

<p><b>ORAU Team</b>  <b>Dose Reconstruction Project for NIOSH</b></p> <p>Y-12 National Security Complex – Occupational Internal Dose</p>	<p>Document Number:  ORAUT-TKBS-0014-5  Effective Date: 05/10/2005  Revision No.: 01  Controlled Copy No.: _____  Page 1 of 44</p>
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## RECORD OF ISSUE/REVISIONS

<b>ISSUE AUTHORIZATION DATE</b>	<b>EFFECTIVE DATE</b>	<b>REV. NO.</b>	<b>DESCRIPTION</b>
Draft	10/24/2003	00-A	New document to establish TBD for the Y-12 National Security Complex – Occupational Internal Dose. Initiated by William E. Murray.
Draft	01/02/2004	00-B	Revised document in response to comments from OCAS and formal review on TBD for the Y-12 National Security Complex – Occupational Internal Dose. Initiated by William E. Murray.
Draft	03/05/2004	00-C	Revised document in response to additional comments from OCAS on TBD for the Y-12 National Security Complex – Occupational Internal Dose. Initiated by William E. Murray.
03/17/2004	03/17/2004	00	First approved issue. Initiated by William E. Murray.
Draft	10/15/2004	01-A	First major revision. Initiated by William E. Murray.
Draft	02/14/2005	01-B	Incorporates internal review comments. Initiated by William E. Murray.
Draft	04/13/2005	01-C	Includes table numbers and titles in body of document. Initiated by William E. Murray.
05/10/2005	05/10/2005	01	Approved issue of Revision 01. No retraining required. Initiated by William E. Murray.

## ACRONYMS AND ABBREVIATIONS

AMAD	aerodynamic median activity diameter
Bq	becquerel
CEDE	committed effective dose equivalent
Ci	curie
cpm	counts per minute
d	day
DF	decontamination factor
dpm	disintegrations per minute
DU	depleted uranium (less than 0.72% <sup>235</sup> U)
EU	enriched uranium
g	gram
hr	hour
HEU	highly enriched uranium (generally between 20% and 90% <sup>235</sup> U)
HPGe	high-purity germanium (detector)
ICRP	International Commission on Radiological Protection
in	inches
keV	kilovolt-electron, 1,000 electron volts
kg	kilogram
L	liter
L <sub>C</sub>	critical level (commonly 5% chance of a false positive)
L <sub>D</sub>	detection level (commonly 5% chance of a false negative)
LEU	low enriched uranium (generally less than 20% <sup>235</sup> U)
MDA	minimum detectable activity
mg	milligram
min	minute
mL	milliliter
mm	millimeter
MPBB	maximum permissible body burden
MPLB	maximum permissible lung burden
mrad	millirad
mrem	millirem
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NU	natural uranium
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory

pCi	picocurie
ppb	parts per billion
ppm	parts per million
RWP	Radiation Work Permit
RU	recycled uranium
SMC	Idaho Specific Manufacturing Capability
U.S.C.	United States Code
VHEU	very highly enriched uranium (greater than 90% <sup>235</sup> U)
wt %	weight percent
yr	year
μCi	microcurie
μg	microgram
μm	micrometer

## 5.1 INTRODUCTION

Technical Basis Documents and Site Profile Documents are general working documents that provide guidance concerning 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 the National Institute for Occupational Safety and Health (NIOSH) 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 facility” as defined in the Energy Employee Occupational Illness Compensation Program Act of 2000 [42 U.S.C. Sections 7384l(5) and (12)].

At the Y-12 Plant, now the Y-12 National Security Complex, uranium isotopes in various chemical and physical forms have been the primary contributors to internal radiation doses to workers since November of 1943. Y-12 missions involved other radionuclides and, at times, some uranium compounds could have contained impurities with radiological health implications. These implications and default analyses are discussed in this report. However, the primary focus on internal dose control has been on uranium compounds and alloys over a wide range of <sup>235</sup>U enrichment. Therefore, this section begins with an overview of concepts that apply broadly to the history of uranium work at Y-12. Attachment 5A summarizes some of the key points from the following sections.

There were programs in the early years of Y-12 operations for which the currently available information is insufficient to provide general guidance for internal dose reconstruction. One program was associated with the research engineering and development (RED) group, using a proton-accelerating cyclotron in Building 9201-2, which became operational on November 11, 1950 and operated until 1980. At the peak of U-235 enrichment operations in the 1943-45 time period there were 1156 “large mass spectrographs”, called “calutrons” in operation. This enrichment production technique was shut down in 1946, although the calutrons in bldg 9204-3 were left operational for R&D in improving the production of the calutron concept and separating other element isotopes. Eight calutrons were still operating as late as 1997. This program produced an inventory of 225 isotopes from nearly every element on the periodic chart. The 86 inch Cyclotron started operation in 1950 and continued to operated until 1980. This cyclotron is no longer in operation. Polonium isotopes and alpha airborne activity are the mentioned internal dose concerns. The 86 inch cyclotron was later used to create neutron-deficient radionuclides as a part of the R&D program above. There were programs in these R&D efforts, which involved plutonium, and workers could have plutonium bioassay results in their records. When claim information indicates that a Y-12 worker was involved with research activities involving the Calutron, cyclotron (accelerator), fusion work, or plutonium (except in the case of recycled uranium (RU) exposure, which is addressed in this section), consideration must be given to possible exposure to radionuclides other than uranium. (Wilcox, Wm. J. Jr., et al. 1999)

### 5.1.1 Uranium Solubility in the Lung

The uranium compounds with which Y-12 has worked ranges from highly soluble to very insoluble. Health physicists have long recognized that nephrotoxicity is the primary hazard associated with soluble uranium compounds (depleted through low enrichment). According to Sterner and Riley (1946), exposures to soluble compounds were monitored from the closing days of World War II by clinical tests of renal function and by fluorometric tests for uranium in urine. However, no urinalysis data have been found for Y-12 prior to 1948, and this issue will have to be addressed in a revision of this document.

Health physicists also recognized early that the lung is the primary organ of concern for the less soluble uranium compounds. During the greater part of the plant's history, control measures for less soluble compounds were guided by the lung and metabolic models presented in the International Commission on Radiological Protection (ICRP) Publication 2 (ICRP 1960) or by similar predecessor models. These models related lung dose to uranium excretion in urine, particularly under chronic exposure conditions.

It is noteworthy that insoluble material as described in ICRP Publication 2 would be classified as moderately soluble type M material in the modern ICRP Publication 66 framework (ICRP 1994). It is also noteworthy that ICRP has recognized that some forms of uranium are less soluble than initially believed. Figure 5-1 shows normalized lung retention for ICRP Publication 2 insoluble material, ICRP Publication 30 (ICRP 1979) class W particles, class Q material as described in the 1990s (Barber and Forrest 1995), and the Publication 66 type M particles. The boxes on the diagram define the modern clearance types F (fast), M (moderate), and S (slow) as a function of lung retention and time as illustrated in Annex D of ICRP Publication 71 (ICRP 1995). The ICRP 2 insoluble model and the class W models fit within the range of type M material.

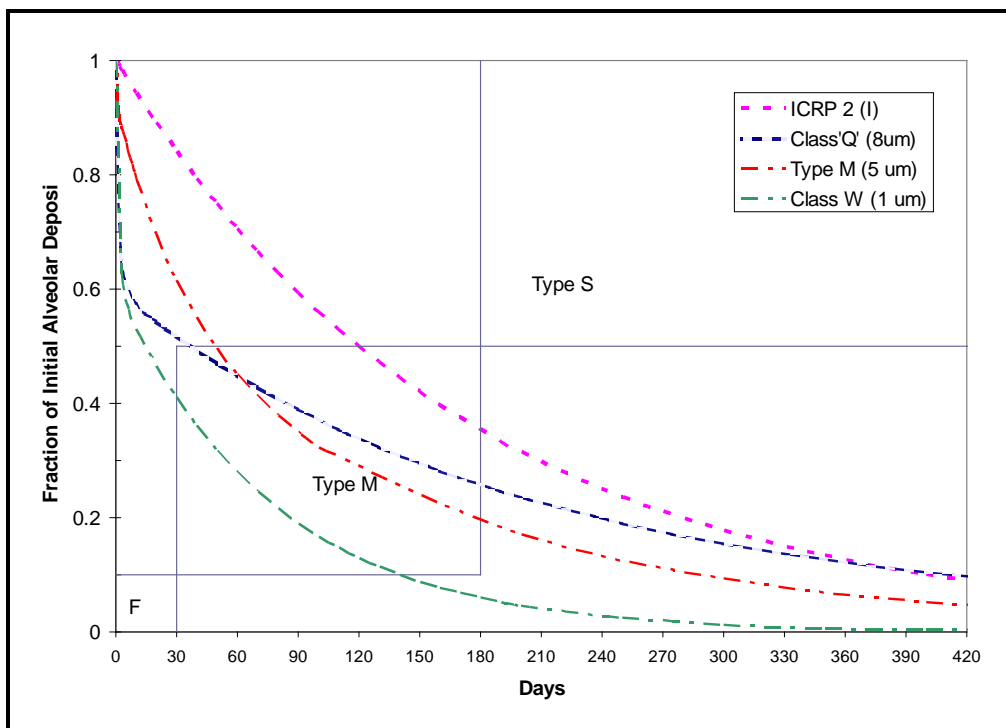


Figure 5-1. Historical solubility types meeting the type M definition.

After *in vivo* lung counting was begun at Y-12 in 1961, it was recognized that a few workers had uranium lung burdens in considerable excess of the amount indicated by urinalysis. Writing in 1969, West and Scott (1969) noted that, "5 exposure cases noted in 1962 and 1963 showed chest burdens of enriched uranium [EU] which exhibited unexpectedly slow decreases after the persons were assigned to non-uranium activities" (West and Scott 1969). The job description of all five individuals was Process Operator (Snapp 2003).

Subsequent investigations suggested that three process combustion ashes from the EU foundry were the most likely exposure materials for these five cases (Steckel and West 1966, p. 31). The particular thermal histories and particle size distributions of these process ashes were considered to be

important factors leading to the low solubility in simulated lung fluid (Steckel and West 1966, p. 30). Particle sizes tended to be relatively larger (0.86 to 1.6  $\mu\text{m}$ ), and the solubility tended to be lower, for materials processed at higher temperatures.

While the exceptional cases with unusually protracted lung clearance are important, it is more important to note that, for the vast majority of individuals, lung clearance took place in approximate accordance with the ICRP Publication 2 (1960) insoluble model, which fits within the current type M framework.

In 1964, a composite urinary excretion curve was described for 157 workers who had been removed from uranium work because of high uranium urinalysis results (Scott 1964). Figure 5-2 shows this empirical composite curve together with expected excretion curves for types F, M, and S uranium. The latter curves are derived from 5- $\mu\text{m}$  aerodynamic median activity diameter (AMAD) excretion curves (Potter 2002). They were normalized to match the Scott curve at 200 d after removal from uranium work areas. The empirical excretion curve matches the type M curve much better than the type F or S curves. Indeed, the Scott curve fits the type M curve quite well over most of the 420-d time range. These results are consistent with the subsequent conclusions of Rucker et al. (2001) that class W is the appropriate default for uranium at Bechtel Jacobs facilities, including Y-12. Exceptions to this default included *high-fired* uranium oxides and compounds formed by the slow oxidation of uranium metal.

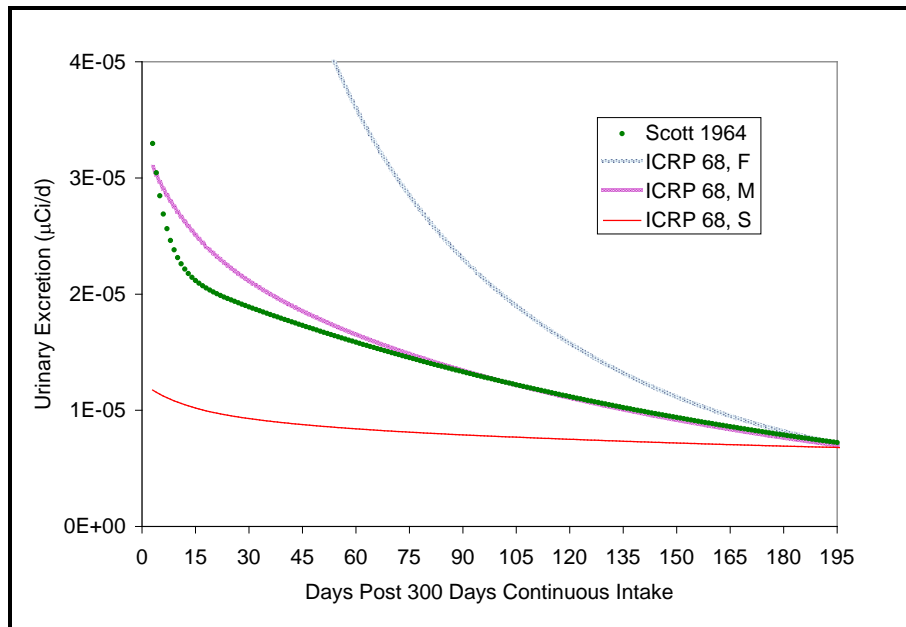


Figure 5-2. Empirical and theoretical uranium excretion after uranium restriction.

