



# ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities | Dade Moeller | MJW Technical Services

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### PUBLICATION RECORD

| EFFECTIVE DATE | REVISION NUMBER | DESCRIPTION   |
|----------------|-----------------|---|
| 10/07/2005     | 00              | New technical basis document for the Lawrence Livermore National Laboratory – Occupational Internal Dose. First approved issue. Training required: As determined by the Task Manager. Initiated by Jay J. Maisler.  |
| 03/16/2010     | 01              | Revision to incorporate Attributions and Annotations section and Advisory Board comments associated with the issuance of a LLNL SEC Class. Incorporates the internal coworker study (ORAUT-OTIB-0065) into the TBD as Attachment B. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Mutty M. Sharfi, Stephen Spanos and Ralph W. Kenning.  |
| 12/13/2010     | 02              | Revision to update Table B-9 uranium 95 <sup>th</sup> percentile coworker intakes for those time periods that were based on the 50 <sup>th</sup> percentile intakes with a GSD that were rounded up to 3. The previous revision used the actual GSDs in Table B-8 that were less than 3 to determine the 95 <sup>th</sup> percentile coworker intakes for those time periods. Added information pertaining to solubility types for aged related fission products for P-32 and C-14 in Section 5.3.2.3 and added associated footnotes to Tables B-25 and B-26. Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Stephen Spanos and Ralph Kenning. |

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**ACRONYMS AND ABBREVIATIONS**

|         |  |
|---------|--|
| AMAD    | activity median aerodynamic diameter                                   |
| AVLIS   | Atomic Vapor Laser Isotope Separation                                  |
| CFR     | Code of Federal Regulations  |
| Ci      | curie  |
| cm      | centimeter   |
| cpm     | counts per minute  |
| CWT     | chest wall thickness   |
| d       | day  |
| DOE     | U.S. Department of Energy  |
| DOL     | U.S. Department of Labor   |
| dpm     | disintegrations per minute   |
| DU      | depleted uranium   |
| D-38    | depleted uranium   |
| EEOICPA | Energy Employees Occupational Illness Compensation Program Act of 2000 |
| F       | fast (solubility rate)   |
| g       | gram   |
| HEPA    | high-efficiency particulate air  |
| HEU     | highly enriched uranium  |
| HPGe    | hyperpure germanium  |
| hr      | hour   |
| HT      | elemental tritium or tritiated gas                                     |
| HTO     | tritiated water vapor  |
| ICP-MS  | inductively coupled plasma–mass spectrometry                           |
| in.     | inch   |
| keV     | kiloelectron-volt, 1,000 electron-volts                                |
| kg      | kilogram   |
| KPA     | kinetic phosphorescence analysis                                       |
| L       | liter  |
| LANL    | Los Alamos National Laboratory   |
| LEGe    | low energy Germanium detector  |
| LEU     | low enriched uranium   |
| LLNL    | Lawrence Livermore National Laboratory                                 |
| LPTR    | Livermore Pool-Type Reactor  |
| LSC     | liquid scintillation counting  |
| M       | moderate (solubility rate)   |
| M&E     | Mechanical and Electrical (Engineering Divisions)                      |
| MDA     | minimum detectable amount  |
| MEQ     | muscle equivalent  |
| MeV     | megaelectron-volt, 1 million electron-volts                            |
| MFP     | mixed fission product  |
| mg      | milligram  |

|             |  |
|-------------|--|
| mm          | millimeter   |
| MT          | metal tritide  |
| Nal         | sodium iodide  |
| nCi         | nanocurie  |
| NESHAP      | National Emission Standard for Hazardous Air Pollutants  |
| NIOSH       | National Institute for Occupational Safety and Health    |
| NTS         | Nevada Test Site   |
| OBT         | organically bound tritium                                |
| ORAU        | Oak Ridge Associated Universities                        |
| pCi         | picocurie  |
| POC         | probability of causation                                 |
| R&D         | research and development                                 |
| S           | slow (solubility rate)                                   |
| SEC         | Special Exposure Cohort                                  |
| SRDB Ref ID | Site Research Database Reference Identification (number) |
| TBD         | technical basis document                                 |
| TRU         | transuranic  |
| U.S.C.      | United States Code                                       |
| WB          | whole-body   |
| WBC         | whole-body counter                                       |
| yr          | year   |
| §           | section or sections                                      |
| μCi         | microcurie   |
| μg          | microgram  |
| μm          | micrometer   |

## 5.1 INTRODUCTION

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

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

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled “Exposure in the Performance of Duty.” That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer “shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation<sup>1</sup>] guidelines established under subsection (c) ...” [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) define “performance of duty” for DOE employees with a covered cancer or restrict the “duty” to nuclear weapons work (NIOSH 2010).

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

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

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

### 5.1.1 Purpose

This TBD documents historical practices at the Lawrence Livermore National Laboratory (LLNL) and provides information for the evaluation of internal and external dosimetry data for unmonitored and monitored workers; it can serve as a supplement to, or substitute for, individual monitoring data.

This document provides a uniform and consistent approach to assessing occupational internal dose at LLNL for dose reconstructions for NIOSH in relation to the EEOICPA. It provides guidance to dose reconstructors on input parameters that are specific to LLNL employees, as well as the approach for employees with either missing or no monitoring information.

This TBD includes guidance on selecting source terms, interpreting *in vivo* and *in vitro* measurement results, and instructions for assessing dose for monitored and unmonitored employees.

### 5.1.2 Scope

Occupational internal dose refers to exposures received by workers inside the facilities at LLNL from ingestion, inhalation, or absorption of radioactive materials in the course of work. This section provides site-specific information to the dose reconstructor for use in reconstructing radiation doses for workers that were employed at LLNL, via multiple pathways of exposure including inhalation, ingestion, and absorption. In some cases, exposures occurred via wounds and were not restricted to intakes inside buildings. The dose reconstructor calculates intakes from information in the files, using information from this section to interpret data.

Two classes of LLNL employees have been added to the Special Exposure Cohort (SEC) as described in the section below. The addition of these classes of employees has limited the scope of potential internal dose reconstructions at LLNL to the following.

- Internal dose from tritium from 1957 to the present;
- Internal dose from mixed fission products (MFPs) from 1974 to the present;
- Internal dose from americium from 1957 to the present;
- Internal dose from plutonium from 1957 to the present;
- Internal dose from uranium from 1958 to the present;
- Internal dose from other radionuclides present at LLNL may be reconstructed from 1957/1958 to the present. The dose reconstructor should refer to the respective section on when the internal dose for that particular radionuclide can be reconstructed.

The internal dose from americium, plutonium, and uranium can be extended for employment prior to the years listed above based on a worker's later years bioassay results and work history.

The remainder of this section describes the SEC, certain historical events that are important to internal dose reconstruction, and the overall approach to internal dose reconstruction for LLNL workers. Section 5.2 discusses source term information. Section 5.3 discusses *in vitro* and *in vivo* bioassay measurement methods. Section 5.4 describes significant incidents with internal dose potential. Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information are presented in Section 5.5.

Attachment A provides materials for reconstruction of occupational internal dose for monitored workers. Attachment B provides coworker intakes. Attachment C describes scaling of coworker dose intakes consistent with type and duration of exposures.

### **5.1.3 Special Exposure Cohort Petition Information for LLNL**

#### **Classes Added to the SEC**

NIOSH has determined, with concurrence from the Secretary of Health and Human Services (HHS), that MFP doses at LLNL cannot be reconstructed with sufficient accuracy between 1950 and 1973, inclusive. For this reason, the following class of LLNL employees has been added to the SEC (Leavitt 2008).

Employees of the DOE, its predecessor agencies, and DOE contractors or subcontractors who were monitored for radiation exposure while working at the Lawrence Livermore National Laboratory from January 1, 1950, through December 31, 1973, for a number of workdays aggregating at least 250 workdays or in combination with workdays within the parameters established for one or more other classes of employees in the Special Exposure Cohort.

NIOSH has subsequently determined that MFP doses for workers not monitored for radiation exposure at LLNL also cannot be reconstructed with sufficient accuracy between 1950 and 1973, inclusive. For this reason, the following class of LLNL employees has been added to the SEC (Sebelius 2010).

All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Lawrence Livermore National Laboratory in Livermore, California from January 1, 1950 through December 31, 1973, 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.

The SEC class includes all workers during the SEC period. Because of the identified dose reconstruction infeasibility, all dose reconstructions for all workers having employment during the SEC period are considered partial dose reconstructions. If monitoring data are available for workers included in the SEC, dose is to be assigned as appropriate based on such data; however, such dose reconstructions are still considered partial dose reconstructions because the HHS has determined that exposure to mixed fission and activation products during the SEC period cannot be bounded.

### **5.2 SOURCE TERM**

LLNL workers handled a variety of radionuclides as part of their routine work. The key elements in the source term were plutonium and tritium, although others were used at various times and in various forms. Gloveboxes, extensive filtered ventilation systems, and other engineering controls were used to minimize personnel exposures from the outset of operations at LLNL (Balanda 1966). Administrative controls were implemented to minimize the potential for an intake from handling radioactive materials. A majority of the buildings at LLNL handled or processed unsealed sources of radioactive materials; persons who were assigned to operate processes or laboratory research had a potential for exposure (ORAUT 2005a; Mansfield 1989). Administrative personnel and others assigned to support the individual buildings had a lesser potential for exposure given the presence of engineering and administrative controls. For the purposes of dose reconstruction, it can be assumed that internal source terms were introduced at LLNL's opening on September 2, 1952.

Before the early 1960s, the only bioassay methodology that LLNL used to monitor employees for intakes of radionuclides was urine bioassay, with the primary focus on excreted tritium. A review of previous bioassay samples indicates that bioassay monitoring started in March 1957. The analysis of bioassay samples continues to this day for plutonium, americium, uranium, MFPs, a variety of tracer radionuclides, iodine, and tritium (Mansfield 2000). Air monitoring in workplaces has been a common surveillance method. A brief description of the building process was provided in ORAUT (2005b, Table 2-2). The buildings that were assigned to handle plutonium are highlighted. Buildings in which uranium was processed and handled are also described. ORAUT (2005b, Table A-1) provides a cross reference of main site building number changes that took place in 1966. The dose reconstructor can refer to this list if needed when provided building location information for a worker's claim.

*In vivo* methodologies began on an investigational basis in 1964, focusing on high-energy (i.e., greater-than-200-keV) gamma emitters. In the 1970s, there were attempts to detect low-energy photon emitters (i.e., 60-keV gamma radiation from the decay of <sup>241</sup>Am and even plutonium L-shell X-rays). LLNL can provide a broad spectrum of *in vivo* counting services with varying degrees of detectability (Mansfield 2000).

A review of in-house procedures used to assess the concentration of radioactivity in urine indicates that quality control steps were an integral part of the process (Miller 1979). For example, LLNL ran duplicates consistently, and comparisons of results to "known quantities" were a critical step. Therefore, *in vitro* results from in-house processing are generally reliable. However, interpretation of those results can be difficult, primarily because they might not have considered the contribution of environmental radioactivity (i.e., uranium, thorium). Because sample collection could have occurred at work (e.g., "in-field" tritium analyses, Monday morning urines), cross-contamination could be an issue.<sup>2</sup>

The primary LLNL missions have been weapons research and development (R&D), controlled nuclear weapons research, peaceful uses of nuclear explosives, biomedical research, and laser fusion research. In addition, work in non-nuclear technologies and materials testing has been ongoing since at least 1976 (LLL ca. 1978). LLNL conducted above-ground (atmospheric) and underground tests at both the Pacific Proving Ground, and at the Nevada Test Site from 1953 to 1992. Although the tests were conducted offsite, test planning and preparation were done at LLNL. In 1971, LLNL managed the CANNIKIN test event at Amchitka Island, Alaska.

Unless site-specific information is available, the particle size is assumed to be 5- $\mu$ m activity median aerodynamic diameter (AMAD), as recommended in ICRP (1995, paragraph 5).

### 5.2.1 Plutonium

If a monitoring result refers to "weapons-grade plutonium" or plutonium, dose reconstructors can use the isotopic mix listed in Table 5-1 for material aging times (Mansfield 2000). Until the fifth year of site operation, fresh plutonium should be assumed. For years 5 through 9, assume a 5-year-old plutonium mixture. After these times, 10-year-old plutonium should be assumed. For assessing Super S solubility, refer to *Estimating Doses for Plutonium Strongly Retained in the Lung* (ORAUT 2010a).

<sup>2</sup> Twenty-four-hour collections typically took place at the employee's home on weekends. However, spot samples were often collected in site restrooms (Mansfield 1989).



Table 5-1. Isotopic information for aging of weapons-grade plutonium (Mansfield 2000).

| Mixture designation                        | Fresh    | 5-yr     | 10-yr    | 20-yr    |
|--|----------|----------|----------|----------|
| Years of aging                             | 0        | 5        | 10       | 20       |
| <b>Specific activity in mixture (Ci/g)</b> |          |          |          |          |
| Pu-238                                     | 6.85E-03 | 6.59E-03 | 6.33E-03 | 5.85E-03 |
| Pu-239                                     | 5.81E-02 | 5.81E-02 | 5.80E-02 | 5.80E-02 |
| Pu-240                                     | 1.37E-02 | 1.37E-02 | 1.37E-02 | 1.37E-02 |
| Pu-241                                     | 5.98E-01 | 4.70E-01 | 3.69E-01 | 2.28E-01 |
| Pu-242                                     | 1.57E-06 | 1.57E-06 | 1.57E-06 | 1.57E-06 |
| Am-241                                     | 0.00E+00 | 4.24E-03 | 7.54E-03 | 1.21E-02 |
| Pu-239+240                                 | 7.18E-02 | 7.18E-02 | 7.17E-02 | 7.17E-02 |
| Pu-alpha                                   | 7.87E-02 | 7.84E-02 | 7.80E-02 | 7.76E-02 |
| <b>Activity ratios</b>                     |          |          |          |          |
| Pu-239+240:Am-241                          | N/A      | 1.69E+01 | 9.51E+00 | 5.93E+00 |
| Pu-239+240:Pu-238                          | 1.05E+01 | 1.09E+01 | 1.13E+01 | 1.23E+01 |
| Pu alpha:Pu-239+240                        | 1.10E+00 | 1.09E+00 | 1.09E+00 | 1.08E+00 |
| Pu alpha:Pu-238                            | 1.15E+01 | 1.19E+01 | 1.23E+01 | 1.33E+01 |
| Pu-241:Pu alpha                            | 7.60E+00 | 6.00E+00 | 4.73E+00 | 2.94E+00 |

## 5.2.2 Uranium

If a monitoring result refers to “uranium,” dose reconstructors can use the isotopic mix listed in Table 5-2 for enrichment levels (Mansfield 2000). LLNL received uranium metal from the Y-12 plant that was known to contain recycled uranium contaminants, including technetium, neptunium, thorium, and plutonium [1]. ORAUT (2006a) discusses the typical concentration of recycled uranium contaminants. Table 5-3a provides the list of recycled uranium ratios to be applied for uranium material received from Y-12. The recycled uranium ratios listed in Table 5-3a reflect upper bound values. Given that there is uncertainty in the specific batches of uranium received by LLNL, the upper bound values in Table 5-3a should be used for dose reconstruction. LLNL also used natural uranium from Fernald for the Atomic Vapor Laser Isotope Separation (AVLIS) project. Table 5-3b provides the list of recycled uranium ratios to be applied for uranium associated with the AVLIS project. In the absence of specific information, the Y-12 recycled uranium ratios in Table 5-3a should be used.

Table 5-2. Isotopic information for various uranium enrichment levels (Mansfield 2000)<sup>a</sup>.

| Mixture            | Radionuclide | Mass fraction | Activity fraction | Specific activity of mix (Ci/g) |
|--------------------|--------------|---------------|-------------------|---------------------------------|
| DU                 | U-238        | 9.975E-1      | 9.02E-1           | 3.72E-7                         |
| DU                 | U-235        | 2.50E-3       | 1.45E-2           |                                 |
| DU                 | U-234        | 5.00E-6       | 8.4E-2            |                                 |
| U-nat.             | U-238        | 9.93E-1       | 4.77E-1           | 7.00E-7                         |
| U-nat.             | U-235        | 7.20E-3       | 2.23E-2           |                                 |
| U-nat.             | U-234        | 5.60E-5       | 5.00E-1           |                                 |
| AVLIS <5% enriched | U-238        | 9.50E-1       | 4.11E-1           | 7.78E-7                         |
| AVLIS <5% enriched | U-235        | 5.00E-2       | 1.39E-1           |                                 |
| AVLIS <5% enriched | U-234        | 5.60E-5       | 4.50E-1           |                                 |
| 95% HEU            | U-238        | 3.96E-2       | 1.98E-4           | 6.72E-5                         |
| 95% HEU            | U-235        | 9.50E-1       | 3.06E-2           |                                 |
| 95% HEU            | U-234        | 1.04E-2       | 9.69E-1           |                                 |

a. When U238, U28, or U38 was reported, these were units of total U mass (Mansfield 2006a).

Table 5-3a. Y-12 recommended recycled uranium contaminant ratios defaults based on upper levels of expected ranges (ORAUT 2006a).

| Isotope | Default levels in pCi/μg U         |  | Default levels in dpm/μg U         |  | Default levels in pCi/pCi or dpm/dpm total U |  |
|---------|------------------------------------|--|------------------------------------|--|--|--|
|         | Highly Enriched Uranium & Oralloys | Depleted, Natural, and 2% Enriched Uranium | Highly Enriched Uranium & Oralloys | Depleted, Natural, and 2% Enriched Uranium | Highly Enriched Uranium & Oralloys           | Depleted, Natural, and 2% Enriched Uranium |
| Tc-99   | 4                                  | 0.3  | 0.88                               | 0.666                                      | 4.00E-02                                     | 0.15                                       |
| Th-228  | 0.5                                | 0.003                                      | 1.11                               | 0.00666                                    | 5.00E-03                                     | 1.50E-03                                   |
| Np-237  | 0.6                                | 0.003                                      | 1.332                              | 0.00666                                    | 6.00E-03                                     | 1.50E-03                                   |
| Pu-238  | 0.2                                |  | 0.444                              |  | 2.00E-03                                     |  |
| Pu-239  | 0.2 (Oralloy only)                 | 0.003                                      | 0.444 (Oralloy only)               | 0.00666                                    | 2E-03 (Oralloy only)                         | 1.50E-03                                   |

If the bioassay results are expressed in units of mass, the dose reconstructor should select the isotopic mix equivalent to natural uranium (see Attachment B) in almost all cases at LLNL. The

Table 5-3b. Fernald recycled uranium contaminant ratios for natural uranium. (ORAUT 2004).

| Isotope | Recycled uranium contaminant ratios for natural uranium |          |                            |
|---------|---|----------|----------------------------|
|         | pCi/μg U  | dpm/μg U | pCi/pCi or dpm/dpm total U |
| Pu-239  | 0.00629   | 0.01396  | 0.00921                    |
| Np-237  | 0.00250   | 0.00555  | 0.00366                    |
| Tc-99   | 0.15435   | 0.34266  | 0.22599                    |

uranium in question would be either depleted or natural uranium. Highly enriched uranium (HEU) which came from Y-12 was used in Building 231, and the Oralloys Shop in Building 321C. HEU was restricted at LLNL, both in areas and amounts used, due to criticality concerns. HEU machining was done in Building 321C in either enclosures/hoods or close capture ventilation systems. Building 231 is an experimental, manufacturing, assembly, test, and materials-handling facility. Building 231 also housed research and development activities. Building 241, which conducted materials science research, had an old glove box line in Room 1600 which contained enriched uranium. As result, internal exposure potential to HEU in Building 241 would have been remote. Building 327 is a radiography facility which conducted nondestructive testing of materials, including HEU. Because of the nature of the work in Building 327, internal exposure potential to HEU would be low. LLNL performed *in vitro* and *in vivo* analysis for HEU.

### 5.2.3 Tritium

Tritium was encountered in several forms: tritium oxide as water or gas (HTO), elemental tritium or tritiated gas (HT), organically bound tritium (OBT), and metal tritides (MTs). Each form has unique characteristics. Tritium handling at LLNL was first performed in Building 231 in the early to mid-1950s. Gas fill operations were performed along with other experimental activities. Tritium handling at Building 331 began in 1958. The second phase of Building 331 was completed in 1962. Tritium-filled microspheres were used at LLNL in Buildings 381 and 391 for the Shiva Nova II experiment in 1977. Stable MTs were handled in Building 331, Rooms 154, 150, 138, 145 (Odell and Arthur 1975), Building 212, and Building 292. Outside these buildings, in cases where exposure to OBT or MTs are not implied by case-specific information, assume HTO. Tritium gas work was done in processing hoods. Compounds of tritium, namely MTs, were usually worked in gloveboxes. The glovebox air

was filtered because experimentation with metal tritides required dry inert environments to prevent contamination of the material under experimentation with water from the environment. Guidance for estimating doses for OBT and MTs is available in ORAUT-OTIB-0066, *Calculation of Dose from Intakes of Special Tritium Compounds* (ORAUT 2007a).

Site locations for potential exposure to tritium are listed in Attachment A. Bioassay results for tritium are usually available in the dosimetry records.

#### 5.2.3.1 **Metal Tritides**

Tritium exposures in the form of MT aerosols were primarily titanium tritide targets for the accelerator beams used in Buildings 212 and 292. Operations at Building 212 were the forerunner to RTNS II (Building 292). Tritium operations were conducted at Building 212 from 1966 until 1978 when the RTNS II became operational. Operations at Building 212 were significantly reduced after this time, and were terminated in 1987. Operations at Building 292 began in 1978 and were terminated in 1987. This facility took over tritium operations that previously had been conducted at Building 212. The titanium MT targets were handled in Building 331, Rooms 150, 138, and 145. The personnel who would have had the potential of exposure to the MT targets would be chemists, researchers, health physics technicians, and accelerator operators and mechanical technicians who performed work on the accelerators.

The targets in Buildings 212 and 292 were changed primarily by the accelerator operators. Occasionally, some of the facility support staff, usually mechanical technicians, changed the targets. The titanium tritide targets for the RTNS I accelerator in Building 212 were changed inside of a hood which was exhausted through a HEPA Filter. The target was inserted in one side of the hood and the target was changed through glove ports on the other side of the hood. Individuals involved in the target change operations wore gloves, protective clothing (typically a lab coat), and a half-mask respirator. A Hazards Control Monitor (Health and Safety Technician) was required to be present for the target change operation. The Hazards Control Monitor was responsible for making radiation measurements and taking air samples to check for airborne contamination. Bioassay samples were collected within 4 hours after the target handling operation. Because of the larger physical size of the titanium tritide targets used on RTNS II in Building 292, they were not changed in a hood enclosure as were the RTNS I targets. The workers changing the targets wore anti-contamination clothing, plastic shoe covers, a paper hood, gloves, a plastic face shield and a half-mask respirator. Health and Safety Technicians provided radiation monitoring support (making radiation measurements, taking swipes and collecting air samples) during the operation. After removing their protective clothing, workers were swiped for clothing and skin contamination by the Health and Safety Technician. Bioassay samples were collected within 4 hours after the target handling operations (Myers 2008).

Solubility type M should be assumed for assigning internal dose from titanium tritides. Based on available information, there is no indication that MT solubility type S was a possibility at LLNL.

ORAUT-OTIB-0066 (ORAUT 2007a) provides guidance on the evaluation of MT intakes. MTs are referred to as *tritium particulates*. In addition to potential exposure in the facilities listed above, the claimant telephone interview can provide indications that a person was exposed to MTs. Building exposure locations are also available in tritium dosimetry records, as an aid to the dose reconstructor in assigning tritium dose from MTs.

#### 5.2.3.2 **Organically Bound Tritium**

Tritium exposures in the form of OBT are primarily found in pump oils associated with fume hoods. Exposure to OBT would occur during maintenance if contact with the skin was made. If claimant

information indicates the need for assigning tritium dose based on OBT, guidance on the calculation of doses from OBT is given in ORAUT-OTIB-0066 (ORAUT 2007a).

#### 5.2.4 Other Limited-Exposure Radionuclides

LLNL has always been a center for research. As such, small-scale use of various radionuclides (in terms of either the number of persons or activity of the source) that are not addressed above has occurred throughout the history of LLNL. The internal dose potentials to these nuclides were remote, given the requirements of handling unsealed sources in fume hoods, hot cells, gloveboxes, and dryboxes. LLNL can provide a broad spectrum of *in vivo* counting services with varying degrees of detectability (Mansfield 2000). This is also evident by the radionuclides assessed by lung counting or whole-body (WB) counting listed in Table 5-7 in Section 5.3.3. The dosimetry records would provide this information to the dose reconstructor as part of the case-specific information. In addition, Attachment C provides a method to scale the intakes provided to present a credible exposure to individuals exposed to rarely encountered radionuclides.

### 5.3 MEASUREMENT METHODS

*In vivo* measurement methods began at the site as far back as 1964, when LLNL used sodium iodide detectors connected to simple multichannel analyzers to evaluate  $^{40}\text{K}$  and fission products in humans. In 1964, a scanning bed, referred to as the "RIDL," was in use at LLNL. This device was a shadow shield counter equipped with a 3-in. by 2-in. sodium iodide detector for detection and quantification of relatively high (i.e., greater-than-200-keV) gamma photons. It had some capability for locating the source of radionuclide deposition in subjects. This device was calibrated with a water tank phantom spiked with  $^{137}\text{Cs}$  and  $^{40}\text{K}$  (Hickman undated).

In April 1965, thin crystal studies began (Schmidt and Anderson 1965). In 1965 and 1966, LLNL identified  $^{65}\text{Zn}$  in people working at the Building 153 cyclotron, which dominated the study groups at that time (Anderson 1964-1967).

Workers were selected for WB or specific organ counting based on the program supervisor and health physics staff. Baseline and termination counts occurred as early as 1966 (Balanda 1966).

From at least 1957, *in vitro* measurement methods were in use, with routine monitoring occurring annually and semiannually during those early days [2]. LLNL selected workers for internal monitoring based on the potential of internal exposure. Employees whose work with radionuclides warranted routine testing for internal radiation were determined based on the supervisor of each operation involved and the Radiation Safety Section Health Physicist assigned to the area (Balanda 1966). Building 331 had a monthly monitoring program in place. During weapons test support periods, routine sampling frequencies for involved employees appear to have been increased to quarterly, with weekly sampling for Building 331 (for tritium) and monthly sampling for Building 251 (Balanda 1966). Workers were selected for internal monitoring based on the potential of internal exposure through the 1980s, in conjunction with the program supervisor and the area Health Physicist (Mansfield 1989). The selection of workers for internal monitoring based on the potential of internal exposure continues at LLNL (Mansfield 2000).

Bioassays were supplemented with workplace airborne monitoring (Mansfield 1989). In addition, nasal smears were collected after incidents as a means of assessing intake potential. The dose reconstructor might come across some of these data in worker dosimetry records (Mansfield 1989, Appendix A).

Site 300 established a routine bioassay monitoring program as early as 1971 where all Site 300 personnel received an employment start and termination WB count. In addition, persons who worked

in the firing areas and other radioactive materials handling (i.e., process) areas were bioassayed for gross alpha and beta, and underwent fluorometric analysis for uranium. Additional WB count or bioassays were requested at the discretion of the Health Physicist (NAG 1971). Starting in 1980 at Site 300, all persons working at, or who can be expected to spend a considerable portion of their time in, areas where an airborne uranium hazard might exist will require an annual lung count and a semiannual urine analysis. Other schedules might be required if radionuclides other than uranium are used and considered a potential internal hazard (Straume 1980). Tables 5-4 and 5-5, compiled from a variety of LLNL documents (cited in the table footnotes), list *in vivo* and *in vitro* sampling frequencies for various employment periods. In the case of *in vitro* methods, sample sizes could have been “spot” (single void), 24-hour collections, or simulated 24-hour collections, with creatinine corrections performed if the results were indicative of “significant” intakes (Mansfield 2000).

Table 5-4. Routine *in vitro* sampling frequencies.<sup>a,b</sup>

| Radionuclide            | Period            | Building  | Measurement type | Frequency                               |
|-------------------------|-------------------|-----------|------------------|---|
| Ar-37                   | 1980 <sup>c</sup> | 151       | Urine            | Weekly, monthly                         |
| Gross alpha, beta (LSC) | 2000–present      | Site-wide | Urine            | As applicable                           |
| HTO                     | 1989 <sup>c</sup> | Site-wide | Urine            | Weekly, monthly                         |
| HTO                     | 2000–present      | Site-wide | Urine            | Quarterly, monthly, biweekly, or weekly |
| Kr-85                   | 1980 <sup>c</sup> | 151       | Urine            | Weekly, monthly                         |
| N-13                    | 1980 <sup>c</sup> | 151       | Urine            | Weekly, monthly                         |
| Workplace-specific      | 1966 <sup>c</sup> | 101       | Urine            | Quarterly, annual                       |
| Workplace-specific      | 1966 <sup>c</sup> | 102       | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 110       | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 112       | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 114C      | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 117       | Urine            | Quarterly, annual                       |
| Workplace-specific      | 1966 <sup>c</sup> | 121       | Urine            | Semiannual                              |
| Workplace-specific      | 1966 <sup>c</sup> | 125       | Urine            | Semiannual                              |
| Workplace-specific      | 1966 <sup>c</sup> | 127       | Urine            | Semiannual                              |
| Workplace-specific      | 1966 <sup>c</sup> | 170       | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 171       | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 172       | Urine            | Weekly, monthly                         |
| Workplace-specific      | 1966 <sup>c</sup> | 173B      | Urine            | Annual                                  |
| Workplace-specific      | 1966 <sup>c</sup> | 190       | Urine            | Semiannual, monthly                     |
| Workplace-specific      | 1966 <sup>c</sup> | 193       | Urine            | Annual                                  |
| O-15                    | 1980 <sup>c</sup> | 151       | Urine            | Weekly, monthly                         |
| P-32                    | 1989 <sup>c</sup> | Site-wide | Urine            | Monthly                                 |
| P-32, C-14 (LSC)        | 2000–present      | Site-wide | Urine            | As applicable                           |
| Pu                      | 2000–present      | Site-wide | Urine            | Semiannual                              |
| TRU                     | 2000–present      | Site-wide | Urine            | Annual, semiannual                      |
| U                       | 1980 <sup>c</sup> | 231       | Urine            | Quarterly                               |
| U                       | 1983 <sup>c</sup> | 251       | Urine            | Quarterly                               |
| U                       | 1985 <sup>c</sup> | Site 300  | Urine            | Semiannual (collected on Mondays)       |
| U                       | 2000–present      | Site-wide | Urine            | Quarterly, monthly                      |
| Weapons-grade Pu        | 1989 <sup>c</sup> | Site-wide | Urine            | Semiannual                              |

- Sources: Mansfield (2000); Balanda (1966, Appendix A); LLNL (1987a); Ozaki (1980); Wilson (1982); Leahy (1983); LRL (1961, 1964); Gibson (1985).
- These are typical *in vitro* monitoring frequencies, and varied depending on workplace conditions, suspected inhalation events, and positive or suspected positive bioassay results.
- Periods listed as a single year were taken from annual reports that did not indicate a beginning date for the stated bioassay period.

Table 5-5. *In vivo* sampling frequencies.<sup>a, b</sup>

| Radionuclide         | Period                 | Building  | Measurement type | Frequency                      |
|----------------------|------------------------|-----------|------------------|--------------------------------|
| As detected          | 2000–present           | Site-wide | WB scans         | Annual                         |
| I-131                | 1989-1999 <sup>c</sup> | Site-wide | Thyroid          | Monthly                        |
| I-131                | 2000–present           | Site-wide | Thyroid count    | Coordinated with work schedule |
| MFPs                 | 1964–present           | Site-wide | WB scans         | Not available                  |
| Pu, Am               | 1970–present           | Site-wide | Lung count       | Annual                         |
| Pu, weapons-grade Pu | 1989 <sup>c</sup>      | Site-wide | Lung             | Annual                         |
| Pu-239               | 1961–present           | Site-wide | Wound            | Incident-specific              |
| U                    | 1985 <sup>c</sup>      | Site 300  | Lung count       | Annual                         |
| U, Th                | 1987                   | Bldg. 321 | Lung count       | Annual                         |
| U, Th                | 2000–present           | Site-wide | Lung count       | Annual, semiannual             |

a. Sources: Mansfield (2000); Balanda (1966, Appendix A); LLNL (1987a); Ozaki (1980); Wilson (1982); Leahy (1983); LRL (1961, 1964); Gibson (1985).

b. These are typical *in vivo* frequencies, and varied depending on workplace conditions, suspected inhalation events, and positive or suspected positive bioassay results.

c. Periods listed as a single year were taken from annual reports that did not indicate a beginning date for the stated bioassay period.

Selected records report the presence of <sup>239</sup>Pu and <sup>240</sup>Pu as determined by accelerator mass spectrometry (also called CAMS – Center for Accelerator Mass Spectrometry) and are reported in units of activity that were converted from units of mass. The accelerator mass spectrometry data in a worker's records could come primarily from two sources: (1) For select individuals with known or suspected uptakes, an intercomparison was performed between alpha spectrometry data and accelerator mass spectrometry data to validate the mass spectrometry techniques, and/or (2) In the case of suspected uptakes or for follow-up, mass spectrometry is used because it typically has a detection limit 10 times to as much as 100 times lower than alpha spectrometry. Where two positive values are reported for the same sample, the higher of the two should be used. If two values are reported at the minimum detectable activity for the same sample, the lower of the two values should be used. If reported as <sup>239</sup>Pu, the alpha spectrometry value is <sup>239</sup>Pu and <sup>240</sup>Pu. In the case of accelerator mass spectrometry, the sum of the <sup>239</sup>Pu and <sup>240</sup>Pu should be used to compare to the alpha spectrometry value.

### 5.3.1 Analysis of Gross Alpha Measurements

Urine samples were analyzed for the presence of alpha radiation from about 1957 through 1996 [3] using three possible procedures, including *Analytical Method for Determining Gross Alpha Activity in Urine, Determination of Plutonium and Americium in Urine by Separation*, and *Urinalysis for Curium by Electrodeposition* (Miller 1979). The results were reported as gross alpha, but all three involved the chemical separation of several different elements, depending on the method that was used (Bihl 2006a). The records are not clear about which method was used and require some review by the dose reconstructor. The methods were capable of separating uranium from the sample before analysis to reduce the potential interference (Miller 1979).

The three urine procedures involving alpha (not including uranium) are described below (Miller 1979):

1. *Analytical Method for Determining Gross Alpha Activity in Urine*. This method involved bismuth phosphate coprecipitation, and then coprecipitation with lanthanum fluoride. Thorium, americium, curium, actinium, and neptunium are carried through to determination with this procedure. This procedure also captures plutonium.
2. *Determination of Plutonium and Americium in Urine by Separation*. This method is specific for plutonium and americium. It involves precipitation with ammonium hydroxide, dissolution in

nitric, and then adsorption on an anion exchange column. The americium does not stick on the column so it comes right through with the wash. If gross alpha was to be measured as well as the plutonium, the effluent was counted on a proportional counter. Meanwhile, the plutonium was eluted from the column, electroplated, and counted in a scintillation counter.

3. *Urinalysis for Curium by Electrodeposition.* This method has the same first steps as procedure 1, but has a third precipitation using lanthanum hydroxide. This separates curium from all the other alpha emitters. The precipitate is electroplated, and then can be counted using "pulse-height analysis," low background proportional counting, or autoradiography.

Gross alpha results were a mix of procedure 1 or 2. The database does not indicate which procedure was used. Sometimes the gross alpha analysis might have been specific for americium, other times it included natural thorium and plutonium. To account for either possibility [4]:

1. If a worker has gross alpha and plutonium results on the same day (i.e., one sample analyzed sequentially), assume there is no plutonium in the gross alpha result and assess it as  $^{241}\text{Am}$ .
2. If there is a gross alpha result without a plutonium result on the same day, assume it contains both plutonium and americium.
  - a. If there are also plutonium results for the individual:
    - i. Assess the plutonium results first.
    - ii. Use the plutonium intake to project the plutonium content of the urine samples on the dates of the gross alpha results.
    - iii. Subtract the plutonium component from each of the gross alpha samples.
    - iv. Use the adjusted sample results to assess a  $^{241}\text{Am}$  intake. If the individual also had  $^{241}\text{Am}$  results (from step 1), include those in the assessment (do not subtract plutonium content from them).
  - b. If there are no plutonium results for the individual:
    - i. If the work location is Building 251
      1. assess the gross alpha results twice, once assuming plutonium and once assuming  $^{241}\text{Am}$ .
      2. Select the nuclide yielding the largest dose to the organ of interest.
    - ii. If the work location is not Building 251 or unknown, assess the gross alpha results assuming plutonium.

Exceptions occur if there is something in the file that shows the worker being exposed to neptunium or curium. Another possible exception would involve thorium. LLNL performed some machining of natural thorium. Thorium machining was done in either enclosures/hoods or close capture ventilation systems. Natural thorium, along with depleted and natural uranium was also used at Site 300 in high explosive tests. Indications are that WB counting was used to monitor for thorium intakes. If a worker was getting regular uranium analyses, and then had a gross alpha bioassay analysis, this suggests that the worker had been working with the uncontained thorium, and the gross alpha result should be interpreted as thorium. The worker's job description should be evaluated to determine if this is

appropriate. If there is a WB count, the thorium intake calculated from the gross alpha urinalysis could be compared against the detection limit of the WB count.

From mid-2002 to late 2003, bioassay records contained an analyte termed "GA-X." These samples, indicated by GA-1, GA-2, or GA-3, represent low, middle, and high alpha energies and were reported along with a total gross alpha that represented the sum of activity in all three regions. In late 2003, the low- and middle-energy regions (GA-1 and GA-2) were combined into a single region labeled GA-1. The new GA-1 region spans alpha energies from about 3.9 MeV to 5.1 MeV, and would normally include  $^{232}\text{Th}$ ,  $^{238}\text{U}$ ,  $^{235}\text{U}$ ,  $^{234}\text{U}$ , and  $^{237}\text{Np}$ . The GA-3 region spans alpha energies from about 5.3 MeV to 6.1 MeV, and would normally include  $^{241}\text{Am}$ ,  $^{228}\text{Th}$ ,  $^{244}\text{Cm}$ ,  $^{249}\text{Cf}$ , and some higher energy naturally occurring radionuclides [5].

### 5.3.2 Analysis of Gross Beta Measurements

Urine samples were also analyzed for beta-emitting radionuclides. The gross beta procedure included some separation chemistry that removed  $^{40}\text{K}$  (Bihl 2006a) before counting. The procedure included a broad spectrum of other beta-emitting radionuclides that would have been captured and counted, although the percent recovery might have differed among the elements. Radioelements likely to have been captured and counted would have been strontium, barium, radium, most of the transition metals unless they form strong amine complexes (such as cobalt, nickel, copper, zinc, and silver), all the rare earths, and all the actinides. This means that beta-emitting progeny of thorium and uranium were likely to be included in the gross beta results. Ruthenium-103 and -106 were likely captured and counted (Bihl 2006a). LLNL used other methods to separate  $^{32}\text{P}$ ,  $^{14}\text{C}$ , and  $^{90}\text{Sr}$  and listed the results for these specific nuclides in the results. A result described as gross beta represented many potential nuclides. Where gross beta samples were reported in units of cpm, the dose reconstructor should assume a conversion of 0.4 cpm/dpm (ORAUT 2009).

There are three different source terms for beta/gamma-emitting radionuclides:

- Fission and activation products from a reactor (through 1980),
- The residual from weapons tests (through 1992), or
- Aged fission products and research-related radionuclides (after 1992).

Note that for intakes before 1981 both reactor mixtures up through 1980 and weapons residue mixtures should be considered. The mixture that gives the highest dose to the organ of interest should be assigned for the relevant work periods. After 1980 only weapons residue should be assumed for the gross beta results up through 1992. Reactor mixtures should only be assumed up through 1980, since the Livermore Pool-Type Reactor (LPTR) which began operations in December of 1957 was permanently shut down on March 31, 1980.

Dosimetry records may contain WBC information for a worker. WBC information can be used to limit the intakes determined by the three gross beta methods listed below. This can be done by multiplying the gross beta intakes by the ratio of the WBC radionuclide intake to the specific gross beta radionuclide intake.

