 0.011 pCi/sample
		1988	0.017, 0.01 pCi/sample
Jan–Jul 1989		0.019, 0.008 pCi/sample	
Aug 1989–Dec 1991	0.018, 0.008 pCi/sample		
1992–present	0.018, 0.009 pCi/sample		
Th-230 (Ionium)	Radium separation, intakes and excreted Ra-226 and Th-230 assumed to be in equilibrium.	Prior 1/94	0.3 dpm
	Th-230 extraction, alpha spectroscopy.	1/94–1995	0.17 dpm
Th-natural	Thorium separation, proportional counting.	Prior 12/87	0.3 dpm
		12/87–1994	0.02 (Th-232) dpm
	Thorium separation, alpha spectroscopy.	1/94–1995	0.08 (Th-228) dpm
Uranium	LiF fusion, fluoroscopy.	1950s–1985	0.4 $\mu\text{g/l}$
	Uranium separation, proportional counting.	Prior 1985	0.3 dpm
	Uranium separation, alpha spectroscopy.	5/85–1995	0.07 (U-234) dpm 0.03 (U-235) dpm 0.09 (U-238) dpm
Am-241, Cm-244	Americium or curium separation, proportional counting.	1950–1989	0.2 pCi
		Prior 7/89	0.3 pCi
	Americium or curium separation, alpha spectroscopy.	7/89–1995	0.05 pCi

- a. Polonium-210 spot and 24-hr sample MDAs were first normalized assuming 86% extraction efficiency and then corrected for a 10% chemical recovery from 1944-1963 and 63% from 1964-1973. No aliquot size corrections were made for polonium.
- b. Plutonium MDAs from before 1957 to 1960 were normalized assuming 85% extraction efficiency and then corrected for 10% chemical recovery due to plating.

### 5.4.1 Fecal Sample Analysis

Average percent recoveries for plutonium, uranium, thorium, and americium for fecal samples were determined from an analysis of fecal sample data collected from July to October 1989 ORAU files (ORAU 2003c). Table 5-21 summarizes these chemical recoveries as determined using Plutonium-242 (<sup>242</sup>Pu), Uranium-232 (<sup>232</sup>U), Thorium-229 (<sup>229</sup>Th), and Americium-243 (<sup>243</sup>Am) as tracers. Specific MDAs for these fecal analyses could not be quantified without reported background sample counts. However, the MDAs appeared to range from 0.01 to 0.04 pCi/sample for <sup>238</sup>Pu and from 0.001 to

0.008 for <sup>239</sup>Pu, assuming that reported error terms associated with the sample activities were 1 sigma.

Table 5-21. Radionuclide chemical recoveries for collected fecal samples.

Nuclide	Samples	Average	Minimum	Maximum
Pu-242	27	0.77	0.32	1.02
U-232	9	0.69	0.13	1.05
Th-229	2	0.37	0.74	1.03
Am-243	2	0.77	0.71	0.82

#### 5.4.2 Whole Body Counting

Whole body counting has most likely been performed since 1960 when the Whole Body Counting facility was completed. However, whole body counting to assess accidental lung deposition of <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>241</sup>Am began in 1969. Chest counting began in the early 1970s using Phoswich detectors.

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

Reported 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 (File Records-2 1990). Personnel records from 1994 reported MDAs associated with chest counting of selected individuals and 171 counts (File Records-3 1994). Table 5-22 summarizes an evaluation of these reported MDAs by radionuclide and current MDAs.

Table 5-22. Reported 1994 chest counting MDAs (nCi).<sup>a</sup>

Radionuclide	MDA range (nCi)	MDA mean ± SD (nCi)	Current MDAs <sup>b</sup> (nCi)
Am-241	< 0.92	0.3 ± 0.1	0.10
Pu-240	-- <sup>c</sup>	--	47 <sup>d</sup>
Pu-239	9–4910	568 ± 842	130 <sup>d</sup>
Pu-238	0.6–1870	214 ± 318	58
Th-234	0–4.35	1.8 ± 0.8	--
Th-232	--	--	31
Th-228	--	--	3.2
U-238	--	--	1.1
U-235	--	--	0.10
U-234	--	--	30

a. Chest counting MDAs assumed a 1.9-cm chest wall thickness.

b. Current MDAs were taken from ORAU (2003).

c. = none.

d. For bioassay samples it would not be possible to differentiate between <sup>239</sup>Pu and <sup>240</sup>Pu due to identical energy peaks at 13.6 keV. Separate MDAs were reported by ORAU (2003).

MDAs are 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  $^{239}\text{Pu}$  (Langham 1962). *In vivo* chest counting was therefore conducted as a secondary indication of worker contamination in conjunction with primary routine bioassay urinalysis. In addition, MDA data was quite limited and unavailable prior to 1990. Dose reconstruction should therefore be based on urine bioassay results to assess missed worker doses. Chest counting data should only be used to assess intake when counting was conducted as a follow-up for a known worker intake.

### **5.4.3 Personnel Air Sampling Data**

A complex array of particulates, aerosols, dusts, and fumes was present during Mound operations. Representative area or personnel air sampling of these airborne materials would require sophisticated sampling equipment, procedures, and calibration efforts. NRC Regulatory Guide 8.25 requires stringent acceptance criteria to demonstrate that such air sampling is representative of breathing zone air (NRC 1992). In addition, area air sampling cannot be used effectively in some workplace scenarios and can greatly underestimate actual worker inhalation intakes. Obtaining air sample data would probably entail a time-consuming search of archived site records.

Personnel breathing zone 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 breathing zone concentrations. In addition, the DOE claims data do not generally provide air concentration data for nuclides other than tritium.

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**ATTACHMENT A**  
**LUNG SOLUBILITY RECOMMENDATIONS FOR SITE PROCESS ACTIVITIES**  
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This attachment lists lung solubility site specific recommendations for Mound process activities. A claimant-favorable solubility should be used if a specific solubility type cannot be determined. In addition, solubility can be determined based on claim-specific intake or bioassay data and therefore cannot be definitely stated in the TBD. Table A-1 summarizes this information by building. Table A-1 information was largely qualitative in nature and derived almost entirely on King (1995).

Table A-1. Recommended lung clearance types for site process activities.<sup>a</sup>

Building	Site process activities	Lung clearance
HH Building	Hydrolysis of irradiated bismuth slugs. Gaseous diffusion and isotope separation activities. Th-230 extraction operations. Pa-231 separation from Sperry Cake.	
	Po-210 Separation Program: Hydrolysis of irradiated bismuth and aluminum chloride solutions to produce gelatinous bismuth-oxychloride sludge. Sludge shipped off site 4/1954.	Po-210: F or M
	Pa-231 Separation Program: Extraction of Pa-231 from Sperry Cake. Extraction of Th-230 and Pa-231 from ore residues.	Pa-231: M Th-230: M
	Ionium Program: Extraction of Th-230 from Airport Cake raffinate during. Raffinate from uranium processing. Expected carbonate and sulfate compounds.	Th-230: M
	Isotope Separation Program: Gaseous thermal using H-3 waste gas. Xenon isotope separation.	(ICRP 1979)
PP Building	Primarily processing of Pu-238 dioxide from Savannah River Site feed materials. Various plutonium compounds used and soluble compounds reported. Various lung clearance types and particle sizes reported. Processing in rooms PP-144, PP-147 and PP-148 and PP-162 and PP-163.	
	Sol Gel processing with soluble EDTA complexes: No heat, vaporization, crushing, or grinding involved in process.	Pu isotopes: M
	Microsphere pre-processing: Majority of particles reported as submicrometer from sieving and sintering operation. Soluble Pu compounds reported.	Pu isotopes: Oxides M Soluble F
	Microsphere Final Production: High temperature plasma processing, Pu condensation aerosols from plasma torch operations.	Pu isotopes: S
	PMC Process: Microsphere production with crushing and sieving. Particle range of product reported as 43-250 µm. Mo used to coat Pu microspheres. Pu machining used.	Pu isotopes: S
	PPO Process: Microsphere production with crushing and sieving. Hot press used to encapsulate.	Pu isotopes: S
	Solid Solution Process: Th nitrate added to a 9:1 Pu/Th mixture. Crushing, sieving, and hot pressing used in process.	Pu isotopes: M Th isotopes: M
R Building	Research and Development laboratories. Use of primarily Po-210, Pu-238/239, H-3, Ac-227, Th-230, Ra-226/228, Pa-231, and U-234 in neutron source and microsphere production, reactor fuel studies, Ionium Program, isotope separation programs, and limited H-3 recovery.	
	Neutron Source Program: Po-210 source processing. Electroplate deposition and precipitate processes. High vapor pressures. Submicrometer particles reported.	Po-210: F or M
	Microsphere Processing: Pu-239 microspheres with 43-250 µm sizes from PP Building were encapsulated.	Pu isotopes: S
	Radium-Actinium Separation: Ac-227 separated from irradiated radium-bromide-226 and K65 ore residues in SW-19 Old Cave. Actinium Processing: Precipitates in SW-19 processed. Soluble Ac reported in precipitates.	Ra isotopes: M Ac-227: F or M