Beginning in September 1994, the plant was placed in a stand-down mode. This effectively stopped all routine work (and chronic exposure potential) in the process areas. Only minimal walkthroughs and area checks continued during the stand-down, which lasted until August 1998. The stand-down significantly influenced the available source term for exposures. Before stand-down, the Y-12 program default modeling assumption was class Q (90% Super-W, 10% Y). During the stand-down, the Y-12 program default assumption of chronic exposure was modified to assume acute exposures occurring at the midpoint of a quarterly sampling frequency. No wet chemical operations were performed during this 4-yr period. Therefore, no soluble component was being produced and

materials that were stopped in process were allowed to oxidize, which provided a larger component of type S material as an exposure source. In August 1998, a partial resumption of activities occurred; wet chemical operations were not restarted and were still not restarted as of October 2003. The predominant material encountered after partial restart in August 1998 is uranium oxide, which fecal sampling has shown to be more consistent with 100% type S material. Based on the changing workplace conditions after partial restart in 1998, the default assumptions were modified to return to chronic exposure but to use type S solubility.

For a workplace as varied as Y-12, it is clear that no single solubility or particle size would apply to all workers. Furthermore, accurate assignment of the uranium lung clearance type to a given bioassay result was considered virtually impossible because of uncertainties about chemical form and limitations of the personnel-tracking system (Barber and Forest 1995, p. 669). As of 2001, Y-12 implemented a radiation work permit (RWP) tracking system that tracks the location of workers, their type of work, and the type of bioassay needed (ORAU 2003, pp. 19 ff.).

Exposure to type M material from 1948 to July 1998 appears to be the more likely absorption type. After July 1998, exposure to absorption type S material is more likely. However, the absorption type can be based on the monitoring data, claimant-favorable assumptions, or both.

### **5.1.2 Particle Size Information**

Several particle size studies in uranium process areas have been conducted at Y-12 (e.g., CCCC 1953; Steckel and West 1966; Barber and Forest 1995; BWXT Y-12 2003a). For different times and different processes, reported particle sizes ranged from less than 1 to over 10  $\mu\text{m}$  (physical). Steckel and West (1966) reported a positive correlation between uranium oxide particle size and process temperature. Barber and Forest (1995) used an 8- $\mu\text{m}$  AMAD based on particle size measurements as the basis for the class Q dosimetry system used in the 1990s. (The class Q system is further described as 10% class Y and 90% modified class W. The modification consists of increasing the class W 50-d compartment to 120 d.) In terms of lung deposition and retention, the 8- $\mu\text{m}$  AMAD class Q material is closer to 5- $\mu\text{m}$  AMAD type M material than to 1- $\mu\text{m}$  AMAD class W material, as shown in Figure 5-3.

Since 2000, Y-12 has implemented the latest guidance provided by the ICRP including the ICRP Publication 66 (ICRP 1994) lung model and ICRP Publication 78 (ICRP 1998) methods and models (BWXT Y-12 2003b, p. 89). This is the justification for the change to the 5- $\mu\text{m}$  AMAD as the default particle size. Y-12 uses the newer weighting factors proposed by the ICRP in its 1990 recommendations (ICRP 1991). For Y-12 dose reconstructions, the default particle size distribution is 5  $\mu\text{m}$  AMAD .

### **5.1.3 Uranium Alpha Activity as a Function of Enrichment**

At Y-12, uranium enrichments range from depleted uranium (DU) [less than 0.72 wt %  $^{235}\text{U}$ ] to very highly enriched uranium (VHEU; greater than 93 wt %  $^{235}\text{U}$ ). Section 5.1.4.1 lists the uranium isotopic mixes with recycled uranium (RU) contaminants that were typical of uranium materials at Y-12. The potential dosimetric complexity is simplified by the similarity of the dose conversion factors for the most important uranium isotopes and by the quantities measured in the *in vitro* bioassay and chest-counting programs.

For work with enriched compounds and alloys, urinalysis programs after 1950 reported either total uranium alpha counts or isotopic uranium results, both of which reflect the dosimetric potential of the

isotopes generally handled. Early *in vivo* chest-counting results were reported either as micrograms of  $^{235}\text{U}$  or milligrams of  $^{238}\text{U}$  based on whether an exposure was believed to be from general enriched

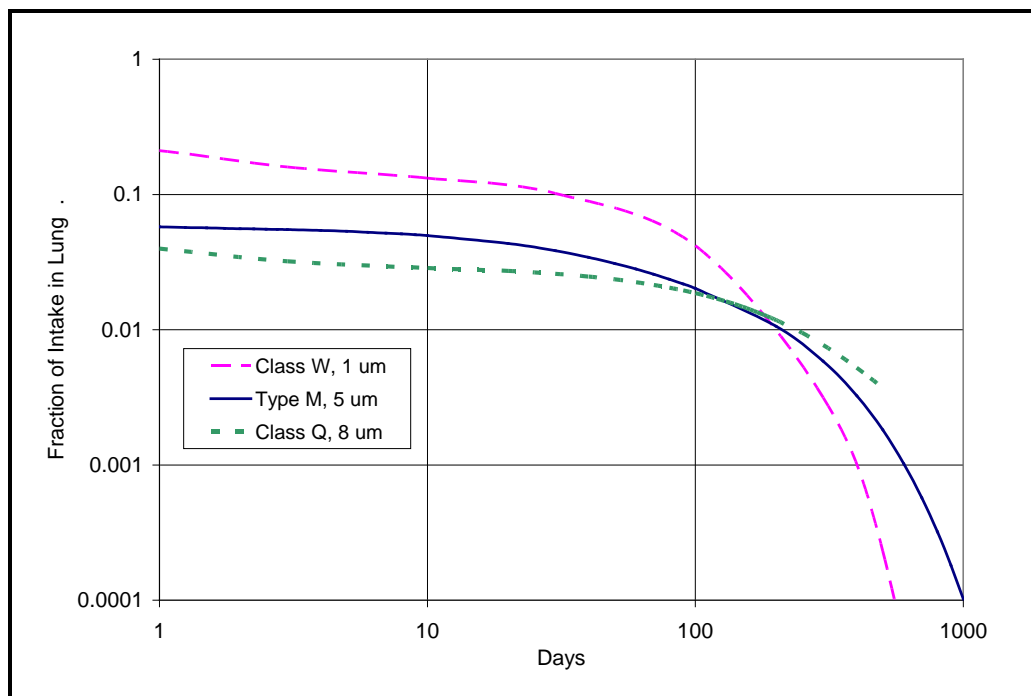


Figure 5-3. Lung deposition and retention of class W, class Q, and type M uranium. (Particle sizes specified are in micrometers AMAD.)

uranium (EU), natural uranium (NU), or DU. Based on these reporting techniques, the following assumptions can be made about data interpretation for the periods from 1950 through 1989 for urinalysis and from 1961 to 1989 for lung counting.

For lung counts a combination of the information in the Type Analysis and Material Type (see Section 5.A.1.3 for details) reporting fields can be used to determine whether the count was believed to be due to NU or DU. For records through 1971, a Type Analysis of 1 with a Material Type of 2 or 3 indicates DU, while a Material Type of 7 indicates NU. For records after 1971, a Type Analysis of 4 with a Material Type of 2 or 3 indicates DU, while a Material Type of 7 indicates NU.

Table 5-1. Enrichment assumptions for dose assessment for various analytical techniques.

Analytical technique (Urinalysis 1948–present) (Lung count 1961–1989)	Measurement information	Default enrichment assumption
Urine by fluorometry (usually for an individual who worked in areas with NU or DU; before 1950, also used for workers in EU areas).	Alpha dpm can be calculated from mass.	Before 1950, 93% enrichment. After 1949, NU.
Urine by gross alpha counting (usually for an individual who worked in areas with EU).	Alpha dpm is the sum of all uranium alphas.	93% enrichment.
Urine by alpha spectrometry (10/1989 to present).	Alpha dpm/d is reported isotopically and total uranium is determined by summing isotopic results.	
Lung count	$\mu\text{g U-235}$	93% enrichment.

Lung count	µg U-238	NU.
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At Y-12, it is current practice to assign dose by applying the dose conversion factor for  $^{234}\text{U}$  to all inferred uranium intakes. This practice is slightly conservative for EU; the resultant overestimate is less than 1% of the effective dose. For DU, the use of the  $^{234}\text{U}$  dose conversion factor leads to an overestimate of 16% for type S uranium and 20% for type M uranium (Eckerman and Kerr 1999, p. 16).

#### 5.1.4 Temporal Pattern of Uranium Exposures

As a rule, routine uranium exposures at Y-12 were considered to be of a chronic nature. For example, in 1958, Patterson stated, "...our interpretation of urinalysis results and our assignment of internal dose assume an exposure under equilibrium conditions of intake and elimination" (Patterson 1958, p. 58). For the stand-down period from September 1994 until August 1998, acute exposures should be assumed to be the more likely mode of exposure. After this time, while complete equilibrium is not expected in modern internal dosimetry models, the presumption of chronic exposure conditions for uranium remains in place as noted in a 2003 report: "The most likely exposure potential for uranium work at the Y-12 Complex is chronic in nature" (BWXT Y-12 2003b, p. 107). Because workers were assigned to various buildings and processes throughout their employment, it is not practical to characterize intakes based on historical operational assignments. The claimant files do not generally contain detailed work assignments, but do contain all bioassay data for each individual worker during the years of operational exposure – primarily *in vitro* (urine) and *in vivo* (lung counts) bioassay. Radiological materials, including isotopic mixes and so forth, are not defined by operational area or building in this document, because internal dose reconstruction relies primarily on the recorded bioassay data and the defaults for unmonitored isotopes that were conservatively established on a plant-wide basis (see Table 5-8 later in this document for plant wide RU contaminant defaults).

#### 5.1.5 Other Radionuclides of Concern Including Recycled Uranium Contaminants

In addition to  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ , the following radionuclides were identified in the Y-12 Technical Basis Document for Internal Dosimetry (BWXT Y-12 2003a):

- $^3\text{H}$ ,  $^{90}\text{Sr}$ ,  $^{99}\text{Tc}$
- $^{228}\text{Th}$ ,  $^{232}\text{Th}$
- $^{232}\text{U}$ ,  $^{233}\text{U}$ ,  $^{236}\text{U}$
- $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{241}\text{Pu}$
- $^{237}\text{Np}$ ,  $^{241}\text{Am}$

The following radionuclides that could interfere with *in vivo* analysis of uranium and thorium were quantified so that their effects on the spectra could be taken into account:

- $^{40}\text{K}$ ,  $^{137}\text{Cs}$

Other radionuclides addressed included:

- $^{60}\text{Co}$  and  $^{95}\text{Zr}/^{95}\text{Nb}$  for organizations outside Y-12 (Cofield 1961)
- $^{210}\text{Po}$

Many of these radionuclides are accompanied by progeny in various stages of equilibrium. In addition, operators of the *in vivo* analysis equipment reviewed accumulating spectra for interferences from medical radioisotopes such as  $^{131}\text{I}$ .

#### 5.1.5.1 Unmonitored Radionuclides from Recycled Uranium

One of the primary sources of significant contaminants (including the production of other than the normal uranium isotopes) that were introduced into the Y-12 plant systems were those associated with RU from 1953 until 1999. These contaminants were a result of fission and activation processes of a variety of uranium enrichment isotope mixtures, which were irradiated in production and test reactors. After completing their useful life in the reactors, the unused uranium in the spent fuel elements or targets was recovered in chemical extraction plants and returned to the inventories in the DOE system along with trace quantities of the contaminants (in some instances U-236 was produced in quantities greater than "trace" levels).

The major source of RU contaminants at Y-12 was in the receipt and processing of returned VHEU from SRS and the Idaho Chemical Processing Plant (ICPP). Tables 5-2 and 5-3 document the quantities of uranium and the calculated mass quantities of plutonium, neptunium, and technetium actually delivered to Y-12. Table 5-4 presents a computer-generated listing of the relative production of a few isotopes of concern, following irradiation of VHEU fuel in a high-flux test reactor. The process streams included a spectrum of chemical forms of uranium, including uranyl nitrate [ $\text{UO}_2(\text{NO}_3)_2$ ] solutions, uranium trioxide ( $\text{UO}_3$ ), uranium-aluminum (U-Al) alloy ingots, uranium scrap, uranium tetrafluoride ( $\text{UF}_4$ ), uranium metal, solvent extraction raffinates, and a variety of secondary process wastes and residues.

