

ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities I Dade Moeller I MJW Technical Services

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FOR DOCUMENTS MARKED AS A TOTAL REWRITE, REVISION, OR PAGE CHANGE, REPLACE THE PRIOR REVISION AND DISCARD / DESTROY ALL COPIES OF THE PRIOR REVISION.

PUBLICATION RECORD

EFFECTIVE DATE	REVISION NUMBER	DESCRIPTION
10/15/2003	00	New document to establish TBD for occupational internal dose – Section 5. First approved issue. Initiated by Edward D. Scalsky.
11/24/2004	01	Approved revision initiated to discuss historical limits and tolerance dose; discussion of separations plant (1944-46) and 231-Z (1945- 46); clarified intakes in the 300 Area uranium fabrication and the laundry facilities; expanded information on ²⁴¹ Am and ²⁴¹ Am MDAs for 1946 and 1967-6/1969; added Section 5.2.4.1 on assignment of tritium doses; added Table 5.2.5-5 (MDAs for nonroutine uranium excreta analyses); revised Table 5.2.6-1 (routine fission product urinalysis detection levels); added discussion of ²¹⁴ Bi and ²⁰⁸ Tl in whole-body counting (Section 5.3.1); expanded Section 5.7 on unmonitored workers to provide additional discussion, tables, and instructions to dose reconstructors for specific areas on the site; included listing of references; plus corrected miscellaneous typos, and table number changes. Incorporates formal internal and NIOSH review comments. Initiated by Edward D. Scalsky
06/22/2007	02	Approved revision initiated to revise or add information in Section 5. Section 5.1: updated tolerance air concentrations and assigned intakes for 1944-48; added summary of Ikenberry report on thyroid monitoring 1945-46; added fission product intakes for 1947. Section 5.2: added statement on types of urinalysis kits normally used. Section 5.2.1: added information about pure ²³⁸ Pu work in 325 Bldg.; added statement to use 5-yr-old Pu mixture for years prior to 1951. Section 5.2.2: added statement about ²⁴⁴ Am recovery operation in PFP in 1960-70s. Section 5.2.3: added stated about ²⁴⁴ Cm recovery in 325 Bldg. Section 5.2.5: added statement about work with 37% enriched U; changed period for 4 µg/L reporting level to 1974; increased ²³⁹ Pu contaminant level in RU to 0.8 nCi/gU; added information about ²³³ U production. Section 5.2.8: added information about promethium use. Section 5.2.11: added information about neptunium urinalyses. Section 5.2.11: added information about neptunium urinalyses and ³⁵ S urinalyses. Revised or added information to Section 5 tables. Introduction: added new boilerplate. All table numbers were reformatted. Section 5.1: removed reliance on air samples and referenced co-worker study (OTIB-0039), Section 5.2: added statement on types of urinalysis kits normally used. Sec. 5.2.1: added information about pure ²³⁹ Pu work in 325 Bldg; added statement to use fresh Pu for 1944-49 and 5-yr-old Pu mixture for 1950-54. Sec. 5.2.2: added statement about ²⁴⁴ Cm recovery operation in PFP in 1960-70s. Sec. 5.2.3: added statement about ²⁴⁴ Cm recovery in 325 Bldg. Sec. 5.2.4: added information about tritium in the 400 Area drinking water. Sec. 5.2.5: Table 5-13: raised Pu-alpha contamination level to 0.8 nCi/gU; added statement about work with 37% enriched U; changed period for 4 µg/L reporting level to 1974; added information about ²³³ U production. Sec. 5.2.8: added some information about promethium use. Sec. 5.2.9: removed discussion about background levels of ²¹⁰ Po. Sec. 5.2.10: added

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		revised Table 5.3.1-1 (5-22) to clarify MDAs, decision levels and reporting levels. Sec. 5.3.4: totally new section on thorium exposure and thorium monitoring. Sec. 5.4: replaced the old mixture rules with use of Table 5.7-2 (5-25); Sec. 5.6.2: (chronic intakes) deleted entire section. Sec. 5.7 (now 5.6): added information about tritium in the 400 area drinking water; added reference to OTIB-0039 and revised Table 5-25 accordingly. Moved old Appendix D into this document as Attachment A; took out all the tables on chronic intakes. Constitutes a total rewrite of the document. Incorporates NIOSH formal review comments. Many grammatical and format changes throughout. Table 5-12: removed the uranium specific activity data for DU and natural U; added statement to use IMBA defaults to footnote a. Section 5.3.4: added table showing buildings with thorium exposure; added text and table explaining adjustment factors for whole body measurements of ²³² Th and associated intakes; added thoron doses for workers processing thorium in 3722 and 3732 buildings. Section 5.6: clarified possible absorption types for Pu and U in the specificfacilities section; added thoron to Table 5-30; revised tritium intakes for unmonitored workers due a coworker analysis of tritium doses. New Tables 5-27 and 5-28 providing ²³² Th intakes based on whole body counting at long times after end of chronic intakes. Section 5.3.4: revised thoron WLMs, removed references to 6-mrem tritium dose and stayed with intake values; clarified how the tritium intake values were determined for the 1955-60, 1970-71, and 1973-83 time periods; corrected a math error resulting in slightly increased intakes for 1984, 85, and 86. Includes Attributions and Annotations and additional references. Incorporates formal internal review comments for Attributions and Annotations section. This revision results in an increase in assigned dose and a PER is required. Training required: As determined by the Task Manager. Initiated by Donald E. Bihl and Edward D. Scalsky.
01/07/2010	03	Revision to add new SEC statement in Section 5.1. All references to OTIB-039 were removed. OTIB-039 was added as Attachment C. All mention of assigning intakes for unmonitored workers for Np, Po-210, or thorium were removed. Attachment A was split with REX codes becoming a new Attachment B and tolerance limits remaining as Attachment A. Table 5-1 on bioassay codes were moved to Attachment B. Text on records missing in REX was added to Section 5.2. In Section 5.2.1, text was added on change in bioassay frequency circa 1960. Text on history of Am exposure and start of bioassay was added in Section 5.2.2. Text on history of Cm exposure was added to Section 5.2.3. Text on history of tritium exposure and discussion of metal tritides and OBT was added to Section 5.2.4. Added new information on sensitivity in 1950. In Table 5-10, a new footnote b on reporting level in 1964 was added. In Table 5-14, there was a change in MDA for the period 9/1964 through 1973. More explanatory text was added to the footnotes. Additional history of ²³³ U exposure and bioassay method in 1970 was added to Section 5.2.5. Updated impurity concentrations in the ²³³ U and added new Tables 5-16 and 5-17 on impurities. In Section 5.2.8, Information was added in regard to ¹⁴⁷ Pm urinalysis development in

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		1962. Added MDA information on urinalysis 1963-64. In vivo counting for ¹⁴⁷ Pm was mentioned. In Section 5.2.9, historical material was removed. New information was referenced in the Hanford Site Description. Added information on 1948 Po-210 urinalysis procedure and statement about monitoring in 1954. Text was added to Section 5.2.10 on purity of Np. Added information about urinalysis method in 1960. Text was added to Section 5.2.11 on thorium urinalysis method circa 1955. Text was removed in Section 5.3.4 on historical exposure to thorium. New information was referenced in the Hanford Site Description. Buildings were added to Table 5-26. Mentioned early thorium urinalysis procedure circa 1955 and first whole body counting 1962-63. New intake guidance for alpha-emitters in tank farm mixtures was added to Section 5.4.1. In Section 5.6, added reference to OTIB-54 for fission product intakes. Corrected operating dates for the Fast Flux Test Facility. Mentioned that routine thyroid counts extended into the 1950s. In Table 5-30, added new intake values for tritium for 1955 and discussed reason in the text. Added statements on the tritium bioassay analysis as discussed in Attachment C. Added guidance for unmonitored worker intakes of ²³³ U in Z Plant. Added new guidance for many of the specific buildings and added some new buildings to the list. Updated Table 5-31 to reflect new guidance in the text. Incorporates formal internal and NIOSH review comments. Provides additional guidance for assigning intakes for exotic radionuclides to unmonitored workers in Section 5.6. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Donald E. Bihl and Fred Duncan.
10/20/2010	04	Revision to make corrections to GSDs and 95 th percentile intake rates in Attachment C (coworker study) for all radionuclides (plutonium, uranium, strontium-90, promethium-137, zinc-65, sodium- 24, cesium-137). Incorporates formal internal and NIOSH review comments. Training required: As determined by the Objective Manager. Initiated by Matthew Arno and Fred Duncan.

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ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
Bq	becquerel
CFR	Code of Federal Regulations
Ci	curie
cm	centimeter
d	day
D&D	decontamination and decommissioning
DAC	derived air concentration
DDCP	dibutyl N,N-diethylcarbamylphosphonate
DIL	derived investigation level
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
dpm	disintegrations per minute
DTPA	diethylenetriaminepentaacetic acid
DU	depleted uranium
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
FFTF	Fast Flux Test Facility
FR	<i>Federal Register</i>
ft	feet
g	gram
GeLi	lithium-drifted germanium
GM	geometric mean
GSD	geometric standard deviation
HERB	Health-Related Energy Research Branch
HIE	Hanford Internal Exposure (database)
HPGe	high-purity germanium
hr	hour
HT	elemental tritium (tritiated gas)
HTO	tritiated water vapor
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Assessment
in.	inch
IREP	Interactive RadioEpidemiological Program
keV	kiloelectron-volt
KPA	kinetic phosphorescence analysis
LANL	Los Alamos National Laboratory
LEPD	low-energy photon detector (also a computer code to indicate use of the LEPD)
LSL-II	Life Sciences Laboratory-II
m	meter
MDA	minimum detectable activity or, for elemental uranium, minimum detectable amount

MDC	minimum detectable concentration
MeV	megaelectron-volt
min	minute
mL	milliliter
MPBB	maximum permissible body burden
MPC	maximum permissible concentration
mrem	millirem
mrep	millirep
mSv	millisievert
nCi	nanocurie
NCRP	National Council on Radiation Protection and Measurements
NIOSH	National Institute for Occupational Safety and Health
NU	natural uranium
ORAU	Oak Ridge Associated Universities
ORE	Occupational Radiological Exposure (database)
pCi	picocurie
PNL	Pacific Northwest Laboratory
PNNL	Pacific Northwest National Laboratory
POC	probability of causation
ppb	parts per billion
ppm	parts per million
PRTR	Plutonium Recycle Test Reactor
PUREX	Plutonium–Uranium Extraction (facility)
RDA	reliably detectable activity
REDOX	Reduction-Oxidation (plant)
REX	Radiological Exposure (database)
RU	recycled uranium
SEC	Special Exposure Cohort
SRDB Ref ID	Site Research Database Reference Identification (number)
SRS	Savannah River Site
TBD	technical basis document
TPU	total propagated uncertainty
TTA	thenoyl trifluoroacetone
U.S.C.	United States Code
UNC	United Nuclear Company
UST	United States Testing Company
yr	year
μCi	microcurie
μg	microgram
μm	micrometer
§	section or sections

5.1 **INTRODUCTION**

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

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

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer ... was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation¹] guidelines established under subsection (c) ..." [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work (NIOSH 2007).

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

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

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

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5.1.1 <u>Purpose</u>

This TBD documents the internal dosimetry program at the Hanford Site and provides the technical basis to be used to evaluate the internal occupational radiation dose for EEOICPA claims.

5.1.2 Special Exposure Cohort Petition Information for Hanford

The current status of Special Exposure Cohort (SEC) petitions for Hanford are:

Classes Added to the SEC

- Employees of DOE, its predecessor agencies, or DOE contractors or subcontractors who were
 monitored or should have been monitored for internal radiological exposures while working at
 the Hanford Engineer Works in the 300 Area fuel fabrication and research facilities from
 October 1, 1943 through August 31, 1946; the 200 Area plutonium separation facilities from
 November 1, 1944 through August 31, 1946; or the 100 B, D, and F reactor areas from
 September 1, 1944 through August 31, 1946; 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 SEC (72 FR 55214).
- Employees of DOE, its predecessor agencies, and DOE contractors or subcontractors who worked from September 1, 1946 through December 31, 1961 in the 300 area, or January 1, 1949 through December 31, 1968 in the 200 areas (East and West) at the Hanford Nuclear Reservation in Richland, Washington, for a number of workdays aggregating at least 250 workdays occurring either solely under this employment or in combination with workdays within the parameters established for one or more other classes of employees in the SEC (73 FR 37459).
- All employees of DOE, its predecessor agencies, and their contractors and subcontractors who worked at the Hanford Site in Richland, Washington, from October 1, 1943 through June 30, 1972, for a number of workdays aggregating at least 250 workdays, occurring either solely under this employment or in combination with workdays within the parameters established for one or more other classes of employees included in the SEC (NIOSH 2009).

Dose reconstruction guidance in this document for periods prior to July 1, 1972 is presented to provide a technical basis for partial dose reconstructions for non presumptive cancers not covered within the SEC class through June 30, 1972. NIOSH found that it was infeasible to reconstruct doses for inadequately monitored radionuclides such as polonium, thorium, and neptunium. Although NIOSH found that it is not possible to completely reconstruct radiation doses for the proposed class, NIOSH intends to use any internal and external monitoring data that may become available for an individual claim (and that can be interpreted using existing NIOSH dose reconstruction processes or procedures). Therefore, dose reconstructions for individuals employed at the Hanford site during the period from October 1, 1943 through June 30, 1972, but who do not qualify for inclusion in the SEC, may be performed using these data as appropriate.

5.1.3 <u>Scope</u>

Hanford operations have played an important role in the development of the U.S. nuclear weapons program. This TBD is part of the overall Hanford Site Profile, which describes plant facilities and processes, historical information, and environmental data in relation to dose reconstruction for Hanford workers. It contains supporting documentation to assist in the reconstruction of occupational internal doses from these activities.

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The methods and concepts of measuring occupational internal doses to workers have evolved since the beginning of operations at the Hanford Site. An objective of this document is to provide supporting technical data to evaluate internal occupational doses that can reasonably be associated with worker radiation exposures. In addition, this document presents the technical basis of methods used to prepare Hanford worker dose information for input to the Interactive RadioEpidemiological Program (IREP).

When the first reactor became operational on the Hanford Site, there were no programs to monitor an employee for internal dose with the exception of measuring particles in the air. The site operated a fuel manufacturing facility, three reactors, and four processing plants from 1943 to 1946 before a bioassay program was in place. The responsibility for personnel monitoring was with the Medical Department. Attachment C, "Internal Dosimetry Coworker Data for the Hanford Site," provides guidance on assigning intakes for the years before bioassay.

In the 1940s, the radiation protection community used the term tolerance to describe dose limits and other values such as air concentration values. This was before the National Council on Radiation Protection and Measurements (NCRP) introduced the terms maximum permissible body burden (MPBB) and maximum permissible concentration (MPC). Air concentration tolerance levels were based on dose rates to significantly impacted organs. Examples at Hanford were 0.01 rep/d from alpha emitters, 1 rep/d to the thyroid from beta and gamma emitters, or total accumulation in the body of a radionuclide with comparable effects to 0.1 µg of radium. The latter limit was used to establish the limit of 0.5 µg for total accumulation of plutonium in the body. The radiation protection program was directed toward preventing workers from acquiring a tolerance dose or being exposed to tolerance air concentrations. Workers were supposed to wear respiratory protection if an air concentration exceeded the tolerance level or if a tolerance level was anticipated to be exceeded for a given job. In some cases, a lower air concentration was established as the respirator-required air concentration. The tolerance air concentrations in 1945 were $5 \times 10^{-10} \,\mu\text{g/cm}^3$ (4 × 10⁻¹¹ μ Ci/cm³) for product (meaning plutonium), $1 \times 10^{-8} \mu \text{Ci/cm}^3$ for fission and activation products, and 1.5×10^{-4} μ g/cm³ (1.1 × 10⁻¹⁰ μ Ci/cm³) for uranium (Cantril 1945). By 1947, the respirator-required air concentrations were $2 \times 10^{-11} \,\mu\text{g/cm}^3$ ($2 \times 10^{-12} \,\mu\text{Ci/cm}^3$) for plutonium, ($1 \times 10^{-9} \,\mu\text{Ci/cm}^3$) for fission products (assumed to mean particulate beta emitters), and $5 \times 10^{-5} \,\mu\text{g/cm}^3$ (3.5 × $10^{-11} \,\mu\text{Ci/cm}^3$) for uranium (Parker 1947; Patterson 1949). Additional discussion on tolerance levels is provided in Attachment A.

According to the history by Wilson (1987), one of the priority tasks for a special studies group that was formed in 1944 was to determine a way to measure plutonium in the body. Limits on the amount of plutonium in the body were set as early as 1944 and, after experimentation with various methods, routine urine sampling and analysis for plutonium were initiated in 1946. Urinalysis for uranium seems to have also started in 1946 and was well established by 1948. Urinalysis for fission products started in this timeframe as well, although the Wilson document indicates that separation from ⁴⁰K was not always successful before 1949. Since then, monitoring for numerous radionuclides has occurred at Hanford because of the complex scope of work over the years, the many research projects, special "campaigns," and other activities. In addition, numerous techniques have been used because of improvements in techniques. The major sources of intakes have been plutonium, ²⁴¹Am either as an ingrown contaminant in the plutonium or as a separated waste product, uranium, fission products, activation products, and tritium. However, the records list a wide spectrum of radionuclides that were monitored and an even longer list of codes that identify the radionuclides, groups of radionuclides, specific measurement techniques, or combinations of radionuclides and techniques. Many of the radionuclides apply to a small set of workers on a research project or to workers (for instance, radiation monitoring technicians) whose tasks might have exposed them to many different sources.

Attachment B provides a fairly exhaustive list of codes for analyses that dose reconstructors might encounter in the bioassay or internal dosimetry records for Hanford workers. Some of the codes were

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used for scheduling bioassay but not for reporting results of the bioassay. For instance, IPA is a code for performing plutonium and americium separation chemistry and alpha spectrometry on an excreta sample, but the results would normally be reported separately for ²³⁸Pu, ²³⁹Pu, and ²⁴¹Am. However, if the sample was not obtained or the results could not be reported due to analysis problems, the record shows just the IPA code with a reason for not obtaining a result [1]. Other codes refer to a type of *in vivo* count or a special type of sample analysis. For instance, LEPD is the code for performing an X-and gamma-ray analysis on an excreta sample using the low-energy photon detector (LEPD: a thinwindow germanium detector). However, if anything was detected, the actual radionuclide was reported [2]. The code GOK shows on *in vivo* count hard-copy records during the 1960s and 1970s. This refers to net counts per minute from an undetermined source in a low-energy region of the spectrum from Nal-based whole-body counters [3].

Other bioassay codes have been used to indicate the following:

- Sample type,
- In vivo count body location,
- Reason for the sample or count,
- Type of kit and some details about the sampling protocol,
- Laboratory used,
- Laboratory turnaround time versus analytical sensitivity,
- Units associated with the result, and
- Reason for not obtaining a valid excreta result or *in vivo* count.

In addition, there are codes that pertain to the nature of the intake:

- Reason for an intake assignment,
- Source of intake (at Hanford or another site),
- Nature of intake, and
- Mode of intake.

Tables that list and explain these codes are provided in Attachment B.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 5.7.

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

Most urinalysis records have, at some time, been entered into one or more databases. However, there are quite a few records missing from the databases, especially for some of the earliest urinalysis records including a significant number of the mixed fission product urinalysis records. In addition, there are examples of duplicate records in the databases. For any case where urinalysis might have been obtained before 1974, the hard-copy file for the case should be thoroughly reviewed for urinalysis results that might be missing from the database. The Hanford Internal Exposure (HIE) database was implemented in 1974 and was followed by the Occupational Radiological Exposure (ORE) database in 1983 and the Radiological Exposure (REX) database in 1993. In principle, the REX database has all the information from the previous databases but, as stated above, there are situations where some data were never entered in a database or did not get transferred from one database to another [4].

There is another anomaly in the results from around 1946 to 1950. There is a urinalysis record with no result and no volume. This might indicate that the sample was not turned in or the analysis failed,

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but experience has shown that this convention was also used to indicate a nondetection. In many cases the actual laboratory urinalysis results card is available in the worker's file and would show if the analysis was performed but the results were below detection or not [5].

Home sampling began very early in the program (1946) (Wilson 1987), and has continued throughout the history of Hanford. Home sampling was used to prevent contamination of samples in the workplace. The sampling protocol that was used most frequently (kit code 1) was a simulated 24-hour sample that was obtained by sampling from evening through morning on two consecutive nights (or equivalence for shift workers). Termination samples were often simulated 12-hour samples (kit code 2) [6]. The dose reconstructor should look at the kit code to determine if sample results should be treated as a day's excretion or adjusted to represent the daily excretion.

In vitro analyses were performed in house until the breakup of the main Hanford contractor (General Electric Company) in 1965. At that time, the U.S. Atomic Energy Commission (AEC) Richland Operations Office established a contract for *in vitro* analyses with the United States Testing Company (UST), which built a commercial low-level radiochemistry laboratory in north Richland and operated it until 1990. The responsibility for awarding and overseeing the contract was subsequently transferred to Battelle Memorial Institute (Battelle) as operators of the Pacific Northwest Laboratory (PNL), now called Pacific Northwest National Laboratory (PNNL). Until 2005, with the exception of a period between 1990 and 1992, *in vitro* analyses were performed in the same facility. However, due to buyouts and mergers, the name of the laboratory changed in the following sequence: UST, International Technology Analytical Services, Quanterra Environmental Services, and Severn Trent Laboratories. The current *in vitro* laboratory is General Engineering Laboratory [7].

Battelle defaulted on the contract with UST in June 1990, after which routine samples were collected and frozen (Lvon et al. 1991, 1992). Between September and November 1990, temporary contracts and agreements were established and samples were analyzed at the following laboratories: Los Alamos National Laboratory (LANL; plutonium), TMA-Norcal (strontium), PNL-Analytical Chemistry Laboratory (325 Building; tritium), and Westinghouse Hanford Company (222-S Building; elemental uranium). In February 1991, International Technology Analytical Services began analyses for plutonium, americium, curium, and isotopic uranium. LANL was replaced by Oak Ridge National Laboratory and Reynolds Electrical and Engineering Company at the Nevada Test Site (plutonium) in April 1991. The contract with International Technology Analytical Services replaced the contract with UST, but the other laboratories continued to process samples until the backlog was worked off. Therefore, the work at the temporary laboratories was finishing during late 1991 through early 1992; Hanford received the last results in March 1992. This interruption in the normal process of bioassay monitoring had two effects that show in the bioassay records for this period: (1) changes in the minimum detectable activities (MDAs) for the various analyses reflecting capabilities at the interim laboratories; and (2) delays in collection, analysis, and follow-up to high routine results from the normal pattern (Lyon et al. 1991, 1992).

5.2.1 <u>Plutonium</u>

As presented in Section 5.1.2, an SEC was designated in which NIOSH found that it was infeasible to reconstruct doses for plutonium before August 31, 1946 due to inadequate monitoring (72 FR 55214). Therefore, dose reconstructions will not include any doses from plutonium before August 31, 1946.

By far the most serious intakes at Hanford involved plutonium and ²⁴¹Am. Routine urinalyses for plutonium started in September 1946 (Wilson 1987). The first plutonium bioassay analysis consisted of lanthanum fluoride precipitation and thenoyl trifluoroacetone (TTA) extraction and gross alpha counting. Electrodeposition on a stainless-steel disk in combination with nuclear track emulsion (autoradiography) started in December 1952. Detection levels for these and subsequent procedures are listed in Table 5-1. The definition of *detection level* no doubt changed over the years, but the

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levels in Table 5-1 fit reasonably with the concept of limit of detection or MDA. For example, Wilson (1987) states,

From statistical evaluations of data collected in 1953, the true detection limit with nuclear-track film was determined. These evaluations showed 0.05 dpm was achievable within reasonable confidence levels. Occasionally recovery, counting, etc., allowed detection levels to be as low as 0.028 dpm and for a short period, a level of 0.027 dpm was reached and used as the detection level ... This practice [of recording lower detection levels] was discontinued and the more conservative 0.05 dpm was used routinely even though lower levels were possible part of the time.

Period	MDA	Decision level	Measured quantity
Before 06/1949	0.96 ^b	0.66	Total Pu alpha
06/1949-11/1952	0.33		Total Pu alpha
12/1952-01/27/1953	0.18		Total Pu alpha
01/28/1953-03/26/1953	0.15		Total Pu alpha
03/27/1953-11/06/1953	0.05		Total Pu alpha
11/07/1953-12/04/1953	0.07		Total Pu alpha
12/1953-04/1955	0.057		Total Pu alpha
05/1955-08/1955	0.027 ^c		Total Pu alpha
09/1955-09/1955	0.04 ^c		Total Pu alpha
10/1955-09/30/1983	0.05 ^d	0.025 ^d	Total Pu alpha
10/01/1983-12/31/1983	0.035	Not established,	Each Pu-238, Pu-239
		recommend use of	
		0.025 consistent	
		with previous years	
01/02/1984–04/1988	0.02		Total Pu alpha
05/1988-05/1990	0.02	0.01	Total Pu alpha
06/1990-11/1991	0.03	0.015	Total Pu alpha
11/1991-04/2000	0.02	0.01	Total Pu alpha
05/2000-08/2001	0.02	X _b + 2.05 × TPU ^e	Total Pu alpha
09/2001-present	0.02	2 × TPU ^e	Total Pu alpha

Table 5-1. Routine plutonium urinalysis detection levels (dpm/sample)^a.

a. From Wilson (1987) except as otherwise noted.

b. Estimated from data in Table 1 in *Bioassay at Hanford* (Healy 1948).

c. The values are probably closer to a decision level than an MDA. It is recommended that dose reconstructors use 0.05 dpm as the MDA [10].

d. During part of this period, results that were less than the detection limit were reported as 0.025. However, if net activity above background and above 0.025 was detected, the actual amount was recorded [11].

e. X_b is mean of blanks and TPU is total propagated uncertainty. The X_b values during that time were 0.0000 and 0.000487 dpm for Pu-238 and Pu-239 respectively. However, it is recommended that dose reconstructors use 2 × TPU as the actual decision level for both periods [12].

Before October 1983, the recorded value was the total alpha activity from plutonium, so it would have included activity from ²³⁸Pu, ²³⁹Pu, and ²⁴⁰Pu. The recorded results would not have accounted for any ²⁴¹Pu or ²⁴¹Am present in the urine [8].

The results might have been reported as plutonium or ²³⁹Pu but, until October 1983, the result was really the total alpha activity from isotopes of plutonium. Results on plutonium urinalysis sheets were recorded in units of disintegrations per minute per sample, but the same results were converted to units of microcuries per sample in the database. The units in the database should have a unit code of 5, meaning μ Ci/sample, but if the code is missing or unreadable, the units are still recognizable because the exponent is normally –7 or –8. A value of 1.1 × 10⁻⁸ was recorded for results for which plutonium was not detected (one-half of the nominal 0.05-dpm MDA). This method of recording was

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used through 1974. In 1975, the units were changed to dpm/sample (unit code 1), and 0.025 was recorded for results for which plutonium was not detected [9].

In October 1983 several changes were made. The lanthanum fluoride with TTA method was replaced by the use of anion exchange columns, autoradiography was replaced by alpha spectrometry analysis, and chemical yield was established for each sample separately by use of a ²⁴²Pu tracer. The results of ²³⁸Pu and ²³⁹⁺²⁴⁰Pu have been reported separately since then. A 2,500-minute count time has been used since 1984. A 10,000-minute count time was introduced for special situations in 1996, but its use was rare [13].

Starting in the mid-1990s, the fecal procedure was enhanced to ensure improved oxidation of highly insoluble plutonium. Added steps included wet ashing with hydrogen peroxide and fusion with hydrogen fluoride. This procedure was tested with special high-fired plutonium oxide samples from the Idaho National Engineering and Environmental Laboratory and was found to work well [14].

Fecal samples were usually not analyzed in total; they were aliquoted after muffling, dry ashing, and wet ashing. Therefore, more than one analysis result for a given sample was possible and are often found in the database [15].

The MDAs that are listed from 1983 to the present are nominal MDAs based on contractual requirements. In general, the laboratory performed slightly better than the contractual MDA, but the true MDA varied slightly over time and the contractual MDA was a reliable estimate [16]. Reporting of errors, which was the total propagated uncertainty (TPU) including uncertainty in the determination of chemical yield, counting efficiency determination, and systematic errors, began in 1981 [17]. The implementation of a distinction between an MDA (type I and type II errors) and a decision level (type I error) occurred in April 1989 (Lyon et al. 1990). A fixed value of 0.01 dpm/sample (half the nominal MDA) was initially used for the decision level for all results. The decision level was allowed to become sample-specific based on the TPU in 2000, and an adjustment was made to the formula in 2001 (Lynch et al. 2000, 2001).

The MDAs in Table 5-1 apply to routine and priority processing of urine samples. Fecal sampling was used for special sampling after potential intakes, and other processing codes (emergency and expedite) have been available for special urine and fecal samples. The contractual MDAs for these samples are provided in Table 5-2.

	Feca	sample MD	A	Urine sample MDA		
Period	Emergency ^a	Expedite	Priority	Emergency ^a	Expedite	
1/1965–10/1983 ^b	0.9-1.5	NA	0.1-0.15	0.5-0.7	NA	
10/1983–1/1985 [°]	9	NA	0.2	0.5	NA	
1/1985-6/1990	9	3	0.2	0.5	0.08	
6/1990–2/1991 ^d	20	4	NA	2	0.4	
2/1991-present	9	3	0.2	0.5	0.08	

Table 5-2. MDAs for nonroutine plutonium excreta analyses (dpm/sample) [18].

NA = not available.

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. MDAs varied according to sample size over the range shown; the lower value was generally applicable except for very large samples (BPNL 1982). MDAs for this period apply to total plutonium alpha.

c. MDAs from this time forward apply to Pu-238 and Pu-239 separately.

d. Emergency and expedited processing of urine and fecal samples was available through the PNNL Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite laboratories but the MDA was not established, probably about 0.2-0.5 dpm/sample considering the state-of-the-art of those laboratories.

Fecal sampling was normally done in response to suspected intakes, but routine fecal sampling was used for some high-risk plutonium workers, mostly operators at the Plutonium–Uranium Extraction

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(PUREX) facility and the Plutonium Finishing Plant, from 1986 through June 1989. The special study showed that, when considered as a group, the mean fecal excretion was statistically significantly different from controls. Enhanced air sampling, which was initiated in response to the study, showed frequent but intermittent releases of plutonium in the workplaces, at levels below the detectability of normal air sampling. When modeled as chronic intake, the intakes and doses were low (less than 10 mrem committed effective dose equivalent), and were documented in the workers' records (Bihl, Buschbom, and Sula 1993; Lyon et al. 1988, 1989). When encountered in the workers' records, results for these fecal samples should be interpreted as associated with chronic intakes, not with an acute intake occurring many days prior to the sample dates.

With the exception of a few standards in radiochemistry laboratories and a ²³⁸Pu purification experiment in the 325 Building C-Cell around 1967 [19], plutonium at Hanford was comprised of a mix of radionuclides, namely ²³⁸Pu, ²³⁹Pu, ²⁴⁰Pu, and ²⁴¹Pu. The activity of ²⁴²Pu in plutonium mixtures at Hanford was too small to contribute significantly to dose. Hanford plutonium mixtures were categorized by their weight percent of ²⁴⁰Pu. When the reactors were operated to produce plutonium for weapons, the target mixture was about 6% ²⁴⁰Pu, a mixture referred to as weapons grade. N Reactor was also operated to produce electrical power for a local public power company. When the reactor was operated to produce power, the mixture in the fuel rods when removed from the reactor was nominally 12% ²⁴⁰Pu, a mixture referred to as fuel grade. At any given time, individual fuel rods would have mixtures that differed from these, as would individual batches of rods starting at the front end of the fuel rod dissolution and plutonium extraction processes. However, when refined and blended, the target mixture was the weapons-grade mixture [20]. There is evidence at LANL that plutonium produced at Hanford has not been found. Tables 5-3 and 5-4 list the relative activities of plutonium isotopes and ²⁴¹Am, which grows in from ²⁴¹Pu, for 6% ²⁴⁰Pu and 12% ²⁴⁰Pu mixtures (PNNL 2003a). In these tables, *aging* refers to the time since the ²⁴¹Am was separated from the plutonium (and then starts to build in again from decay of ²⁴¹Pu).

	Fresh (2-						
Mixture designation:	weeks)	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr
Years of aging: ^a	0.038	5	10	15	20	25	30
		Specific ac	tivity in mix	ture (Ci/g)			
Pu-238	8.56E-03	8.23E-03	7.91E-03	7.60E-03	7.31E-03	7.03E-03	6.75E-03
Pu-239	5.77E-02	5.77E-02	5.77E-02	5.77E-02	5.77E-02	5.77E-02	5.77E-02
Pu-240	1.36E-02	1.36E-02	1.36E-02	1.36E-02	1.36E-02	1.36E-02	1.36E-02
Pu-241	8.24E-01	6.48E-01	5.09E-01	4.00E-01	3.15E-01	2.48E-01	1.95E-01
Pu-242	1.97E-06	1.97E-06	1.97E-06	1.97E-06	1.97E-06	1.97E-06	1.97E-06
Am-241	5.3E-05	5.83E-03	1.04E-02	1.39E-02	1.66E-02	1.87E-02	2.03E-02
Pu-239+240	7.13E-02	7.13E-02	7.13E-02	7.13E-02	7.12E-02	7.12E-02	7.12E-02
Pu-alpha	7.99E-02	7.95E-02	7.92E-02	7.89E-02	7.85E-02	7.83E-02	7.80E-02
Total alpha	7.99E-02	8.53E-02	8.96E-02	9.28E-02	9.52E-02	9.70E-02	9.83E-02
Activity ratios							
Pu-239+240:Am-241	1345	12.2	6.87	5.13	4.28	3.80	3.50
Pu-239+240:Pu-238	8.33	8.67	9.01	9.38	9.74	10.1	10.5
Pu-241:Pu-239+240	11.6	9.09	7.15	5.62	4.42	3.48	2.73
Pu alpha:Pu-239+240	1.12	1.12	1.11	1.11	1.10	1.10	1.10
Pu alpha: Pu-238	9.33	9.66	10.0	10.4	10.7	11.1	11.6
Pu alpha:Am-241	1507	13.6	7.62	5.68	4.73	4.19	3.84
Pu-241: Pu alpha	10.3	8.15	6.43	5.07	4.01	3.17	2.50

Table 5-3. Activity composition of Hanford reference weapons-grade plutonium mixture (6%).

a. Time since separation of Am-241 from the plutonium mix.

The values in these tables can help determine the total intake of plutonium and ²⁴¹Am if there are limited data on the composition of the source of the intake. For instance, only in rare large intakes

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	Fresh (2-						
Mixture designation:	weeks)	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr
Years of aging: ^a	0.038	5	10	15	20	25	30
		Specific ac	tivity in mix	ture (Ci/g)			
Pu-238	1.71E-02	1.64E-02	1.58E-02	1.52E-02	1.46E-02	1.40E-02	1.35E-02
Pu-239	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.26E-02	5.25E-02
Pu-240	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.72E-02	2.71E-02	2.71E-02
Pu-241	3.09E+00	2.43E+00	1.91E+00	1.50E+00	1.18E+00	9.29E-01	7.30E-01
Pu-242	3.93E-06	3.93E-06	3.93E-06	3.93E-06	3.93E-06	3.93E-06	3.93E-06
Am-241	2.0E-04	2.19E-02	3.89E-02	5.22E-02	6.24E-02	7.03E-02	7.63E-02
Pu-239+240	7.98E-02	7.98E-02	7.98E-02	7.97E-02	7.97E-02	7.97E-02	7.97E-02
Pu-alpha	9.69E-02	9.62E-02	9.56E-02	9.49E-02	9.43E-02	9.37E-02	9.32E-02
Total alpha	9.69E-02	1.18E-01	1.35E-01	1.47E-01	1.57E-01	1.64E-01	1.69E-01
	Activity ratios						
Pu-239+240:Am-241	399	3.64	2.05	1.53	1.28	1.13	1.04
Pu-239+240:Pu-238	4.67	4.86	5.05	5.24	5.46	5.69	5.90
Pu-241:Pu-239+240	38.7	30.5	24.0	18.8	14.8	11.7	9.16
Pu alpha:Pu-239+240	1.21	1.21	1.20	1.19	1.18	1.18	1.17
Pu alpha: Pu-238	5.67	5.87	6.05	6.24	6.46	6.69	6.90
Pu alpha:Am-241	485	4.39	2.46	1.82	1.51	1.33	1.22
Pu-241: Pu alpha	31.9	25.3	20.0	15.8	12.5	9.91	7.83

Table 5-4. Activity composition of Hanford reference fuel-grade plutonium mixture (12%).

a. Time since separation of the Am-241 from the plutonium mix.

was ²⁴¹Pu measured as part of the intake, so the activity of that isotope is almost never available. Americium-241 at the time of intake was also often not determined directly. Since 1983, ²³⁸Pu and ²³⁹⁺²⁴⁰Pu were measured separately, so the ratio of one to the other can be used to estimate the category of the plutonium mixture and, from the tables, to estimate the activities of ²⁴¹Pu and ²⁴¹Am. Before 1983, the measured quantity was total alpha from plutonium, which means the total of ²³⁸Pu and ²³⁹⁺²⁴⁰Pu. Therefore, unless ²⁴¹Am was measured or there is other information about the intake, there might be no way to tell from the bioassay how much ²⁴¹Pu and ²⁴¹Am were present at intake.

Most plutonium mixtures at Hanford were nominally weapons grade and, if the ²³⁹⁺²⁴⁰Pu-to-²³⁸Pu ratio from an intake implies weapons grade (e.g., >8), dose reconstructors should use the ratios in Table 5-3. However, if no information is available about the nature of the plutonium mixture that is associated with an intake, or with less than MDA results used to calculate missed dose, and the intake is being calculated from urine sample results, the 12% mixture may be used as a default, favorable to claimant assumption. Because there is less Am-241 in the 6% mix than the 12% mix, use of a 6% mixture is the default starting point for limiting doses based on chest counts. If both types of data are available, it's necessary to compare the two.

With regard to the age of the plutonium, until the fifth year of fuel separation (1949), assume fresh plutonium. For 1950 through 1954, assume a 5-year-old plutonium mixture. After these times, the assumption of 10-year-old plutonium should be made. For intakes since about 1996, 20-year old fuel-grade mixture could be assumed.

There was at least one project in the 1970s that involved irradiated fuel rods from commercial power reactors (Nuclear Waste Vitrification Project). The work took place in the 324 and 325 Buildings and the material was stored in the 303C Building in the 300 Area [21]. Commercial fuel rods have a much higher degree of burnup, and those at Hanford were characterized by much more ²⁴¹Pu and nominally 26% ²⁴⁰Pu. Table 5-5 provides the activity characteristics of the commercial fuel used in the Nuclear Waste Vitrification Project. In addition, the Plutonium Finishing Plant sometimes recycled plutonium from other DOE sites, United Kingdom, and commercial reactors." These materials often had higher ²⁴⁰Pu and ²⁴¹ Am content (Hoyt and Teal 2004, pp. 34-38). Plutonium from the West Valley

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commercial reprocessing site is also stored at Hanford. However, unless the records about the specific intakes being investigated have evidence of these unusual mixtures, the default mixtures above should be used.

Mixture designation:	Fresh (2- weeks)	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr		
Years of aging: ^a	0.038	5	10	15	20	25	30		
	Specific activity in mixture (Ci/g)								
Pu-238	1.71E-01	1.64E-01	1.58E-01	1.52E-01	1.46E-01	1.40E-01	1.35E-01		
Pu-239	3.41E-02	3.41E-02	3.41E-02	3.41E-02	3.41E-02	3.41E-02	3.41E-02		
Pu-240	5.90E-02	5.89E-02	5.89E-02	5.89E-02	5.89E-02	5.88E-02	5.88E-02		
Pu-241	1.34E+01	1.05E+01	8.28E+00	6.51E+00	5.12E+00	4.03E+00	3.17E+00		
Pu-242	1.97E-04	1.97E-04	1.97E-04	1.97E-04	1.97E-04	1.97E-04	1.97E-04		
Am-241	8.22E-04	9.49E-02	1.69E-01	2.26E-01	2.79E-01	3.04E-01	3.31E-01		
Pu-239+240	9.31E-02	9.31E-02	9.30E-02	9.30E-02	9.29E-02	9.29E-02	9.29E-02		
Pu-alpha	2.65E-01	2.58E-01	2.52E-01	2.45E-01	2.39E-01	2.34E-01	2.28E-01		
Total alpha	2.65E-01	3.53E-01	4.20E-01	4.71E-01	5.10E-01	5.38E-01	5.59E-01		
Activity ratios									
Pu-239+240:Am-241	113	0.981	0.551	0.411	0.344	0.305	0.281		
Pu-239+240:Pu-238	0.544	0.568	0.589	0.612	0.636	0.664	0.688		
Pu-241:Pu-239+240	144	113	89.1	70.0	55.1	43.3	34.1		
Pu alpha:Pu-239+240	2.85	2.77	2.71	2.63	2.57	2.52	2.45		
Pu alpha: Pu-238	1.55	1.57	1.59	1.61	1.64	1.67	1.69		
Pu alpha:Am-241	322	2.72	1.49	1.08	0.857	0.770	0.689		
Pu-241: Pu alpha	50.6	40.7	32.9	26.6	21.4	17.2	13.9		

Table 5-5. Activity composition of Hanford reference commercial power fuel-grade plutonium mixture (PNNL 2003a).

a. Time since separation of the Am-241 from the plutonium mixture.