**ATTACHMENT A**  
**LUNG SOLUBILITY RECOMMENDATIONS FOR SITE PROCESS ACTIVITIES**  
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Building	Site process activities	Lung clearance
R Building (cont'd.)	Protactinium Separation: Pa-231 separation from Sperry Cake.	Pa-231: M
	U-234 Separation: Oxalate precipitation followed by ion exchange to separate U-234 from aged Pu-238. Shipped to Oak Ridge National Laboratory.	U isotopes: M Pu isotopes: M
	Pu Reactor Studies: Fuel studies using molten materials and mixtures.	Pu isotopes: M
	Ionium Program: Separation of Th-230 from Sperry Cake raffinate. Depleted U and Th isotopes reported for feed material compared to Cotter Concentrate. Possible Th-228, Th-232 present.	Th isotopes: M
	Metal Tritide Program: Production and studies of metal tritides including uranium tritides.	Metal tritide: S U tritides: S
	H-3 recovery operations: Recovery of H-3 from R Building operations.	(ICRP 1998)
SM Building	Received powdered Pu dioxide and liquid Pu nitrate from Savannah River Site as feed materials. Microsphere and Plutonium metal production. U-234 separation. Heat source projects.	
	Pu Metal Production: Liquid Pu nitrate and powdered Pu dioxide from Savannah River converted to Pu metal for SNAP. Some Pu compounds reported as soluble. Submicrometer to 20 m reported for various processing.	Pu isotopes: Oxides S All others M
	Microsphere Production: High temperature plasma processing of submicrometer particles, Pu aerosols and fumes from plasma torch operations. Majority of particles reported as submicrometer from sieving and sintering operation.	Pu isotopes: S
	Aqueous Waste Recovery: Pu isotopes recovered from high-risk wastewater. Wastewater equipment removed. Soluble Pu compounds reported.	Pu isotopes: M
	U-234 Separation Program: Separation and recovery of U-234 from aged Pu-238 sources using tributyl solvent extraction and ion exchange. Shipped to Oak Ridge National Laboratory.	U-234: M
SW Building	Research and Development laboratories. Neutron source program support. Thorium refinery program. H-3 recovery and research. Pa-separation and purification. Radium and Actinium separation. Th-229 and U-234 separation activities.	
	Metal Tritide Program: Production and studies of metal tritides including uranium tritides. Tritium Program: Thermal diffusion and enrichment, research, waste classification, and effluent capture. Remaining diffusion columns.	Metal tritide: S U tritides: S (ICRP 1998)
	U-234 Separation Program: Separation and recovery of U-234 from aged Pu-238 sources using tri-butyl solvent extraction and ion exchange. Shipped to Oak Ridge National Laboratory.	U isotopes: M Pu isotopes: M
	Thorium Refinery Program: Extraction of thorium and radium from Brazilian monazite sludge (Monex). Oxalate and sulfate compounds reported. Some feed material could have been oxides or hydroxides due to digestion at high temperatures.	Th isotopes: M Ra isotopes: M
	Pa-231 Purification: Processing of Cotter Concentrate residues to recover Th-230 and Pa-231. Use of organic phosphates. Th-230 had 95% of total activity. Reported soluble compounds.	Th isotopes: M Ra isotopes: M Th-230: M Pa-231: M
	Radium and thorium from activities involving production and separation of Ac-227.	Th isotopes: M Ra isotopes: M
	Radium-Actinium Separation: Ac-227 separated from irradiated radium-bromide-226 and K-65 ore in SW-19 Old Cave. Actinium Processing: Precipitates from SW-19 processed in New Cave. Highly soluble Ac compounds reported. Ac-227 production activities conducted without radium.	Ra isotopes: M Ac-227: F or M
	Thorium-229 Separation: Recovery of Th-229 from aged U-238 and U-233 using a chloride complexation process.	Th isotopes: M U isotopes: M
	Neutron Source Program: Po-209 and Po-210 recovery. Source manufacturing of Pu-238/Be and Pu-239/Be sources. Soluble Po compounds reported.	Po-210: F or M Pu isotopes: S Am-241: S

**ATTACHMENT A**  
**LUNG SOLUBILITY RECOMMENDATIONS FOR SITE PROCESS ACTIVITIES**  
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Building	Site process activities	Lung clearance
T Building	Research and development laboratories. Extensive H-3 facilities and operations. Po-210 separation and fuels research. Neutron source program. Source storage operations.	
	Tritium Program: Thermal diffusion facility, H-3 and HTO recycle and enrichment system, H-3 effluent reduction facility, H3 isotope separation system, H-3 engineering and development laboratory. Gas preparation systems for large quantities of H-3 (T-48, T-49, T-50). Cryogenic distillation columns with U capture beds. Uranium tritides, tritiated water, and tritium gas all present.	(ICRP 1998) Metal tritide: S U tritides: S
	Po-210 Separation Program: Dissolution of Al cans with irradiated Bismuth slugs. Processing and separation of Po-210. Storage of irradiated Bi slugs in T236, 237 pool. Soluble Po compounds reported.	Po-210: F or M
	Polonium Fuels Research: Po metal coupled with rare earths in metal matrix. Particle size of < 44 m reported in T260.	Po isotopes: S
	Source Storage and Calibration: Storage of sealed orphan sources at T300A. Calibration of U Mill Tailings surveillance monitors in T-229.	Ra-226+D: M
	Neutron Source Program: Source manufacturing with Po-210 and Pu isotopes coupled with Be. Source testing and calibration. Soluble Po compounds reported.	Po-210: F or M Pu isotopes: S
	WD/WDA Building <sup>b</sup>	Treatment of liquid radioactive wastewater. Treatment systems for alpha and beta emitters. Polonium treatment process. Cotter Concentrate acid waste treatment. H-3 wastewater treatment. Alpha emitter treatment facility. Effluent monitoring.
	Monex Sludge Storage: Storage of Th sludge from Cotter Concentrate processing. No processing in WD Building.	Th isotopes: M Ra isotopes: M
	Effluent Monitoring: Monitoring of wastewater effluents and treatment processes. Th-230 from Cotter processing. Plutonium decontamination activities contributed. Overflow contamination event with Po-210.	Po-210: F or M Pu isotopes: S Th isotopes: S
	Cotter Waste Treatment: Treatment of acidic wastewater from processing of Cotter Concentrate.	Th isotopes: S
	Tritium Waste Treatment: Influent holding tanks in WDA -9.	(ICRP 1998)
	Polonium Treatment: Filtration using 1 m in WD-1. Drumming of sludge, shipment offsite. Soluble Po compounds reported.	Po-210: F or M
	Plutonium Waste Treatment: Waste treatment facility. High-level wastewater from SM and PP contained alpha emitters. Acidic solutions of Pu isotopes, but primarily Pu-238. High risk drumming station.	Pu isotopes: S
	Alpha Waste Treatment: High-level non-plutonium alpha emitters in wastewater from R and SW. Primarily Po-210. Tritium and Pu-238 secondary contaminants.	Po-210: F or M Ac-227: M Pa-231: M

- a. Source: King (1995).
- b. WD/WDA received wastewater from other buildings, so specific information on form is unavailable. Use generic assumptions for nuclides when no specific information on process is available.
- c. Assume tritium (H-3) is tritiated water vapor (HTO) unless specified otherwise. Assume 3% organically bound 3H with T1/2 = 40 d (ICRP 1998).
- d. External irradiation from xenon and krypton dominates worker dose, so internal dose should be disregarded as stated in ICRP 30 (ICRP 1979).

**ATTACHMENT B**  
**SECONDARY RADIONUCLIDES BY LOCATION AND PERIOD**  
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Table B-1 summarizes locations and dates of use for secondary radionuclides; the information is based on MD-22153, *Mound Site Radionuclides by Location* (King 1995). Secondary radionuclides are listed alphabetically by building for all rooms.

Radionuclides listed in Table B-1 should be considered of major concern only where the Comment column states that a primary or major concern exists. It should be noted that other primary or secondary radionuclides might have been present in a building and room along with each radionuclide listed in Table B-1. For example,  $^{227}\text{Ac}$  was listed as used in room R-140, but  $^{226}\text{Ra}$  and  $^{231}\text{Pa}$  were the primary radionuclides of concern. Therefore,  $^{227}\text{Ac}$  was not a primary radionuclide of concern.

**ATTACHMENT B  
SECONDARY RADIONUCLIDES BY LOCATION AND PERIOD**

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Table B-1. Secondary radionuclides by location and period.

