

ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities I Dade Moeller I MJW Technical Services

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ACRONYMS AND ABBREVIATIONS

AMAD aerodynamic median activity diameter

Bq becquerel

CEDE committed effective dose equivalent CER Center for Epidemiologic Research

Ci curie

counts per minute cpm

d day

DF decontamination factor

DHHS U.S. Department of Health and Human Services

dpm disintegrations per minute

depleted uranium (less than 0.72% ²³⁵U) DU

EU enriched uranium

FEMC Feed Materials Production Center

gram

g GSD geometric standard deviation

hr hour

highly enriched uranium (generally between 20% and 90% ²³⁵U) HEU

HPGe high-purity germanium (detector)

Idaho Chemical Processing Plant **ICPP**

ICRP International Commission on Radiological Protection

Integrated Modules for Bioassay Analysis **IMBA**

in. inch

IREP Interactive RadioEpidemiological Program

keV kilovolt-electron, 1,000 electron volts

kilogram kg

L liter

critical level (commonly 5% chance of a false positive) L_{C} detection level (commonly 5% chance of a false negative) L_{D} low enriched uranium (generally less than 20% ²³⁵U) LEU

m meter

MDA minimum detectable activity

milligram mg minute min mL milliliter millimeter mm

maximum permissible lung burden **MPLB**

mrem millirem

nCi nanocurie

NIOSH National Institute for Occupational Safety and Health Document No. ORAUT-TKBS-0014-5 Revision No. 03 Effective Date: 03/12/2012 Page 8 of 79

NU natural uranium

ORAU Oak Ridge Associated Universities

pCi picocurie ppb parts per billion ppm parts per million

RU recycled uranium
RWP Radiation Work Permit
R&D research and development

SEC Special Exposure Cohort

SMC Idaho Specific Manufacturing Capability

SRDB Ref ID Site Research Database Reference Identification (number)

SRS Savannah River Site

U.S.C. United States Code

VHEU very highly enriched uranium (greater than 90% ²³⁵U)

wt % weight percent

yr year

§ section or sections

μCi microcurie μg microgram μm micrometer

5.1 INTRODUCTION

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

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

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

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

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

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

5.1.1 Purpose

The purpose of this technical basis document (TBD) is to describe internal dosimetry systems and practices at the Y-12 Plant (now the Y-12 National Security Complex). This document discusses historical and current practices in relation to the evaluation of internal exposure data for monitored and unmonitored workers. For convenience, this TBD uses the Y-12 Plant or Y-12 to refer to this site.

5.1.2 Scope

The Y-12 Plant, operated by BWXT Y-12, L.L.C. and its predecessors, has played an important role in the development of the U.S. nuclear weapons program. Operations at the Plant have evolved from the separation and enrichment of uranium to the manufacture and assembly of nuclear weapons components to stockpile stewardship and maintenance. This TBD is part of the Y-12 Plant Site Profile, which describes Plant facilities and processes, historical information, and environmental data in relation to dose reconstruction for Y-12 workers. It contains supporting documentation to assist in the reconstruction of occupational internal doses resulting from these activities.

The methods and concepts of measuring occupational internal doses to workers have evolved since the beginning of operations at the Y-12 Plant. One objective of this document is to provide supporting technical data to evaluate internal occupational doses that can reasonably be associated with worker radiation exposures covered by the EEOICPA legislation. In addition, this document presents the technical basis of methods used to prepare Y-12 worker dose information for input into the NIOSH Interactive RadioEpidemiological Program (IREP).

At the Y-12 Plant, uranium isotopes in various chemical and physical forms have been the primary contributors to internal radiation doses to workers since November 1943. Y-12 missions have 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 TBD. However, the primary focus on internal dose control at Y-12 has been on uranium compounds and alloys over a wide range of ²³⁵U enrichment. Therefore, this section begins with an overview of concepts that apply broadly to the history of uranium work at Y-12. Attachment A summarizes some of the key points from the following sections. Attachment B provides coworker intakes for applications when bioassay records are unavailable or incomplete.

There were programs in the early years of Y-12 operations for which currently available information is insufficient to provide general guidance for internal dose reconstruction. At the peak of ²³⁵U enrichment operations from 1943 to 1945, there were 1,156 "large mass spectrographs," called calutrons in operation. This enrichment production technique was shut down in 1946.

However, the calutrons in Building 9204-3 remained operational to permit research and development (R&D) to improve the production of the calutron concept and to separate 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 began operating on November 11, 1950, and operated until the early 1980s. However, the past operation has led to internal dose concerns about polonium isotopes and alpha airborne activity (Livingston and Martin 1952). The 86-inch Cyclotron was later used to create neutron-deficient radionuclides as a part of the R&D program noted above. Some of these R&D efforts involved plutonium, and workers could have plutonium bioassay results in their records (Patterson, West, and McLendon 1957). 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 1999).

5.1.3 **Special Exposure Cohorts**

5.1.3.1 March 1, 1943 through December 31, 1947

On August 15, 2008, as provided for under 42 U.S.C. § 7384q(b), the Secretary of HHS designated the following class of employees as an addition to the SEC:

All employees of the Department of Energy (DOE), its predecessor agencies, and DOE contractors or subcontractors who worked at the Y-12 Plant in Oak Ridge, Tennessee from March 1, 1943, through December 31, 1947 for a number of work days aggregating at least 250 work days occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort (73 FR 58966; October 8, 2008).

As pointed out in the NIOSH evaluation report and the HHS Designation Letter, the class associated with 73 FR 58966, which became effective September 14, 2008, was assembled to clarify and correct the class wording defined in 70 FR 57602, because of implementation issues associated with the 70 FR 57602 SEC class definition.

Through the course of ongoing dose reconstruction and research, NIOSH has determined that, because exposure potential may not have been limited to only specific buildings or groups of workers at Y-12, it is necessary to expand the SEC class definition to include all areas of Y-12, and all employees of DOE, its predecessor agencies, and their contractors and subcontractors who worked at Y-12 during the period from March 1, 1943, through December 31, 1947. The designated class evaluation recommends a class that is consistent with current NIOSH methods for defining a recommended SEC class (NIOSH 2008).

In its evaluation (NIOSH 2008), NIOSH found that it is not possible to reconstruct internal radiation doses for Y-12 employees for all potential radiation exposures. However, NIOSH has decided that the occupational medical dose and external exposures directly associated with the calutron uranium enrichment process can be estimated. Although NIOSH found that it is not possible to reconstruct radiation doses completely for the SEC class, NIOSH intends to use limited available external dose rate measurements that might be applicable to an individual claim, and any internal and external monitoring data that might become available for an individual claim (and can be interpreted using its existing dose reconstruction processes or procedures). Therefore, dose reconstructions for individuals employed at Y-12 during the period from March 1, 1943, through December 31, 1947, but who do not qualify for inclusion in the SEC may be performed using these data as appropriate.

The assessment of NIOSH's ability to reconstruct doses from 1948 onward, is addressed in the following section.

5.1.3.2 January 1, 1948 to December 31, 1957

On October 18, 2011, as provided for under 42 U.S.C. § 7384q(b), the Secretary of HHS designated the following class of employees as an addition to the SEC:

All employees of the Department of Energy, its predecessor agencies, and their contractors or subcontractors who worked at the Y-12 facility in Oak Ridge, Tennessee, during the period from January 1, 1948, through December 31, 1957, for a number of work days aggregating at least 250 workdays, occurring either solely under this employment or in combination with workdays within the parameters established for one or more other classes of employees in the SEC (76 FR 72928; November 28, 2011).

As pointed out in the NIOSH evaluation report and the HHS Designation Letter, the class associated with 76 FR 72928, which became effective November 17, 2011, was assembled to clarify and correct the class wording defined in 71 FR 55477, because of implementation issues associated with the 71 FR 55477 SEC class definition.

Through the course of ongoing dose reconstruction and research, NIOSH has determined that, due to undocumented worker movements across the site, limited claimant-specific information about work locations, and a determination by DOL that employment records do not indicate work locations, it is unable to eliminate any specific worker from potential exposure scenarios based on assigned work location. NIOSH has found that a determination cannot always be made about the specific area an employee worked in, or whether an employee should have been monitored for radiological exposures. Accordingly, NIOSH has determined that it is necessary to remove the area-specific and monitoring criteria from the class description associated with SEC-00028 thorium and cyclotron exposures for the period from January 1, 1948, through December 31, 1957. NIOSH has determined that it is also necessary to expand the SEC class definition to include all areas of Y-12, and all employees of DOE, its predecessor agencies, and their contractors and subcontractors who worked at Y-12 during the period from January 1, 1948, through December 31, 1957. The designated class evaluation recommends a class that is consistent with current NIOSH methods for defining a recommended SEC class (NIOSH 2011).

In its evaluation (NIOSH 2011), NIOSH found that it is not possible to reconstruct internal radiation doses for Y-12 employees with internal exposures to thorium and cyclotron radionuclides. However, NIOSH has decided that the occupational medical dose and external exposures can be estimated. Although NIOSH found that it is not possible to reconstruct radiation doses completely for the SEC class, NIOSH intends to use any internal and external monitoring data that may become available for an individual claim (and that can be interpreted using existing NIOSH dose reconstruction processes or procedures). Therefore, dose reconstructions for individuals employed at Y-12 during the period from January 1, 1948, through December 31, 1957, but who do not qualify for inclusion in the SEC, may be performed using these data as appropriate.

5.2 **URANIUM SOLUBILITY IN THE LUNG**

The uranium compounds that have been used at Y-12 range from highly soluble to very insoluble (ORAUT 2007). 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, very few urinalysis data have been found for Y-12 before 1948. 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 in International Commission on Radiological Protection (ICRP) Publication 2 (ICRP 1959) or by similar predecessor models. These models related lung dose to uranium excretion in urine, particularly under chronic exposure conditions.

Insoluble material described in ICRP Publication 2 (ICRP 1959) would be classified as moderately soluble type M material in the framework of ICRP Publication 66 (ICRP 1994). In addition, 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 1995a). The ICRP Publication 2 insoluble model and the class W models fit within the range of type M material.

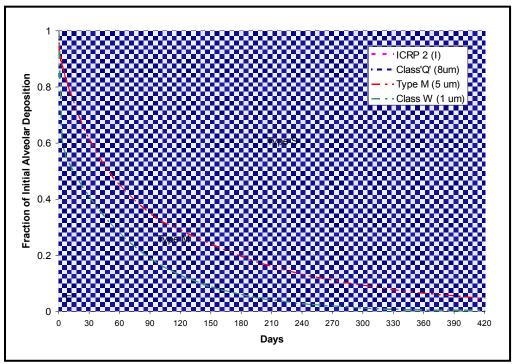


Figure 5-1. Historical solubility types meeting the type M definition.

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. West and Scott (1969) noted that, "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 nonuranium activities." The job description of all individuals was consistent (Snapp 2003b; Barber and Forrest 1995).

Subsequent investigations suggested that three process combustion ashes from the EU foundry were the most likely exposure materials for these 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 µm), 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 (ICRP 1959) 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-µm aerodynamic median activity diameter (AMAD) excretion curves (Potter 2002). They were normalized to match the Scott curve at 200 days 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 guite 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-year 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 was 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 (Eckerman and Kerr 1999, p. 28).

For a workplace as varied as Y-12, it is clear that no single solubility or particle size would apply to all workers. Further, 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 Forrest 1995, p. 669). As of 2001, Y-12 implemented a radiation work permit (RWP) tracking system that tracks the locations of workers, their types of work, and the types of bioassay needed (ORAUT 2007, pp. 19 ff.).

Due to the many different processes and types of work performed at Y-12, and the potential range of solubility types associated with materials and work locations, the absorption type can be based on the monitoring data, assumptions that are favorable to claimants, or both.

5.2.1 Particle Size Information

Several particle size studies in uranium process areas have been conducted at Y-12 (e.g., Ballenger et al. 1953; Steckel and West 1966; Barber and Forrest 1995; Snapp 2003a). For different times and different processes, reported particle sizes ranged from less than 1 to greater than 10 µm (physical).

Steckel and West (1966) reported a positive correlation between uranium oxide particle size and process temperature. Barber and Forrest (1995) used an 8-µm AMAD based on particle size measurements as the basis for the class Q dosimetry system used in the 1990s. (The class Q system is described as 10% class Y and 90% modified class W. The modification consists of increasing the class W 50-d compartment to 120 days.) In terms of lung deposition and retention, the 8-µm AMAD class Q material is closer to 5-µm AMAD type M material than to 1-µm AMAD class W material, as shown in Figure 5-3.

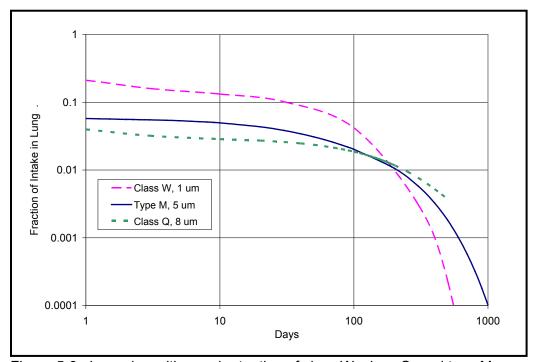


Figure 5-3. Lung deposition and retention of class W, class Q, and type M uranium. (Particle sizes specified are in micrometers AMAD.) (Barber and Forrest 1995).

Since 2000, Y-12 has implemented the latest guidance provided by the ICRP including the Publication 66 (ICRP 1994) lung model and Publication 78 (ICRP 1998) methods and models (Veinot 2003, p. 89). This is the justification for the change to the 5- μ 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 μ m AMAD.

5.2.2 <u>Uranium Alpha Activity as a Function of Enrichment</u>

At Y-12, uranium enrichments range from depleted uranium (DU; less than 0.71 wt % ²³⁵U) to very highly enriched uranium (VHEU; greater than 93 wt % ²³⁵U). Section 5.2.4.1 lists the uranium isotopic mixes with 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 (BWXT Y-12 2000).

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.(Patterson 1958). Early *in vivo* chest-counting results were reported either as micrograms of ²³⁵U or milligrams of ²³⁸U based on whether an exposure was believed to be from EU, natural uranium (NU), or DU (Cofield 1960; Scott and West 1975). Based on these reporting

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techniques, the assumptions in Table 5-1 can be made about data interpretation for the periods from 1950 through 1989 for urinalysis and from 1961 to 1989 for lung counting.

Table 5-1. Enrichment assumptions for dose assessment for 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–present).	Alpha dpm/d is reported isotopically and total uranium is determined by summing isotopic results.	
Lung count Lung count	μg U-235 μg U-238	93% enrichment.

For lung counts, a combination of the information in the Type Analysis and Material Type (see Section A.1.3 in Attachment A for details) reporting fields can be used to determine if 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.