The uranium enrichments ranged from DU, NU, low enriched uranium (LEU - less than 20%  $^{235}\text{U}$ ), highly enriched uranium (HEU - from 20 to approximately 93%  $^{235}\text{U}$ ), and very highly enriched (VHEU - more than 93%  $^{235}\text{U}$ ). The predominant RU and associated contaminants were in HEU and VHEU materials. However, all of the uranium at Y-12 came from other DOE facilities, which had also either generated and/or received RU materials. Therefore, nearly all of the uranium in the DOE facilities contained RU contaminants to varying degrees through being processed in the same equipment, blending with other materials to adjust the degree of enrichment, etc. For example, most of the DU received and processed at Y-12 was seldom chemically processed but was received in forms from which parts were produced by mechanical processing. However, even these materials were received from other plants, such as Fernald, which also had an RU contaminant inventory. Therefore, the DU contaminant levels at Y-12 were inferred from both the Fernald and the Idaho Specific Manufacturing Capability (SMC) projects' recycle reports (ORAUT 2004 and DOE 2000).

The fundamental conclusion from a variety of reports, including a lack of definitive RU contaminant bioassay data, is that for dose reconstruction purposes a default level of RU contaminants should be derived and applied as a percentage increase to the derived uranium intake for each of the four major contaminants. Analytical information derived from a variety of sources allows the calculation or interpolation of the levels of the predominant RU contaminants in the uranium materials received, processed, and handled at Y-12. One of the primary sources of RU contaminant information was the documented report generated as a consequence of a comprehensive study of Recycled Uranium in DOE facilities in 2000. The applicable report for Y-12 was issued in December 2000 and titled Recycled Uranium Mass Balance Project Y-12 National Security Complex Site Report (DOE 2000). It should be noted that there were some recognized inconsistencies in shipping and receipts between the various DOE facilities in the DOE 2000 RU reports. In an attempt to resolve these inconsistencies a summary report in 2003 by the DOE Office of Security, titled Recycled Uranium, United States Production, Enrichment, and Utilization (DOE 2003) was issued though correcting some

inconsistencies, there remain significant inconsistencies in the two reports. For purposes of reconstruction of RU contaminant inventories in the Y-12 facilities and internal dose default analyses, the rationale for resolving those inconsistencies is provided herein. One approach to gain an estimate of the average contaminant concentrations through the years is to document the total receipts of contaminants and uranium and then to derive the average long-term RU contaminant concentrations.

Tables 5-2 and 5-3 list the documented RU contaminants and their primary source in recycled VHEU. For completeness, other DU receipts are listed by the DOE (2000) report without documenting the associated RU contaminant levels. However, it is known that these uranium receipts also contained low-level RU contaminants, the concentrations of which are discussed later in this report.

Table 5-2. Y-12 Uranium receipts summary –presenting average levels of three predominant recycled uranium contaminants through plant history (1953 to 1999).

Site	U Kg	Pu			Np			Tc		
		g	ppb	pCi/μg U	g	ppm	pCi/μg U	g	ppm	pCi/μg U
<b>Y-12 VHEU Receipts Summary – DOE 2000 RU Report</b>										
SRS	125,161	0.0455	0.36	5.2E-03	3,600	29	2E-02	14,268	114	1.9
ICPP	25,696	0.00124	0.05	7.2E-04	66	3	2E-03	231	9	0.15
<b>EU Total</b>	<b>150,857</b>	<b>0.04674</b>	<b>0.3</b>	<b>4.3E-03</b>	<b>3,666</b>	<b>23</b>	<b>1.6E-02</b>	<b>14,499</b>	<b>96</b>	<b>1.64</b>
<b>DU Receipts Summary - DOE 2000 RU Report</b>										
Hanford	1,502	No Data								
ORGDP	192,836									
PGDP	38,423									
<b>DU total</b>	<b>232,761</b>									
<b>Grand total</b>	<b>383,618</b>									

a. ICPP = Idaho Chemical Processing Plant; ORGDP = Oak Ridge Gaseous Diffusion Plant; PGDP = Paducah Gaseous Diffusion Plant.

Table 5-3. Total recycled uranium receipts summary.

Shipping site	U (kg)	Pu			Np			Tc		
		g	ppb	pCi/μg U	g	ppm	pCi/μg U	g	ppm	pCi/μg U
SRS	153,000									
ICPP	25,700									
Hanford	4,400									
West Valley	900									
<b>Total</b>	<b>184,000</b>	<b>0.5</b>	<b>2.7</b>	<b>0.039</b>	<b>54</b>	<b>0.003</b>	<b>0.00021</b>	<b>9,100</b>	<b>49.5</b>	<b>0.85</b>

The results of the DOE Office of Security RU report (DOE 2003) are presented in Table 5-3. As indicated there are significant differences with the results recorded in DOE (2000) report (summarized in Table 5-2) and 5-3. The objective of DOE (2003) was to resolve the differences in the reported shipment amounts from the recycling plants (the “primary sites”) with the reported receipts at the first or “primary receiving sites.” Also the detail in DOE 2003 related to uranium enrichment and specific RU contaminant concentrations from each site were not provided. (Note the lack of detail in table 5-3.) In examining DOE (2003), the shipments from these “primary receiving sites” (following processing and incorporation of recycled materials into their process streams) to what can be called “secondary” receiving sites were not documented. For example, Y-12 received uranium materials from gaseous diffusion plants, Fernald, and others, which also had RU contaminant inventories. Thus the discrepancies in the current values reported in DOE (2000) and DOE (2003) could result from documenting the “primary” shipment and receipts in DOE (2003), while DOE (2000) accounts for “secondary shipments” as well. However, where large differences occur, the most clamant-favorable value is used in deriving the defaults for dose reconstruction, which are generally the results in DOE (2000).

Though plutonium, neptunium, and technetium isotopes were analyzed and documented in all of the facilities as the primary isotopes of concern, there was a significant level of <sup>228</sup>Th detected in nearly all

of the facilities at Y-12 and in reported process stream analyses (DOE 2000). To establish the source of this isotope and provide a basis for including it in the RU contaminants to be considered in the internal dose reconstruction, Tables 5-4 and 5-5 are included to demonstrate the production of this isotope in VHEU fuels and the relative/effective “enrichment” during chemical reprocessing. The values in Table 5-4 were derived for purposes of the DOE (2000) Recycled Uranium report at the ICPP. The ORIGIN2 computer program results illustrate the production of isotopes during use of VHEU fuels (D.R.Wenzel 2000 and D.R.Wenzel “specific tables” of isotopic production---“). The isotopes of interest for purposes of this illustration are both the uranium isotopes and the plutonium, neptunium, technetium, and thorium. Thorium-228 is a contaminant that became an isotope of concern at Y-12, which is the primary receiving site of recycled VHEU fuel materials.

Table 5-4. Primary contaminants (at 3-yr decay) in VHEU fuels and of concern in internal dose reconstruction.

Isotope	Half-life (yr)	Activity (μCi/μg Total U)	Activity (μCi/g Total U)	Percent weight g/100 g U <sup>a</sup>	ppm U	Activity ratio (isotope/U)
Tc-99	2.11E+05	3.8E-04	3.8E+02	2.2E+00	2.2E+04	24
Th-227	18.72 d	3.3E-11	3.3E-05	1.1E-13	1.1E-09	
Th-228	1.913	1.6E-07	1.6E-01	1.9E-08	1.9E-04	0.01
Th-229	7.34E+03	1.4E-12	1.4E-06	6.7E-10	6.7E-06	
Th-230	7.7E+04	1.9E-11	1.9E-05	9.6E-08	9.6E-04	1.2E-06
Th-231	25.52 hr	1.7E-06	1.7E+00	3.2E-10	3.2E-06	0.1
Th-232	1.41E+10	3.5E-15	3.5E-09	3.2E-06	3.2E-02	2.2E-10
<b>Total</b>			<b>1.9</b>			
U-232	6.89E+01	2.7E-07	2.7E-01	1.3E-06	1.3E-02	
U-233	1.59E+05	2.6E-10	2.6E-04	2.7E-06	2.7E-02	
U-234	2.46E+05	6.6E-07	6.6E-01	1.0E+00	1.0E+04	
U-235	7.04E+08	1.7E-06	1.7E-00	7.8E+01	7.8E+05	
U-236	2.34E+07	1.3E-05	1.3E+01	2.0E+01	2.0E+05	
U-237	6.75 d	3.6E-08	3.6E-02	4.4E-11	4.4E-11	
U-238	4.47E+09	5.6E-09	5.6E-03	1.7E+00	1.7E+04	
<b>Total</b>			<b>15.7</b>			
Np-236	1.15E+05	9.4E-11	9.4E-05	7.2E-07	7.2E-03	
Np-237	2.14E+06	9.1E-06	9.1E+00	1.3E+00	1.3E+04	0.6
Np-238	2.117 d	9.2E-10	9.2E-04	3.6E-13	3.6E-09	
Np-239	2.355	4.3E-08	4.3E-02	1.9E-11	1.9E-07	
<b>Total</b>			<b>9.1</b>			
Pu-236	2.851	3.6E-06	3.6E+00	6.8E-07	6.8E-03	0.2
Pu-238	87.74	3.6E-02	3.6E+04	2.1E-01	2.1E+03	2.3E+03
Pu-239	2.41E+04	1.9E-05	1.9E+01	3.1E-02	3.1E+02	1.2
Pu-240	6.54E+03	1.5E-05	1.5E+01	6.4E-03	64	1.0
Pu-241	14.4	1.5E-03	1.5E+03	1.4E-03	14	1.0E+02
Pu-242	3.73E+05	8.8E-09	8.8E-03	2.3E-04	2.3	5.6E-04
<b>Total</b>			<b>4.6E+04</b>			
Am-241	4.32E+02	9.6E-06	9.6E+00	2.8E-04	2.8	0.6

a. There were basically three types of fuel elements with varying enrichments (both at the start and after burn-up). The typical after burn-up enrichments were 78.21% for aluminum-clad, 78.42% for zirconium-clad, and 51.9% for stainless-steel fuel elements. Values represent the 78% after burn-up enrichments as representative, with typical 1%+ for U-234.

Table 5-5. Comparison of the production of three selected isotopes in VHEU spent fuel with levels reported in HEU shipments to Y-12.

Isotope	μCi/g U product from SRS	μCi/g U product from ICPP	μCi/g U in high burn-up VHEU reactor fuel <sup>b</sup>	Approximate Derived Decontamination Factor (DF)
Th-228	0.01 – 0.03	NA <sup>a</sup>	0.16	10
Np-237	0.01 – 0.02	1E-03	9.1	1E+03–1E+04

Pu-238	4E-03	3E-04	3.6E+04	1E+07–1E+08
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a. There were no analytical data for Th-228 reported in the ICPP Y-12 receipt reports. The DF reported is interpolated from Y-12 process stream analytical data. These values were taken directly from column 4 in table 5-4 above.

Comparing the concentrations in the spent fuels with the reported concentrations in the product received at Y-12 provides a rough indication of the decontamination factors (DF) of the chemical extraction processes. Reference to these DFs in Table 5-5 and the average contaminant concentrations in Table 5-6 indicate that plutonium is the most efficiently removed with an effective DF of approximately  $1 \times 10^7$  to  $10^8$ , with a neptunium DF of approximately  $1 \times 10^4$ , while thorium appears to follow the chemistry of uranium more closely, resulting in a DF of only 10, which in turn results in a relative “enrichment” of  $^{228}\text{Th}$  in the VHEU product received at Y-12. Although  $^{228}\text{Th}$  has not been considered an RU contaminant of concern in other DOE facilities and was not routinely analyzed, there were sample analyses of contaminants in surface smears and wipes in most of the Y-12 plant facilities in 1997 and 2001, which included  $^{228}\text{Th}$ . These surface contamination surveys of a large number of Y-12 plant facilities indicate a predominance of  $^{228}\text{Th}$  as a contaminant. The uranium to non-uranium alpha ratios range from 4 to more than  $1 \times 10^4$ , which is not useful for dose reconstruction. However, these data do indicate that  $^{228}\text{Th}$  is one of the primary contaminants at Y-12, which in turn provides a technical base for inclusion of  $^{228}\text{Th}$  in the list of RU contaminant isotopes.

Table 5-6. Ranges of reported contaminant values in the main processing facilities (summary of Tables B-3 and B-4 of ).<sup>a</sup>

Contaminant	Range	Ave. of range	Ave. of range minus the high	Raffinate values <sup>b</sup>
Pu	0.11 - 4.5 ppb	0.5 ppb	0.25 ppb	62 ppb
Np	4.7 – 346 ppm	30.6 ppm	20.8 ppm	2,980 ppm
Tc	0.13 – 211 ppm	83 ppm	66 ppm	641 ppm

- The highest values reported in the processing facilities were in just one of 17 process streams - that being the solvent extraction purification of UN. Uranium levels are still significant in this stream, which indicates a marked enrichment of contaminants relative to U in this stream.
- The raffinate streams are depleted in uranium to a range of 5 ppm U levels in the sludge. Therefore the parts per million of the RU contaminants relative to U are not meaningful (i.e., total Pu – maximum 30 and average 21 pCi/g net sludge, Np – maximum 12 and average 8 pCi/g sludge, Tc – maximum  $1.2\text{E}+4$  and average  $3.7\text{E}+3$  pCi/g sludge).