Building	Rooms	Isotopes	MD-22153 reference	Start year	End year	Comments
R	R crawl space	Ac-227	Table 3	1948	Present	Crawl space above R building.
R	R-113, 114, 115	Ac-227	Table 3	1951	1955	Ac-227 as nitrate, oxide, and oxalate, reportedly all soluble. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-118	Ac-227	Table 3	1955	1955	Soluble actinium as nitrate, oxalate, and oxide. Storage Area. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 major hazard in 1956-1958.
R	R-118, 119, 121, 123	Ac-227	Table 3	1955	1955	Soluble actinium as nitrate, oxalate, and oxide. Storage Area. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 the major hazard in 1956.
R	R-120	Ac-227	Table 3	1951	1953	Actinium-227 processing. Actinium as nitrate, oxalate and oxide, chloride, fluoride. Also rooms 118, 119, 121, 123. 1951-1953: Ac-227 primary hazard with Ra-226 and Th-228 secondary concerns. Th-230 primary concern from 1956-1958.
R	R-129, 130	Ac-227	Table 3	1951	1953	Actinium-227 processing. Actinium oxide, nitrate, and oxalate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-133	Ac-227	Table 3	1951	1953	Actinium as nitrate, oxide, oxalate, fluoride, iodide. Th-230 major hazard 1956-1958.
R	R-137	Ac-227	Table 3	1951	1953	Reduction of actinium. Ac as oxide, nitrate, oxalate, fluoride, iodide. Ac-227, Ra-226, and Th-228 major hazards from 1951-1953.
R	R-140	Ac-227	Table 3	1950	1969	Analytical lab for R-120 work. Pa-231 and Ra-226 primary hazards. Am-241 secondary significance in late 1960s.
R	R-142	Ac-227	Table 3	1949	1953	Radium-actinium separation. Ac oxalate, fluoride, radium bromide, nitrate, and carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-145, 147	Ac-227	Table 3	1949	1954	Radium-actinium separation. Radium carbonate and nitrate. Ac-227 major hazard followed by Ra-226 and Th-228. Pa-231 separation from raffinate. Actinium oxalate, oxide. Pa-231 major hazard from 1955-1956.
R	R-148, 149	Ac-227	Table 3	1949	1954	Reduction and separation of actinium. Radium carbonate. Actinium nitrate, oxalate, oxide. Uranium nitrate. Ac-227 major hazard followed distantly by Ra-226 and Th-228. Separation of Pa-231 from residues. U-234 separation from Pu-238 from 1954-1958 & 1970-1980.
R	R-152	Ac-227	Table 3	1951	1953	Actinium oxide, nitrate, oxalate, fluoride, iodide.
R	R-160	Ac-227	Table 3	1951	1953	Actinium separation. Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-161	Ac-227	Table 3	1951	1953	Purification of Actinium, Ac-227 as oxide, nitrate, oxalate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-169	Ac-227	Table 3	1960	1980	Low level counting room. U-238 separation research from 1980-present.
R	R-171, 172	Ac-227	Table 3	1965	1991	Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. U-235, 238 primary concerns: 1961-1963. Am-241 also present 1965-1991.
R	R-171, 172	Ac-227	Table 3	1949	1954	Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. U-235, 238 primary concern: 1961-1963.
R	R-198	Ac-227	Table 3	1956	1982	Numerous isotopes. Decontamination lab. Ac-227 hazard was secondary to Pu-238. Am-241 present 1956-1982.
R	R-Corr-6	Ac-227	Table 3	1948	1984	Access corridor for hot side of R-Building. Ac-227 followed by Ra-226 and Th-228.
SW	SW-128, 129, 130, 134	Ac-227	Table 5	1970	1979	Support rooms. No processing. Th-230 was major radiological hazard.
SW	SW-132	Ac-227	Table 5	1970	1979	Recovery of Pa-231 and Th-230 from Cottler concentrates. Actinium as oxides or nitrates.
SW	SW-136, 137	Ac-227	Table 5	1964	1979	Provide access port to hot cell in SW-140 hot cell. Actinium as nitrate and oxide. Major concerns Th-228, Ra-226, Ac-227, Pa-231, and U-234, U-235, U-238.

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**SECONDARY RADIONUCLIDES BY LOCATION AND PERIOD**  
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Building	Rooms	Isotopes	MD-22153 reference	Start year	End year	Comments
SW	SW-140	Ac-227	Table 5	1974	1979	Hot cell facility. Actinium processing. Recovery of Pa-231 and Th-230 from Cotter concentrates. Ac-227, Pa-231, Th-230 major radionuclides.
SW	SW-140	Ac-227	Table 5	1974	1979	Hot cell facility. Actinium processing. Recovery of Pa-231 and Th-230 from Cotter concentrates. Ac-227, Pa-231, Th-230 major radionuclides.
SW	SW-142	Ac-227	Table 5	1950	1961	Radium as nitrate, carbonate, bromide. Analytical laboratory. Actinium as fluoride and oxalate. U as deuteride and tritide. Major nuclides: Ra-226, Ac-227, Th-228. Metal tritides.
SW	SW-149, 149a	Ac-227	Table 5	1965	1968	All SW Building Nuclides. Decontamination lab. Tritium was major isotope of concern.
SW	SW-19	Ac-227	Table 5	1951	1953	Ac-227 separated from irradiated Ra-226. Radium as carbonate, nitrate, bromide. Ac-227 major hazard followed by Ra-226 and Th-228. Ac as metal, oxalate, fluoride.
SW	SW-22	Ac-227	Table 5	1966	1975	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, 233, Th-228, Th-230 present.
SW	SW-22	Ac-227	Table 5	1970	1979	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, 233, Th-228, Th-230 present.
WD, WDA	WD-1, 8, 101, 104	Ac-227	Table 7	1949	1990	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
WD, WDA	WD-1, 8, 101, 104	Ac-227	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
WD, WDA	WD Penthouse	Ac-227	Table 7	1949	Present	Filter bank for WD and WDA. All radionuclides are a hazard. Am-241 present 1949-present.
R	R-111	Ac-227 & progeny	Table 3	1951	1965	Soluble actinium. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-116	Am-241	Table 3	1956	1962	Production of alpha sources by plating various metal substrates. Am-241 was major concern, Pu-239 secondary.
R	R-120 R-118, 119, 121, 123	Am-241	Table 3	1956	1962	Production of alpha sources by plating various metal substrates. Am-241 was major concern, Pu-239 secondary.
R	R-140	Am-241	Table 3	Late 1960s	Present	Analysis of Pu compounds via chemical analysis and emission spectroscopy. Vaporization of compounds by arcing. Pu-238 major hazard. Am-241 and Ac-227 of equal secondary concern.
R	R-145, 147	Am-241	Table 3	1956	1959	Chemistry studies of reactor fuels. Pu compounds studied using thermal processes. Pu-239 major hazard. Am-241 a distant secondary concern.
R	R-155	Am-241	Table 3	1956	1969	Reactor fuels program. Materials characterized by electron microprobe. U-238 major concern, then Pu-239.
R	R-159	Am-241	Table 3	1956	1969	Reactor Fuel Group experimentation with U and Pu compounds. Physical properties measured for metal alloys. U-238 major concern and Pu-239 secondary.
R	R-167	Am-241	Table 3	1956	1987	Isotopic separation of Pa-231, Pa-233, characterization and analysis of Ce-141, Ce-144, Am-241, Pa-231. Pa-231 major concern with all others a close second.
R	R-171, 172	Am-241	Table 3	1965	1991	Vaporization of oxide solutions by arcing. Pu-238 major concern with all others a distant second.
R	R-198	Am-241	Table 3	1956	1982	Numerous isotopes. Decontamination lab. Pu-238 major concern. Ac-227 secondary.
T	T-5E	Am-241	Table 5	1955	Present	Nondestructive testing using X-ray techniques and radiography. Tritium major concern, Pu-238 secondary, and Pu-239 tertiary concern.
T	T-19, 19A	Am-241	Table 6	1985	Present	Nondestructive X-ray testing. Tritium major concern, Pu-238 secondary, and Pu-239 tertiary concern.
T	T-208A	Am-241	Table 6	1947	Present	Tritium major concern, Pu-238 secondary, and Pu-239 tertiary concern.
PP	PP-146, 150, 151, 152, 153, 154, 156	Am-241	Table 2	1967	1980	Anion exchange recovered Pu via DOWEX-1 resin. Pu dissolved in nitric acid, hydrofluoric acid, hydroxylamine, sodium carbonate. Other radionuclides present, but of minor concern.
SM	SM-34	Am-241	Table 4	1963	1967	Treatment of solid and liquid wastes. Pu primary hazard, other radionuclides minor.

