

# ORAU TEAM Dose Reconstruction Project for NIOSH

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Page 1 of 81

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Y-12 Plant – Occupational Internal Dose	Effective Date:	08/03/2023
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Revision

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 2 of 81

# PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
03/17/2004	00	New document to establish TBD for the Y-12 National Security Complex – Occupational Internal Dose. First approved issue. Initiated by William E. Murray.
05/10/2005	01	First revision to include table numbers and titles in the body of the document. Approved issue of Revision 01. Initiated by William E. Murray.
10/11/2005	01 PC-1	Page change initiated to incorporate the definition of U.S.C. on page 6 and details for the definition of a DOE facility on page 7. No sections were deleted. First approved page change revision for Revision 01. Initiated by William E. Murray.
01/12/2006	01 PC-2	No changes occurred as a result of formal internal review. Page change initiated to make minor changes to text in Section 5.1, page 7. Deletes and adds text about the default value for the Th-228/Th- 232 equilibrium ratio for lung counts in Section 5.3, page 31. Adds new Figure 5.4 Variation of activity in natural thorium after a single purification on page 31. Also, changes date in C. M. West reference on page 36. No sections were deleted. Training required: As determined by the Task Manager. Initiated by William E. Murray.
02/14/2006	01 PC-3	Approved page change revision. Page changes initiated to incorporate definitions and directions for dose reconstruction for non- presumptive cancers that are excluded from the 1943 through 1947 Special Exposure Cohort. Affected pages are: page 7, page 9 in Section 5.1, pages 20 and 21 in Section 5.2.1.1, and page 36 in the Reference Section. No sections were deleted. Training required: As determined by the Task Manager. Initiated by William E. Murray.
08/03/2006	02	Revised to incorporate additional directions for dose reconstruction for non-presumptive cancers that are excluded from the 1943 through 1947 Special Exposure Cohort. Approved issue of Revision 02. Removed Section 5.4 from the text and Table of Contents, as the most recent page change addresses 1943 to 1947, and coworker data are used for 1948 to 1950, as specified in the coworker TIB. Incorporates NIOSH formal review comments. This revision results in no change to the assigned dose and no PER is required. Training required: As determined by the Task Manager. Initiated by William E. Murray.
10/10/2006	02 PC-1	Page change revision initiated to incorporate Y-12 National Security Complex Special Exposure Cohort class descriptions in newly added Section 5.1.3 on page 10. Minor changes occurred on pages 6, 8, and 9. Added one reference on page 37. Incorporates one new acronym, editorial changes on pages 8, 9, and 10, modifications to SEC instructions in Section 5.1.3.1 on page 10. Two references were deleted from the Reference Section. Incorporates internal and NIOSH formal review comments. No sections were deleted. This revision results in no change to the assigned dose and no PER is required. Training required: As determined by the Task Manager. Initiated by William E. Murray.

Effective Date: 08/03/2023 Page 3 of 81

EFFECTIVE REVISION NUMBER DESCRIPTION DATE 03/12/2012 Revision to add new SEC designation to Section 5.1.3 to clarify, 03 correct, and update the SEC class of covered employees. Added U-232 exception to Section 5.2.2. Added internal coworker study (formerly ORAUT-OTIB-0029) as Attachment B. Revision initiated to add Attributions and Annotations section and make minor editorial corrections. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Steven R. Reed. 08/03/2023 04 Revision to incorporate SEC-00250, address outstanding priority ABRWH issues, and bring into ORAUT-PROC-0031 compliance as specified during DCAS/ORAUT discussions. Changed title of document to use Y-12 Plant rather than Y-12 National Security Complex. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by Objective Manager. Initiated by John M. Byrne and authored by Stephanie L. Felch.

3/03/2023 Page 4 of 81

# TABLE OF CONTENTS

# **SECTION**

# <u>TITLE</u>

# <u>PAGE</u>

Acrony	/ms and	d Abbrev	iations	7
5.1	Introdu 5.1.1 5.1.2 5.1.3	Purpos Scope.	e Exposure Cohort	10 10
5.2	Uraniu 5.2.1 5.2.2 5.2.3 5.2.4	Particle Uraniur Tempor Other F	ility in the Lung Size Information n Alpha Activity as a Function of Enrichment ral Pattern of Uranium Exposures Radionuclides of Concern Including Recycled Uranium Contaminants Unmonitored Radionuclides from Recycled Uranium	14 14 15 16
5.3	5.3.1 5.3.2 5.3.3 5.3.4	Covera Sample Minimu In Vitro 5.3.4.1 5.3.4.2 5.3.4.3 5.3.4.4	Plutonium Tritium	23 24 27 27 27 29 30 30
5.4	In Vivo 5.4.1 5.4.2	Whole Chest 0 5.4.2.1 5.4.2.2	m Detectable Activities, Counting Methods, and Reporting Practices Body Counting Counting Uranium Other Actinides Other Radionuclides	30 31 31 32
5.5	Interna	al Dose F	Reconstruction Guidance	34
Refere	ences			37
Glossa	ary			44
	CHMEN CHMEN		OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS	

LIST OF TABLES

<u>TITLE</u>

5-1 5-2	Enrichment assumptions for dose assessment for analytical techniques Y-12 uranium receipts summary presenting average levels of three predominant RU	15
0-2	contaminants through plant history, 1953 to 1999	17
5-3	Total Y-12 RU receipts summary	
5-4	Primary contaminants (at 3-year decay) in VHEU fuels and of concern in internal dose	
	reconstruction	19
5-5	Comparison of the production of three selected isotopes in VHEU spent fuel with levels reported in HEU shipments to Y-12	19
5-6	Ranges of reported contaminant values in the main processing facilities	
5-7	Typical uranium enrichment materials at Y-12 with calculated and inferred RU	
	contaminant levels	21
5-8	Recommended defaults based on upper levels of expected ranges	22
5-9	L <sub>D</sub> values	
5-10	<sup>233</sup> U to <sup>232</sup> U activity ratios	
5-11	Reported lung counting detection limits for <sup>235</sup> U	
5-12	Reported lung counting detection limits for <sup>238</sup> U	
5-13	Calculated lung counting detection limits for <sup>212</sup> Pb and <sup>228</sup> Ac, 1979 to 1991	
A-1	Uranium urinalysis detection limits	
A-2	Other in vitro detection limits	
A-3	Reported in vivo detection limits for other radionuclides	
B-1	Monthly uranium 24-hour urinary excretion rate analyses, 1952 to 1988	59
B-2	Type F uranium intake periods and rates	
B-3	Type M uranium intake periods and rates	65
B-4	Type S uranium intake periods and rates	66
B-5	Type F uranium intake periods and rates	66
B-6	Type M uranium intake periods and rates	67
B-7	Type S uranium intake periods and rates	67

# LIST OF FIGURES

# **FIGURE**

<u>TABLE</u>

# TITLE

# <u>PAGE</u>

<u>PAGE</u>

5-1	<sup>212</sup> Pb MDAs at various CWTs for a 30-minute count	. 34
B-1	Assumed uranium intake, 01/01/1947 to 12/31/1969, 50th-percentile results, type F	. 68
B-2	Assumed uranium intake, 01/01/1970 to 12/31/1988, 50th-percentile results, type F	. 69
B-3	Assumed uranium intake, 01/01/1947 to 02/28/1978, 50th-percentile results, type M	. 70
B-4	Assumed uranium intake, 01/01/1947 to 04/30/1952, 50th-percentile results, type M	. 70
B-5	Assumed uranium intake, 08/01/1953 to 11/30/1953, 50th-percentile results, type M	.71
B-6	Assumed uranium intake, 11/01/1956 to 02/28/1959, 50th-percentile results, type M	.71
B-7	Assumed uranium intake, 06/01/1961 to 12/31/1962, 50th-percentile results, type M	.72
B-8	Assumed uranium intake, 10/01/1968 to 12/31/1972, 50th-percentile results, type M	.72
B-9	Assumed uranium intake, 03/01/1978 to 09/30/1984, 50th-percentile results, type M	.73
B-10	Assumed uranium intake, 10/01/1984 to 12/31/1988, 50th-percentile results, type M	.73
B-11	Predicted uranium urinary excretion from eight independent intakes, 50th percentile,	
	1952 to 1988	. 74
B-12	Predicted uranium urinary excretion from eight independent intakes, 84th percentile,	
	1952 to 1988	. 75
B-13	Assumed uranium intake, 01/01/1947 to 02/28/1978, 50th-percentile results, type S	.76

Document No. ORAUT-TKBS-0014-5 Revision No. 04 Effective Date: 08/	/03/2023 Page 6 of 8

B-14	Assumed uranium intake, 01/01/1947 to 04/30/1952, 50th-percentile results, type S	76
B-15	Assumed uranium intake, 08/01/1953 to 11/30/1953, 50th-percentile results, type S	77
B-16	Assumed uranium intake, 11/01/1956 to 02/28/1959, 50th-percentile results, type S	77
B-17	Assumed uranium intake, 06/01/1961 to 12/31/1962, 50th-percentile results, type S	78
B-18	Assumed uranium intake, 10/01/1968 to 12/31/1972, 50th-percentile results, type S	78
B-19	Assumed uranium intake, 03/01/1978 to 09/30/1984, 50th-percentile results, type S	79
B-20	Assumed uranium intake, 10/01/1984 to 12/31/1988, 50th-percentile results, type S	79
B-21	Predicted uranium urinary excretion from eight independent intakes, 50th percentile,	
	1952 to 1988	80
B-22	Predicted uranium urinary excretion from eight independent intakes, 84th percentile,	
	1952 to 1988	81

# ACRONYMS AND ABBREVIATIONS

ABRWH	Advisory Board on Radiation and Worker Health
AMAD	activity median aerodynamic diameter
AWE	Atomic Weapons Employer
CEDE	committed effective dose equivalent
CER	Center for Epidemiologic Research
Ci	curie
cpm	counts per minute
CWT	chest wall thickness
d	day
DCF	dose conversion factor
DF	decontamination factor
DL	decision level
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
dpm	disintegrations per minute
DU	depleted uranium
EEOICPA EU	Energy Employees Occupational Illness Compensation Program Act of 2000 enriched uranium
F	fast (absorption type)
FMPC	Feed Materials Production Center
g	gram
GSD	geometric standard deviation
hr	hour
HEU	highly enriched uranium
HTO	tritiated water
ICPP	Idaho Chemical Processing Plant
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
in.	inch
IREP	Interactive RadioEpidemiological Program
kg	kilogram
L	liter
L <sub>C</sub>	critical level
L⊳	detection level
LEU	low-enriched uranium
M	moderate (absorption type)
MDA	minimum detectable activity
mg	milligram
mL	milliliter
mm	millimeter
mrem	millirem

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 8 of 81

Nal(TI)	thallium-drifted sodium iodide
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NU	natural uranium
ORAU	Oak Ridge Associated Universities
ORAUT	ORAU Team
pCi	picocurie
PER	program evaluation report
pH	negative logarithm of the hydrogen ion concentration, a measure of alkalinity or acidity
ppb	parts per billion
ppm	parts per million
R&D	research and development
RU	recycled uranium
RWP	radiation work permit
S	slow (absorption type)
SEC	Special Exposure Cohort
SMC	Specific Manufacturing Capability
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
TBD	technical basis document
TIB	technical information bulletin
UCC U.S.C.	Union Carbide Corporation <i>United States Code</i>
VHEU	very highly enriched uranium
wt%	weight percent
yr	year
§	section or sections
μCi	microcurie
μg	microgram
μm	micrometer

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 9 of 81
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#### 5.1 INTRODUCTION

Technical basis documents (TBDs) and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular U.S. Department of Energy (DOE) or Atomic Weapons Employer (AWE) facilities or categories of DOE or AWE facilities. They will be revised in the event additional relevant information is obtained about the affected DOE or AWE facility(ies), such as changing scientific understanding of operations, processes, or procedures involving radioactive materials. These documents may be used to assist NIOSH staff in the evaluation of Special Exposure Cohort (SEC) petitions and the completion of individual dose reconstructions under Part B of the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA).

In this document the word "facility" is used to refer to an area, building, or group of buildings that served a specific purpose at a DOE or AWE facility. It does not mean nor should it be equated to an "AWE facility" or a "DOE facility." The term "AWE facility" is defined in EEOICPA to mean "a facility, owned by an atomic weapons employer, that is or was used to process or produce, for use by the United States, material that emitted radiation and was used in the production of an atomic weapon, excluding uranium mining or milling." 42 *United States Code* (U.S.C.) § 7384I(5). On the other hand, a DOE facility is defined as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located—(A) in which operations are, or have been, conducted by, or on behalf of, the [DOE] (except for buildings, structures, premises, grounds, or operations … pertaining to the Naval Nuclear Propulsion Program); and (B) with regard to which the [DOE] has or had—(i) a proprietary interest; or (ii) entered into a contract with an entity to provide management and operation, management and integration, environmental remediation services, construction, or maintenance services." 42 U.S.C. § 7384I(12). The DOE determines whether a site meets the statutory definition of an AWE facility and the U.S. Department of Labor (DOL) determines if a site is a DOE facility and, if it is, designates it as such.

Under EEOICPA, a Part B cancer claim for benefits must be based on an energy employee's eligible employment and occupational radiation exposure at a DOE or AWE facility during the facility's designated time period and location (i.e., a "covered employee with cancer"). After DOL determines that a claim meets the eligibility requirements under Part B of EEOICPA, DOL transmits the claim to NIOSH for a dose reconstruction. EEOICPA provides, among other things, guidance on eligible employment and the types of radiation exposure to be included in an individual dose reconstruction. Under EEOICPA, eligible employment at a DOE facility includes individuals who are or were employed by DOE and its predecessor agencies, as well as their contractors and subcontractors at the facility. 42 U.S.C. § 7384I(11). Also under EEOICPA, the types of exposure to be included in dose reconstructions for DOE employees are those radiation exposures incurred in the performance of duty. As such, NIOSH includes all radiation exposures received as a condition of employment at DOE facilities in its dose reconstructions for covered employees, which may include radiation exposures related to the Naval Nuclear Propulsion Program at DOE facilities, if applicable. This is because NIOSH does not determine the fraction of total measured radiation exposure at a DOE facility that is contributed by the Naval Nuclear Propulsion Program at the DOE facility during a specified period of time for inclusion in dose reconstruction.

NIOSH does not consider the following types of exposure as those incurred in the performance of duty as a condition of employment at a DOE facility. Therefore these exposures are not included in dose reconstructions for covered employees [NIOSH 2010]:

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

Document No. ORAUT-TKBS-0014-5 Revision No. 04 Effective Date: 08/03/2023 Pa	Document No. ORAUT-TKBS-	23 Page 10 of 81
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#### 5.1.1 <u>Purpose</u>

This TBD describes internal dosimetry systems and practices at the Y-12 Plant. 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 <u>Scope</u>

The Y-12 Plant, operated by Consolidated Nuclear Security, LLC 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 <sup>235</sup>U enrichment. Therefore, this section begins with an overview of concepts that apply broadly to the history of uranium work at Y-12. Attachment A summarizes some of the key points from the following sections. Attachment B provides co-exposure 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 <sup>235</sup>U 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.

The 86-Inch Cyclotron began operating on November 11, 1950, and operated until the early 1980s. The 86-Inch Cyclotron was later used to create neutron-deficient radionuclides as a part of the research and development (R&D) program noted above. Some of these R&D efforts involved plutonium, and workers could have plutonium bioassay results in their records [Patterson et al. 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 Cohort

The Secretary of the U.S. Department of Health and Human Services has issued six designations of classes of Y-12 employees as additions to the SEC.

Document No. ORAUT-TKBS-0014-5 Revision No. 04 Effective I	Date: 08/03/2023	Page 11 of 81
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#### March 1, 1943, to December 31, 1947

Department of Energy (DOE) employees or DOE contractor or subcontractor employees who worked in uranium enrichment operations or other radiological activities at the Y-12 facility in Oak Ridge, Tennessee from March 1943 through December 1947 and who were employed for a number of work days aggregating at least 250 work days, either solely under this employment or in combination with work days within the parameters (excluding aggregate work day requirements) established for other classes of employees included in the SEC [Leavitt 2005, p. 3].

NIOSH determined that it lacked access to sufficient information to estimate either the maximum radiation dose incurred by any member of the class or to estimate such radiation doses more precisely than a maximum dose estimate [NIOSH 2005].

A second designation extended the class to include all workers in all areas of Y-12 during this period:

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 [Leavitt 2008, p. 3].