If some of the plutonium bioassay was obtained before October 1983 and some after, the two datasets are not compatible. The results from before October 1983 are the sum of activities from ²³⁸Pu, ²³⁹Pu, and ²⁴⁰Pu; the post-October-1983 results are separate for ²³⁸Pu and ²³⁹⁺²⁴⁰Pu. Use known ratios if available or default activity ratios to determine the isotopic components of the total alpha results before combining the datasets for curve fitting.

Plutonium at Hanford could have existed as absorption type M, S, or a highly insoluble form, Super S (ORAUT 2008). Dose reconstructors should model the ²⁴¹Am that is a component of plutonium contamination in the lung the same as the plutonium matrix in which it has grown. In other words, the americium should be treated as absorption type S if the plutonium is type S.

A significant decrease in the number of plutonium bioassays across the Hanford Site occurred starting in 1960. This resulted from a change in policy on the frequency of sampling of workers. A determination of the risk of intake was made and workers were reassigned to sampling frequencies of quarterly, annually, or once every 5 years.

5.2.2 <u>Americium</u>

Americium was usually a trace contaminant in plutonium mixtures, as discussed in Section 5.2.1. However, americium was separated from plutonium at the reprocessing plants [e.g., T Plant and S Plant (also called Reduction-Oxidation (REDOX) Plant in the early years and the PUREX Plant from 1956)] and at the Plutonium Reclamation Facility (a wing in the Plutonium Finishing Plant). There was a major americium recovery operation in the Waste Treatment Facility, 242-Z, from 1964 until 1976. Smaller scale separation of americium from plutonium occasionally occurred earlier at 231-Z and later at the 325 Building in the 300 Area. See the latest revision of ORAUT-TKBS-0006-2, *Hanford Site* –

Site Description, for details (ORAUT 2010c). This americium should be treated as americium (rather than trace americium atoms bound in a plutonium matrix) [22]. The International Commission on Radiological Protection (ICRP) has recommended absorption type M for americium (ICRP 1994).

As presented in Section 5.1.2, an SEC class was designated in which NIOSH found that it was infeasible to reconstruct doses for americium separated from plutonium in the 200 area before December 31, 1968 due to inadequate monitoring (73 FR 37459). Although doses from separated americium cannot be reconstructed during this period, doses can be reconstructed for americium that is part of the plutonium matrix. Doses for separated americium can be reconstructed if the worker's individual record includes chest counts or americium urinalysis results.

Routine chest counting for ²⁴¹Am started in 1968, and special chest counts started in 1966 (see Section 5.3.2).

It has not been discovered yet when an americium urinalysis procedure was first developed at Hanford. There is no mention of americium excreta analysis in Healy (1948); no mention in a 1954 memorandum, "Bioassay Annual Report," that lists numbers of urinalyses for plutonium, fission products, and uranium (Watters 1954); and no mention in a compilation of bioassay procedures, *Bioassay Procedures and Analysis (Old Bioassay Bible)* (Author unknown 1961).

The first americium records in the database show 41 urinalyses for ²⁴¹Am in 1964 on samples that were collected from 19 workers. No americium urinalyses were recorded in 1965 or 1966. The 1964 samples were collected over a short period. The first samples, 12 of them, were collected on August 25 or 26. The last sample was collected on October 29. The results range from 1.3×10^{-6} to 6.4×10^{-6} [23]. No units are given, although the magnitude of the results suggest the units are μ Ci/sample or μ Ci/L. It is not clear whether the units are per sample or per liter. Hanford used per liter from 1967 through July 1969 and, because the volumes can vary from less than a liter to more than a liter, no particular choice is technically better than another. Therefore, the assumption that the units are μ Ci/L is reasonable [24].

One set of four samples for one worker was collected as follow-up to a contamination event. Among the remaining workers, most were sampled on August 25 or 26, and then had at least one additional sample a month or two later. Therefore, it is likely that the August 25 and 26 samples were baselines to establish the bioassay program at the start of the experiments that were being conducted to support a new ²⁴¹Am separation process at the Plutonium Finishing Plant [25].

No information about the radiochemical analysis method or detection level has been found. Assuming the August samples were baselines, a cumulative probability analysis was performed on the results that provided a median value of $1.9 \times 10^{-6} \,\mu$ Ci/L and a 95th percentile of $2.3 \times 10^{-6} \,\mu$ Ci/L. Based on this analysis, the MDA for ²⁴¹Am analyses in 1964 was assumed to be $4.6 \times 10^{-6} \,\mu$ Ci/L [26].

The records show no americium bioassay in 1965 or 1966; there are 168 results in 1967. The latter are a combination of urine and fecal results, and most of the results seem to be duplicated twice in the database. The duplication appears to be a mistake in the database that does not show up in hard-copy listings of these bioassay results. The results are all from special samples for six workers who were involved in the same potential intake accident in 222-S on May 2, 1967. There are internal dosimetry evaluations in the workers' files that document the accident and bioassay results. Many of the urine sample results were listed as <5.41 × 10⁻⁷ μ Ci/L, so it is assumed that this was the MDA for the analysis. This is the same number that was handwritten in the margin of an unpublished article, *Evaluation of Internal Depositions of Americium Using Bioassay Samples*, (Henle 1968). The handwritten note says, "5.41 × 10⁻⁷ μ Ci/L detection level."

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A memorandum to file from J. J. Jech (1969a), Senior Development Engineer in the Personnel Dosimetry Services, states that according to a telephone conversation with M. M. Lardy at UST, the new detection limit for ²⁴¹Am is 2.0 dpm/sample as of July 10, 1969. Lardy described the procedure as dibutyl N,N-diethylcarbamylphosphonate (DDCP) extraction to a planchet and gross-alpha counting (Bihl 2003). According to a letter from Lardy to H. Larson, manager of Personnel Dosimetry Services, a reduction in the detection limit was implemented in 1974 (Lardy 1974). The new limit for ²⁴¹Am in urine was given as 0.1 pCi/sample at the 90% confidence limit. This limit was still listed in a statement of work with UST in 1979 and again in 1982, although it was stated as 0.2 dpm/sample. In the laboratory statement of work for a new contract starting October 1983 (Simpson 1983), the detection level was listed as 0.04 dpm/sample. This improvement was achieved by use of an alpha/gamma coincidence counter.

Until October 1983, the gross alpha count could have included ²⁴²Cm or ²⁴⁴Cm if any was associated with the intake. Assuming the results are ²⁴¹Am is favorable to claimants. However, sometime between October 1983 and October 1985, both the chemistry procedure and the counting technique were changed. The chemistry method was similar to that in the Health and Safety Laboratory 300 manual (Volchok and de Planque 1982) and commonly referred to as the "RICH-RC-50-80" method (Bihl 2003). This method involved sequential precipitation with calcium oxalate and iron hydroxide, removal of plutonium using anion exchange, loading on another column with nitric acid and methanol, and elution of the americium with HCl and methanol. Electrodeposition and counting by alpha spectrometry were also implemented at this time (Bihl 2003). The MDA in the 1985 statement of work (Author unknown 1985) was listed as 0.02 dpm/sample (consistent with the change to alpha spectrometry), and it has stayed there to the present. At present, Eichrom transuranic column exchange is used for the separation of the americium for urine, but the MDA is the same (Bihl 2003). Table 5-6 summarizes what has been uncovered about ²⁴¹Am MDAs for routine urinalysis.

(upin/sample) [Z7].		
Period	MDA	Decision level
1964	4.6E-6 µCi/L	Anything detected
1967–6/1969	5.4E-7 µCi/L ^a	Anything detected
7/1969–2/1974	2.0	Anything detected
3/1974–10/1983	0.2	Anything detected
10/1983–9/1985	0.04	Anything detected
10/1985-05/1988	0.02	Anything detected
05/1988-06/1990	0.02	0.01
07/1990–10/1991	0.03	0.015
11/1991-4/2000	0.02	0.01
5/2000-8/2001	0.02	X _b + 2.05 × TPU ^b
9/2001-present	0.02	2 x TPU
 Not known if MDA or 	اميرما مرمام	•

Table 5-6. Routine ²⁴¹Am urinalysis detection levels (dpm/sample) [27].

a. Not known if MDA or decision level.

b. X_b is the mean of the blanks and TPU is total propagated uncertainty. The X_b value used during that time was 0.00276 dpm for Am-241. However, it is recommended that dose reconstructors use 2 x TPU as the actual decision level for both periods.

The MDAs in Table 5-6 apply to routine and priority processing of urine samples. Fecal sampling was used for special sampling after potential intakes, and other processing codes (emergency and expedite) have been available for special urine and fecal samples. The contractual MDAs for these samples are provided in Table 5-7. These analyses could have been used because of suspected intakes of pure ²⁴¹Am (such as the explosion of an americium exchange column at the Plutonium Finishing Plant in 1976) or to determine the activity of ²⁴¹Am in a plutonium mixture. There is evidence of a few intakes of pure ²⁴¹Am before 1969 that involve unusual circumstances such as using a supposedly sealed source that had ruptured. These intakes were analyzed by urinalysis, so a

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procedure obviously existed at that time, although it is not part of the contract with UST [28]. On rare occasions for a serious intake, samples were analyzed for ²⁴¹Am using a low-energy photon detector (LEPD) before any chemistry. This technique came into existence in 1986 or 1987. Its detection level was about 5 dpm/sample (Author unknown 1987). In general, the LEPD result was used just as a rapid indicator, and a more accurate result was obtained by wet chemistry and alpha spectrometry days later [29].

	Fecal sample MDA			Urine sample MDA	
Period	Emergency ^a	Expedite	Priority	Emergency ^a	Expedite
1/1967–2/1974	(b)	NA	(b)	(b)	NA
2/1974–1981	(C)	NA	4	(c)	NA
1982–9/1983	3.6–12	NA	1.2–5.0	0.7–1.0	NA
	(3.6 most probable) ^d		(1.2 most probable) ^d	(0.7 most probable) ^d	
10/1983–9/1985	200	NA	0.16	1.0	NA
10/1985–6/1989	20	6	0.1	1	0.08
7/1989–10/1991 ^e	20	4	NA	2	0.4
11/1991-present	20	6	0.1	1	0.08

Table 5-7. MDAs for nonroutine ²⁴¹Am excreta analyses (dpm/sample) [30].

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. Probably available but MDAs not found.

c. Emergency analyses were available on request, but the statement of work [based on 1978 statement of work (DOE 1979)] did not specify the MDAs. It implied that an MDA about 10 times the routine (or priority for fecal) MDA was expected.

d. Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples (BPNL 1982).

e. Emergency and expedited processing of urine and fecal samples was available through the PNNL Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite laboratories, but the MDA was not established; it was probably about 0.2 to 0.5 dpm/sample considering the state of the art of those laboratories.

5.2.3 <u>Curium</u>

The curium isotopes of concern were ²⁴²Cm and ²⁴⁴Cm. Most sources of curium at Hanford were minor calibration sources or minor constituents in an actinide mixture; however, flowsheet development for major extraction of ²⁴⁴Cm occurred in the 325 Building in 1966 and 1967 and led to major extraction and purification of curium from the Shippingport waste in 201-C and 325 Buildings in 1967 and 1968. Additional work with curium occurred in the 308 Building in smaller quantities in 1969 through 1973. See the latest revision of the Hanford Site Description for details (ORAUT 2010c).

The curium and americium procedure was the same, so the results would have been reported as curium only if so requested through the bioassay request system until alpha spectrometry was initiated in October 1983. After 1985, the chemistry was the same as americium, but ²⁴¹Am, ²⁴²Cm, and ²⁴⁴Cm were reported separately on request (Author unknown 1987). However, the MDAs were not always identical with ²⁴¹Am. Routine urinalysis MDAs for curium are provided in Table 5-8, and nonroutine excreta analyses are provided in Table 5-9.

5.2.4 <u>Tritium</u>

Tritium was referred to as P-10 in the 1950s (Parker 1950a). The main source of tritium in the 1950s was 108-B, also called the P-10 Plant, in which tritium was extracted from irradiated lithium-aluminum targets intermittently between August 1949 and 1955. A 1967 report states, "Battelle-Northwest and its predecessor at Hanford, the General Electric Company, have been involved in activities with tritium since about 1950, initially as a manufactured product for weapons applications and later as a by-product of heavy water reactor operations. Our most recent experience is from operation of the Plutonium Recycle Test Reactor (PRTR)" (McConnon 1967). The PRTR started up in 1960 and was

(upin/sample) [31]	•		
Period	MDA	Decision level	
7/1969–1981	Not spec	cifically mentioned	
1982–9/1983	Listed for eme	ergency processing only	
10/1983-4/1988	0.02 Anything detected		
5/1988-6/1990	0.02	0.01	
6/1990-10/1991	0.03 0.015		
11/1991-4/2000	0.02	0.01	
5/2000-8/2001	0.02	X _b + 2.05 × TPU ^a	
9/2001-present	0.02 2 × TPU		

Table 5-8. Routine curium urinalysis detection levels (dpm/sample) [31].

a. X_b is the mean of the blanks and TPU is total propagated uncertainty. The X_b values used during that time were 0.00206 and 0.0000 dpm for Cm-242 and Cm-244, respectively. However, it is recommended that dose reconstructors use 2 x TPU as the actual decision level for both periods [32].

Table 5-9.	MDAs for n	onroutine	curium	excreta an	alvses	(dpm/sample)	[33].

	Fecal samples, MDA		Urine samples, MDA		
Period	Emergency ^a	Expedite	Priority	y Emergency ^a Ex	
Before 1982	(b)	NA	(b)	(b)	NA
1982–9/1983	10 ^c	NA	NA	0.5–1.0 (0.5 most probable) ^{c,d}	NA
10/1983–9/1985	240	NA	0.8	10	NA
10/1985-6/1989	240	70	0.8	1	1.2
7/1989–10/1991 ^e	NA	NA	NA	NA	NA
11/1991-present	240	70	0.8	1	1.2

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. Probably available but MDAs not found.

c. Total alpha; would have included any americium present also.

d. Varied according to sample size over the range shown; the lower value was generally applicable except for very large samples (BPNL 1982).

e. Emergency and expedited processing of urine and fecal samples was available through the PNNL Analytical Chemistry Laboratory. Priority fecal analyses were also available through offsite laboratories, but the MDA was not established; it was probably about 0.2-0.5 dpm/sample considering the state of the art of those laboratories.

shut down in 1969. All of the major reactors had some potential for tritium exposure, and tritium was used in tracer quantities in biological research through most of Hanford's history. There was also some work on a tritium target program in the 1990s in the 300 Area and tritium light sources in the 1980s (involving just a few people). Tritium in the groundwater was a source of ingestion for 400 Area workers, principally the Fast Flux Test Facility, from about 1978 to 2003 (Bihl 2005). PNNL has participated in the DOE Tritium Readiness Program in recent years, producing and testing "tritium-producing burnable absorber rods" and examination of the rods in the 324 and 325 Buildings (Lanning 2007).

The history of tritium urinalysis at Hanford is not well documented. Tritium urinalysis was not mentioned at all in Wilson (1987). The earliest report on tritium urinalysis at Hanford dates to 1949 by J. Healy, the leading internal dosimetrist at Hanford for many years (Healy 1949). That procedure was based on "production of acetylene from the active water, with subsequent measurement of the ionization caused by the tritium beta particle" (Healy 1949). No detection level was mentioned in that letter, but one was mentioned in a memorandum (Parker 1950a) that referred to the acetylene method for urinalysis; it provides a sensitivity of about 1.2 μ Ci/L in water. However, that method apparently did not work well because a 1951 letter stated that, "Your problem on the determination of tritium in the urine samples is one that we have been working on for the last two years, and have finally obtained what appears to be a decent method for routine use" (Healy 1951). The copy of the letter is of such poor quality that the method is hard to follow, but it definitely was not liquid scintillation counting. Tritium urinalysis for the workers at the Tritium Extraction Facility (108B) was conducted at 108B until 1955 when the counter was moved to the bioassay lab in the 329 Building (Watters 1955,

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p. 4). A 1961 report, *The Estimation of Whole Body Dose from Tritium by Urine Analysis*, indicated that liquid scintillation was used by that time, but again no detection level was given (Beasley and Rouse 1961, p.4) Liquid scintillation counting was implemented for tritium bioassay at the Savannah River Site (SRS) in 1958, and it is reasonable to expect that Hanford did so at about the same time. In the interview with Lardy (Bihl 2003), Lardy stated that liquid scintillation counting of a 1-mL aliquot of raw urine has been used since UST was awarded the bioassay contract in 1965.

Tritium intakes were accounted for as part of external dose until about 1987, when they were entered in the dose database as an internal dose [34]. Section 5.6 contains a more detailed discussion and guidelines for assigning intakes. Tritium exposure was assumed to be chronic during the exposure period, unless a very large acute intake was known to occur.

Very little data on MDAs has been discovered. A sensitivity of 1.2 μ Ci/L was reported by Parker in 1950, but what constituted sensitivity was not mentioned (Parker 1950a, p. 3). McConnon (1964) states that a tritium bioassay result exceeding 5 μ Ci/L will be reported to the Radiation Monitoring Office the day after the samples are picked up, which indicates a level of concern probably well above the MDA. One P-10 Personnel Sample Analysis card, with entries in 1952, shows several values below 5 μ Ci/L and the smallest as 2.5 μ Ci/L. None of the values is listed as a less-than value. The 1965 statement of work with UST (AEC 1964) shows an MDA of 1 μ Ci/L (which is consistent with the MDA at SRS throughout the 1950s). Table 5-10 lists the most accurate compilation of MDAs for routine tritium urinalysis to date. From 1978 to the present, the MDAs were obtained from statements of work with the bioassay laboratory.

Table 5-10.	Routine tritium urinalysis detection	
levels [35].		

Period	MDA
1949–1960 ^a	≈ 5 µCi/L
1961–1981 ^b	1 µCi/L
1982–10/1991	10 dpm/mL
11/1991-present	20 dpm/mL

a. Dates and MDA are best guesses. The change in 1961 was based on earliest reference to liquid scintillation counting.

 The reporting level continued to be 5 μCi/L at least through August 1964 (GE 1964, p. G-8).

Tritium was present in an organic form in the 108-B facility in the pump oil, which became contaminated over time. According to a retired radiation monitor at the facility, the contamination in pump oil was orders of magnitude below general HT and HTO levels in the facility [36]. The tritium in the lithium-aluminum rods was absorbed in the aluminum as a getter; hence, it would have been present as a metal tritide, albeit a weak one. An article written at the time of operation of 108-B stated that "aluminum is one of the metals least likely to absorb tritium" (Kornberg 1950). Therefore, the tritium was easy to drive off as part of the process. Workers did not handle the aluminum directly and, after the tritium was driven off, the aluminum was placed in cans, sealed, and buried. This work was done in gloveboxes. There was also contaminated mercury in the vacuum lines to which maintenance workers had access about twice a year; special precautions were taken whenever a worker(s) had to be exposed to mercury because of the high contamination [37].

Metal tritides were potentially present as part of the Tritium Target Program work starting in 1988, the metal mostly likely being zirconium. The irradiated targets were examined and the tritium driven off in hot cells. Samples were taken for analysis, but where the analyses were performed has not been discovered yet.

5.2.5 <u>Uranium</u>

Uranium exposure at Hanford involved principally three physical forms: depleted uranium (DU), natural uranium (NU), and slightly enriched (also called recycled) uranium (RU). Uranium-233 was also isolated from irradiated thorium at PUREX from 1966 to approximately 1971. Small numbers of researchers might have experimented with more enriched uranium at different times (e.g., metallurgy on commercial-grade fuel), but such exposure would have been to small groups for limited periods. For instance, there was a radiation work permit for work with "bare, enriched - 37.5% - solid form uranium" for the 306 Building for November 9, 1970, through November 9, 1971 (BN 1970; Jech 1970a). Table 5-11 provides the default uranium mixtures that have been used at the Hanford Site since the 1980s (PNNL 2003a). However, different batches of uranium can have different isotopic mixtures; therefore, the default mixtures in the Integrated Modules for Bioassay Analysis (IMBA) program and the Dose Reconstruction Project tools are acceptable. In general, personnel working in the production facilities (e.g., fuel fabrication, the reactors, fuel dissolution and plutonium processing, and waste management) were exposed to NU during operation of the early reactors (through 1951) and to RU starting in February 1952 at the UO₃ Plant and July 1952 at the 300 Area Fabrication plants (DOE 2000). RU has impurities that build up and track with the uranium over time. Recommended impurity levels based on tolerance specifications or higher than normal concentrations in some drums are provided in Table 5-12 (DOE 2000). Plutonium-239 can be assumed for the plutonium alpha impurity, and ²³²Th can be assumed for the thorium [38].

	Uranium mixture	a,p						
Weight percentage	Weight percentage Recycled (RU) Commercial fuel							
U-234	0.0082	0.0300						
U-235	0.9700	2.9600						
U-236	0.0680	Negligible						
U-238	98.9500	97.0100						
Specific constituent ac	tivity in mixture (µ	ιCi/g, nCi/mg, or pCi/μg) [°]						
U-234	0.5125	1.8750						
U-235	0.0210	0.0639						
U-236	0.0440	Negligible						
U-238	0.3325	0.3260						
Total	0.9099	2.2649						
Specific constituent ac	tivity in mixture (d	lpm/µg) ^c						
U-234	1.1378	4.1625						
U-235	0.0465	0.1419						
U-236	0.0977	Negligible						
U-238	0.7381	0.7236						
Total	2.0200	5.0281						
Constituent fraction of	total uranium acti	vity in mixture						
U-234	0.5632	0.8279						
U-235	0.0230	0.0282						
U-236	0.0484	Negligible						
U-238	0.3654	0.1439						
Total	1.0000	1.0000						

Table 5-11. Radiological characteristics of Hanford uranium mixtures.

a. Commercial fuel data from Rich et al. (1988).

b. RU data based on average of data presented by Sula, Carbaugh, and Bihl (1991).

c. Can be used to represent specific alpha activity in the mixture as well.

Constituent	Maximum allowed ^a	Observed range ^b	Recommended level ^c
Plutonium	10 ppb U	<1–2 ppb U	0.8 nCi Pu-alpha/g U
Neptunium	Not established	0.04–0.16 ppm U	0.4 nCi Np-237/g U
Thorium	750 ppm U	8–10 ppm U	5 pCi Th-232/g U
Tc-99	Not established	3–4 ppm U	0.2 µCi Tc-99/g U
Ru-103, -106	<20 µCi/lb U	<6 µCi/lb U	40 nCi Ru-106/g U
ZrNb-95	<10 µCi/lb U	<4 µCi/lb U	20 nCi ZrNb-95/g U ^d
Other gamma emitters	<2 µCi/lb U	0.09–0.75 µCi/lb U	Negligible

Table 5-12. Impurities in RU at Hanford.

a. From UO₃ Plant operating specifications (Sula, Carbaugh, and Bihl 1991).

b. From analysis of uranium lots 88-1, 88-2, and 88-3 that were processed in 1988, and lots 93-01, 93-02, 93-03, 93-04, and 93-05, processed in 1993.

c. The recommended levels are expected to result in a slight overestimate of dose compared to levels actually observed as presented in the DOE investigation of RU at Hanford (DOE 2000). The plutonium reference level was based on the 10-ppb specification, which was reached or exceeded in a few drums throughout Hanford history (DOE 2000).

d. Interpret as 10 nCi each of the two radionuclides [39].

Uranium compounds at Hanford ranged from very soluble uranyl nitrate and soluble UO_3 to relatively insoluble UO_2 and U_3O_8 . Dissolution tests in simulated lung fluid were conducted on samples from the major uranium handling facilities. Results are shown in Table 5-13. Because the relationship between the old lung fluid studies and the ICRP Publication 66 absorption types (ICRP 1994) is not established, Table 5-13 also shows approximate ICRP 66 absorption types for intakes from the listed facilities. The information from the lung fluid studies indicates that all material types are present onsite. Unless there is claim-specific information about the solubility type, due to the uncertainty of the exact location of the worker at all times, dose reconstructors should use the solubility type that is favorable to the claimant. These absorption type assumptions should be applied to the impurities as well.

ICRP 30 ^a inhalation class from lung fluid studies	Compound and location	Approximate ICRP 66 ^b lung absorption type
80% D	Hanford UO ₃ Plant smear sample dissolution study in 1984 ^{c} (UO ₃	(d)
20% W	powder)	
10% D	Hanford 303-M Building air sample dissolution study ^e (300 Area	(d)
90% Y	Uranium Fuel Production Facilities)	
29% D	Hanford 333 Building air sample dissolution study ^e (300 Area	(d)
71% Y	Uranium Fuel Production Facilities)	
20% D	Hanford 306-W Building Machine Shop air sample dissolution study ^e	(d)
80% Y		
	Uranyl nitrate at PUREX or UO₃ Plant	F
	UCl ₄	M ^t
	U carbonate (assumed form after discharge to the soil)	F ^{t,g}

Table 5-13. Inhalation class for Hanford uranium compounds.

a. ICRP (1982).

b. ICRP (1994)

c. Sula, Carbaugh, and Bihl (1991).

d. Because the conversions from the solubility studies to the ICRP (1994) absorption types are not exact, the dose reconstructor can use the same percentages for D to F, W to M, etc., or can just use the predominant form to maximize dose to the organ of concern; for instance, the 303-M Building uranium might be considered 10% F, 90% S, or all type S.

e. Fisher (1986).

f. Long (1993)

g. Cooke and Holt (1974) [40].

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A note about sampling of UO_3 Plant workers: Because chemical toxicity was the principal concern for uranium exposures at the UO_3 Plant, one sampling scheme was to obtain both a Friday evening sample and Monday morning sample. This sampling scheme was in use as early as the 1950s and possibly earlier. This scheme was changed to Monday-morning-only sampling about the early 1980s. The changeover should be clear in the records.

The Friday-and-Monday sampling scheme was also used in 1962 and 1963 for 313 and 314 Building workers. Dose reconstructors need to be aware of Monday morning only samples and make appropriate adjustments so the intake will not be underestimated. For Friday-and-Monday sampling schemes, the Monday samples can be ignored and the Friday samples can be used.

Wilson (1987) states that the uranium urinalysis program before 1948 was not reliable. The fluorometric method, which fused uranium from raw urine with sodium fluoride and measured the fluorescence when the compound was exposed to ultraviolet light, was implemented sometime during the first half of 1948 (Healy 1948; Wilson 1987). This method was used for elemental uranium analyses, with refinements over the years including some upfront chemistry on the raw urine, until about 1991 when it was replaced by kinetic phosphorescence analysis (KPA) (Bihl 2003). (Note: Lardy said "about 1990" but other evidence indicates late 1991.) Lardy (1970) describes two procedures: (1) wet ashing with nitric acid and hydrogen peroxide, then acidification and counting of a 100- μ L aliquot with a detection level of 0.5 μ g/L, and (2) extraction (after wet ashing) with methyl isobutyl ketone and ammonium hydroxide. The detection limit for the latter was listed as 0.05 µg/L but the recoveries were about the same for both methods, so the latter must have used a 10-times larger aliquot. Based on requirements in later statements of work, it is assumed that the first method was used for routine analyses. Lardy (1970) also describes a third method - a radiometric procedure that uses the same separation chemistry as the second procedure – but the sample "is measured by a gas flow proportional counter or a ZnS (Ag) scintillation counter." The detection limit was given as 0.5 dpm/sample. A 1989 description of the chemistry was wet ashing with HCI and extraction with hexone (Carbaugh et al. 1989). A 100-mL aliquot was used, but the results were reported as per total sample. The chemistry for the KPA involved a 50-mL aliguot that was wet-ashed with acid, passed through an ion exchange column, then eluted with weak acid. Results were reported as per total sample.

When alpha spectrometry was introduced in 1983, two uranium urinalyses procedures were offered: the elemental procedure above and the alpha spectrometric procedure to provide isotopic results. In general, the elemental procedure was used for workers who were exposed to natural or slightly enriched forms of uranium, and the isotopic procedure was used for depleted or more than slightly enriched forms of uranium. In general, personnel who worked in the production facilities were monitored by the elemental analysis, whereas PNL workers were monitored by the isotopic analysis because of the wide scope of research projects that occurred over the years [41].

Alpha spectrometry cannot differentiate between ²³³U and ²³⁴U. Before 1994, the results for this region of the alpha spectrum were reported as ²³³U; they have been reported as ²³⁴U from 1994 to the present unless it was specifically determined that the worker was exposed to ²³³U. Work with ²³³U was rare after the early 1970s, long before alpha spectrometry came into use for bioassay. Therefore, unless ²³³U is specifically mentioned in an intake investigation report, assume that ²³³U results since 1983 are actually ²³⁴U [42].

Table 5-14 summarizes the routine urinalysis detection levels and Table 5-15 summarizes nonroutine detection levels. Although a detection level is presented for 1948, from December 1943 through 1948, intakes for uranium should be assigned in accordance with Section 5.6.2, which assigns intakes based on the Process/Job Title listed as Machining/Operator as presented in Battelle-TBD-6000/PNWD-3738, *Technical Basis Document: Site Profiles for Atomic Weapons Employers That Worked Uranium and Thorium Metals* (Battelle 2006).

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	Elemental (µg/L)		Isotopic	(dpm/sample)
Period	MDA	Decision level	MDA	Decision level
Before 1948	Not specific	ally mentioned	NA	NA
1948–1949	10	Anything detected	NA	NA
1950–1962 ^a	4 (reporting level)	(b)	NA	NA
1963-8/1964	0.14 (reporting level) ^{c,c}			
9/1964–1973	4 ^d			
1974–1981	0.4	Same as MDA	NA	NA
1982–9/1983	0.05–0.25	Same as MDA	NA	NA
	(0.1 most probable) ^e			
10/1983–12/1983	0.03 µg/sample	0.5 μg/sample ^t	0.035	Same as MDA
1/1984–8/1985	0.03 µg/sample	Not known	0.02	Same as MDA
9/1985–6/1990	0.03/0.5 µg/sample ^g	0.2 μg/sample ^h	0.02	Same as MDA
6/1990–10/1991	0.2/0.5 µg/sample ^g	0.2 ^h	0.03	0.15/0.015
11/1991-present	0.06/0.5 µg/sample ^g	0.2 ^t	0.02	0.15/0.010 ^g

Table 5-14. Routine uranium urinalysis detection levels.

NA = not available.

a. Wilson (1959). Lardy (1970) also indicates that the MDA from at least 1970 on was 0.5 μg/L, but the database shows that the use of 4 μg/L continued as a reporting level.

- b. In 1958 to 1962 values were reported well below the 4-µg/L value, but a statement of the actual reporting level was not found.
- c. GE (1963, p. G-8); GE (1964, p. G-9).
- d. The detection level of 4 μ g/L was established in the first contract with UST (AEC 1964) so it was applied starting in 1965 and that is probably when the level changed from 0.14 μ g/L. However, it is favorable to claimants to extend the 4- μ g/L value to September 1964.
- e. MDAs were based on sample size, but 0.1 μg/L was applied to most sample sizes (BPNL 1982). The database shows a change to reporting in μg/sample on July 2, 1982.
- f. Values below this were recorded but not followed up as occupational intakes.
- g. The larger value is the MDA for a special (rapid) analysis for UO₃ Plant workers based on potential chemical toxicity. The need for this special analysis ceased in 1994 after the last processing in the UO₃ Plant [43].
- h. Based on upper level for natural background excretion. See text for discussion.
- i. First value applied to U-234 and U-238; second value applied to U-235 based on natural background in urine. In 2002, the U-235 decision level was lowered to 0.007 dpm PNNL 2003b).

	Analysis	Fecal sample MDA			Urine sa	mple MDA
Period	type	Emergency ^a	Expedite	Priority	Emergency ^a	Expedite
1980 ^b	Elemental	NA	NA	NA	10 µg/L	NA
10/1983-	Elemental	8 µg/sample	NA	0.3 µg/sample	7 µg/sample	NA
9/1985	Isotopic ^c	12 dpm/sample	NA	0.3 dpm/sample	1 dpm/sample	NA
10/1985-	Elemental	8 µg/sample	5 µg/sample	0.3 µg/sample	7 µg/sample	0.5 µg/sample
present	Isotopic	12 dpm/sample	4 dpm/sample	0.3 dpm/sample	1 dpm/sample	0.12 dpm/sample
6/1990-	Elemental	20 µg/sample	4 µg/sample	NA	20 µg/sample	4 µg/sample
10/1991 ^d	Isotopic	20 dpm/sample	4 dpm/sample	NA	2 dpm/sample	0.4 dpm/sample
11/1991-	Elemental	8 µg/sample	5 µg/sample	0.3 µg/sample	7 µg/sample	0.5 µg/sample
present	Isotopic	12 dpm/sample	4 dpm/sample	0.3 dpm/sample	1 dpm/sample	0.12 dpm/sample

Table 5-15. MDAs for nonroutine uranium excreta analyses [44].

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. Earliest mention found in a contract with the laboratory. The ability to perform analyses on fecal samples was also mentioned, but an MDA was not specified.

c. U-234, U-235, U-238.

d. Emergency and expedited processing of urine and fecal samples was available through PNNL's Analytical Chemistry Laboratory. Priority fecal analyses were also available through the offsite laboratories, but the MDA was not established. It was probably about 0.2 to 0.5 dpm/sample considering the state of the art of those laboratories.

Starting about 1995, mass spectrometry has been used as an investigational tool to discriminate between natural background uranium and RU through measurement of ²³⁶U. The presence of ²³⁶U confirms an occupational intake of RU; the detection limit for ²³⁶U is such that urinary excretion of

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uranium greater than 0.2 µg/L (see discussion of natural background excretion below) from an intake of RU should have a detectable amount of ²³⁶U (PNNL 2003a; MacLellan, Wyse, and Scott 1995).

NU from nonoccupational intakes (primarily food and water) is excreted in urine at levels above the analytical MDAs for either the elemental uranium analysis or the alpha spectrometry analysis. The 234 U-to- 238 U ratio can be used to distinguish DU from NU but, considering uncertainties in analytical results, that ratio cannot be used to distinguish RU. Three studies were conducted in 1985, 1990, and 1995 to establish the range of natural background excretion in unexposed persons who lived near the Hanford Site. The third study purposely looked for possible geographic and seasonal differences in the background. All studies found natural excretion to have a lognormal distribution. Although the 50th percentiles and slopes of the excretion curves were different in the studies, each study found 0.2 μ g/d to be between about the 99th and 99.9th percentiles. The 1995 study had one result that greatly exceeded the 0.2 μ g/d value (PNNL 2003a).

Therefore, 0.2 μ g/d was established in 1985 and continues to be used at present by the Site as the environmental decision level for exposures to NU or RU. Only urinary excretion values greater than 0.2 μ g/d, which converts to 0.15 dpm/d for ²³⁴U and ²³⁸U and 0.007 dpm/d for ²³⁵U, are considered indicative of a potential occupational source. Nevertheless, the one result in the 1995 study and many worker-specific investigations of urinary results exceeding 0.2 μ g/d have shown that results well above the environmental screen level do occur from natural sources. Some of these were shown to be due to a specific home water well; others occurred from workers on city water from wells (but apparently not all wells).

Although natural background levels of uranium are present, dose reconstructors should consider any detected uranium as due to occupational exposure and should not make corrections for natural background levels.

Background excretion of uranium in feces probably varies over an even larger range than urinary excretion, but a definitive study for the Hanford area has not been conducted. Fecal samples were rarely obtained for potential uranium intakes; when they were, the investigation report should discuss how the results were interpreted.

Separation of ²³³U from thorium occurred in three distinct campaigns: a small process test in 1965 and two production runs in 1966 and 1970. A total of 820 kg of ²³³U were produced (Walser 1978; Isochem 1967). Section 5.3.4 of the Hanford Site Description (ORAUT 2010c) contains a discussion of the program to create ²³³U by irradiation of thorium.

The ²³³U was recovered at PUREX in the nitrate form, so it would have been absorption type F. Some of the 233 U from the 1965 run was converted to U₃O₈ at Z Plant (Fullam 1965, pp. 3–4; Hopkins 1965, p. 68), which is considered absorption type S. For Z- Plant workers, because the material was being converted to oxide, dose reconstructors should apply absorption type F, M, or S, whichever is most favorable to the claimant. The Hanford bioassay database shows that a few urinalyses were obtained for ²³³U, mostly in 1970. No information has been found about how the analyses were performed. A November 1970 letter from Jech to Lardy at UST indicates that aliquots from plutonium samples were used with a detection limit of "0.5 d/m per 100 ml" (Jech 1970b). Copies of an earlier database show the units to be micrograms per liter, which is consistent with the units for all uranium analyses at that time. However, with the exception of a single value in 1969, the recorded values are much too small to be realistically in micrograms per liter. The recorded values appear to be similar to plutonium alpha measurements at that time, and it is probable but not verified yet that autoradiography was used for these bioassays because of the high specific activity of the ²³³U and the major contaminant ²³²U [45]. Based on the Jech letter, the units are most likely microcuries per sample. A second November 10 letter from Jech indicates that the workers who were sampled were the PUREX workers involved in the ²³³U campaign (Jech 1970c).

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There were impurities in the ²³³U. The measured impurities varied between the three campaigns as did the impurity concentrations. A summary of the highest impurities concentrations is given in Table 5-16.

Table 5-16. Highest level of impurities as measured for the three campaigns.

Impurity	1965 campaign ^a	1966 campaign ^b	1970 campaign ^c
U-232 ppm	2.5	6.9	8.6
Pu μg/μgU	0.002	1.8E-06 ^d	NG
Th μg/μgU	0.094	2.2E-04	NG
Np μg/μgU	NG	2.8E-05	NG
Zr/Nb-95 pCi/µgU	NG	0.35	NG
Ru-103/Ru-106 pCi/µgU	NG	0.046	NG
Pa-233 pCi/µgU	102	4.4	NG
Th-238 pCi/µgU	NG	NG	0.84

NG = not given

a. As measured at Z Plant (Fullam 1965, p. 22)

b. Isochem (1967, p. VII-7).

c. Jackson and Walser (1977, p. 175).

d. A very small batch had a higher concentration of plutonium.

The impurity values were converted to units of picocuries per microgram of uranium using the assumptions that plutonium was ²³⁹Pu, thorium was ²³²Th, and neptunium was ²³⁷Np with specific activities of $6.2 \times 10^4 \text{ pCi/\mug}$ ²³⁹Pu, 0.109 pCi/µg ²³²Th, 710 pCi/µg ²³⁷Np and 2.14 × 10⁷ pCi/µg ²³²U. Using the highest of the measured values of all the campaigns (except for the plutonium value of one small batch in the 1966 campaign), the results are shown in Table 5-17. These impurity concentrations are favorable to the claimant and are recommended for the dose reconstruction if the intake of ²³³U is determined. When determining intakes and dose for ²³³U intakes, determine the intake of ²³³U first then add intakes of the impurities (including ²³²U); use ²³³U and ²³²U not ²³⁴U for dose calculations.

Table 5-17. Thynest impunty concentrations in 0 (pc//pg		
Impurity	Concentration	
U-232	190	
Pu-239	124	
Th-232	1.0E-02	
Np-237	2.0E-02	
Zr/Nb-95	3.5E-01	
Ru-106	4.6E-02	
Pa-233	102	
Th-238	8.4E-01	

Table 5-17. Highest impurity concentrations in ²³³U (pCi/µgU).

Work with ²³³U is covered by the SEC. If dose reconstruction is needed, see Section 5.6 for guidance on intakes of ²³³U for unmonitored workers.

There was exposure to highly enriched uranium (HEU) at Hanford, starting in July 1948, occurring heavily in 1949 through 1956 and sporadically after that. Almost all the known work was done in the 300 Area although there is indication that some work might have occurred in 231-Z. See the Hanford Site Description (ORAUT 2010c) for details. The workers exposed to HEU were most likely also exposed to natural or slightly enriched uranium. There does not seem to be a way to determine from measurements in workers' files whether an intake of uranium included HEU. If exposure to HEU seems likely, the mass urinalysis results should be interpreted as HEU using the 93.5% mixture in IMBA. The proposed SEC ends June 30, 1972 (NIOSH 2009). If claim-specific evidence is found for HEU from July 1, 1972 through October, 1983, the elemental uranium urinalysis and chest counts will be used to reconstruct dose. From October 1983 on, isotopic urinalysis results that indicate HEU (²³⁵U) will be used to reconstruct dose.