For dose reconstruction purposes, the individual isotopes of a uranium mixture intake (234 U, 235 U, 236 U, and 238 U) can be summed for the dose assessment and the 234 U dose conversion factor applied. 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 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). Note that 234 U dose conversion factors are not appropriate substitutes for 232 U, as the DCFs for 232 U are typically 5-10 times larger than those for 234 U.

5.2.3 Temporal Pattern of Uranium Exposures

As a rule, routine uranium exposures at Y-12 were considered to be of a chronic nature. For example, Patterson (1958, p. 58) stated, "...our interpretation of urinalysis results and our assignment of internal dose assume an exposure under equilibrium conditions of intake and elimination." 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" (Veinot 2003, 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 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 TBD, 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 in Section 5.2.4.1 for plant-wide RU contaminant defaults).

5.2.4 Other Radionuclides of Concern Including Recycled Uranium Contaminants

In addition to ²³⁴U, ²³⁵U, and ²³⁸U, the following radionuclides were identified in the Y-12 Technical Basis Document for Internal Dosimetry (Snapp 2003a):

- ³H. ⁹⁰Sr. ⁹⁹Tc
- ²²⁸Th, ²³²Th ²³²U, ²³³U, ²³⁶U
- ²³⁸Pu, ²³⁹Pu, ²⁴¹Pu
- ²³⁷Np. ²⁴¹Am

Radionuclides that could interfere with *in vivo* analysis of uranium and thorium (i.e., ⁴⁰K and ¹³⁷Cs) were quantified so their effects on the spectra could be taken into account.

Other radionuclides addressed included ⁶⁰Co and ⁹⁵Zr/⁹⁵Nb for organizations outside Y-12 (Cofield 1961).

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 ¹³¹I, ^{99m}Tc, ⁶⁷Ga, and ²⁰⁸TI.

Unmonitored Radionuclides from Recycled Uranium 5.2.4.1

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 (BWXT Y-12 2000). These contaminants were a result of fission and activation processes of a variety of uranium enrichment isotope mixtures that 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, ²³⁶U was produced in quantities greater than "trace" levels) (BWXT Y-12 2000).

The major source of RU contaminants at Y-12 was the receipt and processing of returned VHEU from the Savannah River Site (SRS) and the Idaho Chemical Processing Plant (ICPP) (Rich et al. 2000). 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 computergenerated 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 [UO₂(NO₃)₂] solutions, uranium trioxide (UO₃), uranium-aluminum (U-Al) alloy ingots, uranium scrap, uranium tetrafluoride (UF₄), uranium metal, solvent extraction raffinates, and a variety of secondary process wastes and residues (Rich et al. 2000; DOE 2007).

The uranium enrichments ranged from DU, NU, low enriched uranium (LEU - less than 20% ²³⁵U), highly enriched uranium (HEU - from 20 to approximately 93% ²³⁵U), and VHEU (more than 93% ²³⁵U) (Rich et al. 2000). The predominant RU and associated contaminants were in HEU and VHEU materials. However, all the uranium at Y-12 came from other DOE facilities, which had either generated or received RU materials (Rich et al. 2000). 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 the Feed Materials Production Center (FEMC) in Fernald, Ohio, which also had an RU contaminant inventory. Therefore, the DU contaminant levels at Y-12

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

			Pu		Np			Тс		
Site	U (kg)	g	ppb	pCi/µg U	g	ppm	pCi/µg U	g	ppm	pCi/µg U
Y-12 VHEU Receipts Summary – Rich et al. (2000)										
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
EU Total	150,857	0.04674	0.3	4.3E-03	3,666	23	1.6E-02	14,499	96	1.64
Y-12 DU Receipts Summary - Rich et al. (2000)										
Hanford	1,502	No data								
K-25	192,836									
PGDP	38,423									
DU total	232,761				•			•	•	
Grand total	383.618				·			•	·	

a. SRS = Savannah River Site; ICPP = Idaho Chemical Processing Plant; K-25 = Oak Ridge Gaseous Diffusion Plant; PGDP = Paducah Gaseous Diffusion Plant.

Table 5-3. Total Y-12 recycled uranium receipts summary (DOE 2007).

	U		Р	u	Np			Тс		
Shipping site	(kg)	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									
Total	184,000	0.5	2.7	0.039	54	0.003	0.00021	9,100	49.5	0.85

Table 5-4. Primary contaminants (at 3-year decay) in VHEU fuels and of concern in internal dose reconstruction (Wenzel 2000, 2004).

	,	Activity	Activity	Percent weight		Activity ratio
Isotope	Half-life	(μCi/μg total U)	(µCi/g total U)	g/100 g U ^a	ppm U	(isotope/U)
Tc-99	2.11E+05 yr	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 yr	1.6E-07	1.6E-01	1.9E-08	1.9E-04	0.01
Th-229	7.34E+03 yr	1.4E-12	1.4E-06	6.7E-10	6.7E-06	
Th-230	7.7E+04 yr	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 yr	3.5E-15	3.5E-09	3.2E-06	3.2E-02	2.2E-10
Total			1.9			
U-232	6.89E+01 yr	2.7E-07	2.7E-01	1.3E-06	1.3E-02	
U-233	1.59E+05 yr	2.6E-10	2.6E-04	2.7E-06	2.7E-02	
U-234	2.46E+05 yr	6.6E-07	6.6E-01	1.06E-02	1.0E+04	
U-235	7.04E+08 yr	1.7E-06	1.7E-00	7.8E+01	7.8E+05	
U-236	2.34E+07 yr	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	4.41E-07	
U-238	4.47E+09 yr	5.6E-09	5.6E-03	1.7E+00	1.7E+04	
Total			15.7			
Np-236	1.15E+05 yr	9.4E-11	9.4E-05	7.2E-07	7.2E-03	
Np-237	2.14E+06 yr	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 yr	4.3E-08	4.3E-02	1.9E-11	1.9E-07	
Total			9.1			
Pu-236	2.851 yr	3.6E-06	3.6E+00	6.8E-07	6.8E-03	0.2
Pu-238	87.74 yr	3.6E-02	3.6E+04	2.1E-01	2.1E+03	2.3E+03
Pu-239	2.41E+04 yr	1.9E-05	1.9E+01	3.1E-02	3.1E+02	1.2
Pu-240	6.54E+03 yr	1.5E-05	1.5E+01	6.4E-03	64	1.0

Isotope	Half-life	Activity (μCi/μg total U)	Activity (μCi/g total U)	Percent weight g/100 g U ^a	ppm U	Activity ratio (isotope/U)
Pu-241	14.4 yr	1.5E-03	1.5E+03	1.4E-03	14	1.0E+02
Pu-242	3.73E+05 yr	8.8-E-09	8.8E-03	2.3E-04	2.3	5.6E-04
Total			4.6E+04			
Am-241	4.32E+02 yr	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 burnup). The typical after-burnup 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-burnup enrichments as representative, with typical 1%+ for U-234.

were inferred from the FEMC and Idaho Specific Manufacturing Capability (SMC) projects recycle reports (ORAUT 2004; Lewis et al. 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 enables 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 report generated as a consequence of a comprehensive study of RU in DOE facilities in 2000. The report for Y-12 was Recycled Uranium Mass Balance Project Y-12 National Security Complex Site Report (BWXT Y-12 2000). There were some recognized inconsistencies in shipping and receipts among the DOE facilities in the DOE 2000 RU reports. In an attempt to resolve these inconsistencies, a summary report was issued by the DOE Office of Security, Recycled Uranium, United States Production, Enrichment, and Utilization (DOE 2003). Although it corrected some inconsistencies, there remain significant inconsistencies between the two reports. For 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 in the BWXT Y-12 (2000) report without documenting the associated RU contaminant levels. However, these uranium receipts also contained low-level RU contaminants, the concentrations of which are discussed later in this report.

The results of the DOE (2003) report are presented in Table 5-3. As indicated, there are significant differences with the results recorded in the BWXT Y-12 (2000) report (summarized in Table 5-2) and those in Table 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." In addition, the detail in DOE (2003) related to uranium enrichment and specific RU contaminant concentrations from each site was not provided (note the lack of detail in Table 5-3). Examination of DOE (2003) indicates 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, FMPC, and others, which also had RU contaminant inventories. Thus, the discrepancies in the current values reported in BWXT Y-12 (2000) and DOE (2003) could result from documenting the primary shipment and receipts in DOE (2003), while BWXT Y-12 (2000) accounts for secondary shipments as well. However, where large differences occur, the value most favorable to the clamant value is used to derive the defaults for dose reconstruction, which are generally the results in BWXT Y-12 (2000).

Though plutonium, neptunium, and technetium isotopes were analyzed and documented in all the facilities as the primary isotopes of concern, there was a significant level of ²²⁸Th detected in nearly all the facilities at Y-12 and in reported process stream analyses (BWXT Y-12 2000). To establish the source of this isotope and provide a basis for including it in the RU contaminants to be considered in internal dose reconstruction, Tables 5-4 and 5-5 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 the Rich et al. (2000) RU report at the ICPP. The ORIGIN2 computer program results illustrate the production of isotopes during use of VHEU fuels (Wenzel 2000, 2004). The isotopes of interest for this illustration are the uranium isotopes and plutonium, neptunium, technetium, and thorium. Thorium-228 is a contaminant that became an isotope of concern at Y-12, which was the primary receiving site of recycled VHEU fuel materials (BWXT Y-12 2000).

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-burnup VHEU reactor fuel	Approximate derived DF
Th-228	0.01-0.03	NA^a	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

a. There were no analytical data for Th-228 reported in the ICPP Y-12 receipt reports. The decontamination factor (DF) is interpolated from Y-12 process stream analytical data. These values were taken directly from column 4 in Table 5-4.

Comparing the concentrations in the spent fuels with the reported concentrations in the product received at Y-12 provides a rough indication of the DFs 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 × 10⁷ to 10⁸, with a neptunium DF of approximately 1 × 10⁴, 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 Th in the VHEU product received at Y-12. Although 228 Th has not been considered an RU contaminant of concern at 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 that included 228 Th (Oliver 1997; BWXT Y-12 2001). These surface contaminant on surveys of a large number of Y-12 plant facilities indicate a predominance of 228 Th as a contaminant (BWXT Y-12 2001). The uranium-to-nonuranium alpha ratios range from 4 to more than 1 × 10⁴, which is not useful for dose reconstruction. However, these data do indicate that 228 Th is one of the primary contaminants at Y-12, which in turn provides a technical base for inclusion of 228 Th in the list of RU contaminant isotopes.

Table 5-6. Ranges of reported contaminant values in the main processing facilities (summary of tables on pp. B-3 and B-4 of BWXT Y-12 2000).^a

Contaminant	Range	Ave. of range	Ave. of range minus the high	Raffinate values ^b
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

- a. The highest values reported in the processing facilities were in just one of 17 process streams - the solvent extraction purification of NU. Uranium levels are still significant in this stream, which indicates a marked enrichment of contaminants in relation to U in this stream.
- b. 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 in relation 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 1.2E+4 and average 3.7E+3 pCi/g sludge).

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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 (Snapp 2003a). 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 listed in Table 5-7, which is an average representation of six specific generalized enrichment types handled at Y-12. The uranium-to-nonuranium 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 tended 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 the information discussed above to represent the maximum values of the documented RU contaminant levels in the Y-12 process streams and, thus, represent values that are favorable to claimants and that can apply on a plant-wide basis. The application of the default level should be based on the indication by the bioassay of the likelihood of VHEU/HEU or LEU/NU/DU. The appropriate default ratio should be chosen based on the ²³⁵U or ²³⁸U indicated in the bioassay results. For each microgram or picocurie 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.

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

Uranium	Activity		Activity		Activity
isotope	(Ci/g)	Mass fraction	(pCi/µg total U)	RU cont.	(pCi/µg total U)
High burn-up (~		HEU from SRS ^D			
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	
Lower burn-up (~25%) recycled	VHEU from ICPP	j		
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	5.71E-03	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	
Weapons-grade	Oralloy VHEU ^{b, '}	C			
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	
Recycled NU ^d					
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

Uranium	Activity		Activity		Activity
isotope	(Ci/g)	Mass fraction	(pCi/µg total U)	RU cont.	(pCi/µg total U)
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	
Recycled LEU (2	2%) ^a				
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	
Recycled DU ^a					
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 weapons-grade Oralloy VHEU and other burn-up example VHEU were derived from Y-12 data and ICPP reports.
- c. The Oralloy 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").
- d. The recycled DU, recycled NU, and recycled LEU values were derived from FMPC data because this site supplied and received uranium to and from all DOE sites, including Y-12. In addition, the recycled DU information was verified by the analytical data reported by the Idaho SMC facility in (Lewis et al. 2000).

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

Table 6 6. Processing and an analysis and a second control of the					
	Default levels (pCi/µg U)		Default levels (pCi/pCi total U) ^a		
Isotope	VHEU & Oralloy	LEU, NU, & DU	VHEU & Oralloy	LEU, NU, & DU	
Tc-99	4	0.3	4E-02	0.15	
Th-228 ^b	0.5	0.003	5E-03	1.5E-03	
Np-237	0.6	0.003	6E-03	1.5E-03	
Pu-238 ^c	0.2		2E-03		
Pu-239 ^c	0.2 (Oralloy only)	0.003	2E-03-Oralloy	1.5E-03	
Total non-U	1.3	0.01			
Total U	75	2			

- a. Reference to Table 5-7 indicates activity in picocuries per gram of total U for VHEU ranges from 71 to 101; 100 is used for simplicity. For LEU, NU, and DU, the picocuries per gram of total U ranges from 0.9 to 1.6; 2 pCi/q total U is used for simplicity.
- b. 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 is further validated in the process stream analyses documented in BWXT Y-12 (2000).
- c. 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. *In Vitro* Minimum Detectable Activities, Counting Methods, and Reporting Protocols

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, the default levels in picocuries per gram of total U in Table 5-8 can be used to add the RU contaminant activity to the intake. However, if the bioassay units are in uranium alpha activity per unit urine volume and/or U isotope activity units, the default levels in pCi/pCi total U in that table can be used. Again, these values are maximum values expected, while

average values are basically a factor of 10 less than the values for plutonium, neptunium, and thorium and a factor of 3 less than those 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 than those for technetium) should be used.

Reference to Table 5-8 with relationship to the plutonium contaminant, the recommended isotope ²³⁹Pu should be assumed for Oralloy only, because Oralloy at Y-12 did not come directly from the primary RU generating sites (i.e., it was enriched at the gaseous diffusion plants, where ²³⁹Pu was dominant in comparison with ²³⁸Pu) (BWXT Y-12 2000). In most instances the enrichment of uranium is unknown or varies, therefore, the assumption of Oralloy can be used as a default for the majority of dose reconstructions as a favorable to claimant assumption.

The existence of air contamination and personnel exposures from ²³²Th operations is not addressed in this section. This section deals only with RU contaminants.

5.3 IN VITRO URINE ANALYSIS

5.3.1.1 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).