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Building	Rooms	Isotopes	MD-22153 reference	Start year	End year	Comments
SM	SM-38	Am-241	Table 4	1963	1967	Treatment of solid and liquid wastes. Pu primary hazard, other radionuclides minor.
SM	SM-59	Am-241	Table 4	1963	1967	Pu waste recovery. Pu isotopes primary radiological concern, including Pu dioxide, sulfate, nitrate, fluoride.
T	T-310	Am-241	Table 6	1969	1983	Calorimetric measurements and encapsulation of other radionuclides. Pu-238 major concern, H-3 secondary, and Pu-239 tertiary concern.
T	T-319	Am-241	Table 6	1949	Present	Calorimetric measurements and encapsulation of other radionuclides. Pu-238 major concern, H-3 secondary, and Pu-239 tertiary concern.
WD	WD-1	Am-241	Table 7	1949	1990	Materials in aqueous waste. Po-210 major concern, with H-3 and Pu-238 secondary. Pa-231 a distant third.
WD	WD-1	Am-241	Table 7	1961	1974	Low level acidic and basic aqueous wastes Pu wastes. Pu-238 major concern with Pu-239 secondary.
WD	WD-8	Am-241	Table 7	1949	1990	Aqueous waste operations and decontamination of Pu facility. Po-210 major concern, tritium and Pu-238 secondary. Ac-227 and Pa-231 third.
WD	WD-8	Am-241	Table 7	1961	1974	Pu-238 was major concern with Pu-239 secondary. Am-241 present 1949-1990.
WD	WD-9	Am-241	Table 7	1973	Present	Tritium major concern with all other radionuclides secondary.
WD	WD-10	Am-241	Table 7	1961	1974	Aqueous plutonium solutions. Pu-238 major concern with Pu-239 secondary. Room was highly contaminated.
WD	WD-101	Am-241	Table 7	1949	1990	Aqueous waste operations and decontamination of Pu facility. Po-210 major concern, tritium and Pu-238 secondary. Ac-227 and Pa-231 third.
WD	WD-101	Am-241	Table 7	1961	1974	Low-level aqueous plutonium wastes. Pu-238 major concern, then Pu-239.
WD	WD-104	Am-241	Table 7	1949	1990	Aqueous waste operations and decontamination of Pu facility. Po-210 major concern, tritium and Pu-238 secondary. Ac-227 and Pa-231 third.
WD	WD-104	Am-241	Table 7	1961	1974	Low-level aqueous plutonium wastes. Pu-238 major concern, then Pu-239.
WD	WD-107 and basement	Am-241	Table 7.	1976	1981	Ultra filtration and reverse osmosis. All radionuclides in trace amounts and of minor concern.
WD	WD-108	Am-241	Table 7	1949	1981	Highly contaminated stack with filter.
WD	WDA-112	Am-241	Table 7	1966	1979	Techniques developed to separate Pu from wastewater. Pu-238 primary concern with Pu-239 second.
WD	WD-Penthouse	Am-241	Table 7	1949	Present	Filter bank for the building. Any radionuclide in the building potentially present on filters.
SW	SW-219	Am-241	Table 5	1962	1965	Pu neutron source production. Pu-238 was major concern, followed by Pu-239, then Am-241.
T	T-277, 279	Am-241	Table 6	1949	1967	Polonium neutron source program, which used Be, Bo, Li, etc.
HH	HH-5	Pa-231	Table 1	1955	1958	Separation of Pa-231 from raffinates from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake. Pa-231 and Th-230 major nuclides.
HH	HH-6	Pa-231	Table 1	1955	1956	Pilot plant for separation of Pa-231 from Raffinates. Separation of Th-230 from Airport Cake raffinate. Th-230 and Pa-231 predominant nuclides.
R	R-117	Pa-231	Table 3	1948	1990	Storage vault. Pa-231, Th-230, Pu, and Po. Pa-231 was major nuclide before 1955.
R	R-118	Pa-231	Table 3	1956	1958	Soluble actinium as nitrate, oxalate and oxide. Storage Area. Ac-227, Ra-226, Th-228 major hazard in 1955. Th-230 major hazard from 1956-1958.
R	R-118, 119, 121, 123	Pa-231	Table 3	1956	1958	Soluble actinium as nitrate, oxalate and oxide. Storage Area. Ac-227, Ra-226, Th-228 major hazard in 1955. Th-230 major hazard from 1956-1958. Am-241 present 1956-1962.
R	R-120	Pa-231	Table 3	1956	1958	Actinium as nitrate, oxalate and oxide, chloride, fluoride. Also rooms 118, 119, 121, 123. 1951-1953: Ac-227 primary hazard with Ra-226 and Th-228 secondary concerns. Th-230 primary concern from 1956-1958. Am-241 present 1956-1962.
R	R-133	Pa-231	Table 3	1956	1958	Actinium as nitrate, oxide, oxalate, fluoride, iodide. Th-230 major hazard from 1956-1958.
R	R-140	Pa-231	Table 3	1950	1969	Analytical lab for R-120 work. Pa-231 and Ra-226 primary hazards. Am-241 of secondary significance in late 1960s.

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Building	Rooms	Isotopes	MD-22153 reference	Start year	End year	Comments
R	R-142	Pa-231	Table 3	1956	1969	Ac-oxalate, fluoride, radium bromide, nitrate, and carbonate. Radium-actinium separation. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-167	Pa-231	Table 3	1960	1962	Many isotopes. Separation of Pu from irradiated U from 1949-1952. Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
R	R-169	Pa-231	Table 3	1960	1980	Low level counting room. 1980-present: U-238 separation research.
SW	SW-128, 129, 130, 134	Pa-231	Table 5	1970	1979	Support rooms. No processing. Th-230 was major radiological hazard.
SW	SW-132	Pa-231	Table 5	1970	1979	Actinium as oxide or nitrate. Recovery of Pa-231 and Th-230 from Cotter concentrate.
SW	SW-136, 137	Pa-231	Table 5	1964	1979	Actinium as nitrate and oxide. Provide access port to hot cell in SW-140. Th-228, Ra-226, Ac-227, Pa-231, U-234, U-235, and U-238 major nuclides.
SW	SW-140	Pa-231	Table 5	1974	1979	Hot cell facility. Actinium processing of Pa-231 and Th-230 from Cotter concentrate. Ac-227, Pa-231, Th-230 major nuclides.
SW	SW-22	Pa-231	Table 5	1970	1979	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, 233, Th-228, Th-230 present.
WD, WDA	WD-1, 8, 101, 104	Pa-231	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building. All SM Buildings were a concern.
WD, WDA	WD Penthouse	Pa-231	Table 7	1949	Present	Filter bank for WD and WDA. All radionuclides were a hazard. Am-241 present 1949-present.
R	R-167	Pa-231, 233	Table 3	1956	1987	Many nuclides present. Separation of Pu from irradiated U from 1949-1952. Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
SW	SW-19	Ra & progeny	Table 5	1973	Present	Radium as carbonate, nitrate, bromide. Ac-227 separated from irradiated Ra-226. Ac-227 major hazard followed by Ra-226 and Th-228. Actinium as metal, oxalate, fluoride.
R	R-111	Ra-223, 224, 226	Table 3	1951	1965	Soluble actinium. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-118	Ra-223, 224, 226	Table 3	1955	1955	Soluble actinium as nitrate, oxalate and oxide. Storage Area. Ac-227, Ra-226, Th-228 major hazard in 1955. Th-230 major hazard from 1956-1958.
R	R-118, 119, 121, 123	Ra-223, 224, 226	Table 3	1955	1955	Soluble actinium as nitrate, oxalate and oxide. Storage Area. Ac-227, Ra-226, Th-228 major hazard in 1955. Th-230 major hazard from 1956-1958.
R	R-129, 130	Ra-223, 224, 226	Table 3	1951	1953	Actinium oxide, nitrate, and oxalate. Actinium -227 processing. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-140	Ra-223, 224, 226	Table 3	1950	1969	Analytical lab for R-120 work. Pa-231 and Ra-226 primary hazards. Am-241 of secondary significance in late 1960s.
R	R-142	Ra-223, 224, 226	Table 3	1949	1953	Ac-oxalate, fluoride, radium bromide, nitrate, carbonate. Radium-actinium separation. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-169	Ra-223, 224, 226	Table 3	1960	1980	Low level counting room. 1980-present: U-238 separation research.
R	R-171, 172	Ra-223, 224, 226	Table 3	1949	1954	Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. U-235, U-238 primary concern from 1961-1963.
SW	SW-128, 129, 130, 134	Ra-223, 224, 226	Table 5	1970	1979	Support rooms. No processing. Th-230 was major radiological hazard.
SW	SW-132	Ra-223, 224, 226	Table 5	1970	1979	Actinium as oxide or nitrate. Recovery of Pa-231 and Th-230 from Cotter concentrates.
SW	SW-136, 137	Ra-223, 224, 226	Table 5	1964	1979	Actinium as nitrate and oxide. Provide access port to hot cell in SW-140. Th-228, Ra-226, Ac-227, Pa-231, U-234, U-235, and U-238 all present.
SW	SW-140	Ra-223, 224, 226	Table 5	1964	1968	Hot cell facility. Actinium processing. Recovery of Pa-231 and Th-230 from Cotter concentrates. Ac-227, Pa-231, Th-230 major nuclides.
WD, WDA	WD-1, 8, 101, 104	Ra-223, 224, 226	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building. All SM Buildings were a concern. Am-241 present 1949-1990.