NIOSH determined that it lacked the internal dosimetry data necessary to reconstruct the internal exposures from uranium enrichment and other radiological activities. In addition, NIOSH determined that, because exposure potential might not have been limited to only specific buildings or groups of workers at Y-12, the SEC class definition should include all employees in 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, to December 31, 1947 [NIOSH 2008]. NIOSH considers it feasible to adequately reconstruct the occupational medical dose for Y-12 workers for the covered period. Although no exposure data are available for the medical X-rays performed during this time, surrogate data were used to develop an exposure matrix for the Y-12 site profile [Oak Ridge Associated Universities (ORAU) Team (ORAUT) 2007a].

#### January 1, 1948, to December 31, 1957

Department of Energy (DOE) employees or DOE contractor or subcontractor employees who were monitored or should have been monitored for:

- (1) thorium exposures while working in Building 9201-3, 9202, 9204-1, 9204-3, 9206, or 9212 at Y-12 for a number of work days aggregating at least 250 work days from January 1948 through December 1957 or in combination with work days within the parameters (excluding aggregate work day requirements) established for one or more classes of employees in the SEC; or
- (2) radionuclide exposures associated with cyclotron operations in Building 9201-2 at Y-12 for a number of work days aggregating at least 250 work days from January 1948 through December 1957 or in combination with work days within the parameters (excluding aggregate work day requirements) established for one or more classes of employees in the SEC [Leavitt 2006, p. 3].

NIOSH determined that the specific biological monitoring data, air monitoring information, process and radiological source information, and surrogate data from similar operations at other sites that would allow it to estimate, with sufficient accuracy, the potential internal radiological exposures to thorium

Document No. ORAUT-TKBS-0014-5 Rev	ision No. 04    Effe	fective Date: 08/03/2023	Page 12 of 81
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and cyclotron radionuclides are not sufficient to complete dose reconstructions for the class [NIOSH 2006a,b].

A second designation extended the class to include all workers in all areas of Y-12 during this period:

All employees of the Department of Energy, its predecessor agencies, and their contractors and 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 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort [Sebelius 2011, p. 3].

NIOSH determined that, due to undocumented worker movements across the site, limited worker-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 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 determined that it was necessary to remove the area-specific and monitoring criteria from the class description associated with thorium and cyclotron exposures for the period. NIOSH determined that it was also necessary to expand the SEC class definition to include all employees and areas of Y-12 during this period [NIOSH 2011]. In its evaluation [NIOSH 2011], NIOSH found that internal exposure could be reconstructed for all employees from January 1, 1948, to December 31, 1957, with the exception of internal exposure to thorium and cyclotron radionuclides. NIOSH found that external exposure, including occupational medical doses, could be reconstructed for all employees between January 1948 and December 31, 1957.

#### January 1, 1958, to December 31, 1976

All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Y-12 Plant in Oak Ridge, Tennessee, during the period January 1, 1958, through December 31, 1976, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort [Azar 2019a, p. 3].

NIOSH determined that it lacked sufficient information to allow it to estimate with sufficient accuracy the internal exposures to thorium (and associated progeny) for the period from January 1, 1958, to December 31, 1976, and <sup>241</sup>Pu for the period from January 1, 1958, to December 31, 1966 [NIOSH 2018].

Although NIOSH has determined that there are in vivo monitoring data for thorium (i.e., lung counts) during the period from January 1, 1958, to December 31, 1976, these data are recorded in total thorium mass. NIOSH is unable to use these data to determine the associated quantities of <sup>232</sup>Th, <sup>228</sup>Th, and <sup>228</sup>Ra. For this reason, NIOSH cannot determine with sufficient accuracy the internal exposures that might be represented by each thorium lung measurement. NIOSH has also evaluated the available gross alpha air monitoring data at the Y-12 Plant and determined that they cannot be used to accurately reconstruct internal exposure from thorium and associated progeny. NIOSH has not identified biological monitoring data specific to <sup>241</sup>Pu that can be used to reconstruct <sup>241</sup>Pu exposure during the period from January 1, 1958, to December 31, 1966. NIOSH has identified sufficient monitoring data to reconstruct <sup>241</sup>Pu beginning in 1967 [NIOSH 2018].

#### January 1, 1977, to July 31, 1979

All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Y-12 Plant in Oak Ridge, Tennessee, during the period between January 1, 1977, and July 31, 1979, 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 [Azar 2019b, p. 3].

NIOSH determined that it lacked sufficient information to allow it to estimate with sufficient accuracy internal exposures to thorium (and associated progeny) for the period from January 1, 1977, to July 31, 1979, due to a lack of sufficient thorium-related lung count data for this period [NIOSH 2019].

Although NIOSH has determined that there are in vivo monitoring data for thorium (i.e., lung counts) during the period from January 1, 1977, to July 31, 1979, these data are recorded in total thorium mass. NIOSH is unable to use these data to determine the associated quantities of <sup>232</sup>Th, <sup>228</sup>Th, and <sup>228</sup>Ra. For this reason, NIOSH cannot determine with sufficient accuracy the internal exposures that might be represented by each thorium lung measurement. Air-sampling data specific to thorium for the period under evaluation (January 1, 1977, to December 31, 1994) are not available to NIOSH [NIOSH 2019, p. 42]. Consistent with its findings in the earlier Y-12 Plant evaluation reports, NIOSH finds that there is sufficient monitoring and source term information available to reconstruct external dose, including occupational medical dose, with sufficient accuracy for all Y-12 Plant employees during the period from January 1, 1977, to July 31, 1979.

#### **SEC Classes Summary**

Although NIOSH found that it is not possible to reconstruct radiation doses completely for the SEC classes, NIOSH intends to use any internal and external monitoring data that might become available, and can be interpreted using its existing dose reconstruction processes or procedures, for an individual claim. Therefore, for individuals employed at Y-12 during the period from March 1, 1943, to July 31, 1979, who do not qualify for inclusion in the SEC, dose reconstructions may be performed using these data as appropriate. All dose reconstructions for all workers having employment during an SEC period are considered partial dose reconstructions.

For the March 1, 1943, to December 31, 1947 period, NIOSH concludes that external doses for workers directly involved with the calutron uranium enrichment and the occupational medical dose for all workers can be reconstructed [NIOSH 2005, 2008]. NIOSH found that external exposure, including occupational medical doses, could be reconstructed for all employees from January 1, 1948, to July 31, 1979, as determined in the evaluations of SEC-00018 [NIOSH 2005], SEC-00028 [NIOSH 2006a], SEC-00098 [NIOSH 2008], SEC-00186 [NIOSH 2011], SEC-00251 [NIOSH 2018], and SEC-00250 [NIOSH 2019].

#### 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 2007b]. 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.

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

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 14 of 81

default modeling assumption was class Q (90% Super-W, 10% Y) [Barber and Forrest 1995]. 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].

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 absorption 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. 10]. 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 2007b, p. 24].

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., Struxness 1953; Steckel and West 1966; Barber and Forrest 1995; Veinot 2003, Section 5]. 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 activity median aerodynamic diameter (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.)

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. This is the justification for the change to the 5- $\mu$ m AMAD as the default particle size. Y-12 uses the newer ICRP Publication 60 [ICRP 1991] weighting factors [Snapp 2003b, p. 119]. For Y-12 dose reconstructions, the default particle size distribution is 5  $\mu$ m AMAD.

#### 5.2.2 Uranium Alpha Activity as a Function of Enrichment

At Y-12, uranium enrichments range from depleted uranium (DU; less than 0.71 wt% <sup>235</sup>U) to very highly enriched uranium (VHEU; greater than 93 wt% <sup>235</sup>U). 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 (DCFs) 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 [AEC 1959]. Early in vivo chest-counting results were reported either as  $\mu g$  of <sup>235</sup>U or mg of <sup>238</sup>U based on whether an exposure was believed to be from enriched uranium

Document No. ORAUT-TKBS-0014-5 Revision No. 04 Effecti
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(EU), natural uranium (NU), or DU [Cofield 1959a; Scott and West 1975]. Based on these reporting techniques, the assumptions in Table 5-1 can be made about data interpretation for the periods from 1948 to 1989 for urinalysis and from 1961 to 1989 for lung counting.

Table 5-1. Enrichment assumptions for dose assessment for analytical techniques. <sup>a</sup>
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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.	93% before 1950, NU from 1950 to 1989
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 based on the guidance in ORAUT 2018.	93% enrichment
Lung count	μg Ū-235	93% enrichment
Lung count	μg U-238	NU

a. Sources: AEC [1959]; Cofield [1959a]; Scott and West [1975].

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 (<sup>234</sup>U, <sup>235</sup>U, <sup>236</sup>U, and <sup>238</sup>U) can be summed under certain circumstances as outlined in ORAUT-OTIB-0060, *Internal Dose Reconstruction* [ORAUT 2018]. Note that <sup>234</sup>U DCFs are not appropriate substitutes for <sup>232</sup>U, as the DCFs for <sup>232</sup>U are typically 5-10 times larger than those for <sup>234</sup>U.

#### 5.2.3 <u>Temporal Pattern of Uranium Exposures</u>

As a rule, routine uranium exposures at Y-12 were considered to be of a chronic nature. For example, AEC [1959, p. 70] 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" [Snapp 2003b, p. 111]. 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 plantwide basis (see Table 5-8 in Section 5.2.4.1 for plantwide RU contaminant defaults).

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

In addition to <sup>234</sup>U, <sup>235</sup>U, and <sup>238</sup>U, the following radionuclides were identified in the Y-12 *Technical Basis Document for Internal Dosimetry* [Snapp 2003b]:

- <sup>3</sup>H, <sup>90</sup>Sr, and <sup>99</sup>Tc;
- <sup>228</sup>Th and <sup>232</sup>Th;
- <sup>232</sup>U, <sup>233</sup>U, and <sup>236</sup>U;
- <sup>238</sup>Pu, <sup>239</sup>Pu, and <sup>241</sup>Pu; and
- <sup>237</sup>Np and <sup>241</sup>Am.

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

Other radionuclides addressed included <sup>60</sup>Co and <sup>95</sup>Zr/<sup>95</sup>Nb 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 <sup>131</sup>I, <sup>99m</sup>Tc, <sup>67</sup>Ga, and <sup>201</sup>TI.

### 5.2.4.1 Unmonitored Radionuclides from Recycled Uranium

One of the primary sources of significant contaminants (including the production of other than the normal uranium isotopes) that were introduced into the Y-12 plant systems were those associated with RU from 1953 until 1999 [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, <sup>236</sup>U 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 computer-generated listing of the relative production of a few isotopes of concern, following irradiation of VHEU fuel in a high-flux test reactor. The process streams included a spectrum of chemical forms of uranium, including uranyl nitrate  $[UO_2(NO_3)_2]$  solutions, uranium trioxide  $(UO_3)$ , uranium-aluminum (U-AI) alloy ingots, uranium scrap, uranium tetrafluoride  $(UF_4)$ , uranium metal, solvent extraction raffinates, and a variety of secondary process wastes and residues [Rich et al. 2000; DOE 2003].

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 17 of 81
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Table 5-2. Y-12 uranium receipts summary presenting average levels of three predominant RU contaminants through plant history, 1953 to 1999.<sup>a</sup>

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	U	Pu	Pu	Pu	Np	Np	Np	Тс	Tc	Тс
Site	(kg)	(g)	(ppb)	(pCi/µg U)	(g)	(ppm)	(pCi/µg U)	(g)	(ppm)	(pCi/µg U)
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 totals/avg.	150,857	0.04674	0.3	4.3E-03	3,666	23	1.6E-02	14,499	96	1.64

#### Y-12 VHEU receipts summary

#### Y-12 DU receipts summary<sup>b</sup>

Site	U (kg)
Hanford	1,502
Oak Ridge Gaseous Diffusion Plant	192,836
Paducah Gaseous Diffusion Plant	38,423
DU total	232,761
Grand total	383,618

a. Source: Rich et al. [2000].

b. No contaminant data for DU from these sites.

#### Table 5-3. Total Y-12 RU receipts summary.<sup>a</sup>

Shipping site	U (kg)
SRS	153,000
ICPP	25,700
Hanford	4,400
West Valley	900
Total	184,000

Total	U	Pu	Pu	Pu	Nр	Np	Np	Тс	TC	Tc
	(kg)	(g)	(dqq)	(pCi/µa U)	(g)	(ppm)	(pCi/ug U)	(g)	(mqq)	(pCi/μg U)
Totals/averages	184,000	0.5	2.7	0.039	54	0.003	0.00021	9,100	49.5	0.85

a. Source: DOE [2003].

The uranium enrichments ranged from DU, NU, low-enriched uranium (LEU - less than 20% <sup>235</sup>U), highly enriched uranium (HEU - from 20 to approximately 93% <sup>235</sup>U), and VHEU (more than 93% <sup>235</sup>U) [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 (FMPC) in Fernald, Ohio, which also had an RU contaminant inventory. Therefore, the DU contaminant levels at Y-12 were inferred from the FMPC and Idaho Specific Manufacturing Capability (SMC) projects recycle reports [ORAUT 2017; 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

[DOCUMENT NO. ORAUT-TRDS-0014-5] REVISION NO. 04 [Effective Date. 06/03/2023] Page 10 01 0	Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 18 of 81
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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, the FMPC, and others, which also had RU contaminant inventories. Thus, the discrepancies in the 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 claimant 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 <sup>228</sup>Th 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 from 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].

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 19 of 81

Isotope	Half-life	Activity (μCi/μg total U)	Activity (µCi/g total U)	Percent weight g/100 g U <sup>b</sup>	ppm U	Activity ratio (isotope/ total U)
Tc-99	2.11E+05 yr	3.8E-04	3.8E+02	2.2E+00	2.2E+04	2.4E+01
Th-227	18.72 d	3.3E-11	3.3E-05	1.1E-13	1.1E-09	Not applicable
Th-228	1.913 yr	1.6E-07	1.6E-01	1.9E-08	1.9E-04	1.0E-02
Th-229	7.34E+03 yr	1.4E-12	1.4E-06	6.7E-10	6.7E-06	Not applicable
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	1.0E-01
Th-232	1.41E+10 yr	3.5E-15	3.5E-09	3.2E-06	3.2E-02	2.2E-10
Total Th	Not applicable	Not applicable	1.9E+00	Not applicable	Not applicable	Not applicable
U-232	6.89E+01 yr	2.7E-07	2.7E-01	1.3E-06	1.3E-02	Not applicable
U-233	1.59E+05 yr	2.6E-10	2.6E-04	2.7E-06	2.7E-02	Not applicable
U-234	2.46E+05 yr	6.6E-07	6.6E-01	1.0E-02	1.0E+02	Not applicable
U-235	7.04E+08 yr	1.7E-06	1.7E+00	7.8E+01	7.8E+05	Not applicable
U-236	2.34E+07 yr	1.3E-05	1.3E+01	2.0E+01	2.0E+05	Not applicable
U-237	6.75 d	3.6E-08	3.6E-02	4.4E-11	4.4E-07	Not applicable
U-238	4.47E+09 yr	5.6E-09	5.6E-03	1.7E+00	1.7E+04	Not applicable
Total U	Not applicable	Not applicable	15.7E+00	Not applicable	Not applicable	Not applicable
Np-236	1.15E+05 yr	9.4E-11	9.4E-05	7.2E-07	7.2E-03	Not applicable
Np-237	2.14E+06 yr	9.1E-06	9.1E+00	1.3E+00	1.3E+04	6.0E-01
Np-238	2.117 d	9.2E-10	9.2E-04	3.6E-13	3.6E-09	Not applicable
Np-239	2.355 d	4.3E-08	4.3E-02	1.9E-11	1.9E-07	Not applicable
Total Np	Not applicable	Not applicable	9.1E+00	Not applicable	Not applicable	Not applicable
Pu-236	2.851 yr	3.6E-06	3.6E+00	6.8E-07	6.8E-03	0.2E+00
Pu-238	87.70 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	1.2E+00
Pu-240	6.54E+03 yr	1.5E-05	1.5E+01	6.4E-03	6.4E+01	1.0E+00
Pu-241	14.4 yr	1.5E-03	1.5E+03	1.4E-03	1.4E+01	1.0E+02
Pu-242	3.73E+05 yr	8.8E-09	8.8E-03	2.3E-04	2.3E+00	5.6E-04
Total Pu	Not applicable	Not applicable	4.6E+04	Not applicable	Not applicable	Not applicable
Am-241	4.32E+02 yr	9.6E-06	9.6E+00	2.8E-04	2.8E+00	6.0E-01

Table 5-4. Primary contaminants (at 3-year decay) in VHEU fuels and of concern in internal dose reconstruction.<sup>a</sup>

a. Sources: Wenzel [2000, p 104 (Zr column)]; Wenzel [2004, pp. 39, 41, 42, 43, 45, 46].
b. 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.