There is not sufficient information on radiological activities for dose reconstruction during the period from March 1943 through December 1947, including the development of beneficial radiological isotopes, development and testing of a neutron monitor, maintenance and use of a large ²²⁶Ra sealed source, and thorium extraction.

Beginning from 1948, the occupational environmental internal dose from uranium can be calculated using data in the Y-12 National Security Complex – Occupational Environmental Dose (ORAUT 2005).

The earliest fluorometry data was dated 1948. In 1950, workers in production areas were placed on a uranium fluorometry urinalysis program for 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 (Veinot 2003, 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, they were reported as isotopic results based on alpha spectrometry analysis (Souleyrette 2003). Analyses for other radionuclides were performed on an as-needed basis.

5.3.1.2 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-hour absence from the work area. [The July 1 to December 31, 1951, *Health Physics Progress Report* (Ballenger 1952, p. 25), 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 (McRee, West, and McLendon 1965, p. 25):

$$dpm / d = 8 \times (AP \div Eff) \times (Vol \div TI)$$
(5-1)

where:

8 = A constant incorporating time, count, and volume constants, including the 20-mL electroplating aliquot volume. [There is no information in McRee, West, and McLendon (1965) on what the constant includes or what the daily excretion was.]

AP = Average number of counts in 30 minutes on a plate (disk)

Eff = Uranium recovery efficiency (%)

Vol = Volume of total void (mL)

TI = Time between sample void and previous void (hr)

The term (*Vol* ÷ *TI*) also appears in the corresponding equation for fluorometric determinations of NU and DU (McRee, West, and McLendon 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; the corresponding detection level is discussed below.

Since 1989, routine samples have been collected over a 24-hour period, typically while the employee was on a scheduled break from the workplace. Many workers have elected to submit a *simulated* 24-hour 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 (Snapp 2003a, 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 (McLendon 1963). The 1963 edition of the *Y-12 Radiation Safety Manual* (McLendon 1963) 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 (Ashley et al. 1992, p. 22).

The current practice is to schedule samples based on the RWP use 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 days after entry date. If the RWP specified urine only, a urine sample is scheduled approximately 90 days after entry date (Snapp 2003a).

5.3.1.3 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 µg/d

for a nominal 1.4 L/d urine excretion rate (Struxness 1949a, CCCC 1951). In 1952, the instrument sensitivity and sample volumes indicated a minimum detectable activity (MDA) of 7 μ g/d for 1.4 L/d of urine excreted (Ballenger et al. 1953, p. 28).

The activity was calculated from the fluorometric mass reading for NU using a specific activity of 1.55 dpm/µg. For EU (93 wt % 235 U), a specific activity of 150 dpm/µg was used. The corresponding limits of detection are 11 dpm/d for NU and 1,100 dpm/d for EU (93 wt % 235 U). At times, procedures called for the direct conversion of fluorometer current into 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 µg/d or 11 dpm/d for NU. From 1948 to 1950, the MDA was 7 µg/d or 1,100 dpm/d for EU (93 wt % 235 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 minutes 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, 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 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 (McLendon 1963, p. 38). Counters were not used if the background was greater than 5 counts in 30 minutes (Hamrick 1958, p. 7). For a disk counted for 120 minutes (in fact, two disks were counted for 60 minutes 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% (McRee, West, and McLendon 1965 p. 33), L_D would be reduced to 25 dpm/d. In reviewing historical gross alpha urinalysis data, Barber and Forrest (1995) reported a decision level of approximately 20 dpm/d. Average uranium recoveries between 1958 and 1965 have not been identified, and it is not clear that the statistical convention used by Barber and Forrest 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 (Currie 1968). 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 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 $\underline{0.15}$ dpm/d (Souleyrette, Snapp, and Veinot 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 were from 1957), tritium was counted by reacting urine with calcium to evolve hydrogen gas, which was counted using a vibrating reed electrometer. The detection limit for this method is 1 μ Ci/L (1.4 μ Ci/d) (Hursh 1958). This MDA is far below the Plant action level of 0.25 μ Ci/mL (350 μ Ci/d) (Patterson, West, and McLendon 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 (Hursh 1958).

Plutonium in Urine by Gross Alpha Counting

The plutonium in urine procedure used before 1988 involved chemical separation from a 24-hour void or a simulated 24-hour sample before gross alpha counting. The detection limit has not 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 minutes), chemical recovery (40% to 73%), and counting efficiency (0.5 cpm/dpm) to the early uranium-counting method, the MDA for a 24-hour plutonium sample (less 40 mL) would range from about 0.7 to 1.3 dpm/sample. (Inferred from uranium MDA)

Plutonium in Urine by Alpha Spectrometry (1988 to Present)

The *a priori* L_D for both ²³⁸Pu and ^{239,240}Pu is 0.025 dpm/sample, or 0.025 dpm/d, assuming a full 24-hour void volume. Actual sample-specific MDAs are included in the analysis reports (Snapp 2003a, Table 4.2, p. 92).

Other Actinides in Urine and Feces

Table 5-9 lists the analytical laboratory L_D values used since about 1988 (Snapp 2003a, p. 107).

Table 6 6: ED Valace:					
Radionuclide	L _D (dpm/sample)				
Am-241	0.05				
Pu-238	0.025				
Pu-239	0.025				
Th-228	0.15				
Th-232	0.07				
Np-237	0.1				

Table 5-9. L_D values.

Uranium in Feces

Fecal sampling was started in 1999 when the presence of a less soluble component was identified as a result of the stand-down (Veinot 2003, p. 25). In 2002, of 2,800 participants in the bioassay program, 700 Y-12 workers participated in the fecal-sampling program (Souleyrette, Snapp, and Veinot 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 (Veinot 2003, p. 80)]. At present, the Analytical Chemistry division cites the value of 0.15 dpm/sample as a representative MDA for fecal analysis (Souleyrette, Snapp, and Veinot 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 (Veinot and Souleyrette 2003, p. 7).

5.3.2 In Vitro Methods for Individual Radionuclides

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

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5.3.2.1 In Vitro Bioassay for Uranium

Overview

The *in vitro* bioassay program for uranium at Y-12 can be divided into four eras.

- 1. From 1943 to 1947, limited monitoring occurred, for which the data cannot be retrieved (NIOSH 2011).
- 2. 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.
- 3. 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-day 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.
- 4. After October 1989, routine 24-hour 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, Reiss, and Young 1944). By 1946, fluorometric analyses for uranium in urine and blood were in general use to supplement clinical surveillance for soluble uranium exposures (Sterner 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 (Struxness 1949b, p. 7; CCCC 1951, p. 14). By the first half of 1952, a technique involving small (0.2-mL) aliquots of raw urine had been put in place (Ballenger et al. 1953, p. 28). However, after this change in technique was instituted, the urinary uranium concentrations increased greatly, as listed in Table 5-7. This raises the concern that the technique used before May 1952 could have underestimated the urinary uranium concentrations. (See Section 5.4.2 for a 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., micrograms per liter), but results were often converted to disintegrations per minute per day with the assumed specific activity of 1.55 dpm/ μ g for NU. The fluorometric technique had an industry standard sensitivity of about 5 ppb (5 μ g/L) (e.g., Struxness 1949a, 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 μ Ci) corresponding to an ICRP Publication 2 (ICRP 1959) 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 (Struxness 1949b, 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 (Ballenger 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 disks per sample. Each disk was counted once on one proportional counter for 30 minutes 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 (McLendon 1963, 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 minutes 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% (McRee, West, and McLendon 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., Scott 1963, p. 6). The *in vivo* monitoring frequency was determined by urinalysis results and ranged from monthly to once each 18 months (Scott 1963, p. 1). Almost all 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-day 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 ²³³U from a nitrate solution (²³³U 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 ²³²U, 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 ²³³U to ²³²U varied with operation and time. Table 5-10 lists 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.

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Table 5-10. ²³³U to ²³²U ratios.

Process	U-233:U-232 activity ratio
Reduction of UF ₄	1.3
Casting of metal-crucible skull	15–32
Machining final uranium component	160

West and Roberts (1962)

Due to uncertainty in the process, assumptions that are favorable to claimants 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 U tracer, are treated with nitric acid (HNO₃) and hydrogen peroxide (H₂O₂). The sample is wet-ashed, and the uranium is coprecipitated with calcium oxalate (CaC₂O₂). After dissolving the precipitate in hydrochloric acid (HCl), the uranium is further separated by ion exchange chromatography. The uranium is eluted from the column with a solution of dilute HCl to which titanous chloride (TiCl₃) 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 (HF). Next, the uranium isotopes are coprecipitated on neodymium fluoride (NdF₃). The NdF₃ is caught on a 0.1-mm filter, rinsed, dried, and mounted on a planchet for alpha spectrometry (Veinot 2003). Typical recoveries are about 85%. Samples are typically counted for 16 hours on a passivated implanted planar silicon detector.

5.3.2.2 In Vitro Bioassay for Plutonium

Plutonium in Urine by Gross Alpha Counting

Certain workers in the Special Testing Department were tested for plutonium intake by urinalysis (McRee, West, and McLendon 1965, p. 35). Because large urine volumes were required, these employees submitted 24-hour samples through the use of take-home kits. The sampling frequency was monthly (Patterson, West, and McLendon 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-hour 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 U, 242 Pu, and 243 Am tracers, are treated with HNO $_3$ and H $_2$ O $_2$. The sample is wet-ashed, and the uranium, plutonium, and americium are coprecipitated with CaC $_2$ O $_2$. After dissolving the precipitate in 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 NdF $_3$. The NdF $_3$ is caught on a 0.1-mm filter, rinsed, dried, and mounted on a planchet for alpha spectrometry (Veinot 2003). The same

5.3.2.3 *In Vitro* Bioassay for Tritium

procedure is used for isotopic thorium.

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 (Patterson, West, and McLendon 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 (Veinot 2003). Exposure should be assumed to be due to HTO.

5.3.2.4 In Vitro Bioassay for Other Radionuclides

Alpha spectrometry procedures described in Section 3.2 of Snapp (2003a) have been in use since 1989 and include methods for neptunium, plutonium, americium, and thorium. In addition, liquid scintillation methods for strontium and technetium were sometimes performed.

5.3.3 <u>Fecal Sample Analysis</u>

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 other areas of the Plant in 2000 and 2001 (Souleyrette, Snapp, and Veinot 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 days (Souleyrette, Snapp, and Veinot 2002, p. 14) with 24-hour fecal collections preferred. When 24-hour 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 normalize results routinely to Reference Man (Snapp 2003a, p. 72). These normalized results or actual individual daily excretion results can be used to reconstruct dose.

5.3.4 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 in the performance of 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. This section presents information about Plant practices to correct excretion sample results for dietary 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 (Veinot 2003, 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). In addition, the ratios between ²³⁴U and ²³⁸U can provide information to help characterize the sample. Natural background uranium typically has a ²³⁴U-to-²³⁸U 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 (Snapp 2003a).

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. Corrections for samples from the early days when MDAs were significantly larger are not required.

To adjust results for background, the ²³⁴U-to-²³⁸U 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 ²³⁴U-to-²³⁸U ratios greater than 1:1 are considered to be consistent with EU, and all samples with ²³⁴U-to-²³⁸U 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 ²³⁴U to ²³⁸U is 1:1 for dietary uranium.
- b. Determine if the ²³⁸U result exceeds its corresponding critical level value.
- c. If the ²³⁸U result is greater than the critical level, subtract two times that ²³⁸U result from the total uranium result.
- d. If the ²³⁸U result is less than its corresponding critical level value, do not subtract a dietary component.

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

a. The following equations are solved to determine an equation for the occupational ²³⁸U content and the occupational ²³⁴U content in a DU sample:

$$\frac{^{234}U_{\text{diet}}}{^{238}U_{\text{diet}}} = 1 \tag{5-2}$$

$$\frac{^{234}U_{\text{occ}}}{^{238}U_{\text{occ}}} = 0.2 \tag{5-3}$$

$${}^{238}U_{\text{actual}} = {}^{238}U_{\text{diet}} + {}^{238}U_{\text{occ}}$$
 (5-4)

$$^{234}U_{\text{actual}} = ^{234}U_{\text{diet}} + ^{234}U_{\text{occ}}$$
 (5-5)

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

$$^{234}U_{\text{actual}} = ^{238}U_{\text{diet}} + 0.2(^{238}U_{\text{occ}})$$
 (5-6)

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

$$\frac{{}^{234}U_{\text{actual}} - {}^{238}U_{\text{actual}}}{-0.8} = {}^{238}U_{\text{occ}}$$
 (5-7)

c. Once the occupational ²³⁸U quantity is known, the occupational quantity of ²³⁴U can be derived using Equation 5-3:

$$^{234}U_{\rm occ} = 0.2(^{238}U_{\rm occ}) \tag{5-8}$$

- d. Ensure that both the ²³⁴U and ²³⁸U results exceed their corresponding critical level values. If both results are not greater than their corresponding critical level values, a background determination cannot be made. In this case, it is assumed that all the activity observed in the total uranium sample result will contribute to occupational dose.
- e. Use either Equation 5-7 or 5-8 to determine the occupational ²³⁸U component or the occupational ²³⁴U component, respectively. For ease of reference, the ²³⁸U 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:

$$^{238}U_{\text{actual}} - ^{238}U_{\text{occ}} = ^{238}U_{\text{diet}}$$
 (5-9)

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

$$U_{\text{tot}} - 2\left(^{238}U_{\text{diet}}\right) = U_{\text{totalocc}} \tag{5-10}$$

5.4 IN VIVO MINIMUM DETECTABLE ACTIVITIES, COUNTING METHODS, AND REPORTING PRACTICES

5.4.1 Whole-Body Counting

Whole-body counting was not routinely practiced at Y-12 (Snapp 2003a). The primary *in vivo* detection method was chest counting, as described below.

5.4.2 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, 1960). A 9- by 4-in. sodium iodide (thallium doped) [Nal(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 surveyed for remaining surface contamination before entering the counting room. The normal counting time was 20 minutes.

Incremental improvements in the system were made over time. A second 9- by 4-in. NaI(TI) crystal was placed under the subject's back in 1963 or 1964. 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(TI) crystals were added, positioned at the sides under the arms. In 1985, initial development of a high-purity germanium

(HPGe) counting system was begun. This system began routine operation in a new counting room in June 1985 (Souleyrette 2003).

5.4.2.1 Chest Counting for Uranium

With the Nal 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. Measured lung burdens were originally expressed in micrograms of ²³⁵U for EU and milligrams of ²³⁸U for NU or DU. The original limits of detection for ²³⁵U and ²³⁸U were reported as 130 µg 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. Limits are given in terms of 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.

Table 5-11. Reported lung counting detection limits for ²³⁵U.