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Building	Rooms	Isotopes	MD-22153 reference	Start year	End year	Comments
WD, WDA	WD-9	Ra-223, 224, 226	Table 7	1973	Present	Uranium as deuterides and tritides. Pumps for influent tritiated wastewater. Tritium was primary radiological hazard. Am-241 present 1973-present.
R	R-113, 114, 115	Ra-223, 224, 226, 228	Table 3	1951	1955	Ac-227 as nitrate, oxide and oxalate, reportedly all soluble. Ac-227 major hazard followed by Ra-226 and Th-228.
WD, WDA	WD-Penthouse	Ra-223, 224, 226, 228	Table 7	1949	Present	Filter bank for WD and WDA. All radionuclides are a hazard. Am-241 present 1949-present.
WD, WDA	WD-1, 8, 101, 104	Ra-223, 226	Table 7	1949	1990	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
SW	SW-22	Ra-224, 225	Table 5	1966	1975	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, 233, Th-228, Th-230 all present.
SW	SW-22	Ra-224, 226, 228	Table 5	1970	1979	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, 233, Th-228, Th-230 all present.
SW	SW-11, 12, 13, 14, 15, 16	Ra-224, 228	Table 5	1955	1955	Numerous nuclides. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazard. Pa-231 purification process. Pa-231 major concern, U-238 secondary hazard.
WD, WDA	WD-1, 8, 101, 104	Ra-224, 228	Table 7	1955	1955	Liquid radioactive waste processing. "Low level waste" from SM building. All SM Buildings were a concern. Am-241 present 1949-1990.
R	R crawl space	Ra-226	Table 3	1948	Present	Crawl space above R building.
R	R-120, 118, 119, 121, 123.	Ra-226	Table 3	1951	1953	Actinium as nitrate, oxalate and oxide, chloride, fluoride. Ac-227 primary hazard with Ra-226 and Th-228 secondary concerns from 1951-1953. Th-230 primary concern from 1956-1958.
R	R-133	Ra-226	Table 3	1951	1953	Actinium as nitrate, oxide, oxalate, fluoride, iodide. Th-230 major hazard from 1956-1958.
R	R-137	Ra-226	Table 3	1951	1953	Ac as oxide, nitrate, oxalate, fluoride, iodide. Reduction of actinium. Ac-227, Ra-226, Th-228 major hazards from 1951-1953.
R	R-142	Ra-226	Table 3	1949	1953	Radium-actinium separation. Ac-oxalate, fluoride, radium bromide, nitrate, carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-145, 147	Ra-226	Table 3	1949	1954	Radium-actinium separation. Radium carbonate and nitrate. Actinium oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. Pa-231 separation from raffinate. Pa-231 major hazard from 1955-1956.
R	R-148, 149	Ra-226	Table 3	1949	1954	Radium-actinium separation. Reduction of actinium. Radium carbonate. Actinium nitrate, oxalate, oxide. Ac-227 major hazard followed distantly by Ra-226 and Th-228. Separation of Pa-231 from residues. U-234 separation from Pu-238 from 1954-1958 & 1970-1980. Uranium nitrate.
R	R-152	Ra-226	Table 3	1951	1953	Actinium oxide, nitrate, oxalate, fluoride, iodide.
R	R-160	Ra-226	Table 3	1951	1953	Actinium separation. Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-161	Ra-226	Table 3	1951	1953	Purification of Actinium. Ac-227 as oxide, nitrate, oxalate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-167	Ra-226	Table 3	1956	1987	Many nuclides. Separation of Pu from irradiated U from 1949-1952. Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
R	R-171, 172	Ra-226	Table 3	1965	1991	Actinium separation. Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. U-235, U-238 primary concern from 1961-1963. Am-241 also present 1965-1991.
SW	SW-142	Ra-226	Table 5	1950	1961	Analytical laboratory. Radium as nitrate, carbonate, bromide. Metal tritides. Actinium as fluoride and oxalate. U as deuteride and tritide. Major nuclides Ra-226, Ac-227, Th-228.
SW	SW-19	Ra-226	Table 5	1951	1953	Ac-227 separated from irradiated Ra-226. Radium as carbonate, nitrate, bromide. Actinium as metal, oxalate, fluoride. Ac-227 major hazard followed by Ra-226 and Th-228.

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T	T-19, 19A	Ra-226	Table 6	1985	Present	Encapsulated sources. X-ray and non-destructive testing. Am-241 present 1985-present.
T	T-208A	Ra-226	Table 6	1947	Present	Encapsulated sources. X-ray and non-destructive testing. Am-241 also present.
SW	SW-142	Rn & progeny	Table 5	1950	1961	Analytical laboratory. Radium as nitrate, carbonate, bromide. Metal tritides. Actinium as fluoride and oxalate. U as deuteride and tritide. Major nuclides Ra-226, Ac-227, Th-228.
SW	SW-19	Rn & progeny	Table 5	1951	1953	Ac-227 separated from irradiated Ra-226. Radium as carbonate, nitrate, bromide. Actinium as metal, oxalate, fluoride. Ac-227 major hazard followed by Ra-226 and Th-228.
T	T-229	Rn & progeny	Table 6	1978	1988	Neutron source calibration lab. Other radiological instrument calibration.
WD, WDA	WD-9	Rn & progeny	Table 7	1973	Present	Uranium as deuterides and tritides. Pumps for influent tritiated wastewater. Tritium primary radiological hazard. Am-241 present 1973-present.
R	R-142	Rn, various isotopes	Table 3	1949	1953	Radium-actinium separation. Ac-oxalate, fluoride. Radium bromide, nitrate, carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
WD, WDA	WD-9	Rn & progeny	Table 7	1973	Present	Pumps for influent tritiated wastewater. Uranium as deuterides and tritides. Tritium was primary radiological hazard. Am-241 present 1973-present.
HH	HH-6	Th	Table 1	1955	1956	Pilot plant for separation of Pa-231 from Raffinates. Separation of Th-230 from Airport Cake raffinate. Th-230 and Pa-231 were predominant nuclides.
SW	SW-22	Th-228 & progeny	Table 5	1966	1975	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, U-233, Th-228, Th-230 all present.
SW	SW-22	Th-228, 229	Table 5	1966	1975	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, U-233, Th-228, Th-230 all present.
R	R crawl space	Th-228, 229, 230	Table 3	1948	Present	Crawl space above R building.
R	R-111	Th-228, 229, 230	Table 3	1951	1965	Soluble actinium. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-120, Also 118, 119, 121, 123.	Th-228, 229, 230	Table 3	1951	1953	Actinium as nitrate, oxalate and oxide, chloride, fluoride. 1951-1953: Ac-227 primary hazard with Ra-226 and Th-228 secondary concerns. Th-230 primary concern from 1956-1958. Am-241 present 1956-1962.
R	R-129, 130	Th-228, 229, 230	Table 3	1951	1953	Actinium -227 processing. Actinium oxide, nitrate, and oxalate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-133	Th-228, 229, 230	Table 3	1951	1953	Actinium as nitrate, oxide, oxalate, fluoride, iodide. 1956-1958: Th-230 major hazard.
R	R-137	Th-228, 229, 230	Table 3	1951	1953	Reduction of actinium. Ac as oxide, nitrate, oxalate, fluoride, iodide. Ac-227, Ra-226, Th-228 major hazards from 1951-1953.
R	R-142	Th-228, 229, 230	Table 3	1949	1953	Radium-actinium separation. Ac-oxalate, fluoride, radium bromide, nitrate, carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-145, 147	Th-228, 229, 230	Table 3	1949	1954	Radium-actinium separation. Radium carbonates and nitrates Actinium oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. Pa-231 separation from raffinate. Pa-231 major hazard from 1955-1956.
R	R-148, 149	Th-228, 229, 230	Table 3	1949	1954	Reduction of actinium. Separation work. Radium nitrate and carbonate. Actinium nitrate, oxalate, oxide. Uranium nitrate. Ac-227 major hazard followed distantly by Ra-226 and Th-228. Separation Pa-231 from residues. U-234 separation from Pu-238 from 1954-1958 & 1970-1980.
R	R-152	Th-228, 229, 230	Table 3	1951	1953	Actinium oxide, nitrate, oxalate, fluoride, iodide.
R	R-160	Th-228, 229, 230	Table 3	1951	1953	Actinium separation. Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-161	Th-228, 229, 230	Table 3	1951	1953	Purification of Actinium. Ac-227 as oxide, nitrate, oxalate. Ac-227 major hazard followed by Ra-226 and Th-228.