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 fuel⁵	Approximate derived DF <sup>c,d</sup>
Th-228	0.01-0.03	Not applicable <sup>a</sup>	0.16	10
Np-237	0.01-0.02	1E-03	9.1	1E+03–1E+04
Pu-238	4E-03	3E-04	3.6E+04	1E+07–1E+08

a. There were no analytical data for Th-228 reported in the ICPP Y-12 receipt reports.

b. These values were taken directly from column 4 in Table 5-4.

c. DF = decontamination factor.

d. The DF is interpolated from Y-12 process stream analytical data.

Comparing the concentrations in the spent fuels with the reported concentrations in the product received at Y-12 provides a rough indication of the decontamination factors (DFs) of the chemical extraction processes. Reference to these DFs in Table 5-5 and the average contaminant

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 20 of 81
--------------------------------	-----------------	----------------------------	---------------

concentrations in Table 5-6 indicate that plutonium is the most efficiently removed with an effective DF of approximately  $1 \times 10^7$  to  $10^8$ , with a neptunium DF of approximately  $1 \times 10^4$ , while thorium appears to follow the chemistry of uranium more closely, resulting in a DF of only 10, which in turn results in a relative enrichment of <sup>228</sup>Th in the VHEU product received at Y-12. Although <sup>228</sup>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 <sup>228</sup>Th [Oliver 1997; BWXT Y-12 2001]. These surface contaminants [BWXT Y-12 2001]. The uranium-to-nonuranium alpha ratios range from 4 to more than  $1 \times 10^4$ , which is not useful for dose reconstruction. However, these data do indicate that <sup>228</sup>Th is one of the primary contaminants at Y-12, which in turn provides a technical base for inclusion of <sup>228</sup>Th in the list of RU contaminant isotopes.

Table 5-6. Ranges of reported contaminant values in the main processing facilities.<sup>a,b</sup>

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

a. Source: BWXT Y-12 [2000, pp. 156-157].

b. The highest values reported in the processing facilities were in just 1 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.

c. The raffinate streams are depleted in uranium to a range of 5 ppm U levels in the sludge. Therefore, the ppm 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).

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 2003b, p. 68]. 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 according to 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 plantwide basis. 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 <sup>235</sup>U or <sup>238</sup>U indicated in the bioassay results. For each µg or pCi of uranium intake calculated based on bioassay data in the claimant files, an additional intake of each of the four RU contaminants should be added.

	Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 21 of 81
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Table 5-7. Typical uranium enrichment materials at Y-12 with calculated and inferred RU contaminant levels.<sup>a</sup>

Uranium isotope	Activity (Ci/g)	Mass fraction	Activity (pCi/µg total U)	RU contaminant	Activity (pCi/µg total U)
233	9.66E-03	1.000E-04	9.660E-01	Pu-238	6.20E-03
234	6.22E-03	1.280E-02	7.962E+01	Pu-239	1.00E-04
235	2.16E-06	5.220E-01	1.128E+00	Np-237	2.00E-03
236	6.50E-05	2.920E-01	1.898E+01	Tc-99	1.90E+00
238	3.36E-07	1.730E-01	5.796E-02	Th-228	3.17E-02
Totals	Not applicable	9.990E-01	1.007E+02	Total α	4.00E-02
dpm/µg total U	Not applicable	Not applicable	2.24E+02	α dpm/μg total U	8.87E-02
<u> </u>		• • •		U α:Non U α	2.52E+03

### High burn-up (~50%) recycled VHEU from SRS<sup>b</sup>

# Lower burn-up (~25%) recycled VHEU from ICPP<sup>b</sup>

Activity		Activity
(Ci/g)	Mass fraction	(pCi/µg total U)
9.66E-03	3.000E-06	2.90E-02
6.22E-03	1.000E-02	6.22E+01
2.16E-06	7.800E-01	1.68E+00
6.50E-05	2.000E-01	1.30E+01
3.36E-07	1.700E-02	5.71E-03
Not applicable	1.007E+00	7.69E+01
Not applicable	Not applicable	1.71E+02
	(Ci/g) 9.66E-03 6.22E-03 2.16E-06 6.50E-05 3.36E-07 Not applicable	(Ci/g)         Mass fraction           9.66E-03         3.000E-06           6.22E-03         1.000E-02           2.16E-06         7.800E-01           6.50E-05         2.000E-01           3.36E-07         1.700E-02           Not applicable         1.007E+00

Activity				
(pCi/µg total U)				
2.00E-03				
3.30E-05				
2.00E-03				
1.50E–01				
5.00E-02				
5.40E-02				
1.20E–01				
1.4E+03				

#### Weapons-grade Oralloy VHEU<sup>b,c</sup>

Uranium isotope	Activity (Ci/g)	Mass fraction	Activity (pCi/µg total U)
233	9.66E-03	0.000E+00	0.00E+00
234	6.22E-03	1.100E-02	6.84E+01
235	2.16E-06	9.300E-01	2.01E+00
236	6.50E-05	7.500E-03	4.88E–01
238	3.36E-07	6.000E-02	2.02E-02
Totals	Not applicable	1.009E+00	7.09E+01
dpm/µg total U	Not applicable	Not applicable	1.57E+02

RU contaminant	Activity (pCi/µg total U)
Pu-238	2.00E-02
Pu-239	2.00E-02
Np-237	6.00E-02
Tc-99	5.00E-02
Th-228	5.00E-02
Total α	1.50E-01
α dpm/µg total U	3.33E-01
Uα:non-Uα	4.73E+02

		Recycled NU <sup>d</sup>		
Uranium isotope	Activity (Ci/g)	Mass fraction	Activity (pCi/µg total U)	
233	9.66E-03	0.000E+00	0.00E+00	
234	6.22E-03	5.400E-05	3.36E-01	
235	2.16E-06	7.200E-03	1.56E-02	
236	6.50E-05	0.000E+00	0.00E+00	
238	3.36E-07	9.927E-01	3.34E-01	
Totals	Not applicable	9.999E-01	6.85E-01	
dpm/µg total U	Not applicable	Not applicable	1.52E+00	

RU	Activity	
contaminant	(pCi/µg total U)	
Pu-238	0.00E+00	
Pu-239	3.00E-06	
Np-237	2.40E-05	
Tc-99	2.00E-04	
Th-228	3.00E-05	
Total α	5.70E-05	
α dpm/µg total U	1.27E-04	
U α:non-U α	1.20E+04	

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 22 of 81
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	Recycled LEU (2%) <sup>a</sup>			
Uranium	Activity		Activity	
isotope	(Ci/g)	Mass fraction	(pCi/µg total U)	
233	9.66E-03	0.000E+00	0.00E+00	
234	6.22E-03	2.000E-04	1.24E+00	
235	2.16E-06	2.000E-02	4.32E-02	
236	6.50E-05	0.000E+00	0.00E+00	
238	3.36E-07	9.798E-01	3.29E-01	
Totals	Not applicable	1.000E+00	1.62E+00	
dpm/µg total U	Not applicable	Not applicable	3.59E+00	

RU	Activity
contaminant	(pCi/µg total U)
Pu-238	0.00E+00
Pu-239	2.00E-04
Np-237	2.00E-04
Tc-99	9.20E-02
Th-228	3.00E-04
Total α	7.00E-04
α dpm/µg total U	1.55E-03
U α:non-U α	2.31E+03

Recycled DU <sup>a</sup>			
Uranium isotope	Activity (Ci/g)	Mass fraction	Activity (pCi/µg total U)
233	9.66E-03	0.000E+00	0.00E+00
234	6.22E-03	1.000E-05	6.22E-02
235	2.16E-06	2.000E-03	4.32E-03
236	6.50E-05	3.000E-06	1.95E-04
238	3.36E-07	9.980E-01	3.35E-01
Totals	Not applicable	1.000E+00	4.02E-01
dpm/µg total U	Not applicable	Not applicable	8.93E-01

RU contaminant	Activity (pCi/µg total U)
Pu-238	0.00E+00
Pu-239	3.70E-06
Np-237	2.70E-05
Tc-99	4.00E-04
Th-228	4.00E-05
Total α	7.07E-05
α dpm/µg total U	1.57E-04
Uα:non-Uα	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 were 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").</li>

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].

	VHEU & Oralloy	LEU, NU, &	VHEU & Oralloy	LEU, NU, & DU
Isotope	(pCi/µg U)	DU (pCi/µg U)	(pCi/pCi total U) <sup>b</sup>	(pCi/pCi total U) <sup>b</sup>
Tc-99	5.7	0.3	5.7E–2	0.15
Th-228°	0.5	0.003	5E–03	1.5E–03
Np-237	0.6	0.003	6E–03	1.5E–03
Pu-238 <sup>d</sup>	0.2	0	2E–03	0
Pu-239 <sup>d</sup>	0.2 (Oralloy)	0.003	2E–03 (Oralloy)	1.5E–03
Total non-U	1.3	0.01	Not applicable	Not applicable
Total U	75	2	Not applicable	Not applicable

#### Table 5-8. Recommended defaults based on upper levels of expected ranges.<sup>a</sup>

a. For Table 5-8 maximum values, take the highest Table 5-7 value (the average) and multiply by 10 for Pu, Th, and Np and by 3 for Tc. See page 26 of this TBD.

 Reference to Table 5-7 indicates activity in pCi per gram of total U for VHEU ranges from 71 to 101; 100 is used for simplicity. For LEU, NU, and DU, the pCi per gram of total U ranges from 0.9 to 1.6; 2 pCi/g total U is used for simplicity.

c. The Th-228 default is justified in concept due to the demonstrated existence of general plantwide 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].

d. 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 [BWXT Y-12 2000].

From a practical and simplified approach, when the results in the claimant files are given in mass units of uranium per L or lung count, the default levels in pCi 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

[DOCUMENT NO. ORAO 1-1 RD3-00 14-5] REVISION NO. 04 [Ellective Date. 00/05/2025] Page 25 010	Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 23 of 81
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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.

In reference to Table 5-8 with relationship to the plutonium contaminant, the recommended isotope <sup>239</sup>Pu should be assumed for Oralloy only, because Oralloy at Y-12 did not come directly from the primary RU generating sites (i.e., <sup>239</sup>Pu came more predominantly from the gaseous diffusion plants rather than SRS or ICPP by a factor of 1,000 as indicated in Table 5-7) [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 <sup>232</sup>Th operations is not addressed in this section. This section deals only with RU contaminants.

#### 5.3 IN VITRO URINE ANALYSIS

#### 5.3.1 <u>Coverage</u>

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].

Insufficient information exists 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 <sup>226</sup>Ra sealed source, and thorium extraction.

Beginning from 1948, the occupational environmental internal dose from uranium can be calculated using data in ORAUT-TKBS-0014-4, *Y-12 National Security Complex – Occupational Environmental Dose* [ORAUT 2006] for unmonitored workers. Additionally, uranium co-exposure intakes are available starting in 1948 for unmonitored workers as discussed in Attachment B.

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, p. 3]. As of 2003, Y-12 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 [Snapp 2003b, p. 27]. 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.

#### 5.3.2 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. 28] 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 et al. 1965, p. 24]:

$$dpm/d = 8 \times (AP \div Eff) \times (Vol \div Tl)$$
(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 et al. [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*  $\div$  *TI*) also appears in the corresponding equation for fluorometric determinations of NU and DU [McRee et al.1965, p. 26]. 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 (L<sub>D</sub> as defined in Currie [1968]) is discussed below.

Since 1989, routine samples have been collected over a 24-hour period, typically while the worker 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 2003b, p. 74].

#### **Routine Urine Sample Frequency**

Urine samples were collected monthly in 1950, and weekly collection for some workers 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. 30].

As of 2003, samples are scheduled 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 2003b, p. 26].

#### 5.3.3 <u>Minimum Detectable Activities</u>

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

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 (MDA) of 7  $\mu$ g/d for a nominal 1.4 L/d urine excretion rate [Struxness 1949a]. In 1952, the instrument sensitivity and sample volumes indicated an MDA of 7  $\mu$ g/d for 1.4 L/d of urine excreted [Struxness 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% <sup>235</sup>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% <sup>235</sup>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% <sup>235</sup>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. 9] 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. 9]. 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. 42]. Counters were not used if the background was greater than 5 counts in 30 minutes [Hamrick 1958, p. 9]. For two disks counted as described above, assuming a well-known background and alpha of 0.05, the  $L_D$  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 AEC [1959], the  $L_D$  would be 46 dpm/d. With an increase in average uranium recovery to 73% [McRee et al. 1965, p. 32],  $L_D$  would be reduced to 26 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 47 dpm/d through 1964 and 25 dpm/d starting in 1965. However, given the limitations of the rate method of estimating daily urine volumes, uncertainty in the excretion volume is expected to add substantially to the uncertainty associated with the detection limit of a single measurement. Because recoveries were based on batch rather than individual measurements, uncertainties in recovery would add to the uncertainty of the detection limit of a single measurement.

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

The alpha spectrometry system employs large urine aliquots, chemical separation of uranium, sample-specific tracers to determine recovery, and long counting times on low-background detectors. The resultant  $L_D$  is approximately 0.15 dpm/d [Souleyrette et al. 2002, p. 19]. The  $L_D$  varies with the uranium recovery of a particular sample, and is reported with the sample result.

Document No. ORAUT-TKBS-0014-5 Revision N	o. 04 Effective Date: 08/03/2023 F	Page 26 of 81
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#### 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  $\mu$ Ci/L (1.4  $\mu$ Ci/d) [Hursh 1958]. This MDA is far below the plant action level of 0.25  $\mu$ Ci/mL (350  $\mu$ Ci/d) [Patterson et al. 1957, p. 18)]. Results are reported in  $\mu$ Ci per mL.

#### Tritium in Urine by Liquid Scintillation

A few mL of distillate from a urine sample are placed in liquid scintillation fluid for counting [Hursh 1958]. Assuming typical method efficiencies, the MDA is 2,000 dpm/d.

#### Plutonium in Urine by Gross Alpha Counting

The plutonium in urine procedure used before 1988 involved chemical separation from a 24-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<sub>D</sub> for both <sup>238</sup>Pu and <sup>239,240</sup>Pu is 0.025 dpm/sample, or 0.025 dpm/d assuming a full 24-hour void volume [Snapp 2003b, Table 4.2, p. 96]. Actual sample-specific MDAs are included in the analysis reports.

#### **Other Actinides in Urine and Feces**

Table 5-9 lists the analytical laboratory L<sub>D</sub> values used since about 1988.

Table 5-9. L <sub>D</sub> values. <sup>a</sup>				
Radionuclide	L <sub>D</sub> (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 <sup>b</sup>			
a. Source: Snapp	[2003b, p. 111].			

b. Not applicable for fecal.

#### **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. 29]. In 2002, of 2,800 participants in the bioassay program, 700 Y-12 workers participated in the fecal-sampling program [Veinot and Souleyrette 2003, p. 12]. 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 and Souleyrette 2003, p. 20]. The Analytical Chemistry division cites the value of 0.15 dpm/sample as a representative MDA for fecal analysis [Souleyrette et al. 2002, p. 19]. 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. 12].

Document No. ORAUT-TKBS-0014-5 Revision No. 04 Effective Date: 08/03/2023 Page 27 of 81

#### 5.3.4 In Vitro Bioassay Methods for Individual Radionuclides

The following sections discuss the in vitro methods for specific radionuclides in urine. (Section 5.3.5 discusses analysis of fecal samples.)

#### 5.3.4.1 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 minutes per 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 et al. 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. 9; Emerson et al. 1951, p. 16]. By the first half of 1952, a technique involving small (0.2-mL) aliquots of raw urine had been put in place [Struxness 1953, p. 31]. However, after this change in technique was instituted, the urinary uranium concentrations increased greatly. This raises the concern that the technique used before May 1952 could have underestimated the urinary uranium concentrations.

Fluorometric urinalysis continued to be used for NU and DU until 1989, when alpha spectroscopy began to be used. Fluorometry yielded results in mass units (e.g.,  $\mu$ g per L), but results were often converted to disintegrations per minute per day with the assumed specific activity of 1.55 dpm/ $\mu$ g for NU. The fluorometric technique had a sensitivity of about 5 ppb (5  $\mu$ g/L or 7  $\mu$ g/d) (e.g., Struxness [1949a, p. 9]).