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5.2.6 Fission Product Analysis

Fission product urinalysis was the method for monitoring for intakes of fission products until the implementation of whole-body counting in 1960. Routine fission product urinalyses started in January 1947, but ferrous hydroxide precipitation was used on the supernatant from the plutonium lanthanum fluoride procedure, and the results were erratic with occasional breakthrough of ⁴⁰K. Therefore, data before 1948 should be considered unreliable and ignored.

As presented in Section 5.1.2, an SEC was designated in which NIOSH found that it was infeasible to reconstruct doses for fission products before August 31, 1946 due to inadequate monitoring (72 FR 55214). Therefore, dose reconstructions will not include any doses from fission products before August 31, 1946.

The procedure that was initiated in 1948 was to add strontium carrier to the aluminum oxide solution for the plutonium procedure, then precipitate lanthanum hydroxide. This procedure was shown to extract the rare earths and strontium with yields ranging from 90% for cerium to 23% for strontium. The dried planchet was counted for beta activity with an approximate detection level of 30 dpm (Healy 1948; Wilson 1987). The same procedure was in use in 1954 with the addition of a cerium carrier. It was also listed in the compilation of procedures that is referred to as the Old Bioassay Bible (Author unknown 1961), but that same compilation has a separate procedure for ⁹⁰Sr in urine. Another memorandum in the Old Bioassay Bible discusses the start of use of a gas-flow beta proportional counter in November 1958, which resulted in increased counting efficiency. The new detection limit was stated as $1.4 \times 10^{-5} \,\mu$ Ci/sample based on the counting efficiency of ⁹⁰Sr (Author unknown 1961). "Gross fission products" are also mentioned by Lardy (1970) with a brief description that seems to imply the same procedure was still available, although probably not used much. The detection level was given as 5 dpm/sample based on the beta counting efficiency for ⁹⁰Sr. Table 5-18 summarizes the most accurate detection levels for the fission product urinalysis found to date.

detection levels (upin/sample) [40].			
Period	MDA		
1948–2/1956	60 ^a		
3/1956–10/1964	70 ^b		
1965–approx. 1969	31		
1970 ^c	5		

Table 5-18.	Routine fission product urinalysis	
detection lev	vels (dpm/sample) [46].	

a. Reported as 30 dpm/sample but that value was more of a decision level than an MDA.

b. Recorded as 3.1 or $3.17 \times 10^{-5} \mu \text{Ci/sample}$.

c. Listed in the bioassay contract but probably not used; replaced by whole-body counting and ⁹⁰Sr urinalyses.

It is a challenge to interpret the fission product urinalysis in a way that is meaningful as representative of all the possible fission and activation products to which a worker could have been exposed. The procedure separated and counted radionuclides of alkaline and rare earths such as strontium, yttrium, barium, lanthanum, cerium, europium, promethium, zirconium, and niobium. It did not account for radionuclides of ruthenium, cesium, zinc, cobalt, or manganese. The radiochemical yield of the elements that were carried through to the final planchet varied, however, from about 23% for strontium to about 90% for cerium (Healy 1948). The abundances of all the fission products, relative to each other, varied considerably as a function of the time from when the reactor fuel was removed from the core and allowed to cool to when the contamination was inhaled or ingested. ORAUT-OTIB-0054, *Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses*, provides guidance on how to interpret fission product urinalysis results (ORAUT 2007a). Because the fission product urinalysis did not account for radiocesium, ⁹⁰Sr should be used

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as the indicator radionuclide in ORAUT 2007a. Section 5.4.1 contains a discussion about interpreting fission product mixtures.

After whole-body counting came into routine use, regular use of the fission product urinalysis continued for many workers at facilities such as B Plant and the Strontium Semi-Works, where intakes of pure ⁹⁰Sr were possible. Therefore, it was apparently being used as a ⁹⁰Sr bioassay. The records show the use of fission product analysis in this way until early 1964. The same workers show actual ⁹⁰Sr analysis results starting in 1965, probably starting with the new contract with UST. For 1960 through 1964, fission product urinalysis results can be interpreted as ⁹⁰Sr [47].

5.2.7 <u>Strontium</u>

Records of ⁹⁰Sr urinalyses, both routines and specials, begin to appear in the database in 1965. However, the compilation of procedures called the Old Bioassay Bible (Author unknown 1961) had a procedure specific for strontium in urine and fecal salts that included counting of total strontium and then allowing for ⁹⁰Y ingrowth, yttrium separation, and counting of ⁹⁰Y to account for ⁹⁰Sr separately from gross strontium beta, if desired. This procedure was also mentioned in a July 1963 memorandum about discussions between the Analytical Laboratories and Internal Dosimetry clarifying logistics of handling these samples and reporting ⁹⁰Sr results. Handwritten notes on this memorandum indicate that the detection level is about 20 dpm. Nevertheless, the database records show the use of fission product urinalysis into 1964 and ⁹⁰Sr urinalysis apparently starting in 1965. The database contains frequent entries of $1.67 \times 10^{-5} \,\mu\text{Ci/L}$ (37 dpm/L) during 1965 and 1966, which seems to be the reporting level. This is consistent with a draft of the first contract with UST (the official version has not been found) from August 1964 that lists a detection limit for ⁹⁰Sr as 25 pCi/1.5 L, which converts to 56 dpm/1.5L or 37 dpm/L (AEC 1964). Lardy (1970) states that the detection limit is 1 pCi/L (2.2 dpm/L) (at 90% confidence), and describes the procedure as precipitation as the oxalate, then nitrate, removal of yttrium and barium, then reprecipitation as the carbonate and gross-beta counting on gas flow proportional counters. A 1974 letter about terms of the statement of work with UST shows an "analytical limit" (defined as ±25%) at 50 dpm/sample and a reporting level of 2 dpm/sample (ERDA 1975). These values appear again in the 1978 statement of work (DOE 1979) except the analytical limit is defined as ±100%. Robinson (1979), in a letter to R. B. Swoboda (UST bioassay supervisor), requests changes for ⁹⁰Sr urinalyses to lower the analytical limit (±100%) from 50 to 5 dpm/sample, increase the reporting level from 2 to 5 dpm/sample, and add an emergency analysis capability with an analytical limit of 10 dpm/sample and reporting level of 5 dpm/sample. In 1982, the detection limit was listed as 2.5 dpm/sample for ⁹⁰Sr and 5 dpm/sample for ⁸⁹Sr. The contract starting in October 1983 (Simpson 1983) lists the detection limit as 2.0 dpm/sample, and it stayed at that value until 1992 when it was raised to 10 dpm/sample (Acker 1992). However, the procedure stayed the same throughout this period, and the true MDA probably held at about 2 dpm/sample.

Beginning in 1982, actual analytical results were recorded in the database; that is, results were not truncated at a reporting level.

The results of the ⁹⁰Sr procedure usually were reported as ⁹⁰Sr although sometimes a value for ⁸⁹Sr was also reported. Sometime in the 1980s a shortcut was added to the procedure that allowed skipping the ⁹⁰Y ingrowth portion of the procedure if the first beta count was less than 1 dpm [48]. When this happened, the result was reported as Sr total or SRTOT, but the result can be interpreted as ⁹⁰Sr. These results were below the required detection level. Table 5-19 summarizes the routine urinalysis detection levels for ⁹⁰Sr procedure.

Dose reconstructors should consider all strontium at Hanford to be absorption type F [50]. It is favorable to claimants to assume that ⁹⁰Sr and total radiostrontium results are ⁹⁰Sr even though ⁸⁹Sr might be present.

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MDA or MDC		
Might have been available but MDA not known		
1.67E-05 µCi/L (37 dpm/L)		
1.00E-06 µCi/L (2.2 dpm/L)		
50 dpm/sample ^a		
5 dpm/sample		
2.5 dpm/sample		
2 dpm/sample		
30 dpm/sample		
10 dpm/sample ^b		

Table 5-19. Routine ⁹⁰Sr urinalysis detection levels [49].

 Based on an unusual definition of analytical limit and probably conservative on the high side. Results <2 dpm were reported as 2 dpm;

results >2 dpm were reported as measured.

b. Decision level was 5 dpm/sample. Before that time, the MDA was also used as the decision level.

5.2.8 <u>Promethium</u>

Hanford was involved in the manufacture of heat sources using ¹⁴⁷Pm. The Hanford Site Description contains the history of possible exposure to pure ¹⁴⁷Pm (ORAUT 2010c). There were between 500 to 600 bioassay samples obtained from 1966 through 1979, mostly in the 1960s. The database shows only seven ¹⁴⁷Pm urinalyses from 1972 to 1975 but an upswing in the number of analyses from 1976 to 1979, so perhaps the project ended and restarted in 1976 for a few years. In addition, animal studies were conducted with ¹⁴⁷Pm as part of research to develop a human biokinetic model for the behavior of promethium in the body, so animal tenders might have been exposed to low levels of ¹⁴⁷Pm. A small study of 14 human volunteers using ¹⁴³Pm was conducted in 1967 or 1968 (Palmer, Nelson, and Crook 1969). Twelve volunteers received an injection of 0.1 µCi, and two volunteers received an ingestion of 10 µCi. Another study on the behavior of promethium used swine with ^{148m}Pm as the surrogate for ¹⁴⁷Pm (McConnon, Cole, and Smith ca. 1971).

1

The work on the heat sources involved conversion of promethium and cerium nitrates into Pm_2O_3 by separation chemistry then calcining (Howell and King 1968). In addition, there was one mention of cold-pressed, sintered Pm_2O_3 for heart implants. According to ICRP (1995), the nitrate form should be considered absorption type M and the oxide form absorption type S.

The units in the database are microcuries per liter for urine and microcuries per kilogram for feces through 1974, then disintegrations per minute per sample from 1975 to the present. Table 5-20 lists the ¹⁴⁷Pm MDAs or minimum detectable concentrations (MDCs) at various times.

Period	MDA or MDC	
1963–1964	2.00E-06 µCi/250 mL ^a	
1965–3/25/1970	1.67E-05 µCi/L (37 dpm/L)	
3/31/1970–1974	1.00E-05 µCi/L (22 dpm/L)	
1975–1979	50 dpm/sample ^b	
1980–1981	20 dpm/sample	
1982–9/1983	5 dpm/sample	
10/1983–6/1990	4 dpm/sample	
11/1991–present ^c	30 dpm/sample	

Table 5-20. Routine ¹⁴⁷Pm urinalysis detection levels [51].

a. Ludwick (1964, pp. 3.21-3.24)

 Based on an unusual definition of analytical limit and probably conservative on the high side. Results < 25 dpm were reported as 25 dpm; results > 25 dpm were reported as measured.

c. No ¹⁴⁷Pm analyses were performed during 1990 and 1991.

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Fecal samples were analyzed for ¹⁴⁷Pm for some of the potential intake events in the late 1960s. The MDA or at least the lowest reporting level appears to be $1.67 \times 10^{-5} \mu \text{Ci/kg}$ [52]. An MDA for fecal samples does not appear in laboratory statements of work during the 1970s, but reappears in the 1980s as 28 to 110 dpm/sample in 1982 dependent on sample size (roughly 400 dpm/kg); the value was 220 dpm/sample from 1983 to the 1990s [53].

The first promethium urinalysis development was in 1962 (Perkins 1963a,b, pp. 3.9–3.11). It appeared to be under development in late 1962 and finished in February 1963. This procedure extracted rare earths and was followed by a beta count. If sufficient beta activity was detected, then the ¹⁴⁷Pm component was determined by shielding to separate high-energy betas from the low-energy betas from ¹⁴⁷Pm (Perkins 1963a). This procedure was used to analyze samples from the 29 workers who were involved in the February 12, 1963, contamination spread in the 325 Building (AEC 1963, Appendix D).

Another urinalysis procedure that used liquid scintillation counting was developed in 1963 and presented in the 1963 annual report of the Radiological Sciences Group (Ludwick 1964, pp. 3.21-3.24). This method was described as being more amenable to routine processing. The stated detection level was 2 x 10^{-6} µCi for a 250-mL sample, which is close to the reporting level for the routine program later in the 1960s of 1.7 x 10^{-5} µCi/L.

The one description of the routine procedure that was used by UST appeared in documents from 1970, 1974 (Lardy 1970; UST 1974). Promethium and rare earths were precipitated as the fluoride. Interferences such as zirconium, scandium, and IV actinides were removed by extraction by TTA in xylene, first at a pH of less than 1, then at a pH of about 4. The final sample was counted by liquid scintillation. Remaining rare earths were distinguished from ¹⁴⁷Pm by proper setting of the counting window on the liquid scintillation spectrometer.

In vivo counting based on bremsstrahlung from ¹⁴⁷Pm betas was developed with an MDA of 0.5 to 1.5 nCi (Wilson 1987, p. 4.7) but was not used extensively [54].

5.2.9 <u>Polonium</u>

As presented in Section 5.1.2, a class was recommended by NIOSH for addition to the SEC for all employees from October 1, 1943 to June 30, 1972. NIOSH found that it was infeasible to reconstruct doses for polonium, neptunium, and thorium due to inadequate monitoring (NIOSH 2009). The Hanford Site Description (ORAUT 2010c) contains a historical summary of work with polonium.

L

A urinalysis procedure was developed in 1948 apparently in response to an accident involving a Po-Be source in which two workers had potential for intake of ²¹⁰Po (Thorburn 1948). The procedure was described as wet ashing, then solubilization in 2N HCl with 5 g of hydroxylamine hydrochloride and soaking with a silver foil for 48 hours. The polonium was attached to the silver foil, dried, and counted. A detection limit was not given. The control blank was 0.12 ± 0.2 dpm. The letter does not state the sample size but seems to imply that the results were disintegrations per minute per liter. That would imply an MDA of about 1 dpm/L. No other records of ²¹⁰Po urinalysis have been found, so it might have been only rarely used before 1968. The 1954 bioassay annual report stated that the bioassay laboratory was performing routine analyses for polonium on a few individuals (Watters 1954, p. 4).

Considerable activity toward reinitiating a bioassay procedure and establishing a biokinetic model for ²¹⁰Po was found in the files from about 1968 through the mid-1970s. UST was asked to develop a bioassay procedure in March 1968 and did so shortly thereafter.

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The procedure UST developed for ²¹⁰Po in March 1968 was as follows. For urine, gold, mercury, platinum, and tellurium were removed by reduction in hydrazine in an HCl solution. Iron was removed by reduction with ascorbic acid. The polonium was then removed from solution by deposition on silver film by heating at 95°C for 2 hours. The silver film was counted by alpha proportional counting. Fecal samples were first wet-ashed in concentrated nitric acid and peroxide, then treated the same as urine samples. Sometime between 1968 and 1974, copper foil replaced silver foil and alpha spectrometry counting replaced proportional counting (UST 1974). Detection limits for routine urinalysis are shown in Table 5-21 and for nonroutine excreta bioassay in Table 5-22.

Period	MDA or MDC
3/1968–1970	5.4E-7 μCi/L
1971–1974	5.0E-7 μCi/L
1975–1979	1 dpm/sample ^a
1980-9/1983	0.1 dpm/sample ^a
10/1983–present	No longer listed in the contract except for expedited or emergency samples. Probably not used.

Table 5-21. Routine ²¹⁰Po urinalysis detection levels [55].

 Based on an unusual definition of "analytical limit" and probably conservative on the high side. Reporting level listed as 0.5 dpm/sample.

Table 5-22. MDAs for nonroutine ²¹⁰Po excreta analyses (dpm/sample unless otherwise noted)[56].

	Fecal sample MDA			Urine sa	mple MDA
Period	Emergency ^a	Expedite	Priority	Emergency ^a	Expedite
3/1968–1973	NA	NA	5.4E-7 µCi/kg	NA	NA
1974–9/1983	NA	NA	(b)	NA	NA
10/1983–9/1985	340	NA	NA	0.8	NA
10/1985–6/1989	340	100	NA	0.8	0.1

a. At times the emergency category was called "rush" and the routine category was called "normal."

b. Probably available but not listed in the contract.

Because ²¹⁰Po is a natural radionuclide from the ²³⁸U decay chain, it exists naturally in urine and feces. Nothing was found in the records to indicate that a study on natural excretion levels for persons who lived around Hanford had been conducted.

5.2.10 <u>Neptunium</u>

As presented in Section 5.1.2, a class was recommended by NIOSH for addition to the SEC for all employees from October 1, 1943 to June 30, 1972. NIOSH found that it was infeasible to reconstruct doses for polonium, neptunium, and thorium due to inadequate monitoring (NIOSH 2009).

L

At 222-S (1958 through 1962) and PUREX (1958 through June 1972), ²³⁷Np was removed from the dissolved fuel, purified, and packaged for shipment off the site. Until the final purification step, the material had more activity of plutonium than neptunium; however, the final product was very pure. Neptunium target elements, which used the purified neptunium, were manufactured in 231-Z and in the 308 and 3708 Buildings intermittently from 1958 to 1967. See the Hanford Site Description for details (ORAUT 2010c).

A urinalysis method for ²³⁷Np was developed in 1960. There is no evidence in the database that suggests this was used until 1972, but hard-copy results might show up in workers' files for earlier times. The procedure used neptunium separation that was followed by electrodeposition and alpha track counting in the same manner as for plutonium urinalysis samples, with essentially the same sensitivity (0.05 dpm/24-hr sample) (Perkins 1961, p. 4).

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The database contains only four bioassay samples for 237 Np in the 1970s, all for PNNL workers; all four are baseline samples. No routine samples or ending work samples were collected. The value for all four samples was 1×10^{-7} with no units shown. The practice at the time was to record the activity of excreta samples in units of microcuries, but it has not been determined to date if the units are per sample or per liter. No information about the analysis method has been found [57].

A few sample results appear in the database in the 1980s, mostly for PNNL employees. The units are in disintegrations per minute per sample [58]. The 1983 statement of work with UST (Simpson 1983) shows a nonroutine analysis for ²³⁷Np with contractual detection levels of 0.02 dpm/sample for urine samples and 0.1 dpm/sample for fecal samples, which were consistent with alpha spectrometry for plutonium and americium at the time.

5.2.11 Other Limited-Exposure Radionuclides

Hanford has always been a center for research, first as part of Hanford Works, then (1965 to present) as part of PNNL. As such, small-scale use (in terms of either the number of persons or activity of the source) of various radionuclides that are not addressed above has occurred throughout the history of the Hanford Site. The following discussion addresses ¹⁴C, ²³²Th, radon, ⁹⁰Y, ²²⁷Th, ²²⁷Ac, and ³²P, but it is probably not comprehensive.

Carbon-14

Exposure occurred at the 3731 Building in the mid-1950s when irradiated graphite samples were brought to the building from the operating reactors for destructive testing. No information has been uncovered about what bioassay, if any, was done. Carbon-14 was also used as a tracer in biological experiments. One documented study was conducted in the late 1990s in the Life Sciences Laboratory-II (LSL-II) that involved a total of about 4 Ci of ¹⁴C. Urinalyses were obtained on about 20 researchers. The MDA was 10 dpm/mL (Author unknown 1987; Lyon et al. 1991; Carbaugh 1997). Baseline samples were obtained from each worker because natural excretion levels had not been established. ICRP (1995, 1996) assigns ¹⁴C in organic compounds to class SR2.

<u>Thorium</u>

Thorium urinalysis capability apparently existed at least as far back as 1955 (Nickelson et al. 1955, pp. 6d, 7d). However, there is no indication in the database of thorium samples until 1979. The 1955 monthly report indicates a detection limit of 0.077 μ g but does not state sample size. There are some thorium urinalysis samples in the database beginning in 1979. These are identified as either TH or TH232. The TH urinalysis was a total thorium analysis by spectrophotometry, with a detection level of 1 μ g/L. The TH232 analysis was specific for ²³²Th and used alpha spectrometry. The detection levels was listed as 0.5 dpm/sample in 1970 (Lardy 1970) and 0.03 dpm/sample in 1983 (Simpson 1983). Some work was done with ²³²Th slurries in the 3720 Building in the mid-1990s. The plan was to collect baseline urine samples on the few involved workers, then collect special bioassay samples if air samples exceeded a cumulative exposure of 40 derived air concentration (DAC)-hr. The urinalysis MDA was stated to be 0.1 dpm/sample (Bihl 1994; Lyon et al. 1998).

Sulphur-35

There are some urinalysis results for ³⁵S in 1967. According to Lardy (1970), analysis was by ashing then liquid scintillation counting with a detection limit of 10 dpm/sample. However, as with ¹⁴C, the results were reported as microcuries per liter and the reporting level was apparently 1 μ Ci/L.

<u>Radon</u>

A radon generator was used for animal studies in the 108F Building; it was later moved to LSL-II. Monitoring was probably just by air sampling, but no information has been discovered yet. The radon generator was housed in a glovebox and vented to the outside. There should have been only a few potentially exposed researchers [59].

Phosphorous-32

This radionuclide was used for biological tracer studies and, according to one retired researcher, "pipetting was done by mouth in the old days" [60]. Such exposure would be limited to a few researchers and would have to be established through the claimant interview or by some indication of ³²P bioassay samples in the worker's record. More information might be found if such a case is encountered. The whole-body counter was calibrated for ³²P measurements circa 1965, and there might be personnel count results in the records (Keene 1965, p. F-3).

In addition, some unusual radionuclides were isolated in the 325 Building for nuclear medicine studies in the mid to late 1990s.

Yttrium-90

One of these studies isolated ⁹⁰Y from ⁹⁰Sr and packaged and shipped the ⁹⁰Y to users around the world. Only a few workers were involved. The work was monitored by air samplers and no loss of control of the material occurred, so no bioassay was obtained. The material was in an insoluble form, so chest counting would have been the only possible bioassay because of the 64-hour physical half-life, but the need to perform chest counting never arose (Carbaugh 1990; Bauman 1996).

Thorium- and Actinium-227

Another project circa 1995-96 involved milking ²²⁷Th from ²²⁷Ac on an ion exchange column. A bioassay procedure was developed specifically for this project under the assumption that the project was going to continue for several years, but it ended after only a few milkings. Only a couple of researchers were involved (Bihl 1995). No contamination spreads or high air samples occurred. The bioassay procedure had a stated MDA of 0.1 dpm/sample for ²²⁷Th.

5.3 *IN VIVO* MINIMUM DETECTABLE ACTIVITIES, ANALYTICAL METHODS, AND REPORTING PROTOCOLS

In vivo counting equipment and techniques were developed in the late 1950s and have been in routine use for measuring X- and gamma-ray-emitting radionuclides since 1960. [Unless otherwise noted, the *in vivo* information below is from Wilson (1987) and Lynch (2001).] The unit for all *in vivo* measurements is nanocuries.

5.3.1 Whole-Body Counters

The first whole-body counting started in mid-1959, and it became a routine method in 1960. It consisted of a single Nal crystal (9.375-in. diameter and 4-in. thick) in a counting room with 10-in. thick pre-World War II steel plates on all six sides and graded shielding on the inner surfaces (lead, cadmium, and copper) (Wilson 1987; Roesch, McCall, and Palmer 1960). This was called the Iron Room. The counting geometry was a chair that was configured to simulate a 1-m arc. The original count time was 20 minutes, which was reduced to 10 minutes in October 1962 (Swanberg 1962). A second, same-size Nal detector was added in 1963 (Brady 1964). According to personal recollection of H. E. Palmer, the two-detector system improved the detection capabilities somewhat. However, the MDAs in a report from the fall of 1964 were the same MDAs as those in Mr. Palmer's Laboratory Record Notebook in 1960 [61], so the difference between the systems was apparently not great enough to warrant republishing the MDAs. Therefore, the MDAs in Table 5-23 are the only MDAs found for the 1960s and 1970s, and apparently were meant to apply generally to the various whole-body counters in operation during this period. Although they were referred to as detection limits, the calculations were more consistent with what is presently referred to as decision levels.

Shortly after the chair counter in the Iron Room became operational, an entirely new design called the shadow shield counter was developed. The shadow shield consisted of a bed shielded on the bottom and sides by lead. The bed moved under a large NaI crystal (11.5-in. diameter by 4-in. thick) that was

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Table 5-23. Routine whole-body counting detection levels (nCi).^a

Period	Nuclide	MDA	Decision level	Reporting level
	Na-22		1.0	10
	Na-24		0.3	0.3
	Cr-51		50	50
1960–1976 ^b	Fe-59		2.0	10
1900-1970	Co-60		0.4	10
	I-131		0.5 ^c	10
	Cs-137		0.5	0.5
	Zn-65		1.6	0.8
	Na-22		1.0	10
	Na-24		0.5	0.31
	Cr-51		15	15
	Mn-54		2.0	10
	Fe-59		4.0	10
	Co-60		2.0	10
1077 100 1 ^d	Zn-65		3.0	0.75
1977–1984 ^d	Zr/Nb-95		2.0	10
	Ag-110m		2.0	10
	Ru-106		12	12
	Sb-125		3.0	10
	I-131		4.0 ^c	10
	Cs-137		2	0.66 ^e
	Ce-144		100	100
	Na-22		1	1.5
	Na-24		1	0.31
1985–86	Mn-54		1	3
	Fe-59		2	6
	Co-60		1	1
	Zn-65		2	0.75
	Zr-95	3	1	3
	Ru-106	12	4	12
	Eu-154	4.5	5	4.5
	Cs-137	3	1	0.66
	Na-22	1.5		Anything detected
	Mn-54	3		6
(a a -f	Co-60	3		5
1987 ^f	Fe-59	6		Anything detected
	Cs-137	3		6
	No changes for other radio		ing detected is reported	
40000	New formalism for decision			
1992 ^g	decision level.		.,	
4 a a a h	Actual values, regardless of	of amount, repor	ted for Co-60 and Cs-13	37. including negative
1993 ^h	numbers.	· · · · · · · · · · · · · · · · · · ·		,
	Co-60	4		Every result
	Cs-137	4		Every result
	I-131	5	1	Every result
1005 10/1000	Mn-54	3	1	Every result
1995–10/1999 ⁱ	Na-22	2		Every result
	Na-24	1		Every result
	Pr-144 (Ce-144)	230		Every result
Ļ	Other radionuclides		1	Anything detected

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Period	Nuclide	MDA	Decision level	Reporting level
	Co-60	1.25	g	Every result
10/1999-	Cs-137	1.3	g	Every result
present ^{i,j}	Eu-154	3.75	g	Every result
	Other radionuclides			Anything detected

a. Nominal MDAs based on the phantom available at the time, the routine count time, and the least sensitive of various whole-body counters in operation at the time. Listing of an MDA for a given radionuclide does not necessarily mean that that radionuclide was frequently encountered. If smaller MDAs are listed in the database for a given count, use them.

b. Based on 95% confidence of detection [62]. There were some special counts for thorium; see Section 5.3.4.

c. See discussion on thyroid detectors.

d. Based on 99% confidence of detection (Wilson 1987).

e. The reporting level cutoff of 0.66 nCi for Cs-137 appears to start in 1975 [63].

f. From Lyon et al. (1988).

g. From Lyon et al. (1993).

h. [64].

i. Least sensitive of many options during the period. Much better sensitivities were available using the hyperpure germanium (HPGe) system (PNL 1995, Tables 7.2 and 7.3).

j. Physical configurations stayed essentially the same but ABACOS software introduced changes to the methodology for determining MDAs and decision levels. Decision levels were determined by software on a count-by-count basis (Lynch et al. 2000).

also shielded by lead with the exception of the downward-looking face that looked directly onto the body as it passed under the crystal. The shadow shield detector was mounted in a mobile trailer and moved to areas nearer the Hanford worksites. The trailer also had a thyroid detector and a wound counter. The shadow shield detector became operational in 1963 (Brady 1964). The mobile counter was described as having comparable sensitivity to the "larger, conventional whole-body counters installed in massive iron rooms. There is, however, some decreased sensitivity in the lower energy region below about 300 keV, due to increased contribution to the background from scattered radiation" (Swanberg 1963).

A report on the radionuclides detected in workers at the whole-body counter facility in 1961 listed ²⁴Na, ⁶⁰Co, ⁶⁵Zn, ⁹⁵Zr, ⁹⁵Nb, ⁹⁹Mo, ⁹⁹Tc [presumably ⁹⁹Mo], ¹⁰³Ru, ¹⁰⁶Ru, ¹³¹I, ¹³⁷Cs, and ¹⁴⁴Ce (Henle 1962). A similar report that summarized 1961 to 1963 results added ⁴⁶Sc, ⁵¹Cr, and ⁵⁹Fe to the list (Brady 1964).

A shadow shield whole-body detector was added at the Whole Body Counting Facility in 1977. This assembly had two 35% lithium-drifted germanium (GeLi) detectors and a 4-in. by 4-in. by 16-in. Nal detector. It ceased operation in 1987 when the two new counting rooms were added. Wilson (1987) provided a list of MDAs that applied to 1980. These are used to represent this shadow shield detector.

By 1978, there were four shadow shield whole-body counters available for use: one at the Whole Body Counting Facility, two in mobile trailers, and one at the Emergency Decontamination Facility. The latter was designated for use for large acute intakes with potentially high levels of external contamination.

A "standup" counter put in operation in 1983 is still in operation today. It consists of five vertically stacked NaI crystals in a small lead-shielded area. The worker stands in front of the detectors with the detectors to his or her back; the detector array is raised or lowered to best fit the height of the worker. There are four 9.375-in.-diameter by 4-in.-thick detectors and one 11-in.-diameter by 4-in.-thick detector; the latter is located behind the thoracic region. Count time is 200 seconds (Palmer, Rieksts, and Spitz ca. 1984).

A coaxial HPGe scanning array developed in July 1989 is still in operation today (Lyon et al. 1990). For this system the person lies on a bed in a shielded room and the detector array moves under the bed. The configuration of this system, in terms of number and size of the detectors, has changed

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many times. It started as four 68% HPGe detectors; one of the detectors was replaced with a 120% detector in late 1995; in May 1997 the system was upgraded to include seven detectors including three 120% detectors. As a four-detector array, the system was used only when a count on the "standup" counter had detectable activity of an occupationally related radionuclide. However, it was considered the count of record. In 1997, because of its greater resolution and lower decision levels, it started to be used for routine counts for workers who were exposed to mixtures of ¹³⁷Cs and plutonium. The count time is usually 10 minutes, but 20-minute count times are used as confirmation of an initial count with detectable activity. As a consequence, the database usually shows both 10-and a 20-minute counts on the same day or a few days apart if the first count had detectable activity (excluding ⁴⁰K or medical radionuclides) (Lyon et al. 1998).

The radon progeny ²¹⁴Bi and the thoron progeny ²⁰⁸TI were and are present in the background spectra from the shielded rooms used for *in vivo* measurements of workers at Hanford. To account for this contribution when calculating worker results, the background levels of these nuclides have been estimated from background measurements in the rooms with the detectors being shielded with an appropriate phantom. The environmental background count rates, in addition to the continuum background count rates, are subtracted from the gross count rates in the appropriate region of interest of the worker spectra to obtain an activity for these nuclides. This approach was implemented in May 1992 (Lynch 1992).

Even with the background contribution subtracted, activity levels of ²¹⁴Bi were measured periodically that were higher than the room background levels due to activity on the workers' clothes, skin, and hair. To compensate for this, derived investigation levels (DILs) were implemented in October 1995 for ²¹⁴Bi and ²⁰⁸TI measurements from the coaxial germanium counting system. The DIL values were 6.47 nCi for ²¹⁴Bi and 0.75 nCi for ²⁰⁸TI, which represent the upper 99% confidence levels for an unexposed population. Results were considered to be above detection and were reported to the REX database only if the value exceeded the DIL value. The use of the DIL values continued until November 1999 when a new software system was implemented. Since 1999, ²¹⁴Bi and ²⁰⁸TI results have not been reported unless specifically requested.

Use of the first mobile counter at onsite locations stopped in the early 1980s. A new trailer was obtained in 1989 and reconfigured with a new standup counter consisting of five 4-in. by 16-in. by 4-in. thick Nal detectors plus one 4-in. by 8-in. by 4-in. thick detector. The trailer was parked in the 200 East Area and operated remotely starting in 1991 (Lyon et al. 1992). The sensitivity of the detector was comparable to that of the standup counter at the Whole Body Counter Facility. The use of this facility was infrequent, and it was discontinued in August 1995 (Lyon et al. 1996).

From 1960 to 1983, four radionuclides were reported routinely: ²⁴Na, ⁴⁰K, ⁶⁵Zn, ¹³⁷Cs [65]. Potassium-40 is strictly a natural source and can be ignored. Sodium-24 and ⁶⁵Zn were detectable in many whole-body counts in the 1960s. Most of these intakes came from sanitary water drawn from the Columbia River. The onsite reactors and many of the cities around the Site used the Columbia River for sanitary water, although the ²⁴Na intakes came from the drinking water at the reactors and 200 Area (which received water from the 100 Area) because its short physical half-life greatly reduced the activity in city water supplies (Brady 1964). Therefore, intakes of ²⁴Na and ⁶⁵Zn can be considered chronic ingestions, with the exception of the highest body burdens that were statistically different from the general background in Hanford workers and had to come from inhalations. Guidance to distinguish acute inhalations from chronic ingestions based strictly on the measured activity level has not been developed. Therefore, barring evidence in the employee's file that ties the intake to an acute event, the dose reconstructor can assume either chronic ingestion or inhalation in accordance with which is more favorable to the claimant for the organ of concern. Chronic ingestion intakes would have been occurred before implementation of whole-body counting, probably from shortly after the startup of the first once-through-cooling reactor (August 1944), and they would have ceased shortly after the shutdown of the 100 KE reactor (January 1971) [66].

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Net counts in a fifth region of the spectrum were commonly calculated but not usually associated with a radionuclide. This was the low-energy portion of the spectrum noted as the GOK region. The technique was to calculate the activity of the higher energy radionuclides ²⁴Na, ⁴⁰K, etc., then subtract the Compton scatter contribution from those radionuclides to see if any counts were left over in the low-energy region. If sufficient counts were left over, they would have been investigated further to see if an occupational radionuclide was the source, recognizing that the low-energy region was also subject to increased electronic noise and general background scatter in the crystal [67].

If the hard-copy form (In-Vivo Counter Results) shows the "traces of xxx invalidate routine calculation" statement, some radionuclide other than the standard four was detected; this was often ⁶⁰Co. The activity of that radionuclide might be written on the form. Activities that exceeded 10 nCi or 1% of the MPBB were calculated and reported on a Whole Body Counter Evaluation form (Glenn 1968). See Section 5.3.6 for instructions.

Most workers in the early days of whole-body counting had detectable activities of ¹³⁷Cs. Most of this was attributed to fallout. Some workers had even higher levels of ¹³⁷Cs from consumption of wild game. A decision level to establish the difference between occupational and nonoccupational sources of ¹³⁷Cs intake has not been found in the records, and might not have been developed as long as the ¹³⁷Cs measurement did not exceed 1% of an MPBB. Dose reconstructors can use the following guidance: [68]

- Consider the ¹³⁷Cs intake occupational if the same whole-body count detected other fission or activation products (excluding ⁶⁵Zn or ²⁴Na). Also consider it occupational if a fission product or radiostrontium urinalysis showed detectable activity and the sample was obtained within the period between the previous and next whole-body counts.
- If an investigation record clearly shows that the intake was due to a nonoccupational source, disregard the ¹³⁷Cs.
- NCRP Report 94 provides mean body burdens of ¹³⁷Cs for the United States for the years most likely to produce interference with occupational whole-body count results (NCRP 1988). Table 5-24 lists those values. If no other fission or activation products are linked to the intake (excluding ⁶⁵Zn or ²⁴Na) and the ¹³⁷Cs result is less than the values in Table 5-24, the dose reconstructor may assume the ¹³⁷Cs result is due to fallout.

Year	Body burden	Year	Body burden
1953	0.27	1966	9.7
1954	1.1	1967	5.6
1955	2.2	1968	3.5
1956	4.3	1969	2.7
1957	5.1	1970	2.7
1958	6.5	1971	2.7
1959	8.1	1972	2.7
1960	6.8	1973	2.7
1961	4.6	1974	1.6
1962	6.0	1975	1.1
1963	11	1976	1.6
1964	19	1977	1.1
1965	16		

Table 5-24. Mean body burdens of ¹³⁷Cs from fallout in the United States (nCi).^a

a. From NCRP (1988).

5.3.2 Chest Counters

In 1967, PNNL started using the original large Nal detector in the Iron Room for chest counting, with emphasis on uranium workers. The detector was placed directly over and nearly in contact with the chest region with the worker in the supine position. Count time was 30 minutes. MDAs were determined to be 6.7 nCi for "U natural" (presumably based on ²³⁴Th), 0.15 nCi for ²³⁵U, and 0.33 nCi for ²⁴¹Am. However, in the next year a new counting room, called the Lead Room, was built specifically for chest counting. It had four 5-in.-diameter by 0.375-in.-thick Nal detectors, two in front and two in back of the subject. Count time was 30 minutes (Wilson 1987). A lung phantom with variable chest wall thickness was developed for calibration of the new system. MDAs were listed as 0.15 to 0.6 nCi for ²⁴¹Am, 2.0 to 3.7 nCi for ²³⁴Th (assumed to be in equilibrium with ²³⁸U), and 0.17 to 0.37 nCi for ²³⁵U, dependent on a subject's weight-to-height ratio (Andersen 1971; Wilson 1987). (Chest count MDAs are summarized in Table 5-25.) MDAs for direct measurement of ²³⁸Pu and ²³⁹Pu using the 17-keV X-rays were calculated at times, but the values were extremely large in relation to the maximum permissible lung burden, so primary reliance was placed on measuring ²⁴¹Am and applying a plutonium-to-americium ratio. The chest counter was also calibrated to measure bremsstrahlung radiation from ⁹⁰Sr or ¹⁴⁷Pm, although these were probably not routine counts. MDAs for those counts were listed as 25 to 40 nCi and 0.5 to 1.5 µCi for ⁹⁰Sr and ¹⁴⁷Pm, respectively (Andersen 1971). A second chest counting system became operational in 1978 (Wilson 1987). A phoswich detector became available and was used occasionally for special chest counts but was never implemented on a routine basis [69].

A solid-state germanium counting system using three planar HPGe detectors (soon upgraded to six) replaced the Nal detector in the Iron Room chair counter in 1983 (Palmer, Rieksts, and Spitz ca. 1984). The HPGe detectors provided better spectral resolution than the Nal detector, thus lower backgrounds in the region of interest and better discrimination against radon decay products and better detection of low-energy photon emitters in the presence of large activities of high-energy photon emitters (e.g., ¹³⁷Cs or ⁶⁰Co). A thin window on the end of the detector faced the chest for better transmission of low-energy photons. The detectors were positioned over the front of the chest (two over the right lung) with the subject in the supine position. Counting time was 2,000 seconds. MDAs were quoted for "an average size person" as 0.1 nCi for ²⁴¹Am, 0.5 nCi for ¹⁴⁴Ce, 0.7 nCi for ²³⁴Th (²³⁸U), and 0.05 nCi for ²³⁵U (Palmer and Rieksts 1984). These values were quoted as being the reliably detectable activity (RDA), which was defined as 3 standard deviations of the background continuum, and discernable by naked-eye inspection of the spectrum (Carbaugh et al. 1988). Special chest counts as follow-up to high routine chest counts or on special request were twice the normal counting time, so the MDAs were somewhat lower.

Within less than a year, the three-detector system was upgraded to a six-detector array, which enabled reduction of routine counting times to 1,000 seconds with nearly the same RDAs (Palmer, Rieksts, and Spitz ca. 1984; Carbaugh et al. 1988). A second HPGe detector array became operational in July 1989 in a new shielded cell called the Stainless Steel Room because the inner (i.e., visible) lining of the graded shield was stainless steel. Although intended to be a six-detector array, this counter had only four detectors at first because of operational problems with the detectors. Counting times were increased to 2,000 seconds for the six-detector array and 3,000 seconds for the four-detector array (Lyon et al 1990).

In September 1994, the chest counter in the Stainless Steel Room was converted to a four-detector array using larger area detectors (Lyon et al. 1995). The same change was implemented in the Iron Room in June 1996 (Lyon et al. 1997). This configuration continues to the present. The routine counting time was increased to 3,000 seconds for the larger area arrays in November 1995; special counts and recounts were 3,600 seconds.

Table 5-25. Routine chest counting detection levels (nCi) (Andersen 1971; Lyon et al. 1988, 1989, 1990, 1993, 1994; MacLellan et al. 1999; Lynch et al. 2000).

Period	Radionuclide	MDA
1967	Am-241	0.33
	U-238 (Th-234)	6.7
	U-235	0.15
1968–1983	Am-241	0.15–0.6 ^a
	U-238	2.0–3.7 ^a
	U-235	0.17–0.37 ^a
	Sr-90	25–40 ^a
	Pm-147	0.5–1.5 ^a
1983–1986	Am-241	0.24 ^b
	U-238	1.1 ^b
	U-235	0.08 ^b
	Ce-144	0.78 ^b
	Eu-154	0.07
1987	Am-241	0.28 ^c
	U-238	1.8
	U-235	0.12
	Ce-144	0.6
	Eu-154	0.07
1988–6/1989	Am-241	0.28 ^c
	U-238	1.8
	U-235	0.12
	Ce-144	0.6
	Eu-154	0.07
7/1989–1991 ^d	Am-241	0.18 ^c
	U-238	1.2
	U-235	0.08
1992–5/1996 ^e	Am-241	0.18 ^c
	U-238	3
	U-235	0.2
6/1996-10/1999	Am-241	0.28 ^c
	U-238	1.6
	U-235	0.095
11/1999-present	Am-241	0.25 [°]
	U-238	1.5

a. Range for different weight-to-height ratios, a chest wall thickness adjustment for both front and back chest walls. Use highest value for default to cover large persons.