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R	R-171, 172	Th-228, 229, 230	Table 3	1949	1954	Actinium separation. Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. U-235, 238 primary concern from 1961-1963.
SW	SW-142	Th-228, 229, 230	Table 5	1950	1961	Analytical laboratory. Radium as nitrate, carbonate, bromide. Metal tritides. Actinium as fluoride and oxalate. U as deuteride and tritide. Major nuclides: Ra-226, Ac-227, Th-228.
SW	SW-19	Th-228, 229, 230	Table 5	1951	1953	Ac-227 separated from irradiated Ra-226. Radium as carbonate, nitrate, bromide. Actinium as metal, oxalate, fluoride. Ac-227 major hazard followed by Ra-226 and Th-228.
WD, WDA	8, 101, 104	Th-228, 229, 230	Table 7	1949	1990	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
R	R-118	Th-228, 229, 230, 232	Table 3	1955	1955	Storage Area. Soluble actinium as nitrate, oxalate and oxide. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 major hazard from 1956-1958.
R	R-140	Th-228, 230, 232	Table 3	1950	1969	Analytical lab for R-120 work. Pa-231 and Ra-226 primary hazards. Am-241 of secondary significance in late 1960s.
SW	SW-11, 12, 13, 14, 15, 16	Th-228, 232	Table 5	1955	1955	Numerous nuclides. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazards. Pa-231 purification process. U-238 secondary hazard.
WD, WDA	WD-1, 8, 101, 104	Th-228, 232	Table 7	1955	1955	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
R	R-167	Th-229, 230, 232	Table 3	1958	1958	Many isotopes. Separation of Pu from irradiated U (1949-1952), Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
HH	HH-5	Th-230	Table 1	1956	1958	Separation of Pa-231 from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake raffinate. Pa-231 and Th-230 major nuclides.
R	R-117	Th-230	Table 3	1948	1990	Storage vault. Pa-231, Th-230, Pu, and Po. Pa-231 was major nuclide before 1955.
R	R-142	Th-230	Table 3	1956	1969	Radium-actinium separation. Ac-oxalate, fluoride, radium bromide, nitrate, carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-148, 149	Th-230	Table 3	1970	1980	Reduction of actinium. Separation work. Radium carbonate. Actinium nitrate, oxalate, oxide. Uranium nitrate. Ac-227 major hazard followed distantly by Ra-226 and Th-228. Separation of Pa-231 from residues. U-234 separation from Pu-238 from 1954-1958 & 1970-1980.
R	R-142	Th-230 & progeny	Table 3	1956	1969	Radium-actinium separation. Ac-oxalate, fluoride, radium bromide, nitrate, carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-118	Th-230, 232	Table 3	1956	1958	Storage Area. Soluble actinium as nitrate, oxalate and oxide. Ac-227, Ra-226, Th-228 major hazard in 1955. Th-230 major hazard from 1956-1958. Am-241 present 1956-1962.
R	R-120, Also 118, 119, 121, 123.	Th-230, 232	Table 3	1956	1958	Actinium as nitrate, oxalate and oxide, chloride, fluoride. Ac-227 primary hazard with Ra-226 and Th-228 secondary concerns from 1951-1953. Th-230 primary concern from 1956-1958. Am-241 present 1956-1962.
R	R-133	Th-230, 232	Table 3	1956	1958	Actinium as nitrate, oxide, oxalate, fluoride, iodide. Th-230 major hazard from 1956-1958.
R	R-167	Th-230, 232	Table 3	1956	1987	Many isotopes. Separation of Pu from irradiated U from 1949-1952. Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
R	R-169	Th-230, 232	Table 3	1960	1980	Low level counting room. 1980-present: U-238 separation research.
SW	SW-128, 129, 130, 134	Th-230, 232	Table 5	1970	1979	Support rooms. No processing. Th-230 was major radiological hazard.
SW	SW-132	Th-230, 232	Table 5	1970	1979	Actinium as oxide or nitrate. Recovery of Pa-231 and Th-230 from Cotter concentrates.
SW	SW-136, 137	Th-230, 232	Table 5	1964	1979	Provide access port to hot cell in SW-140. Actinium as nitrate and oxide. Nuclides of concern Th-228, Ra-226, Ac-227, Pa-231, U-234, U-235, and U-238.
SW	SW-140	Th-230, 232	Table 5	1974	1979	Hot cell facility. Actinium processing and recovery of Pa-231 and Th-230 from Cotter concentrates. Major nuclides were Ac-227, Pa-231, and Th-230.

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Building	Rooms	Isotopes	MD-22153 reference	Start year	End year	Comments
SW	SW-22	Th-230, 232	Table 5	1970	1979	New Cave. Th-229 separated from U-233. Actinium nitrate and oxide. Other Th and Pa projects included Cotter Concentrate extraction and U ore processing. U-232, U-233, Th-228, Th-230 present.
WD, WDA	WD-1, 8, 101, 104	Th-230, 232	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building.
WD, WDA	WD Penthouse	Th-230, 232	Table 7	1949	Present	Filter bank for WD and WDA. All radionuclides are a hazard. Am-241 present 1949-present.
R	R-118, 119, 121, 123	Th-230, 232 & progeny	Table 3	1956	1958	Storage Area. Soluble actinium as nitrate, oxalate and oxide. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 major hazard in 1956. Am-241 present 1956-1962.
WD, WDA	WD-1, 8, 101, 104	Th-230, 232	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
38 & PP	147, 148	Th-232	Table 2	1967	1980	Precipitation, crushing, hot-pressing, sintering operations. Thorium used as analogue for Pu. Th-nitrate, oxide, oxalate, hydroxide and fluoride.
38 & PP	Corr-141	Th-232	Table 2	1967	Present	Pu Processing building, Th nitrate, oxalate, and hydroxides.
38 & PP	Corr-142	Th-232	Table 2	1967	Present	Pu Processing bldg. Th nitrate, oxalate, and hydroxides present.
38 & PP	PP-143 (A1)	Th-232	Table 2	1967	1980	Pu Processing Building. Precipitation, crushing, hot-pressing, sintering. Thorium used as analogue for Pu. Th-nitrate, oxalate, and hydroxides.
38 & PP	PP-144 (A2)	Th-232	Table 2	1967	1980	Precipitation, crushing, hot-pressing, sintering. Thorium used as analogue for Pu. Th nitrate, oxalate, hydroxides and insoluble forms.
38 & PP	PP-Corr-140	Th-232	Table 2	1967	Present	Pu building, also Th nitrate, oxalate, and hydroxides.
HH	HH-5	Th-232	Table 1	1956	1958	Separation of Pa-231 from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake raffinate. Pa-231 and Th-230 major nuclides.
R	R-129, 130	Th-232	Table 3	1965	1978	Actinium-227 processing. Actinium oxide, nitrate, and oxalate. Ac-227 major hazard followed by Ra-226 and Th-228.
SM	SM-21	Th-232	Table 4	1961	1967	Analytical Laboratory. Physical measurements. Th used as analogue to Pu. EDTA and citrates as complexing agents. Storage area for radioactive materials.
SM	SM-61, 62	Th-232	Table 4	1961	1967	Analytical labs. Studies of Pu. Th-232 used as an analogue for Pu. Complexing agents EDTA and citrates used with Pu.
R	R-118	Th-232 & progeny	Table 3	1956	1958	Storage Area. Soluble actinium as nitrate, oxalate and oxide. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 major hazard from 1956-1958. Am-241 present 1956-1962.
SW	SW-11, 12, 13, 14, 15, 16	Th-232 & progeny	Table 5	1955	1955	Numerous isotopes. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazards. Pa-231 purification process. U-238 secondary hazard.
SM	SM-1	Th-232, 228	Table 4	1962	1967	Pu facility. Treatment of low risk waste.
HH	HH-6	U	Table 1	1955	1956	Pilot plant for separation of Pa-231 from Raffinates. Separation of Th-230 from Airport Cake raffinate. Th-230 and Pa-231 were predominant nuclides.
SM	SM-34	U	Table 4	1963	1971	U as oxide, fluoride and sulfate. Pu major hazard. Am-241 present 1963-1967.
SM	SM-58	U	Table 4	1963	1967	Pu facility waste recovery. Pu- isotopes were primary radiological concern. U oxide, fluoride, nitrate, sulfate.
SW	SW-11, 12, 13, 14, 15, 16	U	Table 5	1968	Present	Numerous nuclides. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazards. Pa-231 purification process. U-238 secondary hazard.
SW	SW-11, 12, 13, 14, 15, 16	U	Table 5	1959	1978	Numerous nuclides. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazards. Pa-231 purification process. U-238 secondary hazard.
SW	SW-9	U	Table 5	1989	Present	U deuteride, tritide, and hydrides. Tritium was primary radiological hazard.
SW	SW-22	U-232, 233	Table 5	1966	1975	New Cave. Th-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction, processing U ore. U-232, U-233, Th-228, Th-230 all present.