Uranium has always been the dominant contributor to collective internal dose at Y-12. Monitoring for other radionuclides has been performed on a limited basis. Both the relatively small concentrations and the difficulty of analyses have contributed to the lack of data. The isotopic characterization of the large variety of uranium enrichments/contaminants is illustrated in Table 5-7, which is an average representation of six specific generalized enrichment types handled at Y-12. The uranium to non-uranium alpha ratios in Table 5-7 are in the same general range as those measured in surface contamination in the plant facilities after years of operation.

From the values in Table 5-6, it is evident that the maximum of the ranges in the plant tend to run a factor of approximately 10 above the average of the range for both plutonium and neptunium, with technetium at a factor of 3. A factor of 10 is also applied to the thorium levels. The defaults in Table 5-8 were chosen consistent with all of the information discussed above with the objective of representing the maximum values of the documented RU contaminant levels in the Y-12 process streams and thus represent claimant-favorable values, and which can apply on a plant-wide basis. The application of the default level should be based upon the indication by the bioassay as to the likelihood of VHEU/HEU or LEU/NU/DU. The appropriate default ratio should be chosen based on the  $^{235}\text{U}$  or  $^{238}\text{U}$  indicated in the bioassay results. For each microgram or pCi of uranium intake calculated

based on bioassay data in the claimant files, an additional intake of each of the four RU contaminants should be added.

From a practical and simplified approach, when the results in the claimant files are given in mass units of uranium per liter or lung count, column 3 values can be used to add the RU contaminant activity to

Table 5-7. Typical uranium enrichment materials at Y-12 with calculated and inferred RU contaminant levels.

Uranium Isotope	Activity (Ci/g)	Mass fraction	Activity (pCi/μg total U)	RU Cont.	Activity (pCi/μg total U)
<b>High burn-up (~50%) recycled VHEU from SRS</b>					
233	9.66E-03	1.00E-04	9.660E-01	Pu-238	6.20E-03
234	6.22E-03	1.28E-02	7.962E+01	Pu-239	1.00E-04
235	2.16E-06	5.22E-01	1.128E+00	Np-237	2.00E-03
236	6.50E-05	2.92E-01	1.898E+01	Tc-99	1.90E+00
238	3.36E-07	1.73E-01	5.796E-02	Th-228	3.17E-02
Total		9.99E-01	1.007E+02	Total alpha	4.00E-02
dpm/μg total U			2.24E+02	α dpm/μg total U	8.87E-02
U α:Non U α				2.52E+03	
<b>Lower burn-up (~25%) recycled VHEU from ICPP</b>					
233	9.66E-03	3.000E-06	2.90E-02	Pu-238	2.00E-03
234	6.22E-03	1.000E-02	6.22E+01	Pu-239	3.30E-05
235	2.16E-06	7.800E-01	1.68E+00	Np-237	2.00E-03
236	6.50E-05	2.000E-01	1.30E+01	Tc-99	1.50E-01
238	3.36E-07	1.700E-02	7.69E-01	Th-228	5.00E-02
Total		1.007E+00	7.69E+01	Total α	5.40E-02
dpm/μg total U			1.71E+02	α dpm/μg total U	1.20E-01
U α:Non U α				1.4E+03	
<b>Weapons-grade Oralloid VHEU</b>					
233	9.66E-03	0.000E+00	0.00E+00	Pu-238	2.00E-02
234	6.22E-03	1.100E-02	6.84E-01	Pu-239	2.00E-02
235	2.16E-06	9.300E-01	2.01E+00	Np-237	6.00E-02
236	6.50E-05	7.500E-03	4.88E-01	Tc-99	5.00E-02
238	3.36E-07	6.000E-02	2.02E-02	Th-228	5.00E-02
Total		1.009E+00	7.09E+01	Total α	1.50E-01
dpm/μg total U			1.57E+02	α dpm/μg total U	3.33E-01
U α:Non U α				4.73E+02	
<b>Recycled NU</b>					
233	9.66E-03	0.000E+00	0.00E+00	Pu-238	0.00E+00
234	6.22E-03	5.400E-05	3.36E-01	Pu-239	3.00E-06
235	2.16E-06	7.200E-03	1.56E-02	Np-237	2.40E-05
236	6.50E-05	0.000E+00	0.00E+00	Tc-99	2.00E-04
238	3.36E-07	9.927E-01	3.34E-01	Th-228	3.00E-05
Total			6.85E-01	Total alpha	5.70E-05
dpm/μg total U			1.52E+00	α dpm/μg total U	1.27E-04
U α:Non U α				1.20E+04	
<b>Recycled LEU (2%)</b>					
233	9.66E-03	0.000E+00	0.00E+00	Pu-238	0.00E+00
234	6.22E-03	2.000E-04	1.24E+00	Pu-239	2.00E-04
235	2.16E-06	2.000E-02	4.32E-02	Np-237	2.00E-04
236	6.50E-05	0.000E+00	0.00E+00	Tc-99	9.20E-02
238	3.36E-07	9.798E-01	3.29E-01	Th-228	3.00E-04
Total			1.62E+00	Total alpha	7.00E-04
dpm/μg total U			3.59E+00	α dpm/μg total U	1.55E-03
U α:Non U α				2.31E+03	
<b>Recycled DU</b>					
233	9.66E-03	0.000E+00	0.00E+00	Pu-238	0.00E+00
234	6.22E-03	1.000E-05	6.22E-02	Pu-239	3.70E-06
235	2.16E-06	2.000E-03	4.32E-03	Np-237	2.70E-05
236	6.50E-05	3.000E-06	1.95E-04	Tc-99	4.00E-04
238	3.36E-07	9.980E-01	3.35E-01	Th-228	4.00E-05
Total			4.02E-01	Total alpha	7.07E-05
dpm/μg total U			8.93E-01	α dpm/μg total U	1.57E-04
U α:Non U α				5.69E+03	

- a. Values derived from a number of sources and should be considered as largely average values.
- b. The recycled DU, recycled NU, and recycled LEU values were derived from Fernald data because this site both supplied and received uranium to and from all the other DOE sites, including Y-12. In addition, the recycled DU information was verified by the analytical data reported by the Idaho SMC facility for DOE (2000).
- c. The weapons-grade Oralloid VHEU and other burn-up example VHEU were derived from Y-12 data and ICPP reports.
- d. The Oralloid RU contaminant data was presented in the reference reports in very general terms (i.e., <0.1 μCi/g U for total Np and Pu and <0.05 μCi/g for "Any other radionuclide").

Table 5-8. Recommended defaults based on upper levels of expected ranges.

Isotope	Default levels in pCi/μg U		Default levels in pCi/pCi total U <sup>c</sup>	
	VHEU & Oralloy	LEU, NU, & DU	VHEU & Oralloy	LEU, NU, & DU
Tc-99	4	0.3	4E-02	0.15
Th-228 <sup>a</sup>	0.5	0.003	5E-03	1.5E-03
Np-237	0.6	0.003	6E-03	1.5E-03
Pu-238 <sup>b</sup>	0.2		2E-03	
Pu-239 <sup>b</sup>	0.2 (Oralloy only)	0.003	2E-03 - Oralloy	1.5E-03
Total non U	1.3	0.01		
Total U	75	2		

- The Th-228 default is justified in concept due to the demonstrated existence of general plant-wide contamination and further indicated by the demonstrated production in VHEU fuels, which in turn is further validated in the process stream analyses documented in DOE (2000).
- VHEU was used in the metal form at Y-12 and called Oralloy. Plutonium analyses for Oralloy were not isotope specific. For Oralloy VHEU only, Pu-239 is the chosen isotope because the total quantities of recycled VHEU (in which Pu-238 was the predominant plutonium isotope) returned to the gaseous diffusion plants was miniscule compared to the LEU returns.
- Reference to Table 5-6 indicates activity in picocuries per gram of total U for VHEU ranges from 71 to 101; 100 is used for simplicity. For the LEU, NU, and DU, the picocuries per gram of total U ranges from 0.9 to 1.6; 2 pCi/g total U is used for simplicity.

the intake. However, when the bioassay units are in uranium alpha activity per unit urine volume and/or U isotope activity units, the values in column 4 can be used. Again, Table 5-8 values are maximum values expected (and claimant-favorable), while average values are basically a factor of 10 less than the values for plutonium, neptunium, and thorium and a factor of 3 less for technetium. If a best estimate dose reconstruction is needed, average values (a factor of 10 less than the values in Table 5-8 for plutonium, neptunium, and thorium and a factor of 3 less for technetium) should be used.

Reference to Table 5-8 with relationship to the plutonium contaminant, the recommended isotope Pu-239 should be assumed for Oralloy only, since Oralloy used at Y-12 did not come directly from the primary RU generating sites, i.e. it was enriched at the GDP facilities in which Pu-239 was dominant compared to Pu-238.

It is important to note that the existence of air contamination and personnel exposures from <sup>232</sup>Th operations is not addressed in this section. This section deals only with RU contaminants.

## 5.2 ***IN VITRO* MINIMUM DETECTABLE ACTIVITIES, COUNTING METHODS, AND REPORTING PROTOCOLS**

### 5.2.1 ***In Vitro* Urine Analysis**

#### 5.2.1.1 Coverage

Uranium enrichment activities began in the fall of 1943. A uranium urinalysis program based on fluorometry was used in conjunction with medical examinations to monitor for kidney damage from exposure to soluble uranium compounds (Sterner and Riley 1946). The earliest fluorometry data was dated in 1948. In 1950 workers in production areas were placed on a uranium fluorometry urinalysis program for the purpose of estimating internal exposure. The program was expanded to include certain maintenance workers in 1954 (McLendon 1960). Y-12 currently requires workers with a potential for internal exposures in excess of 100 mrem/yr committed effective dose equivalent (CEDE) to participate in the bioassay program (BWXT Y-12 2003b, p. 23). Until September 1989, routine urinalysis focused on EU, DU, NU, tritium, and plutonium. Beginning in October 1989, uranium

results were no longer classified as EU, DU, or NU. Rather, uranium results were reported as isotopic results based on alpha spectrometry analysis. Analyses for other radionuclides were performed on an as-needed basis.

### 5.2.1.2 Sample Collection

#### Sample Volumes

For most of the plant's history, the primary urine collection method was a spot sample submitted Monday morning before entering the work area. That is, routine samples were submitted after a minimum of a 48-hr absence from the work area. [In the July 1 to December 31, 1951, Health Physics Progress Report (UCC 1952, p. 25), it was stated that Friday evening samples would be discontinued in favor of Monday morning samples.] The fraction of the daily void volume was estimated on the basis of the time between the sample void and the previous void. This *rate method* of estimating daily void volumes was used explicitly in the calculation of the daily radionuclide excretion.

For example, the following formula was given for calculating EU excretion by electrodeposition and gross alpha counting (UCC 1965, p. 25):

$$dpm/d = 8 \times (AP \div Eff) \times (Vol \div TI) \quad (3.1)$$

where:

- AP* = Average number of counts in 30 min on a plate (disk)
- Eff* = Uranium recovery efficiency (%)
- Vol* = Volume of total void (ml)
- TI* = Time between sample void and previous void (hr)
- 8 = A constant incorporating time, count, and volume constants, including the 20-ml electroplating aliquot volume. [There is no information in UCC (1965) on what the constant includes or what the daily excretion was.]

The term (*Vol* ÷ *TI*) also appears in the corresponding equation for fluorometric determinations of NU and DU (UCC 1965, p. 27). The use of the rate method to estimate daily urinary excretion (and hence, radionuclide elimination) contributed to the uncertainty associated with any given measurement; and the corresponding detection level is discussed below.

Since 1989, routine samples have been collected over a 24-hr period, typically while the employee was on a scheduled break from the workplace. Many workers have elected to submit a *simulated* 24-hr sample. This sample is obtained by collecting the last void in the evening (before retiring), any urine excreted during the night, and the first void the following morning. This procedure is repeated for two consecutive nights (BWXT Y-12 2003b, p. 70).

#### Routine Urine Sample Frequency

Urine samples were collected monthly in 1950, and weekly collection for some employees began in 1951. By 1963, health physics personnel were basing the frequency of participation in the urinalysis program for each department on the most recent urinalysis results of that department. The 1963 edition of the *Y-12 Radiation Safety Manual* (UCC 1963a) stated that the frequency of participation schedules was reviewed monthly and adjusted semiannually to meet the following criterion: "Sample at the frequency necessary to assure, with at least 95% confidence, that 95% of the individuals in a department have a quarterly average below the plant action limit." This criterion was used within the limitations of a maximum frequency of once per week and a minimum frequency of once per quarter. Since the late 1980s, most personnel have been on a quarterly frequency.

The current practice is to schedule samples based on the RWP usage rather than a default frequency. Only if an RWP was used will a person be scheduled for a sample. If the RWP specified a fecal sample, a paired set of urine and fecal samples is scheduled approximately 60 d after entry date. If the RWP specified urine only, a urine sample is scheduled approximately 90 d after entry date.