#### 5.3.2.1 **Reactor Mixtures (1974–1980)**

*Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses*, ORAUT-OTIB-0054 (ORAUT 2007b) discusses how to use gross beta urine bioassay results to assign intakes of fission and activation products. Of the three types of gross beta analysis discussed in OTIB-0054, the gross beta analysis used at LLNL is considered to be best described as "Gross beta analysis that includes all fission and activation products except radioiodines and beta emitters naturally in urine such as  $^{40}\text{K}$ " (see Section 7.1 of OTIB-0054). The default assumption for



this is 10-day-old reactor fuel (Table 7-1 in OTIB-0054). Therefore, the following method is used to assign internal dose from gross beta results from reactor mixtures:

1. Determine the  $^{90}\text{Sr}$  intake by multiplying the gross beta results by the  $^{90}\text{Sr}$  average fraction of beta activity in urine sample for 10-day-old reactor fuel.
2. Determine the associated radionuclides using the ratios for estimating intakes for intakes based on intake activity relative to  $^{90}\text{Sr}$  for 10-day-old reactor fuel in Table 7-3 in OTIB-0054.

### 5.3.2.2 Weapons Residue (1974–1992)

A significant amount of the work involving beta-emitting radionuclides that would have been detected by the gross beta procedure involved analysis of soil cores from the Nevada Test Site (NTS) (Bihl 2007). The fission/activation product mixtures from atomic weapons testing generally were not the same as those for reactor fuel.

Some results of analyses of drill-back core samples from NTS shots are documented in Hicks (1981) in terms of activity per square meter. Results from five shots were analyzed to determine the more abundant contributors to beta exposure in the core samples. The five shots analyzed were Sedan (1962), Schooner (1968), Sulky (1964), Cabriolet (1968), and Palanquin (1965) (Bihl, Brackett, and Smith 2007). All were underground shots. The cores usually arrived within a few days, as opposed to months, after the shot [6]. Exposure to workers depended not only on the age of the samples being worked but, presumably, some build-up of contamination occurred in hoods and ventilation systems. Therefore, exposure was probably a mix of contamination from current cores and older contamination from previous cores. Therefore, the analysis compared ratios for 100 days after each shot as being representative of the age of contamination accessible to the LLNL workers (Bihl, Brackett, and Smith 2007).

The analysis revealed that even among these five shots, all taken in the 1960s, the radionuclides of greatest abundance were not consistent. Some shots showed a high abundance of activation products and others showed mostly fission products. For the Sedan, Schooner, and Cabriolet shots, isotopes of tungsten were most predominant, especially  $^{185}\text{W}$ . Iron-55 was also noticeably present in the Schooner and Cabriolet shots. In the Sulky and Palanquin shots, these radionuclides were minor with the usual list of fission products dominating (e.g.,  $^{91}\text{Y}$ ,  $^{89}\text{Sr}$ ,  $^{95}\text{Nb}$ ,  $^{95}\text{Zr}$ ,  $^{144}\text{Ce}$ ,  $^{103}\text{Ru}$ , and  $^{106}\text{Ru}$ ). When the activity-per-square-meter values were totaled for the five shots and presented as a fraction of the total activity, the fractions of the more significant radionuclides were as follows:

|               |        |
|---------------|--------|
| Tungsten-185  | 0.58   |
| Tungsten-181  | 0.36   |
| Ruthenium-103 | 0.0070 |
| Tungsten-188  | 0.0064 |
| Yttrium-91    | 0.0051 |
| Iron-55       | 0.0046 |
| Niobium-95    | 0.0044 |
| Strontium-89  | 0.0041 |
| Cerium-141    | 0.0036 |
| Zirconium-95  | 0.0033 |
| Ruthenium-106 | 0.0022 |
| Beryllium-7   | 0.0019 |
| Manganese-54  | 0.0017 |
| Cerium-144    | 0.0013 |

Cobalt-58

0.0013

Tungsten-181 is not a beta emitter and produces little organ dose per unit activity in the organ. The same is true for  $^7\text{Be}$  (Bihl, Brackett, and Smith 2007). It is unlikely the tungsten isotopes or  $^7\text{Be}$  were source terms for workers other than for the NTS core analysis program; for instance, they were not likely sources for reactor workers or accelerator workers.

Ruthenium-103 is the third most abundant radionuclide in the five NTS shots, is fairly volatile, and would apply well to the reactor workers. Its half-life compares reasonably well to that of  $^{185}\text{W}$  (39 versus 75 days, respectively) (Kocher 1981) and it produces higher dose in all organs (except ET1) per unit activity in urine than  $^{185}\text{W}$  (Bihl, Brackett, and Smith 2007). It also produces higher dose per unit intake in all organs than  $^{181}\text{W}$  and  $^7\text{Be}$ . Overall, considering abundance, biokinetics, dose per unit activity in urine, and universal application,  $^{103}\text{Ru}$  was considered a reasonably representative beta-emitting radionuclide for all five shots. But some of the other major radionuclides concentrate in different organs. Because there was a mix of radionuclides in any one sample and there was a change in the mix of predominant radionuclides from core to core, it was considered important not to assume that the gross beta urinalysis values were represented by a single radionuclide.

To determine a representative mix, consideration was given to how the major radionuclides concentrate in organs. For instance, barium, beryllium, strontium, and niobium are bone seekers; cerium concentrates mostly in the liver and to some extent bone; yttrium deposits primarily in bone and to some extent the liver; zirconium deposits half to bone and half to all other tissues; ruthenium and iron are mostly distributed evenly to all tissues. Based on all the above considerations, it is recommended that the intake be estimated as consisting of 50%  $^{103}\text{Ru}$  type F, 25%  $^{141}\text{Ce}$  type S, and 25%  $^{89}\text{Sr}$  type F. These values should be used for 1974 through 1992. In terms of activity in urine at equilibrium from chronic exposure, 59% is from  $^{103}\text{Ru}$  and 41% is from  $^{89}\text{Sr}$ . The  $^{141}\text{Ce}$  contribution would be negligible. Beta yield per disintegration from both  $^{103}\text{Ru}$  and  $^{89}\text{Sr}$  is 100% so no adjustment is needed for beta yield. Additional intakes of  $^{141}\text{Ce}$  type S and  $^{89}\text{Sr}$  type F should be included at 50% each of the  $^{103}\text{Ru}$  intakes. Tungsten-181 is not a beta emitter so would not have been detected in the gross beta urinalysis; however, the additional dose to each organ from using  $^{103}\text{Ru}$  accounts for the unmeasured intake/dose from  $^{181}\text{W}$ . Therefore, the following method is used to assign internal dose from gross beta results from weapons residue:

1. Multiply the gross beta results by 0.59 to obtain the  $^{103}\text{Ru}$  contribution.
2. Use the adjusted results to calculate a type F  $^{103}\text{Ru}$  intake.
3. Assign additional intakes relative to the  $^{103}\text{Ru}$  intake:
  - a. Ce-141 (type S) intake =  $0.5 \times ^{103}\text{Ru}$  intake.
  - b. Sr-89 (type F) intake =  $0.5 \times ^{103}\text{Ru}$  intake.

#### 5.3.2.3 Aged Fission Products and Research-Related Radionuclides (Post 1992)

If specific beta bioassay results are available (i.e.,  $^{90}\text{Sr}$ ,  $^{32}\text{P}$ ,  $^{137}\text{Cs}$ , or  $^{14}\text{C}$ ), the dose reconstructor can use that information for determining the internal dose.

If no specific beta bioassay results are available, intakes should be modeled as  $^{90}\text{Sr}$  type F using 55% of the gross beta result. Additional intakes of  $^{137}\text{Cs}$ ,  $^{32}\text{P}$ ,  $^{14}\text{C}$ , and  $^{90}\text{Y}$  should be included, each equal to the  $^{90}\text{Sr}$  intakes. This was based on the following methodology. For post 1992, the radionuclides in Attachment A, which applied to 1992 to 2001, were qualitatively used. Of the radionuclides listed in Attachment A that would have been measured by the gross beta procedure,  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  was considered representative. They tended to be among the more significant radionuclides in many of the beta-exposure facilities and one is a bone seeker and one is a whole-body seeker. In terms of activity in urine at equilibrium from chronic exposure 38% is from  $^{90}\text{Sr}$  and 62% is from  $^{137}\text{Cs}$ . However, beta yield per disintegration from  $^{90}\text{Sr}$  is 200% because the  $^{90}\text{Y}$  beta would have been

counted as well. The beta yield from  $^{137}\text{Cs}$  is 100% so  $^{90}\text{Sr}$  would have constituted 55% of the total beta counts and  $^{137}\text{Cs}$  would have constituted 45% of the total beta counts. Therefore, the gross beta results should be assessed based on 55% type F  $^{90}\text{Sr}$ . The  $^{137}\text{Cs}$  intake is assumed to be equal to the  $^{90}\text{Sr}$  intake, as this is favorable to the claimant. In addition, the listing in Attachment A for the waste operations buildings (513, 514, 514 tank farms, 613, 625) showed that  $^{32}\text{P}$ , and  $^{14}\text{C}$  also constituted significant fractions of the total activity. To account for these sources, the activity intake of  $^{32}\text{P}$ , and  $^{14}\text{C}$  is assumed to be equal to the  $^{90}\text{Sr}$  intake. Yttrium-90 was undoubtedly present with any intake of Sr-90, and it is not known if it was accounted for as part of the reported activity of  $^{90}\text{Sr}$ . Therefore, an intake of  $^{90}\text{Y}$  equal to the intake of  $^{90}\text{Sr}$  should also be included. Therefore, the following method is used to assign internal dose from gross beta results from aged fission products and research-related radionuclides:

1. Determine the  $^{90}\text{Sr}$  (type F) intake assuming 55 percent of the gross beta results.
2. P-32 (type F, M) intake =  $^{90}\text{Sr}$  intake.
3. C-14 (type F, M, S) intake =  $^{90}\text{Sr}$  intake.
4. Cs-137 (type F) intake =  $^{90}\text{Sr}$  intake.
5. Y-90 (type F) intake =  $^{90}\text{Sr}$  intake.

### 5.3.3 Measurement Types and Detection Levels

A variety of program documents reference detection levels for the various *in vivo* and *in vitro* bioassay methods. These methods are listed in Tables 5-6 and 5-7, and the documents are cited in the table footnotes.

Table 5-6. Bioassay *in vitro* detection levels.<sup>a</sup>

| Measurement type | Analysis method                                   | Selected radionuclide <sup>d</sup> | Period       | MDA                    |
|------------------|---|------------------------------------|--------------|------------------------|
| Fecal            | Radiometric alpha spectrometry                    | Pu-239, 238                        | 1977–1980    | 1 nCi/sample           |
| Fecal            | Co-precipitation and proportional-counting        | Pu-239, 238                        | 1990–1999    | 0.04 dpm               |
| Urine            | Co-precipitation, separation (gross alpha method) | Am                                 | 1957–1989    | 0.718 dpm/sample       |
| Urine            | Co-precipitation, separation (gross alpha method) | Am                                 | 1990–1999    | 0.365 dpm/sample       |
| Urine            | 24-hr sample, alpha spec                          | Am-241                             | 2000–present | 0.01 dpm               |
| Urine            | Liquid scintillation                              | C-14                               | 2000–present | 0.002 $\mu\text{Ci/L}$ |
| Urine            | Electrodeposition and proportional counting       | Cm-244                             | 1960–1999    | 0.2 dpm                |
| Urine            | 24-hr sample, alpha spec                          | Cm-244                             | 2000–present | 0.01 dpm               |
| Urine            | Liquid scintillation                              | HTO                                | 1955–1978    | 1 $\mu\text{Ci/L}$     |
| Urine            | Liquid scintillation                              | HTO                                | 1979–present | 0.01 $\mu\text{Ci/L}$  |
| Urine            | Liquid scintillation                              | I-125, I-131, S-35                 | 2000–present | 0.002 $\mu\text{Ci/L}$ |
| Urine            | 24-hr sample, alpha spec                          | Np-237                             | 2000–present | 0.01 dpm               |
| Urine            | Liquid scintillation                              | P-32, P-33                         | 2000–present | 0.002 $\mu\text{Ci/L}$ |
| Urine            | Extraction and gross alpha <sup>b</sup>           | Po-210                             | 1955–1978    | 0.1 pCi/L              |
| Urine            | 24-hr sample, alpha spec                          | Po-210                             | 2000–present | 0.01 dpm               |

| Measurement type | Analysis method   | Selected radionuclide <sup>d</sup> | Period       | MDA   |
|------------------|---|------------------------------------|--------------|---|
| Urine            | Co-precipitation  | Gross alpha                        | 1957–1989    | 0.78 dpm/sample   |
| Urine            | Co-precipitation  | Gross alpha                        | 1990–1999    | 0.365 dpm/sample  |
| Urine            | Co-precipitation, separation and proportional counting                      | Pu-total alpha                     | 1957–1989    | 0.051 dpm/sample  |
| Urine            | Co-precipitation, separation and proportional counting                      | Pu-total alpha                     | 1990–1999    | 0.012 dpm/sample <sup>f</sup>                           |
| Urine            | 24-hr sample, alpha spec  | Pu                                 | 2000–present | 0.01 dpm  |
| Urine            | Gross beta  | MFP                                | 1957–1989    | 600 dpm/L <sup>g</sup>                                  |
| Urine            | Gross beta  | MFP                                | 1990–1999    | 32 dpm/L  |
| Urine            | TRU-spec column with alpha spectrometry                                     | Th-232, Th-228                     | 1989–present | 0.2 dpm/sample (with Th-232 being equivalent to Th-228) |
| Urine            | Fluorometric  | U depleted, natural U, U AVLIS     | 1958–1973    | 5 µg/L  |
| Urine            | Fluorometric  | U depleted, natural U, U AVLIS     | 1974–1978    | 1 µg/L  |
| Urine            | Fluorometric  | U depleted, natural U, U AVLIS     | 1979–1989    | 0.3 µg/L  |
| Urine            | Spot sample KPA   | U depleted, natural U, U AVLIS     | 1990–1999    | 0.05 µg/L   |
| Urine            | Spot sample ICP-MS  | U                                  | 2000–present | 0.002 µg/L  |
| Urine            | Extraction/alpha proportional counting (U-234 alphas measured) <sup>b</sup> | HEU                                | 1955–1970    | 100 dpm/L <sup>e</sup>                                  |
| Urine            | Extraction/alpha proportional counting <sup>b</sup>                         | HEU                                | 1971–1976    | 15 dpm/L  |
| Urine            | Extraction/alpha proportional counting <sup>b</sup>                         | HEU                                | 1977–1986    | 4 pCi/L   |
| Urine            | Alpha spectrometry <sup>c</sup>   | HEU                                | 1987–1999    | 0.3 pCi/L   |

a. Sources: Mansfield (2000); LRL (1960, 1964, 1970); Griffith (1980); Berger (2005); Schmidt and Anderson (1965); ORAUT (2009, 2007a), LLL ca. 1978, Miller (1979). Dupzyk (1990), Simpson (1989), Szalinski .

b. Based on information from Los Alamos National Laboratory (LANL).

c. Based on ICRP (1989) information.

d. Other nuclides might have been detected by the method; these might be reported in workers' files.

e. Based on information from Los Alamos National Laboratory (LANL). Decision level of 50 dpm/L reported. Assume the MDA is 2 times the decision level.

f. MDA range of 0.012 to 0.051 dpm/sample was indicated. DRs should use available sample specific information.

g. Gross beta method MDA was 3.84 cpm/sample with a detector efficiency range of 20 to 31 percent, and 60 percent sample recovery. This results in a MDA range of approximately 400 to 600 dpm/sample depending upon detector efficiency (Miller 1979). DRs should use available sample specific information.

Table 5-7. Bioassay *in vivo* detection levels.<sup>a</sup>

| Measurement type | Analysis method       | Radionuclide   | Period     | MDA                               |
|------------------|-----------------------|----------------|------------|-----------------------------------|
| Lung             | Phoswich <sup>b</sup> | Pu-239, Am-241 | 1966–1969  | 400 nCi, 2 nCi                    |
| Lung             | Phoswich <sup>c</sup> | Pu-239, Am-241 | 1970–1979  | 54 nCi, 0.5 nCi                   |
| Lung             | Phoswich              | Pu-239         | Circa 1978 | 19 nCi                            |
| Lung             | Phoswich              | Am-241         | Circa 1979 | 0.3 nCi                           |
| Lung             | Phoswich              | Am-241         | 1984–1985  | 0.1 – 0.3 nCi (2.3 - 5.25 cm CWT) |

| Measurement type | Analysis method       | Radionuclide                   | Period       | MDA  |
|------------------|-----------------------|--------------------------------|--------------|--|
| Lung             | Dual Phoswich         | Pu-239                         | 1980–present | >16.3 nCi (2.1 cm CWT)   |
| Lung             | --                    | "Pure" Pu-239                  | 1984–1985    | 9 - 200 nCi (1.45 - 5.95 cm CWT)                               |
| Lung             | HPGe arrays           | "Pure" Pu-239                  | 2000–present | 200 nCi  |
| Lung             | --                    | "Pure" Pu-238                  | 1984–1985    | 4 - 100 nCi (1.5 - 4.1 cm CWT)                                 |
| Lung             | ACTII                 | "Pure" Pu-238                  | 1999         | 433 nCi (3.33 cm CWT)  |
| Lung             | HPGe arrays           | "Pure" Pu-238                  | 2000–present | 150 nCi  |
| Lung             | ACTII                 | "Pure" Pu-238                  | 2006         | 87 nCi (3.6 cm CWT)  |
| Lung             | --                    | Weapons-grade Pu               | 1984–1985    | 1 - 4 nCi (1.45 - 5.85 cm CWT)                                 |
| Lung             | HPGe arrays (Am-241)  | Weapons-grade Pu               | 2000–present | 0.15 nCi   |
| Lung             | ACTII                 | Am-241                         | 1999         | 0.14 nCi (3.33 cm CWT)   |
| Lung             | HPGe arrays           | Am-241                         | 2000–present | 0.15 nCi   |
| Lung             | ACTII                 | Am-241                         | 2006         | 0.11 nCi (3.6 cm CWT)  |
| Lung             | --                    | DU, natural U                  | Circa 1977   | 5 nCi  |
| Lung             | --                    | DU                             | 1984–1985    | 0.5 - 2 nCi  |
| Lung             | --                    | Natural U                      | 1984–1985    | 0.7 - 2 nCi  |
| Lung             | --                    | U-235                          | 1984–1985    | ~100 µg (depending upon enrichment)                            |
| Lung             | ACTII                 | U-238 (Th-234)                 | 1999         | 1.27 nCi (3.33 cm CWT)   |
| Lung             | HPGe arrays           | DU, natural U                  | 2000–present | 1 nCi  |
| Lung             | ACTII                 | U-238 (Th-234)                 | 2006         | 0.68 nCi (3.6 cm CWT)  |
| Lung             | ACTII                 | U-235                          | 1999         | 0.07 nCi (3.33 cm CWT)   |
| Lung             | HPGe arrays           | AVLIS U, Th, U-235             | 2000–present | 1 nCi  |
| Lung             | ACTII                 | U-235                          | 2006         | 0.05 nCi (3.6 cm CWT)  |
| Lung             | HPGe arrays           | 40% U, 95% U                   | 2000–present | 0.1 nCi  |
| Lung             | HPGe arrays           | Cm-244                         | 2000–present | 100 nCi  |
| Lung             | HPGe arrays           | Np-237                         | 2000–present | 0.35 nCi   |
| Lung             | ACTII                 | Np-237                         | 2006         | 0.3 nCi (3.6 cm CWT)   |
| Lung             | HPGe arrays           | Pa-233                         | 2000–present | 1 nCi  |
| Lung             | ACTII                 | Pa-233                         | 2006         | 0.13 nCi (3.6 cm CWT)  |
| Thyroid count    | --                    | I-131                          | 1984–1985    | 1 nCi  |
| Thyroid count    | --                    | I-131                          | 2000–present | 0.01 nCi   |
| Thyroid count    | ACTII                 | I-131                          | 2006         | 0.04 nCi   |
| Thyroid count    | Portable NaI          | I-131                          | 2006         | 0.27 nCi   |
| Thyroid count    | --                    | I-125                          | 2000–present | 0.02 nCi   |
| Thyroid count    | ACTII                 | I-125                          | 2006         | 0.31 nCi   |
| Thyroid count    | Portable NaI          | I-125                          | 2006         | 0.02 nCi   |
| WB scan          | Shadow shield counter | K-40                           | 1964–1995    | 150 g K in 20 min (~120 nCi K-40)                              |
| WB scan          | Shadow shield counter | 150- to 300-keV gamma emitters | 1964–present | 500 gammas per second between 150 and 300 keV                  |
| WB Scan          | --                    | Th-232                         | 1970–present | 1 nCi (based on TI-208 with no correction for branching ratio) |
| WB scan          | --                    | Natural Th (TI-208)            | 2000–present | 1 nCi  |
| WB scan          | --                    | I-131 (TB)                     | 1970–1995    | 1 nCi  |
| WB scan          | --                    | Cs-137                         | 1970–1995    | 1 nCi  |
| WB scan          | --                    | Co-60                          | 1970–1995    | 1 nCi  |
| WB scan          | --                    | Ce-144                         | 1970–present | 8 nCi  |
| WB scan          | --                    | Ce-141                         | 1970–present | 2 nCi  |
| WB scan          | --                    | Ru-103                         | 1970–present | 3 nCi  |
| WB scan          | --                    | Ru-106                         | 1970–1995    | 5 nCi  |

| Measurement type | Analysis method | Radionuclide | Period       | MDA          |
|------------------|-----------------|--------------|--------------|--------------|
| WB scan          | --              | Zr-95        | 1970–present | 1 nCi        |
| WB scan          | --              | Mn-54        | 1970–1995    | 1 nCi        |
| WB scan          | --              | Zn-65        | 1970–present | 1 nCi        |
| WB scan          | --              | Na-22        | 1970–1995    | 1 nCi        |
| WB scan          | --              | K-40         | 1996–1999    | 4.46 nCi     |
| WB scan          | --              | Mn-54        | 1996–1999    | 0.63 nCi     |
| WB scan          | --              | Fe-59        | 1996–1999    | 0.98 nCi     |
| WB scan          | --              | Co-60        | 1996–1999    | 0.66 nCi     |
| WB scan          | --              | Ru-106       | 1996–1999    | 6.53 nCi     |
| WB scan          | --              | I-131        | 1996–1999    | 1 nCi        |
| WB scan          | --              | Cs-134       | 1996–1999    | 0.7 nCi      |
| WB scan          | --              | Cs-137       | 1996–1999    | 0.89 nCi     |
| WB scan          | --              | Co-57        | 2000–2005    | 0.97 nCi     |
| WB scan          | --              | Co-60        | 2000–2005    | 0.51 nCi     |
| WB scan          | --              | Cs-137       | 2000–2005    | 0.7 nCi      |
| WB scan          | --              | I-131        | 2000–2005    | 0.66 nCi     |
| WB scan          | --              | K-40         | 2000–2005    | 2.6 nCi      |
| WB scan          | --              | Na-22        | 2000–2005    | 0.44 nCi     |
| WB scan          | --              | K-40         | 2006         | 2.6 nCi      |
| WB scan          | --              | Cs-137       | 2006         | 0.72 nCi     |
| WB scan          | --              | Co-60        | 2006         | 0.43 nCi     |
| WB scan          | --              | I-131        | 2006         | 0.67 nCi     |
| WB scan          | --              | Na-22        | 2006         | 0.42 nCi     |
| WB scan          | --              | Pa-233       | 2006         | 1.5 nCi      |
| WB scan          | --              | Ac-228       | 2007–present | 2.74 nCi     |
| WB scan          | --              | Ba-133       | 2007–present | 0.85 nCi     |
| WB scan          | --              | Bi-207       | 2007–present | 0.47 nCi     |
| WB scan          | --              | Bi-212       | 2007–present | 7.71 nCi     |
| WB scan          | --              | Bi-214       | 2007–present | 1.82 nCi     |
| WB scan          | --              | Cf-249       | 2007–present | 0.83 nCi     |
| WB scan          | --              | Cm-243       | 2007–present | 3.59 nCi     |
| WB scan          | --              | Co-57        | 2007–present | 0.97 nCi     |
| WB scan          | --              | Co-60        | 2007–present | 0.37 nCi     |
| WB scan          | --              | Cs-134       | 2007–present | 0.56 nCi     |
| WB scan          | --              | Cs-137       | 2007–present | 0.7 nCi      |
| WB scan          | --              | Eu-152       | 2007–present | 1.75 nCi     |
| WB scan          | --              | Eu-154       | 2007–present | 1.25 nCi     |
| WB scan          | --              | Eu-155       | 2007–present | 2.58 nCi     |
| WB scan          | --              | I-131        | 2007–present | 0.68 nCi     |
| WB scan          | --              | K-40         | 2007–present | 2.52 nCi     |
| WB scan          | --              | Na-22        | 2007–present | 0.4 nCi      |
| WB scan          | --              | Np-237       | 2007–present | 6.38 nCi     |
| WB scan          | --              | Np-239       | 2007–present | 4.73 nCi     |
| WB scan          | --              | Pa-231       | 2007–present | 21.83 nCi    |
| WB scan          | --              | Pa-233       | 2007–present | 1.6 nCi      |
| WB scan          | --              | Pa-234       | 2007–present | 3.93 nCi     |
| WB scan          | --              | Pb-212       | 2007–present | 2.23 nCi     |
| WB scan          | --              | Pb-214       | 2007–present | 2.36 nCi     |
| WB scan          | --              | Po-210       | 2007–present | 3.40E+04 nCi |
| WB scan          | --              | Ra-224       | 2007–present | 20.6 nCi     |
| WB scan          | --              | Ra-226       | 2007–present | 23.9 nCi     |
| WB scan          | --              | Th-231       | 2007–present | 11.6 nCi     |

| Measurement type | Analysis method | Radionuclide | Period       | MDA      |
|------------------|-----------------|--------------|--------------|----------|
| WB scan          | --              | Th-234       | 2007–present | 15.8 nCi |
| WB scan          | --              | U-235        | 2007–present | 1.61 nCi |

- Sources: Mansfield (2000); LRL (1964, 1970); Schmidt and Anderson (1965); Griffith (1980); Berger (2005); ORAUT (2009, 2007a); LLL (ca. 1978, 1979); King (1977); Author unknown (ca. 1985); Sundsmo and Hickman (2009a,b,c); Hickman (2008).
- Based on LANL information.
- Based on ICRP (1989) information.

In some cases, there is evidence that LLNL implemented monitoring by a particular methodology, but site-specific detection levels for that methodology were not available. In these cases, detection levels for comparable LANL methods were used to complete Tables 5-6 and 5-7 because there is evidence of interlaboratory communications on analytical methods over the years, and because both LLNL and LANL were operated by the same contractor, thus increasing the likelihood for shared procedures (Berger 2005).

In a few instances, neither LLNL nor LANL detection levels were available, requiring a surrogate source of information. ICRP Publication 54 (ICRP 1989) specified nominal detection levels for a variety of measurement methods that were comparable to those used at LLNL.

A review of *in vivo* bioassay records provided by LLNL indicates that the data typically includes the radionuclide assessed, MDA, activity result, height, weight, and chest wall thickness (CWT). In most cases, the dose reconstructor will have all the information necessary to assess the internal dose from *in vivo* bioassay data. In the absence of specific *in vivo* MDA information in the worker's dosimetry records, the dose reconstructor should refer to Table 5-7. The expected intake pattern in most cases is an acute intake. At LLNL, airborne and surface contamination was typically controlled to prevent intakes, so most intakes would have been the result of unexpected releases. Small intermittent releases that were not immediately detectable could have occurred, so an individual could have had multiple acute intakes.

### 5.3.4 Reporting Formats and Codes

There are a variety of codes on urine bioassay records generated at or for LLNL. Table 5-8 is a summary of the codes known at present, along with their interpretation.

Uncertainties associated with bioassay measurements were not stated in the records.

Table 5-8. Bioassay record codes.

| Column no. | Type of entry        | Acceptable entries  |
|------------|----------------------|---|
| 1–6        | Employee number      | 6-digit integer   |
| 8–27       | Employee name        | Last name, comma, other names and/or initials   |
| 28–32      | Nuclide analyzed     | MFP, ALPHA, PU239, PU238, U238, AM241, CM244, or other nuclides (maximum of 5 characters) |
| 34–39      | Date                 | Month, day, year (2 digits each)  |
| 41–48      | Results              | Floating point number between 9999999.0 and 0.000 (F8.3 format)                           |
| 50–51      | Error                | One standard deviation as a percentage of the result                                      |
| 53         | Sample type          | U = urine, B = blood, f = feces, n = nose swipe   |
| 55         | Sample type          | R = routine; S = special  |
| 57–60      | Address              | Building or trailer number (e.g., B253 or 2532)   |
| 61–63      | L code               | 3-digit L code  |
| 65         | Frequency            | W = weekly; M = monthly; Q = quarterly; S = semiannually; A = annually                    |
| 67–70      | Location of exposure | Building number (e.g., B253)  |

| Column no. | Type of entry   | Acceptable entries  |
|------------|-----------------|---|
| 72-76      | Analysis number | Integer value of analysis number assigned to sample by Bioassay Laboratory  |
| 79-80      | Method          | Analytical method used to process samples: 1 = gross alpha technique (since 1957); 2 = gross beta technique (since 1974); 3 = plutonium separation technique (since 1963); 4 = fluorometric uranium technique (since 1974); 5 - 99 = reserved for future revised techniques |

### 5.3.5 Assessment of Intake for Unmonitored Workers

LLNL had an established *in vitro* and *in vivo* bioassay sampling program, and there was clear direction to supervisors on individual employee participation in the program. In the case where a worker may not have been monitored, but should be assigned an intake based on their job description and information in the claimant telephone interview, a means of assessing intakes for unmonitored workers is necessary. There is little correlation between external dosimetry and the bioassay program. From 1953, external dosimetry was administered as a site-wide requirement whereas bioassay was determined based on the potential of exposure by facility supervisors and Health Physics. Dose reconstructors should refer to the coworker data in Attachment B to assign intakes for unmonitored workers. Attachment C provides guidance on assigning the 50th- or 95th-percentile coworker intakes based on job category, and level of contact with the radioactive material.

### 5.3.6 Unmonitored Intakes of LLNL Radionuclides Other than Primary Radionuclides

A short description of the work in relation to each of the listed radionuclides follows. Most were handled as part of basic research, and the number of exposed individuals would have been small.

Actinium-227 was used in Building 251. Building 251 conducted basic research in the chemical and nuclear behavior of the transuranic elements starting in 1955. Building 251 was restricted to "an as needed basis" (LLL 1980). Given the nature of the work conducted, access and radiological controls, and an *in vitro* and *in vivo* bioassay monitoring program in place for workers who worked in areas where radioactive material was routinely handled, unmonitored intakes were unlikely. Starting around mid-1978, a solution of <sup>227</sup>Ac stored in a glovebox in Building 251 was "milked" periodically for its <sup>223</sup>Ra, which was used as an alpha-emitting contaminant in exercises (Gibson 1980). Actinium-227 was also stored in a cave in Room 1235 of Building 251 (Meadows 1981). Given the containment and restrictive use controls, exposure to unmonitored individuals was remote. In 1992 all U.S nuclear testing ceased. In 1993 LLNL decided to discontinue programmatic operations in Building 251 based on new regulatory requirements. Building 251 was placed from an operational mode to standby mode in 1995. During the standby mode period, Building 251 was staffed with three people. Building 251 transitioned from Category II Nonreactor Nuclear Facility in 2002 to a radiological facility in 2005 where equipment and inventory were removed (LLNL 2006a).

Thorium-232 (natural thorium) was associated primarily with Site 300, and two facilities, Buildings 231 and 321C. Building 231 is a large experimental, manufacturing, assembly, test, and materials-handling facility, and Building 321C machined depleted uranium (DU) and thorium. Machining only occurred in 321C. Site 300 was a high-explosive test facility that opened in mid-1955. Depleted uranium, natural uranium, and natural thorium were used with high explosive materials tests at Site 300.

Curium-244 was associated primarily with Building 251, starting in 1955 when the building became operational. As indicated previously, access to Building 251 was restricted to "an as needed basis". Given the nature of the work conducted, access and radiological controls, and an *in vitro* and *in vivo* bioassay monitoring program in place for workers who worked in areas where radioactive material was routinely handled, unmonitored intakes were unlikely. Curium-244 was also used in Building 332 (Plutonium Facility) with similar controls in place. Building 332 became operational in 1961.



Neptunium-237 work was conducted primarily in Building 251 and, as stated above, unmonitored intakes were unlikely. Research on <sup>237</sup>Np was conducted in Room 1319 of Building 281, which included gloveboxes. Building 281 conducted tracer work, dissolution studies, and flow studies. As a result, the potential of internal exposure to <sup>237</sup>Np in Building 281 would have been remote.

Based on the foregoing, the potential for an unmonitored intake of the above-listed radionuclides is limited; however, it is necessary to demonstrate that maximum doses can be assigned when presumptive intakes, beyond those that might be characterized by bioassay records, are implied by case-specific information. In the absence of case specific information, the dose from unmonitored intakes of LLNL radionuclides other than primary radionuclides should not be assigned. Based on the foregoing information on Building 251, any of the radionuclides associated with this building listed above, should not be assigned after 1995.

The following table lists the radionuclides to consider along with the work locations. Workers who are assigned doses from unmonitored intakes of LLNL radionuclides other than primary radionuclides based on the coworker intakes described in the table below will not be assessed for the period prior to 1957 for plutonium and americium, and prior to 1958 for uranium, as they are included in the designated SEC period.

Table 5-9. Radionuclides other than primary radionuclides.

| Radionuclide                  | Building            | Likely exposed individuals  | Period of potential exposure                                      | Dose estimation techniques (in order of preference)  |
|-------------------------------|---------------------|---|---|--|
| Actinium-227                  | 251                 | Chemists, researchers, health physics technicians, technicians, and maintenance | 1978 - 1995   | <ol style="list-style-type: none"> <li>1. Base on bioassay results.</li> <li>2. Assign dose based on coworker intakes for Pu-239 assuming Ac-227 solubility types F, M, and S.</li> </ol>  |
| Neptunium-237                 | 251, 281            | Chemists, researchers, health physics technicians, technicians, and maintenance | 1955 – 1995 (Building 251)<br>1952 - present (Building 281)       | <ol style="list-style-type: none"> <li>1. Base on bioassay results.</li> <li>2. Assign dose based on coworker intakes for Pu-239 assuming Np-237 solubility type M.</li> </ol>   |
| Curium-244                    | 251, 332            | Chemists, researchers, health physics technicians, technicians, and maintenance | 1955 – 1995 (Building 251)<br>1961 - present (Building 332)       | <ol style="list-style-type: none"> <li>1. Base on bioassay results.</li> <li>2. Assign dose based on coworker intakes for Am-241 assuming Cm-244 solubility type M.</li> </ol>   |
| Thorium-232 (natural thorium) | 231, 321C, Site 300 | Chemists, researchers, machinists, health physics technicians, and technicians  | 1952 – present (Buildings 231, 321C)<br>1955 – present (Site 300) | <ol style="list-style-type: none"> <li>1. Base on bioassay results.</li> <li>2. Assign dose assuming a natural thorium mass intake equal to the coworker uranium mass intake. Assume a natural thorium specific activity of <math>4.88 \times 10^2</math> dpm/mg to determine the total thorium activity intake. Assume 50 percent of the total thorium</li> </ol> |

|  |  |  |  |   |
|--|--|--|--|---|
|  |  |  |  | activity intake is Th-232 and 50 percent of the total thorium activity intake is Th-228. Assume solubility types M and S. |
|--|--|--|--|---|

#### 5.4 SIGNIFICANT INCIDENTS WITH INTERNAL DOSE POTENTIAL

During operations at LLNL, a number of incidents increased the potential for intakes of radioactive materials. If a worker recalls involvement in one or more of those incidents, dose reconstructors can use the information in Table 5-10 as input to an incident-specific assessment. This list is not all-encompassing, and many other incidents probably occurred; these are the incidents identified from review of the data capture records. Individual worker records could provide documentation of involvement in other incidents.

Table 5-10. Input parameters for significant incidents.

| Incident date     | Incident description        | Facility            | Other information  |
|-------------------|-----------------------------|---------------------|--|
| March 26, 1963    | Criticality                 | Bldg. 110           | Potential for internal and external exposure (I, Kr, Xe); monitored participants listed by name in 1963 reference (see below). |
| November 6, 1975  | Radium contamination        | Bldg. 343, Rm. 1005 | Crystal was removed from radium dial, resulting in spread of contamination.  |
| November 23, 1971 | Pu-contaminated squib valve | Bldg. 343           | Direct and indirect bioassays performed on 14 individuals  |

Sources: LRL (ca. 1963a,b); Leahy (1975); AEC (1971).

#### 5.5 ATTRIBUTIONS AND ANNOTATIONS

Where appropriate in the preceding text, bracketed callouts have been inserted to indicate information, conclusions, and recommendations to assist in the process of worker dose reconstruction. These callouts are listed in this section with information that identifies the source and justification for each item. Conventional references are provided in the next section that link data, quotations, and other information to documents available for review on the ORAU Team servers.

Thomas LaBone served as a Site Expert for this document and he performed the intake modeling. Mr. LaBone was previously employed by the Savannah River Site and he performed a radiological assessment at LLNL that involved management, direction, or implementation of radiation protection and/or health physics program policies, procedures or practices related to atomic weapons activities at the site. This revision has been overseen by a Document Owner who is fully responsible for the content, including all findings and conclusions. In all cases where information or prior studies or writings are included or relied upon by Mr. LaBone, those materials are fully attributed to the source. Mr. LaBone's Disclosure Statement is available at <http://www.oraucoc.org>.

- [1] Szalinski, Paul A. Integrated Environmental Management. Health Physicist. March 2007. The LLNL *Technical Basis Manual for Internal Dosimetry* (Mansfield 2000) discusses recycled uranium in the DOE complex, so the internal dosimetrists at LLNL are aware of the issues.
- [2] Thomas, Bill R. Integrated Environmental Management. Health Physicist. March 2007. Results for gross alpha in urine were reviewed in the claimant files. The frequency of sample collection was dependant on the particular building where the claimant was assigned. The frequency ranged from samples several times per year to a single sample per year.

- [3] Szalinski, Paul A. Integrated Environmental Management. Health Physicist. March 2007. In Bihl (2006), when asked if the procedures were in place from 1957 to 1996, I. Dupzyk replied that they were, at least to 1987. Because these procedures were based on DOE Environmental Measurement Laboratory Procedures Manual HASL-300 (Cheico 1997), which did not undergo its next significant revision until 1997, it is likely that the procedures were unchanged through 1996.
- [4] Szalinski, Paul A. Integrated Environmental Management. Health Physicist. March 2007. Because the first two gross alpha analysis methods might contain the same radionuclides and there is no information as to which method was used on a given sample, the steps are given to assess the various possibilities that might be encountered.
- [5] Thomas, Bill R. Integrated Environmental Management. Health Physicist. March 2007. The information in this paragraph comes from Explanatory Notes on "Gross Alpha" Results in LLNL claimant files (Author unknown 2006) for Summary of Bioassay Results that were reported after approximately 2002.
- [6] Bihl, Donald E. BPNL. Principal Health Physicist. April 2007. Personal communication with M. H. Chew, October 16, 2006.
- [7] LaBone, Thomas R. ORAU Team. Deputy Principal Internal Dosimetrist. February 2007. The uranium results were recorded as  $\mu\text{g/L}$  and the statistical analyses were performed in those units. However, the Integrated Modules for Bioassay Analysis (IMBA) computer program requires that all excreta data be entered as total excretion per day; hence, the statistical parameters were converted to excretion per day before intake calculations were made using IMBA.
- [8] Brackett, Elizabeth M. ORAU Team. Principal Internal Dosimetrist. February 2007. The uniform absolute error of 1 weights all results equally; other fitting schemes weight high values or low values disproportionately. Because the median and 84th-percentile values were determined from statistical analysis of many samples in each interval, there was no *a priori* reason to weight results from one interval over another.
- [9] LaBone, Thomas R. ORAU Team. Deputy Principal Internal Dosimetrist. February 2007. The breathing rate and particle size distribution are project default values to be used unless site-specific information indicates otherwise. No information has been found concerning intakes at LLNL that shows that the default values should not be used. See, for instance, OCAS-IG-002, *Internal Dose Reconstruction Implementation Guide* (NIOSH 2002), and ICRP Publication 66, *Human Respiratory Tract Model for Radiological Protection* (ICRP 1994).
- [10] Bihl, Donald E. BPNL. Principal Health Physicist. February 2007. The minimum geometric standard deviation (GSD) of 3 is established in ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2007c). It reflects the overall uncertainty associated with biokinetic modeling as well as usual radiochemical analysis, and indicates that even though the spread in coworker excreta results for a given population (e.g., a year of excreta samples) can have a GSD of  $<3$ , the uncertainty of intakes determined using the biokinetic models is no less than 3.
- [11] LaBone, Thomas R. ORAU Team. Deputy Principal Internal Dosimetrist. February 2007. Use of the midpoint of the period represented by the excreta data point is established in ORAUT-OTIB-0019, *Analysis of Coworker Bioassay Data for Internal Dose Assignment* (ORAUT 2005a), and is standard practice for assigning chronic intakes or acute intakes with

unknown dates. See, for instance, ANSI/HPS-N13.39-2001, *Design of Internal Dosimetry Programs* (HPS 2001).