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 28 of 81
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#### 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  $\mu$ Ci) corresponding to an ICRP Publication 2 [ICRP 1960] calculated lung dose of 15 rem/yr (using a quality factor of 10). Samples from workers in EU areas were analyzed by electrodeposition of uranium onto silver discs, which were then counted for gross alpha activity in a proportional counter. Development of the gross alpha counting method was reported to be underway in early 1949 [Struxness 1949b, p. 9]. At first, samples were subjected to an acid digestion step before electrodeposition. In 1951, methodological problems leading to underestimates of a factor of 2.5 were noted and corrected [Ballenger 1952, p. 50]. 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 [AEC 1959, p. 66].

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. 42]. 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-cpm/dpm 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 [AEC 1959], but they improved over time. In 1965, the average recovery for electroplating was 73% [McRee et al. 1965, p. 32]. Method limitations are discussed in a number of reports, including Johnson et al. [1959] and UCC [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* [AEC 1959, p. 69].

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, p.4].

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. 9]). The in vivo monitoring frequency was determined by urinalysis results and ranged from monthly to once each 18 months [Scott 1963, p. 4]. 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 <sup>233</sup>U from a nitrate solution (<sup>233</sup>U uranyl nitrate) in Building 9205 [West and Roberts 1962]. In recognition of the high specific activity of this material, and the potential for gamma radiation from the progeny of contaminant <sup>232</sup>U, operations were doubly contained. Pilot runs were made with less hazardous materials to test the protective measures.

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 29 of 81
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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 <sup>233</sup>U to <sup>232</sup>U 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 DCF.

Table 5-10	. <sup>233</sup> U to	<sup>232</sup> U activity	/ ratios.ª
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Process	Activity ratio
Reduction of UF <sub>4</sub>	1.3
Casting of metal-crucible skull	15–32
Machining final uranium component	160
a Source: West and Pabarta [1062]	•

Source: 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 (HNO<sub>3</sub>) and hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>). The sample is wet-ashed, and the uranium is coprecipitated with calcium oxalate (CaC<sub>2</sub>O<sub>2</sub>). 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 (TiCl<sub>3</sub>) 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. Next, the uranium isotopes are coprecipitated on neodymium fluoride (NdF<sub>3</sub>). The NdF<sub>3</sub> is caught on a 0.1-mm filter, rinsed, dried, and mounted on a planchet for alpha spectrometry [Snapp 2003b, p. 68]. Typical recoveries are about 85%. Samples are typically counted for 16 hours on a passivated implanted planar silicon detector.

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. Any result, other than a baseline sample, that is positive is assumed to be due to an occupational exposure for the purposes of dose reconstruction so no correction is necessary.

#### 5.3.4.2 Plutonium

#### Plutonium in Urine by Gross Alpha Counting

Certain workers in the Special Testing Department were tested for plutonium intake by urinalysis. Because large urine volumes were required, these workers submitted 24-hour samples through the use of take-home kits. The sampling frequency was monthly [Patterson et al. 1957, p. 43]. After two 20-mL aliquots were removed for uranium analysis by electrodeposition, the balance of the sample was treated by chemical separation, precipitation, and evaporation onto a stainless-steel planchet for alpha counting. Health physics personnel converted the reported activity rate (disintegrations per minute per 24-hr void) to mrem 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<sub>3</sub> and H<sub>2</sub>O<sub>2</sub>. The sample is wet-ashed, and the uranium, plutonium, and americium are coprecipitated with CaC<sub>2</sub>O<sub>2</sub>. 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<sub>3</sub>. The NdF<sub>3</sub> is caught on a 0.1-mm

Document No. ORAUT-TKBS-0014-5 Revision No. 04 Effective Date: 08/03/2023 Page 30 of 81
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filter, rinsed, dried, and mounted on a planchet for alpha spectrometry [Snapp 2003b, p. 69]. The same procedure is used for isotopic thorium.

#### 5.3.4.3 Tritium

As of 1957, personnel engaged in processing materials with a potential for tritium contamination submitted three urine samples per month. Samples were submitted at the same stations used for uranium and plutonium samples. The samples were reacted to evolve hydrogen gas, which was then counted for beta activity using a vibrating reed electrometer. Results were reported in  $\mu$ Ci per mL [Patterson et al. 1957, pp. 43-44].

Tritium is measured by liquid scintillation counting. An aliquot of the urine sample is distilled in a heating mantle. 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 [Snapp 2003b, p. 71]. Exposure should be assumed to be due to tritiated water (HTO) vapor.

#### 5.3.4.4 Other Radionuclides

Alpha spectrometry procedures described in Section 3.2 of Snapp [2003b] 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.

#### 5.3.5 Fecal Sample Analysis

Fecal samples have long been used in follow-up investigations when urinalysis or in vivo measurements indicate the likelihood of a substantial intake. West and Scott [1966] describe such an investigation in the 1960s in which fecal, urine, and in vivo measurements were used jointly to investigate clearance. Fecal sampling was discontinued several times in the past, but was reinstated in 1998 due to changes in workplace exposure conditions. At first, fecal sampling was used for a limited number of workers in Building 9212 but was expanded to include Building 9215 in 1999 and other areas of the plant in 2000 and 2001 [Souleyrette et al. 2002, p. 10].

If a work area has been determined to have predominantly insoluble airborne uranium, as of 2002, workers with a moderate to high exposure potential (CEDE greater than 100 mrem) were 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 et al. 2002, p. 18] 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 2003b, p. 79]. These normalized results or actual individual daily excretion results can be used to reconstruct dose.

# 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 2003b]. The primary in vivo detection method was chest counting, as described below.

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 31 of 81

### 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 [1959a, 1959b]. A 9- by 4-in. sodium-drifted iodide [Nal(TI)] 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. Nal(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. Nal(TI) crystals were added, positioned at the sides under the arms. In 1985, initial development of a low-energy germanium counting system was begun [Souleyrette 2003, p. 12]. This system began routine operation in a new counting room in June 1992 [NIOSH 2021, p. 11].

#### 5.4.2.1 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 µg of <sup>235</sup>U for EU and mg of <sup>238</sup>U for NU or DU. The original limits of detection for <sup>235</sup>U and <sup>238</sup>U were reported as 130 µg [Cofield 1959b, pp. 8, 36] and 14.4 mg, respectively. 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 activity (nCi 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.

		U-235	U-235	U-235	Reporting	
Year	Detectors	(µg)	(nCi)	(dpm)	convention	Reference
1959	One 9-in. Nal	130	0.28 <sup>b</sup>	620 <sup>b</sup>	Unknown	Cofield 1959b
1963	Two 9-in. Nal	96	0.21 <sup>b</sup>	470 <sup>b</sup>	Unknown	Scott and West 1967
1965	Two 9-in., 2 5-in. Nal	72	0.16 <sup>b</sup>	360 <sup>b</sup>	Unknown	Scott and West 1967
1975	Two 9-in., 2 5-in. Nal	70	0.15 <sup>b</sup>	330 <sup>b</sup>	Unknown	Scott and West 1975
1983	Two 9-in., 2 5-in. Nal	62	0.13 <sup>b</sup>	290 <sup>b</sup>	1.96 σ <sub>вкс</sub>	King and Barclay 1983
1983	Two 9-in., 2 5-in. Nal	68.5	0.15 <sup>b</sup>	330 <sup>b</sup>	Lc	King and Barclay 1983
1983	Two 9-in., 2 5-in. Nal	137	0.30 <sup>b</sup>	670 <sup>b</sup>	LD	King and Barclay 1983
1990	Two 9-in., 2 5-in. Nal	70	0.15 <sup>b</sup>	330 <sup>b</sup>	(d)	Barber and Forrest 1995
06/1992	High-purity	46 <sup>c</sup>	0.10	220°	(e)	Souleyrette et al. 2002;
	germanium					NIOSH 2021

Table 5-11. Reported lung counting detection limits for <sup>235</sup>U.<sup>a</sup>

a.  $L_c$  = critical level;  $L_D$  = detection level as defined by Currie [1968] with  $\alpha = \beta = 0.05$ , paired observations.

b. Value converted from published value in µg.

c. Value converted from published value in nCi.

d. Type I error = 5%.

e. Individual MDA or L<sub>c</sub> supplied with analytical results.

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 <sup>235</sup>U and <sup>238</sup>U lung count detection thresholds are assumed to be 130  $\mu$ g and 14.4 mg, respectively, through 1990. Beginning in 1991, the critical level (L<sub>C</sub>) or MDA is supplied with the individual results. The sample-specific MDA is reported through 1999 and should be used for calculations in the Integrated Modules for Bioassay

Document No. ORAUT-TKBS-0014-5 Revision No.	lo. 04 [Effective Date: 08/03/2023] Page 32 of 81
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Analysis (IMBA). The sample specific  $L_C$  or  $L_D$  is reported beginning in 2000. Multiply this value by 2 for the MDA for IMBA calculations.

Year	Detectors	U-238 (mg)	U-238 (nCi)	U-238 (dpm)	Reporting convention	Reference
1965	Two 9-in., 2 5-in. Nal	6.5	2.2 <sup>b</sup>	4,900 <sup>b</sup>	Unknown	Scott and West 1975
1983	Two 9-in., 2 5-in. Nal	5	1.7 <sup>b</sup>	3,800 <sup>b</sup>	1.96 σ <sub>вкс</sub>	King and Barclay 1983
1983	Two 9-in., 2 5-in. Nal	7.2	2.4 <sup>b</sup>	5,300 <sup>b</sup>	Lc	King and Barclay 1983
1983	Two 9-in., 2 5-in. Nal	14.4	4.8 <sup>b</sup>	11,000 <sup>b</sup>	LD	King and Barclay 1983
06/1992	High-purity germanium	4.5°	1.5	3,300°	(d)	Souleyrette et al. 2002; NIOSH 2021

Table 5-12. Reported lung counting detection limits for <sup>238</sup>U.<sup>a</sup>

a. L<sub>c</sub> = critical level as defined by Currie [1968] with  $\alpha = \beta = 0.05$ , paired observations.

b. Value converted from published value in mg.

c. Value converted from published value in nCi.

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

No description of the conversion count rate to activity to mass has been found. The analyte reported was based on the area in which the person worked. Individuals working in NU or DU areas had results reported as <sup>238</sup>U, and workers in enriched areas had results reported as <sup>235</sup>U. Assumptions that are favorable to claimants should be based on conversions of 93% enrichment for <sup>235</sup>U (Oralloy VHEU) and natural isotopic abundances for <sup>238</sup>U.

#### 5.4.2.2 Other Actinides

#### Thorium

Thorium-232, <sup>228</sup>Th, and <sup>228</sup>Ra exposures can be assessed using <sup>228</sup>Ac and <sup>212</sup>Pb chest counting data which are available starting in August of 1979. Before August of 1979 Y-12 thorium chest count measurements are presented in mass units (total mg Th), and such measurements cannot be used to assess thorium exposure with sufficient accuracy (see Section 5.1.3).

Because IMBA is not designed to be used with multiple-nuclide chains, it cannot be used for an exact assessment when progeny are used to determine the parent intake. However, an approximation can be made that can be used for many cases. ORAUT-OTIB-0076, *Guiding Reconstruction of Intakes of Thorium Resulting from Nuclear Weapons Programs* [ORAUT 2014], derives the basis and provides examples of how to evaluate thorium chest count data using IMBA. Assumptions about the process history of the material are made so the derived intakes are favorable to the claimant. In most cases, the <sup>212</sup>Pb activity is a more reliable indicator of a thorium intake, so it is used for the thorium for dose reconstruction. Triple-separated thorium is assumed because it results in intakes that are favorable to the claimant. For thorium, absorption types M and S should be evaluated, and the type most favorable to claimants should be used to assign dose.

Because IMBA does not correctly calculate <sup>232</sup>Th and <sup>228</sup>Th doses, the following steps apply only to the intake assessment. Alternate methods must be used for the dose assessment. Given a <sup>212</sup>Pb chest count result for a chronic intake longer than 1 year or a chest count more than 30 days after an acute intake:

- 1. Evaluate the chest burden using the <sup>228</sup>Th biokinetic model in IMBA (i.e., use the <sup>212</sup>Pb result to model the <sup>228</sup>Th intake in IMBA). See Table 5-13 for detection limit information.
- 2. Multiply the intake rate obtained in step 1 by a factor of 1.1 and assign it as the intake rate of <sup>228</sup>Th.
- 3. Divide the <sup>228</sup>Th intake rate by 0.19 to obtain the <sup>232</sup>Th intake rate.

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 33 of 81
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If the conditions above (chronic intake longer than 1 year or chest count more than 30 days after an acute intake) are not met and an over- or underestimate cannot be performed, contact the Principal Internal Dosimetry Scientist for a best estimate.

Reported <sup>228</sup>Ac chest count results can be used to calculate a <sup>228</sup>Ra intake. Determine the <sup>228</sup>Ra intake by assuming a <sup>228</sup>Ra chest count equal to the <sup>228</sup>Ac measured value. Assume a type M material because that is the only material type to which radium is assigned.

The calculated <sup>212</sup>Pb and <sup>228</sup>Ac detection limits for 1979 through 1991 are in Table 5-13. The reported <sup>212</sup>Pb and <sup>228</sup>Ac results should be adjusted by adding the adjustment factor according to the values in the table below before comparing it to 2 times the <sup>212</sup>Pb or <sup>228</sup>Ac detection limit (MDA = 2L<sub>D</sub>). The MDA for post-1994 <sup>228</sup>Ac lung counts (if a sample-specific value is not supplied) is 1.0 nCi [Snapp 1995, p. 61].

	Pb-212	Pb-212 DL	Ac-228	Ac-228 DL
Year	adjustment <sup>c</sup>	(nCi)	adjustment <sup>c</sup>	(nCi)
1979	0.115	0.163	-0.012	0.232
1980	0.094	0.116	-0.039	0.098
1981	0.002	0.287	-0.028	0.145
1982	-0.034	0.209	-0.046	0.180
1983	-0.059	0.224	-0.096	0.187
1984	-0.052	0.227	-0.100	0.137
1985	-0.097	0.319	-0.112	0.218
1986	-0.004	0.192	-0.048	0.159
1987	0.017	0.319	-0.057	0.148
1988	0.212	0.223	-0.043	0.247
1989	0.251	0.237	-0.029	0.402
1990	0.266	0.234	-0.019	0.346
1991	0.127	0.294	-0.012	0.115

Table 5-13. Calculated lung counting detection limits for <sup>212</sup>Pb and <sup>228</sup>Ac, 1979 to 1991.<sup>a,b</sup>

a. Source: ORAUT [2022].

b. DL = decision level; called "detection threshold" at Y-12.

c. Pb-adjustment must be added to the reported Pb-212 chest burden before comparing it to 2 times the Pb DL. The same approach must be applied to the reported Ac results and the Ac DL.

For the years 1992 to 1995, the MDA for <sup>212</sup>Pb derived in DCAS-RPRT-008, *Evaluation of the Pb-212 Detection Limit for a Lung Count at the Y-12 Facility from 1992 through 1995* [NIOSH 2020], is presented as a function of the chest wall thickness (CWT) of the individual being counted. This equation, given at the top of page 17 in the report, simplifies to

$$MDA = 0.148 \exp(0.321CWT)$$
 (5-2)

where the MDA is given in units of nCi and the CWT in cm. The plot in Figure 10 of the report is reproduced as Figure 5-1 below using this equation [ORAUT 2022].

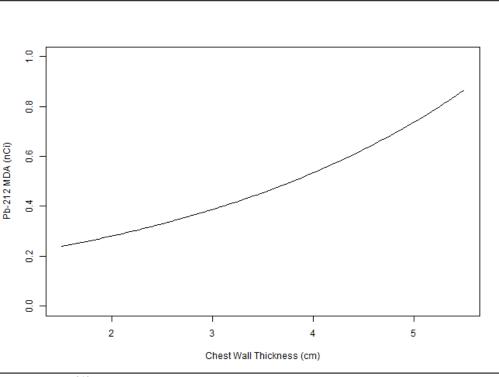


Figure 5-1. <sup>212</sup>Pb MDAs at various CWTs for a 30-minute count [ORAUT 2022].

From January to June 1992, lung counts for Y-12 personnel were performed at the Oak Ridge Gaseous Diffusion Plant (K-25) [NIOSH 2020]. The provided <sup>232</sup>Th sample-specific MDAs should be used to calculate a missed dose.

# Neptunium

At the time the in vivo system was put into routine service in 1961, the reported detection limit for <sup>237</sup>Np without progeny radiation was 2.7 nCi. For <sup>237</sup>Np in full equilibrium with <sup>233</sup>Pa, the detection limit was reported as 0.255 nCi [Cofield 1961, p.12].