### 5.2.1.3 Minimum Detectable Activities

#### **Normal and Depleted Uranium in Urine by Fluorometry (1945 to 1989) and Enriched Uranium (1948 to 1950)**

In fluorometry, the visible radiation emitted from a doped, sodium-fluoride bead illuminated by ultraviolet radiation is measured. Poisson counting statistics used in nuclear particle counting do not apply to this procedure. The detection limits were historically determined by testing the performance of a particular configuration against standards of known content. Early Health Physics Progress Reports give the minimum detectable limit as 5 ppb, which is a minimum detectable activity of 7  $\mu\text{g}/\text{d}$  for a nominal 1.4 L/d urine excretion rate (UCC 1949, 1950). In 1952, the instrument sensitivity and sample volumes indicated an MDA of 7  $\mu\text{g}/\text{d}$  for 1.4 L/d of urine excreted (CCCC 1953, p. 28).

The activity was calculated from the fluorometric mass reading for NU using a specific activity of 1.55 dpm/ $\mu\text{g}$ . For EU (93 wt %  $^{235}\text{U}$ ), a specific activity of 150 dpm/ $\mu\text{g}$  was used. The corresponding limits of detection are 11 dpm/d for NU and 1,100 dpm/d for EU (93 wt %  $^{235}\text{U}$ ). At times, procedures called for the direct conversion of fluorometer current into units of disintegrations per minute, implicitly omitting the mass calculation. Whether the results were expressed in terms of mass or activity, the method remained essentially stable until replaced by long-duration alpha spectroscopy in 1989. Therefore, from 1945 through 1989, the MDA was 7  $\mu\text{g}/\text{d}$  or 11 dpm/d for NU. From 1948 to 1950, the MDA was 7  $\mu\text{g}/\text{d}$  or 1,100 dpm/d for EU (93 wt %  $^{235}\text{U}$ ). Additional variation resulted from individual urine excretion volumes. Given the limitations of the rate method of estimating daily urine volumes, uncertainty in the excretion volume is likely to contribute significantly to the uncertainty associated with the detection limit of a single measurement.

#### **Enriched Uranium in Urine by Gross Alpha Counting (1950 to September 1989)**

Determination of the MDAs for this method is complicated by the standard counting method used. From the mid-1950s to 1989, each sample consisted of two silver disks, each containing uranium electroplated from separate 20-ml raw urine aliquots drawn from the container submitted by the worker. Each disk was counted twice (on two different proportional counters) for 30 min per count. If the two results from a single disk did not agree within tabulated limits, a third count was made (UCC 1966, p. 6) and the two most concordant counts were used. If the average results of the two disks from the same sample did not agree within specified limits, then two more plates were prepared, volume permitting (UCC 1966, p. 6). The expression of potentially censored data of this sort in terms of formal detection limits is not straightforward, and no detailed analysis of the statistics of this process has yet been found. However, an approximate indicator of the detection limit can be determined from published values, if issues about statistical independence are set aside.

The background count rate for the proportional counters was reported as 0.12 cpm in 1963 (UCC 1963a, p. 38). Counters were not used if the background was greater than 5 counts in 30 min (Hamrick 1958, p. 7). For a disk counted for 120 min (in fact, two disks were counted for 60 min each), assuming a well-known background and alpha of 0.05, the detection level  $L_D$  (Currie 1968) is 0.13 cpm. For 1,400 ml/d urine output, a nominal 0.5 cpm/dpm counting efficiency, and an average uranium recovery of 40% as reported by Patterson (1958), the  $L_D$  would be 46 dpm/d. With an increase in average uranium recovery to 73% (UCC 1965 p. 33),  $L_D$  would be reduced to 25 dpm/d. In reviewing historical gross alpha urinalysis data, Barber and Forest (1995) report a decision level of approximately 20 dpm/d. Average uranium recoveries between 1958 and 1965 have not been

identified, nor is it clear that the statistical convention used by Barber and Forest was identical to that used here. It is provisionally assumed that  $L_D$  was 46 dpm/d before 1965 and 25 dpm/d after 1965. However, given the limitations of the rate method of estimating daily urine volumes, uncertainty in the excretion volume is expected to add substantially to the uncertainty associated with the detection limit of a single measurement. Because recoveries were based on batch rather than individual measurements, uncertainties in recovery would also add to the uncertainty of the detection limit of a single measurement.

### Uranium in Urine by Alpha Spectrometry (October 1989 to Present)

The present alpha spectrometry system employs large urine aliquots, chemical separation of uranium, sample-specific tracers to determine recovery, and long counting times on low-background detectors. The resultant  $L_D$  is approximately 0.15 dpm/d (BWXT Y-12 2002, p. 15). The  $L_D$  varies with the uranium recovery of a particular sample, and is reported with the sample result.

### Tritium in Urine by Gas Counting

Throughout most of the plant's history (the earliest recorded data was from 1957), tritium was counted by reacting urine with calcium to evolve hydrogen gas, which was counted in an ion chamber. The MDA for this method has not been identified to date. However, the MDA was almost certainly far below the plant action level of 0.25  $\mu\text{Ci/ml}$  ( $8 \times 10^8$  dpm/d) (UCC 1957, p. 13). Results are reported in microcuries per milliliter.

### Tritium in Urine by Liquid Scintillation

The current system is to place 5 ml of middle distillate from a urine sample in liquid scintillation fluid for counting. Assuming typical method efficiencies, the MDA is 2,000 dpm/d.

### Plutonium in Urine by Gross Alpha Counting

The plutonium in urine procedure used before 1988 involved chemical separation from a 24-hr void or a simulated 24-hr sample before gross alpha counting. The detection limit has not yet been identified, but was certainly far below the corresponding limit for uranium in urine, which was based on 20-ml aliquots. If the counting method was identical (same count time of 60 min), chemical recovery (40% to 73%), and counting efficiency (0.5 cpm/dpm) to the early uranium-counting method, the MDA for a 24-hr plutonium sample (less 40 ml) would range from about 0.7 to 1.3 dpm/sample.

### Plutonium in Urine by Alpha Spectrometry (1988 to Present)

The *a priori*  $L_D$  for both  $^{238}\text{Pu}$  and for  $^{239,240}\text{Pu}$  is 0.025 dpm/sample, or 0.025 dpm/d, assuming a full 24-hr void volume. Actual sample-specific MDAs are included in the analysis reports.

### Other Actinides in Urine and Feces

Table 5-9 lists the analytical laboratory  $L_D$  values used since about 1988 (BWXT Y-12 2003b, p. 107).

Table 5-9.  $L_D$  values.

Radionuclide	$L_D$ (dpm/sample)
$^{241}\text{Am}$	0.05
$^{228}\text{Th}$	0.15
$^{232}\text{Th}$	0.07
$^{237}\text{Np}$	0.1

### Uranium in Feces

Fecal sampling was started in 1998 when the presence of a less soluble component was identified as a result of the stand-down. In 2002, out of 2,800 participants in the bioassay program, 700 Y-12 workers participated in the fecal-sampling program (BWXT Y-12 2002, p. 7). Participation in the

fecal-sampling program is based on the potential for significant exposures to insoluble uranium [i.e., workers who are expected to have an internal exposure to type S uranium and there is a potential to exceed 100 mrem (BWXT Y-12 2003b, p. 80)]. At present, the analytical chemistry division cites the value of 0.15 dpm/sample as a representative MDA for fecal analysis (BWXT Y-12 2002, p. 15). Samples usually consist of a single void. The results in the claim files are assumed to be in units of activity per sample unless otherwise indicated (BWXT Y-12 2003c, p. 7).

## **5.2.2 In Vitro Methods for Individual Radionuclides**

The following section discusses the *in vitro* methods for specific radionuclides in urine. (Section 5.2.3 discusses analysis of fecal samples.)

### **5.2.2.1 In Vitro Bioassay for Uranium**

#### **Overview**

The *in vitro* bioassay program for uranium at Y-12 can be divided into four eras. First, from 1943 to 1947, limited monitoring occurred, for which the data cannot be retrieved. Second, from 1948 to 1950, fluorometric analyses of urine and blood occurred as part of general medical surveillance to prevent kidney damage from exposure to soluble uranium compounds. Third, from 1950 to 1989, collection of routine samples based on uranium exposure potential was initiated. During this third phase, fluorometric analyses were performed on samples submitted by workers in NU and DU areas. Electrodeposition of uranium, followed by gross alpha counting, was used for samples submitted by workers in EU areas. The primary goal for the EU analyses was to control lung doses from insoluble compounds under the assumption that such materials cleared from the lung with a 120-d half-life. During this lengthy era there were modifications in procedures, but the basic approach remained the same. Fecal sampling was used for some follow-up investigations. Fourth, after October 1989, routine 24-hr urine samples were collected. The uranium was extracted by chemical separation and ion exchange, and the extract was counted by alpha spectroscopy for an extended period (1,000 min/sample). Since 2000, fecal samples have been submitted routinely for individuals working with largely insoluble forms of uranium.

#### **Uranium Analysis by Fluorometry (1948 to 1989)**

Uranium processing began in the fall of 1943. Y-12 technical reports describe the use of fluorometry for uranium detection as early as 1944 (Van Wazer and Reiss 1944). By 1946, fluorometric analyses for uranium in urine and blood were in general use to supplement clinical surveillance for soluble uranium exposures (Stern and Riley 1946). Fluorometric uranium urinalysis data from the Tennessee Eastman Corporation era (1943 to 1947) were discussed in a 1981 mortality study of plant personnel (Polednak and Frome 1981).

Early in the Union Carbide management of Y-12, the fluorometric method was reassessed. Extraction methods were used starting in 1949 and 1950 (CCCC 1949, p. 7; UCC 1950, p. 14). By the first half of 1952, a technique involving small (0.2-ml) aliquots of raw urine had been put in place (CCCC 1953, p. 28). However, after this change in technique was instituted the urinary uranium concentrations increased greatly as shown in Table 5-7. This raises the concern that the technique used before May of 1952 could have underestimated the urinary uranium concentrations. (See Section 5.4.2 for discussion of missed doses.)

Fluorometric urinalysis continued to be used for NU and DU until 1989, when alpha spectroscopy began to be used. Fluorometry yielded results in mass units (e.g.,  $\mu\text{g/L}$ ), but results were often converted to disintegrations per minute per day with the assumed specific activity of 1.55 dpm/ $\mu\text{g}$  for

NU. The fluorometric technique had an industry standard sensitivity of about 5 ppb (5 µg/L) (e.g., UCC 1949, p. 7).

### **Uranium Analysis by Electrodeposition and Alpha Counting (1950 to 1989)**

The primary objective of this procedure was to prevent EU lung burdens in excess of the prevailing limit (0.017 µCi) corresponding to an ICRP 2 (ICRP 1960) calculated lung dose of 15 rem/yr (using a quality factor of 10). Samples from workers in EU areas were analyzed by electrodeposition of uranium onto silver discs, which were then counted for gross alpha activity in a proportional counter. Development of the gross alpha counting method was reported to be underway in early 1949 (CCCC 1949, p. 7). At first, samples were subjected to an acid digestion step before electrodeposition. In 1951, methodological problems leading to underestimates of a factor of 2 were noted and corrected (UCC 1952, p. 53). The information available was not sufficient to determine if the records themselves were corrected. In the mid-1950s, the acid digestion step was discontinued altogether due to reported contamination problems, as well as for logistical considerations (Patterson 1958, p. 34).

From the mid-1950s, two 20-ml aliquots of raw urine from each sample were placed in electrodeposition cells, which produced two discs per sample. Each disc was counted once on one proportional counter for 30 min and a second time on a different counter. The results were then averaged. The background count rate was reported as 0.12 cpm in 1963 (UCC 1963a, p. 38). The expected net count rate for a person excreting at the action level of 70 dpm/d was 0.2 cpm, given a urine output of 1.4 L/d, a nominal 0.5 dpm/cpm counting efficiency, and a uranium recovery of 40%. Because each disk was counted twice for 30 min each, the expected net count for the two counts on each disk under these conditions was 12 counts, over a background of 7.2 counts.

Recoveries were estimated on the basis of spiked samples that accompanied each counting run. The typical spike contained relatively little activity. Recovery rates from raw urine tended to be around 40% in the late 1950s (Patterson 1958), but they improved over time: In 1965, the average recovery for electroplating was 73% (UCC 1965, p. 33). Method limitations are discussed in a number of reports, including Johnson et al. (1959) and UCNC (1959). Recovery rates were found to vary with pH, drying procedure, and uranium content, with rates decreasing for uranium concentrations below the plant action level. The precision of any one sample was acknowledged to be relatively low:

*Further, realizing the over-all lack of precision in any one sample result, we normally recommend restriction only on the basis of the 13-week or quarterly cumulative internal dose as indicated by from 2 to 13 samples. (Patterson 1958, p. 57)*

*The precision of an individual result in this type of procedure is poor and for this reason little significance is attached to individual results, particularly those below the "Plant Action Limit (PAL)" level. (UCC 1966).*

These limitations notwithstanding, the salient point is that, when *in vivo* counting became routine in the early 1960s, very few additional workers were found to require restriction from uranium areas (e.g., UCC 1963b, p. 6). The *in vivo* monitoring frequency was determined by urinalysis results and ranged from monthly to once each 18 months (UCC 1963b, p. 1). Almost all of the restrictions that did take place would have occurred on the basis of the urinalysis program alone. In those cases in which restriction was based on *in vivo* analysis alone, lung retention times were often observed to be considerably longer than the assumed 120-d lung half-life on which the urinalysis program was based. In other words, the problem was largely due to the inapplicability of the lung model for some materials and individuals, rather than a failure of the urinalysis program.