- b. Assumed MDA = (RDA)(4.65/3). Am-241 adjusted for 95thpercentile male chest wall (0.2/0.13) (Lynch 2003)
- c. Adjusted for 95th-percentile male chest wall.
- d. Cerium-144 and Eu-154 no longer automatically reported for chest counts because they now can be quantified in the germanium whole-body counter.
- e. Applies to the six-detector array. Better sensitivity was obtained by the four-large-area-detector array in the Stainless Steel Room.

Ultrasonic measurements of chest wall thickness for workers who had activity in the lung began in about 1978 and continues today (Palmer and Rieksts 1979). Therefore, decision levels for nondetected activities use a weight-to-height ratio to estimate chest wall thickness, whereas detected activity is corrected for chest wall thickness using ultrasound [70].

Individual-specific decision levels were reported to the database for each count and each radionuclide starting in 1992 (Lyon et al. 1993).

For *in vivo* counting, the assumption was made that ²³⁴Th was in equilibrium with ²³⁸U [71]. This was a reasonable assumption at Hanford. Uranium that was recently separated from dissolved fuel was certainly not in equilibrium, and uranium being treated at the UO₃ Plant might have been in equilibrium dependent on how long it had taken the material to go through the separation process and be transported to the UO₃ Plant. However, uranium in this part of the fuel cycle was very soluble and not important in relation to chest counting. Chest counts were used to monitor for intakes of insoluble forms of uranium, which were very old forms in terms of time since purification from decay progeny (e.g., machining on metal and uranium metallurgy studies) [72].

5.3.3 <u>Thyroid Counters</u>

Thyroid counting appears to have started on a limited basis for high-risk workers at least as early as 1956 (see Section 5.6 for a discussion on thyroid counting in 1945 and 1946). Wilson (1960) states,

At the present time routine thyroid monitoring is conducted on a limited basis in the REDOX and PUREX facilities. Generally the pattern for coverage in the PUREX facility includes about four to five employees weekly picked from the sampling crews, crane operators, and a Radiation Monitor assigned to the stack area. At the REDOX facility routine monitoring is accomplished on a weekly basis for the shift crane operators.

The letter continues to discuss counts and other data that were obtained in 1959, but there is no indication if those results were placed in workers' files. Radiation monitoring data sheets from 1956 show that results below 10 nCi for ¹³¹I were recorded as less-than values.

The first mobile whole-body counter had a thyroid counter that consisted of a 3- by 3-in. Nal detector (assumed to mean 3-in. diameter by 3-in. thick) that was positioned next to the neck. The MDA was 0.020 nCi for ¹³¹I for a 30-minute count (Wilson 1987). The same detector and MDA were included in a description of *in vivo* counting capabilities at the Whole Body Counting Facility by Andersen (1971) and again by Palmer (1985). In 1986, the first listing of MDAs in the database associated with each count occurred; because the MDA is different for each thyroid count, person-specific calculations were used. There is a cluster of thyroid counts with MDAs that range from 0.020 to 0.050 nCi and another cluster that ranges from 0.22 to 0.298 nCi. The second cluster occurred only in October 1986 and would seem to imply use of a different detector for some reason. The count-specific MDAs after October 1986 vary from 0.07 to 0.006 nCi [73].

The presence of ¹³¹I would have been detected in a whole-body count, but the recorded quantity would have been based on a whole-body calibration rather than a thyroid calibration [74].

It is important to note that, during some years, ¹³¹I that was measured in workers who had radioiodine medical studies or treatment was recorded in the database; the workers' files should have a hard-copy *in vivo* count form that notes the ¹³¹I was due to a medical procedure. Dose reconstructors must look for such a note in the records of anyone who has a recorded ¹³¹I measurement if the ¹³¹I result has impact on the POC.

For counting ¹²⁵I in the thyroid, a thin, 2-in.-diameter Nal crystal with a beryllium window was used starting at least as far back as 1967. The thickness of the crystal has not been determined yet. The MDA was listed as 0.11 nCi for a 1-minute count or 0.07 nCi for a 10-minute count, but there was no mention of which count time was regularly used. There probably were not many workers who were exposed to ¹²⁵I on a regular basis, but there are indications of a contamination spread in 1978 that involved several workers (Palmer and Rieksts 1979). The same counter is described for thyroid

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counting in 1982, except the "reporting level" is given as 0.020 nCi; it is not known if this better sensitivity came from a longer count time, better positioning, or an improved crystal.

By 1985, thyroid counting for ¹²⁵I was performed using two intrinsic germanium detectors, with an MDA of 0.005 nCi for a 2,000-sec count.

Thyroid counting for either of the iodine isotopes has been rare since 1987.

5.3.4 Thorium Exposure and Monitoring

As presented in Section 5.1.2, a class was recommended by NIOSH for addition to the SEC for all employees from October 1, 1943 to June 30, 1972. NIOSH found that it was infeasible to reconstruct doses for polonium, neptunium, and thorium due to inadequate monitoring (NIOSH 2009).

Thorium exposure occurred at many locations throughout Hanford's history; the Hanford Site Description contains details (ORAUT 2010c). Table 5-26 lists buildings where there was a potential for exposure to thorium. Although a thorium urinalysis procedure was in place at least as far back as 1955 (Nickelson et al. 1955, pp. 6d, 7d), there are no thorium urinalysis results in the database until 1979. Whole-body counting specific for ²³²Th first shows up in 1962 (one worker) and 1963 (two workers) with tens of counts per year in the later 1960s and a few counts in the 1970s.

Building, areaDate313, 3001945–1970314, 3001945–1970305,3001945–1946306, 3001955–19703706, 3001954–19633707 A&B, 3001945–1970
314, 300 1945–1970 305,300 1945–1946 306, 300 1955–1970 3706, 300 1954–1963 3707 A&B, 300 1945–1970
305,300 1945–1946 306,300 1955–1970 3706,300 1954–1963 3707 A&B,300 1945–1970
306, 300 1955–1970 3706, 300 1954–1963 3707 A&B, 300 1945–1970
3706, 300 1954–1963 3707 A&B, 300 1945–1970
3707 A&B, 300 1945–1970
,
0700 000 4005 4070
3732, 300 1965–1970
3722, 300 1946–1970
325, 300 1963 ^a
321, 300 1964-1970
202 A, PUREX, 200 East 1965–1970
241-WR vault, 200 West 1965–1978
224-U, 200 West 1966-1967
a. Hot cell work only. Worker exposure
unlikely

Table 5-26. Buildings with thorium processing (Gerber 1992: Isochem 1967: Walser 1978).

Radiation protection practices were similar for thorium and uranium, so dose reconstructors should assume a chronic intake. Because metal, oxide, nitrate, and perhaps other forms of thorium were being handled, either absorption type M or S can be assumed. Radium-228, the first progeny of ²³²Th, would have been removed either during the processing of the ore at Fernald or during baking and sintering in the 300 Area, so the progeny would not have been in equilibrium (West 1965). Workers could have been exposed to thorium in a mixture of ages (i.e., time since purification), but most of the material would have been fairly young. It is favorable to claimants to assume younger material when whole-body counting is the bioassay method (explained below); therefore, the thorium was assumed to have aged 0.5 year since purification. Therefore, for an intake of ²³²Th by activity, dose reconstructors should assign ²²⁸Ra at 0.058 times the ²³²Th intake and assign ²²⁸Th at 0.84 times the ²³²Th intake [75]. The ²³⁰Th specification for the thorium oxide was <1 ppm (Author unknown 1968), which means that ²³⁰Th contamination would have contributed <6% of the total alpha activity and can be ignored.

Routine monitoring for thorium intakes was by whole-body counting based on ²²⁸Ac. Note: Use of whole-body counting for thorium monitoring was apparently different than at other AEC sites that used chest counting. Be sure to recognize that the following discussion and Tables 5-26 to 5-28 apply only to whole-body counts. Urinalyses were used for special investigations. Jech (1967) stated the detection limit for ²³²Th was 1.2 nCi. Jech (1969b) summarized the results of a worker's whole-body count and stated, "...and no detectable thorium (<1 nCi)". The 1.2-nCi detection limit was probably a decision level, so the MDA would have been about 2.4 nCi [76]. Sometimes the whole-body count record shows ²³²Th in the list of radionuclides if the worker was specially earmarked for possible exposure to ²³²Th, but the dose reconstructor should not count on this for every exposed worker. If nothing was detected, no activity is shown. For a few of those workers no record of the whole-body count was found in the database, but a paper record of the count was submitted as part of the DOE files. Although the *in vivo* records show the results as ²³²Th, the measured activity was ²²⁸Ac [77]. The two radionuclides would not have been in equilibrium; both disequilibrium at intake and separate biological processes on the ²³²Th parent and the ²²⁸Ra progeny mean that the ²³²Th activity in the body would not have been the same as the measured ²²⁸Ac. Assuming chronic intake and exposure to 0.5-yr aged mixture, the factors in Table 5-27 should be used to determine the intake of ²³²Th per nanocurie of ²²⁸Ac (reported as ²³²Th) that was measured in a whole-body count. Once the adjusted intake of ²³²Th has been determined, the ratios in the paragraph above must be used to determine the intakes of ²²⁸Ra and ²²⁸Th.

Period of chronic intake before whole-		o Ac-228 in whole of measurement	(reported as Th-232)	er 1 nCi of Ac-228 measured in whole- nt (pCi/d)
body count (d)	Туре М	Type S	Туре М	Type S
182	23.17	11.33	1,810	1,310
364	26.56	9.218	1,150	648
546	29.03	7.834	874	412
728	30.40	6.898	704	297
910	30.73	6.234	580	232
1,092	30.28	5.743	483	191
1,274	29.34	5.370	405	162
1,463	28.09	5.068	341	142
1,645	26.79	4.837	291	127
1,827	25.49	4.649	251	115
2,009	24.24	4.494	218	106
2,184	23.10	4.369	192	98.6

Table 5-27. Thorium-232 intakes from whole-body count measurement	s [78].
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The dose reconstructor can use the whole-body count record with the urinalysis records, if there are any, to determine intakes of ²³²Th. If a person was potentially exposed to thorium but was not monitored for thorium, the next paragraph provides a way to estimate intake from later whole-body counts.

Except for small-scope, limited-time projects that might have occurred occasionally (such as shipping of stored thorium from the PUREX campaigns), exposure to thorium at Hanford stopped in 1970. Nonetheless, later whole-body counts might still be used as a check on the intakes that are estimated for exposure before 1970. Thorium-232 was not routinely reported in whole-body counts, but a prominent ²²⁸Ac peak would have been noticed and investigated. Tables 5-28 and 5-29 provide the ratios of ²³²Th to ²²⁸Ac in the whole body at times after cessation of chronic exposure. The MDA for ²²⁸Ac or ²³²Th was not listed in program documentation after the 1960s Jech letters (1967, 1969a) until 1979 when a few counts for ²³²Th are listed with a detection limit of 0.5 nCi (based on a query of the REX database). This was probably a decision level. Twenty-one ²³²Th counts are recorded in the 1980s with count-specific detection limits that vary from 0.19 to 0.93 nCi, with a median of 0.43 nCi (based on a query of the REX database). However, there was a difference between the detection

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level for a radionuclide specifically targeted for identification by the software and the trigger level for flagging a peak that was not specifically earmarked by the software. Stated another way, the criterion to identify and flag the ²²⁸Ac peak would have been higher than just the decision level. With that in mind, it is reasonable to assume that from 1970 through July 1989, an ²²⁸Ac body burden of 2 nCi would have been noticed and investigated but that burdens less than that might have been overlooked [79].

Years after end	Period of chronic intake (yr)/Ratio					
of chronic intake	1	2	3	4	5	6
0.5	34.1	34.0	31.0	27.5	24.4	21.8
1	36.3	32.8	28.7	25.2	22.4	20.1
1.5	33.9	29.5	25.7	22.7	20.3	18.4
2	29.9	25.9	22.8	20.4	18.4	16.9
2.5	26.1	22.8	20.3	18.4	16.8	15.5
3	22.8	20.3		16.8	15.5	14.4
4	18.3	16.6	15.4	14.3	13.4	12.6
5	15.3	14.2	13.3	12.5	11.9	11.3
6	13.2	14.5	11.8	11.3	10.8	10.4
7	11.8	11.2	10.7	10.3	9.91	9.58
8	10.7	10.2	9.87	9.53	9.22	8.96
9	9.84	9.50	9.19	8.92	8.67	8.44
10	9.17	8.89	8.64	8.41	8.20	8.01
11	8.63	8.39	8.18	7.98	7.80	7.64
12	8.17	7.97	7.79	7.62	7.46	7.31
13	7.78	7.61	7.45	7.30	7.16	7.03
14	7.44	7.29	7.15	7.02	6.89	6.77
15	7.14	7.01	6.88	6.77	6.65	6.54
16	6.88	6.76	6.64	6.54	6.43	6.33
17	6.64	6.53	6.43	6.33	6.23	6.14
18	6.42	6.32	6.23	6.14	6.05	5.97
19	6.23	6.13	6.05	5.96	5.88	5.81
20	6.04	5.96	5.88	5.80	5.72	5.65

Table 5-28.	Ratios of ²³² Th to ²²⁸ Ac in the whole body after end of intake
period, type	M [80].

Table 5-29. Ratios of 232 Th to 228 Ac in the whole body after end of intake period, type S [81].

Years after end of	Period of chronic intake (yr)/Ratio					
chronic intake	1	2	3	4	5	6
0.5	6.34	5.23	4.60	4.19	3.94	3.76
1	5.00	4.36	3.96	3.70	3.54	3.41
1.5	4.23	3.82	3.55	3.38	3.30	3.18
2	3.73	3.45	3.26	3.15	3.06	3.00
2.5	3.39	3.19	3.07	2.98	2.92	2.88
3	3.15	3.01	2.92	2.86	2.82	2.80
4	2.86	2.79	2.75	2.72	2.71	2.70
5	2.72	2.68	2.67	2.66	2.66	2.66
6	2.65	2.64	2.64	2.64	2.64	2.65
7	2.62	2.63	2.63	2.64	2.65	2.66
8	2.63	2.64	2.65	2.66	2.67	2.68
9	2.64	2.65	2.67	2.68	2.69	2.71
10	2.66	2.68	2.69	2.71	2.72	2.73
11	2.69	2.71	2.72	2.74	2.75	2.77
12	2.72	2.74	2.75	2.77	2.79	2.80
13	2.75	2.77	2.79	2.80	2.82	2.83

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Years after end of	Period of chronic intake (yr)/Ratio					
chronic intake	1	2	3	4	5	6
14	2.79	2.80	2.82	2.83	2.85	2.87
15	2.82	2.83	2.84	2.87	2.90	2.91
16	2.86	2.87	2.89	2.91	2.93	2.94
17	2.89	2.91	2.93	2.94	2.96	2.98
18	2.93	2.95	2.96	2.98	3.00	3.01
19	2.96	2.99	3.00	3.02	3.04	3.05
20	3.00	3.02	3.04	3.06	3.07	3.09

5.3.5 Head Counters and Other Counts

Miscellaneous counts have been performed over the years at Hanford, including wound counts, head counts, liver counts, lymph node counts, and various longitudinal scans with collimated detectors to pinpoint the location of external or internal contamination. Results of these in the database are almost always listed as special counts associated with known intakes [83].

Since at least 1978, for intakes of plutonium or americium, head counts have been used to correct chest counts for activity in the bones of the chest region (Palmer and Rieksts1979). Since the mid-1990s, liver counts have been added to the protocol for correcting chest counts to account for possible shine from the liver [84].

Routine head counting for ⁹⁰Sr or ¹⁴⁷Pm did occur for a while in the 1970s. These were not very sensitive and there is the question about what a head count means in relation to the activity in the total skeleton. The same worker will hopefully also have ⁹⁰Sr urinalysis results. The latter should be given preference for confirming or quantifying an intake.

5.3.6 General Notes About Items in the Database

All *in vivo* results appear to be given in nCi [85]. "Limits" were MDAs, which were treated the same as decision levels until 1992. The decision level is listed under "limits" starting in January 1992 [86].

Sometimes a radionuclide is listed without a value or limit. This probably means a detection of what was considered a "trace" amount. More information might be available on the In Vivo Counter Results Form if it was sent to the worker's personal radiation exposure history file. If not, dose reconstructors should assume the result of the count is 100 nCi (Glenn 1968).

Before the advent of GeLi detectors, when a significant peak in a whole-body count of a radionuclide other than ²⁴Na, ¹³⁷Cs, ⁴⁰K, or ⁶⁵Zn occurred, the activity of the trace or "interfering" radionuclide might have been quantified. In addition, the activity of one or more of the regular four radionuclides might have been marked as invalid because of overlap with the interfering peak or because of the impact of the interfering peak on the spectrum stripping calculations [87]. For the small activities that are involved, there is no merit in trying to recalculate or estimate actual quantities. It is favorable to claimants to use the activities of ¹³⁷Cs as given and to include the activity of the interfering radionuclide as given as well. A value of 100 nCi for the interfering radionuclide is appropriate if it is not given directly (Glenn 1968).

The radionuclides that have been routinely reported to the database have changed over the years. From the beginning until 1983, ²⁴Na, ⁴⁰K, ¹³⁷Cs, and ⁶⁵Zn were the only routinely reported radionuclides. In 1983, as part of the switch to the ORE database, only ⁴⁰K and ¹³⁷Cs results (or the MDAs) were routinely reported, and in late 1987, ⁶⁰Co was added [88]. In 1995, with the implementation of a new spectrum analysis software program (NEXEC), the standup counter's energy spectrum was divided into 12 regions and a radionuclide was assigned to each region, including more naturally occurring radionuclides such as ²¹⁴Bi and ²⁰⁸TI [89]. During this time, if a worker had a count

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from the coaxial HPGe whole-body counter, as many as 20 radionuclides might have been listed in the records. The listing of that many radionuclides was simply a bookkeeping approach, and had nothing to do with the sources of exposure [90]. Because of the shutdown of the last reactor in 1986, radionuclides such as ⁵⁹Fe, ²⁴Na, ²²Na, ¹⁴⁴Ce/Pr, and ¹³¹I had decayed away to negligible levels at Hanford (unless a researcher was using a small source for studies). The lack of the need to report all these radionuclides routinely, unless a peak was actually present, was recognized, and when NEXEC was replaced by ABACOS (October 1999), the routinely reported list was reduced to ⁴⁰K, ⁶⁰Co, ¹³⁷Cs, and ¹⁵⁴Eu (Bihl 1998) [91]. <u>Reporting of radionuclides at levels below the MDA or decision level should not be interpreted as implying exposure to those radionuclides.</u>

For chest counting, the database usually lists ²³⁴Th as the potentially measured radionuclide as an indicator of ²³⁸U. Until recently, routinely reported radionuclides for anyone who received a chest count were ²⁴¹Am, ²³⁴Th, and ²³⁵U. This does not imply exposure to both plutonium and americium mixtures and uranium. Very recently, workers have been scheduled for types of chest counts based on their exposure in the workplace, so for plutonium workers for instance only ²⁴¹Am results are determined and reported.

5.4 MIXTURES

Except in a few facilities in the weapons production cycle (such as B Plant or the Waste Encapsulation and Storage Facility after 1968, and the UO₃ Plant), bioassay methods did not measure all the radionuclides in the intake mixture. The emphasis was on measuring exposure to radionuclides with the greatest impact in relation to radiation protection standards (for instance, MPBB or committed effective dose equivalent), or on radionuclides that were most common. Unmeasured radionuclides generally do not have a large impact on dose but might target different organs or might have a larger relative impact over periods of less than 50 years. Therefore, this section attempts to estimate possible mixtures of radionuclides that might have been part of an intake that was indicated by a measured radionuclide. In cases where actual bioassay data are available, dose reconstructors should use those data in preference to the following conservative mixtures.

Plutonium isotopic mixtures and uranium isotopic mixtures are discussed in Sections 5.2.1 and 5.2.5, respectively.

5.4.1 Fission and Activation Products

As presented in Section 5.1.2, an SEC was designated in which NIOSH found that it was infeasible to reconstruct doses for fission products before August 31, 1946 due to inadequate monitoring (72 FR 55214). Therefore, dose reconstructions will not include any doses from fission products before August 31, 1946.

For fission and activation product mixtures through 1988 (1 year after N Reactor shut down), dose reconstructors should use the intake mixture ratios in the latest revision of ORAUT-OTIB-0054, *Fission and Activation Product Assignment for Internal Dose-Related Gross Beta and Gross Gamma Analyses* (ORAUT 2007a or latest revision). Therefore, if an intake of ⁹⁰Sr, ¹³⁷Cs, or ¹⁰⁶Ru was assigned to a worker, the other fission and activation products should be assigned as discussed in that document. ORAUT-OTIB-0054 includes guidance on intake mixtures that applies to workers in different types of work or facilities.

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The fission product urinalysis procedure measured beta activity from any radionuclides of strontium, yttrium, barium, lanthanum, cerium, europium, and promethium. It was calibrated for the ⁹⁰Sr/⁹⁰Y betas so would have underestimated soft beta emitters, but chemical yield was several times higher for cerium and yttrium than for strontium so the recorded result overestimated cerium and yttrium

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activity (Healy 1948; Wilson 1987). Urine results designated as FISPR or FP in the workers' records indicate participation in the fission product bioassay program.

At Hanford some fission products had greater prominence than indicated in ORAUT (2007a). These were ⁶⁵Zn for essentially all workers because of its presence in the Richland drinking water through about 1972, ²⁴Na for workers at the once-through-cooled production reactors through about 1971 (see Section 5.3.1), ¹⁵⁴Eu for workers at N Reactor because of the samarium-ball emergency shutdown system, and ²⁴Na for 400 Area workers at the Fast Flux Test Facility (FFTF) for 1980 through 1994 [92]. From 1960 to the present, these radionuclides would have been measured by whole-body counts. If their activity is not included in the whole-body count record, dose reconstructors should use the standard missed dose procedure for assigning these intakes to workers at the applicable facilities using MDAs or reporting levels in Table 5-23. The N Reactor and FFTF intakes should not be applied unless it is certain the worker had exposure at these facilities because, relative to the Hanford workforce, few workers were actually exposed to these radionuclides at those times. Zinc-65 and ²⁴Na were not measured as part of the fission product urinalysis, however, so during the period before whole-body counting, intakes of these radionuclides should be added using the guidance in Attachment C for the period from startup to 1959. Europium-154 is not addressed in Attachment C, and MDAs are not available in Table 5-23 for all years. When they are not listed in Table 5-23, dose reconstructors should assume the MDA is 3 times the MDA for ⁶⁰Co for 1964 through 1984 [93].

ORAUT (2007a) provides guidance on fission and activation product ratios for waste management workers. However, characterization of the contents of the high-level waste tanks shows that plutonium and americium are also present in ratios relative to fission products such that they might increase the dose to certain organs. Americium is usually associated with ⁹⁰Sr in the supernatant and is generally more available as a contaminant. Plutonium is usually associated with ¹³⁷Cs in the sludge (Carbaugh 1995; Boothe 1992). For workers in waste management facilities or 200 Area workers in general if the work location is not known, and if an intake of fission products is determined and the worker does not have bioassay for plutonium or americium after the intake date, dose reconstructors should assume an intake ratio of 0.15 for either ²³⁹Pu (type M, S, or Super S) or ²⁴¹Am (type M or S) (dependent on which is more favorable to the claimant) relative to the highest fission product intake. The ratio is based on the 95th percentile of alpha to beta/gamma ratios currently in the high-level waste tanks. Current conditions have been shown to generally have higher ratios than past conditions [94].

Tritium and ¹³¹I intakes might have occurred in reactor workers and fuel dissolution plant workers. Dose from tritium was accounted for as part of external dose records for 1949 through 1987. Section 5.6 contains guidance on tritium and ¹³¹I intakes for other years or if there is no evidence of monitoring for tritium in the external dose record for reactor or fuel dissolution plant workers.

5.5 **INTERFERENCES AND UNCERTAINTIES**

5.5.1 <u>Contamination of Samples</u>

Home collection of excreta samples started very early in the bioassay program (Wilson 1987); therefore, contamination of excreta samples can be assumed to be negligible. Laboratory contamination and mix-up of samples in the laboratory are a possibility, although laboratory quality control procedures and performance of test samples were designed to minimize this source of contamination.

It is likely that a contaminated sample shows as an obvious outlier in the dataset for a given worker.

For *in vivo* measurements, contamination can occur as external to the body or, in the case of chest counting, as external to the lung. If a follow-up *in vivo* count on the same day or within a few days

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shows a dramatic decrease in activity or no detectable activity, dose reconstructors should assume external contamination. Radon progeny and medical diagnostic or therapeutic procedures that involve radionuclides can cause interferences to *in vivo* measurements, especially for NaI detectors. However, unless the count was invalidated or noted as being influenced by such interferences, the results should be used as recorded.

5.5.2 <u>Uncertainties</u>

Uncertainties for the bioassay measurements were included in the database starting in late 1981 for excreta measurements [95]. These are listed in the database under Error and represent the TPU (1 sigma) including counting uncertainty, yield uncertainty, and various other systematic uncertainties (UST 1987). Dose reconstructors should use these when available. For excreta, uncertainty can also exist in the sample date. For routine samples, dose reconstructors should assume an uncertainty of ± 2 weeks. This is because one sample date is used for the month regardless of when the sample was actually obtained. For special samples an uncertainty of ± 2 days is reasonable unless the sample is within 2 to 3 days of a known intake.

The period the sample represents is also a source of uncertainty. Most urine samples at Hanford were 24-hour simulated samples (kit code 1), which means the sample was collected over two evening-through-morning periods. Medley, Kathren, and Miller (1994) indicated that this sampling method produced only about half of a true 24-hour sample based on volume for a group of nine workers over a 3-day period. However, Hanford collection protocol was based on percent of day, not volume, so the true bias (when samples were collected according to procedure) was about 75% of a true 24-hour sample [96]. If a worker has enough urine samples to establish the individual-specific excretion pattern, a sample can be normalized to the individual's expected 24-hour excretion. For in vivo results, uncertainties were not reported until 1986 for detected radionuclides and 1993 for the default set of radionuclides. These were 1-sigma counting errors until 1995 [97]. The TPU has been determined and submitted to the records since then. The TPU includes counting uncertainty, calibration uncertainty, and a generic 5% positioning error (for both whole body and lung). The calibration uncertainty includes the uncertainty in source activity, counting error, decay correction, and interpolation using the calibration curve [98]. Uncertainty in reproducibly positioning a person to get the same result was studied at Hanford and found to be about 5% [99]. All calibrations use phantoms, and there is considerable uncertainty in the representativeness of phantoms versus humans. A recent study for whole-body counting at Hanford used a 95th-percentile reference man phantom. There was a low bias of about 25% for the coaxial HPGe detector system for 662- and 1,332-keV gamma rays (Lynch 2007). A similar value of uncertainty (±25%) can reasonably be assumed for the other whole-body detectors (1-m arc, shadow shield, and standup counters) [100].

Uncertainties in chest counting are reduced by use of different calibrations for different chest wall thicknesses and use of ultrasound to measure chest wall thickness. One study showed a 1-sigma uncertainty of about 20% for americium and uranium values in chest counting, not including correction for interferences from bone and liver (PNNL 1990, Table 7-4). Uncertainties would be much higher for an individual with activity in the bone or liver. The uncertainty in lung activity estimates that are affected by contributions from activity in the liver and skeleton would probably range from 100% or more for levels near or below the MDA to 50% or more for activity above the MDA [101]. The uncertainty in the estimate of chest thickness using the height-to-weight correction ratio was at least 50% for the front to back lung counter.

5.6 UNMONITORED WORKERS

For unmonitored workers with external dosimetry, dose reconstructors should use the intakes from the Attachment C coworker analysis in combination with the special instructions in this section. When assigning intakes of fission products, intakes of associated radionuclides as discussed in ORAUT-

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OTIB-0054 (ORAUT 2007a or latest revision) should be included. Short-lived radionuclides, such as ¹⁴⁴Ce, ¹⁰⁶Ru, ⁹⁵Zr, ⁹⁵Nb, ⁵⁴Mn, ⁶⁵Zn, ¹⁴⁰Ba, ¹⁴⁰La, ⁵¹Cr, ⁵⁹Fe, and ^{110m}Ag, should not be assigned to workers after December 31, 1988 [102]. The exception is for ²⁴Na for workers at the FFTF, which operated intermittently from 1980 to 1994. Special consideration is also provided for decontamination and decommissioning (D&D) workers. Section 5.6.2 provides facility-specific details for assignment of intakes/doses for unmonitored workers.

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Under certain conditions, airborne effluents from one facility became air intakes for other facilities. In addition, workers were exposed to diluted effluents when walking between buildings or parking lots or while driving on the site. Therefore, workers in buildings who did not enter contaminated or airborne areas and construction workers almost anywhere could have incurred environmental intakes.

Before 1994, workers with even a remote chance of exposure to workplace external radiation or workplace airborne contamination (rather than exposure from effluents) wore a dosimeter. Through 1971, workers obtained film dosimeters before proceeding past badge houses at the various facilities. From 1972 to 1994, even workers with only a small potential for incurring external dose wore a minimal dosimeter that was called the Hanford basic dosimeter (ORAUT 2010b).

The unmonitored internal dose for workers with no bioassay (or just baseline or termination bioassay) and no evidence of ever having worn a dosimeter should be based only on environmental intakes [103]. Termination whole-body counts were often provided to any worker regardless of exposure risk and should not be considered evidence of being in a bioassay program.

The coworker intakes in Attachment C do not address radioiodines, tritium, or ²³³U. The rationale for intake values for these radionuclides is presented in Section 5.6.1, and the assignments for dose reconstruction based on Section 5.6.1 and Attachment C for unmonitored workers are presented in Section 5.6.2.

5.6.1 Radionuclides Not Presented in Attachment C

Guidance for assignment of intakes of radioiodines, tritium, ²³³U, HEU, ²¹⁰Po, ²⁴¹Am, ²⁴⁴Cm, ²³⁷Np, and ⁹⁹Tc by unmonitored workers is provided in this section.

Radioiodine

A considerable number of thyroid scans (hundreds per month) were routinely performed on workers in the fuel dissolution (canyon) buildings starting in 1945 and continuing into the 1950s (Parker 1948, p. 6; Parker 1950b, p. 6; Parker 1951, p. 6; Parker 1952, p. 8). A review of the thyroid monitoring program was conducted for the Hanford Environmental Dose Reconstruction Project (Ikenberry 1991). The tolerance level for ¹³¹I in air had been established in October 1945 as $1 \times 10^{-7} \,\mu$ Ci/cm³ (Cantril 1945) based on a permissible equilibrium amount in the thyroid of 2 μ Ci. The routine scanning program began in late May or early June 1945. Ikenberry reviewed over 7,900 thyroid checks from October 1944 through August 1946. The decision level for the measurement was estimated to correspond to a thyroid burden of about 27 nCi, and 93% of the measurements were below this activity. The highest measurement that was considered to be reasonably due to a thyroid burden (rather than to external contamination) was 131 nCi.

Since the beginning of routine whole-body counting, detections of ¹³¹I have been rare. For instance, from 1962 through 1969 (the peak production years), only about 130 of about 14,500 workers who had whole-body and thyroid counts had detectable ¹³¹I. The results ranged from 0.001 to 560 nCi, and the highest was a same-day measurement after an accidental exposure in the T Plant canyon. Ninety percent of the detected results were less than 15 nCi [104]. From these two datasets, the assumption was made that was favorable to claimants that at-risk workers had thyroid burdens less

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than 30 nCi, which would result from a daily intake of 20,000 pCi [105]. This intake would apply to all at-risk workers before 1960. From 1960 to 1987, it is reasonable to assume that an unmonitored worker (i.e., no whole-body count) did not incur intakes greater than the thousands of workers who had whole-body counts that showed no detection. Using a nominal detection limit for whole-body counts of 5 nCi (rather than more sensitive thyroid counts), unmonitored workers that are associated with reactors or fuel dissolution plants should be assigned ¹³¹I intakes of 3,400 pCi/d [106]. No intakes of ¹³¹I should be assigned after 1987 because there would have been no significant source term unless the worker's personal information indicates a special exposure, such as for a research project [107].

Tritium

Tritium bioassay results were not included in the database until 1984 [108]. Tritium doses were included in external dose reports from 1949 through 1987, so monitored workers would be determined by tritium dose in external dose records. A zero dose is indicative of a monitored worker, but there are only two of these in the database. No value listed at all for tritium might be indicative of either an unmonitored worker or a monitored worker with a less-than-recordable dose. No tritium doses are listed on external dose reports for 1955 through 1960, 1970, 1971, and 1973 through 1984, so either there was no bioassay monitoring during these periods or the intakes were below some reporting criteria [109]. The 108-B Tritium Extraction Facility shut down in 1955, which probably explains the lack of tritium doses, and probably a lack of tritium monitoring, until the heavy-water-moderated Plutonium Recycle Test Reactor started up in 1960. If a worker's file does not show tritium dose in the external dose records (where a zero is considered a monitored dose), and if the worker might have had exposure to tritium as discussed below, then the unmonitored tritium intakes discussed below and provided in Table 5-30 should be applied.

Year	GM (mrem)	GSD	GM doses normalized to present model (mrem)	Daily intakes resulting in the normalized doses (× 10 ⁶ pCi/d) ^a
1949	29.9	2.9 (3.0) ^b	48.4	2.0
1950	43.6	2.8 (3.0)	70.6	2.9
1951	64.2	3.5	104	4.2
1952	67.1	4.0	109	4.4
1953	40.8	3.7	38.5	1.6
1954	104	3.0	98.2	4.0
1955	40.8	3.7	38.5	1.6
1956–1960	Insufficie	nt data		
1961	62.4	1.9 (3.0)	58.9	2.4
1962	193	3.0	182	7.4
1963	185	3.8	175	7.1
1964	141	4.2	133	5.4
1965	134	3.8	127	5.1
1966	108	3.9	102	4.1
1967	185	4.7	175	7.1
1968	98.6	4.9	93.1	3.8
1969	26.5	2.3 (3.0)	25.0	1.0
1970–1971	Insufficie	nt data		
1972	58.9	2.3 (3.0)	55.6	2.3

Table 5-30. Statistical parameters of Hanford recorded tritium doses and associated intakes [112].

a. Use as injection mode in IMBA or Chronic Annual Dose Workbook.

b. A default GSD of 3.0 should be substituted for a calculated GSD of less than 3.

Before 1949, it is assumed that tritium intakes were limited by the tolerance level. The earliest tritium tolerance limit that has been found to date is an air concentration limit of 0.01 μ Ci/L in 1947 (Parker 1947; Patterson 1949) (see Attachment A for discussion of tolerance levels). The assumption of a

chronic intake at this concentration of 1.2 m^3 /hr for 2,000 hr/yr results in an intake rate of $6.6 \times 10^7 \text{ pCi/d}$. This produces annual organ doses of about 1,500 mrem [110], which is larger than all but a few recorded doses from 1949 through the 1960s. Therefore, this intake rate is favorable to claimants for the highest risk workers, namely those at the 108-B Tritium Extraction Facility. Tritium would have been present at the fuel dissolution plants as well and to a lesser extent at the reactors, but certainly the tritium in the latter facilities would not have been as concentrated as at the 108-B facility. It is assumed that tritium intakes at the reactors and fuel dissolution facilities would have been at most 10% of the intakes at 108-B [111]. Therefore, daily intakes of 7×10^6 pCi/d should be assigned to unmonitored workers at the reactors, fuel dissolution plants, or rovers.

To determine tritium intakes for 1949 through 1972, a coworker analysis was conducted on tritium dose data for that period. The statistical results are biased high because, with two exceptions, doses from tritium of <10 mrem were not recorded; consequently, it is not known how many workers were monitored who had doses less than 10 mrem. The annual geometric means (GMs) and geometric standard deviations (GSDs) are shown in Table 5-30.

The methodology for calculating tritium dose from urine concentration has changed over time. The method introduced in ICRP Publications 2 and 10 (ICRP 1959, 1968) resulted in dose per unit activity in urine slightly higher than the method used in the present. How the tritium doses were calculated before 1959 has not been found yet, but Parker, the head manager of radiation protection at Hanford in the 1940s and 1950s referenced a method by Morgan in 1946 that shows that 40,000 µCi in the body produces the tolerance dose for the time (Morgan 1947). The tolerance dose in the 1940s was 100 mrep/d. Under the assumption that the tritium is distributed in 42 L of body water at equilibrium, 952 µCi/L [40,000 µCi/42L] produced 100 mrem/d. Stated another way, a dose of 0.105 mrem/d would have been calculated from 1 µCi/L in urine. The 1953 body burden of tritium to produce a maximum permissible dose of 300 mrem/wk was 10,000 µCi (NBS 1953). Therefore, 238 µCi/L [10,000 µCi/42L] would have produced 42 mrem/d, or 0.18 mrem/d would have been calculated from 1 µCi/L in urine. In the present model (ICRP 1998), 1.2 × 10⁶ Bg/L results in 20 mSv/yr or a 0.17 mrem/d dose is calculated from 1 µCi/L in urine. Therefore, the recorded doses for 1953 through 1972 are slightly higher per unit activity in urine than would be calculated under the present model. Assuming Hanford used the Morgan model through 1952, those recorded doses were increased by 0.17/0.105 to be consistent with the present model.

After the above calculations were made, hundreds of tritium bioassay records were found in archived records and analyzed. The results are discussed in Attachment C. The analysis of actual tritium bioassay records showed that the above recommendations based on tritium *dose* records were favorable to claimants.

Because the doses in Table 5-30 were based on only the nonzero recorded doses, they were judged to be most applicable to unmonitored workers at the 108-B facility through 1955 and the Plutonium Recycle Test Reactor from 1960 through 1969. The 108-B Tritium Extraction Facility shut down in 1955, and some tritium bioassay has been found for that year. Those records show that the 1953 default doses and intakes are favorable to claimants if they are also used for 1955. It is assumed that tritium doses at the reactors and fuel dissolution facilities would have been at most 10% of the doses in Table 5-30 [113]. The GSDs in Table 5-30 should also be used with a minimum GSD of 3.

There was insufficient dose data in two periods, 1956 to 1960 and 1970 to 1971, for statistical analysis. Disregarding improvements in radiation protection regulations and practices, tritium exposure at the reactors and fuel dissolution plants could have been correlated with production. Using tons of uranium times fuel burnup as a figure of merit for production (see Figure C-1 in Attachment C), average yearly production in 1955 through 1960 was 4 times greater than for 1949 through 1954. The average of the median daily intakes for unmonitored reactor and fuel dissolution plant workers during 1949 through 1954 was 3.2×10^5 pCi/d (one-tenth of the average of the intakes

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in the fifth column in Table 5-30). Four times this intake, 1.3×10^6 pCi/d, should be used for unmonitored reactor and fuel dissolution plant workers during 1956 through 1960. The largest GSD of 4 can be applied to the 1956 through 1960 intakes. Production was essentially equal during 1970 and 1971 in comparison to 1949 through 1954, so the 3.2×10^5 pCi/d intake should be applied to unmonitored reactor and fuel dissolution plant workers during 1970 and 1971 (GSD = 4).

No fuel dissolution occurred from 1973 through 1983 and only one reactor was operating. The 3.2×10^5 pCi/d intake can also be applied to unmonitored reactor workers during this period. A small set of urinalyses were collected from reactor workers in 1982 to 1983. The set was too small for a complete statistical analysis, but the median result was about 4 pCi/mL. This results from daily intakes of about 1.2×10^4 pCi/d [114] and indicates that the 3.2×10^5 pCi/d intake rate is favorable to claimants.

Tritium bioassay results are available for 1984 through 1986 for 100 and 200 Area workers. A coworker analysis was performed on these data [115]. The 50th-percentile urinalysis results were 2.6, 0.37, and 0.21 pCi/mL for 1984, 1985, and 1986, with GSDs of 13, 3.3, and 6.2, respectively. The associated daily intakes were 7,600, 1,080, and 610 pCi/d, respectively [116]. N Reactor shut down in 1987; PUREX ran intermittently in the late 1980s and was shut down in 1992 (see Section 2.2.3 of the Hanford Site Description; ORAUT 2010c). The 1986 value can be applied to workers at those facilities through their respective shutdown dates [117].

By 1992 there was little exposure to tritium at Hanford except for special projects (e.g., tritium target program), low-level tracers, or in 400 Area drinking water as discussed below.