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## GLOSSARY

### absorption type

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

### activity

Amount of radioactivity. The International System unit of activity is the becquerel (1 disintegration per second); the traditional unit is the curie [37 billion ( $3.7 \times 10^{10}$ ) becquerels].

### 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*.

### air sampling

Collection of samples of the ambient atmosphere to detect or measure the presence of radioactive material in the air.

### 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.

### atomic vapor laser isotope separation (AVLIS)

Process for enriching  $^{235}\text{U}$  by ionizing uranium compounds in a vapor based on a laser tuned to the electronic structure of uranium compounds.

### beta radiation

Charged particle emitted from some radioactive elements with a mass equal to 1/1,837 that of a proton. A negatively charged beta particle is identical to an electron. A positively charged beta particle is a positron.

### 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.

**chronic exposure**

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

**claim**

Petition filed by a covered employee or survivor(s) thereof and submitted to the U.S. Department of Labor for compensation under the Energy Employees Occupational Illness Compensation Program. There can be multiple claims filed per worker case.

**claimant**

Individual who has filed for compensation under the Energy Employees Occupational Illness Compensation Program. This individual can be the energy employee (worker), a survivor, or the legal representative of the energy employee.

**cohort**

Group of individuals selected for inclusion in a study.

**contamination**

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

**curie (Ci)**

Traditional unit of radioactivity equal to 37 billion ( $3.7 \times 10^{10}$ ) becquerels, which is approximately equal to the activity of 1 gram of pure  $^{226}\text{Ra}$ .

**depleted uranium (DU)**

Uranium with a percentage of  $^{235}\text{U}$  lower than the 0.7% found in natural uranium.

**dose**

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

**dose equivalent (H)**

In units of rem or sievert, product of absorbed dose in tissue multiplied by a weighting factor and sometimes by other modifying factors to account for the potential for a biological effect from the absorbed dose. See *dose*.

**dosimeter**

Device that measures the quantity of received radiation, usually a holder with radiation-absorbing filters and radiation-sensitive inserts packaged to provide a record of absorbed dose received by an individual.

**electron-volt (eV)**

Unit equal to the energy of one electron moving through a potential difference of 1 volt ( $1.602 \times 10^{-19}$  joules). The common units in nuclear physics and radiology are kiloelectron-volts (thousands) and megaelectron-volts (millions).

**Energy Employees Occupational Illness Compensation Program Act of 2000, as amended (EEOICPA; 42 U.S.C. § 7384 et seq.)**

Law that provides for evaluation of cause and potential compensation for energy employees who have certain types of cancer.

**exchange period**

Period (weekly, biweekly, monthly, etc.) for routine exchange of dosimeters. Also called exchange frequency.

**exposure**

(1) In general, the act of being exposed to ionizing radiation. (2) Measure of the ionization produced by X- and gamma-ray photons in air in units of roentgens. In this document, exposure does not refer to the radiological physics concept of charge liberated per unit mass of air.

**gamma radiation**

Electromagnetic radiation (photons) of short wavelength and high energy (10 kiloelectron-volts to 9 megaelectron-volts) that originates in atomic nuclei and accompanies many nuclear reactions (e.g., fission, radioactive decay, and neutron capture). Gamma photons are identical to X-ray photons of high energy; the difference is that X-rays do not originate in the nucleus.

**glovebox**

Enclosure with special rubber gloves through which an operator can handle radioactive or toxic material without risk of injury or contamination normally operated at a slightly reduced pressure so that air leakage, if any, is inward.

**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%  $^{235}\text{U}$  for use as fissile material in nuclear weapons components and some reactor fuels.

**Integrated Modules for Bioassay Analysis (IMBA)**

Computer program that uses bioassay results and other information to calculate intakes of radionuclides and subsequent doses.

**intake**

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

**internal dose**

Dose received from radioactive material in the body.

**internal dose assessment**

Estimation of an intake of radioactive material and the consequent radiation dose based on bioassay or other measurements in the work environment.

***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**

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

**ionizing radiation**

Radiation of high enough energy to remove an electron from a struck atom and leave behind a positively charged ion. High enough doses of ionizing radiation can cause cellular damage. Ionizing particles include alpha particles, beta particles, gamma rays, X-rays, neutrons, high-speed electrons, high-speed protons, photoelectrons, Compton electrons, positron/negatron pairs from photon radiation, and scattered nuclei from fast neutrons.

**isotope**

One of two or more atoms of a particular element that have the same number of protons (atomic number) but different numbers of neutrons in their nuclei (e.g.,  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$ ). Isotopes have very nearly the same chemical properties.

**kiloelectron-volt (keV)**

Unit of particle energy equal to 1,000 ( $1 \times 10^3$ ) electron-volts.

**lung solubility type (F, M, or S)**

See *absorption type*.

**megaelectron-volt (MeV)**

Unit of particle energy equal to 1 million ( $1 \times 10^6$ ) electron-volts.

**minimum detectable amount (MDA)**

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

**monitoring (personnel)**

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.

**National Institute for Occupational Safety and Health (NIOSH)**

U.S. agency responsible for dose reconstruction under the Energy Employees Occupational Illness Compensation Program. Part of the Centers for Disease Control and Prevention, which is part of the U.S. Department of Health and Human Services, NIOSH is the Federal agency responsible for conducting research and making recommendations for the prevention of work-related injury and illness.

**natural uranium (U, U-nat, NU)**

Uranium as found in nature, approximately 99.27%  $^{238}\text{U}$ , 0.72%  $^{235}\text{U}$ , and 0.0054%  $^{234}\text{U}$  by mass. The specific activity of this mixture is  $2.6 \times 10^7$  becquerel per kilogram (0.7 microcurie per gram). See *uranium*.

**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.

**probability of causation (POC, PC)**

For purposes of dose reconstruction for the Energy Employees Occupational Illness Compensation Act, the percent likelihood, at the 99th percentile, that a worker incurred a particular cancer from occupational exposure to radiation.

**radiation**

Subatomic particles and electromagnetic rays (photons) with kinetic energy that interact with matter through various mechanisms that involve energy transfer. See *ionizing radiation*.

**rem**

Traditional unit of radiation dose equivalent that indicates the biological damage caused by radiation equivalent to that caused by 1 rad of high-penetration X-rays multiplied by a quality factor. The average American receives 360 millirem a year from background radiation. The sievert is the International System unit; 1 rem equals 0.01 sievert. The word derives from roentgen equivalent in man; rem is also the plural.

**routine monitoring**

Monitoring carried out at regular intervals during normal operations. See *special monitoring*.

**sievert**

International System unit for dose equivalent, which indicates the biological damage caused by radiation. The unit is the radiation value in gray (equal to 1 joule per kilogram) multiplied by a weighting factor for the type of radiation and a weighting factor for the tissue; 1 Sv equals 100 rem.

**special monitoring**

Monitoring in addition to the routine monitoring program carried out for special purposes such as estimating the amount of radionuclide deposited in a person after a known or suspected accidental intake or after a known or suspected environmental release. See *routine monitoring*.

**spot sample**

In relation to bioassay, a single void of urine.

**whole-body counter (WBC)**

Equipment used to perform *in vivo* bioassay. Radiation emitted from radioactive material deposited throughout the body is measured.

**whole-body exposure**

Exposure of the entire body to radiation rather than an isolated part. When an ingested radioisotope is uniformly distributed throughout the body tissues rather than being concentrated in certain parts, the irradiation is a whole-body exposure.

**X-ray radiation**

Electromagnetic radiation (photons) produced by bombardment of atoms by accelerated particles. X-rays are produced by various mechanisms including bremsstrahlung and electron shell transitions within atoms (characteristic X-rays). Once formed, there is no difference between X-rays and gamma rays, but gamma photons originate inside the nucleus of an atom.



**ATTACHMENT A**  
**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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Table A-1. Environmental releases by building (1993–2001).<sup>a</sup>

| (Current) building no. | Building activity   | Radionuclide | Activity fraction |
|------------------------|---|--------------|-------------------|
| 131                    | M&E Divisions   | Th-232       | 2.29E-10          |
|                        |   | U-234        | 8.47E-02          |
|                        |   | U-235        | 1.18E-02          |
|                        |   | U-238        | 9.04E-01          |
| 132                    | Analytical & Nuclear Chemistry Laboratories; Forensic Sciences Center | H-3          | 7.3E-09           |
|                        |   | Co-60        | 1.5E-07           |
|                        |   | Th-228       | 1.2E-10           |
|                        |   | Th-230       | 3.5E-10           |
|                        |   | Th-232       | 7.9E-05           |
|                        |   | U-234        | 3.1E-04           |
|                        |   | U-235        | 7.2E-03           |
|                        |   | U-238        | 9.9E-01           |
|                        |   | Pu-238       | 4.4E-03           |
|                        |   | Pu-239       | 6.7E-06           |
|                        |   | Pu-240       | 1.7E-06           |
|                        |   | Pu-241       | 1.6E-05           |
|                        |   | Pu-242       | 2.0E-10           |
|                        |   | Am-241       | 3.1E-07           |
| 151                    | Isotope Sciences, Environmental Services Laboratory                   | Ni-63        | 1.1E-01           |
|                        |   | Zn-65        | 5.1E-07           |
|                        |   | Sr-90        | 3.8E-05           |
|                        |   | Y-90         | 3.7E-06           |
|                        |   | Tc-99        | 1.3E-10           |
|                        |   | Tc-99m       | 5.3E-08           |
|                        |   | Ru-106       | 4.6E-09           |
|                        |   | Sn-113       | 1.4E-07           |
|                        |   | Sb-125       | 1.6E-06           |
|                        |   | I-129        | 4.6E-10           |
|                        |   | Ba-133       | 5.1E-08           |
|                        |   | Cs-134       | 5.1E-08           |
|                        |   | Cs-136       | 7.1E-07           |
|                        |   | Cs-137       | 9.5E-05           |
|                        |   | Ce-144       | 8.0E-07           |
|                        |   | Pm-147       | 7.6E-05           |
|                        |   | Eu-152       | 6.5E-08           |
|                        |   | Eu-154       | 6.5E-08           |
|                        |   | Eu-155       | 1.6E-12           |
|                        |   | Bi-207       | 4.2E-05           |
|                        |   | Po-209       | 1.9E-12           |
|                        |   | Ra-226       | 2.5E-08           |
|                        |   | Pa-231       | 2.7E-07           |
|                        |   | Th-228       | 1.4E-09           |
| Th-229                 | 2.2E-12   |              |                   |
| Th-230                 | 1.5E-09   |              |                   |
| Th-232                 | 6.4E-08   |              |                   |
| U-232                  | 2.7E-11   |              |                   |
| U-233                  | 1.3E-07   |              |                   |

**ATTACHMENT A**  
**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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| <b>(Current) building no.</b> | <b>Building activity</b>                            | <b>Radionuclide</b> | <b>Activity fraction</b> |
|-------------------------------|---|---------------------|--------------------------|
| 151                           | Isotope Sciences, Environmental Services Laboratory | U-234               | 2.0E-03                  |
|                               |   | U-235               | 3.0E-04                  |
|                               |   | U-236               | 7.3E-06                  |
|                               |   | U-238               | 1.9E-02                  |
|                               |   | Np-237              | 8.9E-04                  |
|                               |   | Pu-236              | 5.3E-06                  |
|                               |   | Pu-238              | 2.0E-02                  |
|                               |   | Pu-239              | 1.7E-01                  |
|                               |   | Pu-240              | 3.8E-02                  |
|                               |   | Pu-241              | 6.1E-01                  |
|                               |   | Pu-242              | 9.5E-05                  |
|                               |   | Pu-244              | 5.1E-05                  |
|                               |   | Am-241              | 1.0E-02                  |
|                               |   | Am-243              | 7.6E-04                  |
|                               |   | Cm-242              | 5.1E-06                  |
|                               |   | Cm-244              | 1.5E-05                  |
|                               |   | Cm-246              | 1.2E-05                  |
| Cm-248                        | 8.0E-06   |                     |                          |
| Cf-249                        | 1.5E-04   |                     |                          |
| 175                           | U-AVLIS   | U-234               | 3.11E-01                 |
|                               |   | U-235               | 9.50E-03                 |
|                               |   | U-238               | 6.79E-01                 |
| 177                           | U-AVLIS   | U-234               | 3.05E-01                 |
|                               |   | U-235               | 1.04E-02                 |
|                               |   | U-238               | 6.85E-01                 |
| 179                           |   | U-234               | 4.38E-01                 |
|                               |   | U-235               | 1.24E-01                 |
|                               |   | U-238               | 4.38E-01                 |
| 212                           | Physics and Space (rotating target neutron source)  | H-3                 | 1                        |
| 222                           |   | H-3                 | 6.96E-01                 |
|                               |   | C-14                | 5.44E-10                 |
|                               |   | Ni-63               | 3.04E-01                 |
|                               |   | Th-232              | 6.86E-08                 |
|                               |   | U-234               | 2.84E-08                 |
|                               |   | U-235               | 1.25E-09                 |
|                               |   | U-238               | 7.13E-06                 |
| 223                           |   | Pu-239              | 6.53E-08                 |
|                               |   | Pu-238              | 1.09E-02                 |
|                               |   | Pu-239              | 8.78E-01                 |
|                               |   | Am-241              | 1.25E-04                 |
| 226                           |   | Am-243              | 1.11E-01                 |
|                               |   | H-3                 | 1.00E+00                 |
| 227                           |   | U-238               | 9.67E-06                 |
|                               |   | U-234               | 5.05E-01                 |
|                               |   | U-235               | 2.26E-02                 |
|                               |   | U-238               | 4.73E-01                 |

**ATTACHMENT A**  
**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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| <b>(Current)<br/>building no.</b> | <b>Building activity</b>                          | <b>Radionuclide</b> | <b>Activity<br/>fraction</b> |
|-----------------------------------|---|---------------------|------------------------------|
| 231                               | Safeguards and engineering                        | Th-232              | 1.3E-02                      |
|                                   |   | U-234               | 8.5E-02                      |
|                                   |   | U-235               | 1.2E-02                      |
|                                   |   | U-238               | 8.9E-01                      |
| 235                               | Characterization studies and ion beam experiments | Th-232              | 4.0E-08                      |
|                                   |   | U-234               | 2.7E-01                      |
|                                   |   | U-235               | 1.6E-02                      |
|                                   |   | U-238               | 6.7E-01                      |
|                                   |   | Pu-238              | 3.0E-04                      |
|                                   |   | Pu-239              | 1.1E-02                      |
|                                   |   | Pu-240              | 2.5E-03                      |
|                                   |   | Pu-241              | 2.9E-02                      |
| 235                               | Characterization studies and ion beam experiments | Pu-242              | 1.7E-07                      |
|                                   |   | Am-241              | 1.4E-03                      |
| 241                               | R&D   | C-14                | 2.1E-07                      |
|                                   |   | P-32                | 8.7E-08                      |
|                                   |   | Th-232              | 1.9E-12                      |
|                                   |   | U-234               | 1.0E+00                      |
|                                   |   | U-235               | 1.8E-06                      |
|                                   |   | U-238               | 4.0E-05                      |
| 251                               | Heavy element facility                            | U-233               | 6.58E-09                     |
|                                   |   | Pu-238              | 2.17E-04                     |
|                                   |   | Pu-239              | 5.54E-09                     |
|                                   |   | Pu-243              | 1.45E-06                     |
|                                   |   | Am-241              | 3.29E-01                     |
|                                   |   | Cm-243              | 1.69E-01                     |
|                                   |   | Cm-244              | 5.02E-01                     |
|                                   |   | Cm-248              | 4.18E-05                     |
| Cf-252                            | 1.37E-05  |                     |                              |
|                                   | Laboratories and counting rooms                   | H-3                 | 1.1E-04                      |
|                                   |   | C-14                | 1.0E-07                      |
|                                   |   | P-32                | 1.0E-06                      |
|                                   |   | Sr-90               | 5.5E-06                      |
|                                   |   | Sr-90/Y-90          | 2.8E-08                      |
|                                   |   | Y-90                | 3.7E-08                      |
|                                   |   | Cs-137              | 1.4E-07                      |
|                                   |   | Bi-214              | 3.3E-01                      |
|                                   |   | Pb-214              | 3.3E-01                      |
|                                   |   | Po-218              | 3.3E-01                      |
|                                   |   | Ra-226              | 1.5E-02                      |
|                                   |   | Th-230              | 2.0E-08                      |
|                                   |   | U-234               | 3.7E-04                      |
|                                   |   | U-235               | 1.6E-05                      |
|                                   |   | U-238               | 3.5E-04                      |
|                                   |   | Np-237              | 4.3E-08                      |
|                                   |   | Pu-238              | 2.6E-06                      |
| Pu-239                            | 6.6E-05   |                     |                              |
| Pu-240                            | 1.5E-05   |                     |                              |

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| (Current) building no. | Building activity                 | Radionuclide | Activity fraction |
|------------------------|-----------------------------------|--------------|-------------------|
| 253                    |                                   | Pu-241       | 6.5E-04           |
|                        |                                   | Pu-242       | 9.9E-10           |
|                        |                                   | Am-241       | 3.1E-06           |
| 254                    | Bioassays and analytical services | H-3          | 4.4E-02           |
|                        |                                   | C-14         | 1.4E-02           |
|                        |                                   | P-32         | 4.0E-03           |
|                        |                                   | P-33         | 4.0E-01           |
|                        |                                   | S-35         | 4.1E-02           |
|                        |                                   | Sr-90        | 9.9E-02           |
|                        |                                   | Y-90         | 9.9E-02           |
|                        |                                   | I-125        | 6.3E-02           |
|                        |                                   | Po-209       | 2.6E-05           |
|                        |                                   | Th-230       | 6.0E-03           |
|                        |                                   | U-232        | 2.3E-04           |
|                        |                                   | U-233        | 1.0E-05           |
|                        |                                   | U-234        | 4.1E-04           |
|                        |                                   | U-235        | 1.0E-05           |
|                        |                                   | U-236        | 3.2E-03           |
|                        |                                   | U-238        | 8.9E-07           |
|                        |                                   | Pu-238       | 0.0E+00           |
|                        |                                   | Pu-239       | 9.0E-05           |
|                        |                                   | Pu-240       | 0.0E+00           |
|                        |                                   | Pu-241       | 0.0E+00           |
|                        |                                   | Pu-242       | 5.5E-04           |
|                        |                                   | Np-237       | 4.4E-02           |
|                        |                                   | Np-239       | 1.7E-01           |
| Am-241                 | 3.2E-05                           |              |                   |
| Am-243                 | 9.2E-04                           |              |                   |
| Cm-242                 | 2.5E-04                           |              |                   |
| Cm-244                 | 3.3E-04                           |              |                   |
| Cf-249                 | 2.5E-04                           |              |                   |
| Cf-252                 | 5.4E-03                           |              |                   |
| 255                    | Calibration laboratory            | H-3          | 1.00E+00          |
|                        |                                   | C-14         | 3.50E-10          |
|                        |                                   | P-32         | 2.80E-08          |
|                        |                                   | S-35         | 1.20E-09          |
|                        |                                   | Sr-90        | 4.89E-13          |
|                        |                                   | Y-90         | 4.89E-13          |
|                        |                                   | I-125        | 5.91E-08          |
|                        |                                   | I-131        | 1.80E-07          |
|                        |                                   | Th-230       | 9.53E-11          |
|                        |                                   | Th-232       | 2.50E-15          |
|                        |                                   | U-233        | 2.50E-10          |
|                        |                                   | U-234        | 1.10E-11          |
|                        |                                   | U-235        | 2.90E-13          |
| U-236                  | 8.69E-09                          |              |                   |
| U-238                  | 4.19E-14                          |              |                   |
| Pu-239                 | 2.77E-12                          |              |                   |

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| (Current) building no. | Building activity                                     | Radionuclide               | Activity fraction |
|------------------------|---|----------------------------|-------------------|
| 255                    |   | Pu-242                     | 1.40E-11          |
|                        |   | Np-237                     | 1.70E-09          |
|                        |   | Np-239                     | 1.70E-09          |
|                        |   | Am-241                     | 9.54E-13          |
|                        |   | Am-243                     | 2.54E-11          |
|                        |   | Cm-242                     | 9.09E-12          |
|                        |   | Cm-244                     | 9.49E-13          |
|                        |   | Cf-252                     | 8.39E-11          |
| 281                    | Tracer and dissolution studies                        | H-3                        | 3.7E-02           |
|                        |   | Be-10                      | 1.5E-02           |
|                        |   | C-14                       | 2.8E-01           |
|                        |   | Na-22                      | 1.2E-04           |
|                        |   | Cl-36                      | 1.5E-02           |
|                        |   | Ca-41                      | 1.5E-01           |
|                        |   | Mn-54                      | 4.9E-06           |
|                        |   | Ni-59                      | 1.0E-04           |
|                        |   | Ni-63                      | 3.1E-01           |
|                        |   | Co-60                      | 5.0E-06           |
|                        |   | Sr-90                      | 1.5E-02           |
|                        |   | Tc-99                      | 1.5E-04           |
|                        |   | Sb-125                     | 4.9E-08           |
|                        |   | Eu-152/Tm-171              | 1.7E-08           |
|                        |   | Eu-154/Eu-155              | 2.1E-05           |
|                        |   | Th-232                     | 1.1E-06           |
|                        |   | U-233                      | 1.9E-04           |
|                        |   | U-234                      | 1.9E-04           |
|                        |   | U-235                      | 8.1E-06           |
|                        |   | U-238                      | 1.8E-04           |
|                        |   | U-233/U-238                | 1.9E-06           |
|                        |   | Pu-239                     | 6.3E-04           |
|                        |   | Pu-242                     | 2.0E-03           |
|                        |   | Pu-244                     | 2.4E-06           |
|                        |   | Np-237                     | 1.7E-01           |
|                        |   | Am-241                     | 5.4E-04           |
| Am-241/Np-237          | 1.4E-04   |                            |                   |
| 282                    | Residual contamination                                | H-3                        | 1.0E+00           |
|                        |   | Rb-86/87                   | 7.5E-09           |
| 292                    | Residual contamination rotating target neutron source | H-3                        | 1                 |
| 298                    | Laser fusion program                                  | H-3                        | 9.9E-01           |
|                        |   | U-234                      | 4.1E-04           |
|                        |   | U-235                      | 1.9E-05           |
|                        |   | U-238                      | 6.0E-03           |
| 321                    | Milling and shaping                                   | DU<br>Thorium <sup>b</sup> |                   |
| 321A                   | Milling and shaping                                   | U-234                      | 1.8E-01           |
|                        |   | U-235                      | 2.4E-02           |
|                        |   | U-238                      | 7.9E-01           |

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| <b>(Current) building no.</b> | <b>Building activity</b>        | <b>Radionuclide</b> | <b>Activity fraction</b> |
|-------------------------------|---------------------------------|---------------------|--------------------------|
| 321C                          | Milling, machining, and shaping | U-234               | 6.5E-01                  |
|                               |                                 | U-235               | 8.1E-03                  |
|                               |                                 | U-238               | 3.4E-01                  |
| 322                           | M&E                             | U-234               | 8.1E-01                  |
|                               |                                 | U-235               | 1.2E-02                  |
|                               |                                 | U-238               | 1.8E-01                  |
| 327                           | M&E                             | U-234               | 7.7E-02                  |
|                               |                                 | U-235               | 8.8E-02                  |
|                               |                                 | U-238               | 8.4E-01                  |
| 331                           | Research and laboratories       | H-3 (HTO)           | 4.69E-01                 |
|                               |                                 | H-3 (HT)            | 5.19E-01                 |
|                               |                                 | H-3                 | 1.24E-02                 |
| 332                           | Gloveboxes, HEPA filters        | Pu-239 TRU          |                          |
| 341                           | Lasers Directorate              | U-234               | 5.70E-02                 |
|                               |                                 | U-235               | 7.69E-03                 |
|                               |                                 | U-238               | 9.35E-01                 |
| 361                           | R&D                             | H-3                 | 5.6E-06                  |
|                               |                                 | C-14                | 3.3E-03                  |
|                               |                                 | P-32                | 8.3E-01                  |
|                               |                                 | P-33                | 1.1E-01                  |
|                               |                                 | S-35                | 5.4E-02                  |
| 362                           | R&D                             | H-3                 | 7.22E-01                 |
|                               |                                 | C-14                | 2.78E-01                 |
| 363                           | R&D                             | H-3                 | 9.99E-01                 |
|                               |                                 | C-14                | 1.11E-07                 |
|                               |                                 | P-32                | 1.42E-03                 |
| 364                           | R&D                             | H-3                 | 1.11E-03                 |
|                               |                                 | C-14                | 2.76E-03                 |
|                               |                                 | P-32                | 9.96E-01                 |
| 365                           | R&D                             | H-3                 | 4.04E-03                 |
|                               |                                 | C-14                | 9.96E-01                 |
| 366                           | R&D                             | H-3                 | 5.00E-01                 |
|                               |                                 | P-32                | 4.38E-01                 |
|                               |                                 | P-33                | 6.25E-02                 |
| 377                           | R&D                             | H-3                 | 7.66E-04                 |
|                               |                                 | C-14                | 3.83E-03                 |
|                               |                                 | P-32                | 9.95E-01                 |
|                               |                                 | Ni-63               | 1.15E-04                 |
| 378                           |                                 | Co-57               | 1.44E-03                 |
|                               |                                 | Co-60               | 5.59E-02                 |
|                               |                                 | Sr-85               | 6.52E-02                 |
|                               |                                 | Cd-109              | 3.58E-02                 |
|                               |                                 | Cs-134              | 6.98E-01                 |
|                               |                                 | Cs-137              | 6.98E-02                 |
|                               |                                 | U-233               | 2.14E-04                 |
|                               |                                 | U-234               | 8.66E-07                 |
|                               |                                 | U-235               | 4.84E-08                 |
| U-238                         | 2.00E-06                        |                     |                          |

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| (Current) building no. | Building activity | Radionuclide | Activity fraction |
|------------------------|-------------------|--------------|-------------------|
| 378                    |                   | Pu-236       | 2.13E-03          |
|                        |                   | Pu-239       | 6.99E-05          |
|                        |                   | Pu-240       | 5.59E-05          |
|                        |                   | Pu-242       | 1.05E-02          |
|                        |                   | Pu-244       | 3.58E-04          |
|                        |                   | Np-237       | 1.44E-06          |
|                        |                   | Am-241       | 6.00E-02          |
|                        |                   | Am-243       | 4.01E-04          |
| 381                    |                   | H-3          | 1                 |
| 391                    |                   | H-3          | 1                 |
| 412W                   |                   | Ni-59        | 3.50E-07          |
|                        |                   | Ni-63        | 1.00E+00          |
| 446                    |                   | C-14         | 1                 |
| 513                    | Waste processing  | H-3          | 9.85E-01          |
|                        |                   | C-14         | 2.49E-04          |
|                        |                   | P-32         | 3.17E-06          |
|                        |                   | K-40         | 5.79E-04          |
|                        |                   | Mn-54        | 8.34E-06          |
|                        |                   | Co-57        | 7.28E-07          |
|                        |                   | Co-60        | 1.03E-04          |
|                        |                   | Sr-90        | 7.27E-04          |
|                        |                   | Nb-95        | 8.34E-06          |
|                        |                   | Zr-95        | 1.30E-05          |
|                        |                   | Ru-106       | 6.07E-09          |
|                        |                   | I-125        | 7.49E-07          |
|                        |                   | I-131        | 2.70E-08          |
|                        |                   | Ba-133       | 6.09E-07          |
|                        |                   | Cs-134       | 4.13E-06          |
|                        |                   | Cs-137       | 2.61E-05          |
|                        |                   | Cs-138       | 1.52E-10          |
|                        |                   | Ce-141       | 6.07E-09          |
|                        |                   | Ce-144       | 9.56E-06          |
|                        |                   | Eu-152       | 1.67E-04          |
|                        |                   | Eu-154       | 2.12E-07          |
|                        |                   | Eu-155       | 3.79E-08          |
|                        |                   | Tl-208       | 6.22E-08          |
|                        |                   | Bi-212       | 8.50E-08          |
|                        |                   | Bi-214       | 5.92E-09          |
|                        |                   | Pb-210       | 7.89E-06          |
|                        |                   | Pb-212       | 1.43E-07          |
|                        |                   | Pb-214       | 2.12E-08          |
| Ra-223                 | 4.55E-07          |              |                   |
| Ra-226                 | 8.65E-08          |              |                   |
| Ra-228                 | 4.55E-16          |              |                   |
| Ac-228                 | 1.52E-09          |              |                   |
| Pa-231                 | 8.50E-07          |              |                   |
| Th-226                 | 9.71E-07          |              |                   |
| Th-227                 | 1.67E-06          |              |                   |

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| (Current)<br>building no. | Building activity | Radionuclide | Activity<br>fraction |
|---------------------------|-------------------|--------------|----------------------|
| 513                       |                   | Th-228       | 4.04E-05             |
|                           |                   | Th-232       | 3.51E-06             |
|                           |                   | Th-234       | 3.03E-06             |
|                           |                   | U-233        | 1.52E-05             |
|                           |                   | U-234        | 1.08E-03             |
|                           |                   | U-235        | 1.52E-04             |
|                           |                   | U-238        | 1.04E-02             |
|                           |                   | Pu-238       | 1.74E-09             |
|                           |                   | Pu-239       | 2.28E-04             |
|                           |                   | Pu-240       | 2.03E-08             |
|                           |                   | Pu-241       | 1.27E-06             |
|                           |                   | Pu-242       | 6.87E-07             |
|                           |                   | Am-241       | 7.62E-04             |
|                           |                   | Cm-244       | 3.03E-05             |
|                           |                   | Cf-249       | 2.28E-07             |
| 514                       | Waste processing  | H-3          | 8.61E-01             |
|                           |                   | Be-7         | 7.53E-06             |
|                           |                   | C-14         | 2.40E-02             |
|                           |                   | Na-22        | 9.05E-05             |
|                           |                   | P-32         | 6.53E-02             |
|                           |                   | S-35         | 8.42E-03             |
|                           |                   | K-40         | 6.11E-05             |
|                           |                   | Sc-46        | 5.46E-06             |
|                           |                   | Cr-51        | 5.59E-06             |
|                           |                   | Fe-55        | 5.42E-06             |
|                           |                   | Mn-54        | 1.66E-05             |
|                           |                   | Co-56        | 1.82E-07             |
|                           |                   | Co-57        | 7.56E-05             |
|                           |                   | Co-58        | 5.46E-06             |
|                           |                   | Co-60        | 1.09E-04             |
|                           |                   | Ni-63        | 1.45E-07             |
|                           |                   | Zn-65        | 1.92E-08             |
|                           |                   | Y-88         | 5.33E-05             |
|                           |                   | Sr-89        | 4.96E-11             |
|                           |                   | Sr-90        | 9.13E-04             |
|                           |                   | Nb-94        | 1.19E-06             |
|                           |                   | Nb-95        | 8.46E-06             |
|                           |                   | Zr-95        | 1.20E-07             |
|                           |                   | Tc-99        | 6.05E-05             |
|                           |                   | Ru-103       | 2.18E-09             |
|                           |                   | Ru-106       | 1.33E-06             |
|                           |                   | Cd-109       | 2.89E-08             |
|                           |                   | Sb-125       | 2.08E-05             |
| I-125                     | 1.39E-04          |              |                      |
| I-131                     | 3.63E-06          |              |                      |
| Ba-133                    | 1.02E-04          |              |                      |
| Cs-134                    | 6.37E-05          |              |                      |
| Cs-137                    | 1.16E-03          |              |                      |



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| <b>(Current) building no.</b> | <b>Building activity</b> | <b>Radionuclide</b> | <b>Activity fraction</b> |
|-------------------------------|--------------------------|---------------------|--------------------------|
| 514                           | Waste processing         | Ce-139              | 7.14E-13                 |
|                               |                          | Ce-141              | 1.82E-09                 |
|                               |                          | Ce-144              | 4.87E-03                 |
|                               |                          | Gd-148              | 6.04E-05                 |
|                               |                          | Pm-147              | 3.07E-06                 |
|                               |                          | Sm-151              | 2.45E-07                 |
|                               |                          | Eu-152              | 2.29E-04                 |
|                               |                          | Eu-154              | 2.32E-04                 |
|                               |                          | Eu-155              | 2.54E-05                 |
|                               |                          | Hf-172              | 7.33E-06                 |
|                               |                          | Lu-173              | 1.92E-06                 |
|                               |                          | Lu-174              | 5.42E-06                 |
|                               |                          | W-185               | 5.62E-09                 |
|                               |                          | Po-209              | 9.30E-06                 |
|                               |                          | Po-210              | 9.30E-06                 |
|                               |                          | Bi-207              | 4.71E-06                 |
|                               |                          | Bi-210              | 7.14E-07                 |
|                               |                          | Pb-210              | 5.62E-05                 |
|                               |                          | Ra-226              | 9.73E-06                 |
|                               |                          | Th-228              | 1.47E-05                 |
|                               |                          | Th-229              | 6.04E-05                 |
|                               |                          | Th-230              | 5.81E-05                 |
|                               |                          | Th-232              | 7.87E-05                 |
|                               |                          | U-232               | 6.08E-05                 |
|                               |                          | U-233               | 7.01E-03                 |
|                               |                          | U-234               | 3.60E-03                 |
|                               |                          | U-235               | 2.18E-04                 |
|                               |                          | U-236               | 8.92E-07                 |
|                               |                          | U-237               | 1.82E-09                 |
|                               |                          | U-238               | 8.29E-03                 |
|                               |                          | Pu-236              | 3.67E-08                 |
|                               |                          | Pu-238              | 4.72E-04                 |
|                               |                          | Pu-239              | 6.74E-03                 |
| Pu-240                        | 1.68E-03                 |                     |                          |
| Pu-241                        | 1.49E-04                 |                     |                          |
| Pu-242                        | 1.60E-04                 |                     |                          |
| Pu-244                        | 1.09E-06                 |                     |                          |
| Np-237                        | 6.77E-05                 |                     |                          |
| Np-239                        | 2.41E-06                 |                     |                          |
| Am-241                        | 4.23E-03                 |                     |                          |
| Am-243                        | 8.33E-05                 |                     |                          |
| Cm-244                        | 6.76E-05                 |                     |                          |
| Cf-249                        | 9.30E-06                 |                     |                          |
| 514 Tank Farm                 |                          | H-3                 | 5.45E-01                 |
|                               |                          | Be-7                | 3.33E-07                 |
|                               |                          | C-14                | 2.59E-03                 |
|                               |                          | Na-22               | 5.74E-05                 |
|                               |                          | P-32                | 1.01E-01                 |

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| (Current)<br>building no. | Building activity | Radionuclide | Activity<br>fraction |
|---------------------------|-------------------|--------------|----------------------|
| 514 Tank Farm             |                   | P-33         | 1.86E-04             |
|                           |                   | S-35         | 6.39E-03             |
|                           |                   | K-40         | 5.16E-05             |
|                           |                   | Sc-46        | 1.30E-06             |
|                           |                   | Cr-51        | 4.79E-07             |
|                           |                   | Fe-55        | 6.65E-07             |
|                           |                   | Fe-59        | 1.53E-06             |
|                           |                   | Mn-54        | 2.88E-06             |
|                           |                   | Co-56        | 2.01E-05             |
|                           |                   | Co-57        | 6.68E-03             |
|                           |                   | Co-58        | 2.14E-05             |
|                           |                   | Co-60        | 4.51E-05             |
|                           |                   | Zn-65        | 9.48E-07             |
|                           |                   | Ni-59        | 1.33E-06             |
|                           |                   | Ni-63        | 4.09E-06             |
|                           |                   | Y-88         | 2.12E-05             |
|                           |                   | Y-91         | 5.66E-06             |
|                           |                   | Sr-90        | 2.05E-01             |
|                           |                   | Zr-95        | 1.40E-03             |
|                           |                   | Nb-95        | 3.08E-05             |
|                           |                   | Mo-99        | 7.99E-06             |
|                           |                   | Tc-99        | 2.76E-05             |
|                           |                   | Ru-103       | 2.54E-06             |
|                           |                   | Ru-106       | 4.35E-06             |
|                           |                   | Cd-109       | 8.97E-08             |
|                           |                   | Sn-113       | 4.66E-09             |
|                           |                   | Ag-110m      | 8.65E-07             |
|                           |                   | I-125        | 5.59E-05             |
|                           |                   | I-131        | 1.06E-05             |
|                           |                   | Sb-120m      | 7.32E-07             |
|                           |                   | Sb-124       | 6.25E-06             |
|                           |                   | Sb-125       | 9.93E-06             |
|                           |                   | Te-132       | 2.00E-07             |
|                           |                   | Ba-133       | 4.99E-05             |
| Ba-140                    | 9.99E-07          |              |                      |
| Cs-134                    | 1.42E-05          |              |                      |
| Cs-136                    | 9.31E-07          |              |                      |
| Cs-137                    | 1.08E-01          |              |                      |
| La-140                    | 2.00E-08          |              |                      |
| Ce-139                    | 3.99E-09          |              |                      |
| Ce-141                    | 2.93E-05          |              |                      |
| Ce-144                    | 1.87E-04          |              |                      |
| Nd-147                    | 4.46E-06          |              |                      |
| Pm-147                    | 2.40E-06          |              |                      |
| Gd-148                    | 1.28E-05          |              |                      |
| Sm-151                    | 7.99E-08          |              |                      |
| Eu-152                    | 1.82E-04          |              |                      |
| Eu-154                    | 1.19E-04          |              |                      |

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**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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| <b>(Current) building no.</b> | <b>Building activity</b>      | <b>Radionuclide</b> | <b>Activity fraction</b> |
|-------------------------------|-------------------------------|---------------------|--------------------------|
| 514 Tank Farm                 |                               | Eu-155              | 1.48E-05                 |
|                               |                               | Eu-156              | 1.13E-07                 |
|                               |                               | Tb-160              | 1.20E-08                 |
|                               |                               | Hf-172              | 1.93E-06                 |
|                               |                               | Hf-181              | 2.66E-06                 |
|                               |                               | Lu-173              | 1.60E-06                 |
|                               |                               | Lu-174              | 3.33E-07                 |
|                               |                               | W-185               | 4.79E-09                 |
|                               |                               | Au-195              | 4.26E-06                 |
|                               |                               | Hg-203              | 3.06E-20                 |
|                               |                               | Bi-207              | 1.04E-06                 |
|                               |                               | Bi-210              | 5.06E-07                 |
|                               |                               | Po-209              | 4.50E-06                 |
|                               |                               | Po-210              | 5.06E-07                 |
|                               |                               | Pb-210              | 1.36E-05                 |
|                               |                               | Ra-226              | 1.55E-04                 |
|                               |                               | Pa-233              | 1.60E-09                 |
|                               |                               | Th-228              | 6.02E-03                 |
|                               |                               | Th-229              | 1.28E-05                 |
|                               |                               | Th-230              | 1.27E-05                 |
|                               |                               | Th-232              | 1.14E-04                 |
|                               |                               | U-232               | 1.31E-05                 |
|                               |                               | U-233               | 4.44E-05                 |
|                               |                               | U-234               | 1.63E-03                 |
|                               |                               | U-235               | 7.36E-04                 |
|                               |                               | U-236               | 3.33E-07                 |
|                               |                               | U-237               | 3.46E-04                 |
|                               |                               | U-238               | 1.02E-02                 |
|                               |                               | Pu-236              | 3.46E-08                 |
|                               |                               | Pu-238              | 1.01E-04                 |
|                               |                               | Pu-239              | 1.50E-03                 |
|                               |                               | Pu-240              | 1.48E-04                 |
|                               |                               | Pu-241              | 3.86E-05                 |
|                               |                               | Pu-242              | 4.37E-05                 |
| Pu-244                        | 7.16E-08                      |                     |                          |
| Np-237                        | 2.31E-05                      |                     |                          |
| Np-239                        | 1.01E-04                      |                     |                          |
| Am-241                        | 1.02E-03                      |                     |                          |
| Am-243                        | 1.97E-05                      |                     |                          |
| Cm-244                        | 4.12E-05                      |                     |                          |
| Cf-249                        | 4.63E-06                      |                     |                          |
| 612                           | Waste storage and repackaging | H-3                 | 9.79E-01                 |
|                               |                               | Be-7                | 1.22E-06                 |
|                               |                               | C-14                | 7.04E-04                 |
|                               |                               | Na-22               | 1.87E-04                 |
|                               |                               | P-32                | 1.29E-02                 |
|                               |                               | P-33                | 1.05E-08                 |
|                               |                               | S-35                | 6.98E-06                 |