		U-235	U-235	U-235	Reporting	
Year	Detectors	(µg)	(nCi)	(dpm)	convention	Reference
1959	One 9-in. Nal	130	0.28	620		Cofield 1959
1963	Two 9-in. Nal	96	0.21	470		Scott and West 1967
1965	Two 9-in., 2 5-in. Nal	72	0.16	360		Scott and West 1967
1975	Two 9-in., 2 5-in. Nal	70	0.15	330		Scott and West 1975
1983	Two 9-in., 2 5-in. Nal	62	0.13	290	1.96 σ _{BKG}	King and Barclay 1983
1983	Two 9-in., 2 5-in. Nal	68.5	0.15	330	L _C ^a	King and Barclay 1983
1983	Two 9-in., 2 5-in. Nal	137	0.30	670	L_D^a	King and Barclay 1983
1990	Two 9-in., 2 5-in. Nal	70	0.15	330	(b)	Barber and Forrest 1995
6/1992	HPGe	46	0.10	220	(c)	Veinot 2003

a. As defined by Currie (1968) with $\alpha = \beta = 0.05$, paired observations.

Table 5-12. Reported lung counting detection limits for ²³⁸U.

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

a. As defined by Currie (1968) with $\alpha = \beta = 0.05$, paired observations.

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 U and 238 U lung count detection thresholds are assumed to be 130 μ g and 13.5 mg, respectively, through 1990. Beginning in 1991, the critical level or MDA is supplied with the individual 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 ²³⁸U, and workers in enriched areas had results reported as ²³⁵U. Assumptions that are favorable to claimants should be based on conversions of 93% enrichment for ²³⁵U (oralloy VHEU) and natural isotopic abundances for ²³⁸U.

b. Type I error = 5%.

c. Individual MDA or critical level supplied with analytical results.

b. Individual MDA or critical level supplied with analytical results.

5.4.2.2 **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 ²²⁸Ac and/or ²¹²Pb lung activity (Souleyrette 2003). Thorium lung counting was conducted from 1958 to 1984 with routine lung counts, scheduled at approximately 6-month intervals, starting in 1961 (BWXT, 2005).

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 ²²⁸Ac gamma radiation in an *in vivo* count could be an indication of a ²²⁸Ra intake or an intake of the entire ²³²Th chain. The 239-keV gamma line from ²¹²Pb, a progeny of ²²⁸Th, is a more reliable indicator of ²³²Th, because the thorium isotopes would remain together during processing. However, if the ²²⁸Ra is lost, ²¹²Pb decreases with the 1.9-year half-life of its parent, ²²⁸Th, moderated by the ingrowth of new ²²⁸Ra with its 5.7-year half-life. This complicated disequilibrium pattern operates 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 ²³²Th (corresponding to 15 rem/yr to the lung) varied markedly with the ratio of 228 Th to 232 Th. For full equilibrium (ratio = 1), the MPLB was calculated to be 2.9 nCi of ²³²Th. 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 ²²⁸Ra-to-²³²Th 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.

West (1965, p. 18) reported that thorium at Y-12 was processed less than 1 year after purification by the supplier and, as a consequence, had only about 10% as much ²²⁸Ra as ²²⁴Ra. This means the maximum dose conversion factor per milligram of ²³²Th would be less than that for ²³²Th in full equilibrium with its progeny. For calculating intakes of ²²⁸Th and ²³²Th from lung counts, a default assumption must be made for the ratio of equilibrium of ²²⁸Th to ²³²Th. This ratio will be used to determine the intake of ²²⁸Th from the intake of ²³²Th calculated from the lung count results (assumed to be ²³²Th). As can be seen in Figure 5-4, the activity of ²²⁸Th after initial purification declines over the first 5 years due to the decay of the original ²²⁸Th in the mixture, then slowly builds back up due to the ²²⁸Th resulting from the decay of ²³²Th.

Because the actual time from initial purification is unknown and will change over time, an average of 80% equilibrium of ²²⁸Th to ²³²Th should be assumed. This is a reasonable assumption because the actual equilibrium percentage ranges from 100% at initial purification to below 60% and then slowly rises again. The drop to 80% occurs within the first 6 months and continues dropping over the first year, so the assumption of 80% is consistent with the assumption the thorium was processed within a vear of purification. This translates to a ratio of ²²⁸Th to ²³²Th of 0.8.

Neptunium

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

5.4.2.3 **Chest Counting for Other Radionuclides**

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

Figure 5-4. Variation of activity in natural thorium after a single purification (West 1965).

5.5 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in this document, bracketed callouts have been inserted to indicate information, conclusions, and recommendations provided to assist in the process of worker dose reconstruction. These callouts are listed here in the Attributions and Annotations section, with information to identify the source and justification for each associated item. Conventional References, which are provided in the next section of this document, link data, quotations, and other information to documents available for review on the Project's Site Research Database.

- [1] Mahathy, James M. ORAU Team. Health Physicist. December 2004.

 Data extraction and all statistical analyses were performed by Mr. Mahathy and results were validated by Richard Sparks.
- [2] Brackett, Elizabeth. ORAU Team. Principal Internal Dosimetrist. March 2005. Ms. Brackett modeled the Type M and S intakes as described in this section. These fits were reviewed by Dave Allen of NIOSH.
- [3] Arno, Matthew. ORAU Team. Health Physicist. January 2012.

 Dr. Arno modeled the Type F intakes as described in this section. These fits were reviewed by Tom Labone.

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GLOSSARY

absorption type

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

action level

A radiation or activity level that, when reached, results in a prescribed action. For example, high contact external dose rates can result in the use of extremity dosimetry; elevated airborne concentrations can result in the use of respiratory protection. Action levels are typically higher than minimum detectable levels. See minimum detectable level.

activity median aerodynamic diameter (AMAD)

Diameter of a unit density sphere with the same terminal settling velocity in air as that of the aerosol particle whose activity is the median for the entire aerosol.

acute exposure

Radiation exposure to the body delivered in a short period. See *chronic exposure*.

alpha radiation

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

background radiation

Radiation from cosmic sources, naturally occurring radioactive materials including naturally occurring radon, and global fallout from the testing of nuclear explosives. Background radiation does not include radiation from source, byproduct, or Special Nuclear Materials regulated by the U.S. Nuclear Regulatory Commission. The average individual exposure from background radiation is about 360 millirem per year.

bioassay

Measurement of amount or concentration of radionuclide material in the body (in vivo measurement) or in biological material excreted or removed from the body (in vitro measurement) and analyzed for purposes of estimating the quantity of radioactive material in the body. Also called radiobioassay.

bremsstrahlung

Electromagnetic radiation released as a result of inelastic scattering of a moving charged particle within the nucleus of an atom.

burnup

Depletion of fissionable material in nuclear fuel.

calutron

Accelerator that separates isotopes (e.g., ²³⁵U from ²³⁸U) according to their masses using strong magnetic fields. The name derives from California University cyclotron.

chronic exposure

Radiation dose to the body delivered in small amounts over a long period (e.g., days or years). See *acute exposure*.

class

See inhalation class and absorption type.

class Q

Mixture of 10% class Y uranium and 90% modified class W uranium (the modification being an increase of the class W 50-day compartment to 120 days). An example of class Q material is that which occurs as a thin layer of uranium dioxide (UO_2) on the surface of uranium metal as the metal interacts with oxygen in the air. See *inhalation class* and *absorption type*.

class (type) Q system

Dosimetry system that uses an activity median aerodynamic diameter of 8 micrometers as a basis. See *class Q*.

committed effective dose equivalent (CEDE)

Sum of the effective dose equivalents to various tissues or organs in the body each multiplied by the appropriate tissue weighting factor and committed for a 50-year period following an acute intake or the onset of chronic intake. It does not include contributions from external dose. See *dose*.

contamination

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

cyclotron

Particle accelerator capable of large beam currents where the beam is injected in the center of a circular magnet. A fixed radio frequency signal applied to two D-shaped electrodes accelerates the beam as it passes from one electrode to the other as the potential alternates. The radius of the beam increases as the energy increases.

depleted uranium (DU)

Uranium with a percentage of ²³⁵U lower than the 0.7% found in natural uranium. As examples, spent (used) fuel elements, byproduct tails, residues from uranium isotope separation, and some weapons materials contain DU. DU can be blended with highly enriched uranium to make reactor fuel or used as a raw material to produce plutonium. Y-12 gives the isotopic distribution by mass as 0.001% ²³⁴U, 0.199% ²³⁵U, and 99.8% ²³⁸U.

dose

In general, the specific amount of energy from ionizing radiation that is absorbed per unit of mass. Effective and equivalent doses are in units of rem or sievert; other types of dose are in units of roentgens, rads, reps, or grays.

dose conversion factor

Multiplier for conversion of potential dose to the personal dose equivalent to the organ of interest (e.g., liver or colon).

enriched uranium (EU)

Uranium in which processing has increased the proportion of ²³⁵U to above the natural level of 0.7%. Reactor-grade uranium is usually enriched to approximately 3.5% ²³⁵U;

weapons-grade uranium contains greater than 90% ²³⁵U. At Y-12, a common isotopic distribution by mass is 1% in ²³⁴U, 93% ²³⁵U, and 6% ²³⁸U.

enrichment

Isotopic separation process that increases the percentage of a radionuclide in a given amount of material above natural levels. For uranium, enrichment increases the amount of ²³⁵U in relation to ²³⁸U. Along with the enriched uranium, this process results in uranium depleted in ²³⁵U. See *depleted uranium* and *enriched uranium*.

exposure

In general, the act of being exposed to ionizing radiation. 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). See acute exposure and chronic exposure.

gaseous diffusion plant

Facility where uranium hexafluoride (UF₆) gas is filtered to enrich the ²³⁵U and separate it from ²³⁸U. The process requires enormous amounts of electric power and results in an increase in ²³⁵U enrichment from 1% to about 3%.

half-life

Time in which half of a given quantity of a particular radionuclide disintegrates (decays) into another nuclear form. During one half-life, the number of atoms of a particular radionuclide decreases by one half. Each radionuclide has a unique half-life ranging from millionths of a second to billions of years.

highly enriched uranium (HEU)

Uranium enriched to at least 20% ²³⁵U for use as fissile material in nuclear weapons components and some reactor fuels. Also called high-enriched uranium.

inhalation class

Former respiratory tract inhalation classification scheme developed by the International Council on Radiological Protection for inhaled material according to its rate of clearance from the pulmonary region of the lung. Materials were classified as D (days, half-life less than 10 days), W (weeks, 10 to 100 days), or Y (years, more than 100 days). See absorption type, which superseded this concept.

intake

Radioactive material taken into the body by inhalation, absorption through the skin, injection, ingestion, or through wounds.

internal dose

Dose received from radioactive material in the body.

in vitro bioassay

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

in vivo bioassay

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

low-enriched uranium (LEU)

Uranium enriched to less than 20% ²³⁵U by mass.

maximum permissible lung burden (MPLB)

Historical occupational limit for radionuclides in the lung defined as the quantity of the isotope that could be present at any given time to deliver 15 rem per year to the lung at the end of a 50-year period of chronic exposure. At Y-12 during the 1970s the occupational limit for plutonium was a quantity of plutonium that could be present in the chest at any given time equal to 16 microcuries or 0.25 micrograms of ²³⁹Pu.

minimum detectable amount or activity (MDA)

Smallest amount (activity or mass) of an analyte in a sample that can be detected with a probability β of nondetection (Type II error) while accepting a probability α of erroneously deciding that a positive (nonzero) quantity of analyte is present in an appropriate blank sample (Type I error).

monitoring

Periodic or continuous determination of the presence or amount of ionizing radiation or radioactive contamination in air, surface water, groundwater, soil, sediment, equipment surfaces, or personnel (for example, bioassay or alpha scans). In relation to personnel, monitoring includes internal and external dosimetry including interpretation of the measurements.

natural uranium

Uranium as found in nature, approximately 99.27% 238 U, 0.72% 235 U, and 0.0054% 234 U by mass. The specific activity of this mixture is 2.6 × 10 7 becquerel per kilogram (0.7 microcuries per gram). See *uranium*.

nephrotoxicity

Poisoning of the kidney.

occupational dose

Internal and external ionizing radiation dose from exposure during employment. Occupational dose does not include that from background radiation or medical diagnostics, research, or treatment, but does include dose from occupationally required radiographic examinations that were part of medical screening.

quality factor

Principal modifying factor (which depends on the collision stopping power for charged particles) that is employed to derive dose equivalent from absorbed dose. The quality factor multiplied by the absorbed dose yields the dose equivalent. See *dose* and *weighting factor*.

recycled uranium (RU)

Uranium first irradiated in a reactor then recovered through chemical separation and purification. RU contains minor amounts of transuranic material (e.g., plutonium and neptunium) and fission products (e.g., technetium) or uranium products (e.g., ²³⁶U) after purification.

routine monitoring

Monitoring carried out at regular intervals during normal operations.

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sensitivity

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

uranium (U)

Heavy, metallic, and radioactive element with atomic number 92. Most natural uranium as found in ores is ²³⁸U with trace levels of other isotopes. Uranium-235 (0.7% of natural uranium) is fissile by itself and used in nuclear weapons as well as reactors. Uranium-238 (99.3% of natural uranium) is fissionable by fast neutrons and used in nuclear reactors. Natural uranium contains a minute amount of ²³⁴U. See *depleted uranium*, *enriched uranium*, *highly enriched uranium*, *low-enriched uranium*, *recycled uranium*, and *natural uranium*.

very highly enriched uranium (VHEU)

Uranium enriched to 90% or more of ²³⁵U.

weighting factor (w_T)

The ratio of the stochastic risk arising from tissue *T* to the total risk when the whole body is irradiated uniformly.

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A.1 CONVENTIONS USED IN INTERNAL DOSE REPORTS

The following is adapted from Souleyrette (2003).

A.1.1 Y-12 Urinalysis Data through September 1989

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

<u>Badge/Old SSN</u> – 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 = (might have been used for a limited time to indicate experimental analyses methods).

Volume – volume of sample in milliliters.

Time – Time between last void and sample void. Used to normalize urine result to 24-hour 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.)

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Uranium Result – 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.

A.1.2 Y-12 Urinalysis Data since October 1989

<u>Dept</u> - 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 have been 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 Publication 30 (ICRP 1979) classes D, W, and Y. (Class Q indicates 90% class Super-W (120-day lung retention) and 10% class Y, 8 µm 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

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I = ingestion J = injection

<u>Incident Date</u> – Date of documented radiological exposure incident, if any.

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

A.1.3 In Vivo Data

<u>Dept</u> – Department number of employee at the time of monitoring.

Run Date – Date subject counted.

Run No. – Laboratory identifier of counting session.

Weight – Subject weight in pounds.

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

Surface Contamination Code – A six-digit code indicating if 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 3, there was surface contamination found and the remaining five digits give the survey meter reading of the contamination.

<u>Type Analysis</u> – 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:

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

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

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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 (μ g) – ²³⁵U activity detected in chest cavity. If the count is invalid, an "&" appears as the first character.

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

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

Tc-99 (µCi) - ⁹⁹Tc activity detected in chest cavity.