# Cesium

At Y-12, <sup>137</sup>Cs results were quantified (along with <sup>40</sup>K) so their effects on the spectra could be taken into account and removed from the occupational monitoring results. Because there is no evidence of <sup>137</sup>Cs in the occupational exposure source term at Y-12, no dose of record is associated with these results.

# 5.4.2.3 Other Radionuclides

At the time the in vivo system was put into routine service in 1961, the reported detection limit for <sup>60</sup>Co was 0.66 nCi. For <sup>95</sup>Zr in transient equilibrium with <sup>95</sup>Nb, the reported detection limit was 1 nCi [Cofield 1961, p. 12]. Although bremsstrahlung counting could have been done for <sup>99</sup>Tc, no information is available about the sensitivity of the technique.

# 5.5 INTERNAL DOSE RECONSTRUCTION GUIDANCE

1. Ensure that internal doses are assigned according to the SEC classes outlined in Section 5.1.3 of this document.

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 35 of 81
--------------------------------	-----------------	----------------------------	---------------

- 2. It is preferred to use individual bioassay results to reconstruct internal dose if available in the claim records. Use the guidance in ORAUT-OTIB-0060 [ORAU 2018] to evaluate the bioassay results.
- 3. The urine and fecal sample-specific detection threshold provided in the Y-12 claim records starting in October 1989 should be used to calculate the MDA when using these results in IMBA. At Y-12, the decision level (DL) was called the "detection threshold." Therefore, the MDA is DL × 2.
- 4. For any in vivo results (specifically, lung counts) before 2000, the L<sub>D</sub> that is reported on the "Bioassay Results History Report" is actually the MDA. Beginning in 2000, the L<sub>D</sub> that is reported for in vivo results is the L<sub>C</sub> of detection [Snapp 2014]. Use the provided sample-specific value as the MDA for results before 2000. Multiply the provided sample-specific value by 2 to obtain the MDA for results starting in 2000, where the MDA is the DL × 2).
- 5. At Y-12, <sup>137</sup>Cs results were quantified (along with <sup>40</sup>K) so their effects on the spectra could be taken into account and removed from the occupational monitoring results. Because there is no evidence of <sup>137</sup>Cs in the occupational exposure source term at Y-12, no dose of record is associated with these results.
- 6. Recycled uranium components should be added to uranium intakes starting in 1953. See Table 5-8 and the end of Section 5.2.4.1 for more information.
  - a. The ratios given for VHEU and Oralloy should be assigned unless there is information that justifies a different enrichment assumption. The values given for pCi/pCi total U in Table 5-8 can be used for the ratios. For a best estimate, divide the technetium by 3 and the plutonium, neptunium, and thorium by 10 after applying the ratios. The assumption of VHEU/Oralloy is the default assumption and can be used for the majority of cases as a claimant-favorable assumption.
  - b. For cases involving intakes other than VHEU/Oralloy, apply the RU components for LEU, NU & DU from Table 5-8. The values given for pCi/pCi total U can be used for the ratios. Divide ratios listed in Table 5-8 by 3 for <sup>99</sup>Tc and by 10 for plutonium, neptunium, and thorium for best-estimate cases.
- 7. For unmonitored workers:
  - a. If co-exposure intakes are to be assigned, refer to the guidance in Section B.6.3 of this document. Co-exposure intakes can be assigned starting in 1948. The co-exposure intakes can be extended beyond the last year of available intakes, if necessary, using the 1988 intakes for subsequent years. The maximum values should be used for most applications. Average values can be used for workers who had a lower potential for exposures to radioactive materials using the guidance in the ORAUT-OTIB-0014 [ORAUT 2004] and based on the records provided for the worker or for best estimates.
  - b. Environmental intakes can be assigned starting in 1948 in accordance with ORAUT-TKBS-0014-4 [ORAUT 2006] and ORAUT-OTIB-0014, *Technical Information Bulletin: Assignment of Environmental Internal Doses for Employees Not Exposed to Airborne Radionuclides in the Workplace* [ORAUT 2004]. The environmental intakes can be extended beyond the last year of available intakes, if necessary, using the 2002 intakes for subsequent years.

- 8. Thorium guidance:
  - a. If the worker has in vitro bioassay results (specifically urine and/or fecal results) for thorium, then it can be inferred that the worker was exposed to thorium. These results would be shown on the "Bioassay Results History Report" [Snapp 2014]. Although some bioassays are labeled as dietary, treat all results greater than the MDA as indicating potential occupational intakes and evaluate the bioassay results using the guidance in ORAUT-OTIB-0060 [ORAUT 2018].
  - b. Before August 1979, available thorium-monitoring data are presented in mass units of total thorium. As discussed in the SEC-00251 evaluation report [NIOSH 2018], NIOSH does not have the necessary information to use these data for reconstructing internal dose due to <sup>228</sup>Th, <sup>232</sup>Th, and <sup>228</sup>Ra. For this reason, these data cannot be used for internal dose reconstruction for either thorium or thorium progeny.
  - c. For August 1979 to December 31, 1991, <sup>232</sup>Th, <sup>228</sup>Th, and <sup>228</sup>Ra exposures can be assessed using <sup>228</sup>Ac and <sup>212</sup>Pb chest counting data in accordance with Section 5.4.2.2 to evaluate individual bioassay results. The <sup>228</sup>Ac and <sup>212</sup>Pb results are not always reported from the site. Therefore, if the records include a thorium lung count reported in mass units after July 1979, or if they include data from "Y-12 Lung Counts 1958-1991" [Y-12 lung, no date a] via a "Personnel Exposure" file in the claim files, it is easier to search the "Y-12 Lung Counts Spreadsheet" [Y-12 lung, no date b] by the worker's Social Security Number to determine if <sup>228</sup>Ac and <sup>212</sup>Pb results are available.
  - d. If the ANALYSIS TYPE code associated with a lung count result is listed as either 3, 6 or 7, then it can be inferred that the worker was exposed to thorium [Snapp 2022, p. 4].
  - e. For 1992 to 1995, use the equation in Section 5.4.2.2 to calculate the <sup>212</sup>Pb MDA for the missed dose calculation. Additionally, for the period from January to June 1992, use the sample-specific <sup>232</sup>Th MDAs to calculate a missed dose.
  - f. Beginning in 1992 and continuing through 2004, lung count results showing thorium (typically represented by <sup>228</sup>Ac on the "Bioassay Results History Report") should only be considered as screening results for thorium and are not necessarily indicative of potential for exposure to thorium. These screenings were performed on all workers who were lung counted regardless of work with thorium [Snapp 2014].
  - g. Additional information about work locations involving Y-12 Plant thorium operations and thorium work history are in ORAUT-TKBS-0014-2, Y-12 National Security Complex – Site Description [ORAUT 2007b].

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Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 43 of 81

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Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 44 of 81

# GLOSSARY

#### absorption type

Categories for materials according to their rates of absorption from the respiratory tract to the blood, which replaced the earlier inhalation clearance classes. Defined by the International Commission on Radiological Protection, the absorption types are F: deposited materials that are readily absorbed into blood from the respiratory tract (fast solubilization), M: deposited materials that have intermediate rates of absorption into blood from the respiratory tract (moderate rate of solubilization), and S: deposited materials that are relatively insoluble in the respiratory tract (slow solubilization). 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. X-rays produced in a typical medical X-ray tube frequently originate from inelastic scattering of accelerated electrons in the anode material.

#### burnup

Depletion of fissionable material in nuclear fuel.

#### calutron

Accelerator that separates isotopes (e.g., <sup>235</sup>U from <sup>238</sup>U) according to their masses using strong magnetic fields. The name derives from <u>Cal</u>ifornia <u>U</u>niversity cyclo<u>tron</u>.

#### 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<sub>2</sub>) 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 undesired locations including air, soil, buildings, animals, and persons.

#### cyclotron

Particle accelerator capable of large beam currents where the beam is injected into the center of a circular magnet. A fixed radiofrequency 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 <sup>235</sup>U 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% <sup>234</sup>U, 0.199% <sup>235</sup>U, and 99.8% <sup>238</sup>U.

#### 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 rad, rep, or grays.

# dose conversion factor (DCF)

(1) Multiplier for conversion of potential dose to the personal dose equivalent to the organ of interest (e.g., liver or colon). (2) In relation to radiography, ratio of dose equivalent in tissue or organ to entrance kerma in air at the surface of the person being radiographed.

#### enriched uranium (EU)

Uranium in which processing has increased the proportion of <sup>235</sup>U to <sup>238</sup>U to above the natural level of 0.7%. Reactor-grade uranium is usually enriched to approximately 3.5% <sup>235</sup>U;

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

#### 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 <sup>235</sup>U in relation to <sup>238</sup>U. Along with the enriched uranium, this process results in uranium depleted in <sup>235</sup>U. 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 (UF<sub>6</sub>) gas is filtered to enrich the  $^{235}$ U and separate it from  $^{238}$ U. The process requires enormous amounts of electric power and results in an increase in  $^{235}$ U 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% <sup>235</sup>U for use as fissile material in nuclear weapons components and some reactor fuels. Also called high-enriched uranium. SRS lists the isotopic activity fractions as:

Isotope	Activity fraction
<sup>234</sup> Ú	0.9806
<sup>235</sup> U	0.0194
<sup>238</sup> U	0.0000

The specific activity of highly enriched uranium with these fractions is 6,207 picocuries per microgram.

#### 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 to the organ of interest from sources inside the body (e.g., plutonium or uranium) that are inhaled, ingested, absorbed, or injected through a wound).

Document No. ORAUT-TKBS-0014-5	Revision No. 04	Effective Date: 08/03/2023	Page 47 of 81

#### 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 using instrumentation that detects radiation emitted from the radioactive material in the body.

#### low-enriched uranium (LEU)

Uranium enriched to less than 20% <sup>235</sup>U by mass.

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

The smallest amount of an analyte in a sample that can be detected with a probability  $\beta$  of nondetection while accepting a probability  $\alpha$  of erroneously deciding that a positive (nonzero) quantity of analyte is present in an appropriate blank sample.

#### 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 (NU)

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<sup>7</sup> 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., <sup>236</sup>U) after purification. SRS lists the isotopic activity fractions as:

<u>Isotope</u>	Activity fraction
<sup>234</sup> U	0.8489
<sup>235</sup> U	0.0120
<sup>236</sup> U	0.1388
<sup>238</sup> U	0.0003

The specific activity of recycled uranium with these fractions is 5,296 picocuries per microgram.

#### routine monitoring

Monitoring carried out at regular intervals during normal operations.

# 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 <sup>238</sup>U 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 <sup>234</sup>U. 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 <sup>235</sup>U.

# weighting factor

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

# ATTACHMENT A OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS

# **TABLE OF CONTENTS**

# **SECTION**

# TITLE

PAGE

**PAGE** 

A.1	Conventions Used in Internal Dose Reports A.1.1 Y-12 Urinalysis Data, 1943 to September 1989 A.1.2 Y-12 Urinalysis Data, October 1989 to Present A.1.3 In Vivo Data	50 50
A.2	Uranium Solubility in the Lung	53
A.3	In Vitro Detection Limits	53
A.4	In Vivo Detection Limits	54

# LIST OF TABLES

<u>TITLE</u>

## <u>TABLE</u>

A-1	Uranium urinalysis detection limits	. 53
	Other in vitro detection limits	
A-3	Reported in vivo detection limits for other radionuclides	. 54

# ATTACHMENT A OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS (continued)

# A.1 CONVENTIONS USED IN INTERNAL DOSE REPORTS

The following is adapted from Souleyrette [2003].

# A.1.1 Y-12 Urinalysis Data, 1943 to September 1989

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

Badge/Old Social Security Number - Worker 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)

<u>Volume</u> – volume of sample in mL.

<u>Time</u> – 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.)

<u>Uranium Result</u> – The normalized urine result in disintegrations per minute per day.

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

# A.1.2 Y-12 Urinalysis Data, October 1989 to Present

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

Badge – Worker identification number.

Sample Number – Laboratory sample identifier.

Sample Date – Date sample was voided.

# Sample Type

A = alpha spectroscopy 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.)

# ATTACHMENT A

# OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS (continued)

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).

<u>Retention Class</u> – 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).

<u>Total Uranium Result</u> (dpm) – This is the normalized total uranium urine result in disintegrations per minute per day.

Type Assimilation

- H = inhalation
- I = ingestion
- J = injection

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

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

# A.1.3 In Vivo Data

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

Run Date – Date subject counted.

Run No. – Laboratory identifier of counting session.

Weight – Subject weight in pounds.

Chest Thickness – Subject CWT 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.

# ATTACHMENT A

# OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS (continued)

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

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

For counts after 1971, where Type Analysis is 0:

00 = Background run

01 = Sensitivity check

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

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

For Type Analysis of 3 (any year):

- 00 = Thorium (first examination)
- 02 = Thorium (second or later examination)

<u>U-235</u> ( $\mu$ g) – <sup>235</sup>U activity detected in chest cavity. If the count is invalid, an "&" appears as the first character.

<u>Thorium</u> (mg)  $-^{232}$ Th activity calculated from the progeny isotopes detected in chest cavity.

<u>U-238</u> (mg) - <sup>234</sup>Th activity detected in chest cavity.

<u>Tc-99</u> ( $\mu$ Ci) – <sup>99</sup>Tc activity detected in chest cavity.

# ATTACHMENT A

# OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS (continued)

<u>Current Dept</u> – Additional instance of worker 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 absorption 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. 10]. 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, Section 6]. Based on this study, both depleted and EU 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 varving 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 need 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 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, p. 29]. 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  $L_Ds$  and do not include uncertainties introduced by sample collection or conversion from submitted volumes to daily void volumes. As noted in Section 5.3,  $L_D$  values for some historical techniques remain to be identified and will be reported in later revisions of this TBD as they become available.

Method	Period	Detection limit (mass)	Detection limit (activity) (dpm/d)	Convention
Fluorometry	1950-09/1989	7 μg/dª	11	Not applicable
Gross alpha	1950–1964	Not applicable	46 <sup>b</sup>	LD
Gross alpha	1965-09/1989	Not applicable	25°	LD
Alpha spectrometry	10/1989-present	Not applicable	0.15 <sup>d</sup>	LD

Table A-1. Uranium urinalysis detection limits.

a. Based on 5 ppb sensitivity determination and a nominal 1.4 L/d excretion rate [Struxness 1949a, p. 9].

b. Estimated based on assuming a well-known background and alpha of 0.05, the LD 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 AEC [1959], the LD would be 46 dpm/d.

c. Estimated based on assuming a well-known background and alpha of 0.05, the LD is 0.13 cpm. For 1,400 mL/d urine output, a nominal 0.5 cpm/dpm counting efficiency and an increase in average uranium recovery to 73% [McRee et al. 1965, p. 32] the LD would be 25 dpm/d.

d. Souleyrette et al. [2002, p. 19].

# ATTACHMENT A OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS (continued)

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

Table A-2. Other in vitro detection limits.

a. Estimate assuming typical method efficiencies.

b. Source: Snapp [2003b, p. 111, Table 5.3].

# 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 <sup>137</sup>Cs, <sup>99</sup>Tc, thorium, plutonium, <sup>228</sup>Ac, and tritium, among other radionuclides; there are difficulties in interpreting some of the measurement data that cannot be resolved satisfactorily at this time. At Y-12, <sup>137</sup>Cs results were quantified (along with <sup>40</sup>K) so their effects on the spectra could be taken into account and removed from the occupational monitoring results. Because there is no evidence of <sup>137</sup>Cs in the occupational exposure source term at Y-12, no dose of record is associated with these results.

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.

Radionuclide and solubility type	Reported limit (nCi)	Reported limit (dpm)
Ac-228	1.0 <sup>a</sup>	2,220
Np-237/M	2.7 <sup>b</sup>	5,900
Np-237/M; Pa-233/M, S	0.255 <sup>b</sup>	560
Co-60/M, S	0.66 <sup>b</sup>	1,500
Zr-95/M, S; Nb-95/M, S	1.0 <sup>b</sup>	2,200

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

a. Source: Snapp [1995, p. 61], post-1994 lung counts if sample specific value is not available. For lung counts from 1979 to 1991, see Table 5-13.

b. Source: Cofield [1961, p. 12].

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.

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 µg and 14.4 mg, respectively, through 1990. Beginning in 1991, the L<sub>C</sub> or MDA is supplied with the individual results.

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

# ATTACHMENT A OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS (continued)

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 Y-12 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 <sup>228</sup>Ac 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.).

# ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA

## **TABLE OF CONTENTS**

#### **SECTION** TITLE PAGE **B**.1 Overview 58 **B**.2 **B.3 B.4** B.4.1 **B**.5 Uranium Intake Rates by Material Type ......64 B.6 B.6.3 Dose Assignment ......67

# LIST OF TABLES

# TABLE

# TITLE

B-2 Type F uranium intake periods and rates	
D-2 I YPE I UTATIIUTTI ITTARE PETIOUS ATU TALES	
B-3 Type M uranium intake periods and rates	
B-4 Type S uranium intake periods and rates	
B-5 Type F uranium intake periods and rates	66
B-6 Type M uranium intake periods and rates	67
B-7 Type S uranium intake periods and rates	67

#### LIST OF FIGURES

#### FIGURE

#### TITLE

B-1	Assumed uranium intake, 01/01/1947 to 12/31/1969, 50th-percentile results, type F 68
B-2	Assumed uranium intake, 01/01/1970 to 12/31/1988, 50th-percentile results, type F69
B-3	Assumed uranium intake, 01/01/1947 to 02/28/1978, 50th-percentile results, type M70
B-4	Assumed uranium intake, 01/01/1947 to 04/30/1952, 50th-percentile results, type M70
B-5	Assumed uranium intake, 08/01/1953 to 11/30/1953, 50th-percentile results, type M71
B-6	Assumed uranium intake, 11/01/1956 to 02/28/1959, 50th-percentile results, type M71

# PAGE

PAGE

B-7	Assumed uranium intake, 06/01/1961 to 12/31/1962, 50th-percentile results, type M	72
B-8	Assumed uranium intake, 10/01/1968 to 12/31/1972, 50th-percentile results, type M	'2
B-9	Assumed uranium intake, 03/01/1978 to 09/30/1984, 50th-percentile results, type M	73
B-10	Assumed uranium intake, 10/01/1984 to 12/31/1988, 50th-percentile results, type M	′3
B-11	Predicted uranium urinary excretion from eight independent intakes, 50th percentile,	
	1952 to 1988	74
B-12	Predicted uranium urinary excretion from eight independent intakes, 84th percentile,	
	1952 to 1988	75
B-13	Assumed uranium intake, 01/01/1947 to 02/28/1978, 50th-percentile results, type S	'6
B-14	Assumed uranium intake, 01/01/1947 to 04/30/1952, 50th-percentile results, type S	'6
B-15	Assumed uranium intake, 08/01/1953 to 11/30/1953, 50th-percentile results, type S	77
B-16	Assumed uranium intake, 11/01/1956 to 02/28/1959, 50th-percentile results, type S	77
B-17	Assumed uranium intake, 06/01/1961 to 12/31/1962, 50th-percentile results, type S7	'8
B-18	Assumed uranium intake, 10/01/1968 to 12/31/1972, 50th-percentile results, type S7	'8
B-19	Assumed uranium intake, 03/01/1978 to 09/30/1984, 50th-percentile results, type S	'9
B-20	Assumed uranium intake, 10/01/1984 to 12/31/1988, 50th-percentile results, type S7	'9
B-21	Predicted uranium urinary excretion from eight independent intakes, 50th percentile,	
	1952 to 1988	30
B-22	Predicted uranium urinary excretion from eight independent intakes, 84th percentile,	
	1952 to 1988	31

# B.1 PURPOSE

There are instances of workers 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 co-exposure data from the Y-12 Plant.

# B.2 OVERVIEW

ORAUT-OTIB-0019, *Analysis of Coworker Bioassay Data for Internal Dose Assignment* [ORAUT 2005a] describes the general process for analysis of bioassay data for assignment of doses to individuals based on co-exposure 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 2005a]. The resultant values were input to the 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 [ORAUT 2005b, 2022]. 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 [2005a]. 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 might underestimate the urinary uranium concentrations as discussed in Section 5.3.2. January through April 1952 were included in the modeling because the 50th- and 84th-percentile values. Table B-1 lists the statistical analysis results.

<b>T</b>     <b>D A NA</b>			40501 4000
I able B-1. Monthi	y uranium 24-hour urina	<i>Ty excretion rate analy</i>	ses, 1952 to 1988.

			unnary excreti					
Effective	50th	84th	Normalian of		Effective	50th	84th	Number of
sample	percentile	percentile	Number of		sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples	-	date	(dpm/d)	(dpm/d)	samples
01/15/1952	21.16	64	696	-	02/14/1956	16.03	48.01	1,853
02/14/1952	24.92	84.51	741	-	03/15/1956	14.02	42.59	1,902
03/15/1952	23.86	88.16	918	_	04/15/1956	16.45	46.84	1,886
04/15/1952	25.49	74.15	665		05/15/1956	15.73	47.57	1,980
05/15/1952	21.15	51.31	1,103	_	06/15/1956	13.79	40.67	1,986
06/15/1952	18.06	47.31	1,401		07/15/1956	13.82	41.39	2,068
07/15/1952	14.13	41.9	1,236		08/15/1956	13.39	41.44	843
08/15/1952	11.25	33.06	1,258		09/14/1956	14.11	38.28	1,637
09/14/1952	13.7	35.97	1,590		10/14/1956	11.1	32.33	2,022
10/14/1952	16.71	46.49	1,305		11/14/1956	4.72	15.14	1,835
11/14/1952	16.68	46.41	1,133		12/14/1956	7.3	26.34	1,889
12/14/1952	14.29	37.68	1,368		01/14/1957	9.42	28.04	2,413
01/14/1953	12.61	37.15	1,123		02/13/1957	10.2	38.81	1,967
02/13/1953	10.51	32.28	1,065	-	03/16/1957	11.04	40.78	1,951
03/16/1953	9.18	26.4	1,380	Ī	04/15/1957	10.79	33.8	2,302
04/15/1953	10.34	32.25	1,095	ľ	05/16/1957	10.48	42.11	1,975
05/16/1953	10.43	33.25	1,079	ľ	06/15/1957	9.44	34.06	2,099
06/15/1953	9.81	34.9	1,337		07/15/1957	8.46	28.63	2,471
07/15/1953	11.13	40.08	830	-	08/15/1957	10.56	35.75	2,038
08/15/1953	9.81	33.3	1,261	-	09/14/1957	7.51	24.91	2,552
09/14/1953	19.53	63.39	1,099	-	10/15/1957	12.06	53.49	2,278
10/15/1953	32.43	110.13	677	-	11/14/1957	13.03	44.83	2,387
11/14/1953	26.65	88.11	1,752	-	12/15/1957	11.29	37.96	2,878
12/15/1953	18.47	57.43	1,614	-	01/14/1958	8.36	30.87	2,708
01/14/1954	19.45	84.37	1,655	-	02/13/1958	13.64	53.23	2,208
02/13/1954	16.06	74.2	1,938	Ī	03/16/1958	16.24	59.24	2,219
03/16/1954	19.5	74.74	2,233		04/15/1958	13.3	52.29	2,498
04/15/1954	22.04	78.72	1,688	-	05/16/1958	12.71	56.35	2,396
05/16/1954	22.85	77.01	1,774		06/15/1958	13.7	77.06	2,665
06/15/1954	22.07	73.11	1,801	Ī	07/16/1958	17.44	79.66	2,332
07/16/1954	22.06	83.01	1,303	Ī	08/15/1958	19.45	65.95	2,206
08/15/1954	15.39	63.49	2,571	-	09/15/1958	19.19	86.52	2,669
09/15/1954	8.47	28.08	1,064	Ī	10/15/1958	15.21	58.52	2,115
10/15/1954	11.58	50.14	1,726	Ī	11/14/1958	21.01	73.93	2,258
11/14/1954	22.02	91.14	118		12/15/1958	22.76	80.61	3,071
12/15/1954	14.85	45.87	783		01/14/1959	20.74	77.72	2,949
01/14/1955	7.36	25.15	591	-	02/14/1959	24.9	90.03	2,798
02/14/1955	5.62	19.81	639	-	03/16/1959	18.71	60.78	2,424
03/16/1955	13.71	39.83	909	ŀ	04/16/1959	19.72	66.84	3,126
04/16/1955	16.2	50.07	1,840	ľ	05/16/1959	17.44	56.57	2,725
05/16/1955	13.27	37.94	2,006	ŀ	06/15/1959	12.41	45.55	2,878
06/15/1955	15.71	47.63	1,595	ŀ	07/16/1959	16.69	57.44	2,520
07/16/1955	13.1	36.57	1,573	ľ	08/15/1959	12.79	40.15	3,001
08/15/1955	13.85	42.78	1,874	ľ	09/15/1959	13.66	43.44	2,580
09/15/1955	17.1	55.19	1,592	ŀ	10/15/1959	11.53	40.87	2,983
10/15/1955	16.87	51.49	1,990	ŀ	11/15/1959	14.86	53.07	3,317
11/15/1955	12.99	39.56	1,725	ŀ	12/15/1959	16.04	56.6	2,785
12/15/1955	15.34	45.01	1,732	ŀ	01/15/1960	13.87	43.69	3,395
01/15/1956	14.5	47.71	2,121	ŀ	02/14/1960	13.36	46	3,666
51/10/1000	11.0		<u> </u>	L	52/11/1000	10.00	10	0,000

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
03/15/1960	9.31	34.17	2,662
04/15/1960	9.75	39.57	3,375
05/15/1960	11.19	38.09	3,796
06/15/1960	9.88	37.26	2,468
07/15/1960	10.51	37.64	2,330
08/15/1960	12.76	51.09	3,115
09/14/1960	9.45	32.2	2,392
10/14/1960	8.93	32.44	3,155
11/14/1960	9.85	35.5	2,486
12/14/1960	7.6	27.11	2,443
01/14/1961	7.53	32.63	2,571
02/13/1961	6.99	27.55	2,409
03/16/1961	9.36	37.96	2,397
04/15/1961	8.94	32.38	754
05/16/1961	7.46	26.83	1,556
06/15/1961	9.17	32.46	532
07/15/1961	11.04	38.61	1,035
08/15/1961	5.8	23.46	1,035
09/14/1961	12.96	54.55	621
10/15/1961	8.55	29.21	1,216
11/14/1961	7.46	28.68	1,210
12/15/1961	16.66		555
01/14/1962	11.93	64.1	
	11.95	48.51	1,303
02/13/1962		55.59	1,191
03/16/1962	22.09	81.26	830
04/15/1962	16.35	63.01	1,431
05/16/1962	10.69	39.93	1,462
06/15/1962	19.41	71.41	1,045
07/16/1962	13.57	52.64	1,528
08/15/1962	12.66	58.77	1,420
09/15/1962	19.24	72.07	1,049
10/15/1962	15.98	53.81	1,524
11/14/1962	16.91	65.36	1,470
12/15/1962	22.87	97.5	1,134
01/14/1963	16.8	64.65	1,222
02/14/1963	14.35	65.28	1,307
03/16/1963	17.58	70.17	1,203
04/16/1963	13.52	46.46	1,481
05/16/1963	12.56	44	1,661
06/15/1963	14.88	56	1,038
07/16/1963	13.4	36.41	1,321
08/15/1963	13.29	43.99	902
09/15/1963	13.48	44.18	1,247
10/15/1963	11.65	38.88	576
11/15/1963	9.5	26.83	1,002
12/15/1963	7.73	26.04	1,562
01/15/1964	8	28.09	1,390
02/14/1964	7.96	24.14	1,579
03/15/1964	10.27	32.95	1,172
04/15/1964	4.6	15.35	902

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
05/15/1964	3.58	10.53	1,203
06/15/1964	5.7	21.01	599
07/15/1964	5.66	18.62	903
08/15/1964	4.65	14.2	1,088
09/14/1964	4.62	12.61	477
10/14/1964	3.43	9.95	667
11/14/1964	3.26	10.78	1,112
12/14/1964	7.72	21.81	291
01/14/1965	4.7	14.91	545
02/13/1965	3.99	12.84	646
03/16/1965	6.81	23.26	404
04/15/1965	4.59	21.09	312
05/16/1965	6.7	23.6	571
06/15/1965	9.45	31.48	329
07/15/1965	5.64	18.05	364
08/15/1965	6.41	23.41	609
09/14/1965	2.74	11.57	376
10/15/1965	4.91	18.07	416
11/14/1965	3.44	12.05	571
12/15/1965	9.85	37.35	397
01/14/1966	4.44	19.14	469
02/13/1966	4.41	24.7	668
03/16/1966	10.17	42.75	391
04/15/1966	10.17	48.01	441
05/16/1966	5.04	16.99	739
06/15/1966	6.32	26.44	415
07/16/1966	8.7	39.61	359
08/15/1966	5.01	18.31	624
09/15/1966	8.86	40.76	431
		71.36	
10/15/1966	15.88		392
11/14/1966	8.68	32.4	539
12/15/1966	8.98	40.16	567
01/14/1967	6.79	33.34	339
02/14/1967	5.8	25.69	710
03/16/1967	8.35	49.25	426
04/16/1967	10.81	56.82	364
05/16/1967	3.25	18.88	593
06/15/1967	6.51	25.46	224
07/16/1967	5.49	35.82	390
08/15/1967	7.3	40.93	585
09/15/1967	4.42	19.34	401
10/15/1967	5.21	20.92	412
11/15/1967	5.01	20.71	514
12/15/1967	6.06	33.53	405
01/15/1968	6.5	28.54	393
02/14/1968	10.15	41.09	573
03/15/1968	8.1	39.56	476
04/15/1968	7.07	43.78	411
05/15/1968	11.07	53.86	458
06/15/1968	7.05	29.75	574

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
07/15/1968	5.2	23.25	395
08/15/1968	5.86	23.48	533
09/14/1968	5.48	26.46	421
10/14/1968	3.86	16.16	391
11/14/1968	5.9	23.37	516
12/14/1968	9.71	32.61	533
01/14/1969	8.43	41.12	220
02/13/1969	5.02	20.69	731
03/16/1969	5.65	27.79	655
04/15/1969	7.61	36.08	477
05/16/1969	5.22	24.92	593
06/15/1969	8.26	30.14	546
07/15/1969	7.37	29.64	375
08/15/1969	6.62	28.37	630
09/14/1969	4.7	17.88	475
10/15/1969	6.6	25.36	514
11/14/1969	7.06	22.98	541
12/15/1969	7.00	30.81	401
01/14/1970	9.84	40.81	336
02/13/1970	9.04 8.33	28.27	559
03/16/1970	6.33 7.85		
	6.99	29.62 27.21	316
04/15/1970			378
05/16/1970	4.93	18.99	103
06/15/1970	11.17	44.73	137
07/16/1970	8.88	38.05	399
08/15/1970	8.76	42.72	401
09/15/1970	11.59	55.4	236
10/15/1970	9.81	34.23	500
11/14/1970	13.1	51.16	460
12/15/1970	17.14	67.22	275
01/14/1971	4.41	18.55	12
02/14/1971	11.3	35.27	523
03/16/1971	12.76	50.18	334
04/16/1971	9.52	39.99	458
05/16/1971	5.87	21.42	557
06/15/1971	10.37	41.16	263
07/16/1971	4.67	19.49	396
08/15/1971	10.74	47.49	557
09/15/1971	13.95	62.06	275
10/15/1971	8.25	32.76	463
11/15/1971	8.24	29.09	504
12/15/1971	11.67	35.91	283
01/15/1972	11.54	44.77	289
02/14/1972	12.2	43.92	227
03/15/1972	15.64	56.16	253
04/15/1972	12.09	47.06	246
05/15/1972	12.64	50.99	296
06/15/1972	13.68	34.9	155
07/15/1972	18.95	70.38	257
08/15/1972	20.33	67.87	314