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| (Current)<br>building no. | Building activity             | Radionuclide | Activity<br>fraction |
|---------------------------|-------------------------------|--------------|----------------------|
| 612                       | Waste storage and repackaging | Cl-36        | 3.66E-11             |
|                           |                               | K-40         | 2.03E-05             |
|                           |                               | Sc-46        | 1.25E-06             |
|                           |                               | Cr-51        | 1.34E-06             |
|                           |                               | Fe-55        | 1.23E-06             |
|                           |                               | Mn-54        | 4.20E-06             |
|                           |                               | Co-56        | 7.33E-08             |
|                           |                               | Co-57        | 5.01E-06             |
|                           |                               | Co-58        | 1.34E-06             |
|                           |                               | Co-60        | 1.07E-03             |
|                           |                               | Ni-63        | 4.89E-08             |
|                           |                               | Zn-65        | 1.22E-08             |
|                           |                               | Se-75        | 2.88E-11             |
|                           |                               | Y-88         | 1.45E-07             |
|                           |                               | Y-91         | 1.05E-11             |
|                           |                               | Sr-85        | 8.72E-13             |
|                           |                               | Sr-89        | 3.93E-12             |
|                           |                               | Sr-90        | 6.02E-04             |
|                           |                               | Nb-94        | 5.50E-05             |
|                           |                               | Nb-95        | 1.34E-06             |
|                           |                               | Zr-95        | 1.28E-07             |
|                           |                               | Mo-99        | 1.55E-09             |
|                           |                               | Tc-99        | 1.60E-05             |
|                           |                               | Rh-102       | 2.01E-10             |
|                           |                               | Rh-103m      | 4.10E-13             |
|                           |                               | Ru-106       | 2.58E-08             |
|                           |                               | Cd-109       | 4.02E-05             |
|                           |                               | Cd-115       | 5.25E-09             |
|                           |                               | Ag-110m      | 2.18E-14             |
|                           |                               | I-125        | 2.45E-05             |
|                           |                               | I-131        | 3.15E-09             |
|                           |                               | Sb-124       | 4.45E-14             |
|                           |                               | Sb-125       | 4.99E-06             |
|                           |                               | Sm-151       | 5.10E-11             |
| Ba-133                    | 1.76E-05                      |              |                      |
| Ba-140                    | 7.64E-09                      |              |                      |
| Cs-134                    | 1.58E-05                      |              |                      |
| Cs-137                    | 5.11E-04                      |              |                      |
| Ce-139                    | 2.44E-13                      |              |                      |
| Ce-141                    | 2.85E-08                      |              |                      |
| Ce-144                    | 1.52E-04                      |              |                      |
| Nd-147                    | 1.59E-09                      |              |                      |
| Pm-147                    | 5.95E-10                      |              |                      |
| Pm-151                    | 4.73E-10                      |              |                      |
| Sm-151                    | 8.72E-13                      |              |                      |

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**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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| <b>(Current)<br/>building no.</b> | <b>Building activity</b>      | <b>Radionuclide</b> | <b>Activity<br/>fraction</b> |
|-----------------------------------|-------------------------------|---------------------|------------------------------|
| 612                               | Waste storage and repackaging | Gd-146              | 1.05E-05                     |
|                                   |                               | Gd-148              | 1.57E-05                     |
|                                   |                               | Eu-149              | 2.79E-12                     |
|                                   |                               | Eu-152              | 6.28E-05                     |
|                                   |                               | Eu-154              | 6.10E-05                     |
|                                   |                               | Eu-155              | 5.54E-06                     |
|                                   |                               | Tb-160              | 1.74E-13                     |
|                                   |                               | Hf-172              | 2.44E-06                     |
|                                   |                               | Lu-173              | 1.22E-06                     |
|                                   |                               | Lu-174              | 1.22E-06                     |
|                                   |                               | Ta-182              | 1.40E-11                     |
|                                   |                               | W-185               | 3.66E-09                     |
|                                   |                               | Ir-192              | 8.72E-14                     |
|                                   |                               | Au-195              | 5.41E-12                     |
|                                   |                               | Pt-195m             | 9.59E-10                     |
|                                   |                               | Hg-203              | 8.72E-17                     |
|                                   |                               | Bi-207              | 1.17E-06                     |
|                                   |                               | Bi-210              | 2.44E-07                     |
|                                   |                               | Po-209              | 3.30E-06                     |
|                                   |                               | Po-210              | 2.44E-07                     |
|                                   |                               | Pb-210              | 1.68E-05                     |
|                                   |                               | Ra-223              | 4.73E-12                     |
|                                   |                               | Ra-226              | 1.72E-06                     |
|                                   |                               | Ra-228              | 2.94E-09                     |
|                                   |                               | Th-228              | 1.45E-06                     |
|                                   |                               | Th-229              | 1.57E-05                     |
|                                   |                               | Th-230              | 1.56E-05                     |
|                                   |                               | Th-232              | 1.48E-05                     |
|                                   |                               | Th-234              | 1.13E-12                     |
|                                   |                               | U-232               | 1.58E-05                     |
|                                   |                               | U-233               | 2.08E-05                     |
|                                   |                               | U-234               | 4.93E-04                     |
|                                   |                               | U-235               | 1.85E-04                     |
|                                   |                               | U-237               | 3.39E-08                     |
|                                   |                               | U-238               | 2.01E-03                     |
|                                   |                               | U-239               | 9.60E-12                     |
|                                   |                               | Pu-238              | 1.15E-04                     |
|                                   |                               | Pu-239              | 3.89E-04                     |
|                                   |                               | Pu-240              | 3.54E-05                     |
|                                   |                               | Pu-241              | 1.28E-04                     |
| Pu-242                            | 2.60E-05                      |                     |                              |
| Pu-244                            | 3.56E-06                      |                     |                              |
| Np-237                            | 1.69E-05                      |                     |                              |
| Np-239                            | 6.09E-07                      |                     |                              |
| Am-241                            | 3.74E-04                      |                     |                              |
| Am-242                            | 6.81E-09                      |                     |                              |
| Am-242m                           | 1.57E-09                      |                     |                              |
| Am-243                            | 4.11E-04                      |                     |                              |

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**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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| <b>(Current) building no.</b> | <b>Building activity</b>      | <b>Radionuclide</b> | <b>Activity fraction</b> |
|-------------------------------|-------------------------------|---------------------|--------------------------|
| 612                           | Waste storage and repackaging | Am-244              | 4.45E-06                 |
|                               |                               | Cm-244              | 1.05E-04                 |
|                               |                               | Cf-249              | 3.39E-06                 |
|                               |                               | Cf-250              | 2.24E-12                 |
| 612 Yard                      |                               | H-3                 | 1.00E+00                 |
|                               |                               | C-14                | 2.11E-06                 |
|                               |                               | P-32                | 1.61E-07                 |
|                               |                               | S-35                | 3.40E-08                 |
|                               |                               | K-40                | 4.48E-11                 |
|                               |                               | Cr-51               | 4.84E-11                 |
|                               |                               | Mn-54               | 6.11E-13                 |
|                               |                               | Co-57               | 6.38E-13                 |
|                               |                               | Co-60               | 1.19E-10                 |
|                               |                               | Ni-59               | 1.01E-12                 |
|                               |                               | Ni-63               | 2.92E-11                 |
|                               |                               | Se-75               | 2.45E-08                 |
|                               |                               | Sr-90               | 9.84E-12                 |
|                               |                               | Nb-95               | 3.19E-13                 |
|                               |                               | Tc-99               | 2.44E-12                 |
|                               |                               | Sb-125              | 4.09E-12                 |
|                               |                               | Cs-134              | 1.02E-11                 |
|                               |                               | Cs-137              | 3.60E-10                 |
|                               |                               | Ce-144              | 4.25E-10                 |
|                               |                               | Pm-147              | 1.70E-12                 |
|                               |                               | Sm-151              | 5.26E-13                 |
|                               |                               | Eu-152              | 5.45E-10                 |
|                               |                               | Eu-154              | 2.89E-10                 |
|                               |                               | Eu-155              | 6.11E-12                 |
|                               |                               | Bi-207              | 1.81E-13                 |
|                               |                               | Bi-214              | 4.52E-14                 |
|                               |                               | Ra-226              | 3.65E-11                 |
|                               |                               | Ra-228              | 1.22E-11                 |
|                               |                               | Th-228              | 5.50E-10                 |
|                               |                               | Th-230              | 1.17E-14                 |
|                               |                               | Th-232              | 5.69E-10                 |
|                               |                               | Th-234              | 1.17E-11                 |
|                               |                               | U-233               | 5.40E-10                 |
|                               |                               | U-234               | 1.87E-08                 |
| U-235                         | 2.47E-09                      |                     |                          |
| U-238                         | 1.87E-07                      |                     |                          |
| Pu-238                        | 2.43E-09                      |                     |                          |
| Pu-239                        | 5.46E-10                      |                     |                          |
| Pu-240                        | 2.12E-11                      |                     |                          |
| Pu-241                        | 4.43E-09                      |                     |                          |
| Pu-242                        | 1.02E-08                      |                     |                          |
| Np-239                        | 4.80E-11                      |                     |                          |
| Am-241                        | 1.45E-09                      |                     |                          |
| Am-242                        | 1.38E-12                      |                     |                          |

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| (Current) building no. | Building activity | Radionuclide | Activity fraction |
|------------------------|-------------------|--------------|-------------------|
| 612 Yard               |                   | Am-243       | 4.22E-10          |
|                        |                   | Cm-243       | 4.89E-11          |
|                        |                   | Cm-244       | 3.35E-13          |
|                        |                   | Cm-245       | 3.03E-11          |
| 625                    | Waste operations  | H-3          | 2.14E-01          |
|                        |                   | C-14         | 2.34E-04          |
|                        |                   | P-32         | 3.61E-10          |
|                        |                   | K-40         | 8.78E-05          |
|                        |                   | Mn-54        | 1.36E-06          |
|                        |                   | Co-57        | 5.74E-08          |
|                        |                   | Co-60        | 9.89E-06          |
|                        |                   | Sr-90        | 2.34E-06          |
|                        |                   | Y-88         | 1.70E-10          |
|                        |                   | Zr-95        | 2.55E-06          |
|                        |                   | Ru-106       | 7.01E-07          |
|                        |                   | Cd-109       | 4.40E-08          |
|                        |                   | Sb-125       | 9.78E-07          |
|                        |                   | Ba-133       | 1.01E-07          |
|                        |                   | Cs-134       | 1.11E-06          |
|                        |                   | Cs-137       | 1.05E-05          |
|                        |                   | Ce-141       | 4.46E-09          |
|                        |                   | Ce-144       | 1.57E-06          |
|                        |                   | Eu-152       | 4.47E-06          |
|                        |                   | Eu-154       | 2.55E-06          |
|                        |                   | Eu-155       | 2.55E-07          |
|                        |                   | Bi-214       | 2.98E-08          |
|                        |                   | Pb-212       | 3.83E-08          |
|                        |                   | Pb-214       | 4.89E-09          |
|                        |                   | Pa-231       | 4.89E-08          |
|                        |                   | Ra-226       | 1.51E-05          |
|                        |                   | Ra-228       | 6.59E-04          |
|                        |                   | Th-228       | 1.55E-06          |
|                        |                   | Th-230       | 5.53E-10          |
|                        |                   | Th-232       | 3.62E-06          |
|                        |                   | Th-234       | 4.89E-04          |
|                        |                   | U-233        | 4.46E-08          |
|                        |                   | U-234        | 2.76E-01          |
|                        |                   | U-235        | 4.89E-01          |
|                        |                   | U-238        | 2.32E-03          |
|                        |                   | U-239        | 2.55E-07          |
| Pu-238                 | 7.68E-05          |              |                   |
| Pu-239                 | 3.40E-03          |              |                   |
| Pu-240                 | 3.83E-06          |              |                   |
| Pu-241                 | 1.30E-02          |              |                   |
| Pu-242                 | 4.46E-08          |              |                   |
| Np-237                 | 2.34E-07          |              |                   |
| Am-241                 | 2.87E-04          |              |                   |
| Am-243                 | 5.10E-06          |              |                   |

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**ENVIRONMENTAL RELEASES BY BUILDING (1993–2001)**  
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| (Current) building no. | Building activity | Radionuclide | Activity fraction |
|------------------------|-------------------|--------------|-------------------|
| 625                    | Waste operations  | Cm-243       | 9.78E-09          |
|                        |                   | Cm-244       | 2.34E-09          |
|                        |                   | Cm-245       | 1.38E-09          |
| 2561                   |                   | U-234        | 5.00E-01          |
|                        |                   | U-238        | 5.00E-01          |
|                        |                   | U-234        | 8.01E-02          |
|                        |                   | U-235        | 1.12E-02          |
|                        |                   | U-238        | 8.66E-01          |
| Site 300-801           |                   | H-3          | 1                 |
| Site 300 Pit 7         |                   | H-3          | 1                 |
| Site 300-810A          |                   | U-234        | 8.38E-02          |
|                        |                   | U-235        | 1.16E-02          |
|                        |                   | U-238        | 9.05E-01          |
| Site 300-810B          |                   | U-234        | 8.47E-02          |
|                        |                   | U-235        | 1.13E-02          |
|                        |                   | U-238        | 9.04E-01          |
| Site 300-850           |                   | H-3          | 9.6E-01           |
|                        |                   | U-234        | 3.2E-03           |
|                        |                   | U-235        | 4.4E-04           |
|                        |                   | U-238        | 3.4E-02           |
| Site 300-851           |                   | H-3          | 9.57E-01          |
|                        |                   | U-234        | 9.57E-04          |
|                        |                   | U-235        | 1.34E-04          |
|                        |                   | U-238        | 1.02E-02          |
| Site 300 Well 8 Spring |                   | H-3          | 1                 |

- a. This table was derived from data reported for calendar years 1992 through 2001 as part of the National Emission Standards for Hazardous Air Pollutants (NESHAPs) (40 CFR Part 61, Subpart H) reporting process (Harrach et al. 1994; Surano et al. 1995; Gallegos et al. 1996; Gallegos and Biermann 1997; Gallegos et al. 1998; Biermann et al. 1999; Gallegos et al. 2000, 2001; Harrach et al. 2002, 2003). As part of its required reporting under NESHAPs, LLNL must monitor emissions by radionuclide and total activity at all building release points. This table lists only buildings for which NESHAPs data were reported; this is not a complete list of all buildings. For a listing of major buildings and activities, see ORAUT (2005b). To develop the table, the isotopic listing of each building in the relevant NESHAPs reports was captured in a building-specific spreadsheet. The annual emissions were totaled for each isotope and then for all isotopes in the listing. Activity fractions were obtained by dividing the Activity total by the "all isotopes" total (Berger 2005). The Activity fraction for each radionuclide in the table is that fraction of the total activity released to the environment (i.e., the sum of the fractions for each building is equal to "1" or 100% of the mixture). However, those radionuclides with Activity fractions equal to "zero", those with half-lives too short to contribute significant dose (i.e., isotopes of nitrogen and oxygen), and those that contribute submersion dose only were subsequently deleted from the listing.
- b. LLNL (1987b).



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**B1.      OVERVIEW**

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

Bioassay results for LLNL were obtained as a copy of the site’s MAPPER database converted to an Excel spreadsheet, along with a cover letter explaining characteristics of the database and pointing out possibilities of data recording errors that might exist in the database (Mansfield 2006a). As sent to the ORAU Team, the MAPPER database had approximately 34,900 records. It contained no *in vivo* counting results and had only a few of the many tritium urinalysis results obtained throughout the operation of the Laboratory. The *in vivo* counting results were not available electronically and, therefore, were not analyzed as a coworker database. (*In vivo* results for individual Energy Employees are included in the files sent to NIOSH from LLNL.)

The principal radionuclides with potential for intakes at LLNL were tritium, isotopes of plutonium in a weapons-grade mixture, isotopes of uranium in mixtures typical of depleted uranium, natural uranium or slightly enriched uranium (to 5%), various transuranic radionuclides, activation products, and fission products (ORAUT 2005b). The MAPPER database had small numbers of analyses for other radionuclides, such as <sup>241</sup>Am, curium, and transuranic (TRU) materials; however, the numbers were too small for statistical analysis as an individual coworker data set. Generally, LLNL used the gross alpha analysis to screen for intakes of TRU radionuclides with follow-up analyses being more specific for the radionuclide of concern.

The urinalysis data chosen for this coworker study because of general applicability and number of measurements were for plutonium, uranium, gross alpha, gross beta, and an analysis labeled MFP (mixed fission product). Two previous bioassay laboratory managers confirmed that the gross beta

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results and the MFP results were from the same procedure and should be treated as the same (Bihl 2006b; Mansfield 2006b).

The statistical analyses of the bioassay data for each radionuclide were performed in accordance with the internal dosimetry coworker technical information bulletin (ORAUT 2005a) and the procedure, ORAUT-PROC-0095, *Generating Summary Statistics for Coworker Bioassay Data* (ORAUT 2006b). The resultant values were input to the Integrated Modules for Bioassay Analysis (IMBA) computer program and a fit to the data for each of the four analyzed groups at the 50th- and 84th-percentile values was performed to obtain intake rates for assignment of dose distributions. The 50th-percentile intakes were converted to 95th-percentile intakes based on the following methodology:

$$95\text{th-percentile intake} = 50\text{th-percentile intake} \times \text{GSD}^{1.645}$$

The tables listing the 50th- and 95th-percentile intakes are the values that the dose reconstructor should use when assigning coworker intakes. The 50th-percentile intakes are assigned as a lognormal distribution with the associated GSD listed in each table. The 95th-percentile intakes are assigned as constant distribution.

Coworker intakes can be extended out to later years for aged fission products and research related radionuclides from 1994 to 1995, plutonium from 1991 to 1996, uranium from 1988 to 1996, and americium from 1971 to 1996. Building 251 transitioned from Category II Nonreactor Nuclear Facility in 2002 to a radiological facility in 2005 where equipment and inventory were removed (LLNL 2006a). During this transition of Building 251, there were no major contamination incidents, no radiation over-exposures (in fact, doses were far lower than dose predictions). Individual and collective doses were maintained ALARA. Collective annual whole body doses were at least three times lower than ALARA goals and more than 10 times lower than conservation dose projections. Individual annual external whole body doses were less than 150 mrem (LLNL 2006b). Based on this information about Building 251, extending coworker intakes out to later years is applicable.

Attachment C provides guidance on assigning the 50th- or 95th-percentile intakes based on job category, and level of contact with the radioactive material.

Tables B-1 through B-4 list bioassay sample data statistics for the coworker intakes determined.

Table B-1. Americium coworker sample statistic data.

| Year    | Number of samples | Number of employees | Year    | Number of samples | Number of employees |
|---------|-------------------|---------------------|---------|-------------------|---------------------|
| 1957    | 89                | 78                  | 1979-81 | 127               | 90                  |
| 1958    | 125               | 84                  | 1982    | 117               | 64                  |
| 1959    | 133               | 83                  | 1983    | 127               | 64                  |
| 1960    | 88                | 62                  | 1984    | 140               | 65                  |
| 1961    | 116               | 79                  | 1985    | 157               | 92                  |
| 1962    | 121               | 61                  | 1986    | 210               | 110                 |
| 1963    | 98                | 59                  | 1987    | 160               | 96                  |
| 1964    | 134               | 83                  | 1988    | 141               | 81                  |
| 1965    | 139               | 78                  | 1989    | 165               | 79                  |
| 1966-67 | 83                | 54                  | 1990    | 173               | 91                  |
| 1968    | 89                | 58                  | 1991    | 205               | 104                 |
| 1969    | 106               | 68                  | 1992    | 220               | 119                 |
| 1970    | 122               | 85                  | 1993    | 319               | 144                 |

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|         |     |    |      |     |     |
|---------|-----|----|------|-----|-----|
| 1971-72 | 78  | 35 | 1994 | 325 | 138 |
| 1973-75 | 51  | 35 | 1995 | 300 | 130 |
| 1976-78 | 112 | 75 | 1996 | 173 | 99  |

The MAPPER database was used for the determination of the coworker study datasets, and not the individual claims received by DOL. The cover letter from LLNL transmitting the MAPPER database identified 11 potential issues with the data (Mansfield 2006a). These issues are addressed below.

- **Employee Identification:** While it is helpful to have information linking all of an individual's samples for a given radionuclide for reviewing the data, investigating outliers, and ensuring that there is no bias due to a few individuals, errors in a fraction of the identifiers will not affect the coworker statistics.

Table B-2. Beta coworker sample statistic data.

| Year    | Number of samples | Number of employees | Year | Number of samples | Number of employees |
|---------|-------------------|---------------------|------|-------------------|---------------------|
| 1957-63 | 50                | 41                  | 1986 | 265               | 137                 |
| 1964-65 | 75                | 60                  | 1987 | 227               | 143                 |
| 1966-73 | 43                | 42                  | 1988 | 225               | 123                 |
| 1974-75 | 112               | 57                  | 1989 | 218               | 103                 |
| 1976    | 70                | 35                  | 1990 | 250               | 125                 |
| 1977    | 96                | 46                  | 1991 | 298               | 136                 |
| 1978-79 | 144               | 75                  | 1992 | 287               | 152                 |
| 1980-81 | 148               | 91                  | 1993 | 407               | 179                 |
| 1982    | 120               | 69                  | 1994 | 357               | 153                 |
| 1983    | 152               | 79                  | 1995 | 298               | 126                 |
| 1984    | 200               | 103                 | 1996 | 171               | 98                  |
| 1985    | 199               | 112                 |      |                   |                     |

Table B-3. Plutonium coworker sample statistic data.

| Year | Number of samples | Number of employees | Year | Number of samples | Number of employees |
|------|-------------------|---------------------|------|-------------------|---------------------|
| 1957 | 90                | 79                  | 1977 | 169               | 107                 |
| 1958 | 125               | 84                  | 1978 | 140               | 102                 |
| 1959 | 133               | 83                  | 1979 | 119               | 76                  |
| 1960 | 88                | 62                  | 1980 | 118               | 80                  |
| 1961 | 116               | 79                  | 1981 | 194               | 141                 |
| 1962 | 121               | 61                  | 1982 | 314               | 185                 |
| 1963 | 126               | 73                  | 1983 | 340               | 180                 |
| 1964 | 140               | 87                  | 1984 | 402               | 205                 |
| 1965 | 154               | 82                  | 1985 | 384               | 214                 |
| 1966 | 102               | 57                  | 1986 | 464               | 253                 |
| 1967 | 84                | 57                  | 1987 | 450               | 268                 |
| 1968 | 112               | 66                  | 1988 | 492               | 285                 |
| 1969 | 133               | 84                  | 1989 | 529               | 300                 |
| 1970 | 133               | 87                  | 1990 | 399               | 233                 |
| 1971 | 182               | 111                 | 1991 | 377               | 212                 |
| 1972 | 175               | 95                  | 1992 | 443               | 260                 |
| 1973 | 106               | 75                  | 1993 | 522               | 261                 |
| 1974 | 113               | 83                  | 1994 | 493               | 244                 |
| 1975 | 137               | 85                  | 1995 | 447               | 223                 |

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| Year | Number of samples | Number of employees |
|------|-------------------|---------------------|
| 1976 | 135               | 94                  |

| Year | Number of samples | Number of employees |
|------|-------------------|---------------------|
| 1996 | 252               | 168                 |

- **Sample Dates:** Sample dates are used only in grouping results together into data sets. The shortest statistical analysis interval is 3 months, so the day of the month that a sample was collected is not necessary information for the coworker study.
- **Analyte:** Obvious typographical errors in the analyte were corrected. A list of samples numbers associated with likely erroneous results was provided in the cover letter transmitting the MAPPER database by the site and those involving  $^{238}\text{Pu}$  were not included in the  $^{239}\text{Pu}$  analysis. Tritium coworker data were not evaluated.
- **Results Less Than Zero:** The cover letter notes that these obvious problems were corrected before sending the database to the ORAU Team.

Table B-4. Uranium coworker sample statistic data.

| Year    | Number of samples | Number of employees |
|---------|-------------------|---------------------|
| 1958    | 74                | 67                  |
| 1959–61 | 87                | 70                  |
| 1962–64 | 96                | 44                  |
| 1965    | 107               | 88                  |
| 1966    | 113               | 90                  |
| 1967    | 51                | 31                  |
| 1968    | 141               | 87                  |
| 1969    | 95                | 92                  |
| 1970    | 118               | 101                 |
| 1971    | 35                | 33                  |
| 1972    | 272               | 172                 |
| 1973    | 172               | 138                 |
| 1974    | 139               | 120                 |
| 1975 S1 | 146               | 134                 |
| 1975 S2 | 159               | 128                 |
| 1976 S1 | 139               | 136                 |
| 1976 S2 | 95                | 92                  |
| 1977 S1 | 142               | 133                 |
| 1977 S2 | 151               | 122                 |
| 1978 S1 | 155               | 148                 |
| 1978 S2 | 131               | 120                 |
| 1979 S1 | 213               | 149                 |
| 1979 S2 | 204               | 163                 |
| 1980 S1 | 199               | 108                 |
| 1980 S2 | 141               | 109                 |
| 1981 S1 | 157               | 126                 |
| 1981 S2 | 168               | 131                 |
| 1982 S1 | 245               | 163                 |
| 1982 S2 | 271               | 194                 |
| 1983 S1 | 278               | 195                 |
| 1983 S2 | 317               | 219                 |
| 1984 S1 | 286               | 191                 |

| Year    | Number of samples | Number of employees |
|---------|-------------------|---------------------|
| 1987 Q2 | 254               | 171                 |
| 1987 Q3 | 221               | 139                 |
| 1987 Q4 | 224               | 151                 |
| 1988 Q1 | 273               | 150                 |
| 1988 Q2 | 256               | 154                 |
| 1988 Q3 | 236               | 129                 |
| 1988 Q4 | 175               | 119                 |
| 1989 Q1 | 175               | 126                 |
| 1989 Q2 | 195               | 147                 |
| 1989 Q3 | 188               | 131                 |
| 1989 Q4 | 230               | 146                 |
| 1990 Q1 | 211               | 142                 |
| 1990 Q2 | 192               | 121                 |
| 1990 Q3 | 210               | 120                 |
| 1990 Q4 | 238               | 141                 |
| 1991 Q1 | 298               | 187                 |
| 1991 Q2 | 286               | 153                 |
| 1991 Q3 | 284               | 143                 |
| 1991 Q4 | 214               | 134                 |
| 1992 Q1 | 313               | 204                 |
| 1992 Q2 | 326               | 197                 |
| 1992 Q3 | 279               | 168                 |
| 1992 Q4 | 278               | 174                 |
| 1993 Q1 | 332               | 202                 |
| 1993 Q2 | 304               | 188                 |
| 1993 Q3 | 324               | 201                 |
| 1993 Q4 | 316               | 200                 |
| 1994 Q1 | 247               | 183                 |
| 1994 Q2 | 152               | 122                 |
| 1994 Q3 | 148               | 120                 |
| 1994 Q4 | 120               | 98                  |
| 1995 Q1 | 189               | 153                 |

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| Year    | Number of samples | Number of employees |
|---------|-------------------|---------------------|
| 1984 S2 | 230               | 165                 |
| 1985 S1 | 219               | 159                 |
| 1985 S2 | 213               | 153                 |
| 1986 S1 | 238               | 178                 |
| 1986 S2 | 257               | 194                 |
| 1987 Q1 | 220               | 153                 |

| Year    | Number of samples | Number of employees |
|---------|-------------------|---------------------|
| 1995 Q2 | 114               | 98                  |
| 1995 Q3 | 151               | 110                 |
| 1995 Q4 | 194               | 146                 |
| 1996 Q1 | 197               | 122                 |
| 1996 Q2 | 354               | 206                 |
| 1996 Q3 | 182               | 134                 |

- **Result Error:** Result error has no impact on a coworker analysis; all results are used as reported.
- **Units:** As noted by LLNL, the errors were largely typographical errors and the correct values are apparent.
- **Media:** There are only eight samples with a media code = R or S; all were assumed to be urine samples. The sample with a media code of B was not included in the analyses nor were the two samples with a result of 1747.8, both because the same sample number was attributed to two individuals and because the result appears to be far outside the range of other urine values, indicating a very unusual circumstance or error.
- **Priority:** Priority of a sample is irrelevant to the coworker analysis.
- **Exposure Area:** Because there are limited samples available for the site, no analysis by work area was performed. This field is therefore not used in the study.
- **Result Status:** All comments were reviewed and samples indicated to be invalid (e.g., spiked, cross-contaminated) were excluded from the analysis.
- **Assumptions about Sample Volume:** Discussions were held with LLNL personnel, including the former and present bioassay laboratory managers, regarding this issue. A favorable to claimant resolution was adopted; see the discussion in the coworker study for details.

### B2. LIMITATIONS

NIOSH has determined, with concurrence from the Secretary of HHS, that mixed fission product doses at LLNL cannot be reconstructed between 1950 and 1973, inclusive. For this reason, a class of LLNL employees has been added to the SEC. Therefore, only a limited assessment of internal dose can be performed before 1974 for workers monitored for some type of radiation exposures under the EEOICPA (Leavitt 2008). If monitoring data are available for workers included in the SEC, dose is to be assigned as appropriate based on available monitoring data. However, such dose reconstructions are still considered partial dose reconstructions.

### B3. DISCUSSION OF THE DATASETS

#### B3.1 URANIUM

The MAPPER database contained uranium urinalysis data from 1958 to 1996. Nearly all of the results were reported as  $^{238}\text{U}$  in units of  $\mu\text{g/L}$ . The LLNL cover letter indicates that the results actually represent total uranium (Mansfield 2006a). According to Section 5.2, most uranium exposure was to

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depleted or natural uranium. The AVLIS project converted kilogram quantities of natural uranium to slightly enriched uranium, up to 5%, as a demonstration plant. The uranium used for the AVLIS project came from Fernald. The AVLIS project operated in Buildings 175, 177, and the 482 complex from 1973 through 1999. HEU which came from Y-12 was used through the site's history in only a few selected buildings. HEU was used in Building 231, and the Oralloy Shop in Building 321C. There was another project involving highly enriched uranium in the 1990s, involving the United States and Russian HEU Purchase Agreement. The program monitors the Russian process of converting weapons-usable HEU into low enriched uranium (LEU). A Transparency Monitoring Office in Russia was established in 1996 by DOE and is staffed in part by Livermore workers. When identified in MAPPER, results from workers on these projects were excluded from the uranium dataset because of the higher specific activity of uranium on that project (31 results). Uranium results were left in units of  $\mu\text{g/L}$  for the statistical analysis and curve fitting.

The uranium dataset has no negative (i.e.,  $<0$ ) results. MDAs for the uranium analysis methods were not provided in the LLNL cover letter. Table 5-6 lists the MDA as  $5 \mu\text{g/L}$  through 1973,  $1 \mu\text{g/L}$  for 1974 through 1978,  $0.3 \mu\text{g/L}$  for 1979 through 1989, and  $0.05 \mu\text{g/L}$  for 1990 through 1999. Most of the results less than  $1 \mu\text{g/L}$  were recorded as zero before 1974. In 1974, the database shows a distinctly different pattern, with an overall decrease in the general magnitude of the results, a lower reporting level at around  $0.01 \mu\text{g/L}$ , and sufficient results between  $0.01$  to  $1 \mu\text{g/L}$  to support a reasonably normal or lognormal pattern implying that a change to a more sensitive analysis technique took place. The zero results were included in the ranking of the data but not the curve fitting. In several years before 1974, there were a few results recorded with values between  $1$  and  $0 \mu\text{g/L}$ . Because there were just a few of these and not in all years, they seemed to be anomalous recordings relative to the  $1\text{-}\mu\text{g/L}$  reporting level. Because they might have been misrecorded data, such as misplaced decimals or incorrect unit conversion, and because they were clearly anomalous when included in the lognormal plots of the results, these results were excluded from the final annual datasets. There were 37 of these results throughout the period from 1962 through 1973, as compared to 1200 results used for the coworker study during this period.

The uranium dataset had entries from recounts on the same sample and entries showing multiple same-day samples from a given worker. The recounted samples were identified as having identical information, including sample number, but different results and different uncertainty. The multiple same-day samples were indicated by having the same worker identification and sample dates, however, the sample numbers were different, often by one digit. Because two counts on a single sample or two samples from a given worker on the same day produces a bias when compared to other workers' samples that were given only a single count, the multicounted samples were replaced by the average of the multiple counts.

Duplicate entries that were obvious and samples marked by LLNL as baselines or quality control samples were excluded.

Table B-5 lists the time intervals for separate statistical analyses. Intervals were chosen to obtain approximately 100 results or more per interval; however, 1967 had only 51 results and 1971 had only 34 results. (The median and 84th-percentile values for 1971 were also much lower than surrounding years, perhaps indicating that there was little work with uranium at the site in 1971.) The statistical parameters for each interval (e.g., 50th and 84th percentiles) were assigned to the midpoint of the interval.

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Table B-5. Time intervals for statistical analysis of uranium urine samples.

| Calendar year | Analysis interval            |
|---------------|------------------------------|
| 1958          | Year                         |
| 1959–1961     | Treated as a single interval |
| 1962–1964     | Treated as a single interval |
| 1965–1974     | Year                         |
| 1975–1986     | 6 months                     |
| 1987–1996     | 3 months                     |

### B3.2 GROSS ALPHA, PLUTONIUM, AND AMERICIUM/CURIUM

In one analytical procedure used at LLNL, plutonium, americium, neptunium, curium, actinium, and thorium were extracted from the urine matrix and counted by a gross alpha technique. This is referred to as a “gross alpha” analysis because any activity measured in the urine sample was an indication that one or more of these radionuclides was present. In MAPPER these are called “ALPHA” results.

In another procedure the activity was separated from its urine matrix with anion exchange resin. Plutonium, americium, and curium were retained on the resin, with the plutonium subsequently eluted off. This plutonium fraction of the eluate is referred to as “PU239” in MAPPER and in this report. Pulse height analysis was used for plutonium analysis starting in 1967 or 1968 (the first  $^{238}\text{Pu}$  results show up in 1968) (ORAUT 2006b), so as of 1968 a PU239 result is interpreted to mean  $^{239+240}\text{Pu}$ .

In an interview (Bihl 2006b) with the former bioassay laboratory manager, it was pointed out that the eluate from the anion exchange column that did not contain plutonium was often collected and analyzed by a gross alpha counting technique. This effluent would contain americium and curium and the results were also reported as ALPHA in MAPPER. Note that the two different ALPHA analyses in MAPPER potentially report different radionuclides, so to avoid confusion this analysis is referred to here as “AM/CM” while the results from the procedure described in the first paragraph are referred to as “GA.” A pair of PU239 and AM/CM analyses is referred to as a “split” analysis. There was no reliable way to determine from the ALPHA results in MAPPER which procedure was used, although it is likely that all splits used the anion exchange procedure (Bihl 2006b) and this will be assumed here.

In summary, there are three types of analytical results evaluated here:

- GA – a nonspecific analysis that can potentially report any combination of plutonium, americium, neptunium, curium, actinium, and thorium.
- PU239 – an analysis specific for  $^{239}\text{Pu}$ . Might or might not have an associated AM/CM result.
- AM/CM – a nonspecific analysis that can potentially report any combination of alpha-emitting isotopes of americium and curium. Always has an associated PU239 result.
- AM241 – an analysis that is specific for  $^{241}\text{Am}$ .

And, just to make things clear

ALPHA – an analyte type in MAPPER that could be either GA or AM/CM.

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All PU239 and AM/CM urinalysis results were recorded in units of dpm, which, according to the LLNL cover letter, should be interpreted as dpm/24 hours with the exception of samples that were split for AM/CM and PU239 analyses (Mansfield 2006a) (see additional discussion on splits below). Most GA urinalysis results were recorded in units of dpm, which should also be interpreted as dpm/24 hours. Some GA sample results in 1957 and 1958 had units of cpm; because the appropriate conversion factor between cpm and dpm was not known, the cpm results were excluded from this analysis.

There were no negative (less than zero) PU239 or AM/CM results in the dataset, but numerous results were recorded as zero and some small positive results that were below the reported MDA for the time. For example, in 1990, the recorded PU239 results descend rather orderly down to 0.001 dpm, then there are 3 results at  $1.11 \times 10^{-7}$ ,  $9.3 \times 10^{-8}$ , and  $7.7 \times 10^{-9}$  dpm. All of these types of extremely small results were indicated in the database as not detected (code 99); however, when included as recorded they distorted the curve fitting and so were treated as zero for curve fitting purposes. (Another way to look at this problem is that it appears that most of the time results below 0.001 dpm were recorded as zero, but the policy was not rigorous and some of these very small results were entered. It is also possible these samples were analyzed by a method different from all the others and so were recorded differently.) Thus, all PU239 and AM/CM results less than 0.001 dpm were censored in the analysis (i.e., the rank of the sample was represented in the probability plot but the numerical value was not in fitting the lognormal distribution to data for each time interval). There were no negative gross alpha results, except in 1996. A lower recording level of about 0.01 dpm appears to have been used fairly consistently throughout (1957–1995) with a few results recorded in the 0.001- to 0.009-dpm range intermittently throughout the years. As with PU239, all ALPHA results equal to or less than 0.001 dpm were censored in the analysis.

Starting in 1976, some samples were split and given two analyses, one recorded as PU239 and one recorded as ALPHA (called AM/CM here). According to the LLNL cover letter (Mansfield 2006a), the recorded results were for the split sample, not the total sample, and to obtain a 24-hour result the split-sample results should be doubled. However, in a conference call interview with the former bioassay laboratory manager, present bioassay laboratory manager, and other LLNL personnel, the former manager said that during her time in the laboratory plutonium/alpha split samples were reported as "per total sample volume" (Bihl 2006b). The present bioassay laboratory manager confirmed that the interpretation in the cover letter (that splits should be considered as representing half the volume) was correct for recent times (Mansfield 2006a; Bihl 2006b). No one was sure of the exact time when the change in policy occurred. There was agreement that the change occurred in the mid- to late 1980s. The former bioassay laboratory manager was present in the bioassay lab until 1986 or 1987. As a consequence, the data were analyzed twice: once with split sample results being doubled starting in 1987 and the other with split sample results being doubled starting in 1988. Review of the plutonium data was inconclusive, but continuity of the geometric means for the gross alpha results was better for the 1988 assumption. Thus, a PU239 and an ALPHA result in MAPPER on the same day are considered to be "splits." Splits have different sample numbers and the ALPHA (AM/CM) portion of the split does not contain PU239. Thus, all PU239 and ALPHA (AM/CM) results from splits reported from 1988 to 1996 were multiplied by a factor of 2.

In later years, it appeared that many samples were recounted and some workers had multiple same-day samples. Multiple results on the same day are "recounts" if they have the same sample number. The final result for a recount is the mean of the multiple results, which is assigned to one sample, discarding the others. Multiple results on the same day are partial samples if they have different sample numbers. These results are assumed to be expressed in dpm per partial sample, so the final result for this sample is the sum of the results, which is assigned to one sample, discarding the others.



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Incident samples are problematic for performing group statistics and applying group median values to unmonitored workers because of the large number of samples that often were obtained for a single individual after a known acute intake. LLNL sent a list of known large acute intakes or intakes that occurred to LLNL employees while working at other sites; this list was used to remove sample results from known acute intakes of plutonium. There were 19 samples excluded from the ALPHA results from a non-DOE facility. There were 25 samples excluded from the PU239 results from other DOE sites. There were 18 samples excluded from the PU239 results from wounds. Other sample results removed from the dataset before analysis included duplicate entries and those marked as invalid samples for some reason.