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

A.2 URANIUM SOLUBILITY IN THE LUNG

For a workplace as varied as Y-12, no single solubility type or particle size distribution applies to all workers. Further, accurate assignment of the uranium lung clearance type to a given bioassay result is virtually impossible because of uncertainties about chemical form and limitations of the personnel tracking system (Barber and Forrest 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 favorable to claimant assumptions. Further, several solubility studies were conducted between 1992 and 1999 (Veinot 2003). Based on this study, both depleted and enriched uranium metals and oxides were found to act as Type S material. A more conclusive assessment would require examining activities in each building. A number of buildings seemed to contain Type S material. An important observation in this study was that certain areas in buildings could contain varying solubilities of uranium. One area might contain a very insoluble form, while an adjacent area might contain a more soluble form. Barber and Forrest (1995) showed the necessity to better characterize the type of material found at Y-12. As a result of this study, RWPs were modified for areas where insoluble forms dominated. Areas with mixtures of soluble and insoluble forms of uranium were designated for additional sampling for better characterization. If a worker entered an area that was determined to

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contain Type S material, fecal and urine bioassays were implemented. If the worker entered an area that contained a more soluble form of uranium, urine bioassay only was used to estimate intakes (Veinot 2003). Thus, the presence of fecal sample results in an individual's monitoring records is a strong indicator that the worker was exposed to insoluble uranium compounds.

A.3 IN VITRO DETECTION LIMITS

Tables A-1 and A-2 summarize information developed in Section 5.3. The tabulated values for urinalysis results represent laboratory detection limits $L_{\rm D}$ and do not include uncertainties introduced by sample collection or conversion from submitted volumes to daily void volumes. As noted in Section 5.3, $L_{\rm D}$ values for some historical techniques remain to be identified, and will be reported in later revisions of this TBD as they become available.

Table A-1. Uranium urinalysis detection limits.

Method	Period	Detection limit (mass)	Detection limit (activity)	Convention
Fluorometry	1950–9/1989	7 μg/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 A-2. Other *in vitro* detection limits.

Analyte/solubility type	Method	Period	Detection limit (activity)
Tritium (HTO)	Liquid scintillation	10/1988-present	2,000 dpm/da
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.

A.4 IN VIVO DETECTION LIMITS

From the review of Y-12 documents and conversations with present and former Y-12 staff members, the vast majority of site operations from 1943 to the present involved uranium in a variety of chemical forms and degrees of enrichment. Therefore, the primary internal radiation exposure to Y-12 workers was from uranium. However, the internal dosimetry program has included limited monitoring for ¹³⁷Cs, ⁹⁹Tc, thorium, plutonium, ²²⁸Ac, and tritium, among other radionuclides; there are difficulties in interpreting some of the measurement data that cannot be resolved satisfactorily at this time.

Table A-3 summarizes information developed in Section 5.4. Detection limits for some historical techniques remain to be identified, and will be reported in later revisions of this TBD as they become available.

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.

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

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

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

a. West (1965, p. 26)

For dose reconstruction due to potential thorium intakes, if the monitoring records contain evidence that the worker received *in vivo* chest counts for thorium before 1997, it should be assumed that the worker had the potential for occupational thorium intakes. Beginning in May 1961, Y-12 Plant policy indicated that workers were to be counted at the *in vivo* facility before working with thorium to obtain a baseline or background spectrum. Thereafter, the routine measurements were performed at approximately 6-month intervals as long as the worker was involved with thorium processing. In addition, if an incident occurred or there was reason to believe that an employee received an internal thorium exposure, the affected individual was to be scheduled for an immediate *in vivo* measurement (BWXT, 2005; West 1961; McLendon 1963).

After the initial transuranic hazard assessment performed in the first quarter of 1997 (which included thorium), programmatic steps were implemented to infer when further bioassay sampling was indicated to determine if additional analyses were required to determine CEDE from intakes of the associated transuranic radionuclides. This approach was based on uranium bioassay results. Thus, if the monitoring records indicate increased surveillance for thorium (or other transuranic materials), it is reasonable to conclude that the worker might have had the potential to receive intakes in excess of the rates inferred based on the Table 5-7 ratios. In such cases, missed or measured intakes should be applied based on the available bioassay data.

Oftentimes ²²⁸Ac was reported on *in vivo* chest counts. In such cases after 1997, this should not be inferred to represent a potential for occupational intakes of thorium in excess of the associated transuranic ratio unless there are other indications that the worker was being monitored with greater scrutiny (more frequent *in vivo* counts, urine or fecal sample submissions, etc).

b. Cofield (1961)

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B.1 PURPOSE

There are instances of energy employees who, for a variety of reasons, (1) were not monitored for internal exposure during the course of their employment at a DOE facility or (2) whose records of such monitoring are incomplete or unavailable. In such cases, data from coworkers can be used to approximate an individual's possible exposure. This TBD details the calculation and assignment of intakes based on coworker data from the Y-12 Plant.

B.2 OVERVIEW

ORAUT-OTIB-0019, *Analysis of Coworker Bioassay Data for Internal Dose Assignment* (ORAUT 2005) describes the general process for analysis of bioassay data for assignment of doses to individuals based on coworker results.

Bioassay results were obtained from the Oak Ridge Institute for Science and Education Center for Epidemiologic Research (CER) Dosimetry Database, which contains uranium urinalysis records from the Y-12 Plant for 1950 to 1988. The CER obtained this database from Y-12 to conduct an epidemiology study of site workers. The database results are in units of disintegrations per minute per day, although original urinalysis results were reported in terms of either mass or activity concentrations, depending on the measurement method. Conversion factors from the original results to activity per day are discussed in this TBD. A statistical analysis of these data was performed in accordance with ORAUT-OTIB-0019 (ORAUT 2005). The resultant values were input to the Integrated Modules for Bioassay Analysis (IMBA) computer program, and a fit to the data was performed to obtain intake rates for assigning dose distributions.

B.3 DATA

B.3.1 Selected Bioassay Data

Data were extracted from "Y12 Urinalysis 1950-1988," a version of the CER Dosimetry Database [1]. Sample dates were taken from the Date field and uranium urinalysis results were taken from the DPERM_INT field. The latter field contains uranium bioassay results in units of disintegrations per minute per day. Samples labeled Control were excluded from the analysis, as were those with a code of 9 in the UseFlag field; the documentation provided with the database indicated that the latter meant "do not use data as specified by [Y-12] HP." The reasons for marking particular results are unknown. A review of the database revealed that nearly 20% (85,544 of 479,446) of the results were flagged as not to be used. Of those, 82,892 (97%) had results less than or equal to zero. The excluded results greater than zero were relatively uniformly distributed among all positive results, and accounted for less than 1% (2,652 of 299,967).

B.3.2 Analysis

Because of the large number of sample results, the data were analyzed by month. A lognormal distribution was assumed, and the 50th and 84th percentiles were calculated for each month using the method in ORAUT (2005). Because there were fewer results in 1950 and 1951, these years were analyzed on an annual basis rather than by month. However, the analysis results were not included in the intake modeling because the 50th- and 84th-percentile values were much smaller than those in later years, and may underestimate the urinary uranium concentrations as discussed in Section 5.3.2.1. January through April 1952 were included in the modeling because the 50th- and 84th-

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percentile values were generally larger than subsequent values. Table B-1 lists the statistical analysis results.

Table B-1. Monthly uranium 24-hour urinary excretion rate analyses, 1952 to 1988.

Effective sample date	50th percentile (dpm/d)	84th percentile (dpm/d)	Number of samples	Effective sample date	50th percentile (dpm/d)	84th percentile (dpm/d)	Number of samples
1/15/1952	21.16	64	696	7/16/1970	8.88	38.05	399
2/14/1952	24.92	84.51	741	8/15/1970	8.76	42.72	401
3/15/1952	23.86	88.16	918	9/15/1970	11.59	55.4	236
4/15/1952	25.49	74.15	665	10/15/1970	9.81	34.23	500
5/15/1952	21.15	51.31	1103	11/14/1970	13.1	51.16	460
6/15/1952	18.06	47.31	1401	12/15/1970	17.14	67.22	275
7/15/1952	14.13	41.9	1236	1/14/1971	4.41	18.55	12
8/15/1952	11.25	33.06	1258	2/14/1971	11.3	35.27	523
9/14/1952	13.7	35.97	1590	3/16/1971	12.76	50.18	334
10/14/1952	16.71	46.49	1305	4/16/1971	9.52	39.99	458
11/14/1952	16.68	46.41	1133	5/16/1971	5.87	21.42	557
12/14/1952	14.29	37.68	1368	6/15/1971	10.37	41.16	263
1/14/1953	12.61	37.15	1123	7/16/1971	4.67	19.49	396
2/13/1953	10.51	32.28	1065	8/15/1971	10.74	47.49	557
3/16/1953	9.18	26.4	1380	9/15/1971	13.95	62.06	275
4/15/1953	10.34	32.25	1095	10/15/1971	8.25	32.76	463
5/16/1953	10.43	33.25	1079	11/15/1971	8.24	29.09	504
6/15/1953	9.81	34.9	1337	12/15/1971	11.67	35.91	283
7/15/1953	11.13	40.08	830	1/15/1972	11.54	44.77	289
8/15/1953	9.81	33.3	1261	2/14/1972	12.2	43.92	227
9/14/1953	19.53	63.39	1099	3/15/1972	15.64	56.16	253
10/15/1953	32.43	110.13	677	4/15/1972	12.09	47.06	246
11/14/1953	26.65	88.11	1752	5/15/1972	12.64	50.99	296
12/15/1953	18.47	57.43	1614	6/15/1972	13.68	34.9	155
1/14/1954	19.45	84.37	1655	7/15/1972	18.95	70.38	257
2/13/1954	16.06	74.2	1938	8/15/1972	20.33	67.87	314
3/16/1954	19.5	74.74	2233	9/14/1972	18.14	54.91	234
4/15/1954	22.04	78.72	1688	10/14/1972	15.48	64.45	225
5/16/1954	22.85	77.01	1774	11/14/1972	12	42.03	369
6/15/1954	22.07	73.11	1801	12/14/1972	20.42	61.5	164
7/16/1954	22.06	83.01	1303	1/14/1973	15.63	41.34	347
8/15/1954	15.39	63.49	2571	2/13/1973	15.79	57.05	271
9/15/1954	8.47	28.08	1064	3/16/1973	15.71	49.06	325
10/15/1954	11.58	50.14	1726	4/15/1973	16.2	48.06	268
11/14/1954	22.02	91.14	118	5/16/1973	14.46	47.02	366
12/15/1954	14.85	45.87	783	6/15/1973	15.04	56.06	248
1/14/1955	7.36	25.15	591	7/15/1973	14.38	43.07	326
2/14/1955	5.62	19.81	639	8/15/1973	16.5	52.5	296

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	50th	84th			50th	84th	
Effective	percentile	percentile	Number of	Effective	percentile	percentile	Number of
sample date	(dpm/d)	(dpm/d)	samples	sample date	(dpm/d)	(dpm/d)	samples
3/16/1955	13.71	39.83	909	9/14/1973	12.97	56.63	509
4/16/1955	16.2	50.07	1840	10/15/1973	12.31	40.96	485
5/16/1955	13.27	37.94	2006	11/14/1973	10.62	29.86	486
6/15/1955	15.71	47.63	1595	12/15/1973	14.2	46.52	393
7/16/1955	13.1	36.57	1573	1/14/1974	12.16	38.96	418
8/15/1955	13.85	42.78	1874	2/13/1974	11.16	34.34	378
9/15/1955	17.1	55.19	1592	3/16/1974	20.33	56.38	287
10/15/1955	16.87	51.49	1990	4/15/1974	12.99	40.27	325
11/15/1955	12.99	39.56	1725	5/16/1974	9.14	22.89	405
12/15/1955	15.34	45.01	1732	6/15/1974	9.75	29.71	222
1/15/1956	14.5	47.71	2121	7/16/1974	7.54	20.57	270
2/14/1956	16.03	48.01	1853	8/15/1974	8.44	24.96	330
3/15/1956	14.02	42.59	1902	9/15/1974	8.77	24.28	264
4/15/1956	16.45	46.84	1886	10/15/1974	14.19	137.02	298
5/15/1956	15.73	47.57	1980	11/14/1974	9.16	53.77	368
6/15/1956	13.79	40.67	1986	12/15/1974	8.11	35.07	250
7/15/1956	13.82	41.39	2068	1/14/1975	8.93	37.19	252
8/15/1956	13.39	41.44	843	2/14/1975	11.49	44.05	360
9/14/1956	14.11	38.28	1637	3/16/1975	16.92	57.92	291
10/14/1956	11.1	32.33	2022	4/16/1975	10.37	46.11	319
11/14/1956	4.72	15.14	1835	5/16/1975	11.74	39.05	297
12/14/1956	7.3	26.34	1889	6/15/1975	10.66	33.4	186
1/14/1957	9.42	28.04	2413	7/16/1975	11.04	38.53	188
2/13/1957	10.2	38.81	1967	8/15/1975	11.07	32.85	338
3/16/1957	11.04	40.78	1951	9/15/1975	8.9	31.58	171
4/15/1957	10.79	33.8	2302	10/15/1975	11.92	36.58	186
5/16/1957	10.48	42.11	1975	11/15/1975	9.18	32.82	298
6/15/1957	9.44	34.06	2099	12/15/1975	12.88	40.99	148
7/15/1957	8.46	28.63	2471	1/15/1976	9.97	30.51	196
8/15/1957	10.56	35.75	2038	2/14/1976	10.81	32.89	321
9/14/1957	7.51	24.91	2552	3/15/1976	9.19	28.35	225
10/15/1957	12.06	53.49	2278	4/15/1976	8.65	27.03	211
11/14/1957	13.03	44.83	2387	5/15/1976	8.3	24.08	347
12/15/1957	11.29	37.96	2878	6/15/1976	8.24	23.91	185
1/14/1958	8.36	30.87	2708	7/15/1976	3.82	12.47	194
2/13/1958	13.64	53.23	2208	8/15/1976	6.51	18.64	332
3/16/1958	16.24	59.24	2219	9/14/1976	5.39	14.99	193
4/15/1958	13.3	52.29	2498	10/14/1976	9.5	26.06	228
5/16/1958	12.71	56.35	2396	11/14/1976	13.24	40.32	297
6/15/1958	13.7	77.06	2665	12/14/1976	13.72	43.42	202