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HH	HH-5	U-233	Table 1	1955	1958	Separation of Pa-231 from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake raffinate. Pa-231 and Th-230 major nuclides.
R	R-167	U-233	Table 3	1956	1987	Many isotopes. Separation of Pu from irradiated U (1949-1952), Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
R	R-167	U-233	Table 3	1958	1958	Numerous nuclides. Separation of Pu from irradiated U from 1949-1952. Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958. Am-241 secondary 1956-1987.
T	T-310	U-233, 235	Table 6	1969	1983	Calorimetry. Pu isotopes and tritium were primary hazard. Am-241 present 1969-1983.
T	T-319	U-233, 235	Table 6	1949	Present	Calorimetry. Pu isotopes and tritium were primary hazard. Am-241 present 1949-present.
R	R-162	U-233, 235, 238	Table 3	1958	1969	Analytical lab. Pu and fission products major concern 1948-1951. U-233, U-235, U-238 major concerns 1958-1959.
HH	HH-5	U-234	Table 1	1955	1958	Separation of Pa-231 from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake raffinate. Pa-231 and Th-230 major nuclides.
R	R-148, 149	U-234	Table 3	1970	1980	Reduction of actinium. Separation work. Radium carbonate. Actinium nitrate, oxalate, oxide. Uranium nitrate. Ac-227 major hazard followed distantly by Ra-226 and Th-228. Separation of Pa-231 from residues. U-234 separation from Pu-238 from 1954-1958 & 1970-1980.
WD, WDA	WD-1, 8, 101, 104	U-234	Table 7	1961	1974	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
WD, WDA	WD-10	U-234	Table 7	1961	1974	High activity influent storage tanks in room. Highly contaminated area. Pu-238 was primary concern. Am-241 present 1961-1974.
WD, WDA	WD-108	U-234	Table 7	1949	1981	Room having stack with filter. Highly contaminated area. Uranium and transuranics present. Am-241 from 1949-1981.
WD, WDA	WDA-110	U-234	Table 7	1966	1970	Alpha emitter wastewater treatment. Pu isotopes were major radiological hazard.
WD, WDA	WDA-112	U-234	Table 7	1966	1979	Extraction of Pu from wastewater. Pu-238 was primary radiological hazard. Am-241 present 1966-1979.
R	R-118	U-234, 235, 238	Table 3	1956	1958	Storage Area. Soluble actinium as nitrate, oxalate and oxide. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 major hazard from 1956-1958.
R	R-118, 119, 121, 123	U-234, 235, 238	Table 3	1956	1958	Storage Area. Soluble actinium as nitrate, oxalate and oxide. Ac-227, Ra-226, Th-228 major hazards in 1955. Th-230 major hazard in 1956. Am-241 present 1956-1962.
R	R-120, also rooms 118, 119, 121, 123.	U-234, 235, 238	Table 3	1956	1958	Actinium as nitrate, oxalate and oxide, chloride, fluoride. 1951-1953: Ac-227 primary hazard with Ra-226 and Th-228 secondary concerns. Th-230 primary concern from 1956-1958. Am-241 present 1956-1962.
R	R-133	U-234, 235, 238	Table 3	1956	1958	Actinium as nitrate, oxide, oxalate, fluoride, iodide. Th-230 major hazard from 1956-1958.
R	R-140	U-234, 235, 238	Table 3	1950	1969	Analytical lab for R-120 work. Pa-231 and Ra-226 primary hazards. Am-241 of secondary significance in late 1960s.
R	R-142	U-234, 235, 238	Table 3	1956	1969	Radium-actinium separation. Ac-oxalate, fluoride, radium bromide, nitrate, carbonate. Ac-227 major hazard followed by Ra-226 and Th-228.
R	R-145, 147	U-234, 235, 238	Table 3	1968	1975	Radium-actinium separation. Radium carbonate and nitrate. Ac-227 major hazard followed by Ra-226 and Th-228. Pa-231 separation from raffinate. Actinium oxalate, oxide. Pa-231 major hazard 1955-1956.
R	R-167	U-234, 235, 238	Table 3	1949	1952	Numerous nuclides. Separation of Pu from irradiated U from 1949-1952. Extraction of Pa-231 from residues from 1954-1955. U-233 major nuclide in 1958.
R	R-169	U-234, 235, 238	Table 3	1960	1980	Low level counting room. 1980-present: U-238 separation research.
R	R-198	U-234, 235, 238	Table 3	1956	1982	Numerous isotopes. Decontamination lab. Ac-227 hazard was secondary hazard to Pu-238. Am-241 present 1956-1982.

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SW	SW-128, 129, 130, 134	U-234, 235, 238	Table 5	1970	1979	Support rooms. No processing. Th-230 was major radiological hazard.
SW	SW-132	U-234, 235, 238	Table 5	1970	1979	Recovery of Pa-231 and Th-230 from Cotter concentrates. Actinium as oxide or nitrate.
SW	SW-136, 137	U-234, 235, 238	Table 5	1964	1979	Provide access port to hot cell in SW-140. Actinium as nitrate and oxide. Concerns were Th-228, Ra-226, Ac-227, Pa-231, U-234, U-235, and U-238.
SW	SW-140	U-234, 235, 238	Table 5	1974	1979	Hot cell facility. Actinium processing and recovery of Pa-231 and Th-230 from Cotter concentrates. Ac-227, Pa-231, Th-230 major nuclides of concern.
SW	SW-149, 149a	U-234, 235, 238	Table 5	1965	1968	All SW Building Nuclides. Decontamination lab. Tritium was major isotope of concern.
SW	SW-22	U-234, 235, 238	Table 5	1970	1979	New Cave. Thorium-229 separated from U-233. Actinium as nitrate and oxide. Other Th and Pa projects including Cotter Concentrate extraction and U ore processing. U-232, 233, Th-228, Th-230.
WD, WDA	WD-1, 8, 101, 104	U-234, 235, 238	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
WD, WDA	WD-1, 8, 101, 104	U-234, 235, 238	Table 7	1949	1990	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
WD, WDA	WD-1, 8, 101, 104	U-234, 235, 238	Table 7	1974	1979	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.
WD, WDA	WD-Penthouse	U-234, 235, 238	Table 7	1949	Present	Filter bank for WD and WDA. All radionuclides are a hazard. Am-241 present 1949-present.
HH	HH-5	U-235	Table 1	1955	1958	Separation of Pa-231 from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake raffinate. Pa-231 and Th-230 are major nuclides.
T	T-236	U-235, U-238, U-239	Table 6	1985	Present	Dock. Tritium was primary hazard from 1985-present.
R	R-169	U-235, 238	Table 3	1980	Present	Low level counting room. U-238 separation research from 1980-present.
R	R-171, 172	U-235, 238	Table 3	1961	1963	Ac as nitrate, oxalate, oxide. Ac-227 major hazard followed by Ra-226 and Th-228. U-235, 238 primary concern from 1961-1963.
WD, WDA	WD-107	U-235, 238	Table 7	1976	1981	Ultra-filtration and reverse osmosis. Th-230 was primary isotope of concern in trace amounts only. Am-241 present 1976-1981.
WD, WDA	WD-9	U-235, 238, 239	Table 7	1973	Present	Pumps for influent tritiated wastewater. Uranium as deuterides and tritides. Tritium was primary radiological hazard. Am-241 present 1973-present.
R	R-155	U-235, U-238	Table 3	1956	1969	Microprobe Lab, 1956-1969. U-238 major radionuclide.
R	R-159	U-235, U-238	Table 3	1956	1969	Numerous other isotopes. U-238 major radionuclide
HH	HH-5	U-238	Table 1	1955	1958	Separation of Pa-231 from 1955-1956. Decommissioned by 1958. Th-230 extraction from Airport Cake raffinate. Pa-231 and Th-230 are major nuclides.
R	R-105, 106	U-238	Table 3	1965	1978	Uranium deuteride and uranium tritide. Tritium gas was the major radiological hazard followed by uranium.
R	R-108	U-238	Table 3	1948	1965	Uranium-deuteride and tritide. Tritium gas was the major radiological hazard followed by uranium.
R	R-112	U-238	Table 3	1973	1993	U-Nat deuteride and tritide. Tritium hazard >> uranium hazard.
R	R-116	U-238	Table 3	1960	1965	Numerous isotopes, U-238 as deuteride and tritide. Tritium hazard >> uranium hazard.
R	R-128	U-238	Table 3	1987	Present	Uranium as deuteride and tritide. Tritium hazard >> Uranium hazard. Am-241 used 1956-1962.
R	R-134	U-238	Table 3	1986	Present	U deuteride and tritide. Tritium primary radiological hazard. Low radionuclide inventory.
R	R-168	U-238	Table 3	1958	1958	Physical properties of U-238 salts and oxide including fluorides. Major hazard was U-238.
R	R-170	U-238	Table 3	1960	1964	U-238 mass spectroscopy.
SW	SW-11, 12, 13, 14, 15, 16	U-238	Table 5	1955	1955	Numerous nuclides. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazards. Pa-231 purification process. U-238 secondary.