For statistical analysis the data were broken down into two categories:

- All PU239 and GA results are considered to be  $^{239}\text{Pu}$ .
- All AM/CM, AM-241, and GA results are considered to be americium/curium.

Note that the GA results play a dual role here, being considered to measure  $^{239}\text{Pu}$  on one hand and americium/curium on the other. In general, the time interval for separate statistical analyses was the calendar year with the goal of having about 100 or more results per analysis period. There were sufficient  $^{239}\text{Pu}$  results to permit analyses for each year from 1957 to 1996, but a number of years had to be combined for the americium/curium: 1966 + 1967, 1971 + 1972, 1973 + 1974 + 1975, 1976 + 1977 + 1978, and 1979 + 1980 + 1981. The results of the statistical analysis (the 50th- and 84th-percentile excretion rates) were assigned to the midpoint of the analysis interval, which is July 1 for an annual interval.

### B3.3 GROSS BETA AND MFP

There is only one procedure in the 1979 bioassay procedures manual for gross beta analysis, "Gross Beta Determination of Fission Products in Urine," and, according to the former bioassay laboratory, that was the only procedure used (Miller 1979; Bihl 2006b). As stated above, the gross beta and MFP results in MAPPER were treated as a single dataset. The gross beta procedure consisted of wet-ashing to white salts, dissolution in nitric acid, coprecipitation with ammonium hydroxide, plating on stainless-steel planchets, and gross beta counting.

MAPPER had gross beta or MFP data starting in 1957. In general, the results were recorded as dpm or dpm/L, but some results were recorded as cpm, cpm/L,  $\mu\text{Ci}$ , or  $\mu\text{Ci/L}$ . The cpm and cpm/L results were excluded from the statistical analysis to avoid having to estimate the cpm-to-dpm conversion factor. It was also not certain that the unit was entered correctly; that is, the magnitude of the result might have been correct, but the unit should have been entered as dpm. The  $\mu\text{Ci}$  and  $\mu\text{Ci/L}$  samples were also excluded because the magnitudes of the results were not obviously consistent with the  $\mu\text{Ci}$  unit and error in the recording of the units was considered possible. Results reported as dpm/L were converted to dpm/24-hour sample by multiplying by 1.4 L/d reference excretion (ICRP 1975).

The only negative numbers in the database were recorded in 1996. Negative numbers and zeros were included in the ranking of the results but were not included in the fitting of the lognormal curve.

Starting in 1984, there were situations where two samples were listed for the same worker in the same day. Some of these had the same sample numbers but different results. These were assumed to be recounts on the same sample. To reduce bias from multiple counts on a single sample, the average of the same-day results were used. Also starting in 1984, there were situations where two or

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more samples were listed for the same worker in the same day but had different sample numbers. It is not known if these were actually different voidings from the worker or different aliquots of the same sample. To reduce bias, these were reduced to a single result. If the results were reported as dpm/L, the average of the two results was used before normalization to 24-hour samples; if the results were reported as dpm, the two results were added to get the total dpm/day.

Duplicate entries that were obvious and samples marked by LLNL as baselines or quality control samples were excluded. No samples were excluded due to known large acute intakes. There was a single high outlier in the dataset in 1985, which appeared to be an error (5,718,000 dpm/L without a follow-up sample or recount and marked as "negative" in the MAPPER field that shows the dosimetrist's disposition of the result). This result was excluded.

The initial statistical analyses on the gross beta results resulted in poor fits and large geometric standard deviations (GSDs) for many of the years. This situation appeared to be due in part because there was a scattering of results between 1 to 0 dpm/d that were not consistent with a lognormal or even a normal distribution. These results were few in number and were scattered over several decades (e.g., a few between 0.1 to 1 dpm and a few between 0.01 to 0.1 dpm). An analysis of the 99-code break points (i.e., the bioassay result below which almost all samples were labeled as nondetections) for the gross beta analyses implied that the decision level for the analysis was in the tens of dpm/day before 1982 and hundreds of dpm/L for 1982 through 1996. Because these random, small results were well below the detection or decision level and appeared to be outliers relative to the rest of the distribution, all results less than 1 dpm/d were treated as if they were zero. As a consequence, they were included in the ranking but were not part of the fitting of the lognormal curve.

The entire dataset for 1996 was suspect. The 1996 dataset had a definite low bias, with negative numbers running into the thousands and with a total of 46 results <0, 101 results recorded exactly as zero, and only 4 results >0. The distribution for this year was the mirror image of a lognormal distribution. Statistical parameters were not generated for this year.

Table B-6 lists the time intervals for separate statistical analyses. Intervals were chosen to obtain approximately 100 results or more per interval (although in some years consideration was given to how many results were >0). Before 1974, there were too few samples to perform meaningful statistics so the intakes were determined only for 1974 through 1995. The statistical parameters for each interval (e.g., 50th and 84th percentiles) were assigned to the midpoint of the interval.

Table B-6. Time intervals for statistical analysis of gross beta urine samples.

| Calendar year | Analysis interval   |
|---------------|---|
| 1957–1973     | Intakes not determined (168 samples total for the 17-yr period) |
| 1974–1975     | Treated as a single interval                                    |
| 1976          | Year (70 samples)   |
| 1977          | Year  |
| 1978–1979     | Treated as a single interval                                    |
| 1980–1981     | Treated as a single interval                                    |
| 1982–1995     | Year  |
| 1996          | Not analyzed  |

The procedure did not state exactly which elements would have been quantitatively captured on the planchet. This question was posed to a Hanford radiochemist; his reply was, "The LLNL procedure

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will recover and measure nearly everything except <sup>40</sup>K, <sup>137</sup>Cs, tritium, carbon, sulfur, and halogens. It will recover and measure strontium, barium, radium, most of the transition metals, all the rare earths, and all the actinides. Some of the transition metals that form strong ammine complexes probably are not completely recovered by this method; they include cobalt, nickel, copper, zinc, and silver. Nickel is probably not recovered at all. Iron should be recovered and measured completely. Ruthenium is probably mostly recovered by this method" (Soderquist 2007).

**B3.4 ANALYSIS**

For each of the radionuclide groups above, a lognormal distribution for the data in the intervals specified in Tables B-1 and B-2 was assumed (ORAUT 2006b). The 50th- and 84th-percentile values were calculated using the method described in ORAUT (2006a). Table B-7 shows the statistical analysis results for uranium.

Table B-7. Uranium urine bioassay data for 50th and 84th percentiles in units of µg/L and mg/day along with the period and effective bioassay date, 1958 through 1996.

| Period               | Effective bioassay date | 50th <sup>a</sup> (µg/L) | 84th <sup>b</sup> (µg/L) | 50th <sup>a</sup> (mg/day) | 84th <sup>b</sup> (mg/day) | Employees monitored |
|----------------------|-------------------------|--------------------------|--------------------------|----------------------------|----------------------------|---------------------|
| 1958                 | 6/30/1958               | 6.89E+00                 | 1.39E+01                 | 9.64E-03                   | 1.95E-02                   | 67                  |
| 1959-61              | 6/30/1960               | 6.13E+00                 | 1.48E+01                 | 8.58E-03                   | 2.07E-02                   | 70                  |
| 1962-64              | 6/30/1963               | 5.77E+00                 | 1.35E+01                 | 8.08E-03                   | 1.89E-02                   | 44                  |
| 1965                 | 6/30/1965               | 7.61E+00                 | 1.54E+01                 | 1.07E-02                   | 2.16E-02                   | 88                  |
| 1966                 | 6/30/1966               | 1.62E+00                 | 6.24E+00                 | 2.26E-03                   | 8.74E-03                   | 90                  |
| 1967                 | 6/30/1967               | 6.91E+00                 | 1.40E+01                 | 9.67E-03                   | 1.96E-02                   | 31                  |
| 1968                 | 6/30/1968               | 5.42E+00                 | 9.34E+00                 | 7.58E-03                   | 1.31E-02                   | 87                  |
| 1969                 | 6/30/1969               | 3.68E+00                 | 7.73E+00                 | 5.15E-03                   | 1.08E-02                   | 92                  |
| 1970                 | 6/30/1970               | 1.91E+00                 | 5.41E+00                 | 2.68E-03                   | 7.58E-03                   | 101                 |
| 1971                 | 6/30/1971               | 9.16E-01                 | 3.89E+00                 | 1.28E-03                   | 5.45E-03                   | 33                  |
| 1972                 | 6/30/1972               | 8.32E+00                 | 2.03E+01                 | 1.16E-02                   | 2.84E-02                   | 172                 |
| 1973                 | 6/30/1973               | 4.31E+00                 | 1.33E+01                 | 6.04E-03                   | 1.87E-02                   | 138                 |
| 1974                 | 6/30/1974               | 4.57E-02                 | 2.61E-01                 | 6.41E-05                   | 3.66E-04                   | 120                 |
| 1975 S1 <sup>c</sup> | 3/31/1975               | 6.25E-02                 | 1.87E-01                 | 8.75E-05                   | 2.62E-04                   | 134                 |
| 1975 S2 <sup>c</sup> | 9/30/1975               | 7.56E-02                 | 2.95E-01                 | 1.06E-04                   | 4.13E-04                   | 128                 |
| 1976 S1 <sup>c</sup> | 3/31/1976               | 3.08E-02                 | 2.09E-01                 | 4.32E-05                   | 2.93E-04                   | 136                 |
| 1976 S2 <sup>c</sup> | 9/30/1976               | 2.75E-02                 | 1.79E-01                 | 3.86E-05                   | 2.50E-04                   | 92                  |
| 1977 S1 <sup>c</sup> | 3/31/1977               | 2.11E-02                 | 1.03E-01                 | 2.95E-05                   | 1.44E-04                   | 133                 |
| 1977 S2 <sup>c</sup> | 9/30/1977               | 2.42E-02                 | 1.90E-01                 | 3.39E-05                   | 2.66E-04                   | 122                 |
| 1978 S1 <sup>c</sup> | 3/31/1978               | 3.27E-02                 | 1.65E-01                 | 4.58E-05                   | 2.32E-04                   | 148                 |
| 1978 S2 <sup>c</sup> | 9/30/1978               | 3.32E-02                 | 1.72E-01                 | 4.64E-05                   | 2.40E-04                   | 120                 |
| 1979 S1 <sup>c</sup> | 3/31/1979               | 2.14E-02                 | 1.07E-01                 | 2.99E-05                   | 1.50E-04                   | 149                 |
| 1979 S2 <sup>c</sup> | 9/30/1979               | 3.71E-02                 | 1.74E-01                 | 5.19E-05                   | 2.44E-04                   | 163                 |
| 1980 S1 <sup>c</sup> | 3/31/1980               | 2.48E-02                 | 2.75E-01                 | 3.47E-05                   | 3.85E-04                   | 108                 |
| 1980 S2 <sup>c</sup> | 9/30/1980               | 8.37E-03                 | 6.44E-02                 | 1.17E-05                   | 9.02E-05                   | 109                 |
| 1981 S1 <sup>c</sup> | 3/31/1981               | 9.34E-03                 | 8.28E-02                 | 1.31E-05                   | 1.16E-04                   | 126                 |
| 1981 S2 <sup>c</sup> | 9/30/1981               | 8.42E-02                 | 3.66E-01                 | 1.18E-04                   | 5.12E-04                   | 131                 |
| 1982 S1 <sup>c</sup> | 3/31/1982               | 3.96E-02                 | 1.98E-01                 | 5.55E-05                   | 2.77E-04                   | 163                 |
| 1982 S2 <sup>c</sup> | 9/30/1982               | 8.18E-02                 | 3.61E-01                 | 1.15E-04                   | 5.06E-04                   | 194                 |
| 1983 S1 <sup>c</sup> | 3/31/1983               | 2.84E-02                 | 2.23E-01                 | 3.98E-05                   | 3.12E-04                   | 195                 |
| 1983 S2 <sup>c</sup> | 9/30/1983               | 3.69E-02                 | 2.04E-01                 | 5.17E-05                   | 2.85E-04                   | 219                 |

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| Period               | Effective bioassay date | 50th <sup>a</sup> (µg/L) | 84th <sup>b</sup> (µg/L) | 50th <sup>a</sup> (mg/day) | 84th <sup>b</sup> (mg/day) | Employees monitored |
|----------------------|-------------------------|--------------------------|--------------------------|----------------------------|----------------------------|---------------------|
| 1984 S1 <sup>c</sup> | 3/31/1984               | 2.78E-02                 | 1.55E-01                 | 3.90E-05                   | 2.17E-04                   | 191                 |
| 1984 S2 <sup>c</sup> | 9/30/1984               | 4.76E-02                 | 1.59E-01                 | 6.66E-05                   | 2.22E-04                   | 165                 |
| 1985 S1 <sup>c</sup> | 3/31/1985               | 3.33E-02                 | 2.18E-01                 | 4.66E-05                   | 3.05E-04                   | 159                 |
| 1985 S2 <sup>c</sup> | 9/30/1985               | 2.86E-02                 | 1.10E-01                 | 4.00E-05                   | 1.54E-04                   | 153                 |
| 1986 S1 <sup>c</sup> | 3/31/1986               | 2.01E-02                 | 1.37E-01                 | 2.82E-05                   | 1.92E-04                   | 178                 |
| 1986 S2 <sup>c</sup> | 9/30/1986               | 5.12E-03                 | 5.04E-02                 | 7.16E-06                   | 7.05E-05                   | 194                 |
| 1987 Q1 <sup>c</sup> | 2/15/1987               | 6.01E-03                 | 5.42E-02                 | 8.41E-06                   | 7.59E-05                   | 153                 |
| 1987 Q2 <sup>c</sup> | 5/15/1987               | 1.12E-02                 | 6.62E-02                 | 1.56E-05                   | 9.27E-05                   | 171                 |
| 1987 Q3 <sup>c</sup> | 8/15/1987               | 2.04E-02                 | 1.05E-01                 | 2.85E-05                   | 1.47E-04                   | 139                 |
| 1987 Q4 <sup>c</sup> | 11/15/1987              | 1.90E-02                 | 1.23E-01                 | 2.66E-05                   | 1.72E-04                   | 151                 |
| 1988 Q1 <sup>c</sup> | 2/15/1988               | 1.38E-02                 | 1.15E-01                 | 1.93E-05                   | 1.61E-04                   | 150                 |
| 1988 Q2 <sup>c</sup> | 5/15/1988               | 6.78E-02                 | 1.58E-01                 | 9.49E-05                   | 2.22E-04                   | 154                 |
| 1988 Q3 <sup>c</sup> | 8/15/1988               | 6.88E-02                 | 1.66E-01                 | 9.64E-05                   | 2.32E-04                   | 129                 |
| 1988 Q4 <sup>c</sup> | 11/15/1988              | 1.04E-01                 | 2.75E-01                 | 1.45E-04                   | 3.84E-04                   | 119                 |
| 1989 Q1 <sup>c</sup> | 2/15/1989               | 7.95E-02                 | 1.88E-01                 | 1.11E-04                   | 2.63E-04                   | 126                 |
| 1989 Q2 <sup>c</sup> | 5/15/1989               | 1.04E-01                 | 2.58E-01                 | 1.46E-04                   | 3.61E-04                   | 147                 |
| 1989 Q3 <sup>c</sup> | 8/15/1989               | 7.69E-02                 | 2.16E-01                 | 1.08E-04                   | 3.02E-04                   | 131                 |
| 1989 Q4 <sup>c</sup> | 11/15/1989              | 1.04E-01                 | 2.83E-01                 | 1.46E-04                   | 3.97E-04                   | 146                 |
| 1990 Q1 <sup>c</sup> | 2/15/1990               | 5.56E-02                 | 1.42E-01                 | 7.78E-05                   | 1.99E-04                   | 142                 |
| 1990 Q2 <sup>c</sup> | 5/15/1990               | 4.67E-02                 | 1.19E-01                 | 6.54E-05                   | 1.67E-04                   | 121                 |
| 1990 Q3 <sup>c</sup> | 8/15/1990               | 4.64E-02                 | 1.02E-01                 | 6.49E-05                   | 1.42E-04                   | 120                 |
| 1990 Q4 <sup>c</sup> | 11/15/1990              | 5.33E-02                 | 8.95E-02                 | 7.46E-05                   | 1.25E-04                   | 141                 |
| 1991 Q1 <sup>c</sup> | 2/15/1991               | 7.23E-02                 | 1.35E-01                 | 1.01E-04                   | 1.89E-04                   | 187                 |
| 1991 Q2 <sup>c</sup> | 5/15/1991               | 8.44E-02                 | 1.59E-01                 | 1.18E-04                   | 2.22E-04                   | 153                 |
| 1991 Q3 <sup>c</sup> | 8/15/1991               | 6.28E-02                 | 1.11E-01                 | 8.79E-05                   | 1.55E-04                   | 143                 |
| 1991 Q4 <sup>c</sup> | 11/15/1991              | 3.92E-02                 | 7.68E-02                 | 5.48E-05                   | 1.08E-04                   | 134                 |
| 1992 Q1 <sup>c</sup> | 2/15/1992               | 3.84E-02                 | 7.15E-02                 | 5.37E-05                   | 1.00E-04                   | 204                 |
| 1992 Q2 <sup>c</sup> | 5/15/1992               | 3.67E-02                 | 6.04E-02                 | 5.13E-05                   | 8.45E-05                   | 197                 |
| 1992 Q3 <sup>c</sup> | 8/15/1992               | 4.19E-02                 | 7.38E-02                 | 5.86E-05                   | 1.03E-04                   | 168                 |
| 1992 Q4 <sup>c</sup> | 11/15/1992              | 3.11E-02                 | 4.56E-02                 | 4.35E-05                   | 6.38E-05                   | 174                 |
| 1993 Q1 <sup>c</sup> | 2/15/1993               | 3.58E-02                 | 5.45E-02                 | 5.01E-05                   | 7.63E-05                   | 202                 |
| 1993 Q2 <sup>c</sup> | 5/15/1993               | 4.38E-02                 | 6.67E-02                 | 6.14E-05                   | 9.34E-05                   | 188                 |
| 1993 Q3 <sup>c</sup> | 8/15/1993               | 4.34E-02                 | 6.85E-02                 | 6.07E-05                   | 9.58E-05                   | 201                 |
| 1993 Q4 <sup>c</sup> | 11/15/1993              | 5.26E-02                 | 7.10E-02                 | 7.36E-05                   | 9.94E-05                   | 200                 |
| 1994 Q1 <sup>c</sup> | 2/15/1994               | 4.76E-02                 | 6.45E-02                 | 6.67E-05                   | 9.03E-05                   | 183                 |
| 1994 Q2 <sup>c</sup> | 5/15/1994               | 4.32E-02                 | 6.54E-02                 | 6.05E-05                   | 9.16E-05                   | 122                 |
| 1994 Q3 <sup>c</sup> | 8/15/1994               | 5.07E-02                 | 7.46E-02                 | 7.09E-05                   | 1.04E-04                   | 120                 |
| 1994 Q4 <sup>c</sup> | 11/15/1994              | 5.34E-02                 | 7.86E-02                 | 7.47E-05                   | 1.10E-04                   | 98                  |
| 1995 Q1 <sup>c</sup> | 2/15/1995               | 6.47E-02                 | 9.70E-02                 | 9.06E-05                   | 1.36E-04                   | 153                 |
| 1995 Q2 <sup>c</sup> | 5/15/1995               | 5.13E-02                 | 7.84E-02                 | 7.18E-05                   | 1.10E-04                   | 98                  |
| 1995 Q3 <sup>c</sup> | 8/15/1995               | 4.63E-02                 | 6.61E-02                 | 6.48E-05                   | 9.26E-05                   | 110                 |
| 1995 Q4 <sup>c</sup> | 11/15/1995              | 4.71E-02                 | 7.43E-02                 | 6.59E-05                   | 1.04E-04                   | 146                 |
| 1996 Q1 <sup>c</sup> | 2/15/1996               | 5.24E-02                 | 9.03E-02                 | 7.34E-05                   | 1.26E-04                   | 122                 |
| 1996 Q2 <sup>c</sup> | 5/15/1996               | 4.86E-02                 | 8.39E-02                 | 6.81E-05                   | 1.18E-04                   | 206                 |
| 1996 Q3 <sup>c</sup> | 8/15/1996               | 5.53E-02                 | 8.90E-02                 | 7.75E-05                   | 1.25E-04                   | 134                 |

- a. 50th-percentile value of the fitted line.  
b. 84th-percentile value of the fitted line.

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- c. S1 means first 6 months of the year, S2 means second 6 months of the year, Q1 means months 1-3 of the year, Q2 means months 4-6 of the year, Q3 means months 7-9 of the year, Q4 means months 10-12 of the year.

### **B4. INTAKE MODELING**

#### **B4.1 ASSUMPTIONS**

All urinalysis results were assumed to be representative of a full-day (24-hour) urinary excretion or were normalized to be representative of a full day before creating the lognormal plots with the exception of the uranium data, which were normalized to daily excretion before intake modeling [7]. Each result used in the intake calculation was assumed to have a normal distribution and a uniform absolute error of 1 was applied to all results, which weighted all results equally [8]. A chronic exposure pattern was assumed unless the data clearly showed a sharp, short-term, increase in excretion. Intakes were assumed to be from inhalation using a default breathing rate of 1.2 m<sup>3</sup>/hr and a 5- $\mu$ m AMAD particle size distribution [9].

When calculating doses to individuals from bioassay data, a GSD of 3 has been used to account for biological variation and uncertainty in the models. It was considered inappropriate to assign a value less than 3 for the coworker data. Therefore, a GSD of 3 was assigned for any intake period in which the calculated GSD was <3 [10].

#### **B4.2 BIOASSAY FITTING AND INTAKES**

The IMBA Expert ORAU-Edition, Version 4.0.9 computer program was used to fit the bioassay results to a series of chronic inhalations. The intake assumptions were based on observed patterns in the bioassay data. Intervals with constant chronic intake rates were chosen by selecting periods during which the bioassay results were of similar magnitudes. A new chronic intake period was started where the data indicated a significant sustained change in the results. The effective bioassay dates used in IMBA to calculate the intake rates were the midpoints of the sampling periods [11]. The 50th- and 84th-percentile excretion values were fit as independent datasets producing separate 50th- and 84th-percentile intakes (ORAUT 2005a). The GSDs were calculated as the ratio of the 84th- to the 50th-percentile intakes for each intake interval (ORAUT 2005a).

This TBD does not specify inhalation absorption types for the various radionuclide or group of radionuclides; therefore, the bioassay results were entered into IMBA with assumed lung absorption types chosen to be consistent with International Commission on Radiological Protection (ICRP) Publication 68 (ICRP 1995). The plots at the end of this attachment show the resultant 50th-percentile intakes. The bioassay data used in the fits are shown as solid blue dots (●) (dark spots when printed) and data that are not used in the fits are shown as red dots (●) (light dots when printed).

Some materials have very long radiological half-lives and/or are retained in the body for long periods. In such cases, the excretion results for different chronic intake periods are not independent. For example, an intake in the 1950s could contribute to urinary excretion in the 1980s and later. To avoid potential underestimation of intakes for people who worked at LLNL for relatively short periods, each chronic intake of long-lived or long-retained material was independently fit using only the bioassay results from the single intake period. This fitting method will result in a best estimate of dose if the person worked in only one period and a potential overestimate if an individual worked in multiple periods.

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**B4.2.1 Uranium**

The 50th- and 84th-percentile uranium urinary excretion concentrations in µg/L were converted to mg/day assuming a daily urine volume of 1.4 L/d (ICRP 1975). These data are shown in Table B-8. The intakes were determined from the mg/d excretion values. For ease of use by dose reconstructors, the intakes were also converted to activity using the specific activity of natural uranium (682 pCi/mg), which is favorable to claimants relative to using the specific activity of depleted uranium. While workers at LLNL might not have been chronically exposed to natural uranium, chronic intakes will approximate a series of acute intakes with unknown intake dates. Plots of the fits of models to urinary excretion are shown in Figures B-1 through B-24; note that all figures are at the end of this attachment.

The intake rates, GSDs, and periods in which they are applicable are given in Table B-8. Table B-9 lists the 50th- and 95th-percentile coworker uranium intakes to be assigned for dose reconstruction.

Table B-8. Uranium mass and activity intake rates for four different periods.<sup>a</sup>

| Type | Dates           | 50th percentile (mg/d) | 84th percentile (mg/d) | 50th percentile (pCi/d) | 84th percentile (pCi/d) | GSD               |
|------|-----------------|------------------------|------------------------|-------------------------|-------------------------|-------------------|
| F    | 1/1/58–12/31/73 | 2.51E-02               | 5.81E-02               | 1.71E+01                | 3.96E+01                | 2.32 <sup>a</sup> |
| F    | 1/1/74–12/31/87 | 1.65E-04               | 8.52E-04               | 1.13E-01                | 5.81E-01                | 5.16              |
| F    | 1/1/88–12/31/96 | 2.87E-04               | 3.73E-04               | 1.96E-01                | 2.54E-01                | 1.30 <sup>a</sup> |
| M    | 1/1/58–12/31/73 | 1.03E-01               | 2.39E-01               | 7.04E+01                | 1.65E+02                | 2.32 <sup>a</sup> |
| M    | 1/1/74–12/31/87 | 6.76E-04               | 3.49E-03               | 4.61E-01                | 2.38E+00                | 5.16              |
| M    | 1/1/88–12/31/96 | 1.20E-03               | 1.582E-03              | 8.16E-01                | 1.08E+00                | 1.32 <sup>a</sup> |
| S    | 1/1/58–12/31/73 | 1.24E+00               | 2.93E+00               | 8.49E+02                | 1.99E+03                | 2.35 <sup>a</sup> |
| S    | 1/1/74–12/31/87 | 8.55E-03               | 4.19E-02               | 5.83E+00                | 2.86E+01                | 4.90              |
| S    | 1/1/88–12/31/96 | 1.75E-02               | 3.01E-02               | 1.19E+01                | 2.05E+01                | 1.72 <sup>a</sup> |

a. Use the default GSD of 3.0 instead of the calculated GSD.

Table B-9. 50th- and 95th-percentile uranium mass and activity intake rates for four different periods.<sup>a</sup>

| Type | Dates           | 50th percentile (mg/d) | 50th percentile (pCi/d) | GSD  | 95th percentile (mg/day) | 95th percentile (pCi/d) |
|------|-----------------|------------------------|-------------------------|------|--------------------------|-------------------------|
| F    | 1/1/58–12/31/73 | 2.51E-02               | 1.71E+01                | 3.00 | 1.53E-01                 | 1.04E+02                |
| F    | 1/1/74–12/31/87 | 1.65E-04               | 1.13E-01                | 5.16 | 2.45E-03                 | 1.68E+00                |
| F    | 1/1/88–12/31/96 | 2.87E-04               | 1.96E-01                | 3.00 | 1.75E-03                 | 1.19E+00                |
| M    | 1/1/58–12/31/73 | 1.03E-01               | 7.04E+01                | 3.00 | 6.28E-01                 | 4.29E+02                |
| M    | 1/1/74–12/31/87 | 6.76E-04               | 4.61E-01                | 5.16 | 1.01E-02                 | 6.86E+00                |
| M    | 1/1/88–12/31/96 | 1.20E-03               | 8.16E-01                | 3.00 | 7.31E-03                 | 4.97E+00                |
| S    | 1/1/58–12/31/73 | 1.24E+00               | 8.49E+02                | 3.00 | 7.56E+00                 | 5.17E+03                |
| S    | 1/1/74–12/31/87 | 8.55E-03               | 5.83E+00                | 4.90 | 1.17E-01                 | 7.96E+01                |
| S    | 1/1/88–12/31/96 | 1.75E-02               | 1.19E+01                | 3.00 | 1.07E-01                 | 7.25E+01                |

a. Use the intakes in pCi/d assuming all the activity is from <sup>234</sup>U, which is favorable to claimants.

There were no bioassay results for uranium before 1958 in the MAPPER database and, according to the laboratory manager circa 1958, few or none might have been obtained, so workers exposed to uranium during 1952 through 1957 who did not have subsequent uranium bioassay would be

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considered unmonitored. Therefore, unmonitored uranium doses will not be assessed for the period before 1958, which is included in the designated SEC period.

**B4.2.2 Plutonium and Americium**

Table B-10 provides the 50th- and 84th-percentile values for <sup>239</sup>Pu urinary excretion per day. Plots of the fits of models to urinary excretion are shown in Figures B-25 through B-47. The 50th- and 84th-percentile intakes based on urinary excretion modeled as <sup>239</sup>Pu are summarized in Tables B-11 and B-12. Either absorption type M or S might have been present in the LLNL workplace, so the dose reconstructor should use whichever is more favorable to the claimant. All GSDs are larger than 3 so they can be used directly from the table. The plutonium intakes in Tables B-11 and B-12 can be considered pure <sup>239</sup>Pu; that is, other isotopes in a plutonium mixture should be based on isotopic ratios from Table 5-1. Tables B-13 and B-14 list the 50th- and 95th-percentile coworker plutonium intakes to be assigned for dose reconstruction.

Table B-10. Fiftieth- and 84th-percentiles for <sup>239</sup>Pu<sup>a</sup> urine bioassay data for 1957 through 1996.

| Year | Effective bioassay date | 50th-percentile excretion rate (dpm/d) | 84th-percentile excretion rate (dpm/d) |
|------|-------------------------|--|--|
| 1957 | 7/1/1957                | 4.470E-02                              | 2.829E-01                              |
| 1958 | 7/1/1958                | 9.435E-02                              | 1.002E+00                              |
| 1959 | 7/1/1959                | 1.362E-01                              | 1.361E+00                              |
| 1960 | 7/1/1960                | 7.974E-02                              | 3.566E-01                              |
| 1961 | 7/1/1961                | 3.559E-02                              | 4.136E-01                              |
| 1962 | 7/1/1962                | 4.280E-02                              | 1.113E+00                              |
| 1963 | 7/1/1963                | 2.858E-02                              | 2.956E-01                              |
| 1964 | 7/1/1964                | 3.106E-02                              | 1.133E-01                              |
| 1965 | 7/1/1965                | 1.571E-02                              | 1.547E-01                              |
| 1966 | 7/1/1966                | 1.007E-02                              | 1.547E-01                              |
| 1967 | 7/1/1967                | 6.051E-03                              | 3.878E-02                              |
| 1968 | 7/1/1968                | 4.899E-03                              | 2.202E-02                              |
| 1969 | 7/1/1969                | 1.462E-03                              | 1.526E-02                              |
| 1970 | 7/1/1970                | 2.788E-04                              | 6.192E-03                              |
| 1971 | 7/1/1971                | 2.427E-03                              | 2.898E-02                              |
| 1972 | 7/1/1972                | 3.008E-03                              | 2.499E-02                              |
| 1973 | 7/1/1973                | 4.306E-03                              | 3.266E-02                              |
| 1974 | 7/1/1974                | 2.651E-03                              | 2.339E-02                              |
| 1975 | 7/1/1975                | 1.395E-02                              | 9.313E-02                              |
| 1976 | 7/1/1976                | 3.893E-03                              | 5.441E-02                              |
| 1977 | 7/1/1977                | 3.085E-03                              | 2.961E-02                              |
| 1978 | 7/1/1978                | 1.541E-03                              | 3.706E-02                              |
| 1979 | 7/1/1979                | 7.989E-03                              | 4.731E-02                              |
| 1980 | 7/1/1980                | 1.759E-02                              | 1.376E-01                              |
| 1981 | 7/1/1981                | 5.392E-03                              | 3.582E-02                              |
| 1982 | 7/1/1982                | 4.136E-04                              | 7.478E-03                              |
| 1983 | 7/1/1983                | 1.650E-03                              | 1.078E-02                              |
| 1984 | 7/1/1984                | 8.048E-04                              | 9.488E-03                              |
| 1985 | 7/1/1985                | 6.502E-04                              | 7.737E-03                              |
| 1986 | 7/1/1986                | 9.787E-04                              | 1.257E-02                              |
| 1987 | 7/1/1987                | 2.037E-03                              | 9.097E-03                              |

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|      |          |           |           |
|------|----------|-----------|-----------|
| 1988 | 7/1/1988 | 1.207E-03 | 9.843E-03 |
| 1989 | 7/1/1989 | 3.082E-03 | 1.816E-02 |
| 1990 | 7/1/1990 | 1.092E-03 | 1.479E-02 |
| 1991 | 7/1/1991 | 2.890E-04 | 3.935E-03 |
| 1992 | 7/1/1992 | 1.308E-04 | 3.465E-03 |
| 1993 | 7/1/1993 | 2.425E-04 | 4.860E-03 |
| 1994 | 7/1/1994 | 2.670E-04 | 5.789E-03 |
| 1995 | 7/1/1995 | 2.991E-04 | 5.973E-03 |
| 1996 | 7/1/1996 | 1.357E-04 | 4.863E-03 |

a. The data for 1970 were excluded from the analysis because they are anomalously low.

Table B-11. Fiftieth- and 84th-percentile Type M <sup>239</sup>Pu intake rates from coworker plutonium urine bioassay data for 1957 through 1996.

| From     | To         | 50th percentile (dpm/day) | 84th percentile (dpm/day) | GSD   |
|----------|------------|---------------------------|---------------------------|-------|
| 1/1/1957 | 12/31/1960 | 22.97                     | 123.02                    | 5.36  |
| 1/1/1961 | 12/31/1963 | 8.09                      | 123.02                    | 15.20 |
| 1/1/1964 | 12/31/1964 | 8.09                      | 26.19                     | 3.24  |
| 1/1/1965 | 12/31/1967 | 1.88                      | 26.19                     | 13.95 |
| 1/1/1968 | 12/31/1968 | 1.88                      | 6.32                      | 3.37  |
| 1/1/1969 | 12/31/1981 | 0.82                      | 6.32                      | 7.69  |
| 1/1/1982 | 12/31/1990 | 0.23                      | 1.88                      | 8.11  |
| 1/1/1991 | 12/31/1996 | 0.04                      | 0.98                      | 22.59 |

Table B-12. Fiftieth- and 84th-percentile Type S <sup>239</sup>Pu intake rates from coworker plutonium urine bioassay data for 1957 through 1996.

| From     | To         | 50th percentile (dpm/day) | 84th percentile (dpm/day) | GSD   |
|----------|------------|---------------------------|---------------------------|-------|
| 1/1/1957 | 12/31/1960 | 580.54                    | 2,280.00                  | 3.93  |
| 1/1/1961 | 12/31/1963 | 126.67                    | 2,280.00                  | 18.00 |
| 1/1/1964 | 12/31/1965 | 126.67                    | 1,203.20                  | 9.50  |
| 1/1/1966 | 12/31/1968 | 47.40                     | 163.17                    | 3.44  |
| 1/1/1969 | 12/31/1969 | 13.41                     | 163.17                    | 12.16 |
| 1/1/1970 | 12/31/1981 | 13.41                     | 120.09                    | 8.95  |
| 1/1/1982 | 12/31/1990 | 4.24                      | 33.96                     | 8.01  |
| 1/1/1991 | 12/31/1996 | 0.88                      | 20.58                     | 23.38 |

Table B-13. 50th- and 95th-percentile Type M <sup>239</sup>Pu intake rates from coworker plutonium urine bioassay data for 1957 through 1996.

| From     | To         | 50th percentile (dpm/d) | 84th percentile (dpm/d) | GSD   | 95th percentile (dpm/d) |
|----------|------------|-------------------------|-------------------------|-------|-------------------------|
| 1/1/1957 | 12/31/1960 | 22.97                   | 123.02                  | 5.36  | 363.61                  |
| 1/1/1961 | 12/31/1963 | 8.09                    | 123.02                  | 15.2  | 711.35                  |
| 1/1/1964 | 12/31/1964 | 8.09                    | 26.19                   | 3.24  | 55.95                   |
| 1/1/1965 | 12/31/1967 | 1.88                    | 26.19                   | 13.95 | 143.54                  |
| 1/1/1968 | 12/31/1968 | 1.88                    | 6.32                    | 3.37  | 13.87                   |
| 1/1/1969 | 12/31/1981 | 0.82                    | 6.32                    | 7.69  | 23.51                   |



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|          |            |      |      |       |      |
|----------|------------|------|------|-------|------|
| 1/1/1982 | 12/31/1990 | 0.23 | 1.88 | 8.11  | 7.20 |
| 1/1/1991 | 12/31/1996 | 0.04 | 0.98 | 22.59 | 6.75 |

For assessing Super S solubility, refer to ORAUT-OTIB-0049, *Estimating Doses for Plutonium Strongly Retained in the Lung* (ORAUT 2010a).

Table B-15 provides the 50th- and 84th-percentile values for <sup>241</sup>Am urinary excretion per day. Plots of the fits of models to urinary excretion are shown in Figures B-48 through B-54. Intakes were modeled assuming all of the americium/curium results were <sup>241</sup>Am. The biokinetic model for curium is basically the same as that for americium and produces doses comparable to those from <sup>241</sup>Am for longer-lived isotopes of curium (ICRP 1995). Thus, using <sup>241</sup>Am as the modeled radionuclide is favorable to

Table B-14. 50th- and 95th-percentile Type S <sup>239</sup>Pu intake rates from coworker plutonium urine bioassay data for 1957 through 1996.

| From     | To         | 50th percentile (dpm/d) | GSD   | 95th percentile (dpm/d) |
|----------|------------|-------------------------|-------|-------------------------|
| 1/1/1957 | 12/31/1960 | 580.54                  | 3.93  | 5515.79                 |
| 1/1/1961 | 12/31/1963 | 126.67                  | 18    | 14709.47                |
| 1/1/1964 | 12/31/1965 | 126.67                  | 9.5   | 5140.78                 |
| 1/1/1966 | 12/31/1968 | 47.4                    | 3.44  | 361.76                  |
| 1/1/1969 | 12/31/1969 | 13.41                   | 12.16 | 816.85                  |
| 1/1/1970 | 12/31/1981 | 13.41                   | 8.95  | 493.38                  |
| 1/1/1982 | 12/31/1990 | 4.24                    | 8.01  | 129.97                  |
| 1/1/1991 | 12/31/1996 | 0.88                    | 23.38 | 157.12                  |

claimants. The 50th- and 84th-percentile intakes based on urinary excretion modeled as <sup>241</sup>Am are summarized in Table B-16. Table B-17 lists the 50th- and 95th-percentile coworker americium intakes to be assigned for dose reconstruction. The coworker americium intakes should be assigned for cases where there was potential for internal exposure from americium or curium. Separated americium was used in Building 251 as early as 1955 for tracer studies. Curium was used primarily in Buildings 251 around the same time frame as americium, and 332. Americium coworker intakes should only be assigned in the case for work in Building 251. This can be determined based on the worker's job description, work location, claimant telephone interview, and DOE dosimetry records information. In the absence of specific information, the plutonium coworker intakes, as part of a weapons grade mixture, should be assigned.