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	50th	84th			50th	84th	
Effective sample date	percentile (dpm/d)	percentile (dpm/d)	Number of samples	Effective sample date	percentile (dpm/d)	percentile (dpm/d)	Number of samples
7/16/1958	17.44	79.66	2332	1/14/1977	13.97	44.66	169
8/15/1958	19.45	65.95	2206	2/13/1977	13.41	41.7	268
9/15/1958	19.19	86.52	2669	3/16/1977	12.91	40.62	245
10/15/1958	15.21	58.52	2115	4/15/1977	8.86	24.33	191
11/14/1958	21.01	73.93	2258	5/16/1977	8.39	22.81	302
12/15/1958	22.76	80.61	3071	6/15/1977	9.62	31.33	180
1/14/1959	20.74	77.72	2949	7/15/1977	7.46	28.02	190
2/14/1959	24.9	90.03	2798	8/15/1977	9.29	25.59	342
3/16/1959	18.71	60.78	2424	9/14/1977	11.22	54.94	207
4/16/1959	19.72	66.84	3126	10/15/1977	10.86	37.43	244
5/16/1959	17.44	56.57	2725	11/14/1977	10.75	47.78	341
6/15/1959	12.41	45.55	2878	12/15/1977	9.31	35.7	151
7/16/1959	16.69	57.44	2520	1/14/1978	9.28	35.13	184
8/15/1959	12.79	40.15	3001	2/13/1978	9.82	29.72	293
9/15/1959	13.66	43.44	2580	3/16/1978	4.34	13.13	223
10/15/1959	11.53	40.87	2983	4/15/1978	8.07	26.41	196
11/15/1959	14.86	53.07	3317	5/16/1978	5.53	18.15	291
12/15/1959	16.04	56.6	2785	6/15/1978	4.73	14.79	164
1/15/1960	13.87	43.69	3395	7/16/1978	6.58	20.43	185
2/14/1960	13.36	46	3666	8/15/1978	5.1	24.98	362
3/15/1960	9.31	34.17	2662	9/15/1978	7.02	22.71	198
4/15/1960	9.75	39.57	3375	10/15/1978	7.51	22.85	205
5/15/1960	11.19	38.09	3796	11/14/1978	6.81	24.14	390
6/15/1960	9.88	37.26	2468	12/15/1978	4.93	15.71	155
7/15/1960	10.51	37.64	2330	1/14/1979	5.48	22.99	180
8/15/1960	12.76	51.09	3115	2/14/1979	5.73	24.87	398
9/14/1960	9.45	32.2	2392	3/16/1979	7.5	55.07	219
10/14/1960	8.93	32.44	3155	4/16/1979	4.12	20.11	213
11/14/1960	9.85	35.5	2486	5/16/1979	5.45	26.64	357
12/14/1960	7.6	27.11	2443	6/15/1979	6.54	24.51	198
1/14/1961	7.53	32.63	2571	7/16/1979	6.15	26.52	205
2/13/1961	6.99	27.55	2409	8/15/1979	2.81	8.47	407
3/16/1961	9.36	37.96	2397	9/15/1979	7.52	23.8	180
4/15/1961	8.94	32.38	754	10/15/1979	6.69	21.68	205
5/16/1961	7.46	26.83	1556	11/15/1979	5.68	16.34	360
6/15/1961	9.17	32.46	532	12/15/1979	10.15	28.45	219
7/15/1961	11.04	38.61	1035	1/15/1980	5.43	19.51	238
8/15/1961	5.8	23.46	1137	2/14/1980	5.39	18.48	462
9/14/1961	12.96	54.55	621	3/15/1980	9.96	31.5	206
10/15/1961	8.55	29.21	1216	4/15/1980	7.03	28.92	273

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Effective sample date	50th percentile (dpm/d)	84th percentile (dpm/d)	Number of samples	Effective sample date	50th percentile (dpm/d)	84th percentile (dpm/d)	Number of samples
11/14/1961	7.46	28.68	1222	5/15/1980	6.08	23.84	473
12/15/1961	16.66	64.1	555	6/15/1980	7.52	24.86	215
1/14/1962	11.93	48.51	1303	7/15/1980	4.43	13.44	231
2/13/1962	14.1	55.59	1191	8/15/1980	4.33	14.91	488
3/16/1962	22.09	81.26	830	9/14/1980	5.59	16.59	237
4/15/1962	16.35	63.01	1431	10/14/1980	8.8	27.7	253
5/16/1962	10.69	39.93	1462	11/14/1980	3.7	14.66	468
6/15/1962	19.41	71.41	1045	12/14/1980	6.65	26.31	255
7/16/1962	13.57	52.64	1528	1/14/1981	6.78	27.85	305
8/15/1962	12.66	58.77	1420	2/13/1981	5.54	25.15	462
9/15/1962	19.24	72.07	1049	3/16/1981	6.2	20.98	325
10/15/1962	15.98	53.81	1524	4/15/1981	4.81	19.11	269
11/14/1962	16.91	65.36	1470	5/16/1981	3.66	15.89	535
12/15/1962	22.87	97.5	1134	6/15/1981	5.97	21.83	251
1/14/1963	16.8	64.65	1222	7/15/1981	6.47	31.68	26
2/14/1963	14.35	65.28	1307	8/15/1981	4.89	16.79	575
3/16/1963	17.58	70.17	1203	9/14/1981	6.13	22.23	315
4/16/1963	13.52	46.46	1481	10/15/1981	5.14	21.72	314
5/16/1963	12.56	44	1661	11/14/1981	4.35	18.28	605
6/15/1963	14.88	56	1038	12/15/1981	5.73	29.53	294
7/16/1963	13.4	36.41	1321	1/14/1982	4.53	18.34	351
8/15/1963	13.29	43.99	902	2/13/1982	4.13	15.44	540
9/15/1963	13.48	44.18	1247	3/16/1982	5.01	18.71	482
10/15/1963	11.65	38.88	576	4/15/1982	5.72	32.11	323
11/15/1963	9.5	26.83	1002	5/16/1982	6.89	24.15	584
12/15/1963	7.73	26.04	1562	6/15/1982	5.47	20.5	366
1/15/1964	8	28.09	1390	7/16/1982	4.66	17.11	320
2/14/1964	7.96	24.14	1579	8/15/1982	6.48	31.75	581
3/15/1964	10.27	32.95	1172	9/15/1982	2.92	10.86	351
4/15/1964	4.6	15.35	902	10/15/1982	6.66	25.77	332
5/15/1964	3.58	10.53	1203	11/14/1982	3.24	12.97	637
6/15/1964	5.7	21.01	599	12/15/1982	2.94	13.34	308
7/15/1964	5.66	18.62	903	1/14/1983	3.67	15.42	348
8/15/1964	4.65	14.2	1088	2/14/1983	5.93	23.87	634
9/14/1964	4.62	12.61	477	3/16/1983	5.51	26.4	316
10/14/1964	3.43	9.95	667	4/16/1983	4.51	17.99	296
11/14/1964	3.26	10.78	1112	5/16/1983	4.83	19.89	633
12/14/1964	7.72	21.81	291	6/15/1983	2.99	12.46	288
1/14/1965	4.7	14.91	545	7/16/1983	2.74	9.92	271
2/13/1965	3.99	12.84	646	8/15/1983	5.17	19.39	690

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	50th	84th			50th	84th	
Effective	percentile	percentile	Number of	Effective	percentile	percentile	Number of
sample date	(dpm/d)	(dpm/d)	samples	sample date	(dpm/d)	(dpm/d)	samples
3/16/1965	6.81	23.26	404	10/15/1983	3.36	16.08	279
4/15/1965	4.59	21.09	312	11/15/1983	2.69	14.1	706
5/16/1965	6.7	23.6	571	12/15/1983	2.08	8.98	325
6/15/1965	9.45	31.48	329	1/15/1984	2.72	11	326
7/15/1965	5.64	18.05	364	2/14/1984	2.59	14.01	653
8/15/1965	6.41	23.41	609	3/15/1984	4.2	20.09	329
9/14/1965	2.74	11.57	376	4/15/1984	4.14	21.85	360
10/15/1965	4.91	18.07	416	5/15/1984	3.51	15.94	637
11/14/1965	3.44	12.05	571	6/15/1984	3.42	15.07	335
12/15/1965	9.85	37.35	397	7/15/1984	3.49	15.65	352
1/14/1966	4.44	19.14	469	8/15/1984	2.96	17.55	697
2/13/1966	4.41	24.7	668	9/14/1984	3.97	20.81	356
3/16/1966	10.17	42.75	391	10/14/1984	6.23	26.17	367
4/15/1966	10.75	48.01	441	11/14/1984	2.68	8.13	644
5/16/1966	5.04	16.99	739	12/14/1984	4.62	20.08	348
6/15/1966	6.32	26.44	415	1/14/1985	2.93	12	397
7/16/1966	8.7	39.61	359	2/13/1985	1.93	8.23	605
8/15/1966	5.01	18.31	624	3/16/1985	4.45	31.7	425
9/15/1966	8.86	40.76	431	4/15/1985	1.67	8.6	391
10/15/1966	15.88	71.36	392	5/16/1985	1.66	8.97	625
11/14/1966	8.68	32.4	539	6/14/1985	0.76	5.03	318
12/15/1966	8.98	40.16	567	7/15/1985	1.91	8.85	383
1/14/1967	6.79	33.34	339	8/14/1985	1.97	7.66	569
2/14/1967	5.8	25.69	710	9/14/1985	2.76	11.77	369
3/16/1967	8.35	49.25	426	10/14/1985	1.82	6.05	419
4/16/1967	10.81	56.82	364	11/13/1985	1.53	6.04	544
5/16/1967	3.25	18.88	593	12/14/1985	2.71	11.36	379
6/15/1967	6.51	25.46	224	1/13/1986	2.79	11.56	472
7/16/1967	5.49	35.82	390	2/13/1986	1.1	5.61	593
8/15/1967	7.3	40.93	585	3/14/1986	2.41	13.98	388
9/15/1967	4.42	19.34	401	4/15/1986	1.35	5.24	497
10/15/1967	5.21	20.92	412	5/15/1986	1.12	5.13	555
11/15/1967	5.01	20.71	514	6/15/1986	1.63	7.52	401
12/15/1967	6.06	33.53	405	7/15/1986	2.42	11.67	408
1/15/1968	6.5	28.54	393	8/14/1986	2.54	10.13	572
2/14/1968	10.15	41.09	573	9/14/1986	1.79	8.35	352
3/15/1968	8.1	39.56	476	10/14/1986	0.95	4.22	550
4/15/1968	7.07	43.78	411	11/14/1986	2.6	11.31	626
5/15/1968	11.07	53.86	458	12/14/1986	1.17	6.57	323
6/15/1968	7.05	29.75	574	1/14/1987	0.8	8.03	437

Effective Date: 03/12/2012

INTERNAL DOSIMETRY COWORKER DATA FOR Y-12
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ATTACHMENT B

Effective	50th percentile	84th percentile	Number of	Effective	50th percentile	84th percentile	Number of
7/15/1968	(dpm/d) 5.2	(dpm/d) 23.25	samples 395	sample date 2/13/1987	(dpm/d) 9.62	(dpm/d) 57.02	samples 555
8/15/1968	5.86		533	3/15/1987	5.04	18.9	345
9/14/1968		23.48			2.5		404
	5.48	26.46	421	4/15/1987		13.62	
10/14/1968	3.86	16.16	391	5/15/1987	2.43	11.68	525
11/14/1968	5.9	23.37	516	6/15/1987	2.2	19.91	328
12/14/1968	9.71	32.61	533	7/15/1987	8.16	46.96	34
1/14/1969	8.43	41.12	220	8/15/1987	13.59	70.44	24
2/13/1969	5.02	20.69	731	9/14/1987	4.01	19.3	54
3/16/1969	5.65	27.79	655	10/15/1987	0.44	2.13	474
4/15/1969	7.61	36.08	477	11/14/1987	1.78	14.61	494
5/16/1969	5.22	24.92	593	12/14/1987	2.13	11.13	327
6/15/1969	8.26	30.14	546	1/14/1988	2.19	14.84	338
7/15/1969	7.37	29.64	375	2/13/1988	2.37	10.64	550
8/15/1969	6.62	28.37	630	3/15/1988	4.62	24.43	371
9/14/1969	4.7	17.88	475	4/14/1988	4.03	26.36	345
10/15/1969	6.6	25.36	514	5/15/1988	1.52	7.09	606
11/14/1969	7.06	22.98	541	6/14/1988	2.05	13.32	363
12/15/1969	7.71	30.81	401	7/14/1988	4.03	19.34	416
1/14/1970	9.84	40.81	336	8/14/1988	2.96	17.83	563
2/13/1970	8.33	28.27	559	9/14/1988	2.38	14.04	432
3/16/1970	7.85	29.62	316	10/14/1988	0.75	3.7	434
4/15/1970	6.99	27.21	378	11/13/1988	2.18	10.51	103
5/16/1970	4.93	18.99	103	12/14/1988	3.69	13.94	152
6/15/1970	11.17	44.73	137				

B.4 INTAKE MODELING

B.4.1 <u>Assumptions</u>

All results were assumed to be representative of a full day (24 hours) of urinary excretion. Each result in the intake calculation was assumed to have a normal distribution, and a uniform absolute error of 1 was applied to all results to weight all results equally. Because of the nature of work at Y-12, a chronic exposure pattern best approximates the true exposure conditions for most workers with a potential for intakes. Intakes were assumed to be by inhalation using a default breathing rate of 1.2 m³/hr and a 5-µm AMAD particle size distribution.

The database reported all results as "uranium." Because a variety of enrichments have been present at the Y-12 site, ²³⁴U was assumed for the IMBA intake modeling. This does not affect the fitting of the data for intake determination (i.e., the same total intakes would be obtained for any assumed enrichment) because all uranium isotopes behave the same biokinetically and the isotopes that were considered in this analysis have long half-lives in relation to the assumed intake period. ICRP Publication 68 provides dose coefficients (also referred to as dose conversion factors) for ²³⁴U that are

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7% to 31% larger than those for ²³⁵U, ²³⁶U, and ²³⁸U (ICRP 1995b). Because of the isotopic compositions of the source terms, use of the ²³⁴U dose conversion factor overestimates doses.

Although there are no bioassay results from before 1950 in the database, the first intake period was assumed to begin on January 1, 1947. Before 1947, the calutron was in operation. There are no bioassay measurements for the period, and conditions were likely quite different than in later years, so this period was not included in the modeling. The ORAUT-TKBS-0014-2, *Y-12 National Security Complex – Site Description* (ORAUT 2007) states, "Y-12 was shut down in December 1946 and employment was cut drastically," in reference to the calutron and associated uranium isotope separation programs. Primary operations from 1947 to 1951 consisted of salvage, recovery, and recycling operations with uranium preparation and machining beginning in 1949. It was assumed, therefore, that exposure conditions beginning in 1947 would have been similar to those in the early 1950s.

B.4.2 Bioassay Data Fitting

The IMBA computer program was used to fit the bioassay results to a series of inhalation intakes [2, 3]. IMBA allows the input of 200 urine sample results, which is insufficient to include all Y-12 monthly results. However, this can be expanded to 400 results by using the user-defined bioassay function and applying all the urine parameters to this function. Data from January 1952 through December 1988 were fit as a series of chronic intakes.