### Uranium-233 by Gross Alpha Counting

In early 1962, Y-12 undertook the fabrication of metallic  $^{233}\text{U}$  from a nitrate solution ( $^{233}\text{U}$  uranyl nitrate) in Building 9205 (West and Roberts 1962). In recognition of the high specific activity of this material, and the potential for gamma radiation from the progeny of contaminant  $^{232}\text{U}$ , operations were doubly contained. Pilot runs were made with less hazardous materials to test the protective measures. As part of these precautions, eight workers submitted urine samples for analysis by electrodeposition and gross alpha counting as described above, with the same recoveries and detection efficiencies. Ratios of  $^{233}\text{U}$  to  $^{232}\text{U}$  varied with operation and time. Table 5-10 list ratios for various exposure scenarios. In addition to absorption types M and S, exposure to type F material is considered likely in this process. Uranium-232 has the larger dose conversion factor.

Table 5-10.  $^{233}\text{U}$  to  $^{232}\text{U}$  ratios.

Process	U-233:U-232 activity ratio
Reduction of $\text{UF}_4$	1.3
Casting of metal-crucible skull	15–32
Machining final uranium component	160

Due to uncertainty in the process, claimant-favorable assumptions should be made about solubility and uranium activity ratios.

### Uranium Analysis by Alpha Spectrometry (1989 to Present)

Up to 2 L of urine, along with a  $^{232}\text{U}$  tracer, are treated with nitric acid ( $\text{HNO}_3$ ) and hydrogen peroxide ( $\text{H}_2\text{O}_2$ ). The sample is wet-ashed, and the uranium is coprecipitated with calcium oxalate ( $\text{CaC}_2\text{O}_2$ ). After dissolving the precipitate in hydrochloric acid ( $\text{HCl}$ ), the uranium is further separated by ion exchange chromatography. The uranium is eluted from the column with a solution of dilute  $\text{HCl}$  to which titanous chloride ( $\text{TiCl}_3$ ) has been added to reduce actinides that could be in an elevated oxidation state. The final fraction of the eluate is first treated with ascorbic acid to reduce the presence of any ferric iron and next with hydrofluoric acid ( $\text{HF}$ ). Then the uranium isotopes are coprecipitated on neodymium fluoride ( $\text{NdF}_3$ ). The  $\text{NdF}_3$  is caught on a 0.1-mm filter, rinsed, dried, and mounted on a planchet for alpha spectrometry (BWXT Y-12 2003b). Typical recoveries are about 85%. Samples are typically counted for 16 hr on a passivated implanted planar silicon detector.

#### 5.2.2.2 *In Vitro* Bioassay for Plutonium

##### Plutonium in Urine by Gross Alpha Counting

In 1957, certain workers in the Special Testing Department were tested for plutonium intake by urinalysis. Because large urine volumes were required, these employees submitted 24-hr samples through the use of take-home kits. The sampling frequency was monthly (UCC 1957, p. 38). After two 20-ml aliquots were removed for uranium analysis by electrodeposition, the balance of the sample was treated by chemical separation, precipitation, and evaporation onto a stainless-steel planchet for alpha counting. Health physics personnel converted the reported activity rate (disintegrations per minute per 24-hr void) to millirem per day to the critical organ.

##### Uranium, Plutonium, and Americium by Alpha Spectrometry (October 1989 to Present)

Up to 2 L of urine, along with  $^{232}\text{U}$ ,  $^{242}\text{Pu}$ , and  $^{243}\text{Am}$  tracers, are treated with  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$ . The sample is wet ashed, and the uranium, plutonium, and americium are coprecipitated with  $\text{CaC}_2\text{O}_2$ . After dissolving the precipitate in  $\text{HCl}$ , the uranium, plutonium, and americium are further separated by the use of two ion exchange columns and an additional oxalate precipitation. The uranium, plutonium, and americium in the final fractions are coprecipitated with  $\text{NdF}_3$ . The  $\text{NdF}_3$  is caught on a 0.1-mm

filter, rinsed, dried, and mounted on a planchet for alpha spectrometry (BWXT Y-12 2003b). The same procedure is used for isotopic thorium.

### 5.2.2.3 *In Vitro* Bioassay for Tritium

As of 1957, personnel engaged in processing materials with a potential for tritium contamination submitted three urine samples per month. Samples were submitted at the same stations used for uranium and plutonium samples. The samples were reacted to evolve hydrogen gas, which was then counted for beta activity in an ionization chamber. Results were reported in microcuries per milliliter. (UCC 1957, pp. 38–39)

Tritium is currently measured by liquid scintillation counting. An aliquot of the urine sample is distilled in a heating mantle, and the middle fraction of the distillate is collected. A 5-ml portion of the collected distillate is mixed with liquid scintillation cocktail. The beta activity of the tritium is then measured by liquid scintillation counting (BWXT Y-12 2003b). Exposure should be assumed to be due to HTO.

### 5.2.2.4 *In Vitro* Bioassay for Other Radionuclides

The following alpha spectrometry procedures described in Section 3.2 of BWXT Y-12 (2003b) have been in use since 1989.

#### Neptunium-237 in Urine by Alpha Spectrometry

In this procedure,  $^{239}\text{Np}$  is used as an internal yield monitor. Americium-243, which is in equilibrium with its daughter,  $^{239}\text{Np}$ , is added during the sample preparation process. The separation time of  $^{239}\text{Np}$  from its parent,  $^{243}\text{Am}$ , is recorded (so that the count can be corrected for decay), and the beta activity is measured to calculate the radiochemical yield. The tracer is added to the urine sample (or fecal material) along with concentrated  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$ , and the sample is wet ashed. Neptunium is coprecipitated with  $\text{CaC}_2\text{O}_2$  and mixed hydroxides. After dissolving the precipitate in concentrated  $\text{HNO}_3$  and ashing in a muffle furnace, the oxidation state of neptunium is adjusted to the tetravalent state by using ferrous sulfamate as a reducing agent. Neptunium is separated from other actinides by the anion exchange method. Neptunium is eluted with 0.5-molar HCl solution containing  $\text{TiCl}_3$ . The eluate is collected, and neptunium is coprecipitated on  $\text{NdF}_3$  and filtered on 0.1-mm filter paper. The  $\text{NdF}_3$  is rinsed, dried, and mounted on a planchet for beta counting and alpha spectrometry.

#### Isotopic Thorium ( $^{228}\text{Th}$ , $^{230}\text{Th}$ , and $^{232}\text{Th}$ ) by Alpha Spectrometry

Up to 2 L of urine along with a  $^{229}\text{Th}$  tracer is treated with  $\text{HNO}_3$  and  $\text{H}_2\text{O}_2$ . The sample is wet ashed, and the thorium is coprecipitated with  $\text{CaC}_2\text{O}_2$ . After dissolving the precipitate in  $\text{HNO}_3$  and mixing with methanol, the thorium is further separated by ion exchange chromatography. The thorium is eluted from the column with 8-molar HCl. Hydrofluoric acid (HF) is added, and the thorium isotopes are coprecipitated with  $\text{NdF}_3$ . The  $\text{NdF}_3$  is caught on a 0.1-mm filter, rinsed, dried, and mounted on a planchet for alpha spectrometry. The same procedure is used for uranium, plutonium, and americium determinations. This method accommodates substantial quantities of common interferences, such as iron, aluminum, titanium, zirconium, and uranium. However, plutonium can present some interference. If plutonium is expected, an alternate method should be chosen (BWXT Y-12 2003b).

### 5.2.3 Fecal Sample Analysis

Fecal samples have long been used in follow-up investigations when urinalysis or *in vivo* measurements indicate the likelihood of a substantial intake. West and Scott (1966) describe such an investigation in the 1960s in which fecal, urine, and *in vivo* measurements were used jointly to investigate clearance. Fecal sampling was discontinued several times in the past, but was reinstated

in 1998 due to changes in workplace exposure conditions. At first, fecal sampling was used for a limited number of workers in Building 9212 but was expanded to include Building 9215 in 1999 and to other areas of the plant in 2000 and 2001 (BWXT Y-12 2002, p. 6).

If a work area has been determined to have predominantly insoluble airborne uranium, it is current practice for workers with a moderate to high exposure potential (CEDE greater than 100 mrem) to submit both urine and fecal samples. The relative elimination by urinary and fecal pathways is used to determine the solubility mixtures for each individual. For combined urine and fecal monitoring, the routine sampling interval is 53 d (BWXT Y-12 2002, p. 14) with 24-hr fecal collections preferred. When 24-hr fecal samples are not available, the results can be normalized using the ratio of the Reference Man excretion rate of 135 g/d to the mass of the submitted sample. The Y-12 practice was to routinely normalize results to Reference Man. These normalized results or actual individual daily excretion results can be used to reconstruct dose.

#### 5.2.4 Interferences

As investigation levels decreased over time, the contribution of natural background to worker uranium excretion assumed greater importance. Since 1989, Y-12 has corrected the measured uranium excretion values for background uranium interference when performing dose assessments. However, the uranium results reported in response to NIOSH requests for dose records have not been corrected for dietary uranium.

Y-12 used the following methods to correct sample results reported after 1989 for dietary levels of uranium. The range of background uranium values expected to be observed in urine samples is 0.14 to 0.57 dpm and the range of background uranium values expected to be observed in the fecal samples is 2.32 to 2.75 dpm (BWXT Y-12 2003b, p. 75). Therefore, at Y-12, the reference point is established at the midpoint of the quoted ranges of values for urine (0.35 dpm/d) and fecal samples (2.5 dpm/sample). The ratios between  $^{234}\text{U}$  and  $^{238}\text{U}$  can also provide information to help characterize the sample. Natural background uranium typically has a  $^{234}\text{U}$  to  $^{238}\text{U}$  ratio of approximately 1:1. EU also has a characteristic ratio, typically greater than 3:1. DU, on the other hand, typically has a characteristic ratio of less than 0.9:1.

Based on the above information, the background uranium corrections used at Y-12 are as follows:

For bioassay sample results less than the detection limit for the analytical technique, no action is required. Note, that corrections for samples from the early days when MDAs were significantly larger are not required.

To adjust results for background, the  $^{234}\text{U}$ -to- $^{238}\text{U}$  ratios are reviewed for each sample to determine if the sample is consistent with EU or DU. Conservative ratios are applied in this determination: All samples with  $^{234}\text{U}$ -to- $^{238}\text{U}$  ratios greater than 1:1 are considered to be consistent with EU, and all samples with  $^{234}\text{U}$ -to- $^{238}\text{U}$  ratios less than 1:1 are considered to be consistent with DU.

The background contribution is subtracted from EU samples as follows:

- a. Assume that the ratio of  $^{234}\text{U}$  to  $^{238}\text{U}$  is 1:1 for dietary uranium.
- b. Determine if the  $^{238}\text{U}$  result exceeds its corresponding critical level value.
- c. If the  $^{238}\text{U}$  result is greater than the critical level, then subtract two times that  $^{238}\text{U}$  result from the total uranium result.

- d. If the  $^{238}\text{U}$  result is less than its corresponding critical level value, do not subtract a dietary component.