Table B-15. Fiftieth- and 84th-percentiles for <sup>241</sup>Am urine bioassay data for 1957 through 1996.

| Year | Effective bioassay date | 50th-percentile excretion rate (dpm/d) | 84th-percentile excretion rate (dpm/d) |
|------|-------------------------|--|--|
| 1957 | 7/1/1957                | 4.313E-02                              | 2.785E-01                              |
| 1958 | 7/1/1958                | 9.435E-02                              | 1.002E+00                              |
| 1959 | 7/1/1959                | 1.362E-01                              | 1.361E+00                              |
| 1960 | 7/1/1960                | 7.974E-02                              | 3.566E-01                              |
| 1961 | 7/1/1961                | 3.559E-02                              | 4.136E-01                              |
| 1962 | 7/1/1962                | 4.280E-02                              | 1.113E+00                              |
| 1963 | 7/1/1963                | 4.462E-02                              | 4.147E-01                              |

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| Year    | Effective bioassay date | 50th-percentile excretion rate (dpm/d) | 84th-percentile excretion rate (dpm/d) |
|---------|-------------------------|--|--|
| 1964    | 7/1/1964                | 3.329E-02                              | 1.106E-01                              |
| 1965    | 7/1/1965                | 2.384E-02                              | 1.910E-01                              |
| 1966-67 | 12/31/1966              | 9.702E-03                              | 1.660E-01                              |
| 1968    | 7/1/1968                | 5.216E-03                              | 2.327E-02                              |
| 1969    | 7/1/1969                | 1.179E-03                              | 1.462E-02                              |
| 1970    | 7/1/1970                | 8.473E-05                              | 2.928E-03                              |
| 1971-72 | 12/31/1971              | 2.371E-03                              | 2.889E-02                              |
| 1973-75 | 7/1/1974                | 1.395E-02                              | 1.283E-01                              |
| 1976-78 | 7/1/1977                | 3.564E-02                              | 7.259E-01                              |
| 1979-81 | 7/1/1980                | 2.059E-02                              | 1.807E-01                              |
| 1982    | 7/1/1982                | 4.221E-02                              | 2.041E-01                              |
| 1983    | 7/1/1983                | 1.466E-02                              | 9.062E-02                              |
| 1984    | 7/1/1984                | 3.748E-02                              | 2.872E-01                              |
| 1985    | 7/1/1985                | 2.131E-02                              | 1.345E-01                              |
| 1986    | 7/1/1986                | 5.588E-02                              | 2.278E-01                              |
| 1987    | 7/1/1987                | 7.659E-02                              | 5.604E-01                              |
| 1988    | 7/1/1988                | 4.988E-02                              | 3.161E-01                              |
| 1989    | 7/1/1989                | 2.334E-02                              | 2.421E-01                              |
| 1990    | 7/1/1990                | 3.713E-02                              | 2.369E-01                              |
| 1991    | 7/1/1991                | 1.872E-02                              | 1.024E-01                              |
| 1992    | 7/1/1992                | 5.301E-02                              | 2.260E-01                              |
| 1993    | 7/1/1993                | 1.601E-02                              | 1.010E-01                              |
| 1994    | 7/1/1994                | 2.848E-02                              | 2.101E-01                              |
| 1995    | 7/1/1995                | 3.952E-02                              | 1.900E-01                              |
| 1996    | 7/1/1996                | 7.815E-02                              | 3.557E-01                              |

Table B-16. Fiftieth- and 84th-percentile Type M <sup>241</sup>Am intake rates from coworker plutonium urine bioassay data for 1957 through 1996.

| From     | To         | 50th percentile (dpm/d) | 84th percentile (dpm/d) | GSD  |
|----------|------------|-------------------------|-------------------------|------|
| 1/1/1957 | 12/31/1960 | 10.1                    | 43.8                    | 4.36 |
| 1/1/1961 | 12/31/1965 | 3.48                    | 43.8                    | 12.6 |
| 1/1/1966 | 12/31/1970 | 0.49                    | 5.72                    | 11.8 |
| 1/1/1971 | 12/31/1996 | 1.93                    | 12.7                    | 6.54 |

Table B-17. 50th- and 95th-percentile Type M <sup>241</sup>Am intake rates from coworker plutonium urine bioassay data for 1957 through 1996.

| From     | To         | 50th percentile (dpm/d) | GSD  | 95th percentile (dpm/d) |
|----------|------------|-------------------------|------|-------------------------|
| 1/1/1957 | 12/31/1960 | 10.1                    | 4.36 | 113.84                  |
| 1/1/1961 | 12/31/1965 | 3.48                    | 12.6 | 224.74                  |
| 1/1/1966 | 12/31/1970 | 0.49                    | 11.8 | 28.41                   |
| 1/1/1971 | 12/31/1996 | 1.93                    | 6.54 | 42.38                   |

There were no bioassay results for plutonium or gross alpha before 1957 in the MAPPER database and, according to the laboratory manager circa 1958, few or none might have been obtained (Bihl

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2006b). Therefore, unmonitored plutonium doses will not be assessed for the period before 1958, which is included in the designated SEC period.

**B4.2.3 Gross Beta/MFP**

Interpreting gross beta urinalyses requires

- At least approximate knowledge of the ratio of the various possible fission/activation products at the time of intake,
- How the mix of those radionuclides will be altered by human biokinetics and be manifested in urine, and
- How the latter mix will contribute to gross beta counts given the impact of the chemistry and counting technique.

Note: See Section 5.2 for source term information.

**Reactor Mixtures (1974–1980)**

As shown in Figures B-55 and B-56, the gross beta urine data given in Table B-18 were modeled as 100% Type F <sup>90</sup>Sr. The resulting intakes were then multiplied by 0.024, which is the average fraction of <sup>90</sup>Sr in urine following intakes of 10-day old reactor fuel in OTIB-0054 (ORAUT 2007b, Table 7-1). Table B-19 lists the fiftieth- and 84th-percentile intake rates of Type F <sup>90</sup>Sr for 1974 through 1980. The 50th- and 95th-percentile <sup>90</sup>Sr intake rates, which are given in Table B-20, should be used in conjunction with associated radionuclides' ratios for estimating intakes for relevant radionuclides based on intake activity relative to <sup>90</sup>Sr for 10-day old reactor fuel from Table 7-3 in OTIB-0054. As noted previously, the LPTR operated from December of 1957 to March 31, 1980.

Table B-18. Fiftieth- and 84th-percentiles for gross beta urine bioassay data for 1974 through 1995.

| Year    | Effective bioassay date | 50th-percentile excretion rate (dpm/d) | 84th-percentile excretion rate (dpm/d) |
|---------|-------------------------|--|--|
| 1974–75 | 12/31/1974              | 19.22                                  | 199.1                                  |
| 1976    | 7/1/1976                | 17.74                                  | 136.8                                  |
| 1977    | 7/1/1977                | 18.13                                  | 103.8                                  |
| 1978–79 | 12/31/1978              | 1.474                                  | 55.76                                  |
| 1980–81 | 12/31/1980              | 13.04                                  | 165.8                                  |
| 1982    | 7/1/1982                | 22.56                                  | 236.8                                  |
| 1983    | 7/1/1983                | 96.75                                  | 636.4                                  |
| 1984    | 7/1/1984                | 9.806                                  | 99.19                                  |
| 1985    | 7/1/1985                | 20.17                                  | 145.1                                  |
| 1986    | 7/1/1986                | 17.59                                  | 203.3                                  |
| 1987    | 7/1/1987                | 64.88                                  | 395.0                                  |
| 1988    | 7/1/1988                | 125.1                                  | 777.1                                  |
| 1989    | 7/1/1989                | 62.62                                  | 544.2                                  |
| 1990    | 7/1/1990                | 62.73                                  | 455.0                                  |
| 1991    | 7/1/1991                | 14.79                                  | 145.5                                  |
| 1992    | 7/1/1992                | 7.883                                  | 76.81                                  |
| 1993    | 7/1/1993                | 1.128                                  | 19.69                                  |
| 1994    | 7/1/1994                | 0.1914                                 | 4.956                                  |
| 1995    | 7/1/1995                | 0.2789                                 | 4.694                                  |

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Table B-19. Fiftieth- and 84th-percentile intake rates of Type F  $^{90}\text{Sr}$  for 1974 through 1980.

| From     | To        | 50th percentile (dpm/d) | 84th percentile (dpm/d) | GSD  |
|----------|-----------|-------------------------|-------------------------|------|
| 1/1/1974 | 3/31/1980 | 1.51                    | 14.71                   | 9.75 |

Table B-20. 50th- and 95th-percentile Type F  $^{90}\text{Sr}$  intake rates for 1974 through 1980. (These intake rates are used in conjunction with OTIB-0054 (ORAUT 2007b) to assign intakes of fission product mixtures from reactors based on the associated radionuclides ratios relative to  $^{90}\text{Sr}$  for 10-day old reactor fuel in Table 7-3 in OTIB-0054.)

| From     | To        | 50th percentile (dpm/d) | GSD  | 95th percentile (dpm/d) |
|----------|-----------|-------------------------|------|-------------------------|
| 1/1/1974 | 3/31/1980 | 1.51                    | 9.75 | 63.96                   |

### Weapons Residue (1974–1992)

As shown in Figures B-57 and B-58, the gross beta urinary excretion for 1974 through 1992 was modeled Type F  $^{103}\text{Ru}$ . Ruthenium-103 intakes were then assigned using 59% of the geometric means and 84th percentiles. Additional intakes of Type S  $^{141}\text{Ce}$  and Type F  $^{89}\text{Sr}$  were included at 50% each of the  $^{103}\text{Ru}$  intakes. Tungsten-181 is not a beta emitter and would not have been detected in the gross beta urinalysis (Kocher 1981); however, the additional dose to each organ from using  $^{103}\text{Ru}$  compensates for the unmeasured intake/dose from  $^{181}\text{W}$ . Table B-21 lists the 50th- and 84th-percentile  $^{103}\text{Ru}$  intakes. Table B-22 lists the 50th- and 84th-percentile  $^{141}\text{Ce}$ , and  $^{89}\text{Sr}$  intakes. Table B-23 lists the 50th- and 95th-percentile  $^{103}\text{Ru}$  intakes to be assigned for dose reconstruction. Table B-24 lists the 50th- and 95th-percentile  $^{141}\text{Ce}$ , and  $^{89}\text{Sr}$  intakes to be assigned for dose reconstruction.

Table B-21. Fiftieth- and 84th-percentile intake rates of Type F  $^{103}\text{Ru}$  for 1974 through 1992.

| From     | To         | 50th percentile (dpm/d) | 84th percentile (dpm/d) | GSD  |
|----------|------------|-------------------------|-------------------------|------|
| 1/1/1974 | 12/31/1982 | 62.72                   | 605.52                  | 9.65 |
| 1/1/1983 | 12/31/1983 | 411.48                  | 2,706.57                | 6.58 |
| 1/1/1984 | 12/31/1986 | 62.72                   | 605.52                  | 9.65 |
| 1/1/1987 | 12/31/1987 | 269.51                  | 1,975.44                | 7.33 |
| 1/1/1988 | 12/31/1988 | 531.74                  | 3,304.41                | 6.21 |
| 1/1/1989 | 12/31/1990 | 269.51                  | 1,975.44                | 7.33 |
| 1/1/1991 | 12/31/1992 | 62.72                   | 605.52                  | 9.65 |

### Aged Fission Products and Research-Related Radionuclides (After 1992)

As shown in Figures B-59 and B-60, the gross beta urinary excretion data for 1993 to 1995 were modeled as Type F  $^{90}\text{Sr}$ . Intakes of  $^{32}\text{P}$ ,  $^{14}\text{C}$ ,  $^{137}\text{Cs}$ , and  $^{90}\text{Y}$  equal to the  $^{90}\text{Sr}$  intakes were then applied. Table B-25 lists the 50th- and 84th-percentile  $^{90}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{32}\text{P}$ ,  $^{137}\text{Cs}$ , and  $^{14}\text{C}$  intakes. Table B-22 lists the 50th and 84th percentile  $^{141}\text{Ce}$ , and  $^{89}\text{Sr}$  intakes. Tables B-26 lists the 50th- and 95th-percentile  $^{90}\text{Sr}$ ,  $^{90}\text{Y}$ ,  $^{32}\text{P}$ ,  $^{137}\text{Cs}$ , and  $^{14}\text{C}$  intakes to be assigned for dose reconstruction.

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Table B-22. Fiftieth- and 84th-percentile intake rates of Type S  $^{141}\text{Ce}$  and Type F  $^{89}\text{Sr}$  for 1974 through 1992.

| From     | To         | 50th percentile (dpm/d) | 84th percentile (dpm/d) | GSD  |
|----------|------------|-------------------------|-------------------------|------|
| 1/1/1974 | 12/31/1982 | 31.36                   | 302.76                  | 9.65 |
| 1/1/1983 | 12/31/1983 | 205.74                  | 1,353.28                | 6.58 |
| 1/1/1984 | 12/31/1986 | 31.36                   | 302.76                  | 9.65 |
| 1/1/1987 | 12/31/1987 | 134.76                  | 987.72                  | 7.33 |
| 1/1/1988 | 12/31/1988 | 265.87                  | 1,652.21                | 6.21 |
| 1/1/1989 | 12/31/1990 | 134.76                  | 987.72                  | 7.33 |
| 1/1/1991 | 12/31/1992 | 31.36                   | 302.76                  | 9.65 |

Table B-23. 50th- and 95th-percentile Type F  $^{103}\text{Ru}$  intake rates for 1974 through 1992. These intake rates are used to assign intakes of fission and activation product mixtures from weapons test residuals.

| From     | To         | 50th percentile (dpm/d) | GSD  | 95th percentile (dpm/d) |
|----------|------------|-------------------------|------|-------------------------|
| 1/1/1974 | 12/31/1982 | 62.72                   | 9.65 | 2611.88                 |
| 1/1/1983 | 12/31/1983 | 411.48                  | 6.58 | 9127.05                 |
| 1/1/1984 | 12/31/1986 | 62.72                   | 9.65 | 2611.88                 |
| 1/1/1987 | 12/31/1987 | 269.51                  | 7.33 | 7139.55                 |
| 1/1/1988 | 12/31/1988 | 531.74                  | 6.21 | 10723.46                |
| 1/1/1989 | 12/31/1990 | 269.51                  | 7.33 | 7139.55                 |
| 1/1/1991 | 12/31/1992 | 62.72                   | 9.65 | 2611.88                 |

Table B-24. 50th- and 95th-percentile Type S  $^{141}\text{Ce}$  and Type F  $^{89}\text{Sr}$  intake rates for 1974 through 1992. These intake rates are used to assign intakes of fission and activation product mixtures from weapons test residuals. Note that the indicated intake rate is assigned for each radionuclide.

| From     | To         | 50th percentile (dpm/d) | GSD  | 95th percentile (dpm/d) |
|----------|------------|-------------------------|------|-------------------------|
| 1/1/1974 | 12/31/1982 | 31.36                   | 9.65 | 1305.94                 |
| 1/1/1983 | 12/31/1983 | 205.74                  | 6.58 | 4563.52                 |
| 1/1/1984 | 12/31/1986 | 31.36                   | 9.65 | 1305.94                 |
| 1/1/1987 | 12/31/1987 | 134.76                  | 7.33 | 3569.91                 |
| 1/1/1988 | 12/31/1988 | 265.87                  | 6.21 | 5361.73                 |
| 1/1/1989 | 12/31/1990 | 134.76                  | 7.33 | 3569.91                 |
| 1/1/1991 | 12/31/1992 | 31.36                   | 9.65 | 1305.94                 |

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Table B-25. Fiftieth- and 84th-percentile intake rates of Type F <sup>90</sup>Sr, <sup>90</sup>Y, <sup>32</sup>P, <sup>137</sup>Cs, and <sup>14</sup>C for after 1992. (Note that the indicated intake rate is assigned for each radionuclide.)<sup>a</sup>

| From     | To         | 50th percentile (dpm/d) | 84th percentile (dpm/d) | GSD  |
|----------|------------|-------------------------|-------------------------|------|
| 1/1/1993 | 12/31/1993 | 5.11                    | 89.5                    | 17.5 |
| 1/1/1994 | 12/31/1995 | 0.909                   | 19.1                    | 20.9 |

a. Refer to Section 5.3.2.3 for solubility types for Y-90, P-32, Cs-137, and C-14.

Table B-26. 50th- and 95th-percentile Type F <sup>90</sup>Sr, <sup>90</sup>Y, <sup>32</sup>P, <sup>137</sup>Cs, and <sup>14</sup>C intake rates for after 1992. (Note that the indicated intake rate is assigned for each radionuclide.)<sup>a</sup>

| From     | To         | 50th percentile (dpm/d) | GSD  | 95th percentile (dpm/d) |
|----------|------------|-------------------------|------|-------------------------|
| 1/1/1993 | 12/31/1993 | 5.11                    | 17.5 | 566.52                  |
| 1/1/1994 | 12/31/1995 | 0.909                   | 20.9 | 134.96                  |

a. Refer to Section 5.3.2.3 for solubility types for Y-90, P-32, Cs-137, and C-14.

Because there were few bioassays before 1974, guidance on assigning intakes of beta/gamma-emitting radionuclides before 1974 were not developed. Intakes and doses before 1974 are covered under the SEC designation (Leavitt 2008).

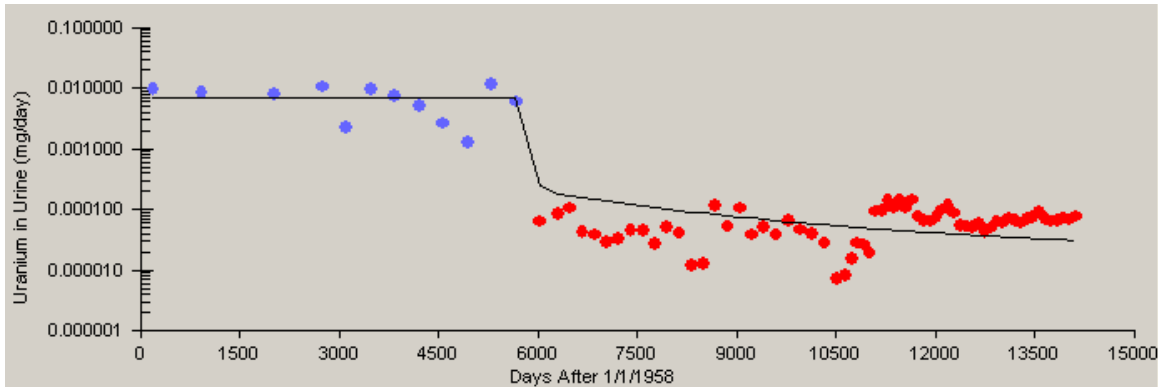


Figure B-1. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type F natural uranium from January 1958 through December 1973 (blue dots).

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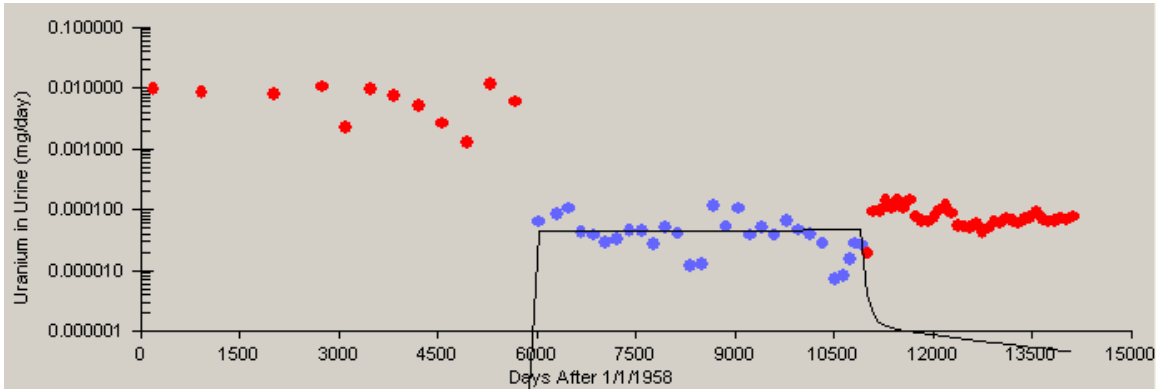


Figure B-2. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type F natural uranium from January 1974 through December 1987 (blue dots).

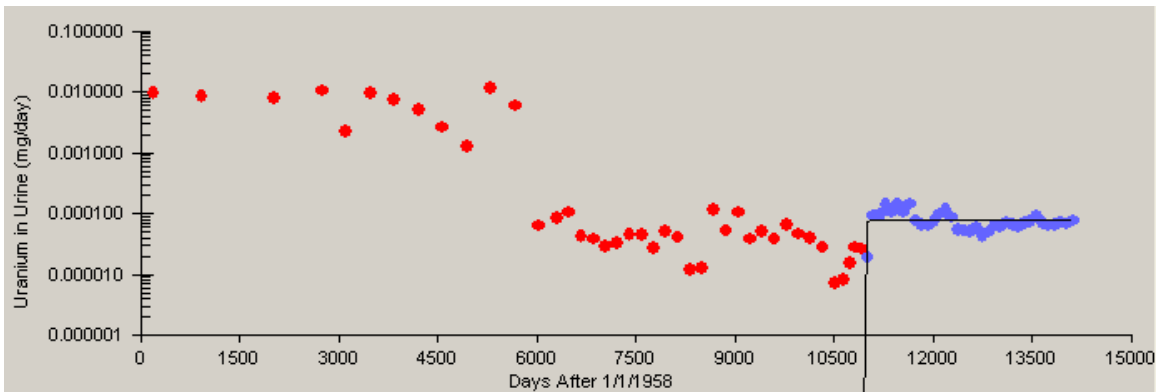


Figure B-3. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type F natural uranium from January 1988 through December 1996 (blue dots).

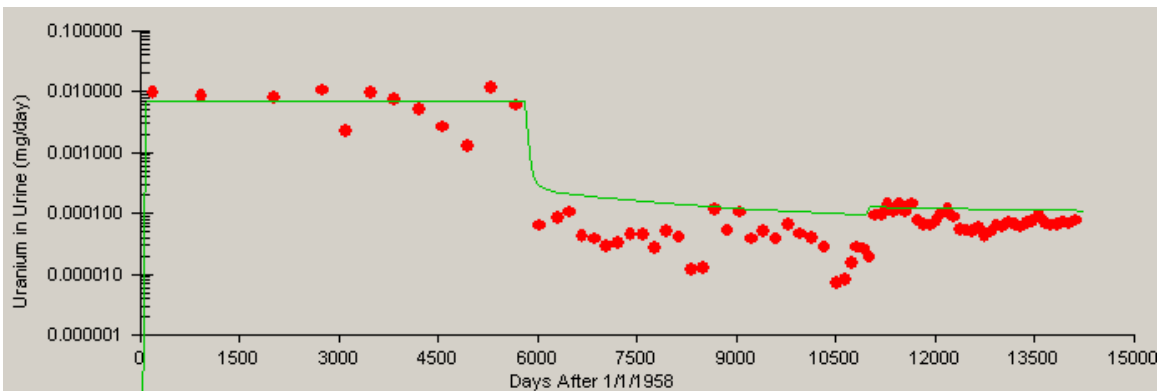


Figure B-4. Predicted and observed 50th-percentile urinary excretion assuming three separate chronic inhalation intakes of Type F natural uranium.

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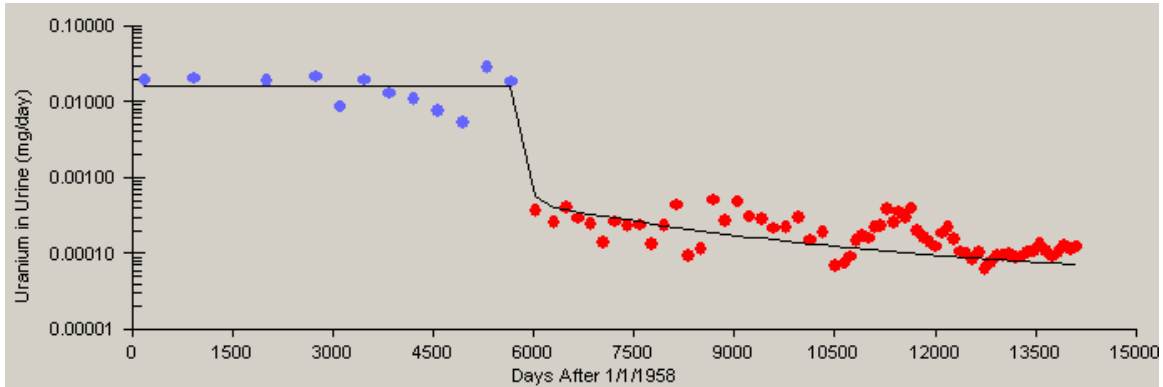


Figure B-5. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type F natural uranium from January 1958 through December 1973 (blue dots).

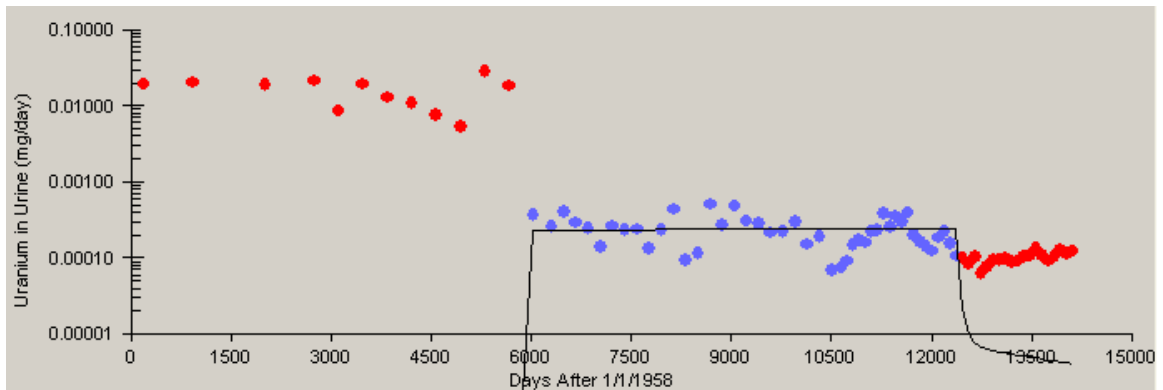


Figure B-6. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type F natural uranium from January 1974 through December 1987 (blue dots).

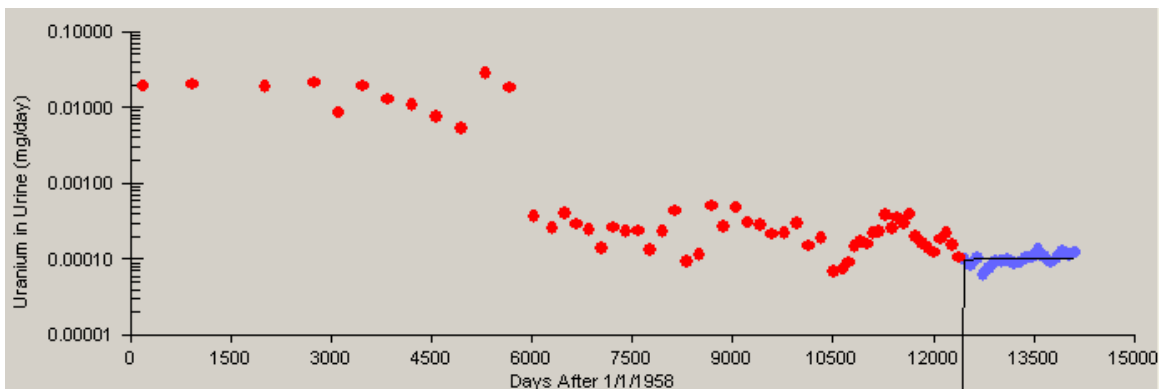


Figure B-7. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type F natural uranium from January 1988 through December 1996 (blue dots).



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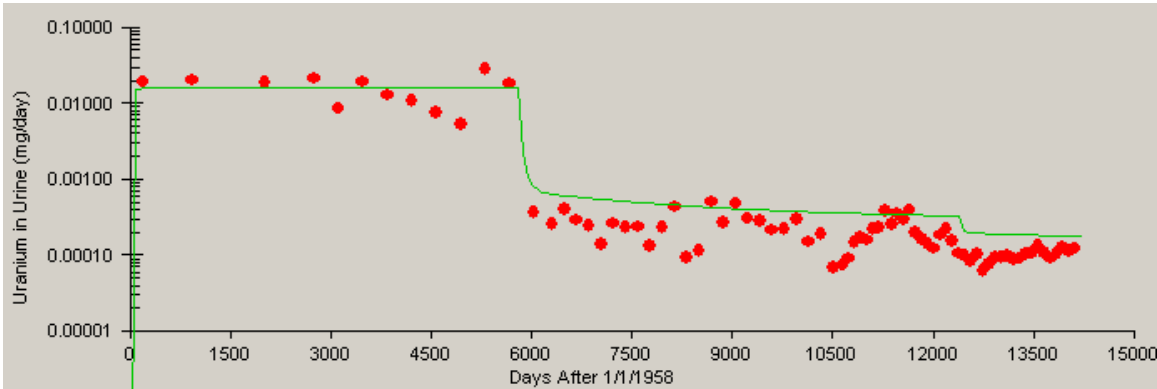


Figure B-8. Predicted and observed 84th-percentile urinary excretion assuming three separate chronic inhalation intakes of Type F natural uranium.

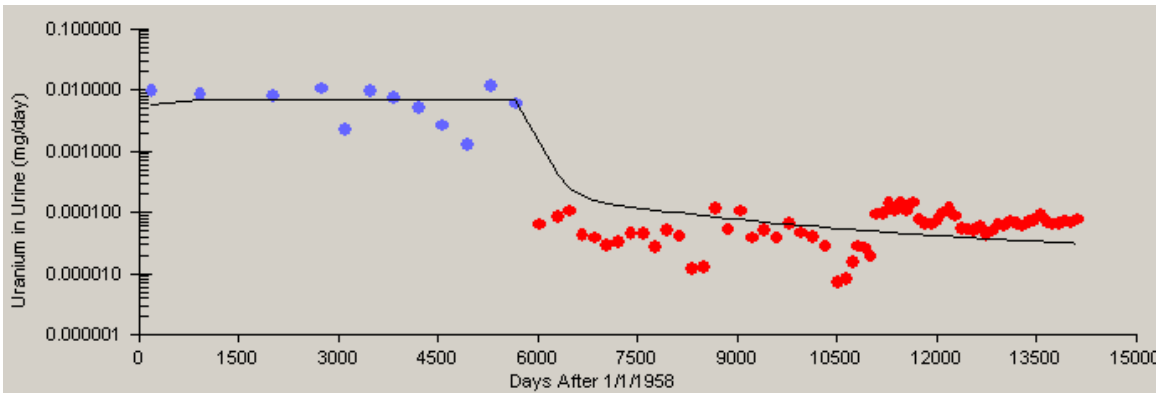


Figure B-9. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M natural uranium from January 1958 through December 1973 (blue dots).

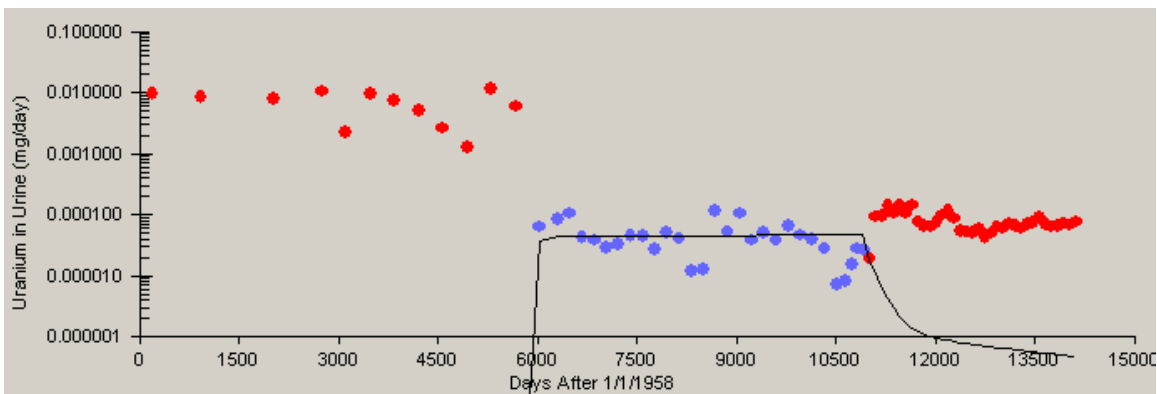


Figure B-10. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M natural uranium from January 1974 through December 1987 (blue dots).

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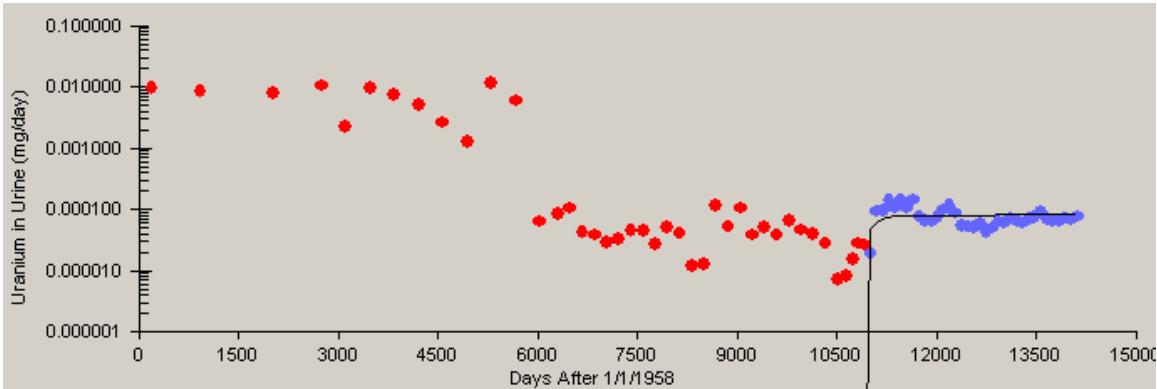


Figure B-11. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M natural uranium from January 1988 through December 1996 (blue dots).

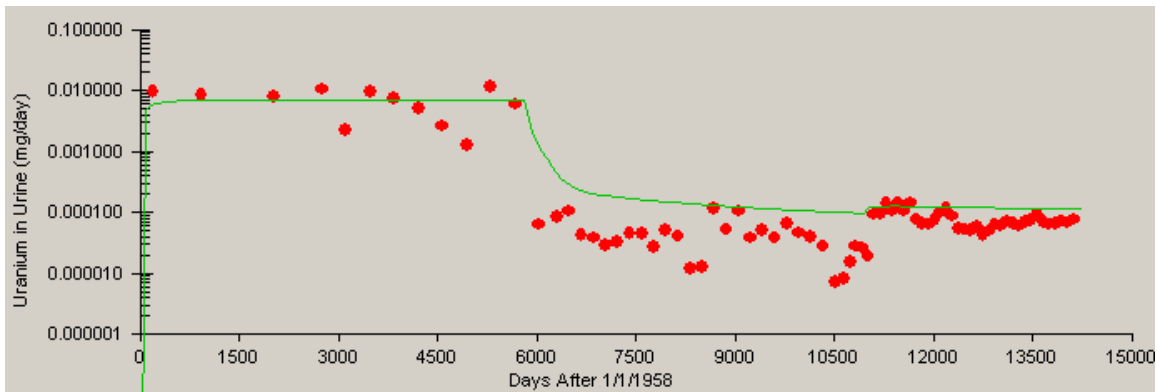


Figure B-12. Predicted and observed 50th-percentile urinary excretion assuming three separate chronic inhalation intakes of Type M natural uranium.

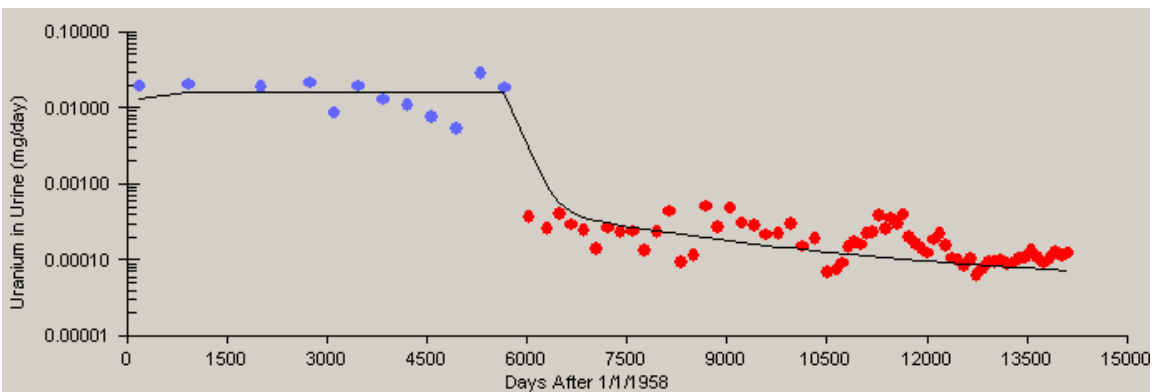


Figure B-13. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M natural uranium from January 1958 through December 1973 (blue dots).

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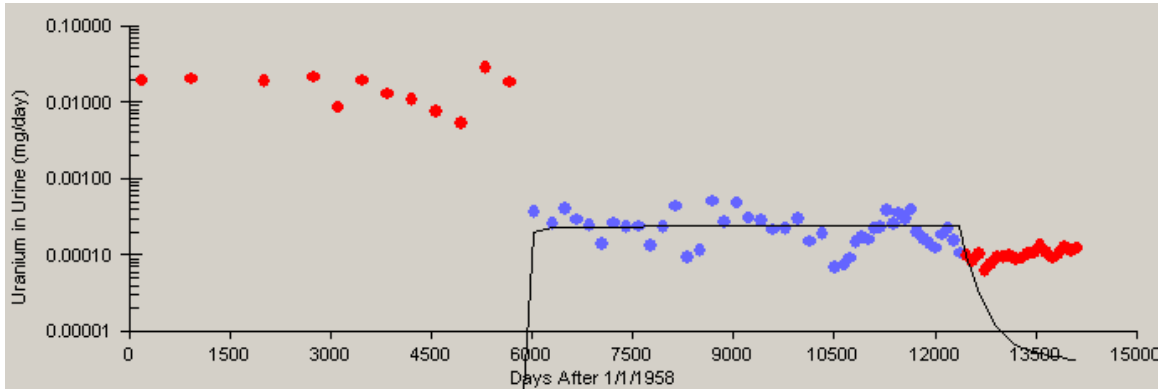


Figure B-14. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M natural uranium from January 1974 through December 1987 (blue dots).

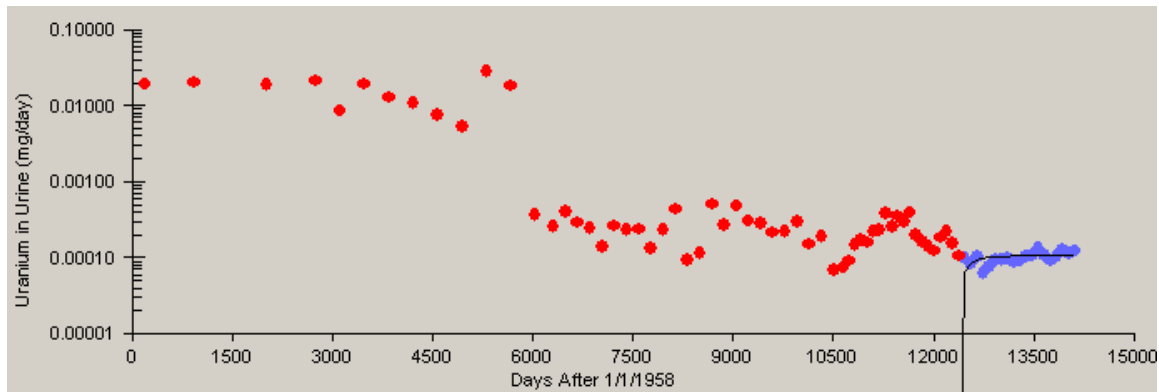


Figure B-15. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M natural uranium from January 1988 through December 1996 (blue dots).

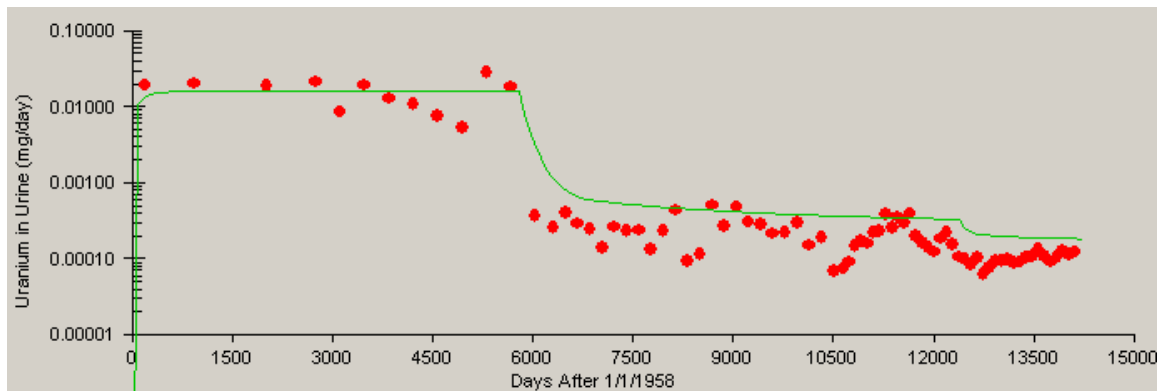


Figure B-16. Predicted and observed 84th-percentile urinary excretion assuming three separate chronic inhalation intakes of Type M natural uranium (blue dots).