The initial intake assumptions were based on periods that coincided with major operations on the site. Operations from 1947 to 1951 were very specific and, therefore, were modeled as one intake period. However, the bioassay data had some distinct patterns, so the intake dates were adjusted to obtain a better approximation of the data. There appeared to be low-level chronic intakes of uranium throughout long periods, with briefer, larger intakes superimposed on them. To model this pattern, three long-term chronic exposures were assumed to cover 1947 through 1988. For the type M and S fits, five shorter chronic exposures were modeled on top of the early period to account for the intermittent rises in the urine results. Type F uranium was fit as a series of chronic exposures corresponding to the patterns in the urine results.

Because the uranium isotopes at Y-12 have very long half-lives and the material is retained in the body for long periods for solubility types M and S, excretion results are not independent. For example, an intake in the early 1950s could contribute to urinary excretion in the 1980s and later. To avoid potential underestimation of intakes for people who worked at Y-12 for relatively short periods, each intake was fit independently using only the bioassay results from the single intake period. This is likely to result in overestimation of intakes, particularly for assumed type S exposures that extend through multiple assumed intake periods. Type F intake periods were fit simultaneously, subject to the limitations of IMBA, which required that the intake modeling be split into two time periods.

B.5 URANIUM INTAKE RATES BY MATERIAL TYPE

All the bioassay results were entered into IMBA and are displayed in the figures at the end of this attachment. However, as described in Section B.4.2, each period was fit separately for solubility types M and S and into two periods for type F, so only the results during the intake period were selected for use in fitting each period. Excluded results are shown in red in the figures.

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B.5.1 Type F

Uranium urine results were fit assuming type F material. Figures B-1 and B-2 show the individual fits to the 50th-percentile values. The same intake periods were applied to the 84th-percentile values because the values followed a similar pattern; results of the individual fits are not shown here but they fit reasonably well. Table B-2 summarizes the intake periods and corresponding intake rates for the 50th and 84th percentiles with their geometric standard deviations (GSDs).

The GSDs were determined by dividing the 84th-percentile intake rates by the 50th-percentile intake rates.

Table B-2. Type F uranium intake periods and rates.

		Uranium intak		
Start	Stop	50th percentile	84th percentile	GSD
1/1/1947	5/31/1952	85.35	264.1	3.09
6/1/1952	8/31/1953	43.89	131.3	2.99
9/1/1953	10/31/1953	104.3	339.5	3.26
11/1/1953	12/31/1954	66.43	254.1	3.83
1/1/1955	12/31/1956	47.03	145.2	3.09
1/1/1957	12/31/1957	36.07	145.6	4.04
1/1/1958	12/31/1959	59.48	207.8	3.49
1/1/1960	12/31/1961	34.68	134	3.86
1/1/1962	12/31/1963	53.27	171.9	3.23
1/1/1964	12/31/1969	22.97	97.28	4.24
1/1/1970	12/31/1971	35.8	115.6	3.23
1/1/1972	12/31/1975	47.51	168.4	3.54
1/1/1976	12/31/1977	35.09	121.7	3.47
1/1/1978	12/31/1983	19.68	85.61	4.35
1/1/1984	12/31/1988	9.098	51.26	5.63
7/1/1987	8/31/1987	43.498	102.61	2.36

B.5.2 <u>Type M</u>

Uranium urine results were fit assuming type M material. Figures B-3 to B-10 show the individual fits to the 50th-percentile values. The same intake periods were applied to the 84th-percentile values because the values followed a similar pattern; results of the individual fits are not shown here but they fit reasonably well. Table B-3 summarizes the intake periods and corresponding intake rates for the 50th and 84th percentiles with their geometric standard deviations (GSDs).

Table B-3. Type M uranium intake periods and rates.

		Uranium intak		
Start	Stop	50th percentile	84th percentile	GSD
1/1/1947	2/28/1978	169.34	598.93	3.54
1/1/1947	4/30/1952	354.59	1,154.9	3.26
8/1/1953	11/30/1953	547.22	1,963.1	3.59
11/1/1956	2/28/1959	226.90	825.31	3.64
6/1/1961	12/31/1962	248.68	866.81	3.49
10/1/1968	12/31/1972	160.67	601.60	3.74
3/1/1978	9/30/1984	80.03	355.93	4.45
10/1/1984	12/31/1988	44.203	223.85	5.06

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Figure B-11 shows a representation of the predicted 50th-percentile values from all intakes, and Figure B-12 shows the 84th-percentile predictions. These depict the expected excretion rates from an individual who was exposed for all the periods at the 50th- and 84th- percentile intake rates, respectively, in Table B-3. The GSDs were determined by dividing the 84th-percentile intake rates by the 50th-percentile intake rates.

B.5.3 Type S

The intake periods for the type M fits were applied to the type S material fits. Figures B-13 to B-20 show the individual fits for the 50th-percentile values. Table B-4 summarizes the intake rates for the 50th- and 84th-percentile values. The GSDs were determined as noted for the type M intake rates.

Table B-4. Type S uranium intake periods and rates.

		Uranium intak		
Start	Stop	50th percentile	84th percentile	GSD
1/1/1947	2/28/1978	1,844.4	6,544.2	3.55
1/1/1947	4/30/1952	5,210.4	16,970	3.26
8/1/1953	11/30/1953	17,983	64,559	3.59
11/1/1956	2/28/1959	5,694.6	20,560	3.61
6/1/1961	12/31/1962	6,849.4	23,632	3.45
10/1/1968	12/31/1972	3,290.3	12,415	3.77
3/1/1978	9/30/1984	1,280.3	5,802.9	4.53
10/1/1984	12/31/1988	884.85	4,340.6	4.91

Figures B-21 and B-22 show the predicted 50th- and 84th-percentile excretion rates, respectively, from all type S intakes.

B.6 ASSIGNMENT OF INTAKES AND DOSES

B.6.1 Intake Rate Summary

Several intake periods overlapped, so they were combined to make distinct periods with one intake rate and associated GSD for each. For 1947 through 1977, similar magnitude GSDs were assigned the largest GSD of the similar group for simplicity. Tables B-5 through B-7 summarize the intake periods. These are equivalent to the intake periods in Table B-2 through B-4 but provide a chronological layout of the changing intake rates over time, simplify the GSDs, and provide the 95th percentile intake rates.

B.6.2 Contribution from Contaminants in Recycled Uranium

Throughout the DOE complex, spent fuel from fission reactors has been processed to recover uranium for recycling. Because the uranium streams at Y-12 could have contained recycled uranium, the dose from the added constituents, including plutonium, ²³⁷Np, and ⁹⁹Tc, must be included. See Section 5.2.4 for information about intake values in relation to the uranium intake amounts.

B.6.3 Dose Assignment

For most cases, individual doses are calculated from the 50th-percentile intake rates. Dose reconstructors should select the material type that is the most favorable to claimants. Doses based

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on 50th-percentile intake rates are assigned as lognormal distribution with the applicable GSD. There are situations when the 95th percentile of the coworker distribution and a constant distribution are more appropriate than the 50th percentile and lognormal GSDs. For cases where the 50th-percentile intake rates are not appropriate, dose reconstructors should use the 95th-percentile intake rates. The 95th-percentile intakes should be assigned as a constant rather than lognormal distribution.

Table B-5. Type F uranium intake periods and rates, dpm/day.

data 2 a. Typo i didinam inta		50th		95th
Start	Stop	percentile	GSD	percentile
1/1/1947	5/31/1952	85.4	3.54	683
6/1/1952	8/31/1953	43.9	3.54	351
9/1/1953	10/31/1953	104	3.54	834
11/1/1953	12/31/1954	66.4	4.24	715
1/1/1955	12/31/1956	47.0	3.54	376
1/1/1957	12/31/1957	36.1	4.24	388
1/1/1958	12/31/1959	59.5	3.54	476
1/1/1960	12/31/1961	34.7	4.24	373
1/1/1962	12/31/1963	53.3	3.54	426
1/1/1964	12/31/1969	23.0	4.24	247
1/1/1970	12/31/1971	35.8	3.54	286
1/1/1972	12/31/1975	47.5	3.54	380
1/1/1976	12/31/1977	35.1	3.54	281
1/1/1978	12/31/1983	19.7	4.35	221
1/1/1984	6/30/1987	9.10	5.63	156
7/1/1987	8/31/1987	43.5	3.00	265
9/1/1987	12/31/1988	9.10	3.54	156

Table B-6. Type M uranium intake periods and rates, dpm/day.

		50th		95th
Start	Stop	percentile	GSD	percentile
1/1/1947	4/30/1952	524	3.74	4,588
5/1/1952	7/31/1953	169	3.74	1,483
8/1/1953	11/30/1953	717	3.74	6,275
12/1/1953	10/31/1956	169	3.74	1,483
11/1/1956	2/28/1959	396	3.74	3,470
3/1/1959	5/31/1961	169	3.74	1,483
6/1/1961	12/31/1962	418	3.74	3,661
1/1/1963	9/30/1968	169	3.74	1,483
10/1/1968	12/31/1972	330	3.74	2,890
1/1/1973	2/28/1978	169	3.74	1,483
3/1/1978	9/30/1984	80.0	4.45	933
10/1/1984	12/31/1988	44.2	5.06	636

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Table B-7.	IVNA	: iiranii im	INTAKA	nerione	วทด	rates	anm/aav
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		50th	,	95th
Start	Stop	percentile	GSD	percentile
1/1/1947	4/30/1952	7,054	3.77	62,595
5/1/1952	7/31/1953	1,844	3.77	16,366
8/1/1953	11/30/1953	19,827	3.77	175,929
12/1/1953	10/31/1956	1,844	3.77	16,366
11/1/1956	2/28/1959	7,539	3.77	66,895
3/1/1959	5/31/1961	1,844	3.77	16,366
6/1/1961	12/31/1962	8,693	3.77	77,138
1/1/1963	9/30/1968	1,844	3.77	16,366
10/1/1968	12/31/1972	5,134	3.77	45,559
1/1/1973	2/28/1978	1,844	3.77	16,366
3/1/1978	9/30/1984	1,280	4.53	15,367
10/1/1984	12/31/1988	885	4.91	12,125

Recycled uranium contaminants, when appropriate for the period, also factor into this comparison.

The lognormal distribution is selected in the Interactive RadioEpidemiological Program (IREP), and the calculated dose is entered as Parameter 1 and the associated GSD as Parameter 2. The GSD is associated with the intake, so it is applied to all annual doses from the intake period.

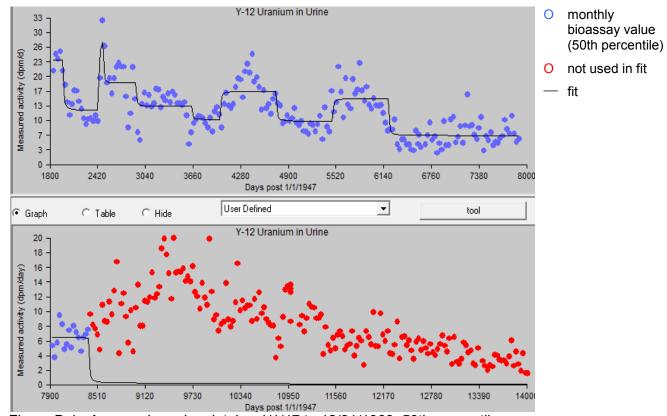


Figure B-1. Assumed uranium intake, 1/1/47 to 12/31/1969, 50th-percentile results, type F.

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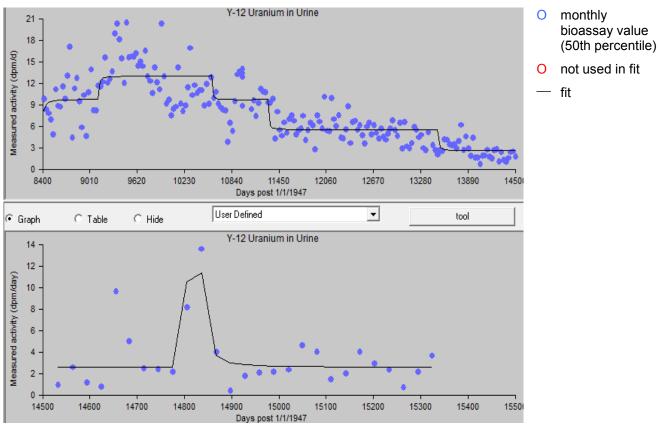


Figure B-2. Assumed uranium intake, 1/1/70 to 12/31/1988, 50th-percentile results, type F.

monthly

fit

bioassay value (50th percentile)

not used in fit

ATTACHMENT B INTERNAL DOSIMETRY COWORKER DATA FOR Y-12

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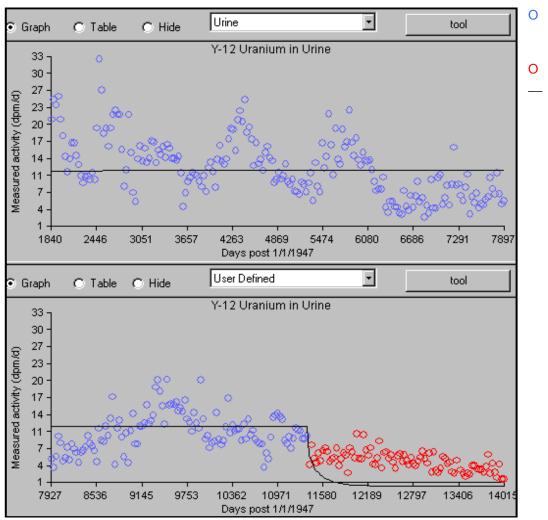


Figure B-3. Assumed uranium intake, 1/1/47 to 2/28/78, 50th-percentile results, type M.

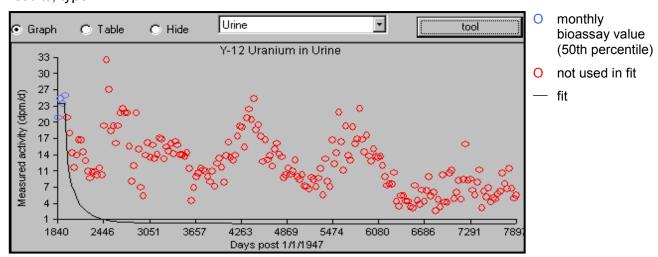


Figure B-4. Assumed uranium intake, 1/1/47 to 4/30/52, 50th-percentile results, type M.

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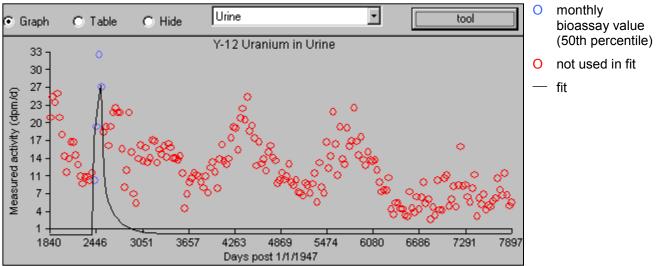


Figure B-5. Assumed uranium intake, 8/1/53 to 11/30/53, 50th-percentile results, type M.