For DU samples (e.g., those with a  $^{234}\text{U}$ -to- $^{238}\text{U}$  ratio less than 1:1), the following steps are used to determine the dietary correction for DU samples:

- a. The following equations are solved to determine an equation for the occupational  $^{238}\text{U}$  content and the occupational  $^{234}\text{U}$  content in a DU sample:

$$\frac{^{234}\text{U}_{\text{diet}}}{^{238}\text{U}_{\text{diet}}} = 1 \quad (3-2)$$

$$\frac{^{234}\text{U}_{\text{occ}}}{^{238}\text{U}_{\text{occ}}} = 0.2 \quad (3-3)$$

$$^{238}\text{U}_{\text{actual}} = ^{238}\text{U}_{\text{diet}} + ^{238}\text{U}_{\text{occ}} \quad (3-4)$$

$$^{234}\text{U}_{\text{actual}} = ^{234}\text{U}_{\text{diet}} + ^{234}\text{U}_{\text{occ}} \quad (3-5)$$

- b. Using Equation 3-5, and substituting the  $^{238}\text{U}_{\text{diet}}$  for the  $^{234}\text{U}_{\text{diet}}$  based on Equation 3-2 and substituting  $0.2 (^{238}\text{U}_{\text{occ}})$  based on Equation 3-3 for the  $^{234}\text{U}_{\text{occ}}$ , the following relationship results:

$$^{234}\text{U}_{\text{act}} = ^{238}\text{U}_{\text{diet}} + 0.2(^{238}\text{U}_{\text{occ}}) \quad (3-6)$$

Because the  $^{238}\text{U}_{\text{diet}}$  can be replaced with  $^{238}\text{U}_{\text{actual}} - ^{238}\text{U}_{\text{occ}}$  derived from Equation 3-4, Equation 3-5 can be solved to determine the occupational quantity of  $^{238}\text{U}$ . The resultant equation for determining the occupational quantity of  $^{238}\text{U}$  is:

$$\frac{^{234}\text{U}_{\text{act}} - ^{238}\text{U}_{\text{act}}}{-0.8} = ^{238}\text{U}_{\text{occ}} \quad (3-7)$$

- c. Once the occupational  $^{238}\text{U}$  quantity is known, the occupational quantity of  $^{234}\text{U}$  can be derived using Equation 3-3:

$$^{234}\text{U}_{\text{occ}} = 0.2(^{238}\text{U}_{\text{occ}}) \quad (3-8)$$

- d. Ensure that both the  $^{234}\text{U}$  and  $^{238}\text{U}$  results exceed their corresponding critical level values. If both of these results are not greater than their corresponding critical level values, then a background determination cannot be made. In this case, it is assumed that all of the activity observed in the total uranium sample result will contribute to occupational dose.
- e. Use either Equation 3-7 or 3-8 to determine the occupational  $^{238}\text{U}$  component or the occupational  $^{234}\text{U}$  component, respectively. For ease of reference, the  $^{238}\text{U}$  component was chosen for the remaining steps.
- f. Subtract the occupational component of the sample from the corresponding observed component to determine the dietary contributor:

$${}^{238}\text{U}_{act} - {}^{238}\text{U}_{occ} = {}^{238}\text{U}_{diet} \quad (3-9)$$

- g. Once the dietary component has been calculated, subtract twice this dietary value from the total uranium result to remove the background:

$$U_{tot} - 2({}^{238}\text{U}_{diet}) = U_{totalocc} \quad (3-10)$$

### 5.3 ***IN VIVO* MINIMUM DETECTABLE ACTIVITIES, COUNTING METHODS, AND REPORTING PRACTICES**

#### 5.3.1 **Whole-Body Counting**

Whole-body counting was not routinely practiced at Y-12. The primary *in vivo* detection method was chest counting, as described below.

#### 5.3.2 **Chest Counting**

The Y-12 *in vivo* chest-counting facility was developed in the late 1950s and was put into routine use in 1961. The original facility is described in Cofield (1959) and Cofield (1960). A 9- by 4-in. sodium iodide (thallium doped) [NaI(Tl)] crystal was placed over the chest of a subject reclining in a cot in a shielded room. Subjects showered, shampooed, and changed into a clean garment before counting. They were then surveyed for any remaining surface contamination before entering the counting room. The normal counting time was 20 min.

Incremental improvements in the system were made over time. A second 9- by 4-in. NaI(Tl) crystal was placed under the subject's back in 1963 or 1964. It was found that this position provided greater counting efficiency than the position over the chest. A second advantage was that skin contamination seldom occurred on the back. The ratio of the count rates from the two crystals could be used to identify surface contamination missed by the survey meter. In 1965, two 5-in. NaI(Tl) crystals were added, positioned at the sides under the arms. In 1985, initial development of a high-purity germanium (HPGe) counting system was begun. The new system began routine operation in a new counting room in June 1992.

#### 5.3.2.1 **Chest Counting for Uranium**

With the NaI scintillation system used for much of the plant's history, the amount of uranium deposited in the lungs was inferred from the worker's spectrum by use of a prediction equation. This included the subject's weight and constants derived from the spectra of control subjects. Compton scatter from natural  ${}^{40}\text{K}$  and fallout  ${}^{137}\text{Cs}$  in the body was a particularly important contributor to background in the  ${}^{235}\text{U}$  region of interest around 186 keV and the  ${}^{238}\text{U}$  region of interest around 93 keV. The  ${}^{137}\text{Cs}$  contribution to the regions of interest changed over time as fallout levels varied, requiring frequent updates to the control subject spectrum library.

Measured lung burdens were originally expressed in micrograms of  ${}^{235}\text{U}$  for EU and milligrams of  ${}^{238}\text{U}$  for NU or DU. The original limits of detection for  ${}^{235}\text{U}$  and  ${}^{238}\text{U}$  were reported as 130  $\mu\text{g}$  and 13.5 mg, respectively (Cofield 1959). Improvements in hardware and data reduction procedures led to lower detection limits, as indicated in Tables 5-11 and 5-12 below. Limits are given in terms of both mass units and of activity (nanocurie and dpm), with the published value in regular type and the converted value in italics. When known, the reporting convention used to define the detection limit is indicated in the tables. The alternative definitions listed by King and Barclay (1983) illustrate the importance of this factor.

Due to the uncertainty about actual methods for determining the lung counting detection limits, for dose reconstruction purposes and based on review of the Y-12 data, the  $^{235}\text{U}$  and  $^{238}\text{U}$  lung count detection thresholds are assumed to be 130  $\mu\text{g}$  and 13.5 mg, respectively, through 1990. Beginning in 1991, the critical level or MDA is supplied with the individual results.

Table 5-11. Reported lung counting detection limits for  $^{235}\text{U}$ .

Year	Detectors	U-235 ( $\mu\text{g}$ )	U-235 (nCi)	U-235 (dpm)	Reporting convention	Reference
1959	One 9-in. NaI	130	0.28	620		Cofield 1959
1963	Two 9-in. NaI	96	0.21	470		Scott and West 1967
1965	Two 9-in., 2 5-in. NaI	72	0.16	360		Scott and West 1967
1975	Two 9-in., 2 5-in. NaI	70	0.15	330		Scott and West 1975
1983	Two 9-in., 2 5-in. NaI	62	0.13	290	1.96 $\sigma_{\text{BKG}}$	King and Barclay 1983
1983	Two 9-in., 2 5-in. NaI	68.5	0.15	330	$L_C^a$	King and Barclay 1983
1983	Two 9-in., 2 5-in. NaI	137	0.30	670	$L_D^a$	King and Barclay 1983
1990	Two 9-in., 2 5-in. NaI	70	0.15	330	$b$	Barber and Forest 1995
6/1992	HPGe	46	0.10	220	$c$	BWXT Y-12 2003b

- As defined by Currie (1968) with  $\alpha = \beta = 0.05$ , paired observations.
- Type I error = 5%.
- Individual MDA or critical level supplied with analytical results.

Table 5-12. Reported lung counting detection limits for  $^{238}\text{U}$ .

Year	Detectors	U-238 (mg)	U-239 (nCi)	U-238 (dpm)	Reporting convention	Reference
1959	One 9-in. NaI	13.5	4.5	10,000		Cofield 1959
1965	Two 9-in., 2 5-in. NaI	6.5	2.2	4,900		Scott and West 1967
1983	Two 9-in., 2 5-in. NaI	5	1.7	3,800	1.96 $\sigma_{\text{BKG}}$	King and Barclay 1983
1983	Two 9-in., 2 5-in. NaI	7.2	2.4	5,300	$L_C^a$	King and Barclay 1983
1983	Two 9-in., 2 5-in. NaI	14.4	4.8	11,000	$L_D^a$	King and Barclay 1983
1991	HPGe	4.5	1.5	3,300	$b$	BWXT Y-12 2003b

- As defined by Currie (1968) with  $\alpha = \beta = 0.05$ , paired observations.
- Individual MDA or critical level supplied with analytical results.

No description of the conversion count rate to activity to mass has been found. The analyte reported was based on the area in which the employee worked. Individuals working in NU or DU areas had results reported as  $^{238}\text{U}$ , and workers in enriched areas had results reported as  $^{235}\text{U}$ . Claimant-favorable assumptions should be based on conversions of 93% enrichment for  $^{235}\text{U}$  and natural isotopic abundances for  $^{238}\text{U}$ .

### 5.3.2.2 Chest Counting for Other Actinides

#### Thorium

The *in vivo* lung count was the only monitoring technique for monitoring thorium exposure in the body during the plant's first decades. Thorium lung activity was inferred from  $^{228}\text{Ac}$  and/or  $^{212}\text{Pb}$  lung activity. Thorium lung counting was conducted from 1958 to 1984 with routine lung counts, scheduled at approximately six-month intervals, starting in 1961.

The interpretation of thorium spectra is complicated by the tendency of decay chain members to become separated during operations, particularly those involving heat. The boiling point of radium is below the melting point of thorium. The  $^{228}\text{Ac}$  gamma radiation in an *in vivo* count could be an indication of a  $^{228}\text{Ra}$  intake or an intake of the entire  $^{232}\text{Th}$  chain. The 239-keV gamma line from  $^{212}\text{Pb}$ , a progeny of  $^{228}\text{Th}$ , is a more reliable indicator of  $^{232}\text{Th}$ , as the thorium isotopes would remain together during processing. However, if the  $^{228}\text{Ra}$  is lost,  $^{212}\text{Pb}$  decreases with the 1.9-yr half-life of its parent,  $^{228}\text{Th}$ , moderated by the ingrowth of new  $^{228}\text{Ra}$  with its 5.7 yr half-life. This complicated disequilibrium

pattern operates both before and after chain members are taken into the body. Some knowledge of the likely state of equilibrium is therefore necessary to translate observed activities into dose.

The maximum permissible lung burden (MPLB) of  $^{232}\text{Th}$  (corresponding to 15 rem/yr to the lung) varied markedly with the ratio of  $^{228}\text{Th}$  to  $^{232}\text{Th}$ . For full equilibrium (ratio = 1) the MPLB was calculated to be 2.9 nCi of  $^{232}\text{Th}$ . For a ratio of 0.1, the lung burden was 12 nCi (West 1965, p. 22). For a ratio of 0.8, an MPLB of 3.2 nCi is indicated. For this ratio, with a  $^{228}\text{Ra}$ -to- $^{232}\text{Th}$  ratio of 0.6, an MDA of 0.2 lung burdens, or 0.6 nCi, is indicated (West 1965, p. 26). In mass units, this is 5.5 mg.

It was reported in 1965 that thorium at Y-12 was processed less than 1 yr after purification by the supplier and as a consequence had only about 10% as much  $^{228}\text{Ra}$  as  $^{224}\text{Ra}$  (West 1965, p. 18). This means that the maximum dose conversion factor per milligram of  $^{232}\text{Th}$  would be less than that for  $^{232}\text{Th}$  in full equilibrium with its progeny. However, unless specific information is available in the claimants' data, the dose reconstructor must make claimant-favorable assumptions. The thorium sensitivity varied due to the dependence of this technique on the ratios of  $^{232}\text{Th}$  and  $^{228}\text{Ra}$  to the progeny being measured. Data interpretation was based on a careful evaluation of the work histories. In the absence of sufficient data to determine the  $^{228}\text{Th}$ -to- $^{232}\text{Th}$  ratios and state of equilibrium, the claimant-favorable assumption is to assume full equilibrium.

### Neptunium

At the time the *in vivo* system was put into routine service in 1961, the reported detection limit for  $^{237}\text{Np}$  without progeny radiation was 2.7 nCi (Cofield 1961). For  $^{237}\text{Np}$  in full equilibrium with  $^{233}\text{Pa}$ , the detection limit was reported as 0.255 nCi (Cofield 1961).

#### 5.3.2.3 Chest Counting for Other Radionuclides

At the time the *in vivo* system was put into routine service in 1961, the reported detection limit for  $^{60}\text{Co}$  was 0.66 nCi. For  $^{95}\text{Zr}$  in transient equilibrium with  $^{95}\text{Nb}$ , the reported detection limit was 1 nCi. Although bremsstrahlung counting could have been done for  $^{99}\text{Tc}$ , no information is available about the sensitivity of the technique.

### 5.4 UNMONITORED DOSE (NOT COMPLETE FOR ALL OPERATIONS AND/OR RADIONUCLIDES)

There are two eras of operations at Y-12 for which sufficient monitoring information might not be available in the worker records. From 1943 to 1947, limited or no monitoring was performed. No data have been found for this period. The primary site activities for this period were uranium enrichment by the Calutron process. From 1948 to 1950, fluorometric analyses of urine and blood were conducted as part of general medical surveillance to prevent kidney damage from exposure to soluble uranium compounds; these data cannot be found at this time. Coworker data will be applied to this time period.

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## GLOSSARY

### **activity median aerodynamic diameter (AMAD)**

The diameter of a sphere having a density of 1 gram per cubic centimeter and the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol.

### **acute exposure**

Exposure of short duration.

### **background (radiation)**

Radiation from:

- Naturally occurring radioactive materials that have not been technologically enhanced
- Cosmic sources
- Global fallout as it exists in the environment (from the testing of nuclear explosive devices)
- Radon and its progeny in concentrations or levels existing in buildings or the environment that have not been elevated as a result of current or previous activities
- Consumer products that contain nominal amounts of radioactive material or produce nominal amounts of radiation.

### **bioassay**

The determination of kinds, quantities, or concentrations and, in some cases, locations of radioactive material in the human body, whether by direct measurement or by analysis and evaluation of radioactive materials excreted or removed from the human body. Also called *radiobioassay*.

### **chronic exposure**

Exposure of long duration, delivered by fractionation or protraction.