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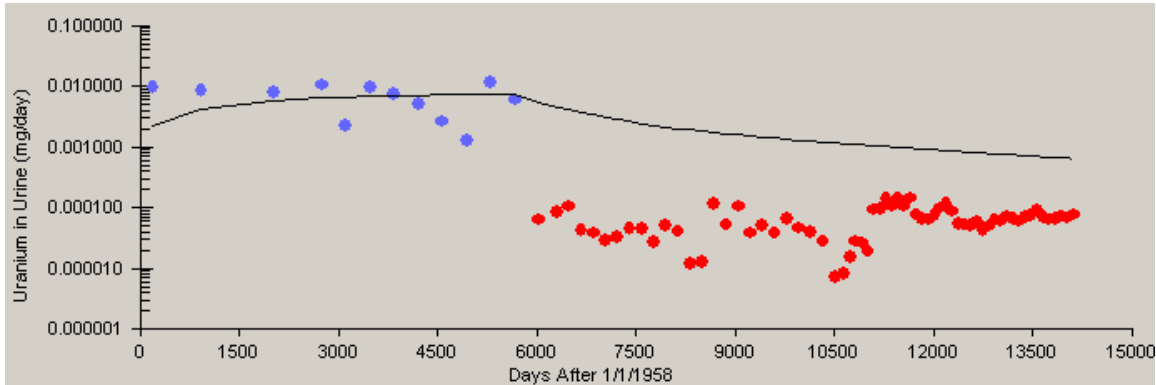


Figure B-17. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S natural uranium from January 1959 through December 1973 (blue dots).

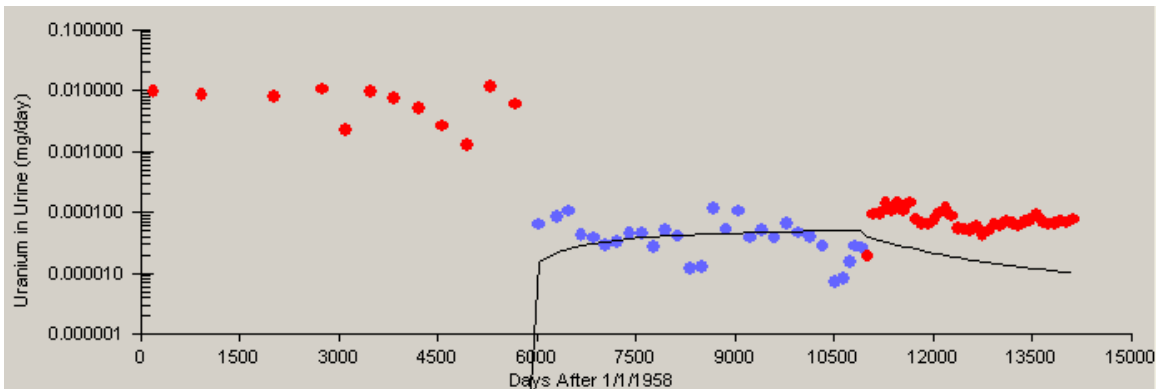


Figure B-18. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S natural uranium from January 1974 through December 1987 (blue dots).

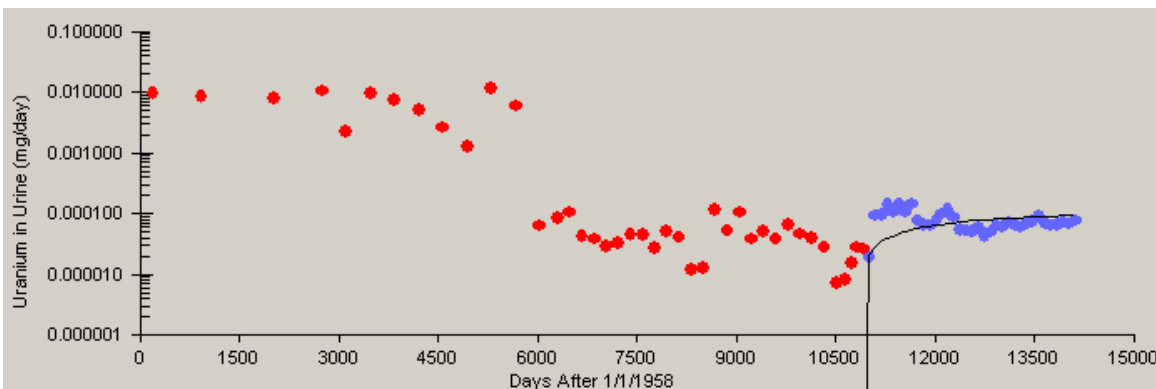


Figure B-19. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S natural uranium from January 1988 through December 1996 (blue dots).

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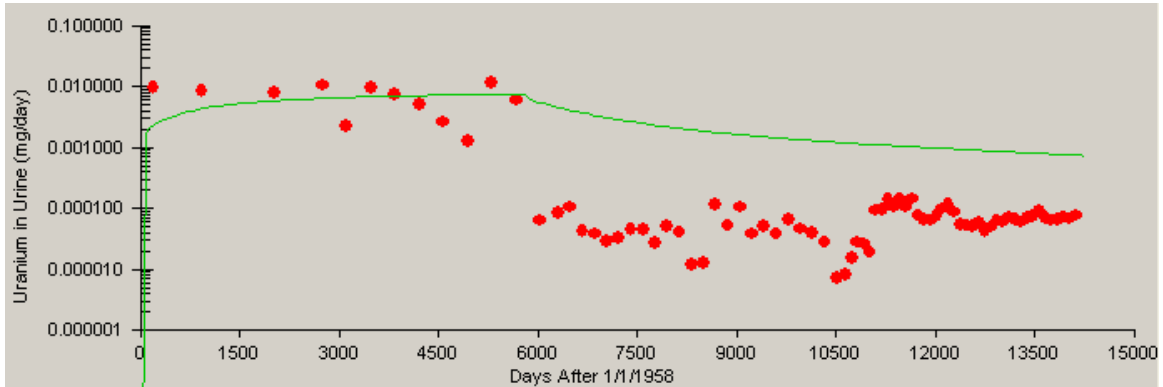


Figure B-20. Predicted and observed 50th-percentile urinary excretion assuming three separate chronic inhalation intakes of Type S natural uranium.

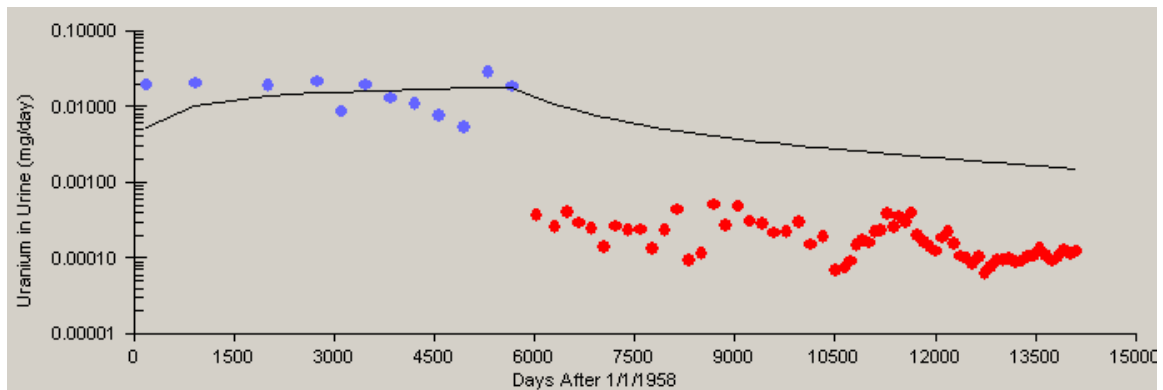


Figure B-21. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S natural uranium from January 1958 through December 1973 (blue dots).

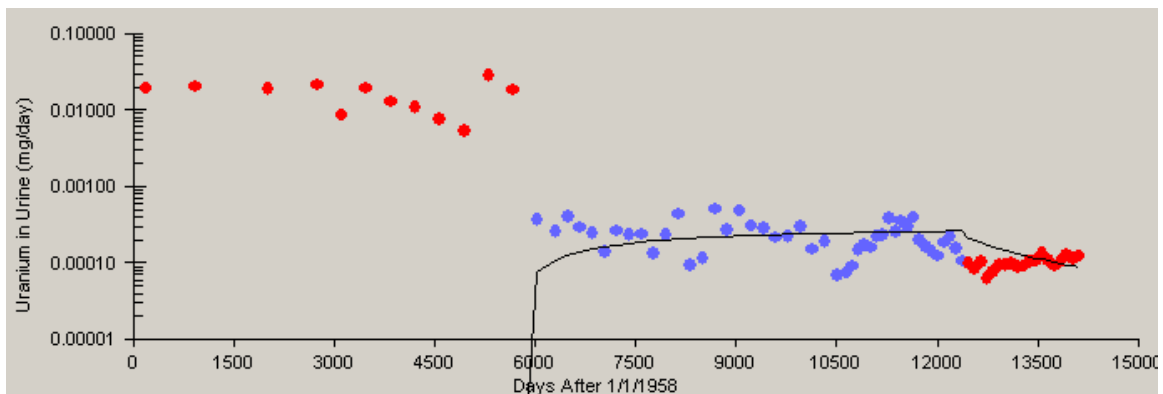


Figure B-22. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S natural uranium from January 1974 through December 1987 (blue dots).

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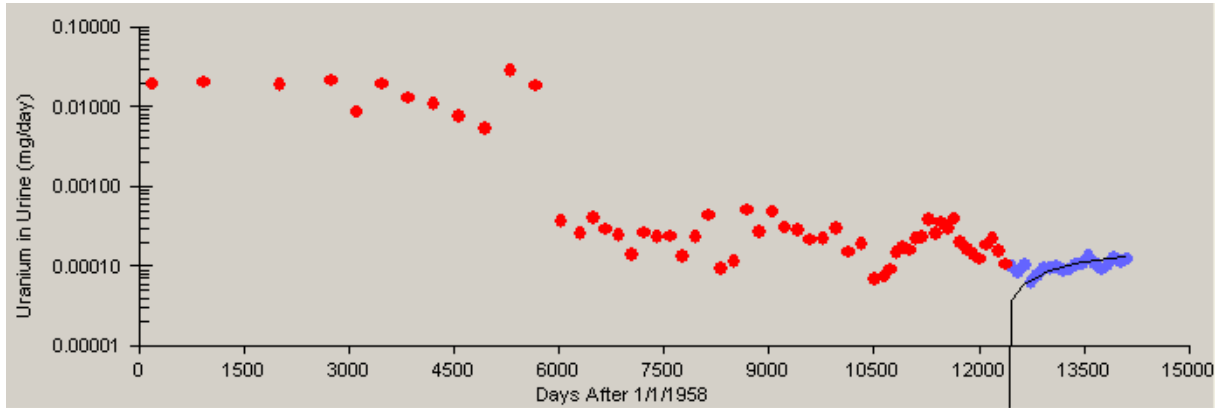


Figure B-23. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S natural uranium from January 1988 through December 1996 (blue dots).

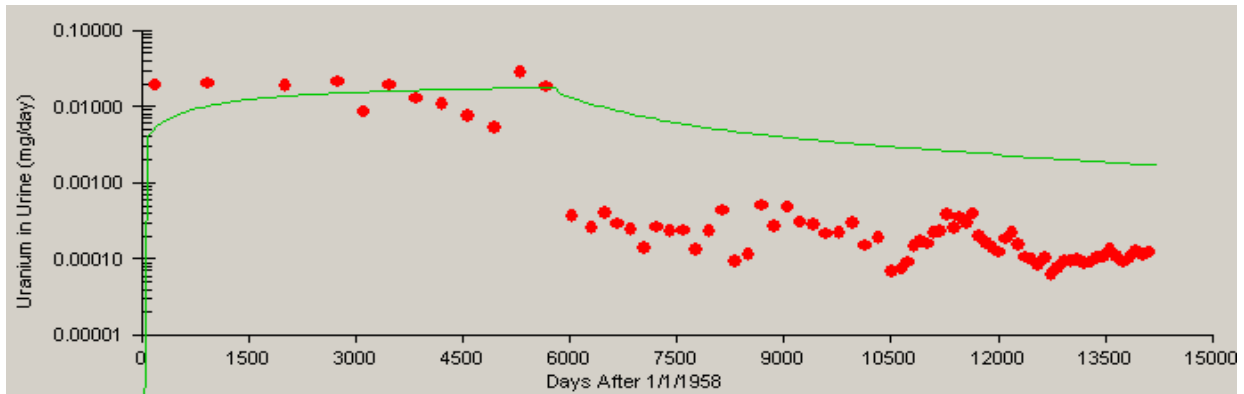


Figure B-24. Predicted and observed 84th-percentile urinary excretion assuming three separate chronic inhalation intakes of Type S natural uranium.

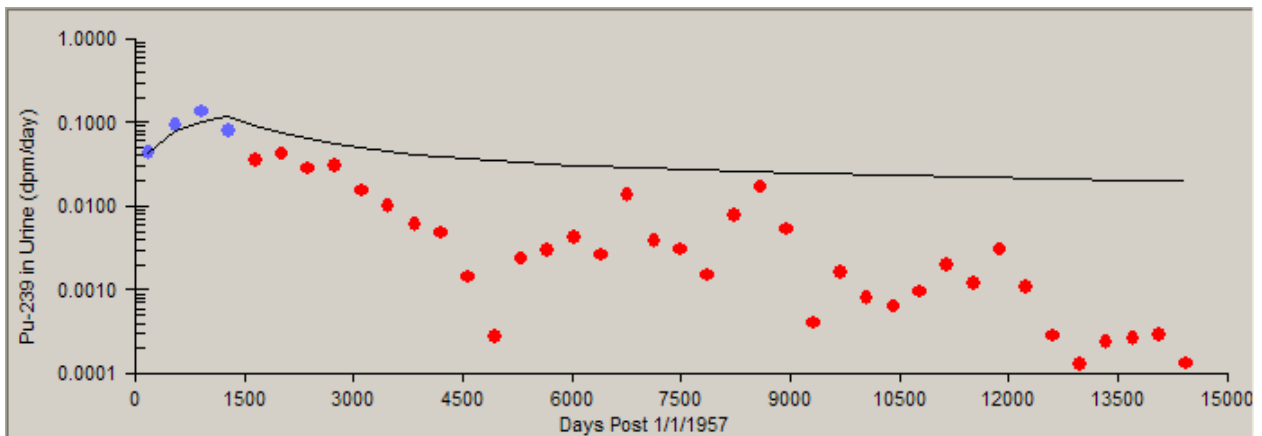


Figure B-25. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1957 through December 1960.

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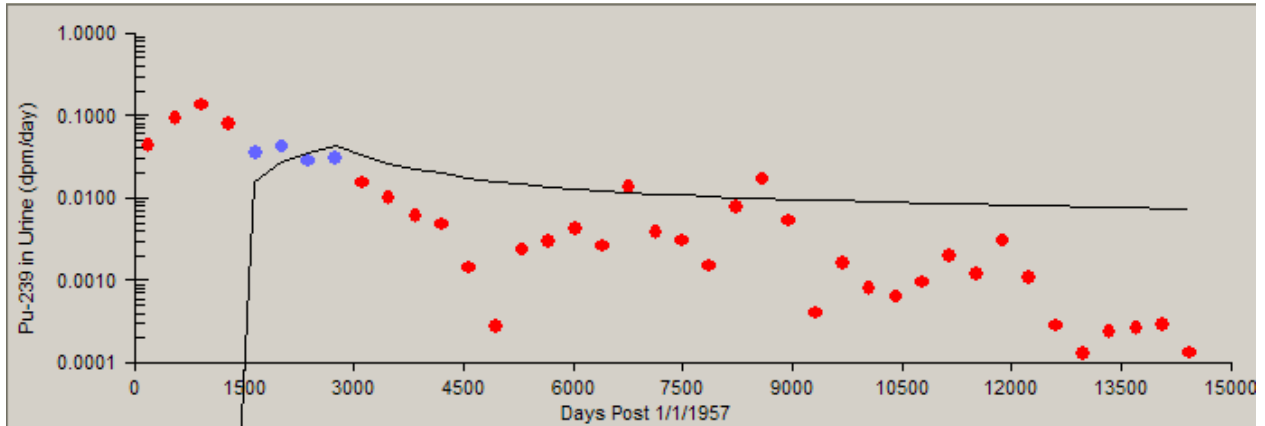


Figure B-26. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1961 through December 1964.

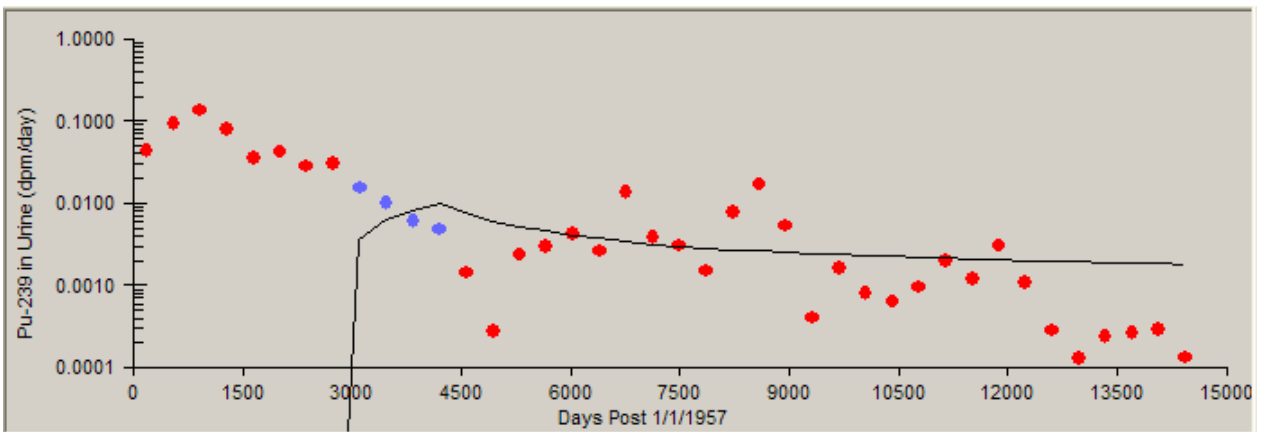


Figure B-27. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1965 through December 1968.

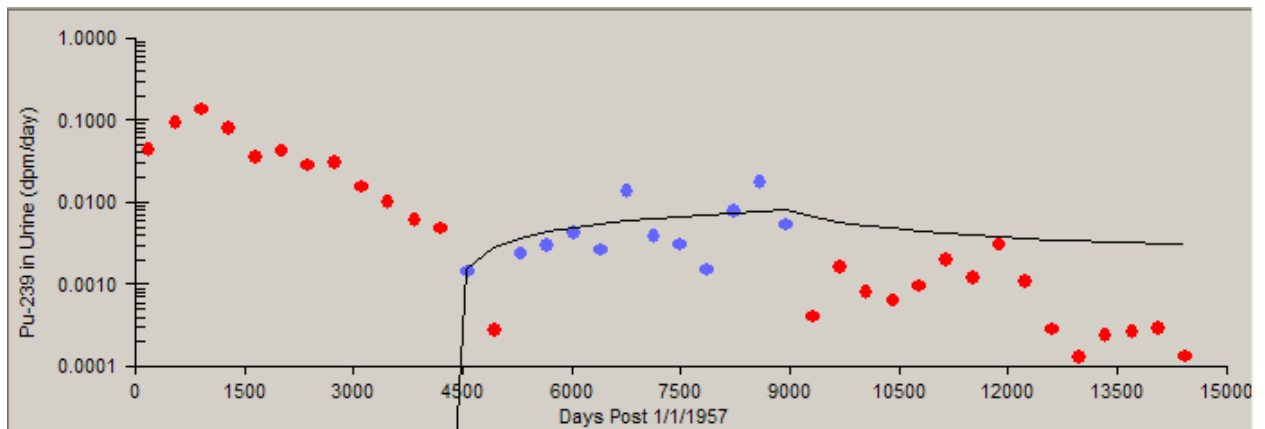


Figure B-28. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1969 through December 1981.

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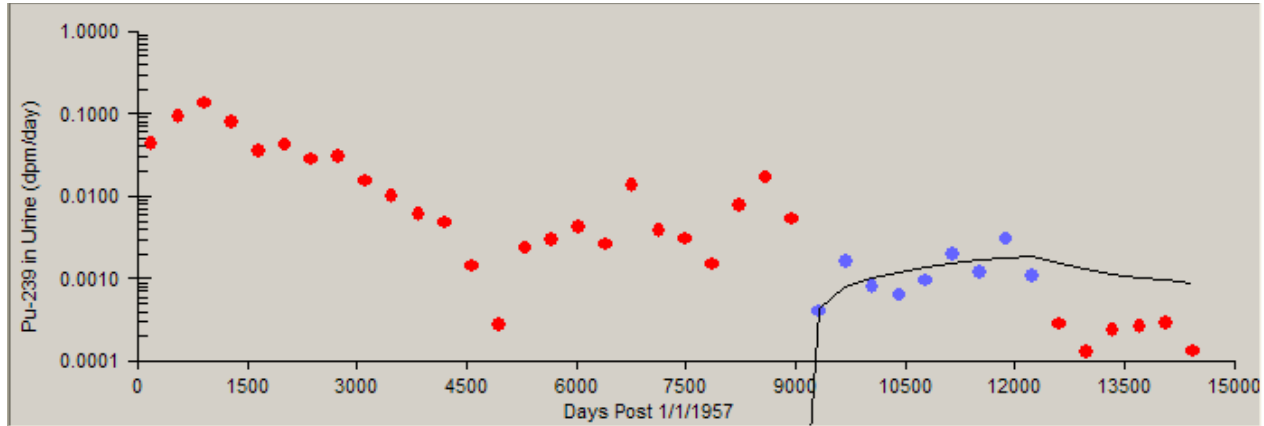


Figure B-29. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1982 through December 1990.

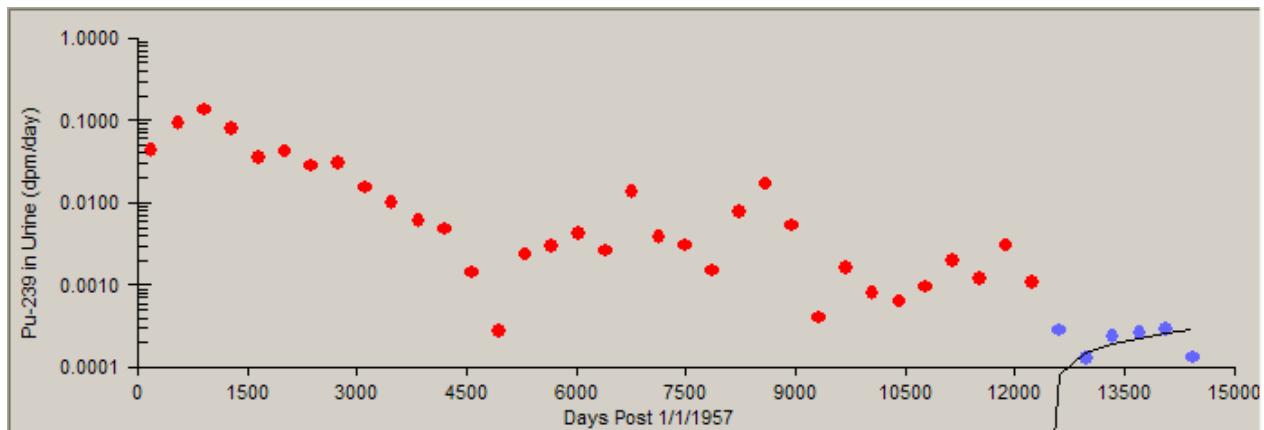


Figure B-30. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1957 through December 1996.

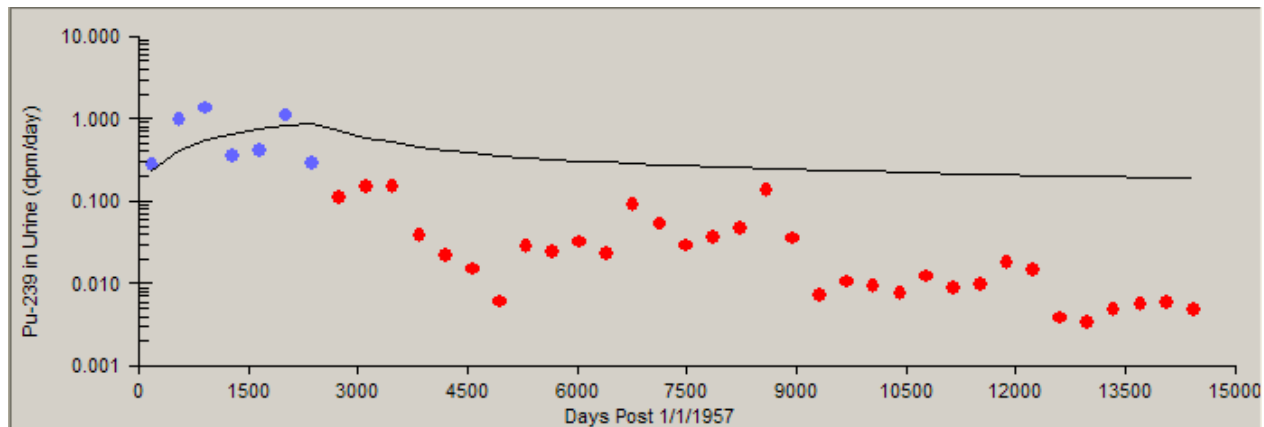


Figure B-31. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1957 through December 1963.



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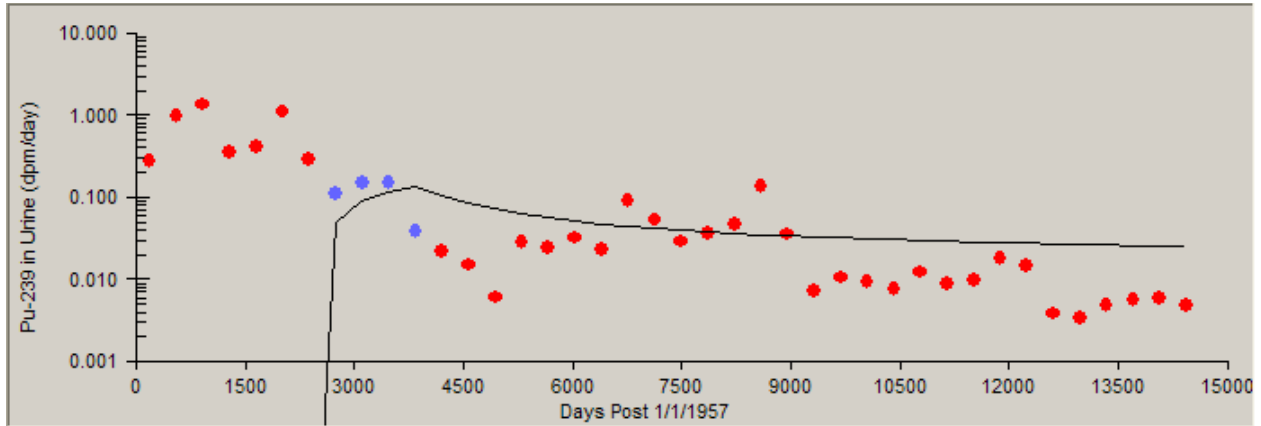


Figure B-32. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1964 through December 1967.

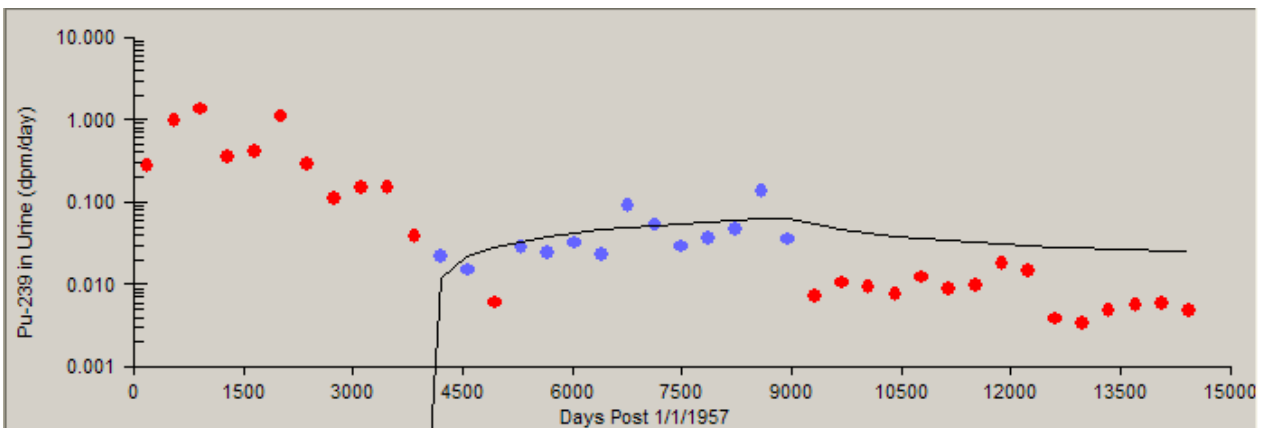


Figure B-33. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1968 through December 1981.

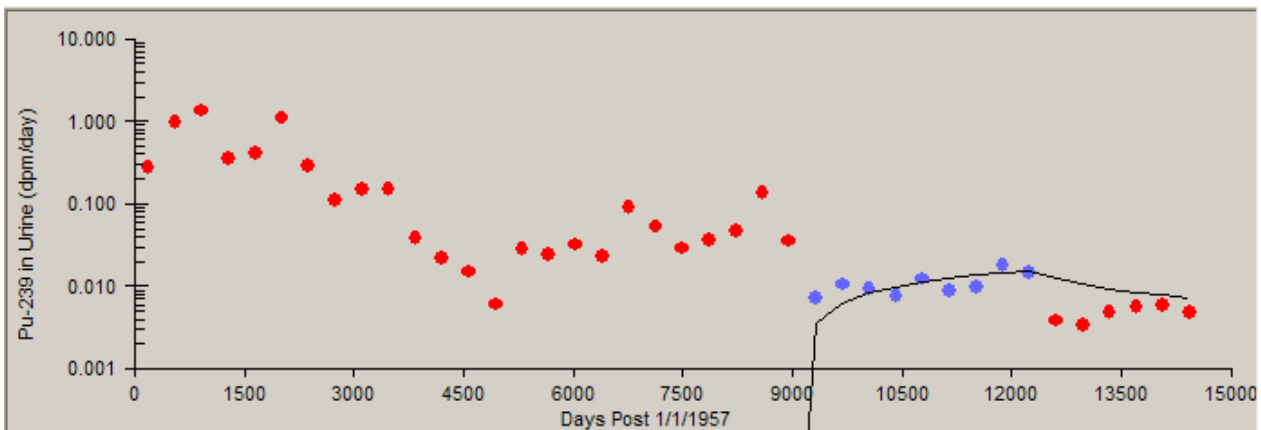


Figure B-34. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1982 through December 1990.

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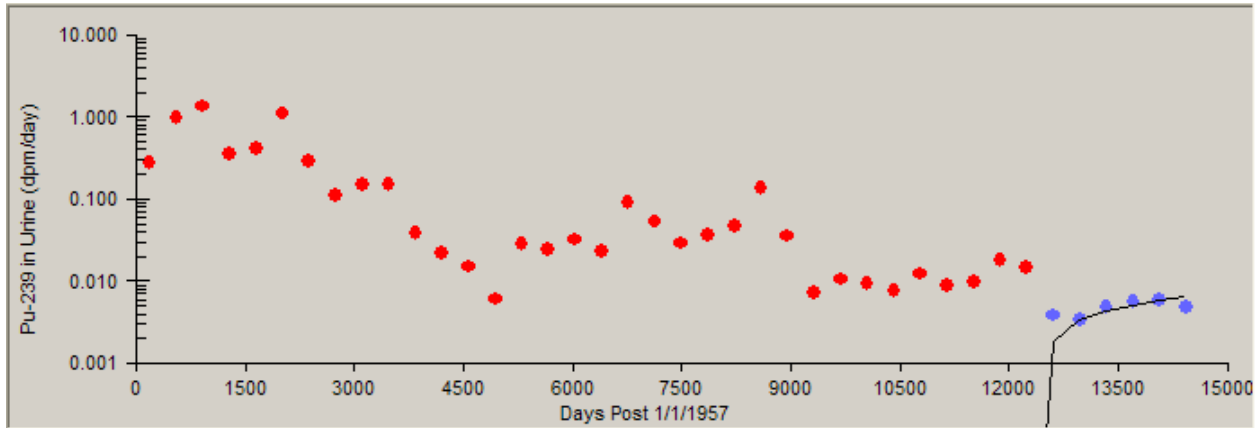


Figure B-35. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M plutonium from January 1991 through December 1996.

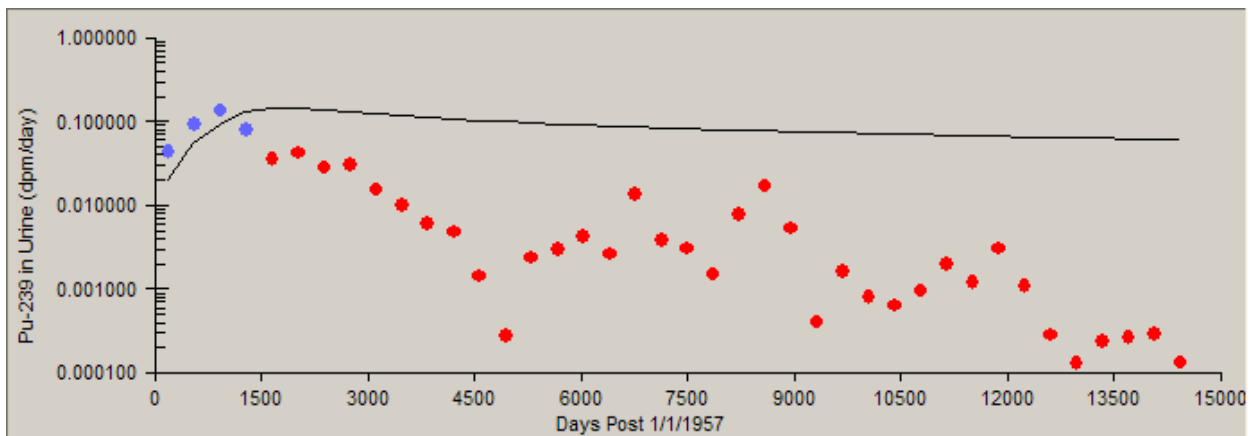


Figure B-36. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1957 through December 1960.

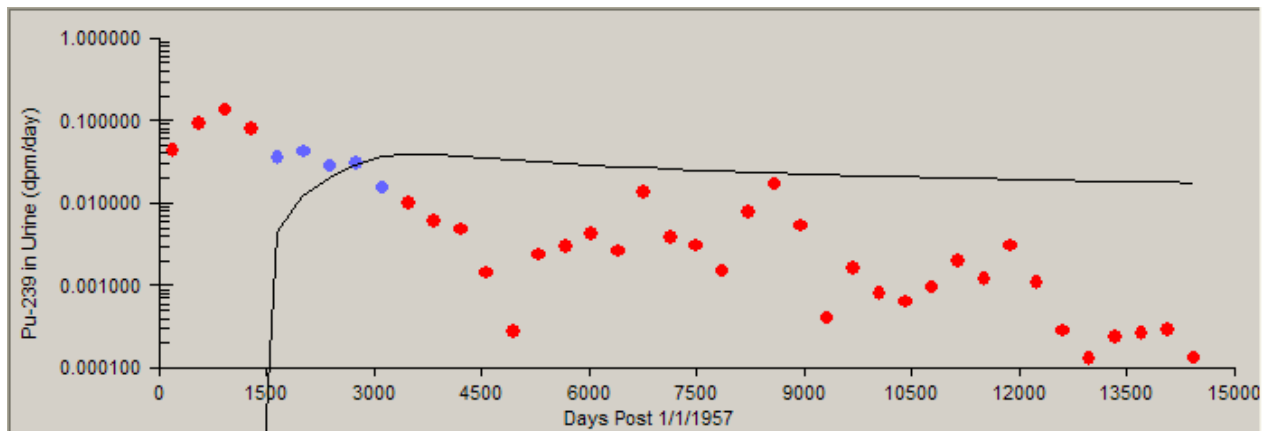


Figure B-37. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1961 through December 1965.

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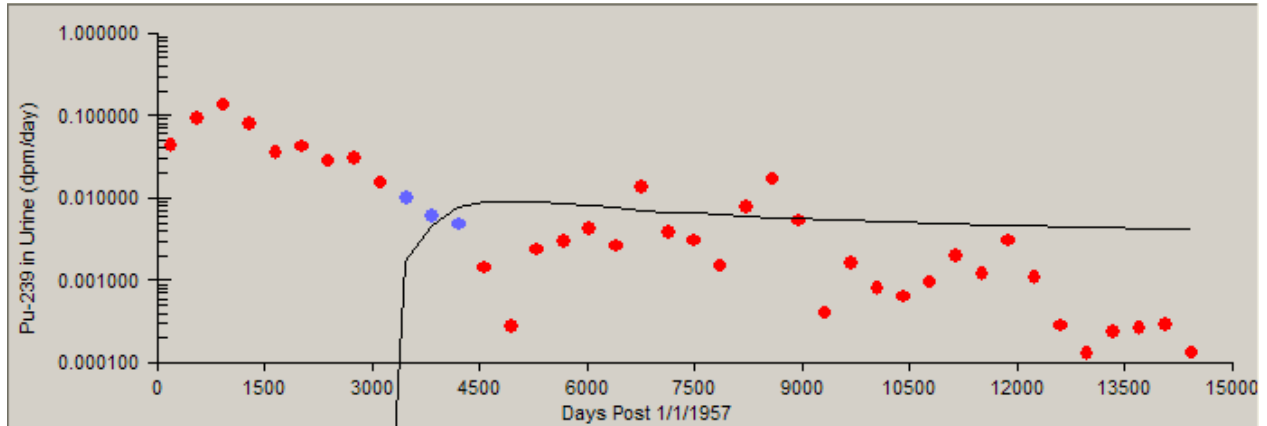


Figure B-38. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1966 through December 1968.

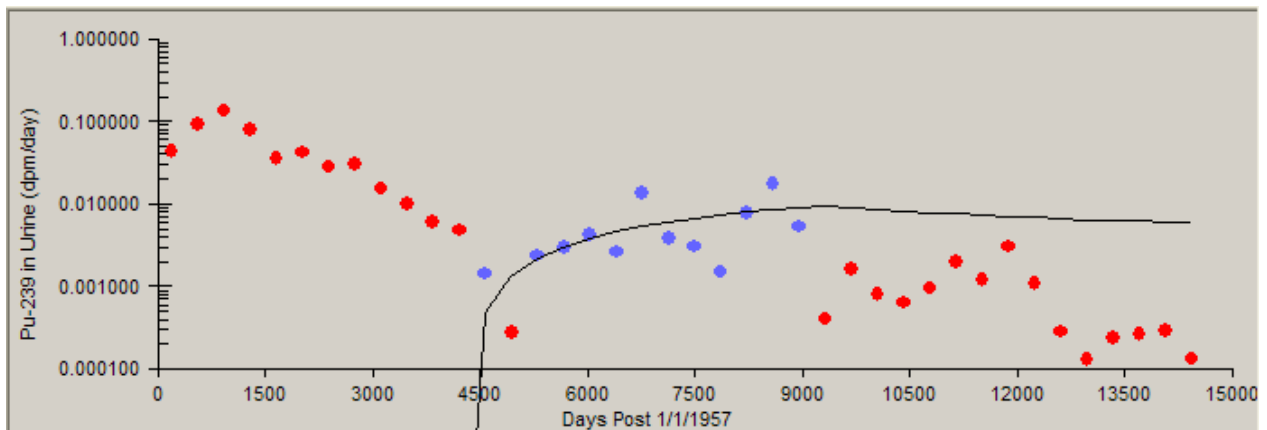


Figure B-39. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1969 through December 1981.