Drinking water for workers in the Hanford 400 Area, of which the principal facility was the FFTF, was obtained from an unconfined aquifer that had low-level tritium contamination. The FFTF was started in 1980. The exact date well water was provided for drinking at the site has not been discovered, but it is reasonable to assume that it predated the startup of FFTF by a year or so. An analysis of the tritium concentrations in the wells that provide the drinking water and estimation of the tritium intakes from that source was performed (Bihl 2005). Some 400 Area workers were on routine tritium bioassay, and dose reconstructors should use those data when available. For other 400 Area workers, calendar day ingestion intakes of 48,000 pCi/d for 1978 through 1985 and 14,000 pCi/d for 1986 through present should be assigned (Bihl 2005). However, the annual doses from the post-1985 intakes are less than 1 mrem and can be ignored [118].

Unmonitored intakes for certain radionuclides are determined differently. Generally these were radionuclides not related to the primary radionuclides or mixtures; i.e., plutonium, uranium, tritium, and mixed fission and activation products. Bioassay monitoring for the secondary radionuclides at certain times was rare or nonexistent. Most of the exposure to these other radionuclides is covered under the SEC.

In general workplace monitoring and action levels, such as for air concentrations, were based on total alpha or total beta measurements with action levels set for the most restrictive radionuclide of the list of radionuclides likely to be encountered. For instance, the total alpha measurements were usually compared to the maximum permissible concentration for ²³⁹Pu and the total beta measurements were compared to the maximum permissible concentration for ⁹⁰Sr. An exception was for air samples collected at facilities where uranium was the predominant radioelement. Containments and other barriers (e.g., gloveboxes, hot cells) were also similar for radionuclides emitting similar radiations. Protocols for establishing of contamination zones and performing workplace and person surveys and other radiation protection practices were the same for the secondary radionuclides as for the primary radionuclides. Therefore, it is reasonable to apply the plutonium coworker intakes (Attachment C) to unmonitored intakes of ²⁴¹Am, curium, ²¹⁰Po, and ²³⁷Np. Thorium intakes should be based on coworker intakes for uranium. However, these guidelines do not apply to all times. Additionally, there

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are situations when the 95th percentile of the coworker distribution and a constant distribution are more appropriate than the 50th percentile and lognormal GSDs. The 95th percentile is used when routine bioassay was not available or not used. The 95th percentile intake can be found using

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

Specifics are given for each situation below.

Uranium-233

Operators, radiation monitors, and equipment maintenance workers at PUREX who were involved in the ²³³U campaigns might have had intakes without specific monitoring for ²³³U. The same might be true for chemists, chemical technicians, and radiation monitors associated with the Plutonium Chemistry Laboratory in Z-Plant in 1965. The highest urinalysis result (4.39 pCi/d) from a batch of 1970 PUREX worker records was used to estimate potential chronic intakes for the period from 1965 through 1970 [119]. Because the material was absorption type F, urinary excretion equilibrates quickly and the same daily intake can be applied for 1 to 5 years of chronic exposure. The daily intake that produced 4.39 pCi/d uranium in urine was 16 pCi [120]. The end of Section 5.2.5 lists contaminants that must be added to this intake. If this intake is applied to Z- Plant workers, because the material was being converted to oxide, dose reconstructors should apply absorption type F, M, or S, whichever is most favorable to the claimant.

Highly Enriched Uranium (HEU)

Hanford canned and irradiated HEU fuel elements called C and J slugs from 1949 to 1956. Numerous, small-scale, special projects with HEU, often alloyed with other metals, occurred at various locations in the 1940s, 50s, and 60s. See the Site Description for details. Exposure to HEU prior to July 1, 1972 is covered by the SEC, and intakes will not be assigned for HEU for this period for partial dose reconstructions for unmonitored workers. No work with unsealed HEU between 1972 and 1983 (when isotopic uranium urinalyses began) has been discovered. However, small-scale projects cannot be ruled out. Intakes will not be assigned for HEU for the period between July 1, 1972 and October 1983 for partial dose reconstructions for unmonitored workers. However, if there is evidence of work with HEU between July 1, 1972 and October 1983, and the worker was monitored with routine chest counts and elemental uranium urinalysis even though not specifically monitored for HEU, results should be interpreted using HEU as the source of intake and intakes and doses can be calculated.

Thorium

Thorium fuel elements or target elements were produced at Hanford at various times for various reasons since 1945. Many facilities handled thorium at one time or another. See Table 5-26 and the Site Description for details. Major production of thoria target elements occurred in the 3732 and 3722 Buildings in 1965 through 1970. Dissolution of the target elements and separation of the thorium and ²³³U occurred in three campaigns at PUREX in 1965, 1966, and 1970. After the 1970 campaign PUREX thorium was stored in the WR Vault in the 200 West Area and eventually shipped offsite (mostly to the Fernald Feed Materials Production Center) in the 1970s. Whole body counting was the principal method of routine bioassay beginning in 1964 with a few counts in 1962 and 1963. Chest counting for thorium replaced whole body counting in 1979. Several workers had thorium urinalyses in 1969 and 1970. Exposure prior to July 1, 1972 is covered by the SEC because it was determined to be infeasible to reconstruct doses. Only five whole body counts specific for ²³²Th were conducted from 1970 through 1978 and there were a few urinalyses for "total actinides" during this period, but it is possible that workers involved with shipping of the thorium from the WR Vault (primarily storage and transfer of liquid thorium nitrate solution) or involved with decontamination of the 3722 and 3732

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Buildings did not receive bioassay monitoring. If there is evidence of exposure to thorium from shipping of the material in the WR Vault or decontamination of the 3722 or 2732 Buildings without bioassay, unmonitored workers should be assigned intakes of thorium (with progeny per Section 5.3.4) using the 95th percentile of the uranium coworker intakes (constant distribution).

Polonium-210

Exposure to ²¹⁰Po prior to July 1, 1972 is covered by the SEC because it was determined to be infeasible to reconstruct doses. Bioassay monitoring was used for significantly exposed workers from 1968 on. If an intake estimate is required, unmonitored workers from July 1, 1972 on should be assigned intakes at the same values as the plutonium coworker intakes using the 50th percentile values and the GSDs from Attachment C.

Americium-241

Significant quantities of pure ²⁴¹Am or mixtures enriched in ²⁴¹Am were present in several Z-Plant facilities through 1976 and at times at a few other facilities. See the Site Description for details. Exposure prior to July 1, 1972 is covered by the SEC because it was determined to be infeasible to reconstruct doses. Bioassay monitoring via chest counts was used for significantly exposed workers from 1968 on and urinalysis was used for special monitoring after incidents with potential for intake. If an intake estimate is required from July 1, 1972 on, unmonitored workers should be assigned intakes at the same values as the plutonium coworker intakes using the 50th percentile values and the GSDs from Attachment C.

Curium-244

Curium-244 was extracted and purified from Shippingport fuel in 1966 through 1968 at the 201-C facility and the hot cells in the 325 Building. Unrelated work with curium was performed in the 308 Building from 1969 through 1973. See the Site Description for details. Exposure prior to July 1, 1972 is covered by the SEC, and intakes will not be assigned for this period for partial dose reconstructions for unmonitored workers. Incident follow-up urinalysis for curium was available and used rarely from 1968 on. Limited use of routine urinalysis was conducted starting in 1974. If an intake estimate is required, unmonitored workers should be assigned intakes at the 95th percentile of the plutonium coworker intakes (constant distribution) prior to 1974 and at the 50th percentile (with GSDs) from 1974 on.

Neptunium-237

Neptunium-237 was extracted and purified from fuel from 1958 until PUREX shutdown in 1972, with small-scale flowsheet development starting in November 1956. Neptunium target elements were fabricated, irradiated, and dissolved from 1959 through 1961 and 1966 through 1970. Many facilities were involved at different times. See the Site Description for details. Exposure prior to July 1, 1972 is covered by the SEC because it was determined to be infeasible to reconstruct doses. Excreta bioassay for neptunium was available from 1960 but was not used routinely until the 1980s. No evidence of significant exposure to neptunium after 1972 has been found, but small research projects with the material can not be ruled out. If an intake estimate is required from July 1, 1972 on, unmonitored workers should be assigned intakes at the 95th percentile of the plutonium coworker intakes (constant distribution) prior to 1984 and at the 50th percentile (with GSDs) from 1984 on.

Technetium-99

Technetium-99 was extracted and purified from fuel in a few campaigns during 1962 through 1964 with later shipments of the material in 1967 and 1970. The work was mostly conducted in the hot

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cells in the 325 Building, although some or all of the purified material was subsequently stored somewhere in the 200 Area. The 1970 shipment involved a mix of previously extracted material with recently extracted material with the work being performed at REDOX. No evidence of significant exposure to ⁹⁹Tc after 1970 has been found. No evidence of bioassay for ⁹⁹Tc in the 1960s or 1970s has been found so the exposure is considered unmonitored. Exposure prior to July 1, 1972 is covered by the SEC, and intakes will not be assigned for this period for partial dose reconstructions for unmonitored workers.

5.6.2 Assignments for Unmonitored Workers

When information about what facility or type of facility an employee worked at is available, guidance on how to assign mixtures of the various radionuclides is given below. Dose reconstructors should check the Site Description for years of operation of specific facilities. It is important to note that ingestion intake of ⁶⁵Zn should be assigned to all workers from August 31, 1946 through 1972 in accordance with Attachment C. For the years 1949-55. 1961-69, and 1972, note that it is the tritium doses in Table 5-31 that are to be assigned by dose reconstructors, and if the derived daily intake values are used in a dose calculation tool, the calculated doses should be verified with the doses in Table 5-31.

		Daily intake		
Radioactive		(pCi/d)/dose-tritium		
material	Period	only (mrem/yr)	Distribution	
Pu	September 1,	See Attachment C	See Attachment C	
	1946-present			
U	Through present	See Attachment C	See Attachment C	
Sr-90	September 1, 1946–present	See Attachment C	See Attachment C	
Pm-147	See Attachment C	See Attachment C	See Attachment C	
Zn-65	September 1, 1946–present	See Attachment C	See Attachment C	
Na-24	September 1, 1946–present	See Attachment C	See Attachment C	
Cs-137	September 1, 1946–present	See Attachment C	See Attachment C	
Other	September 1,	See ORAUT-OTIB-	See ORAUT-OTIB-	
fission/activation	1946-present	0054 (ORAUT	0054 (ORAUT 2007a)	
products		2007a)		
I-131 (vapor)	1944–1959	2 E+04	Lognormal, GSD =3	
I-131 (vapor)	1960–1987	3.4 E+03	Lognormal, GSD =3	N Reactor only 1973 through 1983.
1-131 (vapor)	1988-present	0		
Tritium (HTO)	1944–1948	6.6 E+06	Lognormal, GSD = 3	Reactors, fuel dissolution, rovers
Tritium (HTO)	1949	2.0E+06/48.4	Lognormal, GSD = 3	108B
Tritium (HTO)	1950	2.9E+06/70.6	Lognormal, GSD = 3	108B
Tritium (HTO)	1951	4.2E+06/104	Lognormal, GSD = 3.5	108B
Tritium (HTO)	1952	4.4E+06/109	Lognormal, GSD = 4.0	108B
Tritium (HTO)	1953	1.6E+06/38.5	Lognormal, GSD = 3.7	108B
Tritium (HTO)	1954	4.0E+06/98.2	Lognormal, GSD = 3.0	108B
Tritium (HTO)	1955	1.6E+06/38.5	Lognormal, GSD = 3.7	108B
Tritium (HTO)	1949	2.0E+05/4.8	Lognormal, GSD =3.0	Reactors, fuel dissolution, rovers
Tritium (HTO)	1950	2.9E+05/7.1	Lognormal, GSD = 3.0	Reactors, fuel

Table 5-31. Intakes for workers with external monitoring but essentially no bioassay (pCi/d) [121].

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		Daily intake		
Radioactive		(pCi/d)/dose-tritium		
material	Period	only (mrem/yr)	Distribution	
				dissolution, rovers
Tritium (HTO)	1951	4.2E+05/10.4	Lognormal, GSD =3.5	Reactors, fuel
				dissolution, rovers
Tritium (HTO)	1952	4.4E+05/10.9	Lognormal, GSD =4.0	Reactors, fuel
	4050			dissolution, rovers
Tritium (HTO)	1953	1.6E+05/3.9	Lognormal, GSD =3.7	Reactors, fuel
	4054			dissolution, rovers
Tritium (HTO)	1954	4.0E+05/9.8	Lognormal, GSD =3.0	Reactors, fuel
Tritium (HTO)	1955	1.6E+05/3.9	Lognormal, GSD =3.7	dissolution, rovers Reactors, fuel
	1955	1.6E+05/3.9	Lognormal, GSD =3.7	dissolution, rovers
Tritium (HTO)	1956–1960	1.3 E+06	Lognormal, GSD = 4	Reactors, fuel
	1930-1900	1.5 L+00	Loghonnai, GGD – 4	dissolution, rovers
Tritium (HTO)	1961	2.4E+05/5.9	Lognormal, GSD = 3.0	Reactors (except
	1001	2.42100/0.0		PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1962	7.4E+05/18.2	Lognormal, GSD = 3.0	Reactors (except
				PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1963	7.1E+05/17.5	Lognormal, GSD = 3.8	Reactors (except
			0	PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1964	5.4E+05/13.3	Lognormal, GSD = 4.2	Reactors (except
				PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1965	5.1E+05/12.7	Lognormal, GSD = 3.8	Reactors (except
				PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1966	4.1E+05/10.2	Lognormal, GSD = 3.9	Reactors (except
				PRTR), fuel dissolution,
	4007			rovers
Tritium (HTO)	1967	7.1E+05/17.5	Lognormal, GSD = 4.7	Reactors (except PRTR), fuel dissolution,
				,
Tritium (HTO)	1968	3.8E+05/9.3	Lognormal, GSD = 4.9	rovers Reactors (except
	1900	5.82+05/9.5	Loghormal, GSD = 4.9	PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1969	1.0E+05/2.5	Lognormal, GSD = 3.0	Reactors (except
(110)	1000	1102100/210		PRTR), fuel dissolution,
				rovers
Tritium (HTO)	1960	2.4E+06/58.9	Lognormal, GSD = 3.0	PRTR
Tritium (HTO)	1961	2.4E+06/58.9	Lognormal, GSD = 3.0	PRTR
Tritium (HTO)	1962	7.4E+06/182	Lognormal, $GSD = 3.0$	PRTR
Tritium (HTO)	1963	7.1E+06/175	Lognormal, $GSD = 3.8$	PRTR
Tritium (HTO)	1964	5.4E+06/133	Lognormal, GSD = 4.2	PRTR
Tritium (HTO)	1965	5.1E+06/127	Lognormal, GSD = 3.8	PRTR
Tritium (HTO)	1966	4.1E+06/102	Lognormal, GSD = 3.9	PRTR
Tritium (HTO)	1967	7.1E+06/175	Lognormal, GSD = 4.7	PRTR
Tritium (HTO)	1968	3.8E+06/93.1	Lognormal, GSD = 4.9	PRTR
Tritium (HTO)	1969	1.0E+06/25.0	Lognormal, GSD = 3.0	PRTR
Tritium (HTO)	1970–1971	3.2 E+05	Lognormal, GSD = 4	Reactors, fuel
				dissolution
Tritium (HTO)	1972	2.3E+6/55.6	Lognormal, GSD = 3	N Reactor, PUREX
				plant

		Daily intake		
Radioactive		(pCi/d)/dose-tritium		
material	Period	only (mrem/yr)	Distribution	
Tritium (HTO)	1973–1983	3.2 E+05	Lognormal, GSD =4	N Reactor
Tritium (HTO)	1984	7.6 E+03	Lognormal, GSD = 13	N Reactor, PUREX
Tritium (HTO)	1985	1.1 E+03	Lognormal, GSD = 3.3	N Reactor, PUREX
Tritium (HTO)	1986–1987	6.1 E+02	Lognormal, GSD = 6.2	N Reactor, PUREX
Tritium (HTO)	1988–1992	6.1 E+02	Lognormal, GSD = 6.2	PUREX only
Tritium (HTO)	1978–1985	4.8 E+04	Lognormal, GSD = 3	Ingestion of drinking water in the 400 Area
U-233 (type F) + contaminants	1965, 1966, 1970	16	Constant	PUREX
U-233 (type S)	1965, 1966	Use Pu intakes from Attachment C	From Attachment C, 50th percentiles with GSDs	234-5Z
Am-241 (type M)	July 1, 1972- present	Use Pu intakes (type M) from Attachment C	50th percentiles with GSDs	
Cm-244 (type M)	July 1, 1972- 1973	Use Pu intakes (type M) from Attachment C	95th percentiles, constant distribution	
Cm-244 (type M)	1974-present	Use Pu intakes (type M) from Attachment C	50th percentiles with GSDs	
Np-237 (type M)	July 1, 1972- 1983	Use Pu intakes (type M) from Attachment C	95th percentiles, constant distribution	
Np-237 (type M)	1984-present	Use Pu intakes (type M) from Attachment C	50th percentiles with GSDs	
Po-210 (type F or M)	July 1, 1972- present	Use Pu intakes (type M) from Attachment C	50th percentiles with GSDs	
Thorium (type M or S)	July 1, 1972- present	Use U intakes (type M or S) from Attachment C	95th percentiles, constant distribution	See Section 5.3.4 for progeny intakes
Tc-99 (type F or M)	July 1, 1972- present	None	None	No known exposures during this period.
D&D radiological workers	2002-present	See discussion, last paragraph in this section.		

As presented in Section 5.1.2, an SEC was designated in which NIOSH found that it was infeasible to reconstruct doses for plutonium or fission products before August 31, 1946 due to inadequate monitoring (72 FR 55214). Therefore, dose reconstructions will not include any doses from plutonium or fission products before August 31, 1946. The first extruded uranium rods arrived in October 1943 and machining began in December.

It is important to note that as presented in Attachment C, from December 1943 through 1948, intakes for uranium should be assigned in accordance with the Process/Job Title listed as Machining/Operator as presented in Battelle-TBD-6000/PNWD-3738, *Technical Basis Document: Site Profiles for Atomic Weapons Employers That Worked Uranium and Thorium Metals* (Battelle 2006).

Attachment C intake rates exhibit a decreasing trend over time and may be assigned past 1988 when overestimates are appropriate for dose reconstructions.

100 Area Reactors

- <u>While operating, 1944 to 1987</u>: Use fission and activation products (from September 1, 1946 on) from Attachment C (except ¹⁴⁷Pm) and ORAUT-OTIB-0054 (ORAUT 2007a) and ¹³¹I and tritium intakes from Table 5-31.
- <u>When not operating, 1988 to the present</u>: Use fission and activation products from Attachment C only without ¹³¹I or tritium and without short-lived fission and activation products after 1988 for best estimates.

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300 Area Test Reactors, 1944 to 1978: Use fission and activation products (from September 1, 1946 on) from Attachment C (except ¹⁴⁷Pm) and ORAUT-OTIB-0054 (ORAUT 2007a) and tritium for PRTR from Table 5-31.

400 Area (includes 405 Building FFTF), 1980 to 1994 (1978 to 1985 for tritium): Use fission and activation products (reactors) from Attachment C (except ¹⁴⁷Pm) and ORAUT (2007a). Note that this reactor continued to be used intermittently until 1994. Use tritium ingestion from drinking water from Table 5-31.

200 Area Fuel Separation Facilities

- <u>T-Plant, 224-T, 1945 to 1956; B-Plant, 224-B, 1945 to 1952; REDOX, 224-S, 1952 to 1967;</u> <u>PUREX 1956 to 1992</u>: Use fission and activation products (from September 1, 1946 on), plutonium (from September 1, 1946 on, types M, S, or Super S), and uranium (type F from January 1, 1949 on) from Attachment C (except ¹⁴⁷Pm) and ORAUT (2007a) but may ignore ²⁴Na from Attachment C for best estimates. May use ²³³U (and contaminants) intakes for PUREX 1965-70 from Table 5-31 in lieu of plutonium and typical uranium. Use tritium and ¹³¹I intakes from Table 5-31. No short-lived fission and activation products should be applied after 1988 for best estimates.
- <u>B-Plant, 225-B, 1953 to the present</u>: Use fission and activation products from Attachment C (except ¹⁴⁷Pm) and ORAUT (2007a waste management option) but may ignore ²⁴Na for best estimates. Use ⁶⁵Zn ingestion-only intakes for 1953 to 1972.

Plutonium Finishing Plants (various Z buildings and 233-S), September 1, 1946 to present: Use plutonium intakes from Attachment C (M, S, or Super S). If it is more favorable to the claimant:

- For July 1, 1972 through 1976, intakes of ²⁴¹Am may be assigned in lieu of plutonium.
- For 234-5Z, for 1958, 1965, and 1966, intakes of ²³³U with contaminants may be assigned in lieu of plutonium.
- For 231-Z, ²⁴¹Am may be assigned in lieu of plutonium (from July 1, 1972 on) and the same for ²³³U in 1957 and 1958.

221-U, 224-U (UO₃ Plant), 1952 to 1993: Use uranium type F intakes from Attachment C.

WR Vault, 1965-1979: If evidence of involvement with storing or shipping of thorium (primarily storage and transfer of liquid thorium nitrate solution), from July 1, 1972 on, use uranium type F or M from Attachment C.

222-S Laboratory: Use fission and activation products (fuel dissolution) from Attachment C and ORAUT (2007a). Include ¹⁴⁷Pm for 1964 through 1965.

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108-B Tritium Extraction Facility, 1949 to 1956: Use tritium intakes from Table 5-31. Exposure to organically bound tritium and type F tritides were possible.

C-Plant (Hot Semiworks, Strontium Semiworks)

- <u>1949 to 1956</u>: Use fission and activation products (fuel dissolution) from Attachment C (except ¹⁴⁷Pm) and ORAUT (2007a); ²⁴Na may be ignored for best estimates. Use plutonium (M, S, Super S) and uranium (F) from Attachment C.
- <u>1962 to 1967</u>: Use fission and activation products from Attachment C and ORAUT (2007a) (fuel dissolution), ²⁴Na may be ignored for best estimates. Use ⁶⁵Zn ingestion intakes only. Include ¹⁴⁷Pm for 1964 through 1967.

300 Area Fuel Fabrication Facilities (303, 305, 306, 313, 314, 333), 1944 to 1988: Use insoluble uranium intakes from Attachment C. It is important to note that in Attachment C, from December 1943 through 1948, intakes for uranium are assigned in accordance with the Process/Job Title listed as Machining/Operator as presented in Battelle-TBD-6000/PNWD-3738, *Technical Basis Document: Site Profiles for Atomic Weapons Employers That Worked Uranium and Thorium Metals* (Battelle 2006).

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209-E and 120 Critical Mass Laboratories, 1950 to 1986: Use plutonium intakes (M, S, or Super S) from Attachment C.

308 Plutonium Fuels Pilot Plant, 1960 to 1990: Use fission and activation products (fuel dissolution), plutonium (M, S, or Super S), and uranium (M or S), and ¹⁴⁷Pm intakes from Attachment C and ORAUT (2007a). Use ¹³¹I intakes from Table 5-31; ²⁴Na intakes may be ignored for best estimates. Intakes of ²⁴⁴Cm from July 1, 1972 through 1973 may be used in lieu of plutonium if it is more favorable to the claimant.

324 Building, 1966 to 2001: Use fission and activation products (fuel dissolution) from Attachment C (except ¹⁴⁷Pm) and ORAUT (2007a). The fuel was well-aged before arriving, so ²⁴Na and other short-lived radionuclides in ORAUT (2007a) may be ignored for best estimates and may use ⁶⁵Zn ingestion intakes only. From July 1, 1972 on, include intakes of ²⁴¹Am at plutonium coworker intake levels from Attachment C.

325 Building, 1953 to the present: Use fission and activation products (fuel dissolution), plutonium (M, S, or Super S), and ¹⁴⁷Pm intakes from Attachment C and ORAUT (2007a); ²⁴Na may be ignored for best estimates; however, because of multiple uses of this facility, short-lived radionuclides might have been present past 1987.

327 Building, 1953 to 1987: Use fission and activation products (fuel dissolution) and plutonium (M,S, or Super S) intakes from Attachment C and ORAUT (2007a). Include ¹⁴⁷Pm for 1966 through 1970.; ²⁴Na may be ignored for best estimates.

3706 Building (Radiochemistry Laboratory, Technical Building), 1945 to about 1963: Assign fission and activation products (from September 1, 1946 on) and plutonium (from September 1, 1946 on, types M, S, or Super S) intakes from Attachment C (except ¹⁴⁷Pm) and ORAUT (2007a). Assign uranium intakes (types M or S from January 1, 1949 on) from Attachment C.

3732 Building (Process Equipment Development Laboratory) and 3722 Building, July 1, 1972-1980: Note: thorium work ceased in 1970 but decontamination of these facilities occurred shortly after shutdown. The time period has not been determined. Exposure prior to July 1, 1972 is covered by the SEC because it was determined to be infeasible to reconstruct doses, so no intakes are assigned. Assign thorium based on uranium coworker intakes from Attachment C (type, F, M, or S).

3708 Building: Assign intakes of ²⁴¹Am or ²⁴⁴Cm based on plutonium coworker intakes from Attachment C for July 1, 1972 to 1973.

Tank Farms, Evaporators, and 340 Liquid Waste Handling Buildings, September 1, 1946 to the present: Assign fission and activation products (waste management) from Attachment C (except ¹⁴⁷Pm) and ORAUT-OTIB-0054 (ORAUT 2007a). Assign plutonium (M, S, or Super S) or americium intakes at 0.15 times the ¹³⁷Cs fission product intake from Attachment C (to account for an alpha component in the waste). ²⁴Na may be ignored for best estimates, and ²⁴Na, ⁶⁵Zn, and radionuclides with short half-lives may be ignored after 1988.

D&D and Remediation, 2002 to the present: For employees of the site's remediation contractor whose job description indicates potential for exposure to airborne contamination, assign 40-DAC-hr of unmonitored intakes per year (based on the investigation level of the lapel air sampling program). Various radionuclides are possible; however, choose the most favorable to the claimant of the following (choose one): 260 pCi/d of either ⁹⁰Sr (type F) ¹³⁷Cs (type F), ¹⁵²Eu (type M), or ¹⁴C (particulate type M or S), or 0.26 pCi/d of ²³⁹Pu (Type M, S, or Super S) [122].

Rovers: Could have been exposed at many different facilities. Use the same intakes as for the fuel separations facilities, except do not include ²³³U.

Unknown facility: Use judgment based on any available information and ORAUT (2007a). If the work can be associated with the 100 Area, assume exposure at a reactor. If it can be associated with the 200 Area, then consider assuming the worst case, which would be the fuel separations facilities. If it can be associated with the 300 Area, it would be helpful to determine if the work was associated with fuel fabrication or one of the research facilities. After 1964, fuel fabrication was done by the same contractor(s) as operated the 100 Area reactors (e.g., Douglas or United Nuclear Corporation). Assume 308 Building if no other 300 Area information is available. If the work location is listed as the 600 Area, assign environmental intake only.

5.7 ATTRIBUTIONS AND ANNOTATIONS

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

Donald Bihl served as the initial Subject Expert for this document. Mr. Bihl was previously employed at Hanford and his work 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 (and earlier revisions) have been overseen by a Document Owner who is fully responsible for the content, including all findings and conclusions. Mr. Bihl continues to serve as a Site Expert for this document because he possesses or is aware of information relevant for reconstructing radiation doses experienced by claimants who worked at the site. In all cases where such information or prior studies or writings are included or relied upon by Mr. Bihl, those materials are fully attributed to the source. Mr. Bihl's Disclosure Statement is available at www.oraucoc.org.

Fred Duncan assumed responsibility as Document Owner for this document in September, 2008. Mr. Duncan replaced Edward Scalsky when Mr. Scalsky's employer declared a new corporate conflict of interest for the Hanford Site. Mr. Scalsky continues to participate on this document team in the appropriate role of Subject Expert in compliance with the NIOSH Conflict or Bias policy.

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- [1] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. These were the protocols for reporting results from the bioassay laboratory to the dosimetry database.
- [2] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. These were the protocols for reporting results from the bioassay laboratory to the dosimetry database.
- [3] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussions with former *in vivo* counting program staff E. H. Palmer and G. A. Rieksts. The GOK region also shows on whole-body count forms from the 1960s and 1970s.
- [4] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. The dates for HIE and ORE are from Wilson (1987). The other remarks are from personal experience and discussions with various program managers of the Radiological Records Program.
- [5] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and review of the bioassay database.
- [6] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and review of the bioassay database.
- [7] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Information in the previous paragraph came from personal experience, common knowledge passed down from previous managers of the Hanford Internal Dosimetry Program, and review of the various statements of work with the bioassay laboratory.
- [8] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is due to the nature of the analysis, which involved separation of the plutonium then gross alpha analysis. Plutonium -241 is a beta emitter, and ²⁴¹Am would have been removed during the chemistry.
- [9] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. In addition, most of this information is readily noticeable based on queries of the database, which show the nature of how the data were recorded and when the changes in recording practices occurred. Detection levels are also discussed in Wilson (1987). The recording of 0.025 dpm/sample for nondetections during 1975 to 1982 and recording of actual results for measured results >0.025 dpm/sample was also verified when the former Personnel Dosimetry Program Manager, K. Heid, was asked that direct question shortly after his retirement in the late 1980s.
- [10] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2004. This information is based on discussion in Wilson (1987) and general understanding of how the concepts of detection level and MDA have changed over the years.
- [11] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. In addition, most of this information is readily noticeable based on queries of the database, which show the nature of how the data were recorded and when the changes in recording practices occurred. Detection levels are also discussed in Wilson (1987). The recording of 0.025 dpm/sample for nondetections during 1975 to 1982 and recording of actual results for measured results >0.025 dpm/sample was

also verified when the former Personnel Dosimetry Program Manager, K. Heid, was asked that direct question shortly after his retirement, in the late 1980s.

- [12] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2006. This information was provided by J. A. MacLellan, the Bioassay Laboratory Contract Technical Administrator, and C. L. Antonio, Internal Dosimetrist in the Hanford Internal Dosimetry Program. It is also discussed in a letter to the Hanford Internal Dosimetry Program file (Antonio 2002).
- [13] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience.
- [14] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and discussions with J. A. MacLellan, the Bioassay Laboratory Contract Technical Administrator.
- [15] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. In addition, a summary of bioassay procedures is contained in PNNL (2003b). That two results were occasionally recorded for the same sample, because both splits were analyzed, is also evident from a query of the database.
- [16] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The MDAs were extracted from various statements of work for the bioassay laboratory. The true MDAs for various times were determined as part of the laboratory's quality control program and from a double-blind oversight program operated by the Hanford Internal Dosimetry Program.
- [17] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The requirement to report TPU shows in the 1983 statement of work (Simpson 1983). A query of the database shows that reporting of the uncertainty was sporadic in 1980 and became routine in 1981.
- [18] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was extracted from various statements of work with the bioassay laboratory, including AEC (1964), ERDA (1975), DOE (1979), Simpson (1983), Author unknown (1985, 1987), and Acker (1992).
- [19] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. There is some mention of the ²³⁸Pu operations in Gerber (1992), although that reference does not give the date of 1967. The original reference (an incident write-up) using the year 1967 has not been located.
- [20] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussions with PUREX engineers in the 1980s and from Sula, Carbaugh, and Bihl (1989).
- [21] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The composition of the plutonium used in the Nuclear Waste Vitrification Project was added to the Internal Dosimetry Program technical basis document based on a memorandum from D. Bihl to Program staff, "Unusual Plutonium Mixture" (Bihl 1992), which in turn was based on a description of the mixture in a more formal document, only a portion of which was found in the files.

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- [22] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. In mixtures that are mostly americium by mass or where the americium atoms have been separated from the plutonium atoms, the americium behaves as americium, not as plutonium.
- [23] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2004. Information in the previous five sentences was based on a query of the REX database.
- [24] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2004. No information was found to indicate whether the units were activity per sample or activity per liter during 1964. Guidance was required; therefore, the units used in 1967 to 1969 were assumed.
- [25] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2004. This section is a continuation of the discussion about the 41 analyses introduced in the previous paragraph. This discussion is leading to an estimation of the MDA for the americium analysis in 1964.
- [26] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2004. This is based on calculations by the author on the 1964 dataset in an attempt to determine the MDA of the procedure at that time.
- [27] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Most of the information in this table is discussed in the text. The values during 1990 and 1991 resulted from the use of several DOE laboratories after the default of the contract with the commercial laboratory. The values from October 1991 are the same as for the plutonium analysis. In addition, see Table 5-1, the discussion in the plutonium section, and Attribution 12.
- [28] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience, results of a query of the REX database, and research on some of the cases in the files.
- [29] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience.
- [30] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), ERDA (1975), DOE (1979), Simpson (1983), Author unknown (1985, 1987), and Acker (1992).
- Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003.
 This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), Acker (1992), and PNNL (2003b, Appendix B).
- [32] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was provided by J. A. MacLellan, the Bioassay Laboratory Contract Technical Administrator, and C. L. Antonio, Internal Dosimetrist in the Hanford Internal Dosimetry Program. It is also discussed in a letter to the Hanford Internal Dosimetry Program file (Antonio 2002).
- [33] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay

laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Acker (1992).

- [34] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. It is also evident from a query on the dosimetry database. It was standard practice to include the tritium doses as external dose until the mid-1980s.
- [35] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).
- [36] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. January 2009. Based on a conversation with W. Baumgartner, who worked as a radiation monitor for a time at the 108-B tritium plant
- [37] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. January 2009 Based on a conversation with W. Baumgartner, who worked as a radiation monitor for a time at the 108-B tritium plant.
- [38] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Plutonium impurity would have resulted from incomplete separation of plutonium from the irradiated uranium, of which ²³⁹Pu was the isotope with the highest activity. The thorium impurity was probably a mix of ²³²Th, ²²⁸Th, and ²³⁴Th because all three isotopes were in PUREX; however, the assumption of 100% ²³²Th is efficient and favorable to claimants.
- [39] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2006. Because both the DOE Hanford RU document (DOE 2000) and the Hanford internal dosimetry manual (PNNL 2003a) express the entity as ⁹⁵Zr/⁹⁵Nb, it is hard to know for sure if the activity is for each radionuclide or a total of both; however, DOE (2000) also refers to ¹⁰⁶Ru/¹⁰⁶Rh. In the latter case it is clear that the activity refers to the sum of the parent and progeny, so it was deemed reasonable to assume that the 20 nCi of ⁹⁵Zr/⁹⁵Nb also refers to the sum of the parent and progeny. Because the radionuclides are usually found close to equilibrium, the recommendation was made to use 10 nCi ⁹⁵Zr and 10 nCi ⁹⁵Nb.
- [40] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Hanford Internal Dosimetry Program staff member M. Long did some research on uranium discharged to the soil by asking various Hanford and PNNL soil chemists. Long concluded that uranium would be in a carbonate complex that most likely would be quite soluble (Long 1993).
- [41] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience.
- [42] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. For some reason the reporting of activity in the ^{233/234}U energy region was set up as ²³³U early on; the switch to reporting as ²³⁴U was personally negotiated with the laboratory by Bihl.
- [43] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The rapid method was coded QUS for "quick uranium soluble." Sula, Carbaugh, and Bihl (1989, Appendix C) refers to it as the "less sensitive method." The kit code for the QUS was 7.

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- [44] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).
- [45] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2005. The ²³³U results were recorded on the same data sheets as for regular uranium analyses, and the units were printed on the form. However, the magnitude of the results argues against units of micrograms per liter, and normal uranium analysis methods would not have produced results that small (ranging from 2×10^{-7} to 4×10^{-6}). A decision had to be made as to the units for the dose reconstructors to apply. It was reasoned that autoradiography was used to measure the tracks from the ²³³U alpha particles in the same manner as was being used for plutonium bioassay. As such the units would have been microcuries per sample. Someone probably forgot to mark the special units on the uranium bioassay form.
- [46] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Data in the table was compiled from the references in the text and from a query of the bioassay database, which showed the distinct reporting levels used at different times.
- [47] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is evident from a query on the bioassay data that shows use of the fission product urinalysis from 1960 to 1964. Microfiche records of several of the workers with fission product urinalysis were reviewed and showed evidence of working at B Plant and Semi-Works. The same workers showed ⁹⁰Sr urinalyses beginning in 1965. Therefore, it was concluded that the use of the fission product urinalysis after 1960 was for workers potentially exposed to radiostrontium.
- [48] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This was explained to Bihl verbally during a regular contractual meeting between the Hanford Internal Dosimetry Program representative, the Battelle contract representative, and staff from the bioassay laboratory in about the late 1980s. This practice was considered standard, had been going for quite awhile, and continued throughout the duration of the contract.
- [49] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).
- [50] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. No use of strontium titanate at Hanford has been discovered.
- [51] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).
- [52] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information is based on query of the database and review of the data.
- [53] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).

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- [54] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. March 2009. Only four records of ¹⁴⁷Pm *in vivo* counts were found in the REX database. It is possible that some early counts were not entered into the database, but it is unlikely that this type of count was performed other than rarely.
- [55] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).
- [56] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This information was compiled from the various statements of work with the bioassay laboratory, including AEC (1964), Lardy (1970), ERDA (1975), DOE (1979), BPNL (1982), Simpson (1983), Author unknown (1985, 1987), and Adkins (1992).
- [57] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2005. This is based on queries of the REX database.
- [58] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2005. This is based on queries of the REX database.
- [59] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with the project manager, Dr. D. Fisher, a PNNL employee.
- [60] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on a casual discussion with Dr. M. Sikov, a retired PNNL radiobiologist, now deceased.
- [61] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Bihl had access to a series of Laboratory Record Notebooks associated with the *in vivo* counting facility. Laboratory Record Notebooks are proprietary, and permission was not sought to be able to copy them.
- [62] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The earliest detection levels came from a slide in a training class on whole-body counting at Hanford from about 1964. The author was not stated, although it was most likely E. H. Palmer, the In Vivo Monitoring Program Manager at the time (Author unknown 1964). Some of the detection levels were also shown, with calculations, in Palmer's 1960 Laboratory Record Notebook. Based on the calculations, Bihl estimated that the recorded detection levels most closely approximated the modern-day concept of decision level. The 10-nCi recording level for some of the radionuclides came from *Non-Routine Whole Body Counter Calculations* (Glenn 1968).
- [63] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. July 2005. This is based on query of the REX database which shows a clear cutoff of reported results at 0.66 nCi for ¹³⁷Cs.
- [64] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience. It is also evident from query on the REX database that shows the start of reporting of negative numbers on March 29, 1993.
- [65] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The reporting of these radionuclides is evident from a query on the database. It was also

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obvious from hard-copy printouts of *in vivo* data from the first computer databases that were used by the *in vivo* counting program. A yearly printout was made of all the counts for each calendar year and stored in the Radiological Records library. These printouts had hard-coded columns for ¹³⁷Cs, ²⁴Na, ⁶⁵Zn, and ⁴⁰K, whereas other radionuclides were noted in a comments field. For reasons not understood by Bihl, there were some counts for which one or more of the four regularly reported radionuclides were missing. So as a general rule they were nearly always reported, but clearly there are some exceptions in the database.

- [66] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Although the persistence of burdens of ²⁴Na and ⁶⁵Zn were discovered when whole-body counting became routine and the sources were investigated over the next couple of years, it is favorable to claimants and reasonable to assume that the radionuclides were in the drinking water starting soon after the first once-through-cooling reactor started at Hanford.
- [67] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal discussions with former *in vivo* counting program staff, H. E. Palmer and G. A. Rieksts. Palmer was the *in vivo* counting program manager for many years, and Rieksts was a scientist in the program for many years. Both are retired. The GOK region is also shown on the early whole-body counting results forms.
- [68] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Because nearly every U.S. resident had detectable burdens of ¹³⁷Cs in the 1960s and 1970s, the next three bullets were provided as guidance on when to assign occupational intakes of ¹³⁷Cs
- [69] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal discussions with H. E. Palmer and G. A. Rieksts.
- [70] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and discussion with G. Rieksts. In addition, see for instance the 1992 *In Vivo Bioassay Statement of Work* (Lynch et al. 1992, p. 3.6).
- [71] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with program managers H. E. Palmer and T. P. Lynch. In addition, it is implied in most of the annual reports, such as Lyon et al. (1988) and others.
- [72] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and discussion with H. E. Palmer.
- [73] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2005. This is based on queries on the REX database for ¹³¹I results.
- [74] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2005. This is based on discussion with T. P. Lynch and G. A. Rieksts.
- [75] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2006. This is based on calculations by R. Traub; however, similar values can be extracted from the graph in West (1965).
- [76] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. June 2005. The terms "detection limit" or "less-than" during the 1960s appear to be most closely associated with the present concept of decision level. The MDA is approximately twice the

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decision level; therefore, the recommendation was made to assume that the MDA was twice the reported detection level.