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
09/14/1972	18.14	54.91	234
10/14/1972	15.48	64.45	225
11/14/1972	12	42.03	369
12/14/1972	20.42	61.5	164
01/14/1973	15.63	41.34	347
02/13/1973	15.79	57.05	271
03/16/1973	15.71	49.06	325
04/15/1973	16.2	48.06	268
05/16/1973	14.46	47.02	366
06/15/1973	15.04	56.06	248
07/15/1973	14.38	43.07	326
08/15/1973	16.5	52.5	296
09/14/1973	12.97	56.63	509
10/15/1973	12.31	40.96	485
11/14/1973	10.62	29.86	486
12/15/1973	14.2	46.52	393
01/14/1974	12.16	38.96	418
02/13/1974	11.16	34.34	378
03/16/1974	20.33	56.38	287
04/15/1974	12.99	40.27	325
05/16/1974	9.14	22.89	405
06/15/1974	9.75	29.71	222
07/16/1974	7.54	20.57	270
08/15/1974	8.44	24.96	330
09/15/1974	8.77	24.28	264
10/15/1974	14.19	137.02	298
11/14/1974	9.16	53.77	368
12/15/1974	8.11	35.07	250
01/14/1975	8.93	37.19	252
02/14/1975	11.49	44.05	360
03/16/1975	16.92	57.92	291
04/16/1975	10.37	46.11	319
05/16/1975	11.74	39.05	297
06/15/1975	10.66	33.4	186
07/16/1975	11.04	38.53	188
08/15/1975	11.07	32.85	338
09/15/1975	8.9	31.58	171
10/15/1975	11.92	36.58	186
11/15/1975	9.18	32.82	298
12/15/1975	12.88	40.99	148
01/15/1976	9.97	30.51	196
02/14/1976	10.81	32.89	321
03/15/1976	9.19	28.35	225
04/15/1976	8.65	27.03	211
05/15/1976	8.3	24.08	347
06/15/1976	8.24	23.91	185
07/15/1976	3.82	12.47	194
08/15/1976	6.51	18.64	332
09/14/1976	5.39	14.99	193
10/14/1976	9.5	26.06	228

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
11/14/1976	13.24	40.32	297
12/14/1976	13.72	43.42	202
01/14/1977	13.97	44.66	169
02/13/1977	13.41	41.7	268
03/16/1977	12.91	40.62	245
04/15/1977	8.86	24.33	191
05/16/1977	8.39	22.81	302
06/15/1977	9.62	31.33	180
07/15/1977	7.46	28.02	190
08/15/1977	9.29	25.59	342
09/14/1977	11.22	54.94	207
10/15/1977	10.86	37.43	244
11/14/1977	10.75	47.78	341
12/15/1977	9.31	35.7	151
01/14/1978	9.28	35.13	184
02/13/1978	9.82	29.72	293
03/16/1978	4.34	13.13	233
04/15/1978	8.07	26.41	196
05/16/1978	5.53	18.15	291
06/15/1978	4.73	14.79	164
07/16/1978	6.58	20.43	185
08/15/1978	5.1	20.43	362
09/15/1978	7.02	24.90	198
10/15/1978	7.02	22.71	205
11/14/1978		22.05	
12/15/1978	6.81 4.93	15.71	390 155
01/14/1979	5.48	22.99	180
02/14/1979	5.73	24.87	398
03/16/1979	7.5	55.07	219
04/16/1979	4.12	20.11	213
05/16/1979	5.45	26.64	357
06/15/1979	6.54	24.51	198
07/16/1979	6.15	26.52	205
08/15/1979	2.81	8.47	407
09/15/1979	7.52	23.8	180
10/15/1979	6.69	21.68	205
11/15/1979	5.68	16.34	360
12/15/1979	10.15	28.45	219
01/15/1980	5.43	19.51	238
02/14/1980	5.39	18.48	462
03/15/1980	9.96	31.5	206
04/15/1980	7.03	28.92	273
05/15/1980	6.08	23.84	473
06/15/1980	7.52	24.86	215
07/15/1980	4.43	13.44	231
08/15/1980	4.33	14.91	488
09/14/1980	5.59	16.59	237
10/14/1980	8.8	27.7	253
11/14/1980	3.7	14.66	468
12/14/1980	6.65	26.31	255

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
01/14/1981	6.78	27.85	305
02/13/1981	5.54	25.15	462
03/16/1981	6.2	20.98	325
04/15/1981	4.81	19.11	269
05/16/1981	3.66	15.89	535
06/15/1981	5.97	21.83	251
07/15/1981	6.47	31.68	26
08/15/1981	4.89	16.79	575
09/14/1981	6.13	22.23	315
10/15/1981	5.14	21.72	314
11/14/1981	4.35	18.28	605
12/15/1981	5.73	29.53	294
01/14/1982	4.53	18.34	351
02/13/1982	4.13	15.44	540
03/16/1982	5.01	18.71	482
04/15/1982	5.72	32.11	323
05/16/1982	6.89	24.15	584
06/15/1982	5.47	20.5	366
07/16/1982	4.66	17.11	320
08/15/1982	6.48	31.75	581
09/15/1982	2.92	10.86	351
10/15/1982	6.66	25.77	332
11/14/1982	3.24	12.97	637
12/15/1982	2.94	13.34	308
01/14/1983	3.67	15.42	348
02/14/1983	5.93	23.87	634
03/16/1983	5.51	26.4	316
04/16/1983	4.51	17.99	296
05/16/1983	4.83	19.89	633
06/15/1983	2.99	12.46	288
07/16/1983	2.74	9.92	271
08/15/1983	5.17	19.39	690
10/15/1983	3.36	16.08	279
11/15/1983	2.69	14.1	706
12/15/1983	2.08	8.98	325
01/15/1984	2.72	11	326
02/14/1984	2.59	14.01	653
03/15/1984	4.2	20.09	329
04/15/1984	4.14	21.85	360
05/15/1984	3.51	15.94	637
06/15/1984	3.42	15.07	335
07/15/1984	3.49	15.65	352
08/15/1984	2.96	17.55	697
09/14/1984	3.97	20.81	356
10/14/1984	6.23	26.17	367
11/14/1984	2.68	8.13	644
12/14/1984	4.62	20.08	348
01/14/1985	2.93	12	397
02/13/1985	1.93	8.23	605
03/16/1985	4.45	31.7	425

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
04/15/1985	1.67	8.6	391
05/16/1985	1.66	8.97	625
06/14/1985	0.76	5.03	318
07/15/1985	1.91	8.85	383
08/14/1985	1.97	7.66	569
09/14/1985	2.76	11.77	369
10/14/1985	1.82	6.05	419
11/13/1985	1.53	6.04	544
12/14/1985	2.71	11.36	379
01/13/1986	2.79	11.56	472
02/13/1986	1.1	5.61	593
03/14/1986	2.41	13.98	388
04/15/1986	1.35	5.24	497
05/15/1986	1.12	5.13	555
06/15/1986	1.63	7.52	401
07/15/1986	2.42	11.67	408
08/14/1986	2.54	10.13	572
09/14/1986	1.79	8.35	352
10/14/1986	0.95	4.22	550
11/14/1986	2.6	11.31	626
12/14/1986	1.17	6.57	323
01/14/1987	0.8	8.03	437

Effective	50th	84th	
sample	percentile	percentile	Number of
date	(dpm/d)	(dpm/d)	samples
02/13/1987	9.62	57.02	555
03/15/1987	5.04	18.9	345
04/15/1987	2.5	13.62	404
05/15/1987	2.43	11.68	525
06/15/1987	2.2	19.91	328
07/15/1987	8.16	46.96	34
08/15/1987	13.59	70.44	24
09/14/1987	4.01	19.3	54
10/15/1987	0.44	2.13	474
11/14/1987	1.78	14.61	494
12/14/1987	2.13	11.13	327
01/14/1988	2.19	14.84	338
02/13/1988	2.37	10.64	550
03/15/1988	4.62	24.43	371
04/14/1988	4.03	26.36	345
05/15/1988	1.52	7.09	606
06/14/1988	2.05	13.32	363
07/14/1988	4.03	19.34	416
08/14/1988	2.96	17.83	563
09/14/1988	2.38	14.04	432
10/14/1988	0.75	3.7	434
11/13/1988	2.18	10.51	103
12/14/1988	3.69	13.94	152

# 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 m<sup>3</sup>/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, <sup>234</sup>U 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 DCFs) for <sup>234</sup>U that are 7% to 31% larger than those for <sup>235</sup>U, <sup>236</sup>U, and <sup>238</sup>U [ICRP 1995b]. Because of the isotopic compositions of the source terms, use of the <sup>234</sup>U DCF 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 from those in later years, so this period was not included in the modeling. The ORAUT-TKBS-0014-2, *Y-12 National* 

*Security Complex – Site Description* 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 [ORAUT 2007b]. 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 [ORAUT 2022]. 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 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.

# B.5.1 <u>Type F</u>

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.

ATTACHMENT B	
INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)	ļ

Start	Stop	50th percentile	84th percentile	GSD
01/01/1947	05/31/1952	85.35	264.1	3.09
06/01/1952	08/31/1953	43.89	131.3	2.99
09/01/1953	10/31/1953	104.3	339.5	3.26
11/01/1953	12/31/1954	66.43	254.1	3.83
01/01/1955	12/31/1956	47.03	145.2	3.09
01/01/1957	12/31/1957	36.07	145.6	4.04
01/01/1958	12/31/1959	59.48	207.8	3.49
01/01/1960	12/31/1961	34.68	134	3.86
01/01/1962	12/31/1963	53.27	171.9	3.23
01/01/1964	12/31/1969	22.97	97.28	4.24
01/01/1970	12/31/1971	35.8	115.6	3.23
01/01/1972	12/31/1975	47.51	168.4	3.54
01/01/1976	12/31/1977	35.09	121.7	3.47
01/01/1978	12/31/1983	19.68	85.61	4.35
01/01/1984	12/31/1988	9.098	51.26	5.63
07/01/1987	08/31/1987	43.498	102.61	2.36

Table B-2. Type F uranium intake periods and rates (dpm/d).

# 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 GSDs.

Start	Stop	50th percentile	84th percentile	GSD
01/01/1947	02/28/1978	169.34	598.93	3.54
01/01/1947	04/30/1952	354.59	1,154.9	3.26
08/01/1953	11/30/1953	547.22	1,963.1	3.59
11/01/1956	02/28/1959	226.90	825.31	3.64
06/01/1961	12/31/1962	248.68	866.81	3.49
10/01/1968	12/31/1972	160.67	601.60	3.74
03/01/1978	09/30/1984	80.03	355.93	4.45
10/01/1984	12/31/1988	44.203	223.85	5.06

Table B-3. Type M uranium intake periods and rates (dpm/d).

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 <u>Type S</u>

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.

Start	Stop	50th percentile	84th percentile	GSD
01/01/1947	02/28/1978	1,844.4	6,544.2	3.55
01/01/1947	04/30/1952	5,210.4	16,970	3.26
08/01/1953	11/30/1953	17,983	64,559	3.59
11/01/1956	02/28/1959	5,694.6	20,560	3.61
06/01/1961	12/31/1962	6,849.4	23,632	3.45
10/01/1968	12/31/1972	3,290.3	12,415	3.77
03/01/1978	09/30/1984	1,280.3	5,802.9	4.53
10/01/1984	12/31/1988	884.85	4,340.6	4.91

Table B-4. Type S uranium intake periods and rates (dpm/d).

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 Tables 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.

Start	Stop	50th percentile	GSD	95th percentile
01/01/1947	05/31/1952	85.4	3.54	683
06/01/1952	08/31/1953	43.9	3.54	351
09/01/1953	10/31/1953	104	3.54	834
11/01/1953	12/31/1954	66.4	4.24	715
01/01/1955	12/31/1956	47.0	3.54	376
01/01/1957	12/31/1957	36.1	4.24	388
01/01/1958	12/31/1959	59.5	3.54	476
01/01/1960	12/31/1961	34.7	4.24	373
01/01/1962	12/31/1963	53.3	3.54	426
01/01/1964	12/31/1969	23.0	4.24	247
01/01/1970	12/31/1971	35.8	3.54	286
01/01/1972	12/31/1975	47.5	3.54	380
01/01/1976	12/31/1977	35.1	3.54	281
01/01/1978	12/31/1983	19.7	4.35	221
01/01/1984	06/30/1987	9.10	5.63	156
07/01/1987	08/31/1987	43.5	3.00	265
09/01/1987	12/31/1988	9.10	3.54	156

Table B-5. Type F uranium intake periods and rates (dpm/d).

Table B-6. Type M uranium intake periods and rates (dpm/d).				
Start	Stop	50th percentile	GSD	95th percentile
01/01/1947	04/30/1952	524	3.74	4,588
05/01/1952	07/31/1953	169	3.74	1,483
08/01/1953	11/30/1953	717	3.74	6,275
12/01/1953	10/31/1956	169	3.74	1,483
11/01/1956	02/28/1959	396	3.74	3,470
03/01/1959	05/31/1961	169	3.74	1,483
06/01/1961	12/31/1962	418	3.74	3,661
01/01/1963	09/30/1968	169	3.74	1,483
10/01/1968	12/31/1972	330	3.74	2,890
01/01/1973	02/28/1978	169	3.74	1,483
03/01/1978	09/30/1984	80.0	4.45	933
10/01/1984	12/31/1988	44.2	5.06	636

Table B-6. Type M uranium intake periods and rates (dpm/d).

Table B-7. Type S uranium intake periods and rates (dpm/d).

Start	Stop	50th percentile	GSD	95th percentile
01/01/1947	04/30/1952	7,054	3.77	62,595
05/01/1952	07/31/1953	1,844	3.77	16,366
08/01/1953	11/30/1953	19,827	3.77	175,929
12/01/1953	10/31/1956	1,844	3.77	16,366
11/01/1956	02/28/1959	7,539	3.77	66,895
03/01/1959	05/31/1961	1,844	3.77	16,366
06/01/1961	12/31/1962	8,693	3.77	77,138
01/01/1963	09/30/1968	1,844	3.77	16,366
10/01/1968	12/31/1972	5,134	3.77	45,559
01/01/1973	02/28/1978	1,844	3.77	16,366
03/01/1978	09/30/1984	1,280	4.53	15,367
10/01/1984	12/31/1988	885	4.91	12,125

# 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 RU, the dose from the added constituents, including plutonium, <sup>237</sup>Np, and <sup>99</sup>Tc, 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

Doses can be calculated using these intakes starting in 1948. The co-exposure intakes can be extended beyond the last year of available intakes, if necessary, using the intakes for 1988 for subsequent years. 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 on 50th-percentile intake rates are assigned as lognormal distribution with the applicable GSD. There are situations when the 95th percentile of the co-exposure 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.

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

The lognormal distribution is selected in 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.

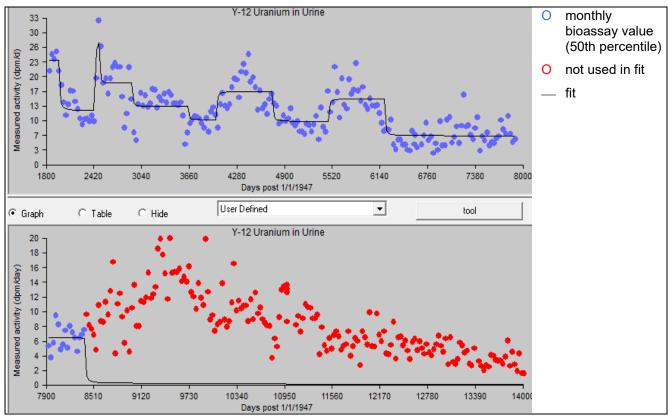
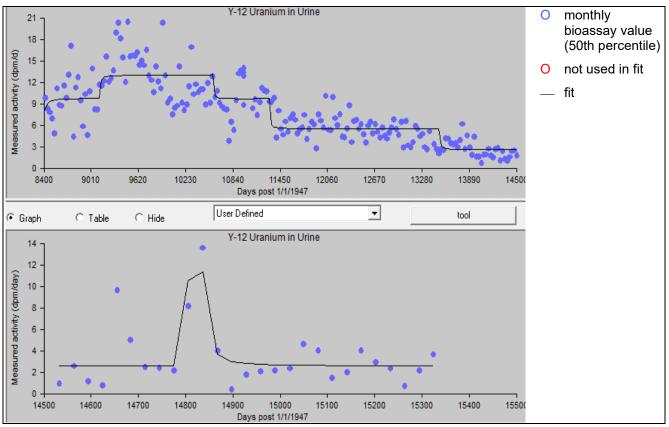


Figure B-1. Assumed uranium intake, 01/01/1947 to 12/31/1969, 50th-percentile results, type F.



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

Figure B-2. Assumed uranium intake, 01/01/1970 to 12/31/1988, 50th-percentile results, type F.

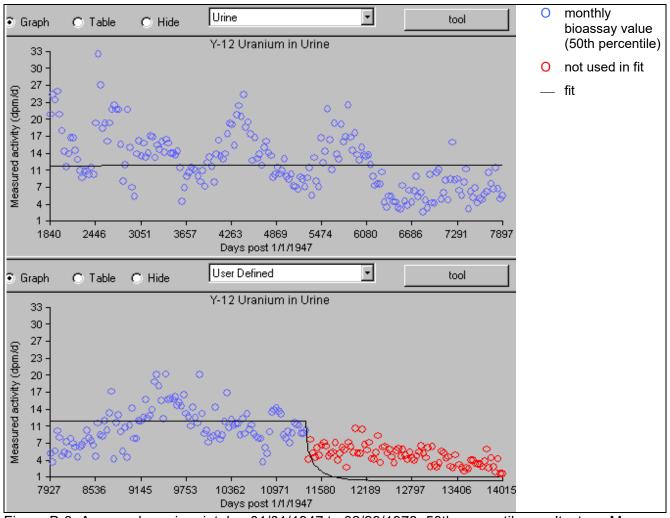




Figure B-3. Assumed uranium intake, 01/01/1947 to 02/28/1978, 50th-percentile results, type M.