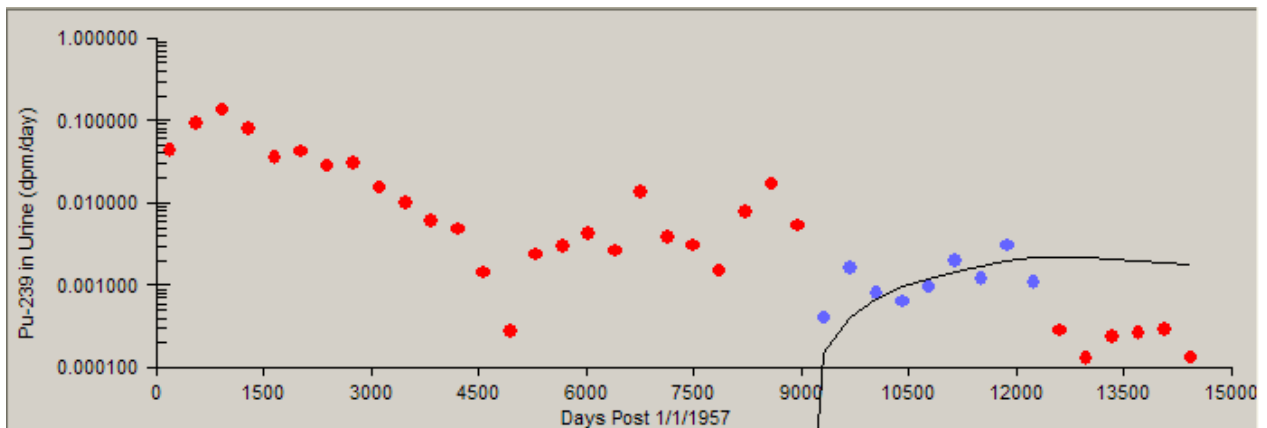


Figure B-40. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1982 through December 1990.

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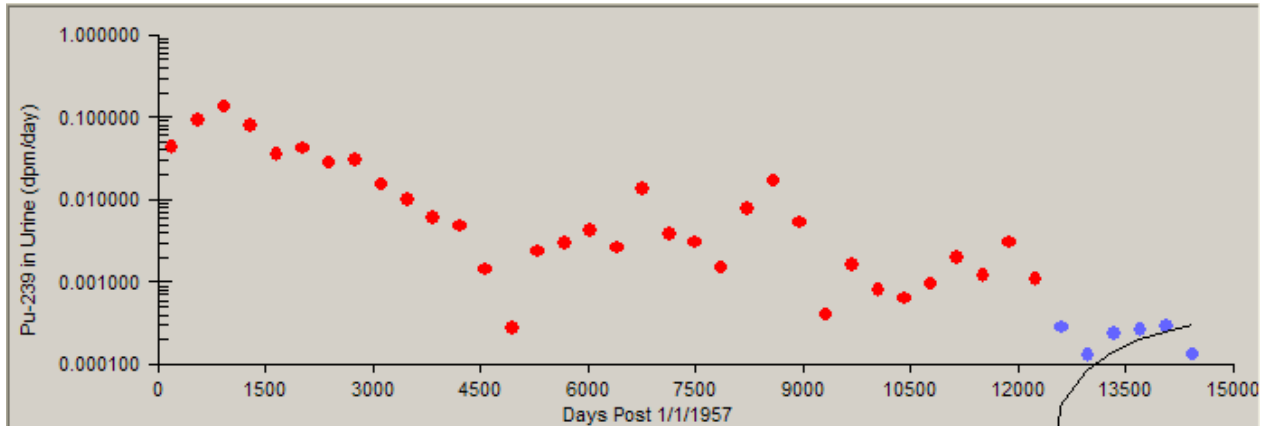


Figure B-41. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1991 through December 1996.

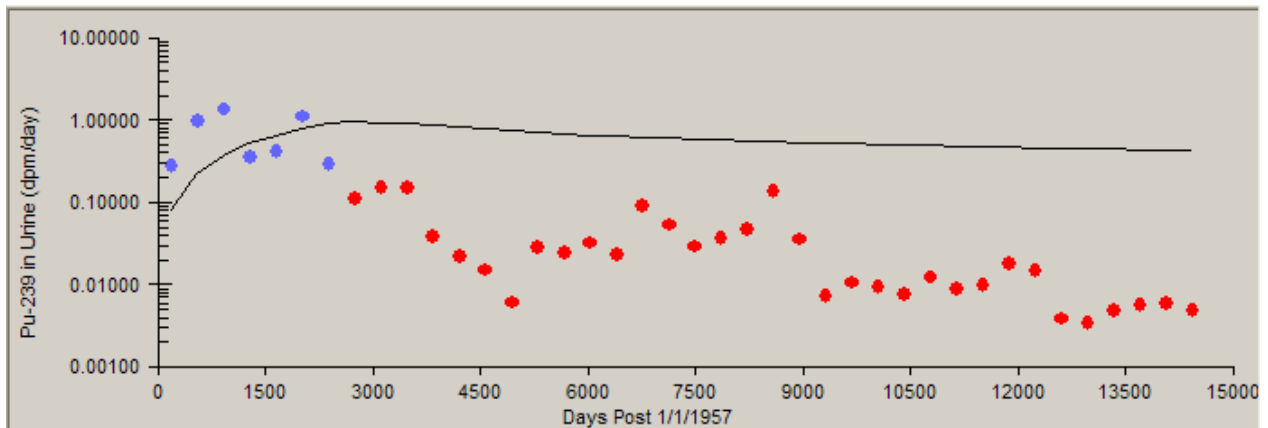


Figure B-42. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1957 through December 1963.

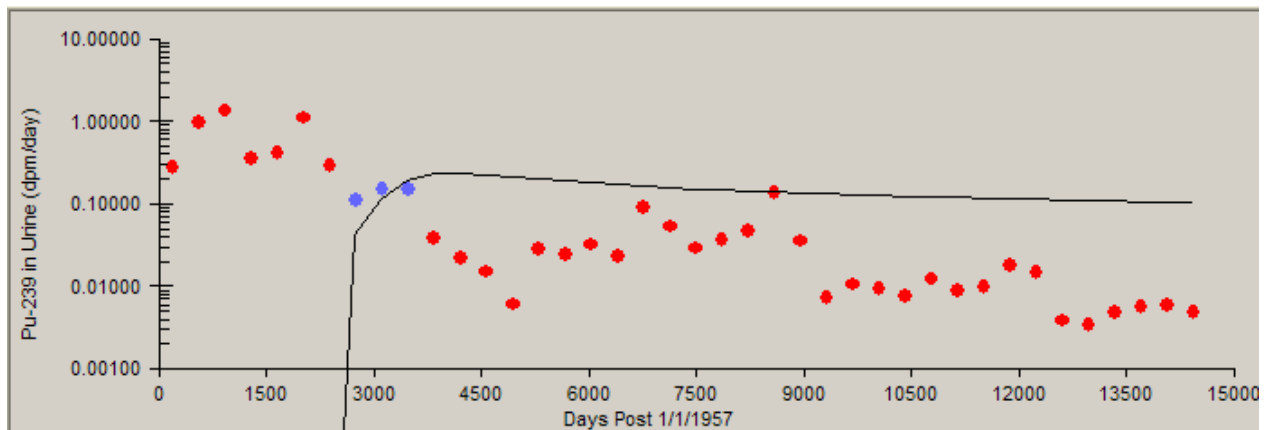


Figure B-43. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1964 through December 1966.

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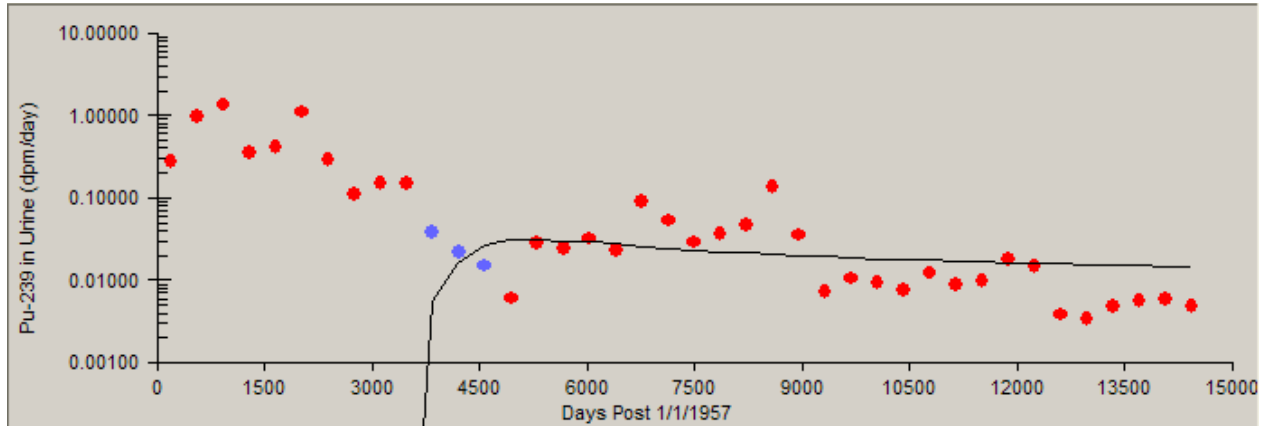


Figure B-44. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1967 through December 1969.

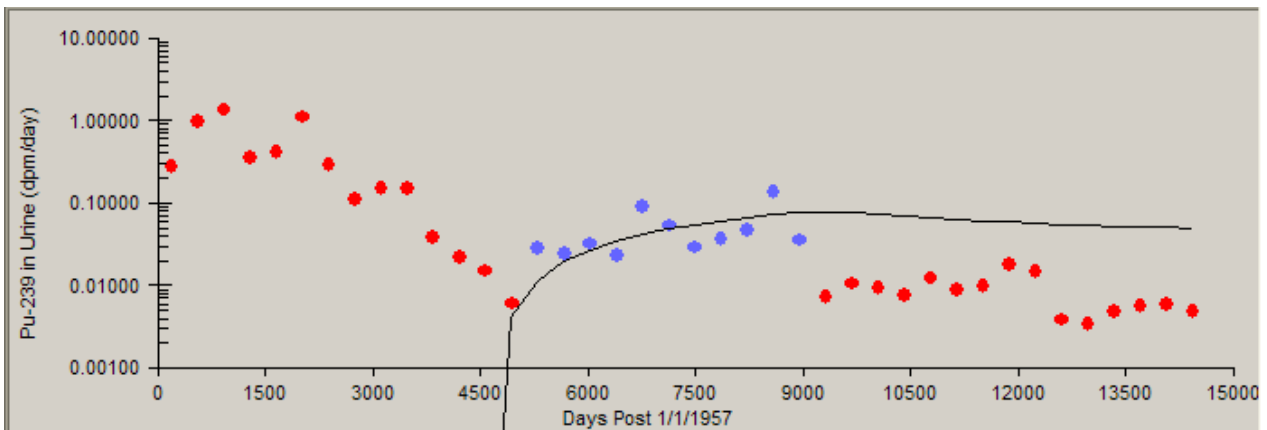


Figure B-45. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1970 through December 1981.

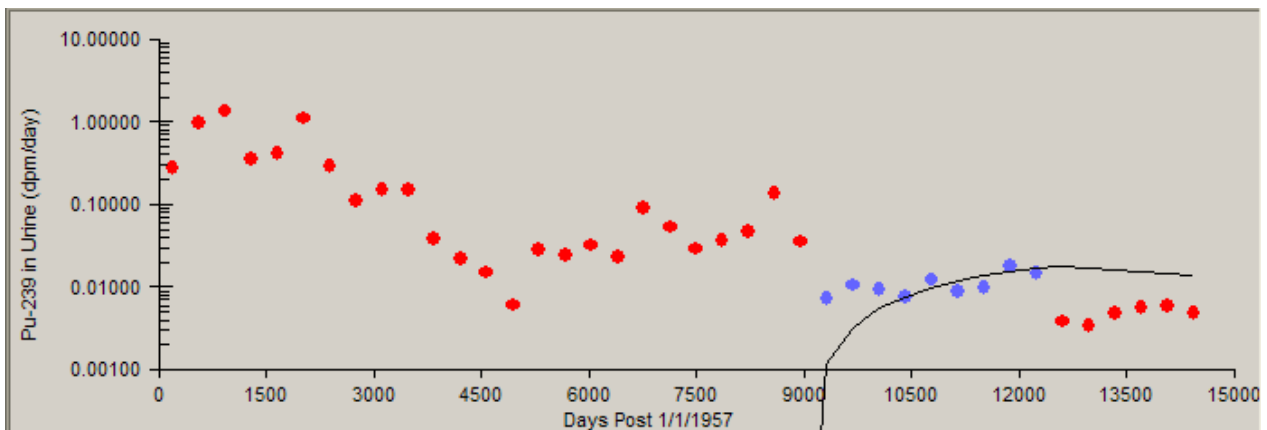


Figure B-46. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1982 through December 1990.

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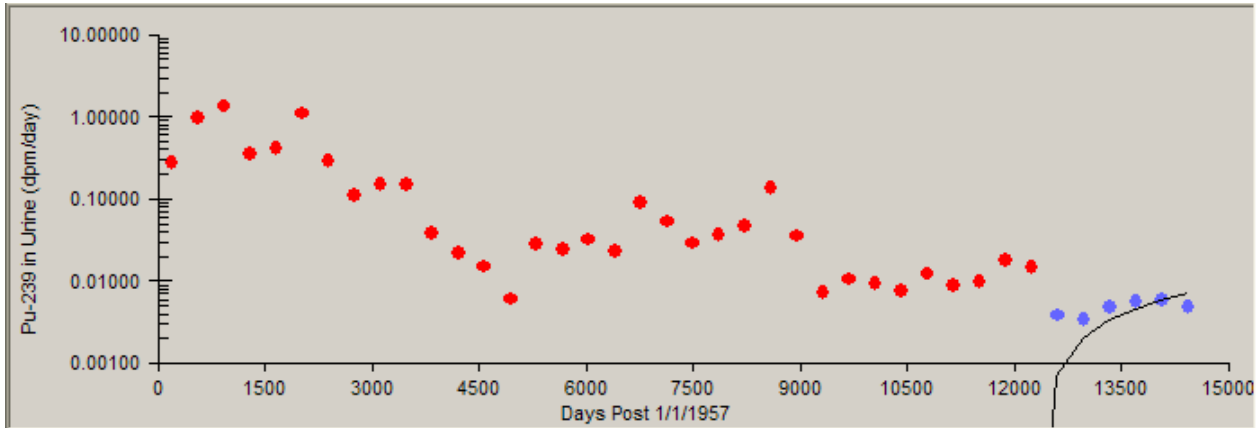


Figure B-47. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type S plutonium from January 1991 through December 1996.

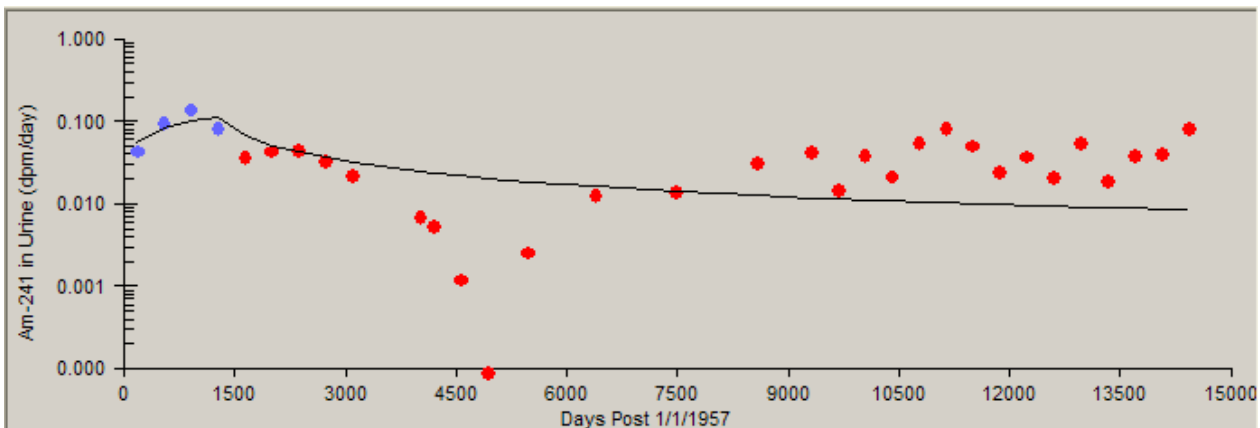


Figure B-48. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1957 through December 1960.

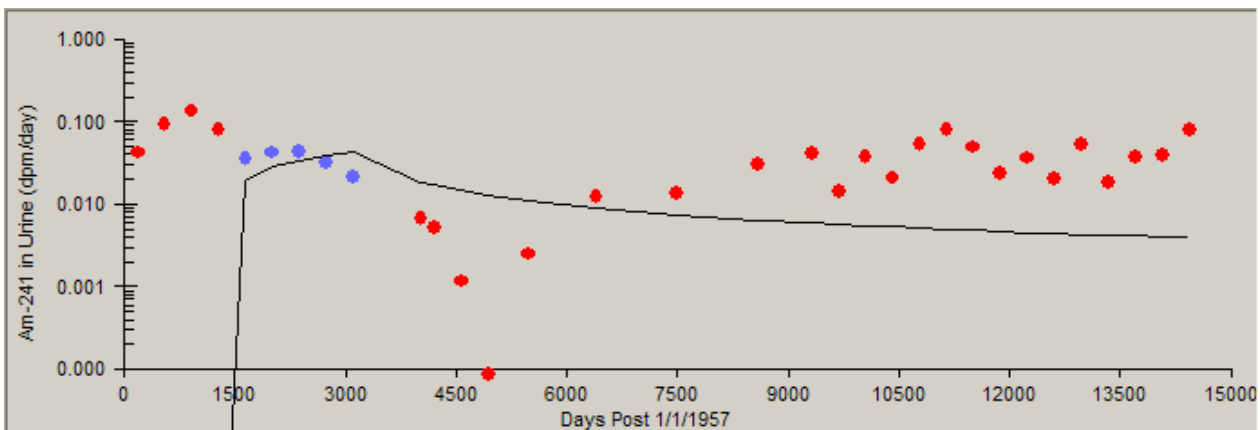


Figure B-49. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1961 through December 1965.

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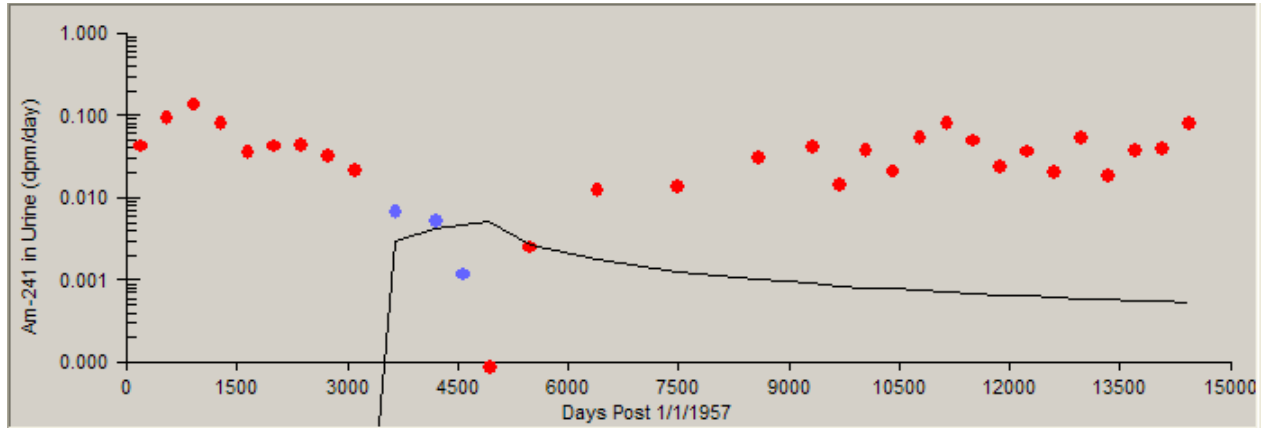


Figure B-50. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1966 through December 1970.

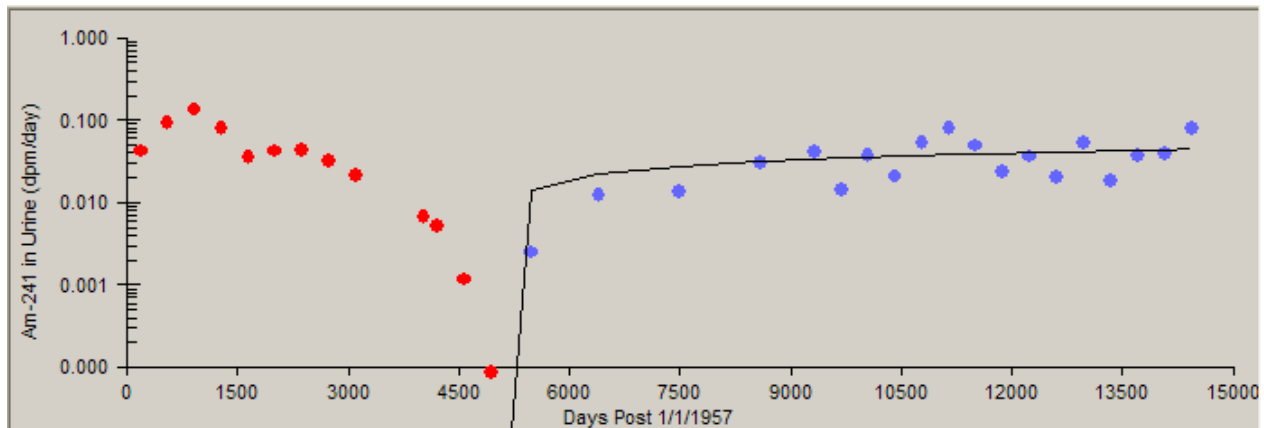


Figure B-51. Predicted and observed 50th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1973 through December 1979.

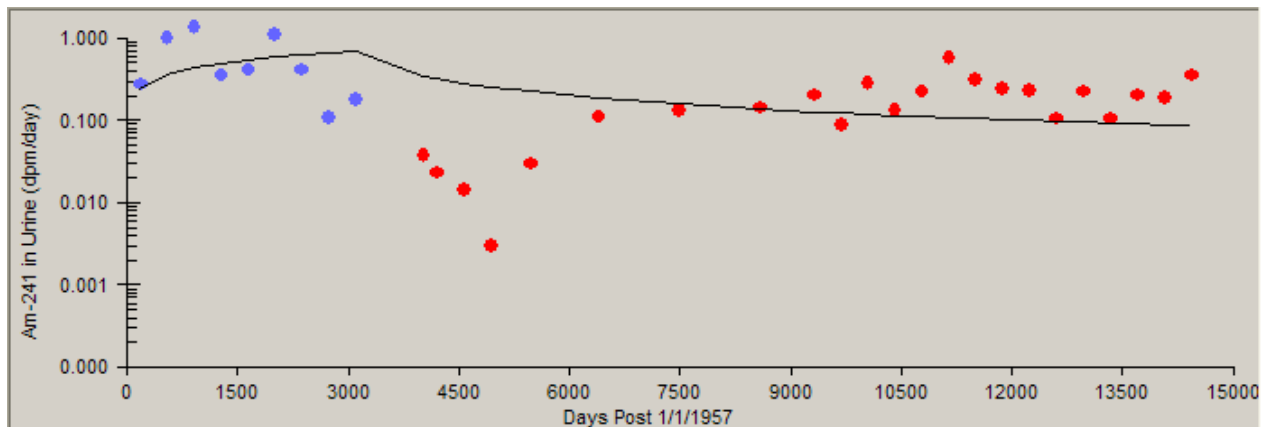


Figure B-52. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1957 through December 1965.

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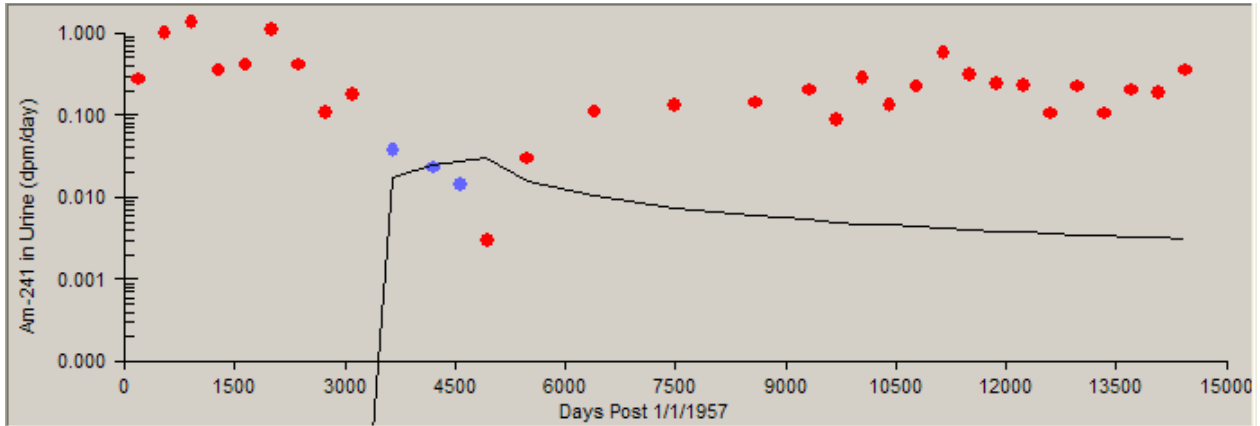


Figure B-53. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1966 through December 1970.

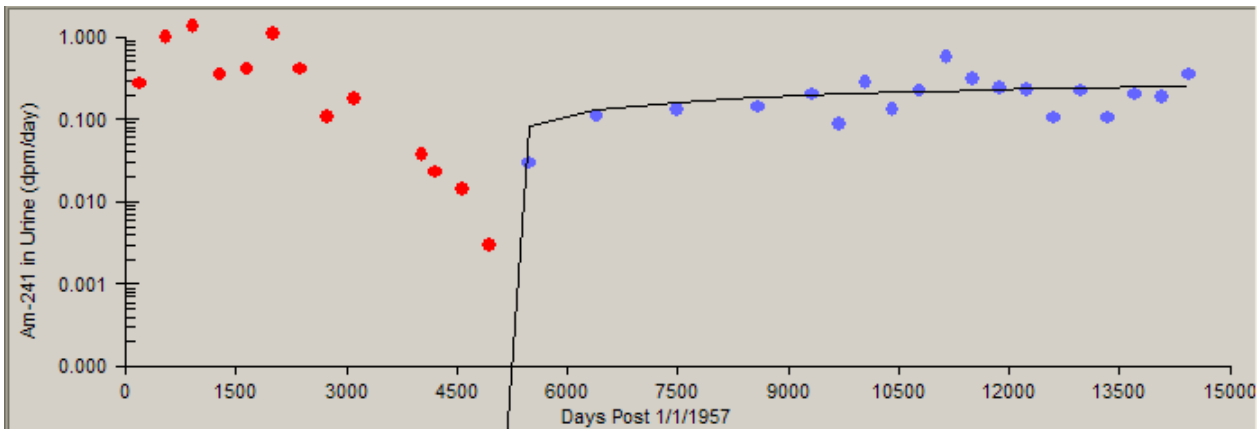


Figure B-54. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type M <sup>241</sup>Am from January 1971 through December 1996.

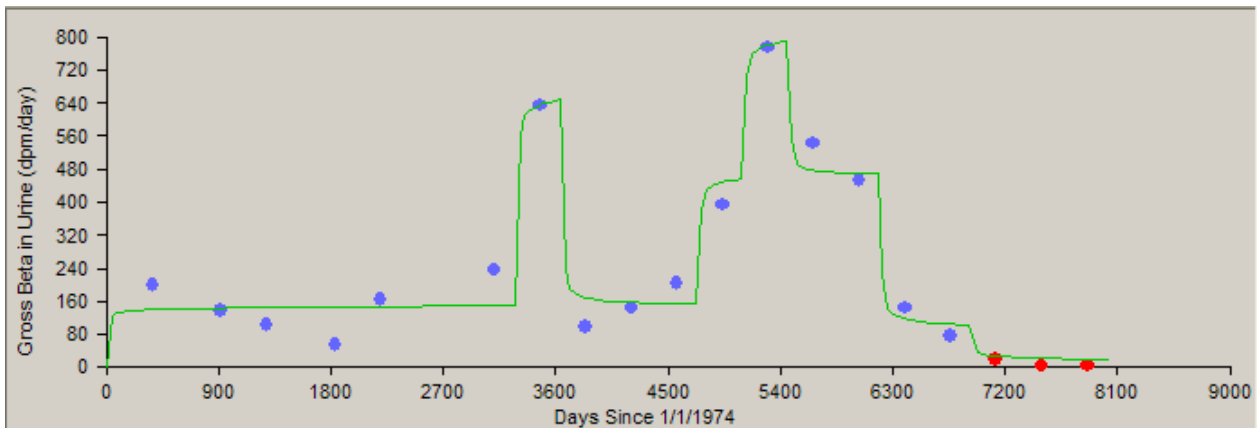


Figure B-55. Predicted and observed 50th-percentile gross beta urinary excretion assuming a chronic inhalation intake of Type F <sup>90</sup>Sr from January 1974 through December 1992.



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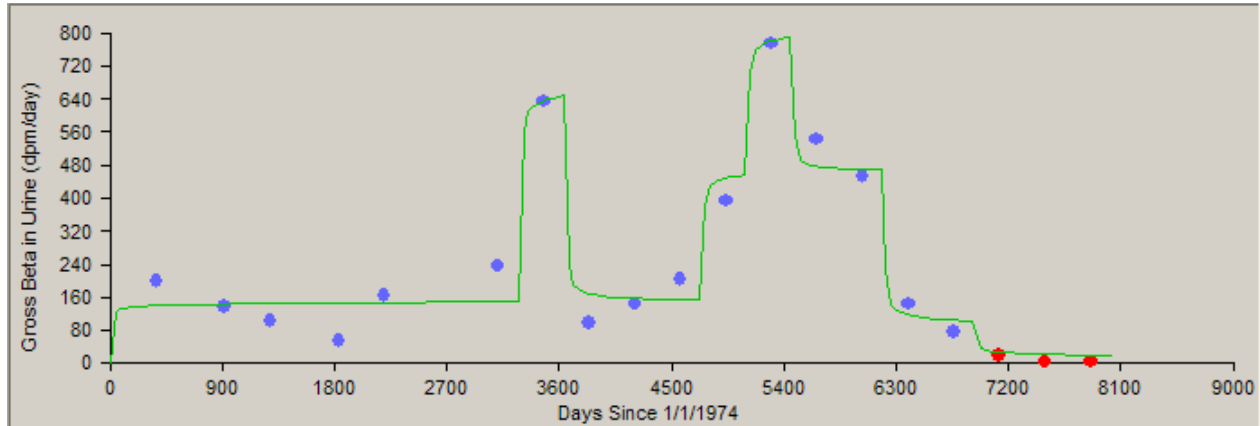


Figure B-56. Predicted and observed 84th-percentile gross beta urinary excretion assuming a chronic inhalation intake of Type F <sup>90</sup>Sr from January 1974 through December 1992.

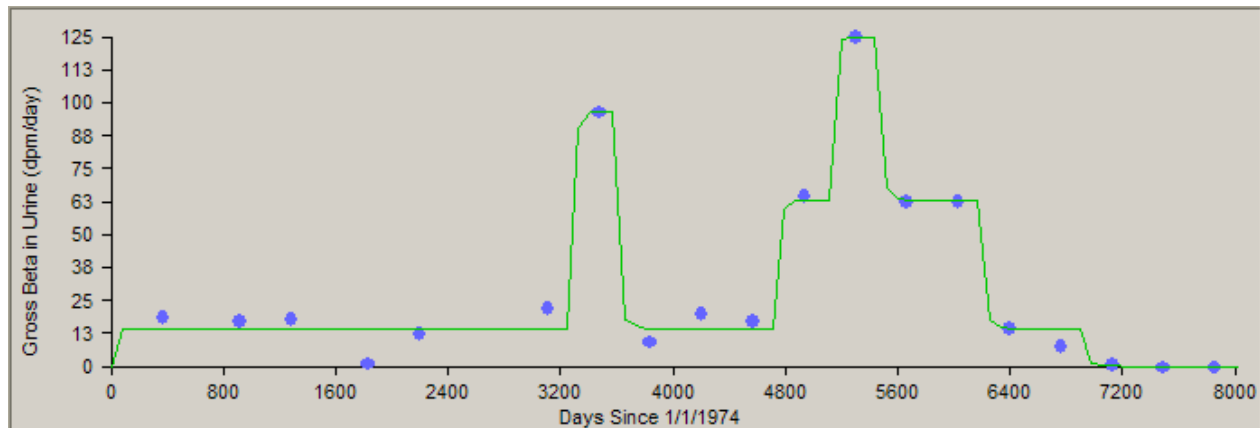


Figure B-57. Predicted and observed 50th-percentile gross beta urinary excretion assuming a chronic inhalation intake of Type F <sup>103</sup>Ru from January 1974 through December 1992.

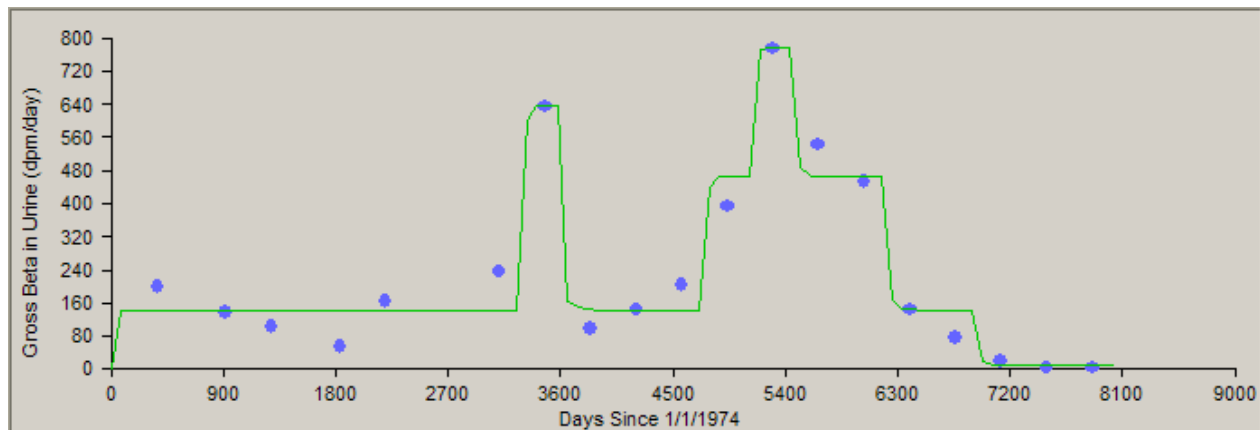


Figure B-58. Predicted and observed 84th-percentile gross beta urinary excretion assuming a chronic inhalation intake of Type F <sup>103</sup>Ru from January 1974 through December 1992.

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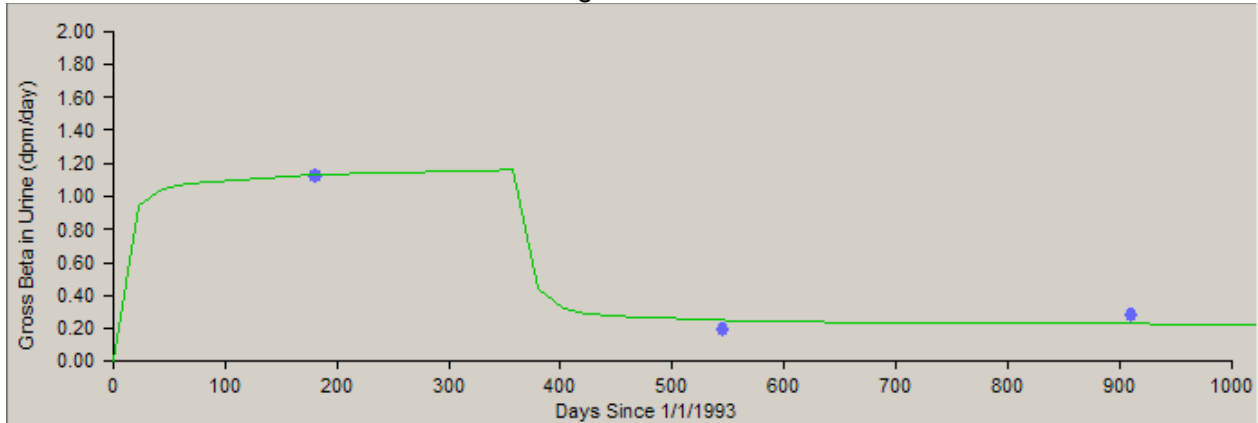


Figure B-59. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type F <sup>90</sup>Sr from January 1993 through December 1995.

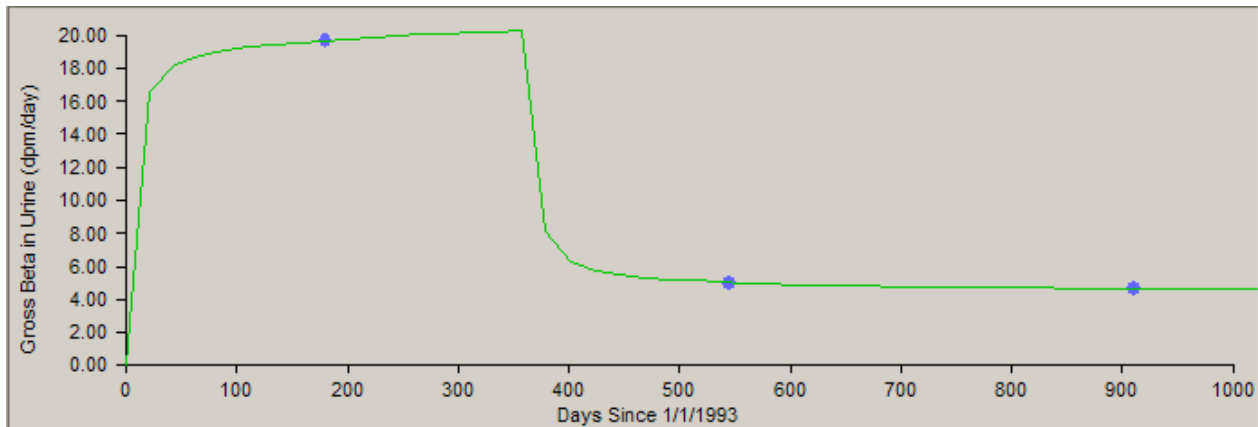


Figure B-60. Predicted and observed 84th-percentile urinary excretion assuming a chronic inhalation intake of Type F <sup>90</sup>Sr from January 1993 through December 1995.

**ATTACHMENT C**  
**SCALING OF COWORKER DOSE INTAKES CONSISTENT**  
**WITH TYPE AND DURATION OF EXPOSURES**

**Assigned Intakes Using Case-Specific Information**

The major presumptive exposures at LLNL were made up of the primary radionuclides plutonium, uranium, and tritium. Other radionuclides might have resulted in intakes to personnel but, due to the small amounts of material, the limited operations, and the use of engineering controls such as hot cells, fume hoods, gloveboxes, and dry boxes, potential exposures are relatively unlikely. When exposures are possible, the intakes were likely to be small, and highly dependent on work category. Work locations are another important consideration when evaluating for a potential unmonitored intake. Although it is not possible in all cases to precisely locate LLNL employees, it is certainly true that the nonprimary radionuclides presented a limited hazard to most LLNL personnel. In this attachment, a method is presented to scale assigned intakes based on job category and likely duration of exposure to limit the margin of overestimation.

**Level of contact.** For the purpose of dose reconstruction, the amount of exposure should be considered when assigning coworker dose intakes. For personnel with full contact with the material, the 95th percentile of the calculated coworker intakes should be assigned. For others, the 50th percentile is more appropriate, as listed in Table C-1.

Table C-1. Levels of contact for certain job categories.

| Contact level                  | Description   | Example job titles  | Percentile of CW intake            |
|--------------------------------|---|---|------------------------------------|
| Primary contact                | Contact with material in unencapsulated and uncontained form. Normal contact with the material is under less-controlled environments such as in fume hoods and inside chemical apparatus.                               | Chemists, researchers, metallurgists  | 95th                               |
| Secondary contact              | Normal contact with the material is in containment or process equipment, occasional contact with airborne material during excursions.   | Operators, janitors, decommissioning and decontamination workers, health physics personnel routinely assigned   | 50th                               |
| Incidental contact             | Contact with contamination and airborne radioactive material only during an excursion or incident or as part of an exposure during a maintenance, repair, waste disposal, decontamination, or decommissioning activity. | Janitors, decommissioning and decontamination workers, maintenance, health physics personnel not routinely assigned construction activities associated with waste disposal or demolition of legacy structures | 50th                               |
| Contact unlikely, but presumed | Nonradiological job categories, construction activities not associated with waste or decontamination and decommissioning, but evidence supports a potential incidental exposure.  | Construction activities   | 50th                               |
| Contact unlikely, not presumed | Nonradiological job categories, and no evidence to suggest a presumptive exposure.  |   | Assume environmental internal dose |