- [77] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2006. This is based on discussion with T. P. Lynch; however, this is an educated guess by Lynch based on present practices and his knowledge of the detection systems at the time. Documentation that the ²³²Th body burdens in the 1960s and 1970s were based on ²²⁸Ac has not been found. It is also not known if an adjustment for disequilibrium between ²²⁸Ac and ²³²Th was made before the results were reported. It was considered favorable to claimants to assume the disequilibrium adjustment was not made and to direct the dose reconstructors to revise the ²³²Th results based on disequilibrium factors in the text.
- [78] Traub, Richard. Battelle Memorial Institute. Principal Health Physicist. November 2006. This is based on calculations by R. Traub using Matlab[™]. The calculations incorporated the latest ICRP biokinetic models with independent systemic biokinetics for the major progeny.
- [79] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2006. Because ²²⁸Ac is usually considered part of natural background, it is not routinely calculated or reported in a whole-body count spectrum. If it had been known that the worker was exposed to thorium, then any detectable activity would have been calculated and reported, but otherwise it would take a more significant peak to prompt an investigation about possible thorium exposure. The exact level of activity that would have caused an investigation has not been found; the 2-nCi level was an assumption based on the fact that levels well below 1 nCi were being recorded in the early 1970s and detection levels have generally improved over time. Therefore, 2 nCi is favorable to claimants.
- [80] Traub, Richard. Battelle Memorial Institute. Principal Health Physicist. December 2006. These were calculated by R. Traub using Matlab[™]. The calculations incorporated the latest ICRP biokinetic models with independent systemic biokinetics for the major progeny.
- [81] Traub, Richard. Battelle Memorial Institute. Principal Health Physicist. December 2006. These were calculated by R. Traub using Matlab[™]. The calculations incorporated the latest ICRP biokinetic models with independent systemic biokinetics for the major progeny.
- [82] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2006. Because of the very high activities of fission products that were handled in the PUREX canyon, the ventilation system was built to ensure exhaust of contaminants through the canyon filtration and exhaust system. The air flow rate through the canyon exhaust stack was nominally 100,000 ft³/min. Fuel with high thorium and ²³³U content was dissolved in the canyon in the same way as normal fuel and thoron was exhausted out the canyon stack.
- [83] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and also evident from a query of the REX database.
- [84] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and discussion with T. Lynch, *in vivo* monitoring program manager. The correction for shine from the liver is also mentioned in BPNL (1990).
- [85] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on review of the REX database and a sampling of whole-body count results forms in the 1960s and 1970s.

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- [86] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with T. Lynch, *in vivo* monitoring program manager. Document research has not found a reference that states this precisely, but Lynch (1992) shows the detailed work being done in relation to decision levels at that time.
- [87] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is evident from a review of various whole-body count results forms.
- [88] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. These changes were evident from review of the REX database.
- [89] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Implementation of NEXEC is discussed in Lyon et al. (1996). PNL (1995, Table 4.1) shows the 12 regions for the stand-up counter, and Table 4.5 shows the 20 regions for the coaxial germanium counter.
- [90] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on personal experience and discussion with T. Lynch.
- [91] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The minutes of the November 1998 meeting of the Dosimetry Advisory Committee (Bihl 1998) show the proposal by T. Lynch to reduce the routinely reported radionuclides in whole-body counts to ¹³⁷C, ⁶⁰Co, ¹⁵⁴Eu, ⁴⁰K, and -²⁴Na. By the time the ABACOS software was implemented in November 1999, ²⁴Na had been dropped because Hanford contractors believed there was no significant source of ²⁴Na left. Subsequent minutes of the Dosimetry Advisory Committee were scanned, but documentation of the decision to drop ²⁴Na was not found.
- [92] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. May 2006. The FFTF was sodium cooled, so neutron activation of the sodium was a potential source of contamination (Section 2.1.9).
- [93] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. March 2007. The principal ¹⁵⁴Eu photopeak is at 1.27 MeV with 35% abundance; the photopeak that was used to quantify ⁶⁰Co was at 1.33 MeV with 100% abundance. The detector efficiencies per emitted photon would have been nearly the same for the two photons; the background counts in the respective energy regions would also have been similar. Therefore, the MDAs would have differed by the ratio of the yields, or 1.00/0.35 ≈ 3.
- [94] Napier, Bruce A. Battelle Memorial Institute. Principal Health Physicist. December 2008. B. A. Napier conducted a study of present alpha to beta-gamma emitting radionuclides in all the high-level waste tanks from the TWINS 2001 Tank Farm Inventory Database (http://twins.pnl.gov/twins3/twins.htm). The ratios in the tanks were fit to a lognormal distribution and the 95th-percentile ratio was obtained. The ratios from the TWINS database were compared to a database for the Hanford Environmental Dose Reconstruction Project (Napier 1992) after correction for removal of strontium and cesium at B Plant and the West Encapsulation Facility and after decay correction to 2001. The study showed that in general the tanks historically had smaller alpha to beta/gamma ratios because the fuel being processed had less cooling time and, of course, present waste has had many additional years of decay. Shorter-cooled and shorter-decayed fuel has more activity from beta- and gammaemitting radionuclides.

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- [95] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is evident from a query on the database. Error limits are listed as "±10% at the 95% confidence level at 10 times the detection limits" in the earliest statement of work for the bioassay laboratory (AEC 1964), and similar statements show up in subsequent statements of work, but uncertainties for each sample were not reported at that time. There was probably a change to the statement of work in 1981 that instigated or accompanied the change in that year to start reporting uncertainties for each sample, but a copy of that statement of work has not been found. Propagation and reporting of uncertainty for each sample shows up in the 1983 bioassay laboratory statement of work (Simpson 1983) and in each statement of work thereafter.
- [96] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The collection instructions are to collect starting with the last excretion before retiring for sleep, any other voidings during the night (or sleep period if on shift), and first excretion on rising in the morning for two consecutive nights. This covers about 18 hours or about 75% of a 24-hour period.
- [97] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with the *in vivo* monitoring program manager T. Lynch.
- [98] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with the *in vivo* monitoring program manager T. Lynch.
- [99] Bihl, Donald. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with the *in vivo* monitoring program manager T. Lynch.
- [100] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The 95th-percentile phantom did not exist during the period that the 1-m arc or the shadow shield detectors were in use, and that phantom was not measured on the standup counter, so the impact of large workers on results from those detectors has not been quantified. T. P. Lynch believed the 25% bias was a reasonable figure to use for the latter detectors as well as the coaxial counter based on his general experience with the various detectors.
- [101] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. This is based on discussion with the *in vivo* monitoring program managers T. P. Lynch and G. A. Rieksts.
- [102] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2005. N Reactor, the last source of short-lived fission products, shut down in January 1987 after a brief operating period, and was also shut down for most of 1986. The December 31, 1988, date was arbitrarily chosen to allow time for the short half-life radionuclides to decay such that they were no longer significant contributors to contamination.
- [103] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. Workers without at least a basic dosimeter were not allowed into radiation areas or general areas of the site (200 Area, 300 Area, etc.) where there was some risk of exposure; some companies gave beginning work or termination bioassay to everyone regardless of their work location or tasks, and those bioassays are not indicative of exposure. Therefore, after consultation with other ORAU Team experts (including J. Fix, E. Scalsky, and E. Brackett), the conclusion was reached that workers without at least a basic dosimeter were not at risk for occupational intakes, and the instruction was made to assign environmental intakes only.

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- [104] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2005. This is based on queries of the REX database and an incident record in the worker's file about the T Plant incident.
- [105] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2005. Equilibrium thyroid burden from daily inhalation of 20,000 pCi of type F¹³¹I is 29.5 nCi after about 80 days. This calculation was performed using IMBA. This burden exceeds 93% of all the measurements made on at-risk workers in 1945 and 1946 and exceeds 90% of the measurements made from 1962 to 1969, so it was deemed to be favorable to claimants to apply the dose to unmonitored workers in years before implementation of routine *in vivo* counts.
- [106] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2005. The whole-body burden from daily inhalation of 3,400 pCi of type F¹³¹I is 5 nCi after only 10 days of intake and reaches an equilibrium burden of 7.2 nCi after about 80 days. This calculation was performed using IMBA.
- [107] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2005. N Reactor shut down in May 1986 and restarted very briefly in January 1987. With an 8-day half-life, ¹³¹I activities would have been negligible by the end of 1987.
- [108] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. May 2006. This is based on query of the REX database.
- [109] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. May 2006. This is based on query of the REX database.
- [110] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. May 2006. This was calculated using IMBA.
- [111] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. May 2006. There were considerable differences in potential exposure to tritium between the 108B facility, which involved extracting and packaging curie quantities of tritium in a relatively small space, and exposures at large, heavily ventilated facilities with unconcentrated tritium such as the reactors and fuel separation facilities. Most of the tritium at the latter facilities was exhausted through the stacks or discharged to the ground in cribs. It is likely that the relative exposure to workers at the reactors or fuel separations plants was less than an order of magnitude relative to exposure at 108B. However, lacking air sample data, the order of magnitude ratio was considered reasonable and favorable to claimants.
- [112] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. Joseph Lochamy. XCEL Engineering. Scientist. Richard Sparks. CDE Dosimetry Services. Principal Health Physicist. December 2006. The tritium dose data is from a query on REX by Bihl, a lognormal statistical analysis was performed by Lochamy in accordance with ORAUT-PROC-0095, *Generating Summary Statistics for Coworker Bioassay Data* (ORAUT 2006), and a validation of statistical analysis was performed by Sparks.

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[113] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. May 2006. There were considerable differences in potential exposure to tritium between the 108B facility, which involved extracting and packaging curie quantities of tritium in a relatively small space, and exposures at large, heavily ventilated facilities with unconcentrated tritium such as the reactors and fuel separation facilities. Most of the tritium at the latter facilities was exhausted

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through the stacks or discharged to the ground in cribs. It is likely that the relative exposure to workers at the reactors or fuel separations plants was less than an order of magnitude relative to exposure at 108B. However, lacking air sample data, the order of magnitude ratio was considered reasonable and favorable to claimants.

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- [114] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. November 2006. This was calculated using IMBA.
- [115] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist; Joseph Lochamy. Scientist. XCEL Engineering, Inc.; and Richard Sparks. CDE Dosimetry Services, Inc. Principal Health Physicist. December 2006. The tritium dose data is from a query on REX by Bihl, a lognormal statistical analysis was performed by Lochamy in accordance with ORAUT-PROC-0095, *Generating Summary Statistics for Coworker Bioassay Data* (ORAUT 2006), and a validation of statistical analysis was performed by Sparks.
- [116] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2006. These were calculated using IMBA.
- [117] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. December 2006. Although the tritium source term would have been smaller in the late 1980s and early 1990 because of reduced processing of fuel, it is favorable to claimants to use the values for 1984 to 1986 for the later years.
- [118] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. June 2005. These were calculated using IMBA. Ingestion of 40,000 pCi/d produces approximately 1 mrem/yr to all organs.
- [119] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. June 2006. There were not enough samples for a formal coworker analysis (n=11). The units were not known exactly; based on the magnitude of the results, microcuries per liter was assumed (see Section 5.2.5).
- [120] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. June 2006. This was calculated using IMBA.
- [121] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. June 2006. This is a summary of information developed in the text.
- [122] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. June 2006. This is based on discussion with the D&D contractor's dosimetry expert E. W. Carlson. It reflects radionuclides being encountered during D&D operations of old facilities, burial sites, liquid effluent discharge zones, etc. Because most of the work is outside, the contractor uses lapel air samplers to monitor for intakes. The 40-DAC-hr value comes from the requirement to initiate special bioassay if an individual's cumulative intake in a year as measured by the air samplers reaches or exceeds 40 DAC-hr (PNNL 2003b).
- [123] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. October 2003. The air concentration tolerance value as applied to fission products was deduced from the fact that other values were given for "product," ¹³¹I, and uranium. In addition, the magnitude of the limit implies general fission products.

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- [124] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. This is from a query on the REX database by the REX custodian.
- [125] Arno, Matthew. Foxfire Scientific. Dose Reconstructor. June 11, 2007. The periods for how data was recorded are taken directly from examination of the HERB database.
- [126] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. July 2005. The uranium intakes were identified by their pattern of excretion associated with each individual followed by review of each individual's radiological exposure file, to which Bihl had access. The description in the radiological exposure file was used to determine the specifics of the intakes. Most intakes investigated in this way were not excluded from the database; only intakes considered unusual and not relevant to a coworker analysis were excluded.
- [127] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. July 2005. The promethium intakes were identified by the patterns of excretion that were associated with each individual and followed by review of each individual's radiological exposure file, to which Bihl had access. The description in the radiological exposure file was used to determine the specifics of the intakes. Most intakes investigated in this way were not excluded from the database; only intakes considered unusual and not relevant to a coworker analysis were excluded.
- [128] Arno, Matthew. Foxfire Scientific. Dose Reconstructor. June 2007. Samples with known volumes are normalized using a urinary excretion rate of 1.4 L/d. Samples without a known volume are assumed to be representative of a full day of urinary excretion because no additional information is available.
- [129] Arno, Matthew. Foxfire Scientific. Dose Reconstructor. June 2007. Ten-year aged fuel-grade plutonium is the default assumption per the main text of this document. However, before 1955, it is not possible to have 10-year aged plutonium. Therefore, the most favorable to claimant credible mixture is used.
- [130] Arno, Matthew. Foxfire Scientific. Dose Reconstructor. June 2007. Internal dose conversion factors for ²³⁴U, ²³⁵U, and ²³⁸U are all similar, but those for ²³⁴U are the most favorable to claimants and are therefore used as a simplification.
- [131] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. July 2005. Plot created by Bihl from data in Heeb (1994, Appendix A).
- [132] Bihl, Donald E. Battelle Memorial Institute. Principal Health Physicist. March 2009. The Hanford Site Description (ORAUT 2010c) lists the buildings and periods of work with mixtures highly enriched in ¹⁴⁷Pm or with purified ¹⁴⁷Pm. In general the early work was in developing separations techniques and was done by only a few chemists and chemical technicians, and major production started in 1966. Most of the work was done in the hot cells in the 325 Building with early low-level work in fume hoods in the 325 Building or the 1-F cubicle in 222-S. Urinalyses were taken for 29 workers in the 1973 contamination spread in the 325 Building (AEC 1963), but only the calculated body burdens were found. These were converted to intakes and urine excretion using the Langham equation as stated in the report. The calculated urine excretion values were then recalculated to intakes using IMBA and present-day models. The 95th percentile of these recalculated intakes were comparable, albeit slightly higher, compared to annualized intakes from Table C-28 for the period of major production from 1966 to 69. Considering that the February 1963 bioassays resulted from a major contamination spread, and that work conditions were similar in the two periods, it was

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concluded that the 1966 to 1969 coworker intakes were reasonable surrogates for intakes by workers in the 1961 to 1965 period that is not associated with contaminating events.

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GLOSSARY

absorbed dose

Amount of energy (ergs or joules) deposited in a substance by ionizing radiation per unit mass (grams or kilograms) of the substance and measured in units of rads or grays See *dose*.

active

Hanford term, circa 1940s and 1950s, meaning *radioactive*. Example, "production of acetylene from the active water, with subsequent measurement of the ionization caused by the tritium beta particle." See *radioactive*.

age or aging

In relation to reactor fuel and mixtures of plutonium isotopes, time since the step in the refinement process that separates americium from the mixture.

alpha particle (α)

See alpha radiation.

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.

ashing

The driving off of all moisture in a sample; accomplished by heating in an oven or by dissolving the sample in a liquid (often nitric acid), then evaporating to dryness. The latter technique is called *wet ashing*.

atom

Smallest amount of an element that cannot be chemically divided or broken up. It consists of a central core (the nucleus) surrounded by one or more electrons. See *element* and *nucleus*.

beta particle (β)

See beta radiation.

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.

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.

cooling

In relation to reactor fuel, the time since removal of the fuel from the reactor core.

curie (Ci)

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

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dose

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

dosimeter

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

electromagnetic radiation

Energy that results from the acceleration of electric charge and the associated electric and magnetic fields. The energy can be regarded as waves propagated through space or as a stream of photons. The electromagnetic radiation spectrum ranges from short wavelengths (X- and gamma rays) to intermediate wavelengths (ultraviolet, visible, and infrared light) to long wavelengths (radar and radio). Only the higher energy (shorter wavelength) forms of electromagnetic radiation can ionize matter. Lower energy electromagnetic radiation, such as visible and infrared light, radar, and radio waves, does not have enough energy to ionize matter.

electron

Basic atomic particle with negative charge and a mass 1/1,837 that of a proton. Electrons surround the positively charged nucleus of the atom. See *element*.

element

One of the known chemical substances in which the atoms have the same number of protons. Elements cannot be broken down further without changing their chemical properties. Chemical symbols for the elements consist of either a single letter or a combination of letters, some of which descend from the Latin names [e.g., Au from *aurum* (gold), Fe from *ferrum* (iron)]. This glossary indicates *elements* by their names. Specific *isotopes* appear as their standard chemical symbols with the number of protons and neutrons in the nucleus. For example, the isotope of uranium that contains 92 protons and 143 neutrons can appear as ^{235}U , *U-235*, or *uranium-235*. See *radioactive isotope*.

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.

exposure

(1) In general, the act of being exposed to ionizing radiation. See *acute exposure* and *chronic exposure*. (2) Measure of the ionization produced by X- and gamma-ray photons in air in units of roentgens.

film dosimeter

Package of film for measurement of ionizing radiation exposure for personnel monitoring purposes. A film dosimeter can contain two or three films of different sensitivities, and it can contain one or more filters that shield parts of the film from certain types of radiation. When developed, the film has an image caused by radiation measurable with an optical densitometer.

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fission

Splitting of the nucleus of an atom (usually of a heavy element) into at least two other nuclei and the release of a relatively large amount of energy. This transformation usually releases two or three neutrons.

fuel-grade

Plutonium isotopic mixture with approximately 12% ²⁴⁰Pu.

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.

gamma ray, particle, or photon (γ)

See gamma radiation.

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.

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.

ionization

Process by which a neutral atom or molecule loses or gains electrons and therefore acquires a net electrical charge. See *ionizing radiation*.

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. See alpha radiation, beta radiation, gamma radiation, neutron radiation, and X-ray radiation.

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., ²³⁴U, ²³⁵U, and ²³⁸U). Isotopes have very nearly the same chemical properties. See *element*.

nucleus

Central core of an atom, which consists of positively charged protons and, with the exception of ordinary hydrogen, electrically neutral neutrons. The number of protons (atomic number) uniquely defines a chemical element, and the number of protons and neutrons is the mass number of a nuclide. The plural is nuclei.

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neutron (n)

Basic nucleic particle that is electrically neutral with mass slightly greater than that of a proton. There are neutrons in the nuclei of every atom heavier than normal hydrogen. See *element*.

neutron radiation

Radiation that consists of free neutrons unattached to other subatomic particles emitted from a decaying radionuclide. Neutron radiation can cause further fission in fissionable material such as the chain reactions in nuclear reactors, and nonradioactive nuclides can become radioactive by absorbing free neutrons. See *neutron*.

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.

nuclide

Stable or unstable isotope of any element. Nuclide relates to the atomic mass, which is the sum of the number of protons and neutrons in the nucleus of an atom. A radionuclide is an unstable nuclide.

operating area

Designation of Hanford major operational work areas among the respective fuel fabrication (e.g., 300 Area), reactor operations (e.g., 100B, 100C, 100D, 100DR, 100F, 100H, 100KE, 100KW, and 100N), chemical separations (e.g., U-, T-, B-, UO3, REDOX, and PUREX Plants), plutonium finishing (Z-Plant), research and development (e.g. 300 and 3000 Areas), and transportation, communication, and general site support (e.g., 600, 700, and 1100 Areas).

photon

Quantum of electromagnetic energy generally regarded as a discrete particle having zero rest mass, no electric charge, and an indefinitely long lifetime. The entire range of electromagnetic radiation that extends in frequency from 10²³ cycles per second (hertz) to 0 hertz.

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.

radioactive

Of, caused by, or exhibiting radioactivity.

radioactive isotope

Natural or synthetic form of an atom that emits radioactivity when it decays. See *isotope*.

radioactivity

Property possessed by some elements (e.g., uranium) or isotopes (e.g., ¹⁴C) of spontaneously emitting energetic particles (electrons or alpha particles) by the disintegration of their atomic nuclei. See *radionuclide*.

radionuclide

Radioactive nuclide. See radioactive and nuclide.

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reactor

Device in which a fission chain reaction occurs under controlled conditions to produce heat or useful radiation for experimental purposes or to generate electrical power or nuclear fuel.

reliably detectable activity

Measure for *in vivo* counting in the 1980s defined as three standard deviations of the spectral continuum with a peak discernable by the naked eye.

rep

Historical quantity of radiation (usually other than X-ray or gamma radiation) originally defined as 83 ergs absorbed per gram in the body and redefined in the 1940s or early 1950s as the amount that would liberate the same amount of energy (93 ergs per gram) as 1 roentgen of X-or gamma rays. Replaced by the gray in the International System of Units; 1 rep is approximately equal to 8.38 milligray. The word derives from <u>roentgen equivalent physical</u>.

simulated 24-hour urine sample

Collection of all urine samples beginning with the void before retiring for the evening and ending with the first void after rising the next morning, for two consecutive nights, to simulate a 24-hour urine sample.

Special Exposure Cohort (SEC) [42 U.S.C. § 7384I(14)]

... "member of the Special Exposure Cohort" means a Department of Energy employee, Department of Energy contractor employee, or atomic weapons employee who meets any of the following requirements:

- (A) The employee was so employed for a number of work days aggregating at least 250 work days before February 1, 1992, at a gaseous diffusion plant located in Paducah, Kentucky, Portsmouth, Ohio, or Oak Ridge, Tennessee, and, during such employment—
 - *(i)* was monitored through the use of dosimetry badges for exposure at the plant of the external parts of employee's body to radiation; or
 - (ii) worked in a job that had exposures comparable to a job that is or was monitored through the use of dosimetry badges.
- (B) The employee was so employed before January 1, 1974, by the Department of Energy or a Department of Energy contractor or subcontractor on Amchitka Island, Alaska, and was exposed to ionizing radiation in the performance of duty related to the Long Shot, Milrow, or Cannikin underground nuclear tests.
- (C) (i) Subject to clause (ii), the employee is an individual designated as a member of the Special Exposure Cohort by the President for purposes of the compensation program under section 7384q of this title.
 - (ii) A designation under clause (i) shall, unless Congress otherwise provides, take effect on the date that is 180 days after the date on which the President submits to Congress a report identifying the individuals covered by the designation and describing the criteria used in designating those individuals.

thoron

Informal name for ²²⁰Rn.

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U.S. Atomic Energy Commission (AEC)

Federal agency created in 1946 to assume the responsibilities of the Manhattan Engineer District (nuclear weapons) and to manage the development, use, and control of nuclear energy for military and civilian applications. The U.S. Energy Research and Development Administration and the U.S. Nuclear Regulatory Commission assumed separate duties from the AEC in 1974. The U.S. Department of Energy succeeded the U.S. Energy Research and Development Administration in 1979.

weapons-grade

Plutonium isotopic mixture with nominally 4% to 6% ²⁴⁰Pu.

whole-body counter (WBC)

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

X-ray

See X-ray radiation.

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 TOLERANCE DOSE AND TOLERANCE VALUES AT HANFORD

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Various health physics textbooks and histories document the history of the tolerance dose. The description below was extracted from Jacob Shapiro's textbook (Shapiro 1990) and from *Radiation Protection Criteria and Standards: Their Basis and Use* (Parker 1960). H. M. Parker was the manager of the radiation protection activities at the start of Hanford and eventually became the manager of the Hanford Laboratories.

The tolerance dose was based on a rate of radiation exposure that produced no obvious harm. The basis for harm changed as understanding of health effects of radiation improved.

The first tolerance dose was established in 1934 by the International X-Ray and Radium Protection Commission as 0.2 R/d. In 1936 the U.S. Advisory Committee on X-Ray and Radium Protection reduced the tolerance dose to 0.1 R/d. This value was used at the Manhattan Project sites during World War II. For instance, Parker (1960) states that in 1945 the values accepted as a working basis for occupational exposure were:

- 100 mr/d for external X and gamma radiation
- 10 to 14 Ci/cc for radon in air of working rooms, and
- 0.1 µg of radium as the maximum allowable amount deposited in the body.

In 1949, the NCRP recommended reduction of the permissible dose to 0.3 rem/wk. In 1953, the National Committee on Radiation Protection (predecessor of NCRP) developed maximum permissible amounts of certain radionuclides in the body and MPCs in air and water, which were published as National Bureau of Standards Handbook 52 (NBS 1953). At that point tolerance values were replaced with MPBBs and MPCs.

Hanford's radiation protection standards were in accordance with these national and international standards. For instance, the following are examples of the "tolerance limits for prolonged exposure" at Hanford in 1945 (Cantril 1945):

0.1 R/d
0.1 rep ^a /d
0.02 rep/d
0.025 rep/d
0.01 rep/d
0.1-µg total accumulation
1×10^{-14} Ci/cc
1.0 × 10 ⁻¹³ Ci/cc
1.2 × 10 ⁻⁹ Ci/cc
10 ⁻⁵ µg/cc
5 × 10 ⁻¹⁰ µg/cc
0.5-µg total accumulation
1.5 × 10⁻⁴ µg/cc

a. The rep (roentgen equivalent physical) was a unit of dose from particulate radiation invented by H. Parker. It was a precursor to the rad. It was defined as an absorbed dose of 83 ergs/g of tissue and was later changed to 93 erg/g.

- b. Assumed to refer to fission products [123].
- c. "Product" was a euphemism for plutonium.

By March 1949, the Hanford limits had changed to incorporate the 0.3 rem/wk guidance from the National Committee. Examples are quoted below from Patterson (1949), but these same air

ATTACHMENT A TOLERANCE DOSE AND TOLERANCE VALUES AT HANFORD

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concentrations limits were discussed as Hanford limits in a lecture series by Parker (1947). The Patterson letter refers to these values as permissible tolerances.

External Radiation Limits

Whole body: 0.3 rem/wk or whatever the National Committee recommended.

Hands only: 1.0 rem/wk or whatever the National Committee recommended.

Internal Emitter Limits

0.3 rem/wk to the significant organ or whatever the National Committee recommended.

Drinking Water

Uranium: $100 \ \mu g/L$ (believe this is high).

Plutonium: 0.01 μ g/L (intend to revise to not more than 0.001 μ g/liter).

Mixed fission products: $0.1 \,\mu c/L$ (to be changed when new figure provided by K. Z. Morgan's subcommittee). [Patterson (1949) used " μc ," which has been assumed to mean microcuries.]

Air Contamination Limits

Respiratory protection was required when the following limits were exceeded or expected to be exceeded:

- Uranium: >0.05 μg/L
- Plutonium: >2 × 10⁻⁸ μ g/L (to be revised perhaps to 2 × 10⁻⁹ μ g/L)
- Mixed fission products: >10⁻⁶ µc/L [123]

Other air contamination limits included:

- Tritium: Gas 0.1 µc/L, vapor 0.01 µc/L
- Carbon: 0.02 µc/L
- Argon: 1.6 × 10⁻³ μc/L
- Iodine $1.5 \times 10^{-6} \,\mu c/L$
- Xenon: 0.01 µc/L

The tolerance values for air and drinking water were based on the dose to the significant organ and state-of-the-art understanding of the biokinetics of the element in the body. An assumption of continuous inhalation or drinking of water was usually made, although in the example below a tolerance value based on a single inhalation was also established. A sample calculation was found (Parker 1945) that provides the calculation of the tolerance value for "potentially long-continued exposure" to ¹³¹I and a "one-shot tolerance value," the latter assuming a single 8-hour exposure. For the chronic exposure, Parker assumed a thyroid radiation tolerance of 1 R/d, which resulted from an equilibrium thyroid burden of 1.95 μ Ci, which would result from a daily intake of 0.85 μ Ci. Parker assumed the volume of air that was breathed per 8-hour workday was 8 × 10⁶ cm³. For the acute intake, Parker referenced a "Project Handbook" that condoned a dose of 100 R; however, he stated that this was too close to the dose that was given patients in the treatment of hyperthyroidism, so he arbitrarily reduced the dose by a factor of 10. He then calculated an air concentration for a single 8-hour exposure to be 1.2 × 10¹¹ Ci/cm³.

ATTACHMENT B CODES USED IN BIOASSAY AND INTERNAL DOSE RECORDS

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This attachment is adapted from PNNL (2003b). These codes apply to information in the REX database and include present and historical uses. Different codes were implemented at different times according to the needs at the time; the early dates have few if any codes.

Code	Description	Comment
AAAA1	Americium	Probably Am-241
AAAA2	Americium	Probably Am-241
AAA3	Americium	Probably Am-241
AAAA4	Americium	Probably Am-241
AAAA5	Americium	Probably Am-241
AAAA6	Americium	Probably Am-241
AAAA7	Americium	Probably Am-241
AC225	Actinium-225	
ACS	Actinium-227, thorium-227	Scheduling code
AC227	Actinium-227	
AC228	Actinium-228	
AG110	Silver-110	
AM241	Americium-241	
AM242	Americium-242	
AM243	Americium-243	
BA140	Barium-140	
BETA	Beta	
BI213	Bismuth-213	
BI214	Bismuth-214	
BK249	Berkelium-249	

Table B-1. Codes and radionuclides associated with bioassay at Hanford [124].

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Code	Description	Comment
BR 82	Bromine-82	
C 14	Carbon-14	
CE141	Cerium-141	
CE143	Cerium-143	
CE144	Cerium-144	
CF249	Californium-249	
CM242	Curium-242	
CM244	Curium-244	
CO 58	Cobalt-58	
CO 60	Cobalt-60	
CR 51	Chromium-51	
CS134	Cesium-134	
CS137	Cesium-137	
EU152	Europium-152	
EU154	Europium-154	
EU155	Europium-155	
EU156	Europium-156	
EV155	?	Probably a typographical error for Eu-155 that got left in the
		database
EV156	?	Probably a typographical error for Eu-156 that got left in the
21.00		database
FE 59	Iron-59	
FP	Fission products	
GA	Gross alpha	
GB	Gross beta	
GELI	Gamma-GeLi detector	Excreta scheduling code for a gamma scan with a germanium
•		detector
GOK		See text
GS	Gamma Nal detector	Excreta scheduling code for a gamma scan with a Nal detector
H 3	Tritium	
I 125	lodine-125	
I 129	Iodine-129	
I 131	Iodine-131	
I 133	Iodine-133	
IAM	Isotopic americium	Excreta scheduling code for americium separation and alpha
		spectrometry
ICA	?	Probably scheduling code for americium and curium via alpha
		spectrometry
ICM	Cm isotopic	Excreta scheduling code for curium isotopes via alpha spectrometry
IEU	Eu isotopic	Excreta scheduling code for europium separation and isotopic
		analysis
IPA	Isotopic Pu and Am-241	Excreta scheduling code
IPIU	Isotopic Pu, isotopic U	Excreta scheduling code
	Isotopic Pu and Sr	Excreta scheduling code
IPS		Excreta scheduling code: Sr tot means radiostrontium by gross beta
IPS IPSA	Isotopic Pu, Sr tot & Am-241	Excreta scheduling code; Sr tot means radiostrontium by gross beta Excreta scheduling code for isotopes of Pu and radiostrontium
IPS IPSA IPSR	Isotopic Pu, Sr tot & Am-241 Seq Pu isotopic Sr-total	Excreta scheduling code for isotopes of Pu and radiostrontium
IPS IPSA	Isotopic Pu, Sr tot & Am-241	

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Code	Description	Comment
IPUL	Low-level isotopic Pu	Pu-238 and Pu-239 using a 10,000-min count
IRA	Radium isotopic	Excreta scheduling code
IR192	Iridium-192	Excreta scheduling code
ISCP	Sequential Sr-90 Ce Pm	Excreta scheduling code
ISPEC	Gamma spectroscopy	Excreta scheduling code
ISR	Sr isotopic	Excreta scheduling code
ITH	Thorium isotopic	Excreta scheduling code
ITPAC	Seq isotopic Pu, Cm & Am-241	Excreta scheduling code
IU	U isotopic	Excreta scheduling code
IUPU	Isotopic plutonium/NU	Excreta scheduling code
K 40	Potassium	
LA140	Lanthanum-140	
LEPD	Low-energy photon detector	Excreta scheduling code for low-energy photon scan
MFP	Mixed fission products	
MN 54	Manganese-54	
MO 99	Molybdenum-99	
NA 22	Sodium-22	
NA 24	Sodium-24	
NAI	Gamma Nal detector	Excreta scheduling code
NB 95	Niobium-95	
NP237	Neptunium-237	
NP239	Neptunium-239	
PB210	Lead-210	
PB212	Lead-212	
PM147	Promethium-147	
PO210	Polonium-210	
PR144	Praseodymium-144	
PU	Plutonium alpha	Total alpha from Pu isotopes after separation
PUMIX	Plutonium alpha	Total alpha from Pu isotopes and Am-241
PU238	Plutonium-238	
PU239	Plutonium-239	When pertaining to excreta samples, it's actually Pu-239+240
PU240	Plutonium-240	
PU241	Plutonium-241	
PU242	Plutonium-242	
QUS	U	Quick Uranium Soluble; excreta scheduling code for elemental U
QUS 1	U	Same as QUS
QUS 2	U	Same as QUS
RA224	Radium-224	
RA225	Radium-225	
RA226	Radium-226	
RA228	Radium-228	
RH106	Rhodium-106	
RND	Radon daughters	
RU103	Ruthenium-103	
RU106	Ruthenium-106	
S 35	Sulfur-35	
SB124	Antimony-124	
SB125	Antimony-125	
SCP	Sequential Sr-total Ce Pm	Excreta scheduling code
55.		

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Code	Description	Comment
SM153	Samarium-153	
SR	Strontium	Total radiostrontium by beta counting
SR 89	Strontium-89	· · · · · · · · · · · · · · · · · · ·
SR 90	Strontium-90	When pertaining to excreta samples, Sr-90 by yttrium ingrowth
TAC	Total actinides	
TC 99	Technetium-99	
TH227	Thorium-227	
TH228	Thorium-228	
TH230	Thorium-230	
TH232	Thorium-232	
TH234	Thorium-234	
TL208	Thallium-208	
U	Elemental uranium	
URAN	Elemental uranium	
U DEP	Depleted uranium	
U NAT	NU	
U 233	Uranium-233	See uranium discussion in text
U 234	Uranium-234	Actually U-234+233, but usually U-234
U 235	Uranium-235	
U 236	Uranium-236	
U 238	Uranium-238	
UMIX	Uranium mix	Total uranium, used for intakes not bioassay
UMS	U-235 U-236 U-238 U-234	
US	U	
XX 0	Isotope will have no result	
ZN 65	Zinc-65	
ZR 95	Zirconium-95	

Table B-2. Sample type codes.

Code	Type of sample
В	Blood
F	Feces
S	Sputum
Т	Tissue
U	Urine

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Table B-3. Bioassay reason codes.

Code	Name	Description
BL	Baseline	Measurement performed to establish reference level against which subsequent measurements will be compared. In general, could be for new employees, or for established employees before commencing work with radioactive materials, beginning specific type of radiation zone work, or making offsite trip where potential intakes could occur.
PR	Periodic	Measurement performed at regularly scheduled interval.
EA	End of Assignment	Measurement performed after completion of specific work assignment, but not end of employment.
SP	Special	Measurement performed as part of specific investigation of potential internal dose. Could include response to off-normal work conditions or follow-up of abnormal periodic measurements.
CR	Contractor Request	Measurement requested by employer for reasons other than periodic, baseline, end of assignment, or special investigation.
RA	Reanalysis A	First reanalysis of sample by taking another aliquot and repeating same radiochemical or chemical analysis.
RB	Reanalysis B	Second reanalysis of sample by taking another aliquot and repeating same radiochemical or chemical analysis.
R1	Recount 1	First recount of original excreta sample or repeat in vivo examination.
R2	Recount 2	Second recount of original excreta sample or repeat in vivo examination.
QR	Quality and Research	Measurement performed as part of quality control, quality assurance, or research work.
TM or TS	Termination	Final bioassay at termination of employment.
12	Contract Work	<i>In vivo</i> measurement performed under contract to customers rather than for Hanford employees.
20	Source Count	<i>In vivo</i> source count made for system calibration or as function check, usually using known check source.
30	Background Count	In vivo system background measurement performed for system calibration or as functional check.

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Table B-4. Excreta sample kit codes^a.

Ki	t code ^b		
D/R	P/U	Media	Sample description
1	Р	Urine	Approximate 24-hr urine collection. Collected at home over 2-d period. Used for routine sampling and when larger volume sample is desired. Designated sample date is day after kit delivery to employee.
2	Q	Urine	Approximate 12-hr urine collection for termination sampling only. Collected at home overnight. Designated sample date is day after date of kit delivery to employee.
3	R	Urine	Total 24-hr urine collection. Collected at home and at work (if necessary) to collect all urine voided during 24-hr period. Generally used for sampling immediately following occurrence or for work restriction sampling. Designated sample date is day after delivery or date on which sample collection began.
4	S	Urine	Single void (spot urine) collection. Collection in single bottle, used for initial indications of intake. Designated sample date is date of voiding.
5	Т	Feces	Collection of single fecal voiding usually for investigation of potential intake. Sample date is day after kit delivery or date on which sample was actually voided.
6	U	Urine	Partial day or approximate 12-hr collection. Usually collected at home overnight. Used for collection following occurrence or when large-volume urine sample is necessary, such as for tritium or uranium determination. Designated sample date is date of delivery to employee.
7	V	Urine	Approximate 12-hr collection Sunday-Monday sample (Friday delivery only). Generally used for workers chronically exposed to soluble uranium. Designated sample date is Sunday in sampling period.
8	W	Urine before 1986. Feces starting in 1986.	Associated with urine sampling in 1950s through 1970s; used to mean undesignated or unknown. Starting in 1986: collection of single fecal voiding used for special program for plutonium oxide workers. Designated sample date for shift workers is Tuesday of long shift change, and for day workers is appropriate Sunday.
9	Х	Urine	Kit designed for collection of urine outside local service area. Transportation handled by private carrier. Generally used for termination samples not collected locally.
A	Y	Urine	Simulated 48-hr urine collection. Collected at home over 4-d period. Used for IPUL (low-level isotopic plutonium) sampling. Designated sample date is 2 d after kit delivery to employee.
В	Not applicable	Urine	12-hr urine collection for termination sampling only. Collected at home overnight. Kit delivered in normal manner, but brought to designated onsite location by worker for pickup by contractor. Designated sample date is day after date of kit delivery to employee. Delivery only, no home pickup required.

a. Before about 1983, kit codes were called collection codes.

b. D/R = Delivery and Retrieval; P/U = Pick-Up only (the latter series of codes were not used before about 1990, but should have no impact on dose reconstruction).

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Table B-5.	In	vivo	body	[,] location	codes.
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Computer code	Body location
ABD	Abdomen
CHT	Chest result
CH1	Chest result
CH2	Chest result corrected by ultrasound measurement of chest wall thickness
HED	Head
HND	Hand
KNE	Knee
LG1	Lung result (chest result corrected for skeleton burden interference)
LG2	Lung result (chest result corrected for skeleton and liver burden interference)
LV1	Liver
LV2	Liver result corrected for skeleton burden interference
LV3	Liver result corrected for skeleton and lung burden interference
LYM	Lymph nodes
SKL	Skull (head) – old code no longer in use
SK1	Total activity in skeleton based on head count
SK2	Skeleton result based on something other than head count
SPL	Special
THX	Thorax
THY	Thyroid
TRT	Throat – old code no longer in use
WBC	Whole body
WND	Wound

Table B-6. Excreta unit codes.

Computer code	Description of units
1	dpm/sample
2	dpm/volume analyzed
3	μg/L until 07-01-82 μg/sample after 07-01-82
4	μg/g until 07-01-82 μg/sample after 07-01-82
5	µCi/sample
6	µCi/L
7	nCi
8	μCi

Table B-7. Excreta processing codes.^a

Processing code	Description
R	Routine processing
Р	Priority processing
Х	Expedite processing (added about 1985)
Ē	Emergency processing

a. Used in conjunction with contract with commercial laboratory starting in 1965. Used to designate turnaround time and MDAs; that is, different processing codes had different MDAs.

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Table B-8. Excreta laboratory codes.

Code	Analytical laboratory		
IT	IT Analytical Services, Richland		
LA	Los Alamos National Laboratory		
OR	Oak Ridge National Laboratory		
PL	PNNL Analytical Chemistry Laboratory		
QN	Quanterra		
RE	REECo (Reynolds Electrical & Engineering Company,		
	Nevada Test Site)		
ST	Severn Trent Laboratories, Richland		
TA	TMA/Norcal, Richmond, California		
WH	Westinghouse Hanford Company, 222-S Laboratory		

Table B-9. Excreta no-sample codes.

No-sample code	Description		
CN	Kit not out. Sample kit not out at time of scheduled pickup.		
CS	Cancelled sample or analysis.		
CT	Sample lost due to bioassay analysis contract termination.		
FA	Failed Analysis. Valid analytical result could not be obtained.		
IS	Insufficient sample. Sample provided by worker but volume		
	insufficient to meet contractual requirements.		
LC	Lost container. Sample kit not retrieved.		
ND	Not delivered. Sample scheduled but kit never delivered.		
NS	No sample. Kit retrieved but no sample provided by worker.		

Table B-10. In vivo invalid result codes.