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SW	SW-11, 12, 13, 14, 15, 16	U-238	Table 5	1968	Present	Numerous nuclides. Th-232 extraction from monazite sand. Th-232, Th-228, Ra-224 major hazards. Pa-231 purification process. U-238 secondary.
SW	SW-142	U-238	Table 5	1969	1985	Analytical laboratory. Radium as nitrate, carbonate, bromide. Metal tritides. Actinium as fluoride and oxalate. U as deuteride and tritide. Major nuclides Ra-226, Ac-227, Th-228.
SW	SW-146, 147	U-238	Table 5	1967	Present	Uranium as deuteride, tritide. Tritium was radiological concern from 1967-1985.
SW	SW-150, 152	U-238	Table 5	1968	Present	Uranium as deuterides, tritides. Tritium was primary radiological hazard.
SW	SW-208	U-238	Table 5	1965	Present	Uranium as deuterides and tritides. Tritium was primary radiological hazard.
SW	SW-210	U-238	Table 5	1968	1969	Uranium as deuterides and tritides. Tritium was primary radiological hazard.
SW	SW-219	U-238	Table 5	1985	Present	Uranium as deuterides and tritides. Tritium was primary radiological hazard.
SW	SW-238, 240	U-238	Table 5	1968	Present	Uranium-238 and tritium. Tritium was major hazard.
SW	SW-8	U-238	Table 5	1960	1985	Primarily deuterium and tritium research. Tritium was primary radiological hazard.
T	T-266, 267	U-238	Table 6	1988	Present	Tritium engineering & development. Tritium was primary radiological hazard. Uranium as deuterides and tritides from 1988-present.
T	T-274	U-238	Table 6	1986	Present	Uranium as deuterides and tritides. Tritium was primary radiological hazard.
T	T-37	U-238	Table 6	1985	Present	Decontamination area. Provided access to other parts of building. Tritium was primary radiological hazard. Uranium as deuterides and tritides.
T	T-48, 49, 50	U-238	Table 6	1983	Present	Glovebox labs and access corridor. Tritium was primary hazard. Uranium as deuterides and tritides.
T	T-59	U-238	Table 6	1985	Present	Tritium thermal diffusion facility. Tritium was primary hazard. Uranium as deuterides and tritides.
T	T-61, 61a	U-238	Table 6	1973	Present	Uranium as deuteride and tritide. Tritium recovery.
WD, WDA	WD-1, 8, 101, 104	U-238	Table 7	1955	1955	Liquid radioactive waste processing. "Low level waste" from SM building. Am-241 present 1949-1990.

**ATTACHMENT C**  
**CODING INSTRUCTIONS FOR URINALYSIS RECORDS**

The following coding instructions are summarized in Table 5-9 for urinalysis records reported on Urinalysis Cards, Weekly Reports, and 24-hour reports at Mound.

Table C-1. Coding instructions for urinalysis records.

<b>Code</b>	<b>Description</b>
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	Disintegrations / minute
DM-24	Disintegrations / minute / 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 D  
UNCERTAINTY ASSESSMENT**

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**D.0 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 the bioassay measurements were not stated in the records.

**D.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.

**D.2 ASSIGNMENT OF GROSS ALPHA RESULTS AS PLUTONIUM**

Before alpha spectrometry, gross alpha activity based on chemical extractions was administratively assigned to <sup>238</sup>Pu or <sup>239</sup>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 allocated improperly among the organs.

**D.3 MISSING BIOASSAY RECORDS**

The record of urinalysis data is generally complete for the Mound site. Some data are available for the Monsanto Chemical Company facilities operated prior to the beginning of operations at the Mound laboratory as well. Reportable worker exposures in the early years could also have been undetected due to low recovery caused by plating on sample containers before sample analysis. Intakes for missing bioassay results should be estimated using the MDAs reported in Table 5-5 for spot and 24-hr samples.

Tritium bioassay data were not reported from 1948 to 1956. The dose reconstructor was advised to use a value of 1000 mrem corrected to 340 mrem ICRP68 dosimetry for unmonitored workers. This value was assumed to be the approximate upper 95 percentile and considered a claimant-favorable dose for all workers. Uncertainty of extrapolating doses from 1959–1962 to the 1947–1956 time period are considered minor and compensated by use of the upper 95 percentile dose estimate. Doses should not be assigned to administrative or other non-radiological workers.

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### D.4 ASSUMPTION OF EQUILIBRIUM

Dose uncertainty related to degree of equilibrium is potentially important when assigning dose from bioassay for secondary radionuclides. For example, there are indications that  $^{230}\text{Th}$  doses were assigned based on the presumption of equilibrium of  $^{230}\text{Th}$  with  $^{226}\text{Ra}$  in some instances. This could result in errors of up to a factor of 10 where acute intakes of unspecified feed materials such as raffinate, tailings, and various production residues occurred. For instance, raffinate produced by sulfuric acid leaching of ore would have a relatively low concentration of radium compared to thorium due to radium sulfate precipitation. Therefore, the inferred  $^{230}\text{Th}$  intake could be greatly underestimated. The assumption of equilibrium is therefore largely dependent on the true nature of feed materials that workers were exposed to 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  $^{228}\text{Th}$  and  $^{224}\text{Ra}$ , and between  $^{227}\text{Ac}$  and  $^{223}\text{Ra}$ , are less than the  $^{230}\text{Th}$  case because (1)  $^{238}\text{U}$  chain and possible cases of disequilibria are not involved, and (2) both  $^{223}\text{Ra}$  and  $^{224}\text{Ra}$  have short half-lives in days and therefore quickly achieve equilibrium with their respective parent radionuclides.

### D.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 involved. These processes were used in buildings and reconstruction necessitates deviation from the default ICRP Publications 66 and 68 assumption of an AMAD of 5  $\mu\text{m}$  for occupational exposure (ICRP 1994, 1995). Alpha emitters with larger 5- $\mu\text{m}$  particles and lung clearance types S or M tend to have smaller inhalation effective dose coefficients than do smaller 1- $\mu\text{m}$  particles. Use of a smaller AMAD of 1  $\mu\text{m}$  increases worker dose following inhalation of type M  $^{239}\text{Pu}$  by a factor of 1.5.

Of the secondary radionuclides, the difference in dose conversion factors for 1- $\mu\text{m}$  versus 5- $\mu\text{m}$  particles with clearance types S or M was greatest for  $^{231}\text{Pa}$  by a factor of 2. The opposite trend was seen for type F alpha emitters, where 5- $\mu\text{m}$  particles of uranium or  $^{227}\text{Ac}$  have inhalation effective dose coefficients that are 20% larger than corresponding dose coefficients for 1- $\mu\text{m}$  particles. Therefore, uncertainty in dose coefficients related to particle size distribution is dependent on lung clearance type and is comparatively small based on data provided 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.

### D.6 SOLUBILITY AND LUNG CLEARANCE TYPE

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

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### D.7 RADIONUCLIDE USE BY LOCATION

The uncertainty associated with radionuclide use by location was almost entirely documented by King (1995) and was considered minor compared with other sources of uncertainty. Radionuclide use by location has been sufficiently documented for primary radionuclides (Table 5-1). There is little uncertainty associated with documentation for tritium,  $^{210}\text{Po}$ , and the plutonium isotopes and their processes by building. Protactinium-231 had limited use and, therefore, there is 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 to those for natural uranium. Isotopic radium and thorium data were also limited. Almost no information was available for  $^{244}\text{Cm}$  and  $^{237}\text{Np}$  other than as a contaminant in the Savannah River Plant source material.

Information on the location and use of radioactive gases was primarily only available for  $^{222}\text{Rn}$  (radon) from 1979 to the present. Very limited information was available for  $^{220}\text{Rn}$  (thoron) and  $^{219}\text{Rn}$  (actinon), and the measurements were questionable due to excessive decay times before analysis. However, measurements conducted in the underground tunnel in 1979 confirmed the presence of  $^{222}\text{Rn}$  at  $88,000 \text{ pCi L}^{-1}$ ,  $^{220}\text{Rn}$  at  $28,000 \text{ pCi L}^{-1}$ , and  $^{219}\text{Rn}$  at  $640,000 \text{ pCi L}^{-1}$ . 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 4.0 s and direct deposition of actinon daughters 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 compensated for this uncertainty.

### D.8 ANALYTICAL PROCEDURES

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

Systematic errors occurred in analytical procedures in the early years due primarily to ineffective plating techniques and low recoveries. Low recoveries were due largely to colloidal plating of metabolized  $^{238,239}\text{Pu}$  and  $^{210}\text{Po}$  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  $^{210}\text{Po}$  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.

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Additional errors occurred in the early years as a result of the inability to accurately assign gross alpha activity determined with low background proportional counting to specific alpha-emitting radionuclides. Gross alpha activity was thus 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 detection limits.