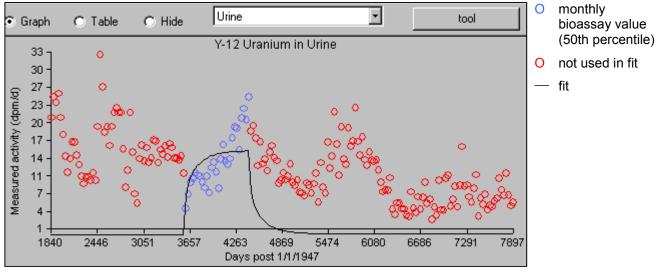


Figure B-6. Assumed uranium intake, 11/1/56 to 2/28/59, 50th-percentile results, type M.

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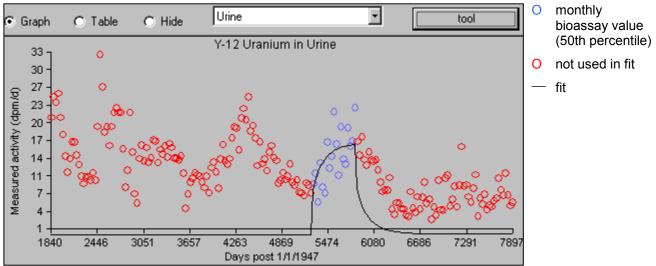


Figure B-7. Assumed uranium intake, 6/1/61 to 12/31/62, 50th-percentile results, type M.

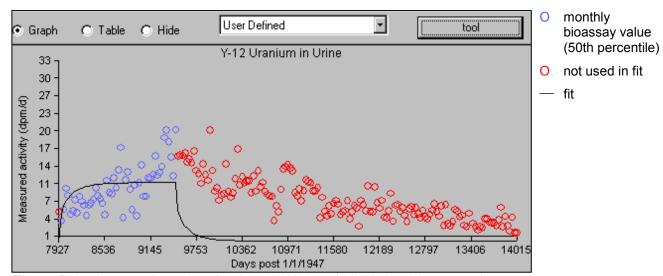


Figure B-8. Assumed uranium intake, 10/1/68 to 12/31/72, 50th-percentile results, type M.

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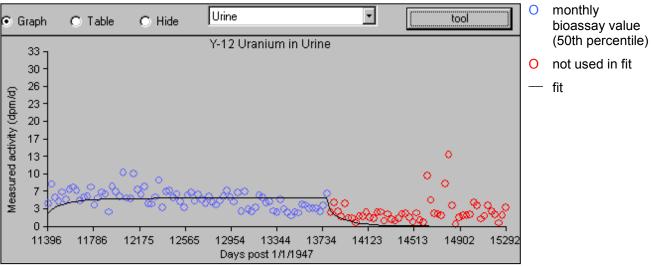


Figure B-9. Assumed uranium intake, 3/1/78 to 9/30/84, 50th-percentile results, type M.

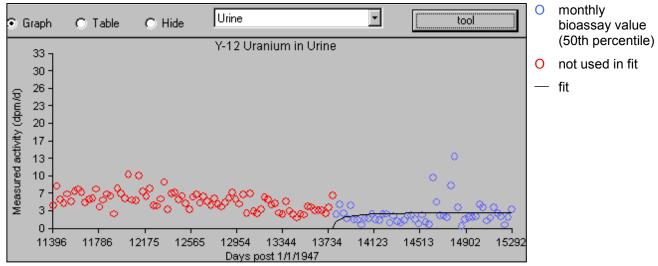


Figure B-10. Assumed uranium intake, 10/1/84 to 12/31/88, 50th-percentile results, type M.

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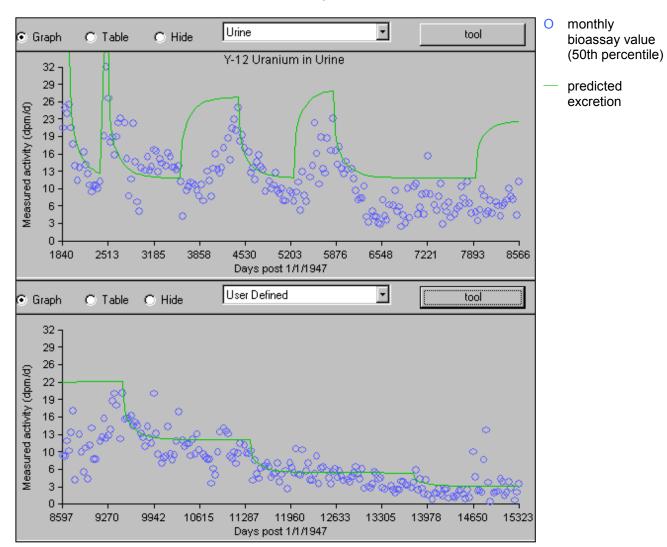


Figure B-11. Predicted (type M, 5 µm) uranium urinary excretion from eight independent intakes, 50th percentile, 1952 to 1988.

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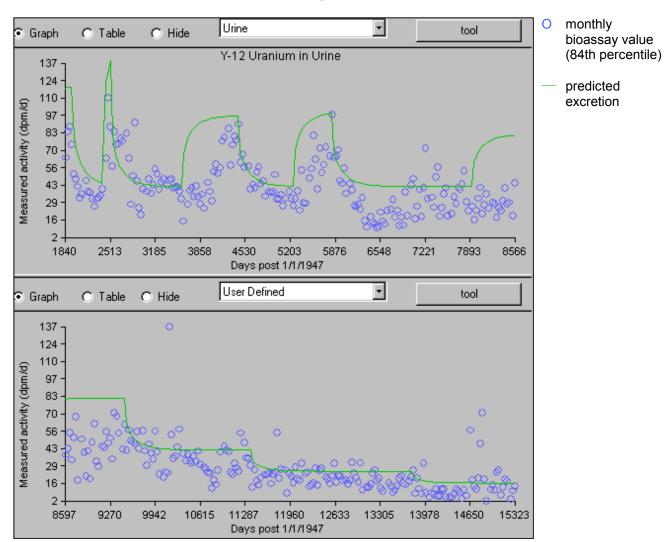


Figure B-12. Predicted (type M, 5 μ m) uranium urinary excretion from eight independent intakes, 84th percentile, 1952 to 1988.

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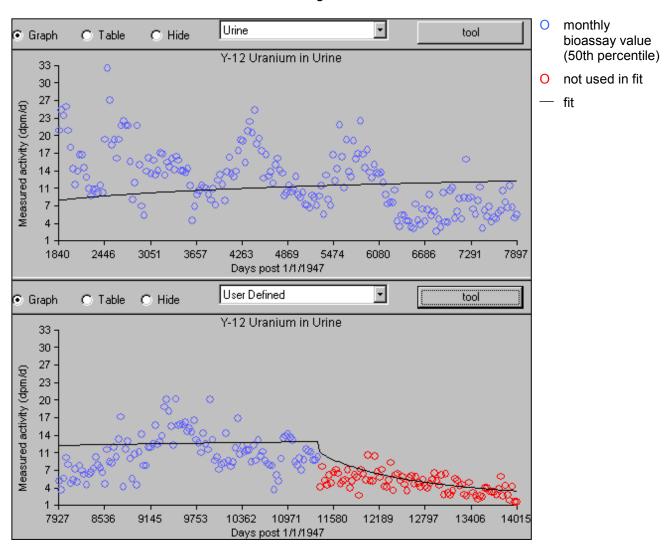


Figure B-13. Assumed uranium intake, 1/1/47 to 2/28/78, 50th-percentile results, type S.

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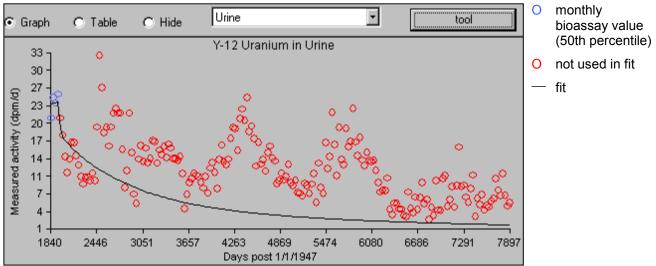


Figure B-14. Assumed uranium intake, 1/1/47 to 4/30/52, 50th-percentile results, type S.

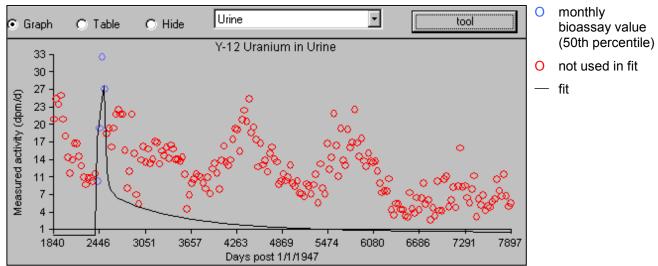


Figure B-15. Assumed uranium intake, 8/1/53 to 11/30/53, 50th-percentile results, type S.

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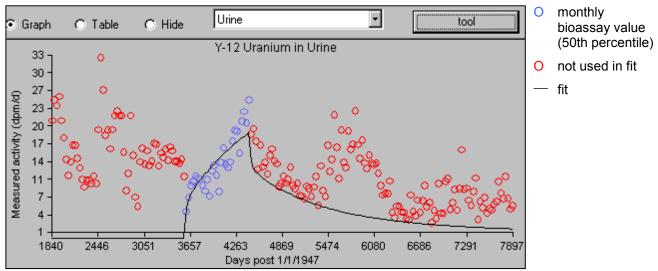


Figure B-16. Assumed uranium intake, 11/1/56 to 2/28/59, 50th-percentile results, type S.

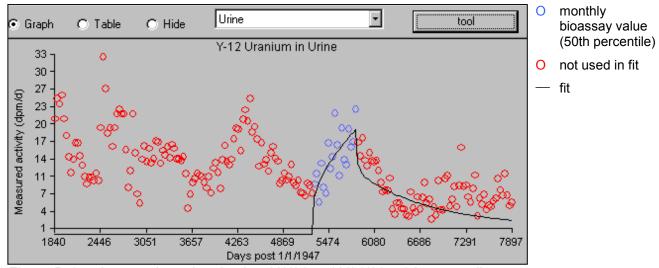


Figure B-17. Assumed uranium intake, 6/1/61 to 12/31/62, 50th-percentile results, type S.

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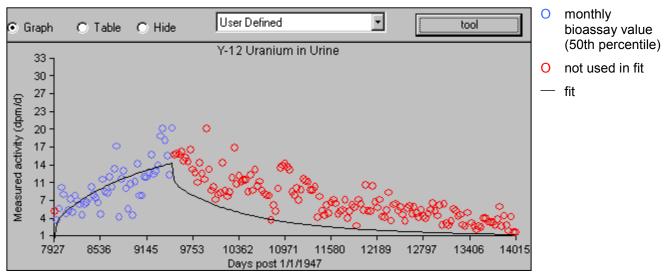


Figure B-18. Assumed uranium intake, 10/1/68 to 12/31/72, 50th-percentile results, type S.

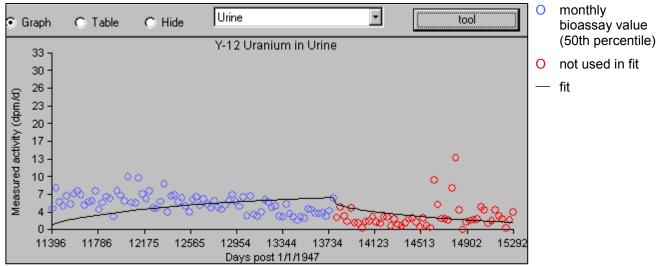


Figure B-19. Assumed uranium intake, 3/1/78 to 9/30/84, 50th-percentile results, type S.

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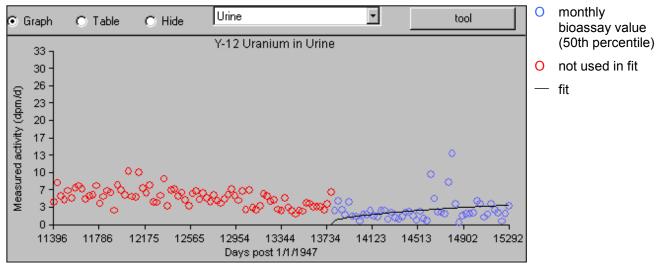


Figure B-20. Assumed uranium intake, 10/1/84 to 12/31/88, type S.

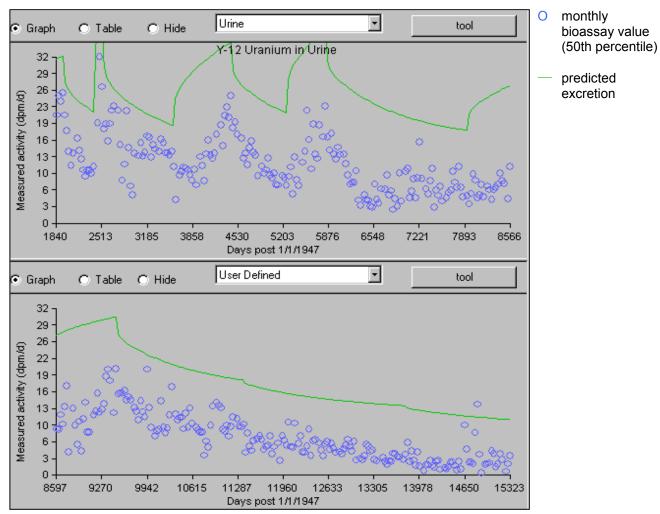


Figure B-21. Predicted (type S, 5 μ m) uranium urinary excretion from eight independent intakes, 50th percentile, 1952 to 1988.

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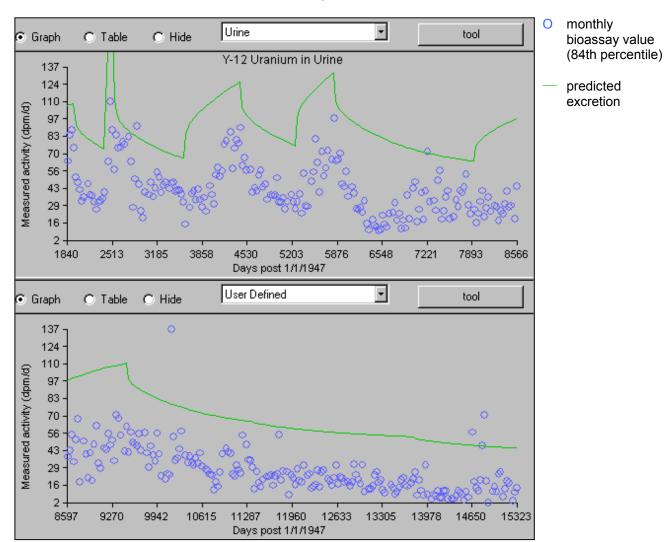


Figure B-22. Predicted (type S, 5 µm) uranium urinary excretion from eight independent intakes, 84th percentile, 1952 to 1988.