Code ^a	Reason for no results
С	External contamination other than radon detected on subject. Measurement invalid; no results
	obtained.
F	Failure of equipment or faulty setup of equipment. Measurement invalid; no results obtained.
I	Interference from localized activity in another part of subject's body. Measurement invalid; no results obtained.
L	Location of internal or external activity qualitatively determined by mapping, masking, or collimating. Could include one or more measurement counts. Measurements are qualitative for identifying
	location of activity and do not yield quantifiable estimates of activity.
М	Medically administered radioactivity interfered with measurement. Measurement invalid; no results
	obtained.
Р	Preliminary count, when followed by more quantitative record count. Used to indicate measurement
	taken, but not record count.
R	Radon interference from subject's clothing, hair, or skin. Measurement invalid; no results obtained.
S	Subject's actions interrupted completion of count. Measurement invalid; no results obtained.
Х	Measurement invalid; no results obtained. Other no-result codes do not apply. See comment field for brief description.

a. The comment field might contain a brief explanation in addition to the codes listed.

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Table B-11. INTERTRAC mode-of-intake codes.

Code	Mode of intake	
ABS	Absorption	
ING	Ingestion	
INH	Inhalation	
NON	None (no intake)	
UNK	Unknown	
WND	Wound	

Table B-12. INTERTRAC evaluation reason codes.

Code	Reason for evaluation
A	Annual chronic intake evaluation
С	Contractor-requested evaluation
Н	High routine bioassay evaluation
I	Incident evaluation
N	New hire measurement or previous employment record
	indicated exposure before Hanford employment
R	Reevaluation

Table B-13. INTERTRAC source-of-intake codes.

Code	Source of intake
DHE	Intake at DOE site while employed at Hanford
HAN	Intake at Hanford
NHE	Intake at non-DOE site while employed at Hanford
NOC	Nonoccupational intake
PTH	Intake occurred before Hanford employment

Table B-14. INTERTRAC miscellaneous codes.

Code type	Code	Description
Intake confirmed	Y	Yes
	N	No
Nature of intake	А	Acute
	С	Chronic
Recorded dose	Y	Yes
	N	No
	0	Undetermined - old evaluation assessing body
		burden rather than dose, or evaluation in process
	Z	Recorded dose is 0 mrem
Source known	Y	Yes
	N	No
Type of evaluation	Р	Preliminary
	F	Final

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Table B-15. Special whole-body count resolution codes (RC) (used in 1983 only).

Code	Description
А	Investigation in progress
В	Recent intake < or = to 1% Maximum Permissible Annual Dose
С	Previous deposition
D	Under investigation with additional examinations scheduled
E	Investigation completed, see radiation exposure records
F	Unresolved
G	Deposition from previous non-Hanford employment
Н	Exposure received offsite by Hanford employee
	Activity derived from medical diagnostic or therapeutic procedure

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C.1 PURPOSE

There are instances of energy employees who, for a variety of reasons, were not monitored for internal exposure during the course of their employment at a DOE facility. In addition, there are incidents in which an employee's monitoring records are incomplete or unavailable. In such cases, data from coworkers can be used to approximate an individual's possible exposure. The purpose of this attachment is to provide the details of the calculation and assignment of intakes based on coworker data from the Hanford site (including PNNL) for the purpose of estimating unmonitored exposures. Many of the periods that are addressed by this coworker analysis include those that are covered by the groups of Hanford workers that have been included in the SEC. The information provided in this attachment for those periods can be used when dose reconstruction is required outside the scope of the SEC inclusions.

C.2 OVERVIEW

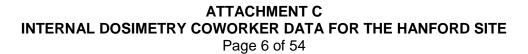
ORAUT-OTIB-0019, *Analysis of Coworker Bioassay Data for Internal Dose Assignment* (ORAUT 2005), describes the general process for analyzing bioassay data for the assignment of doses based on coworker results.

Bioassay results for Hanford were obtained as a copy of the REX database sent to the NIOSH Health-Related Energy Research Branch (HERB) before the start of the dose reconstruction project. The REX data necessary to generate the bioassay statistics for the coworker data project were converted to another database program. Data in the converted tables were spot-checked against REX at Hanford, and no discrepancies were found.

To determine the completeness of the REX database, it was compared with statistics from monthly reports that were prepared by Hanford for 1946 through 1959 (where records were available). REX was found to contain data for over 90% of the plutonium bioassay samples that were indicated as collected in the monthly reports. For uranium, REX contained data for approximately 77% of the samples. Figures C-1 and C-2 depict the bioassay sample data available from each source for plutonium and uranium, respectively. Table C-39 contains the SRDB reference numbers for the monthly reports that were used for this comparison.

As is typical of a site with large operating reactors and spent fuel dissolution operations, there was potential for exposure at Hanford to many radionuclides including fission and activation products, plutonium, uranium, and tritium. In addition, Hanford handled thorium for part of its history. With the advent of *in vivo* counting, detection of a wide range of gamma-emitting fission and activation products became possible. In addition, bioassay monitoring of workers on small-scale or short-term projects for unusual radionuclides was performed at different times. Therefore, there are a number of different radionuclides in the excreta and *in vivo* data. For most of the radionuclides in the database, there were too few measurements to allow reliable statistical evaluation. Exposure to such radionuclides was arguably rare or limited to only a few workers on a specific project and would not be appropriate for this general coworker study.

The urinalysis data that were chosen for this coworker study because of general applicability and number of measurements were from plutonium, uranium, radiostrontium, and promethium analyses,



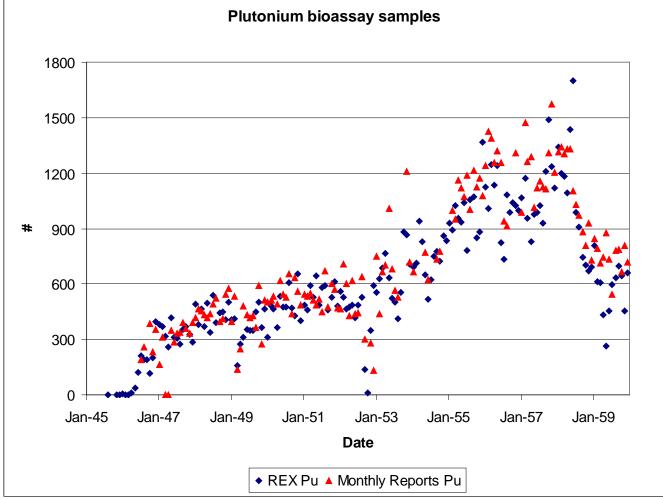


Figure C-1. Plutonium bioassay data in REX and from Hanford monthly reports.

although the ¹⁴⁷Pm intakes have limited applicability (Sections C.3.1.4 and C.5.1.4). Before whole-body counting became routine in 1960, bioassay monitoring for intakes of some fission products was performed using a radiochemical procedure called "fission product urinalysis." This procedure involved chemical separation and counting (gross beta) of rare-earth fission products and radioisotopes of strontium. However, the results are difficult to interpret because the actual radionuclide or composition of the mixture of radionuclides in the urine was not determined and different radionuclides had different chemical yields and detection efficiencies. Therefore, intakes of ²⁴Na, ⁶⁵Zn, ⁹⁰Sr, and ¹³⁷Cs were extrapolated from periods when they were measured, and other fission products were accounted for using air concentration limits (Section C.5).

With the exception of naturally-occurring ⁴⁰K, only three gamma-emitting radionuclides that were measured by whole-body counting were routinely reported: ¹³⁷Cs, ²⁴Na, and ⁶⁵Zn. The available documentation does not state the reasons for choosing these three radionuclides. Other fission or activation products were reported only when detected above certain levels of significance that were determined through different methods at different times. A review of the *in vivo* database showed that there was enough data to warrant statistical evaluation only for these three routinely reported radionuclides.

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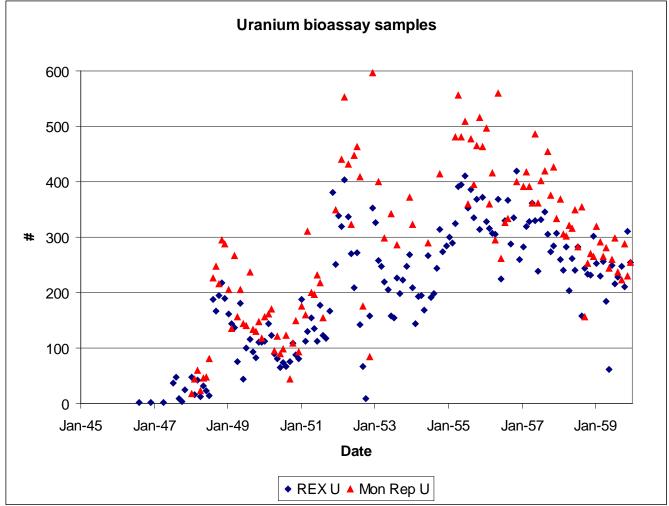


Figure C-2. Uranium bioassay data in REX and from Hanford monthly reports.

Dose from intakes of tritium was treated as external dose until about 1986 and reported as part of penetrating dose in the radiological records. Dose from tritium was included in the coworker analysis of penetrating external dose and was therefore not duplicated in this coworker analysis. The completeness of those records was subsequently evaluated as discussed in Section C.3.3. [The coworker external analysis is in Attachment B of ORAUT-TKBS-0006-6, *Hanford Site – Occupational External Dose* (ORAUT 2010b).]

The statistical analyses of the bioassay data for each radionuclide were performed in accordance with ORAUT (2005). The resultant values were input to the IMBA computer program, and a fit to the data for each of the seven radionuclides at the 50th- and 84th-percentile values was performed to obtain intake rates for assignment of dose distributions.

C.3 DATA

C.3.1 <u>Selected Bioassay Data</u>

Data for each of the database codes that were considered were extracted from a series of files that contained a version of the HERB dataset.

Throughout most of Hanford's history, bioassay data were truncated or censored below some level. Various terms can be applied to such a level, such as *detection limit*, *less-than level*, *reporting level*, *lower limit of detection, decision level*, or *MDA*. In this attachment, *reporting level* denotes the level below which a measurement was not recorded. *Decision level* and *MDA*, as best as could be determined from available documentation, are used in accordance with the definitions in Health Physics Society Standard N13.30 (HPS 1996). The analysis established time intervals for which single values for the 50th- and 84th-percentile values were calculated. These intervals were determined such that there was a sufficient total number of measurements and a sufficient number of measurements greater than the reporting level to perform meaningful statistical analysis.

For some data, reporting levels were not used for all samples, and there was therefore a large batch of samples at or just above the reporting level, a smattering of samples below the reporting level, and some samples with zero, blank, or null values. This mix of reporting practices led to poor curve fits when values at the reporting level were treated as zeros. Therefore, this analysis used the linear distribution (ORAUT 2005) for values that were considered to represent nondetections. The following radionuclide-specific sections discuss exceptions to this treatment.

C.3.1.1 Plutonium Urinalysis

Routine urinalysis for plutonium at Hanford began in 1946. Over the years, several factors in the methods for measuring and reporting plutonium urinalysis data changed:

- Chemical method,
- Radioactivity counting method,
- Reporting units,
- Reporting levels and data censoring (that is, not reported below a certain activity),
- Method for indicating data below the reporting level, and
- Consistency of use of the previous two protocols (the data for some years show that the reporting level was not used consistently and/or that different methods of reporting data below the reporting level were used).

Some of these changes affected how the urinalysis data were treated for the statistical analysis. The issues and resolutions are discussed in the following paragraphs.

From 1946 to the third quarter of 1983, plutonium urinalysis results were measured and recorded as a gross alpha count on a chemically separated sample. The measured result was the total activity of ²³⁸Pu, ²³⁹Pu, and ²⁴⁰Pu but did not include ²⁴¹Pu or ²⁴¹Am. Since the fourth quarter of 1983, the

chemically separated sample was measured by alpha spectrometry (for routine and priority samples). The recorded results were a value for ²³⁹Pu that was actually the total activity of ²³⁹Pu and ²⁴⁰Pu and a value for the ²³⁸Pu activity. The statistical analyses were conducted on the total plutonium alpha activity for 1946 to September 1983 and on the ²³⁹⁺²⁴⁰Pu activity for October 1983 to 1988. Because most plutonium exposures at Hanford would have involved a mixture of isotopes, the results of these different measurement techniques were normalized (Section C.4).

Routine plutonium urinalysis samples were usually simulated 24-hour samples, which were collected over two 12-hour periods (usually consecutive evenings). Some samples were not 24-hour samples, especially those collected shortly after a suspected acute intake. The collected volume of urine was part of the record starting irregularly in 1958 and consistently in 1959, so the 1959 to 1988 results with volumes of less than 400 mL were not used in the statistics. This criterion removed some of the incident samples and some samples from persons who did not follow instructions and provided less than simulated 24-hour samples.

Incident samples were problematic for performing group statistical analysis. Hanford generally collected many samples from a worker involved with a known large acute intake, which were referred to as special or follow-up samples. The number of samples varied dependent on the severity of the intake, whether treatment was involved, the willingness of the exposed worker, and general internal dosimetry practices at the time. However, in many cases the number of follow-up samples ranged from tens to hundreds. These samples were not comparable to routine annual or semiannual samples from the majority of the workforce, and they tended to bias the statistics on the high side. It was too difficult to remove all of these samples from the database, and minor acute intakes were frequent enough at Hanford that they could represent the normal course of exposure to plutonium workers. However:

- If there were a large number of samples from a single worker, usually bunched after a specific time, these were removed. A specific number was not rigorously applied as the definition of large, but it was generally greater than about 20 in 1 year. In addition, once a person was selected to be removed, data for subsequent years in that worker's history were removed until the excretion was near or less than the reporting level.
- If a record of an acute intake was judged to be unrepresentative, that record was removed. Any intake that was treated by diethylenetriaminepentaacetic acid (DTPA) met this criteria because DTPA-treated urinary excretion is enhanced. Intake through a wound or the skin was considered unrepresentative. Inhalations, however, were considered representative unless they were caused by an event that was judged to be unrepresentative, such as explosion, fire, or major contamination spread.

Another challenge to data interpretation was results listed as "PU" or "PU239" with blank quantities. For most years, the meaning of a blank was not found in documentation. However, the meaning could be inferred from the number of blank results and how they fit with the other data for the year. For instance, there were many blank results each year in the early years, and it was inferred that the blanks meant a sample was analyzed and nothing was detected. In later years there were many sample results that were recorded exactly at the recording level but only a few blanks. It was favorable to claimants to leave out the blanks in those years under the assumption that a blank was a sample collected for which the analysis failed in some way. Each year's distribution of sample results was scrutinized, and instructions were written on how to disposition blank results on a year-by-year basis.

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The method that was used to indicate no detection of plutonium, similar to the issue of blanks, varied over time and appeared not to be consistent even during a given period. For instance, zero, blank, and the reporting level were apparently used to indicate nondetections during some periods. In general, Hanford used reporting levels for plutonium urinalysis results until September 1981. Whether a value that was recorded at the reporting level meant no detection, or whether the reporting level or higher was a true detection and nondetections were recorded as zero or blank, was indeterminate at the broad level.

The meanings of the reporting level, zero, and blanks had to be determined on a year-by-year basis, and the decision was included in the specific instructions on how to rank such sample results. For example, the 1966 to 1974 reporting level was $1.1 \times 10^{-8} \mu \text{Ci/sample}$ (0.244 dpm/sample) and the 1975 to 1981 reporting level was 0.025 dpm/sample. These numbers were considered to be one-half of the detection level and were used to indicate no detection of plutonium. The linear distribution was used to distribute and rank blanks, zeros, and values at these reporting levels. For other years, however, the reporting level was less apparent because there were many values that were probably a reporting level, but other values were less than the reporting level. The latter samples were included in the linear fit, and the apparent reporting level was set as the top of the linear distribution.

1946	3.6E-7 µCi/sample
1947–1948	3.0E-7 µCi/sample
1949–1951	1.5E-7 µCi/sample
1952	8.1E-8 µCi/sample
1953–1957	2.3E-8 µCi/sample
1958–1965	1.2E-8 µCi/sample
1966–1974	1.1E-8 µCi/sample
1975–Sep. 9, 1981	0.025 dpm/sample

The following were set as the top of the linear distribution:

On September 10, 1981, the recording practice changed from recording 0.025 dpm/sample to indicate a nondetection to recording the exact result as measured. After September 9, 1981, therefore, any result greater than zero was used as recorded. Zero and negative results were included in the ranking of samples but not in the fitting of the line. The linear distribution was not used for data after September 9, 1981.

Units for plutonium analyses in the Hanford records were microcuries per sample from 1946 to 1974 and disintegrations per minute per sample from 1975 to present. The earlier values were converted to disintegrations per minute before statistical analysis.

The time interval for separate statistical analyses varied. The choice of the interval was based on the number of results versus the number of results over the reporting level. If there were several hundred sample results and 10% or more exceeded the reporting level, the basic time interval was guarterly. Table C-1 lists the intervals. The unusual division of the third and fourth guarters of 1981 was applied because of the change in recording practice that is discussed above.

C.3.1.2 **Uranium Urinalysis**

Routine urinalysis for uranium began in 1947, but the results were considered unreliable until improvements in the procedure were made (Healy 1948). The 1947 data were analyzed but not used for intake modeling. Data analyzed for this study were from elemental analysis procedures. Starting

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Table C-1.	Time intervals for statistical
analysis of	plutonium urine samples.

Calendar year	Analysis interval
1946	Year
1947	Year
1948	Half years
1949	Year
1950	Year
1951	Year
1952	Year
1953	Half years
1954–Jun. 1981	Quarter years
3Q 1981	Jul. 1–Sep. 9
4Q 1981	Sep. 10–Dec. 31
1982–1988	Quarter years

in 1983 an alpha spectrometry procedure has been used for some workers, but these data were less robust and were not representative of the overall Hanford workforce. The results were recorded as micrograms per liter from 1947 to July 1, 1982, and as micrograms per sample from July 2, 1982, to present. The latter were converted to micrograms per liter using the sample volume that was recorded as part of the information in the database [125]. Elemental uranium samples were not usually 24-hour samples; a mix of sample collection periods was used from 1947 to 1988 with overnight sampling being one of the more frequent methods. Therefore, samples of less than 400 mL were not removed from the uranium data.

In general, Hanford did not have many large, acute intakes of uranium; only six samples were removed because they were collected after unusual acute intakes (e.g., a worker fell into a vat of uranyl nitrate) [126].

A small number of duplicate entries were found in the records from 1965 and 1968 that were true duplicates rather than, for instance, two analyses of the same sample; one of each pair was removed from the data before statistical analysis.

Reporting levels were:

1948–1974	4.0 μg/L
1975–1881	0.4 μg/L
1982 to present	Actual values were recorded as measured

Use of the reporting levels for uranium urinalysis appears to have been more consistent than was the case for plutonium, with the exception of one period. From 1958 to 1966 there were many results lower than 4 μ g/L. It is not known if these indicated detection (i.e., whether the MDA was really reduced during this period). Therefore, these were treated as less-than results and included in the linear distribution. In 1967, Hanford returned to consistent use of the 4- μ g/L reporting level.

Through 1981, zeros, blanks, or less-than results were treated as nondetections and included in the linear distribution. Starting in 1982:

• No reporting level was used.

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- The linear distribution was not used in the statistical analysis.
- Blanks were considered invalid samples and not used in the statistical analysis.
- Zero and negative numbers were included in the ranking of samples but not in the fitting of the line.

Table C-2 lists the time intervals for separate statistical analyses. The PUREX and UO₃ plants were shut down during the 1970s, so there were few exposures and few samples (ORAUT 2010a). There were only 65 samples collected in 1973; the results were comparable to neighboring years, so the 1973 statistical parameters were included.

Calendar year	Analysis interval
1947	Year
1948	Half years
1949–1969	Quarter years
1970–1983	Year
1984–1988	Quarter years

Table C-2. Time intervals for statistical analysis of uranium urine samples.

C.3.1.3 **Strontium Urinalysis**

Specific urinalysis for radiostrontium began in 1965. Before that, the fission product urinalysis method was used to monitor for radiostrontium and some other fission product radionuclides. Both ⁸⁹Sr and ⁹⁰Sr could have been counted depending on whether the sample was beta-counted before or after a period for ingrowth of ⁹⁰Y. For this study, all the results were assumed to be ⁹⁰Sr because it was generally more prevalent and because it produces higher (more favorable to claimants) doses per unit intake than ⁸⁹Sr. Results were recorded as microcuries per liter for 1965 to 1974 and as disintegrations per minute per sample from 1975 to the present. The earlier results were converted to disintegrations per minute per sample before statistical analysis using the recorded volume of the sample.

Most strontium urine samples were simulated 24-hour samples, so samples that were less than 400 mL were removed from the data before statistical analysis. Thirteen sample results were removed because they were follow-up samples to known large acute intakes; one sample result was removed because it was a recount. There were some obvious duplicates that were removed.

Reporting levels were:

1965–1969	1.67E-5 μCi//L
1970–1974	1.00E-6 μCi/L
1975–Mar. 1979	2 dpm/sample
Apr. 1979–1981	5 dpm/sample
1982–1988	Actual values were recorded as measured

Blanks with volumes greater than 400 mL were rare; these were considered invalid and were not used. Through 1981, zero or less-than results were treated as nondetections and included in the linear distribution. Starting in 1982:

• No reporting level was used.

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- The linear distribution was not used in this analysis.
- Zero and negative numbers were included in the ranking of samples but not in the fitting of the • line.

Table C-3 lists the time intervals for separate statistical analyses. Before 1970 there were few samples in a given year and very few samples that exceeded the reporting level, so samples were arouped into two 3-year intervals. Before 1979, in general, there were only a few hundred (and sometimes less than a hundred) samples in a year. Whole-body counting was the primary bioassay method for fission products; the exception was for the workers at the WESF where exposure to pure ⁹⁰Sr was possible. The data from 1979 were split into two periods because of the change in reporting level from 2 to 5 dpm/sample on April 1.

analysis of Stunne samples.				
Analysis interval				
As one group				
As one group				
Years				
Quarter				
As one group				
Years				
Quarters				

	Time intervals for statistical
analysis of	⁹⁰ Sr urine samples.

C.3.1.4 **Promethium Urinalysis**

Promethium-147 is a fission product with greater abundance in 1-year-aged weapons-grade fuel (1 year since removal from the reactor) than either ⁹⁰Sr or ¹³⁷Cs. However, it behaves in the body somewhat similarly to ⁹⁰Sr and had an early MPBB that was a factor of 30 less than that for ⁹⁰Sr (ICRP 1959) and, later, an annual limit on intake that was a factor of 14 less than that for ⁹⁰Sr (ICRP 1979, 1982). The presence of ¹⁴⁷Pm would have been detected as part of the fission product urinalysis even though the results of the fission product urinalysis were calibrated for and interpreted as ⁹⁰Sr. When whole-body counting and ⁹⁰Sr urinalysis replaced the fission product urinalysis as the bioassay method of choice for fission products, ¹⁴⁷Pm was considered an insignificant contributor to internal dose and ¹⁴⁷Pm bioassay for workers exposed to general fission products was not performed. Promethium-147 as an unconcentrated fission product in general contamination was treated in this analysis as a miscellaneous fission product (Section C.5.8).

Specific bioassay for ¹⁴⁷Pm was initiated when PNNL began manufacturing ¹⁴⁷Pm heat sources in the 325 Building in 1966. The number of workers in the bioassay program and the number of samples were small in comparison to the numbers for the plutonium, uranium, or strontium bioassays, with a high of 65 workers in 1968. There was almost no sampling from 1972 to 1975, which probably indicates a cessation of the original heat source program, and only 20 workers were sampled from 1976 to 1979. The exact end date of the heat source program has not been determined. For purposes of this study, it was assumed that exposure occurred from 1966 to 1979, although it is likely that any exposure from 1972 to 1975 was only due to residual activity.

Results were recorded as microcuries per liter from 1966 to 1974 and as disintegrations per minute per sample from 1975 to 1979. The earlier results were converted to disintegrations per minute per sample using the sample volume.

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Most routine samples were simulated 24-hour samples, so samples with volumes smaller than 400 mL were removed from the data before statistical analysis. There were a large number of duplicate entries in the database from 1966 to 1968, and these were removed. There was a glovebox rupture and large contamination spread that mainly affected three workers in 1967, and an overpressurization and blowback of a sampling line affected another worker in 1968; samples that were associated with these incidents were removed [127].

Reporting levels were:

1966–Mar. 25, 1970	1.67E-5 µCi/L
Mar. 31, 1970–1974	1.00E-5 µCi/L
1975–1979	25 dpm/sample

The linear distribution was applied to results at or below the reporting levels. There were no blanks with volumes greater than 400 mL.

Table C-4 lists the time intervals for separate statistical analyses. The split in 1970 occurred because of the change in reporting level on March 31.

	ii anaiysis ol i Fill unne samples.
Calendar year	Analysis interval
1966, 1967	As one group
1968, 1969, 1970 through March 25	As one group
Mar. 31–Dec. 31, 1970; 1971	As one group
1972–1974	Not enough samples for statistics
1975, 1976, 1977, 1978, 1979	As one group

Table C-4. Time intervals for statistical analysis of ¹⁴⁷Pm urine samples.

C.3.1.5 Zinc-65 in Whole-Body Counts

Zinc-65 was routinely reported in whole-body counts from 1960 to 1983. There were only about 370 recordings of ⁶⁵Zn in whole-body counts in 1984, but that was considered sufficient to include in the statistical analysis. After 1984 there were very few detections of ⁶⁵Zn in whole-body counts. Zinc-65 was mostly a concern during the operation of the once-through-cooled reactors (through 1971) when large activities of ⁶⁵Zn were discharged to the Columbia River.

The three largest local cities drew their sanitary water mostly from the Columbia River, as did the reactors on the site. Brady (1964) shows that ⁶⁵Zn passed through the purification systems in sufficient concentration to produce measurable body burdens in many Hanford workers. Brady also attributed some of the ⁶⁵Zn to ingestion of fish from the Columbia River or from crops irrigated by Columbia River water.

Figure C-35 shows a decrease in the median body burden of ⁶⁵Zn starting in 1972 (with the exception of a spike in body burden in the fourth quarter of 1976), which appears to show the effect of the shutdown of the once-through-cooled reactors. However, it was apparent from the distribution of measured activities in the workers that inhalation in the workplace also had to be a viable intake pathway; ingestion of water cannot explain the higher body burdens at the higher percentile values of the quarterly distributions. Therefore, both inhalation and ingestion pathways were modeled.

In whole-body counting, the decision level varies with every individual and every count because of the interperson variability in activity of ⁴⁰K, the intraperson variability in activity of interfering radionuclides

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a person could have on them on any given day (especially radon and thoron progeny), and the day-to-day variability in the natural background in the counting cell. In addition and especially during the early years of the *in vivo* counting program, there were improvements to the program including type or number of detectors, counting times, and improved electronics. It was common for more than one whole-body counting system to be used at a time. Therefore, the stated detection limits were approximate and were meant to apply to a general range; little documentation on detection levels has been found. A single reporting level is not apparent in the recorded data. All whole-body counts were recorded in nanocuries.

Because of these factors, it was impossible to state a distinct threshold value above which all results meant detection and below which all results meant nondetection. For 1960 to 1974, a detection level of 0.8 nCi for ⁶⁵Zn was found in the documentation. Relative to the distribution of results in most years, this value was a reasonable one to use as the reporting level. Blanks and zero results were distributed using the linear distribution with 0.8 nCi as the upper range; values between zero and 0.79 nCi were used as recorded and mixed in with the numbers from the distribution.

For 1975 to 1984, the value of 0.75 nCi was distinctly used as the reporting level, with only one value that fell between zero and 0.75 nCi. Blanks and zeros were distributed using the linear distribution.

Table C-5 lists the time intervals for separate statistical analyses.

analysis of ⁶⁵ Zn whole-body counts.				
Calendar year	Analysis interval			
1960	Year			
1961-1962	Half years			
1963–1974	Quarter years			
1975	Half years			
1976–1983	Quarter years			
1984	Year			

Table C-5. Time intervals for statistical

C.3.1.6 Sodium-24 in Whole-Body Counts

Similar to ⁶⁵Zn, ²⁴Na was reported routinely from 1960 to 1983 with a significant but much smaller number of results in 1984; after 1984 there were very few recorded results. Sodium-24 was released to the Columbia River from the once-through-cooled reactors, but its short half-life (15 hours) prevented significant concentrations in local municipal drinking water systems. Its presence was correlated with reactor workers and was to some extent believed to be due to drinking water at the reactors (Brady 1964). However, as with ⁶⁵Zn there were body burdens that were too high to be explained by ingestion of drinking water. As a consequence, ²⁴Na was modeled using both inhalation and ingestion pathways.

As explained in the ⁶⁵Zn section, a distinct threshold value for detection was not apparent from the data. For 1960 the lowest reported value was 0.56 nCi, so this value was used as the reporting level. Blanks and zero results were distributed using the linear distribution with 0.56 nCi as the upper range. For 1961 to 1974, a detection level of 0.3 nCi for ²⁴Na was found in the documentation. This value was reasonable for use as the reporting level even though there were many results recorded at smaller values. Blanks and zero results were distributed using the linear distribution with 0.3 nCi as the upper range. Values between zero and 0.29 nCi were used as recorded and mixed with the

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numbers from the distribution. For 1975 to 1984, a value of 0.31 nCi was distinctly being used as the reporting level with no measurements recorded lower than 0.31 nCi. Blanks and zeros were distributed using the distribution with 0.3 nCi as the upper range.

Table C-6 lists the time intervals for separate statistical analyses.

analysis of - Na whole-body counts.				
Calendar year	Analysis interval			
1960	Year			
1961–1962	Half years			
1963–1983	Quarter years			
1984	Half years			

Table C-6.	Time	int	er	/als	for	statistical
	24					

C.3.1.7 Cesium-137 in Whole-Body Counts

Cesium-137 was recorded routinely with every whole-body count from 1960 to 1983. For 1984 to 1988, ¹³⁷Cs was generally recorded only if it was detected above a reporting level, but the database also shows valid whole-body counts including blank values for ¹³⁷Cs. Why this occurred is not known. Because of its long half-life and abundance in the waste from the separations plants, ¹³⁷Cs has long been considered one of the principal contaminants at Hanford and has often been used as an indicator of potential intakes. The linear distribution was not used to evaluate the cesium-137 data except for the period of 1984 through 1988.

Table C-7 lists the time intervals for separate statistical analyses.

whole-body counts.					
Calendar year Analysis interval					
1960–1974	Quarter years				
1975	Only two values reported and neither above the reporting level. This year was skipped.				
1976–1983	Quarter years				
1984–1988	Half years				

Table C-7. Time intervals for statistical analysis of ¹³⁷Cs whole-body counts.

From 1984 to June 1986, ¹³⁷Cs results were recorded differently than in other years. When ¹³⁷Cs was not detected, the radionuclide was sometimes listed in the results as a blank but, more often, ¹³⁷Cs was not listed at all. Instead, it was treated like the many other fission product radionuclides that could be detected by *in vivo* counting but are not listed if they are not detected. For this period the total number of whole-body counts was determined by tallying the number of ⁴⁰K counts, which were recorded for every valid whole-body count. Each whole-body count for which ¹³⁷Cs was not listed was considered the same as a whole-body count with a blank or zero result for ¹³⁷Cs.

The numbers of results recorded as greater than the reporting level (0.65 nCi for 1984 to September 19, 1986, and 3 nCi for September 22, 1986, to 1988) were very small (generally less than 2% of the total counts during each half-year period). The rank-only method forces a fit to these small numbers of detected results and extrapolates the fits to the almost 4,000 counts in each half-year period that had no detection. Therefore, this analysis used the linear distribution for 1984 to 1988 because it provides better fits (visually and by the R^2 values).

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C.3.2 Analysis

For each of the seven radionuclides, a lognormal distribution for the data in the intervals in Tables C-1 to C-7 was assumed. The 50th- and 84th-percentile values were calculated using the method in ORAUT (2005). Tables C-33 to C-39 show the statistical analysis results for plutonium, uranium, ⁹⁰Sr, ¹⁴⁷Pm, ⁶⁵Zn, ²⁴Na, and ¹³⁷Cs.

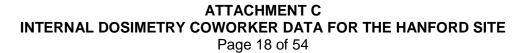
C.3.3 Tritium Dose Evaluation

The tritium doses in the main text of this document are based on calculated summary doses in the Hanford external dosimetry records. However, bioassay data are typically preferred for dose reconstruction over summary doses that are calculated contemporaneously with the exposure because assumptions for this project are not necessarily consistent with those of a radiation protection program.

In 2007, several boxes of tritium data were identified that appeared to contain tritium urinalysis data from 108-B workers for the late 1940s and 1950s. Additional boxes were subsequently identified and expanded the period into the 1960s. In 2008, the contents of these boxes became available to the ORAU Team. The hard-copy records were entered into a database to permit analysis. The calculated doses from these data can be compared to the summary dose data that was previously the only available data. The objective of this comparison was to determine whether the tritium doses to be assigned to unmonitored workers as provided in the main text of this document (Section 5.6) are believed to be favorable to claimants. As a practical matter, the objective became whether there is any reason to believe that they are not favorable given the limitations of the comparisons that can be performed.

Figure C-3 summarizes the data. Table C-40 contains a list of the SRDB reference numbers for the hard-copy tritium bioassay data. Table C-39 contains a list of the SRDB reference numbers for the monthly reports.

The number of bioassay samples in the hard-copy records follows the trend of the number of tritium bioassay samples that were reported as being taken in the Hanford monthly reports for most years for which a comparison could be made except 1961 through 1964. No bioassay data was found for 1961 through 1963 and very little for 1964. There does appear to be a gap in the hard-copy records for 1961 through 1964 and 1966 where the number of workers with reported doses in the external dosimetry database exceeds the number for which hard-copy records were found. In addition, the monthly report data is known to be incomplete because the reports for all months in the timeframe of interest are not available. Annual tritium bioassay totals or reports with tritium bioassay information for all months in a given year are available only for 1950 and 1963. The tritium bioassay in the hardcopy records exceeded that reported in the available monthly reports for 1949 through 1952, 1954, and 1967. However, the tritium bioassay in the hard-copy records are also known to be incomplete based on known instances where the retrieved bioassay data are less than that reported in the incomplete monthly reports, especially 1961 through 1964 and 1966. An additional complicating factor is that the monthly reports track analyzed bioassay samples rather than collected samples, which might introduce an offset into any comparison because the tritium bioassay in the hard-copy records is recorded based on the sample collection date.



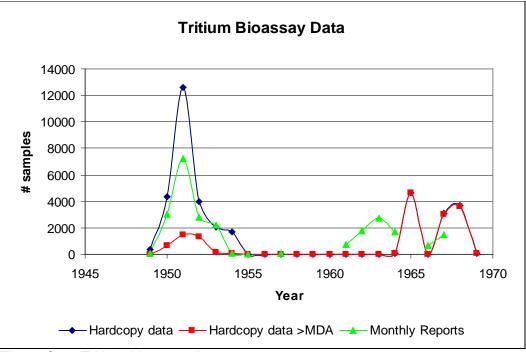


Figure C-3. Tritium bioassay data.

The number of individuals with recorded doses in the REX database was compared to the number of individuals for whom doses were calculated based on the hard-copy records. In addition, the doses were compared on an individual-specific basis where dose data for an individual was present in both the REX data and in the hard-copy records (Figure C-4). Relatively good success was achieved in matching individuals in both datasets. The number of individuals with data in both datasets closely tracked the minimum number of individuals in either of the data sources for a given year.

Using an updated version of a tool for calculation of coworker doses from tritium bioassay for the Mound and LANL coworker studies, tritium doses were calculated for each worker and year for which data was available. The doses by individual were compared on an annual basis to determine if a correlation between the main text (REX) values and the dose based on the hard-copy records could be determined. The most relevant comparison of the two data sources is the GM of the annual doses for the individuals in each dataset for each year because this is the actual dose to be used for unmonitored workers. The distributions of annual doses for all individuals in a given year were fitted to a lognormal distribution for each year. Figure C-5 depicts the comparison of the calculated GM doses from these two data sources. As can be seen, the values in the main text are higher in all but 1 year than the values calculated from the hard-copy records. The values are similar in magnitude and generally higher than the doses based on the hard-copy records as well.

The doses that would be assigned by either method are not significant at less than 200 mrem/yr in all years and generally much lower. Given that the values in the main text are favorable to the claimant in comparison with the values that were calculated from the hard-copy records, and that they cover more years than those values, the values assigned in the main text are deemed more appropriate for assignment of coworker doses.



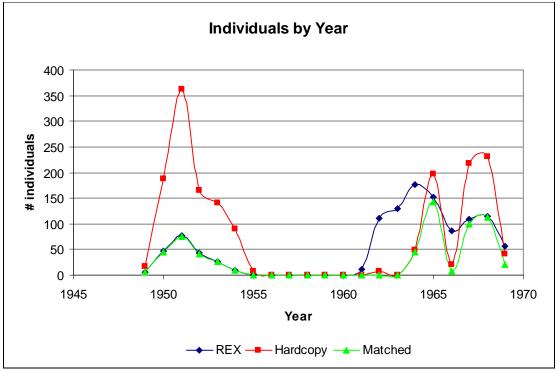


Figure C-4. Individual tritium bioassay or dose data by year.

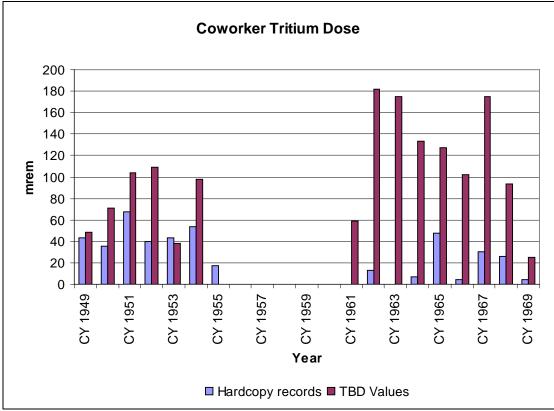


Figure C-5. Tritium dose from hard-copy records and the main text (Section 5.6).

C.4 INTAKE MODELING

C.4.1 <u>Assumptions</u>

All urinalysis results were assumed to be representative of a full-day (24-hour) urinary excretion or were either eliminated or normalized to be representative of a full day before creating the lognormal plots [128]. Each result that was 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. A chronic exposure pattern was assumed, even though this is unlikely for most exposures at Hanford, because it approximates a series of acute intakes with unknown dates. Intakes were assumed to be from inhalation using a default breathing rate of 1.2 m³/hr and a 5-µm activity median aerodynamic diameter particle size distribution unless otherwise specified (NIOSH 2002). Intakes of ⁶⁵Zn and ²⁴Na were determined using whole-body counting results. As discussed in Sections C.3.1.5 and C.3.1.6, both inhalation and ingestion intakes were possible, so both were modeled.

The results for ¹⁴⁷Pm and ⁹⁰Sr urinalysis were taken directly from the statistical analysis of the database, as were the results for ⁶⁵Zn, ²⁴Na, and ¹³⁷Cs whole-body counting.

For plutonium, ²³⁹Pu was used for the IMBA intake modeling, but the database results were presumed to be gross plutonium alpha measurements through the third quarter of 1983. To convert from gross plutonium alpha to ²³⁹Pu, fresh fuel-grade plutonium was assumed for 1945 to 1949, 5-year-aged fuel-grade plutonium was assumed for 1950 to 1954, and 10-year-aged fuel-grade plutonium was assumed for 1955 and after [129]. Table C-8 lists the percentages of ²³⁹Pu that were used. Starting in October 1, 1983, alpha spectrometry was used in the analysis to specifically identify the ²³⁹Pu; therefore, no correction factor was used.

Period	Plutonium grade	Percentage ²³⁹ Pu of plutonium alpha emitters
1946–1949	Fresh fuel-grade	82.4%
1950–1954	5-yr-aged fuel-grade	83.0%
1955-09/30/1983	10-yr-aged fuel-grade	83.5%
10/01/83-present	10-yr-aged fuel-grade	100% (isotopic analysis)

Table C-8. Plutonium mixture usage.

For uranium, because the IMBA program requires urine results in units of activity per day, the total uranium values in micrograms per liter were multiplied by 1.4 to normalize them to the Reference Man excretion rate of 1,400 mL/d. Because a variety of enrichments was possible, the intake modeling used mass concentration units and ²³⁴U was assumed for all the IMBA intake modeling as it has the most favorable to claimant dose conversion factors (ICRP 1994) [130]. This did not affect the data fits for intake determination (i.e., the same total intakes would be obtained for any enrichment that was assumed) because all uranium isotopes have the same biokinetic behavior and the isotopes in this analysis have long half-lives in relation to the assumed intake period.

To convert to units of activity for dose calculation, the 0.7054 pCi/µg specific activity of NU was assumed through 1952, and the 0.9099 pCi/µg specific activity of RU was used for later years (see the main text). Like the intake modeling, ²³⁴U was used to calculate the doses. The ICRP Publication 68 dose coefficients (also referred to as dose conversion factors) for ²³⁴U are 7% to 31% larger than those for ²³⁵U, ²³⁶U, and ²³⁸U (ICRP 1995). Because of the isotopic compositions of the source terms, the ²³⁴U dose conversion factor yields doses that are favorable to claimants.