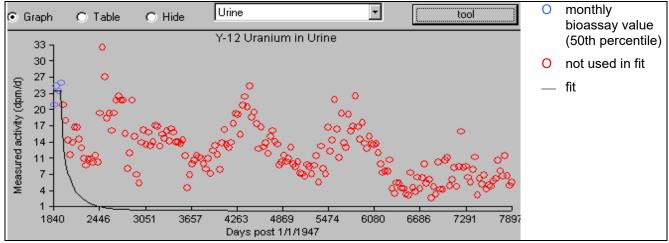


Figure B-4. Assumed uranium intake, 01/01/1947 to 04/30/1952, 50th-percentile results, type M.



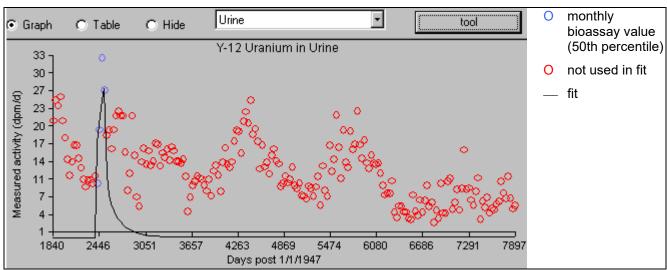


Figure B-5. Assumed uranium intake, 08/01/1953 to 11/30/1953, 50th-percentile results, type M.

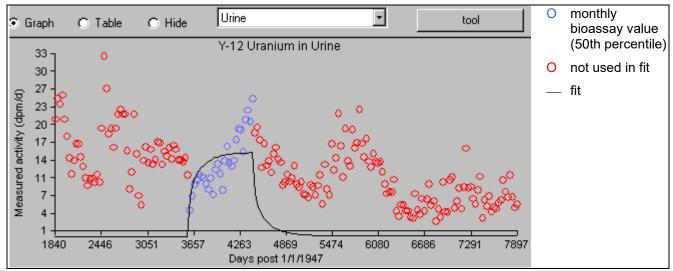


Figure B-6. Assumed uranium intake, 11/01/1956 to 02/28/1959, 50th-percentile results, type M.



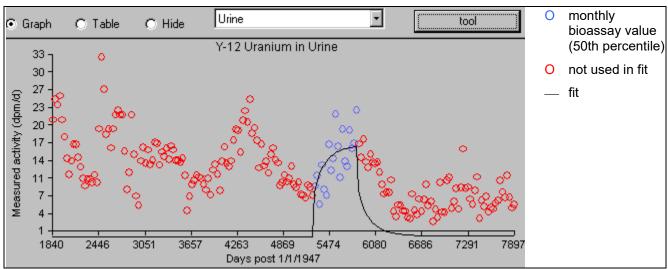


Figure B-7. Assumed uranium intake, 06/01/1961 to 12/31/1962, 50th-percentile results, type M.

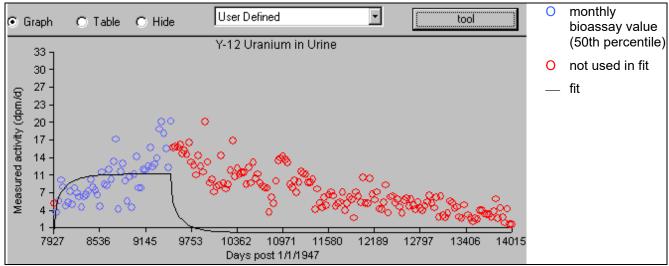
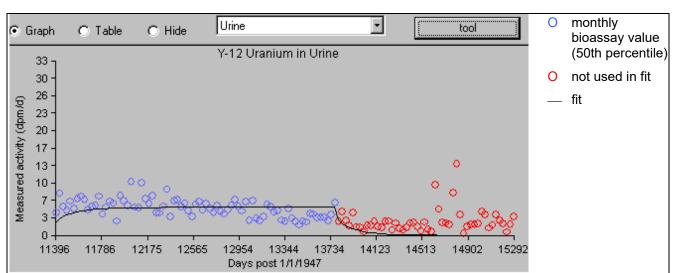


Figure B-8. Assumed uranium intake, 10/01/1968 to 12/31/1972, 50th-percentile results, type M.



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

Figure B-9. Assumed uranium intake, 03/01/1978 to 09/30/1984, 50th-percentile results, type M.

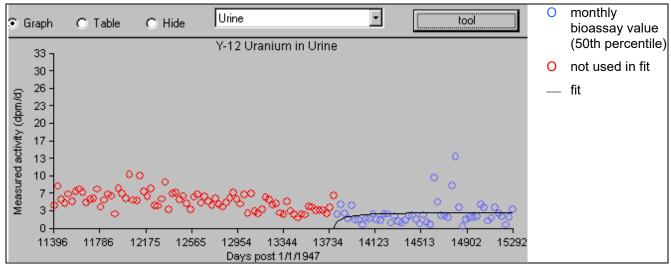
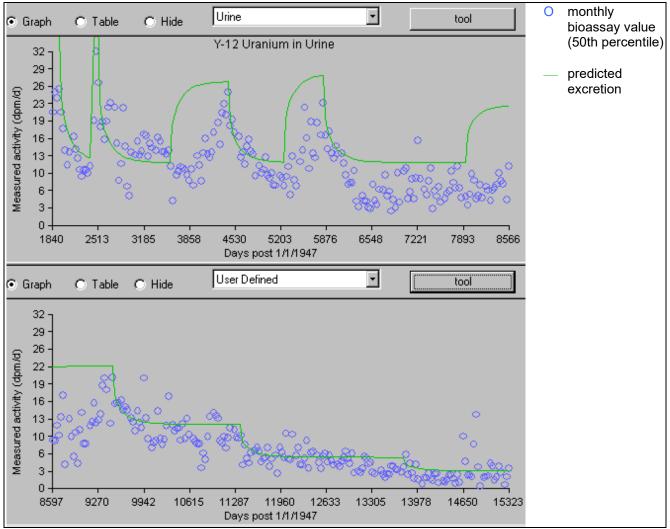
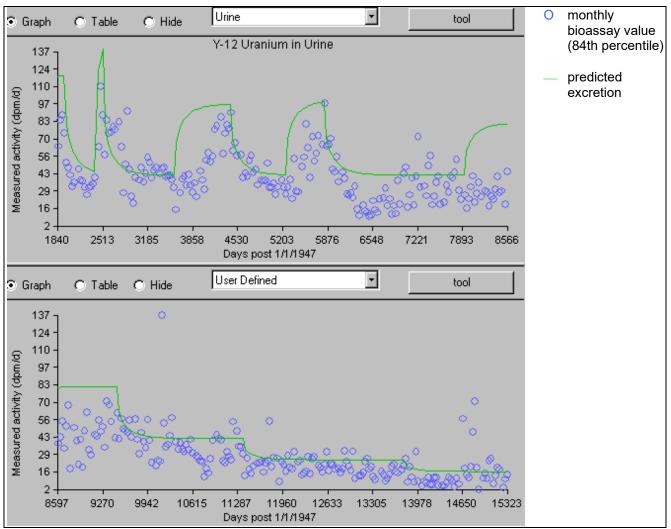


Figure B-10. Assumed uranium intake, 10/01/1984 to 12/31/1988, 50th-percentile results, type M.



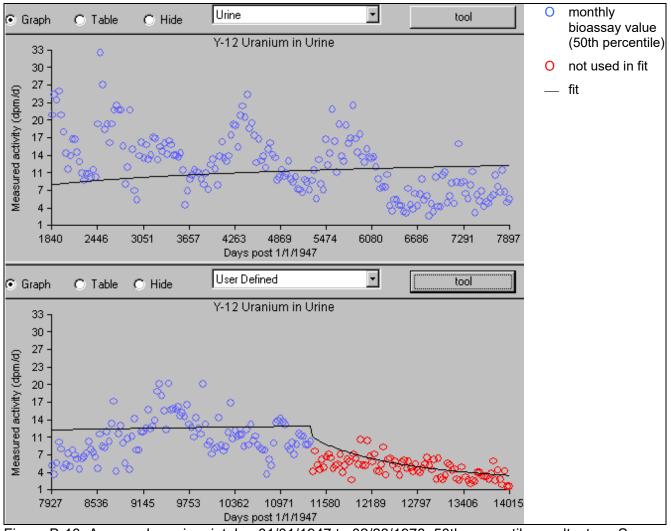
ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

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



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

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



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

Figure B-13. Assumed uranium intake, 01/01/1947 to 02/28/1978, 50th-percentile results, type S.

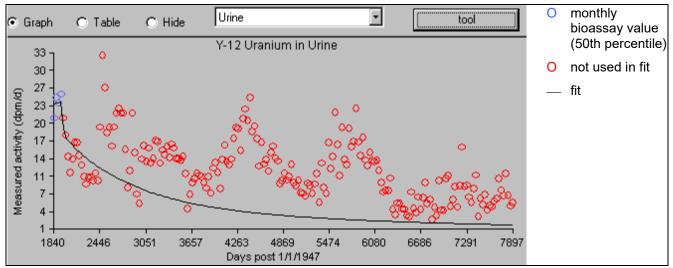


Figure B-14. Assumed uranium intake, 01/01/1947 to 04/30/1952, 50th-percentile results, type S.



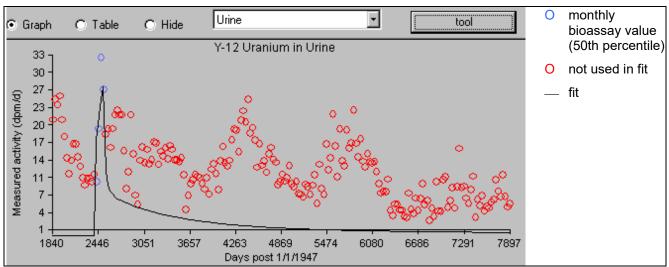


Figure B-15. Assumed uranium intake, 08/01/1953 to 11/30/1953, 50th-percentile results, type S.

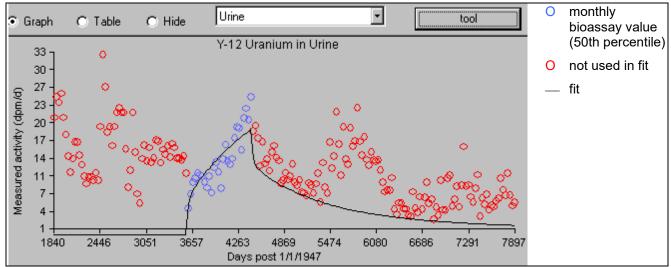


Figure B-16. Assumed uranium intake, 11/01/1956 to 02/28/1959, 50th-percentile results, type S.



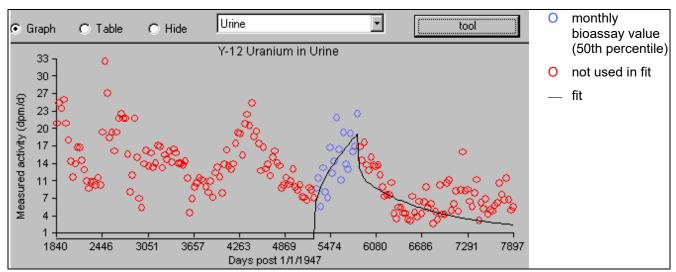


Figure B-17. Assumed uranium intake, 06/01/1961 to 12/31/1962, 50th-percentile results, type S.

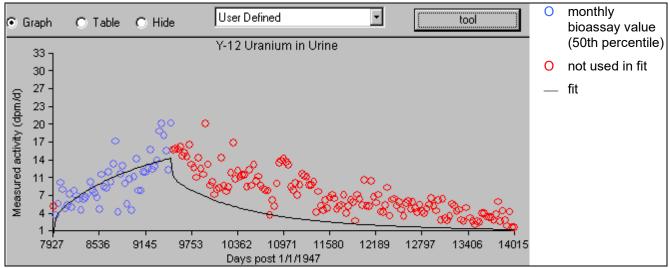
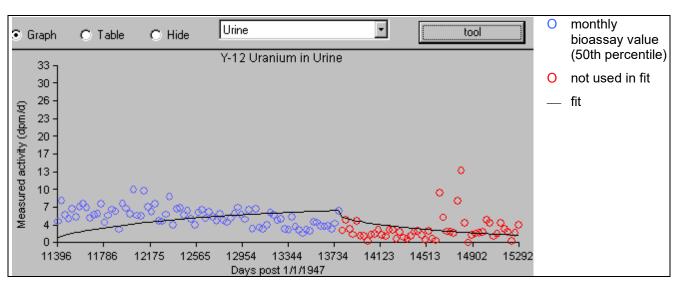


Figure B-18. Assumed uranium intake, 10/01/1968 to 12/31/1972, 50th-percentile results, type S.



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

Figure B-19. Assumed uranium intake, 03/01/1978 to 09/30/1984, 50th-percentile results, type S.

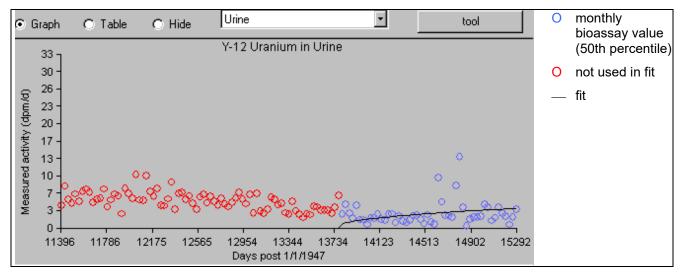
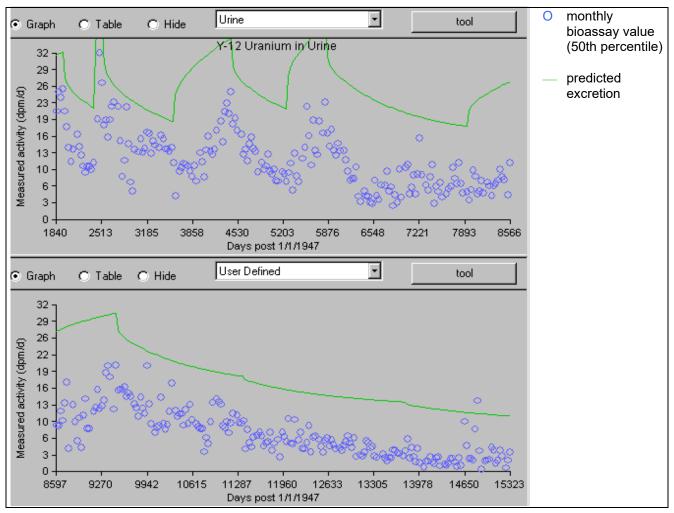
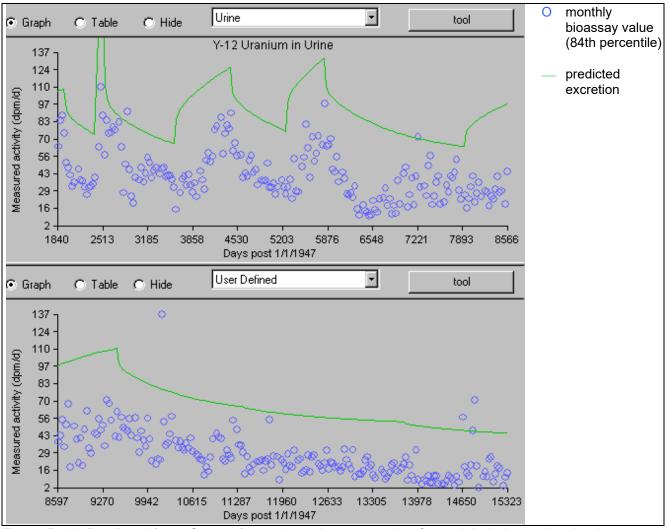


Figure B-20. Assumed uranium intake, 10/01/1984 to 12/31/1988, 50th-percentile results, type S.



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

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



ATTACHMENT B INTERNAL DOSIMETRY CO-EXPOSURE DATA (continued)

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