### **class D, W, or Y material**

ICRP 30 classifies inhaled radioactive materials as type D, W, or Y (days, weeks, or years) depending on their retention times in the pulmonary region. Class D materials have a pulmonary half-life of less than 10 d; W materials have a half-life from 10 to 100 d; and Y materials have a half-life greater than 100 d. The times actually used for these classes of materials in ICRP 30 in the calculation of annual limits on intake are 0.5 d, 50 d and 500 d for class D, W, and Y materials, respectively. Note: A class Y uranium has been defined as having an effective pulmonary half-life of 100 d and the systemic distribution, retention, and excretion parameters of class W uranium.

### **class (type) Q system**

A class Q dosimetry system uses an 8- $\mu\text{m}$  AMAD as a basis. The class Q dosimetry system is described as 10% class Y and 90% modified class W (the modification being an increase of the class W 50-d compartment to 120 d).

### **committed effective dose equivalent (CEDE)**

The sum of the committed dose equivalents to various tissues or organs in the body for 50 yr after intake. CEDE is expressed in units of rem (or sievert).

**depleted uranium (DU)**

A byproduct of the enrichment process, depleted uranium has a reduced abundance of  $^{235}\text{U}$  relative to the 0.72% abundance in natural uranium.

**dose**

A general term for absorbed dose, dose equivalent, effective dose equivalent, committed dose equivalent, committed effective dose equivalent, or total effective dose equivalent.

**enriched uranium (EU)**

Uranium with an increased abundance of  $^{235}\text{U}$ . Enriched uranium varies from greater than the natural  $^{235}\text{U}$  abundance of 0.72% up to almost 100%. Enriched uranium can also contain increased abundances of  $^{234}\text{U}$ ,  $^{236}\text{U}$ ,  $^{232}\text{U}$ ,  $^{233}\text{U}$  and others.

**exposure**

(1) The general condition of being subjected to radiation, such as by exposure to radiation from external sources or to radiation sources inside the body. (2) In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air. The product of exposure time to a radioactive aerosol and the average concentration during exposure, divided by the value of the derived air concentration for the radioactive material in question (expressed in derived air concentration-hours).

**intake**

The amount of radionuclide (mass or activity) taken into the body by inhalation, absorption through the skin, injection, ingestion, or through wounds.

***in vitro* measurement**

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

***in vivo* measurement**

The measurements of radioactive material in the human body using instrumentation that detects radiation emitted from the radioactive material in the body.

**maximum permissible lung burden (MPLB)**

During the 1970s the occupational limit for plutonium was expressed in terms of a quantity of plutonium that could be present in the chest at any given time, equal to 16  $\mu\text{Ci}$  (0.25  $\mu\text{g}$ )  $^{239}\text{Pu}$ .

**minimum detectable amount or activity (MDA)**

The smallest amount (mass) or activity (per mass or per sample) of an analyte in a sample that will be detected with a probability  $\beta$  of nondetection (type II error) while accepting a probability of erroneously deciding that a positive (nonzero) quantity of analyte is present in an appropriate blank sample (type I error).

**minimum detectable concentration (MDC)**

The minimum detectable amount expressed in units of concentration.

**monitoring**

The measurement of radiation levels, airborne radioactivity concentrations, radioactive contamination levels, quantities of radioactive material, or individual doses.

**natural uranium**

The mixture of uranium as it is found in nature (i.e., 99.27%  $^{238}\text{U}$ , 0.72%  $^{235}\text{U}$ , and 0.0054%  $^{234}\text{U}$  by weight). The alpha activity comes mostly, and in about equal amounts, from  $^{238}\text{U}$  and  $^{234}\text{U}$ . The specific activity of this mixture is  $2.6 \times 10^7$  Bq/kg (0.7 pCi/g).

**nephrotoxicity**

Poisoning of the kidney.

**quality factor**

A modifying factor used to calculate the dose equivalent from the absorbed dose.

**recycled uranium (RU)**

Uranium recovered from spent fuel from fission reactors.

**routine bioassay monitoring**

Any bioassay measurement made on a predetermined, periodic schedule, to establish a worker's internal exposure status relative to previous periods of time.

**sensitivity (detection sensitivity)**

The minimum amount of contaminant (or dose) that can be repeatedly measured (or calculated) by a particular analysis. See *minimum detectable amount*.

**type F, M, or S material**

ICRP 66 classifies inhaled radioactive materials as F, M, or S (fast, medium, or slow) depending on their retention time in the pulmonary region. These designations are similar to the clearance classes D, W, and Y, but refer strictly to the rate of absorption from the lungs to the blood.

**ATTACHMENT 5A  
OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS**

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## 5A.1 CONVENTIONS USED IN INTERNAL DOSE REPORTS

The following is adapted from Souleyrette (2003).

### 5A.1.1 Y-12 Urinalysis Data through September 1989

Dept – Department number of employee at the time of monitoring.

Badge/Old SSN – Employee identification number.

Void Date – Date sample was voided.

#### Program Code

- 1 = EU urinalysis by gross alpha
- 2 = NU or DU by fluorophotometry (used for EU before 1950)
- 6 = control sample
- 9 = (may have been used for a limited time to indicate experimental analyses methods).

Volume – volume of sample in ml.

Time – Time between last void and sample void. Used to normalize urine result to 24-hr void based on individual employee daily excretion rate. (A zero time in conjunction with a large void volume and program code 9 probably indicates an experimental measurement technique.)

Uranium Result (dpm) – The normalized urine result in disintegrations per minute per day.

Background Value – Background was stored for later results. The column Uranium Result (dpm) is background corrected already.

### 5A.1.2 Y-12 Urinalysis Data since October 1989

Dept - Department number of employee at the time of monitoring.

Badge - Employee identification number.

Sample Number – Laboratory sample identifier.

Sample Date - Date sample was voided.

#### Sample Type

- A = Alpha spec
- F = fecal
- U = urine
- S = Start or stop date

(Note: For urinalysis, A is expected because all urinalyses were done this way since October 1989. The other entries are for database flexibility.)

Reason – Reason for scheduling a sample:

1S = acute exposure

2S = follow-up sample to an acute exposure

3 = routine (chronic) exposure

2R = follow-up to routine (chronic) exposure

4 = background sample

5 = baseline sample

6 = random sample

8 = start date of exposure \*

9 = stop date of exposure \*

0 = acute exposure

\* (These should appear only in separate records introducing a series related to a chronic intake study).

Retention Class – Assigned retention class: ICRP-30 classes D, W, and Y. (Class Q indicates 90% class Super-W (120-d lung retention) and 10% class Y, 8  $\mu$ m AMAD.)

Volume (ml) or Weight (g) – sample volume (urine) or weight (fecal).

Total Uranium Result (dpm) - This is the normalized total uranium urine result in disintegrations per minute per day.

Type Assimilation

H = inhalation

I = ingestion

J = injection

Incident Date – Date of documented radiological exposure incident, if any.

Incident Time – Time of documented radiological exposure incident, if any.

### **5A.1.3 In Vivo Data**

Dept – Department number of employee at the time of monitoring.

Run Date – Date subject counted.

Run No. – Laboratory identifier of counting session.

Weight (lbs.) – Subject weight in pounds.

Chest Thickness (in.) – Subject chest wall thickness in inches (could be in tenths of inches for some years).

Surface Contamination Code – A six-digit code indicating whether or not surface contamination was detected.

- If the first position is a 1, there was no surface contamination found, and the remaining five digits should be zeros.

- If the first digit is a 2 or a 3, there was surface contamination found and the remaining five digits give the survey meter reading of the contamination.

Type Analysis – Code indicating the activity regions analyzed. There are two distinct sets of codes depending on the date of the count.

From October 1962 to 1971, the codes are:

- 0 = Background run
- 1 = Uranium
- 2 = New hires/control counts
- 3 = Thorium
- 4 = Neptunium
- 5 = Miscellaneous
- 6 = Second thorium count.

Since 1972, the codes are:

- 1 =  $^{235}\text{U}$
- 3 = Thorium
- 4 =  $^{238}\text{U}$  and  $^{235}\text{U}$
- 6 = Thorium and  $^{235}\text{U}$  (first thorium count)
- 7 =  $^{235}\text{U}$  and Thorium (second thorium count).

Material Type – This code provides additional details for cases where Type Analysis = 0, 1, 2, or 3.

For counts after 1971, where Type Analysis is 0:

- 00 = Background run
- 01 = Sensitivity Check.

For Type Analysis of 1 (any year) or Type Analysis of 2 (after 1971):

- 01 = Unknown
- 02 = DU
- 03 = DU
- 07 = Normal uranium
- 13 = EU
- 19 = EU.

For Type Analysis of 3 (any year):

- 00 = Thorium (first exam)
- 02 = Thorium (second or later exam).

U-235 ( $\mu\text{g}$ ) –  $^{235}\text{U}$  activity detected in chest cavity. If the count is invalid, an '&' appears as the first character.

Thorium (mg) –  $^{232}\text{Th}$  activity calculated from the daughter isotopes detected in chest cavity.

U-238 (mg) –  $^{234}\text{Th}$  activity detected in chest cavity.

Tc-99 ( $\mu\text{Ci}$ ) -  $^{99}\text{Tc}$  activity detected in chest cavity.

Current Dept – Additional instance of employee department number used historically for reporting purposes.

## 5A.2 URANIUM SOLUBILITY IN THE LUNG

For a workplace as varied as Y-12, it is clear that no single solubility type or particle size distribution applies to all workers. Furthermore, accurate assignment of the uranium lung clearance type to a given bioassay result is considered virtually impossible because of uncertainties about chemical form and limitations of the personnel-tracking system (Barber and Forest 1995, p. 669). Exposure to type M material from 1948 to June 1998 appears to be the most likely absorption type. After June 1998, exposure to absorption type S material is more likely. However, the absorption type can be based on the monitoring data or claimant-favorable assumptions.

## 5A.3 IN VITRO DETECTION LIMITS

Tables 5A-1 and 5A-2 summarize information developed in Section 5.2. The tabulated values for urinalysis results represent laboratory detection limits  $L_D$  and do not include uncertainties introduced by sample collection or conversion from submitted volumes to daily void volumes. As noted in Section 5.2,  $L_D$  values for some historical techniques remain to be identified, and will be reported in subsequent revisions as available.

Table 5A-1. Uranium urinalysis detection limits.

Method	Period	Detection limit (mass)	Detection limit (activity)	Convention
Fluorometry	1950–9/1989	7 $\mu\text{g}/\text{d}$	11 dpm/d	
Gross alpha	1950–1964		47 dpm/d	$L_D$
Gross alpha	1965–9/1989		26 dpm/d	$L_D$
Alpha spectrometry	10/1989–present		0.15 dpm/d	$L_D$

Table 5A-2. Other *in vitro* detection limits.

Analyte/solubility type	Method	Period	Detection limit (activity)
Tritium (HTO)	Liquid scintillation	10/1988–present	2,000 dpm/d <sup>a</sup>
Isotopic plutonium/M, S	Alpha spectrometry	10/1989–present	0.025 dpm/sample
Am-241/M	Alpha spectrometry	10/1989–present	0.050 dpm/sample
Th-228/M, S	Alpha spectrometry	10/1989–present	0.150 dpm/sample
Th-232/M, S	Alpha spectrometry	10/1989–present	0.070 dpm/sample
Np-237/M	Alpha spectrometry	10/1989–present	0.100 dpm/sample

a. Estimate.

## 5A.4 IN VIVO DETECTION LIMITS

From the review of Y-12 documents and conversations with present and former Y-12 staff, the vast majority of site operations from 1943 to the present involved uranium in a variety of chemical forms and degree of enrichment. Therefore, it is concluded that the primary internal radiation exposure to Y-12 workers was from uranium. However, the internal dosimetry program has included limited monitoring for  $^{137}\text{Cs}$ ,  $^{99}\text{Tc}$ , thorium, plutonium,  $^{228}\text{Ac}$ , and tritium, among other radionuclides. There are difficulties in interpreting these measurement data that cannot be resolved satisfactorily at this time. These issues should not have a large impact on the worker's internal dose because exposure to uranium is the source of the greatest part of the internal dose.

Table 5A-3 summarizes information developed in Section 5.3. Detection limits for some historical techniques remain to be identified, and will be reported in subsequent revisions as available.

Table 5A-3. Reported *in vivo* detection limits for other radionuclides.

Radionuclide /solubility type	Reported limit (nCi)	Reported limit (dpm)	Citation year
Th-232/M, S	0.6	1,300	1965
Np-237/M	2.7	5,900	1961
Np-237/M–Pa-233/M, S	0.255	560	1961
Co-60/M, S	0.66	1,500	1961
Zr-95/M, S–Nb-95/M, S	1.0	2,200	1961

Although internal monitoring has existed from the earliest days, the data from these measurements are not available for years before 1950. Guidance for the period from 1948 to 1950 is provided, but it is not feasible to provide any guidance for exposures before 1948. This will be provided in revisions to this document.

For dose reconstruction purposes and based on review of the Y-12 data, the <sup>235</sup>U and <sup>238</sup>U lung count detection thresholds are assumed to be 130 µg and 13.5 mg, respectively, through 1990. Beginning in 1991, the critical level or MDA is supplied with the individual results.