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C.4.2 Bioassay Fitting

The IMBA computer program was used to fit the bioassay results to a series of chronic inhalation or ingestion intakes. The exception is that one acute intake was used in the modeling of ²⁴Na exposure because that resulted in the best fit. Modeling was performed only for intervals when data were available. Having been based on whole-body counts, the data for ⁶⁵Zn and ²⁴Na were available for 1960 to 1984. The least amount of available data was for ¹⁴⁷Pm from 1966 to 1979. Plutonium data was from 1946 to 1988, uranium data was from 1948 to 1988, and ⁹⁰Sr data was from 1965 to 1988.

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.

C.4.3 Radionuclides and Material Types

For each considered radionuclide or group of radionuclides, the bioassay results were entered into IMBA with assumed material types in terms of lung absorption type or uptake factor from the gastrointestinal tract. The types were chosen to be consistent with ICRP Publication 68 (ICRP 1995) and the main text of this document. The figures at the end of this attachment show the resultant 50th-percentile intakes as plots. The annual bioassay data that were used in the fits are shown as solid blue dots (•) (dark dots when printed in black and white), and data that were not used in the fits are shown as red dots (•) (light dots when printed).

The type S compounds at Hanford have very long radiological half-lives, and the materials are retained in the body for long periods. Therefore, the excretion results for different chronic intake periods are not independent for type S materials. 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 Hanford for relatively short periods, each chronic intake of type S material was independently fit using only the bioassay results from the single intake period. This same process was used for type M plutonium and ¹⁴⁷Pm as well. This method likely results in an overestimate of intakes, particularly for assumed type S exposures that extend through multiple assumed intake periods.

C.4.3.1 Plutonium

Both type M and S materials were present at Hanford and were common enough to apply to general workers, therefore, both types were modeled. This section provides intakes for ²³⁹⁺²⁴⁰Pu of both types. Dose reconstructions must also include intakes of other plutonium isotopes and ²⁴¹Am as shown in Table 5-1.

C.4.3.1.1 **Type M**

This section shows intakes when the plutonium urinalysis results were fit as type M material. As discussed above, each chronic intake period was fit independently. Figures C-7 to C-10 show the fits to the 50th-percentile values for each intake period. The same intake periods were applied to the 84th-percentile values because the values followed a similar pattern. Table C-9 summarizes the intake periods and corresponding intake rates for the 50th- and 84th-percentile values. Figure C-11 depicts the predicted excretion rates from all 50th-percentile type M intakes.

Table C-9.	турети	r u intake penou	s anu raies.		
		Plutonium inta	Plutonium intake rate (dpm/d)		
Start	Stop	50th-percentile	84th-percentile	GSD	
01/01/1946	12/31/1948	57.7	149.2	2.59	
01/01/1949	12/31/1952	21.1	54.11	2.56	
01/01/1953	12/31/1981	1.003	3.196	3.19	
01/01/1982	12/31/1988	0.1336	1.132	8.47	

Table C-9. Type M²³⁹⁺²⁴⁰Pu intake periods and rates.

The large decrease in 50th-percentile bioassay results and intakes after 1981 resulted from a switch in reporting practices from a reporting level of 0.025 dpm to reporting all results. The MDA also decreased from 0.05 dpm to 0.02 dpm in 1984.

C.4.3.1.2 **Type S**

This section shows intakes when the plutonium urinalysis results were fit as type S material. As discussed above, each chronic intake period was fit independently. Figures C-12 to C-15 show the fits to the 50th-percentile values for each intake period. The same intake periods were applied to the 84th-percentile values because the values followed a similar pattern. Table C-10 summarizes the intake period and corresponding intake rate for the 50th- and 84th-percentile values. Figure C-16 depicts the predicted excretion rates from all 50th-percentile type S intakes.

Table C-10. Type S²³⁹⁺²⁴⁰Pu intake period and rates.

		Plutonium inta		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1946	12/31/1948	1,579	4,094	2.59
01/01/1949	12/31/1952	495.8	1,270	2.56
01/01/1953	12/31/1981	12.21	38.82	3.18
01/01/1982	12/31/1988	2.654	20.45	7.71

The large decrease in 50th-percentile bioassay results and intakes after 1981 resulted from a switch in reporting practices from a reporting level of 0.025 dpm to reporting all results. The MDA also decreased from 0.05 dpm to 0.02 dpm in 1984.

C.4.3.2 Uranium

Uranium at Hanford existed as mostly type F or S, but neither classification should be considered a perfect match to the uranium compounds, and type M cannot be ruled out. Therefore, all three types should be considered possible. Thorium exposure was associated with uranium exposure in 300 Area facilities from 1950 to 1970. Section C.5.2 contains guidance for adding thorium to uranium intakes.

C.4.3.2.1 **Type F**

This section shows intakes when uranium urinalysis results were fit using a type F material. Figure C-17 shows the fit to the 50th-percentile values from all intakes. The figure depicts the expected excretion rate from an individual who was exposed for all the periods at the 50th-percentile intake rate. The same intake periods were applied to the 84th-percentile values because the values followed a similar pattern. Table C-11 summarizes the intake periods and corresponding intake rates for the 50th- and 84th-percentile values.

			Uranium inta		
	Start	Stop	50th-percentile	84th-percentile	GSD
Ī	01/01/1948	12/31/1952	17.42	58.35	3.35
Ī	01/01/1953	12/31/1956	12.3	39.45	3.21
Ī	01/01/1957	12/31/1961	17.73	61.65	3.48
Ī	01/01/1962	12/31/1974	10.61	31.09	2.93
	01/01/1975	12/31/1983	1.281	7.037	5.49
	01/01/1984	12/31/1988	0.4903	1.895	3.86

Table C-11. Type F uranium intake periods and rates.

The decrease in intake rate in 1975 resulted from a decrease in the reporting level from 4 μ g/L to 0.4 μ g/L. That decrease could also have resulted in part from the shutdowns of the PUREX and UO₃ plants from 1973 to 1983.

C.4.3.2.2 **Type M**

The intake periods for the type F fits were applied to the type M material fits. Figure C-18 shows the fit to the 50th-percentile values from all intakes. The same intake periods were applied to the 84th-percentile values because the values followed a similar pattern. Table C-12 summarizes the intake periods and corresponding intake rates for the 50th- and 84th-percentile values.

		Uranium inta	ke rate (µg/d)	
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1948	12/31/1952	73.12	244.5	3.34
01/01/1953	12/31/1956	49.35	159	3.22
01/01/1957	12/31/1961	72.79	252.6	3.47
01/01/1962	12/31/1974	42.78	124.9	2.92
01/01/1975	12/31/1983	4.414	25.99	5.89
01/01/1984	12/31/1988	1.919	7.043	3.67

Table C-12. Type M uranium intake periods and rates.

C.4.3.2.3 **Type S**

The intake periods for the type F and M fits were applied to the type S material fits. As discussed, each chronic intake period for type S material was fit independently. Figures C-19 to C-24 show the individual fits for the 50th-percentile values. The 84th–percentile values were fit similarly. Table C-13 summarizes the intake rates for the 50th- and 84th-percentile values. Figure C-25 shows the predicted excretion rates from all 50th-percentile value type S intakes.

		Uranium inta	ke rate (µg/d)	
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1948	12/31/1952	1,308	4,373	3.34
01/01/1953	12/31/1956	1,035	3,321	3.21
01/01/1957	12/31/1961	1,349	4,660	3.45
01/01/1962	12/31/1974	665.1	1,953	2.94
01/01/1975	12/31/1983	83.67	403.3	4.82
01/01/1984	12/31/1988	46.84	171.6	3.66

Table C-13. Type S uranium intake periods and rates.

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C.4.3.2.4 **Strontium-90**

Only type F solubility was analyzed for ⁹⁰Sr. Figure C-26 shows the fit to the 50th-percentile values from all intakes. Table C-14 summarizes the intake rates for the 50th- and 84th-percentile values.

Table C-14. Type 1 SI Intake pendus and fates.				
		Sr-90 intake rate (dpm/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1965	12/31/1967	90.47	241.5	2.67
01/01/1968	12/31/1970	60.5	241.5	3.99
01/01/1971	12/31/1988	3.698	9.405	2.54

Table C-14.	Type F ⁹⁰ S	r intake	periods	and rates.
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C.4.3.3 Promethium-147

C.4.3.3.1 **Type M**

Each chronic intake period for type M¹⁴⁷Pm was fit independently. Figures C-27 and C-28 show the individual fits for the 50th-percentile values. The 84th-percentile values were fit similarly. Table C-15 summarizes the intake rates for the 50th- and 84th-percentile values. Figure C-29 shows the predicted excretion rates from all 50th-percentile value type M intakes.

Table C-15. Type M ¹⁴⁷Pm intake periods and rates.

		¹⁴⁷ Pm intake rate (dpm/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1966	12/31/1969	4,976	15,620	3.14
01/01/1970	12/31/1979	1,720	4,483	2.61

C.4.3.3.2 **Type S**

Each chronic intake period for type S¹⁴⁷Pm was fit independently. Figures C-30 and C-31 show the individual fits for the 50th-percentile values. The 84th–percentile values were fit similarly. Table C-16 summarizes the intake rates for the 50th- and 84th-percentile values. Figure C-32 shows the predicted excretion rates from all 50th-percentile value type S intakes.

Table C-10. Type 3 Fill intake pendus and fates.					
		Pm-147 intake rate (dpm/d)			
Start	Stop	50th-percentile	84th-percentile	GSD	
01/01/1966	12/31/1969	131,500	411,400	3.13	
01/01/1970	12/31/1979	34,810	90,850	2.61	

Table C-16. Type S¹⁴⁷Pm intake periods and rates

C.4.3.4 Zinc-65

Intakes of ⁶⁵Zn include a single acute intake in addition to the normal chronic intakes that were modeled. This acute intake was included to optimize the fit of the modeling results to the data. The data show a sudden increase in the last quarter of 1976 that is followed by a steady decrease over the next several quarters, which is indicative of an acute intake during the last quarter of 1976.

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C.4.3.4.1 Inhalation Intakes

Absorption type S was used for ⁶⁵Zn inhalation intakes. Each chronic intake period for type S ⁶⁵Zn was fit independently. However, the acute intake on October 1, 1976, was modeled with the contemporaneous chronic intake. Figures C-33 to C-38 show the individual fits for the 50th-percentile values. The 84th-percentile values were fit similarly. Table C-17 summarizes the intake rates for the 50th- and 84th-percentile values. Figure C-39 shows the predicted excretion rates from all 50th-percentile value type S intakes.

		Zinc-65 intake rate (pCi/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1960	12/31/1963	44.56	181.2	4.07
01/01/1964	12/31/1967	53.68	199.6	3.72
01/01/1968	12/31/1970	35.29	124.2	3.52
01/01/1971	12/31/1972	29.88	94.31	3.16
01/01/1973	12/31/1977	12.58	42.59	3.39
01/01/1978	12/31/1984	7.411	21.83	2.95
10/01/1976 ^a	-	2,555 ^b	14,260 ^b	5.58

Table C-17. Type S zinc-65 inhalation intake periods and rates.

This is the date of the acute intake. a.

b. This intake has units of pCi for an acute intake rather than pCi/d.

C.4.3.4.2 Ingestion Intakes

The ingestion intakes of ⁶⁵Zn were modeled together. Figure C-40 shows the fit to the 50th-percentile values from all intakes. The 84th-percentile values were fit similarly. Table C-18 summarizes the intake rates for the 50th- and 84th-percentile values. The once-through-cooled reactors were being shutdown from 1964 to 1971 (ORAUT 2010a). Ingestion intakes after the once-through-cooled reactors were shut down (after 1972) were not considered plausible.

		Zn-65 intake rate (pCi/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1960	12/31/1963	26.21	105.0	4.01
01/01/1964	12/31/1967	27.64	103.4	3.74
01/01/1968	12/31/1970	16.69	57.34	3.44
01/01/1971	12/31/1972	13.06	39.86	3.05

Table C-18. Zinc-65 ingestion intake periods and rates.

C.4.3.5 Sodium-24

C.4.3.5.1 Inhalation Intakes

Absorption type F was used for ²⁴Na inhalation intakes. Figure C-41 shows the fit to the 50th-percentile values from all intakes. The 84th-percentile values were fit similarly. Table C-19 summarizes the intake rates for the 50th- and 84th-percentile values.

C.4.3.5.2 Ingestion Intakes

The ingestion intakes of ²⁴Na were modeled together. Figure C-42 shows the fit to the 50th-percentile values from all intakes. These depict the expected excretion rates from an individual who was

		Na-24 intake rate (pCi/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1960	09/30/1963	310.3	942.6	3.04
10/01/1963	12/31/1963	1921	6,939	3.61
01/01/1964	09/30/1964	452.7	1,410	3.11
10/01/1964	03/31/1965	1,037.6	3,654	3.52
04/01/1965	12/31/1973	452.7	1,410	3.11
01/01/1974	12/31/1984	250.8	680.7	2.71

Table C-19.	Type F ²⁴ Na inh	alation intake	periods and rates.
	Typer runni	alation intake	

exposed for all the periods at the 50th- and 84th-percentile intake rates. Table C-20 summarizes the intake rates for the 50th- and 84th-percentile values. Ingestion intakes after the once-through-cooled reactors were shut down (by 1971 because of the very short half-life of ²⁴Na) were not considered plausible.

		Na-24 intake rate (pCi/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1960	09/30/1963	205.6	624.7	3.04
10/01/1963	12/31/1963	1274	4,599	3.61
01/01/1964	09/30/1964	311.9	986.3	3.16
10/01/1964	03/31/1965	687.6	2,421	3.52
04/01/1965	12/31/1971	311.9	986.3	3.16

Table C-20. Sodium-24 ingestion intake periods and rates.

C.4.3.6 Cesium-137

Only absorption type F was analyzed for ¹³⁷Cs. Figure C-43 shows the fit to the 50th-percentile values from all intakes. Table C-21 summarizes the intake rates for the 50th- and 84th-percentile values.

		Cs-137 intake rate (pCi/d)		
Start	Stop	50th-percentile	84th-percentile	GSD
01/01/1960	12/31/1960	118.6	173.7	1.46
01/01/1961	12/31/1961	39.08	61.74	1.58
01/01/1962	12/31/1962	55.23	81.79	1.48
01/01/1963	12/31/1964	246.8	374.8	1.52
01/01/1965	12/31/1966	132.6	213.9	1.61
01/01/1967	12/31/1967	44.34	85.13	1.92
01/01/1968	12/31/1976	28.38	58.36	2.06
01/01/1977	12/31/1988	10.13	26.08	2.57

Table C-21. Type F ¹³⁷Cs intake periods and rates.

C.5 ASSIGNMENT OF INTAKES AND DOSES

Tables C-22 to C-31 summarize the 50th-percentile intake rates, GSDs, and 95th percentile intake rates for each primary radionuclide over their respective periods and solubility classes. 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 at least 3 was assigned for each of the intake periods for the 50th percentile intakes (ORAUT 2007b). The GSDs for different intake periods have

also been adjusted in a favorable to claimant manner for consistency between intake periods for calculational efficiency. The 95th percentile intakes should be assigned as a constant rather than lognormal distribution.

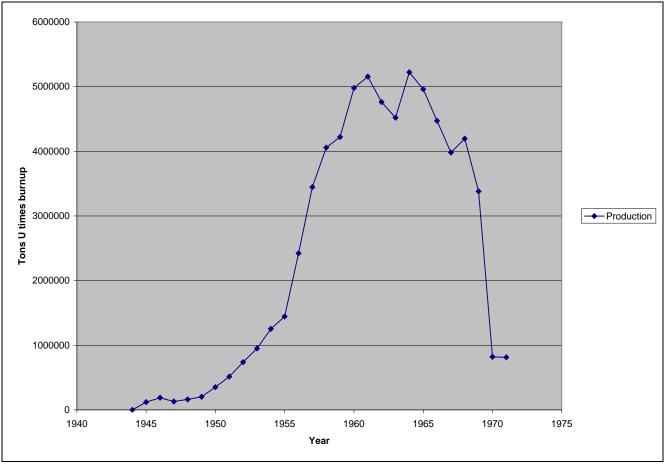


Figure C-6. Production rates in tons of uranium times fuel burnup, 1944 to 1972 [131].

The bioassay results that were used to establish coworker intakes did not cover all the years of possible exposure at Hanford. Extrapolation from monitored years to unmonitored years is discussed as part of the overall discussion of intakes for each radionuclide. The history of production at Hanford is relevant to those discussions. Figure C-6 shows how a figure of merit for production (tons of uranium processed in the reactors times the burnup) varied from 1944 to 1972. This unit is especially relevant for the production of fission products and to a reasonable extent for production of activation products. The figure shows that the period of highest production at Hanford was from about 1958 to 1968, which overlaps with the period of whole-body counting and ⁹⁰Sr urinalysis.

For assignment of coworker intakes after 1988, the intake rates for 1988 for each radionuclide may be assigned. Given that the internal dosimetry and radiation safety programs generally improved with time, especially after 1988, the assignment of the 1988 intake rates for periods after 1988 is considered to be favorable to claimants. For radionuclides with no assigned intake rate in 1988, no intake post-1988 should be assigned.

C.5.1 <u>Plutonium</u>

Dose reconstructors should assume a 12% ²³⁹Pu (fuel-grade) mixture for coworker intakes per Table C-22. Tables C-22 and C-23 list the ²³⁹Pu intakes and associated GSDs for each period for type M and S solubility classes, respectively. A small amount of plutonium was produced in 1944 (T Plant started operations in December 1944), and more was produced in 1945. As presented in Section 5.1.2, an SEC was designated in which NIOSH found that it was infeasible to reconstruct doses for plutonium before August 31, 1946 due to inadequate monitoring (72 FR 55214). Therefore, dose reconstructions will not include any doses from plutonium before August 31, 1946.

Table C-22. Type M plutonium-239 intake periods and rates.

		Plutonium intake rate (dpm/d)					
Start	Stop	50th-percentile	GSD	95th-percentile			
09/01/1946	12/31/1948	57.7	3.00	352			
01/01/1949	12/31/1952	21.1	3.00	129			
01/01/1953	12/31/1981	1.003	3.19	6.76			
01/01/1982	12/31/1988	0.1336	8.47	4.49			
1/1/1989	Present	0.1336	8.47	4.49			

a. Assumed to be 25 times the 1946 to 1948 rate.

	71 1						
		Plutonium intake rate (dpm/d)					
Start	Stop	50th-percentile	GSD	95th-percentile			
09/01/1946	12/31/1948	1579	3.00	9622			
01/01/1949	12/31/1952	495.8	3.00	3021			
01/01/1953	12/31/1981	12.21	3.18	81.9			
01/01/1982	12/31/1988	2.654	7.71	76.4			
1/1/1989	Present	2.654	7.71	76.4			

Table C-23. Type S plutonium-239 intake period and rates.

a. Assumed to be 25 times the 1946 to 1948 rate.

C.5.2 Uranium

Tables C-24 to C-26 list the uranium intakes and associated GSDs for each period. The values were adjusted from micrograms per day to picocuries per day for input into IMBA. Uranium bioassay started in 1947, but the early results were not considered reliable. The first extruded uranium rods arrived in October 1943 and machining began in December. From December 1943 through 1948, intakes for uranium should be assigned in accordance with the Process/Job Title listed as Machining/Operator as presented in Battelle-TBD-6000/PNWD-3738, *Technical Basis Document: Site Profiles for Atomic Weapons Employers That Worked Uranium and Thorium Metals* (Battelle 2006).

Starting in 1952, uranium at Hanford was RU with impurities. Impurities must be included with intakes of uranium; Section 5.2.5 contains the activities of impurities to apply to the uranium intakes in Tables C-24 to C-26 from 1952 to the present. The absorption types for the uranium impurities are provided in ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2007b).

C.5.3 <u>Strontium-90</u>

Table C-27 lists the ⁹⁰Sr intakes and associated GSDs for each period. Production of fission products in general from 1965 to 1967 was as great as at any time in the history of Hanford, and buildup of ⁹⁰Sr

Table C-24. Uranium Type F intake periods and rates.

		Uranium Type F intake (pCi/d)				
		50th-percentile 95th-percent				
Start	Stop	intake	GSD	intake		
		Not applicable		N/A		
12/01/1943	12/31/1948	(N/A)	N/A			
01/01/1949	12/31/1951	12.29	3.48	95.6		
01/01/1952	12/31/1952	15.85	3.48	123		
01/01/1953	12/31/1956	11.19	3.48	87.0		
01/01/1957	12/31/1961	16.13	3.48	125		
01/01/1962	12/31/1974	9.654	3.00	58.8		
01/01/1975	12/31/1983	1.166	5.49	19.2		
01/01/1984	12/31/1988	0.446	3.86	4.11		
1/1/1989	Present	0.446	3.86	4.11		

Table C-25. Uranium Type M intake periods and rates.

		Uranium Type M intake (pCi/d)				
		50th-percentile		95th-percentile		
Start	Stop	Intake	GSD	intake		
		See (Battelle		N/A		
12/01/1943	12/31/1948	2006)	5			
01/01/1949	12/31/1951	51.6	3.47	399		
01/01/1952	12/31/1952	66.5	3.47	515		
01/01/1953	12/31/1956	44.9	3.47	348		
01/01/1957	12/31/1961	66.2	3.47	513		
01/01/1962	12/31/1974	38.9	3.00	237		
01/01/1975	12/31/1983	4.02	5.89	74.2		
01/01/1984	12/31/1988	1.75	3.67	14.8		
1/1/1989	Present	1.75	3.67	14.8		

Table C-26. Uranium Type S intake periods and rates.

		Uranium Type S intake (pCi/d)				
Start	Stop	50th-percentile Intake	GSD	95th-percentile Intake		
		See (Battelle				
12/01/1943	12/31/1948	2006)	5	N/A		
01/01/1949	12/31/1951	923	3.45	7078		
01/01/1952	12/31/1952	1,190	3.45	9126		
01/01/1953	12/31/1956	942	3.45	7224		
				9409		
01/01/1957	12/31/1961	1,227	3.45			
01/01/1962	12/31/1974	605	3.00	3686		
01/01/1975	12/31/1983	76.1	4.82	1012		
01/01/1984	12/31/1988	42.6	3.66	361		
1/1/1989	Present	42.6	3.66	361		

as a contamination source had occurred over 20 years of operation. Air concentration limits were essentially unchanged from 1947 to the present (Parker 1947; Patterson 1949). The tolerance air concentration in 1945 was 10 times the 1947 value. Therefore, it is reasonable to apply the ⁹⁰Sr intake rates from the 1965 to 1967 period to the 1947 to 1964 period and to apply 10 times those rates to the 1944 to 1946 period.

		SI-90 III.	akerale	(apin/a)
		50th-percentile		95th-percentile
Start	Stop	Intake	GSD	Intake
12/01/1944	12/31/1946	904.7 ^a	3.00	5512
01/01/1947	12/31/1964	90.47 ^b	3.00	551.2
01/01/1965	12/31/1967	90.47	3.00	551.2
01/01/1968	12/31/1970	60.5	3.99	589
01/01/1971	12/31/1988	3.698	3.00	22.5
1/1/1989	Present	3.698	3.00	22.5

 Table C-27. Type F ⁹⁰Sr intake periods and rates.

 Sr-90 intake rate (dpm/d)

a. Intake rate assumed to be 10 times the rate for 1965 to 1967.

b. Intake rate assumed to be the same as the rate for 1965 to 1967.

Review of claimant mixed fission product bioassay data for the 1958 through 1963 period revealed an essentially constant GM urinary excretion rate, which supports the assumption that use of the 1965 to 1967 data is appropriate for 1947 through 1964.

C.5.4 Promethium-147

Table C-28 lists the ¹⁴⁷Pm intakes and associated GSDs for each period. Promethium-147 work occurred mostly in the 325 Building starting in 1961, 222-S from October 1964 through sometime in 1965, 201-C from 1964 through 1967, the 308 and probably the 309 Buildings approximately 1965 into the 1970s, and the 327 Building during major production (assume 1966-1970). See the Hanford Site Description for details (ORAUT 2010c). Insufficient bioassay data were available for statistical analysis until 1966; intakes for unmonitored potentially exposed workers before 1966 were set equal to the 1966 to 1969 intakes because, for the most part, the same buildings and same facilities within those buildings (and in general even the same workers) were used for both periods. In addition, intakes from a February 1963 contamination spread were analyzed and found to be comparable at the 95th percentile to the 1966 to 1969 intake rates [132].

I

		· · ·	Pm-147 intake (dpm/d)								
		-	Туре М	1	•	Type S	5				
		50th-percentile		95th-percentile	50th-percentile		95th-percentile				
Start	Stop	intake	GSD	intake	intake	GSD	intake				
10/01/1961	12/31/1965 ^a	4,976	3.14	32,684	131,500	3.13	859,204				
01/01/1966	12/31/1969	4,976	3.14	32,684	131,500	3.13	859,204				
01/01/1970	12/31/1979	1,720	3.00	10,481	34,810	3.00	212,114				

Table C-28. Promethium-147 intake periods and rates.

a. Intakes set equal to the 1966-69 intakes.

C.5.5 <u>Zinc-65</u>

Table C-29 lists the zinc intakes and associated GSDs for each period. Similar to ⁹⁰Sr, the intake rates before 1960 were assumed based on smaller production and similar air concentration limits, with the exception of the 1944 to 1946 period. Review of claimant mixed fission product bioassay data for the 1958 through 1963 period revealed an essentially constant GM urinary excretion rate, which supports the assumption that use of the 1960 to 1963 data is appropriate for 1947 through 1959. Because the intakes are based on whole-body counting, which measures activity in the body from all intake pathways, the dose reconstructor should apply the more favorable to claimant of the inhalation or ingestion intake <u>but not both in the same year</u>. If ingestion is used and the exposure period

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continues to 1973 or later, then the intake mode must be switched from ingestion to inhalation for the latter years. Because ingestion was determined not to be plausible after 1972, the intakes for inhalation must be assumed after this date, even when ingestion has been assumed before this date.

		•	Zn-65 intake (pCi/d)								
		lr	nhalatio	n		า					
Start	Stop	50th- percentile intake	GSD	95th- percentile intake	50th- percentile intake	GSD	95th- percentile intake				
12/01/1944	12/31/1946 ^a	445.6	4.07	4484	262	4.01	2573				
01/01/1947	12/31/1959 ^b	44.56	4.07	448	26.2	4.01	257				
01/01/1960	12/31/1963	44.56	4.07	448	26.2	4.01	257				
01/01/1964	12/31/1967	53.68	4.07	540	27.64	4.01	271				
01/01/1968	12/31/1970	35.29	3.52	280	16.69	3.44	127				
01/01/1971	12/31/1972	29.88	3.52	237	13.06	3.05	82				
01/01/1973	12/31/1977	12.58	3.52	99.7	NA ^c						
01/01/1978	12/31/1984	7.411	3.00	45.2	NA						
10/1/1976 ^d	-	2,555 ^e	5.58	43,212	NA						

Table C-29. Zinc-65 intake periods and rates.

a. Intake rates assumed to be 10 times the rates for 1960 to 1963.

b. Intake rates assumed to be the same as the rates for 1960 to 1963.

c. NA = not applicable.

d. This is the date of the acute intake.

e. This intake has units of pCi for an acute intake rather than pCi/d.

C.5.6 <u>Sodium-24</u>

Table C-30 lists the ²⁴Na intakes and associated GSDs for each period. As an assumption favorable to claimants, the highest intake rate within a given year was used to set the intake rate for the entire year. Similar to ⁹⁰Sr, the intake rates before 1960 were assumed based on smaller production and similar air concentration limits with the exception of the 1944 to 1946 period, which was supported by the review of the fission product urine data as discussed previously. Because the intakes were based on whole-body counting, which measures activity in the body from all intake pathways, the dose reconstructor should apply the more favorable to claimant of the inhalation or ingestion intakes <u>but not both in the same year</u>. If ingestion is used and the exposure period continues to 1972 or later, then the intake mode must be switched from ingestion to inhalation for the latter years; that is, the intakes for inhalation must be assumed after 1971, even when ingestion has been assumed before this date.

				Na-24 int	ake (pCi/d)			
		Inhal	ation T	ype F	Ingestion			
Start	Stop	50th- percentile intake	GSD	95th- percentile intake	50th- percentile intake	GSD	95th- percentile intake	
12/1/1944	12/31/1946 ^a	3,103	3.04	19325	2,056	3.04	12804	
01/01/1947	12/31/1959 ^b	310.3	3.04	1932	205.6	3.04	1280	
01/01/1960	12/31/1962	310.3	3.04	1932	205.6	3.04	1280	
01/01/1963	12/31/1963	1,921	3.61	15872	1,274	3.61	10526	
01/01/1964	12/31/1965	1,038	3.52	8228	687.6	3.61	5681	

Table C-30. Sodium-24 intake periods and rates.

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		Na-24 intake (pCi/d)						
		Inhalation Type F				'n		
Start	Stop	50th- percentile intake GSD		95th- percentile intake	50th- percentile intake	GSD	95th- percentile intake	
01/01/1966	12/31/1971	452.7	3.11	2927	311.9	3.61	2577	
01/01/1972	12/31/1973	452.7	3.11	2927	NA ^c	NA		
01/01/1974	12/31/1984	250.8	3.11	1622	NA	NA		

a. Intake rates assumed to be 10 times the rates for 1960 to 1962.

b. Intake rates assumed to be the same as the rates for 1960 to 1962.

c. NA = not applicable.

C.5.7 Cesium-137

Table C-31 lists the ¹³⁷Cs intakes and associated GSDs for each period. Similar to ⁹⁰Sr, the intake rates before 1960 were assumed based on smaller production and similar air concentration limits, with the exception of the period from 1944 to 1946.

	Type i cesiun	1-157 Illiake perious and fales.						
		Cs-137 intake rate (pCi/d)						
		50th-percentile		95th-percentile				
Start	Stop	intake	GSD	intake				
12/1/1944	12/31/1946	1,186 ^ª	3.00	7227				
01/01/1947	12/31/1959	118.6 ^b	3.00	723				
01/01/1960	12/31/1960	118.6	3.00	723				
01/01/1961	12/31/1961	39.08	3.00	238				
01/01/1962	12/31/1962	55.23	3.00	337				
01/01/1963	12/31/1964	246.8	3.00	1503				
01/01/1965	12/31/1966	132.6	3.00	808				
01/01/1967	12/31/1967	44.34	3.00	270				
01/01/1968	12/31/1976	28.38	3.00	173				
01/01/1977	12/31/1988	10.13	3.00	61.7				
1/1/1989	Present	10.13	3.00	61.7				

Table C-31. Type F cesium-137 intake periods and rates.

Intake rate assumed to be 10 times the rate for 1960. a.

Intake rate assumed to be the same as the rate for 1960. h

C.5.8 Additional Radionuclides

A large number of different radionuclides were present at Hanford at various times, but the available bioassay data for radionuclides in addition to those in this attachment were considered to be too few to be statistically reliable for intake estimation. Workers who were exposed to ¹³⁷Cs, ⁹⁰Sr, ²⁴Na, and ⁶⁵Zn could also have been exposed to other fission and activation products. From 1960 to 1988, intakes of most fission or activation products would have been detectable in whole-body counts, but the recording practice for fission and activation products other than ¹³⁷Cs, ²⁴Na, and ⁶⁵Zn was not amenable to statistical analysis. Guidance for assigning intakes of additional radionuclides is provided in ORAUT-OTIB-0054 (ORAUT 2007a) and for specific buildings in Section 5.6.

Assignment to unmonitored workers of radionuclides that were not analyzed in this study should be in accordance with guidance in Section 5.6.

STATISTICAL SUMMARIES AND PLOTS

In the following tables, the GM is the 50th-percentile value of the fitted line and GM*GSD is the 84th-percentile value.

	Statistica	i summary c		our urinar	y excretion r	ates (dpm/d)	, 1946 to	1988.
Effective			Effective			Effective		
bioassay	GM	GM*GSD	bioassay	GM	GM*GSD	bioassay	GM	GM*GSD
date	(50th)	(84th)	date	(50th)	(84th)	date	(50th)	(84th)
07/01/1946	0.25402	0.66092	02/15/1964	0.01826	0.06779	08/15/1976	0.00801	0.02006
07/01/1947	0.20476	0.50991	05/15/1964	0.02133	0.08054	11/15/1976	0.00898	0.02764
04/01/1948	0.21047	0.53721	08/15/1964	0.02435	0.08191	02/15/1977	0.00871	0.02336
10/1/1948	0.22645	0.60437	11/15/1964	0.02067	0.07232	05/15/1977	0.00875	0.02371
07/01/1949	0.10752	0.28933	02/15/1965	0.01409	0.04706	08/15/1977	0.00835	0.02161
07/01/1950	0.10249	0.25412	05/15/1965	0.01380	0.04542	11/15/1977	0.00812	0.02061
07/01/1951	0.10358	0.26263	08/15/1965	0.01248	0.03685	02/15/1978	0.00878	0.02468
07/01/1952	0.05707	0.14968	11/15/1965	0.01556	0.04811	05/15/1978	0.00906	0.02664
04/01/1953	0.01647	0.04436	02/15/1966	0.01197	0.03675	08/15/1978	0.00836	0.02192
10/1/1953	0.01724	0.04873	05/15/1966	0.01152	0.03613	11/15/1978	0.00825	0.02180
02/15/1954	0.01704	0.04637	08/15/1966	0.01354	0.04801	02/15/1979	0.00871	0.02373
05/15/1954	0.01746	0.04932	11/15/1966	0.01200	0.04220	05/15/1979	0.00849	0.02278
08/15/1954	0.01765	0.05080	02/15/1967	0.01330	0.05292	08/15/1979	0.00851	0.02273
11/15/1954	0.01697	0.04609	05/15/1967	0.02204	0.11370	11/15/1979	0.00818	0.02095
02/15/1955	0.01671	0.04478	08/15/1967	0.01175	0.04248	02/15/1980	0.00821	0.02140
05/15/1955	0.01695	0.04624	11/15/1967	0.01338	0.05001	05/15/1980	0.00865	0.02388
08/15/1955	0.01746	0.05021	02/15/1968	0.01216	0.04585	08/15/1980	0.00870	0.02426
11/15/1955	0.01681	0.04538	05/15/1968	0.01078	0.03891	11/15/1980	0.00900	0.02488
02/15/1956	0.01703	0.04723	08/15/1968	0.01397	0.06615	02/15/1981	0.00799	0.02036
05/15/1956	0.01716	0.04803	11/15/1968	0.01192	0.04489	05/15/1981	0.00827	0.02204
08/15/1956	0.01724	0.04828	02/15/1969	0.01321	0.04680	8/5/1981	0.00851	0.02341
11/15/1956	0.01756	0.04956	05/15/1969	0.00952	0.02753	11/5/1981	0.00109	0.00644
02/15/1957	0.01659	0.04369	08/15/1969	0.01101	0.03788	02/15/1982	0.00069	0.00973
05/15/1957	0.01699	0.04599	11/15/1969	0.01095	0.03868	05/15/1982	0.00053	0.00572
08/15/1957	0.01663	0.04360	02/15/1970	0.00974	0.03019	08/15/1982	0.00083	0.00647
11/15/1957	0.01712	0.04671	05/15/1970	0.00888	0.02567	11/15/1982	0.00149	0.01120
02/15/1958	0.01576	0.06661	08/15/1970	0.00980	0.03067	02/15/1983	0.00113	0.00661
05/15/1958	0.01089	0.03745	11/15/1970	0.01307	0.05338	05/15/1983	0.00087	0.00497
08/15/1958	0.01001	0.03106	02/15/1971	0.00933	0.02895	08/15/1983	0.00094	0.00738
11/15/1958	0.01240	0.05037	05/15/1971	0.00993	0.03206	11/15/1983	0.00083	0.00589
02/15/1959	0.01265	0.05288	08/15/1971	0.01126	0.03955	02/15/1984	0.00132	0.00760
05/15/1959	0.01163	0.04018	11/15/1971	0.01089	0.03976	05/15/1984	0.00062	0.01534
08/15/1959	0.01058	0.03276	02/15/1972		0.04290	08/15/1984		0.00810
11/15/1959	0.01098	0.03388	05/15/1972	0.01013	0.03254	11/15/1984	0.00099	0.00860
02/15/1960	0.01237	0.04317	08/15/1972	0.00973	0.03035	02/15/1985	0.00116	0.01028
05/15/1960	0.01002	0.02983	11/15/1972	0.01073	0.03642	05/15/1985	0.00056	0.00531
08/15/1960	0.00980	0.02880	02/15/1973	0.01107	0.03977	08/15/1985	0.00076	0.00541
11/15/1960	0.00980	0.02931	05/15/1973	0.00948	0.02877	11/15/1985	0.00073	0.00389
02/15/1961	0.00977	0.02866	08/15/1973	0.00945	0.02812	02/15/1986	0.00065	0.00635
05/15/1961	0.00984	0.02971	11/15/1973	0.00875	0.02403	05/15/1986	0.00069	0.01717

Table C-32. Statistical summary of ²³⁹Pu 24-hour urinary excretion rates (dpm/d), 1946 to 1988.^a

Effective			Effective			Effective		
bioassay	GM	GM*GSD	bioassay	GM	GM*GSD	bioassay	GM	GM*GSD
date	(50th)	(84th)	date	(50th)	(84th)	date	(50th)	(84th)
08/15/1961	0.01022	0.03099	02/15/1974	0.00885	0.02454	08/15/1986	0.00099	0.00725
11/15/1961	0.00990	0.02813	05/15/1974	0.00900	0.02532	11/15/1986	0.00050	0.00463
02/15/1962	0.00948	0.02621	08/15/1974	0.00911	0.02602	02/15/1987	0.00069	0.00551
05/15/1962	0.01091	0.03307	11/15/1974	0.01119	0.03852	05/15/1987	0.00065	0.00471
08/15/1962	0.01235	0.04221	02/15/1975	0.00910	0.02516	08/15/1987	0.00086	0.00488
11/15/1962	0.01061	0.03407	05/15/1975	0.00953	0.02738	11/15/1987	0.00048	0.00411
02/15/1963	0.01100	0.03651	08/15/1975	0.00802	0.02030	02/15/1988	0.00080	0.00395
05/15/1963	0.01276	0.04553	11/15/1975	0.00838	0.02233	05/15/1988	0.00052	0.00312
08/15/1963	0.01166	0.03917	02/15/1976	0.00857	0.02433	08/15/1988	0.00064	0.00339
11/15/1963	0.01479	0.05371	05/15/1976	0.00835	0.02245	11/15/1988	0.00020	0.00203

a. For 1946 through 1949, the Pu-239 values were calculated as 0.824 of the total Pu alpha values; for 1950 through 1954, the Pu-239 values were calculated as 0.830 of the total Pu alpha values, for 1951 through 08/15/1983, the Pu-239 values were calculated as 0.835 of the total Pu alpha values.

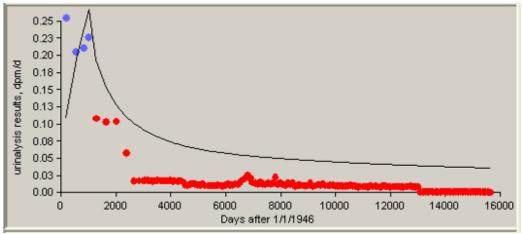


Figure C-7. 50th-percentile plutonium urinalysis data for intakes of Type M material, 1946 to 1948.

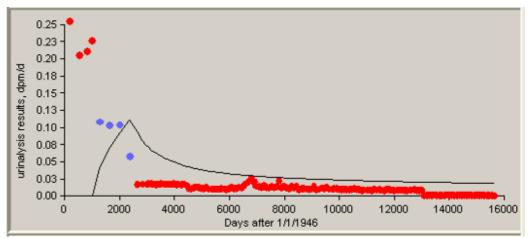


Figure C-8. 50th-percentile plutonium urinalysis data for intakes of Type M material, 1949 to 1952.

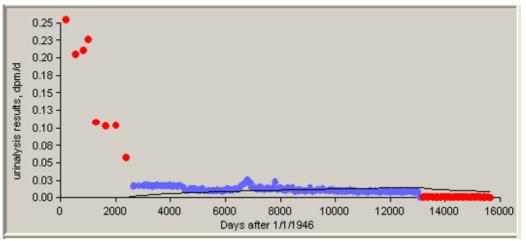


Figure C-9. 50th-percentile plutonium urinalysis data for intakes of Type M material, 1953 to 1981.

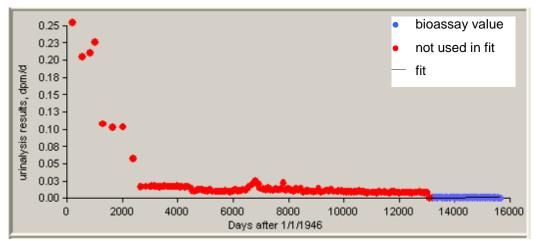


Figure C-10. 50th-percentile plutonium urinalysis data for intakes of Type M material, 1982 to 1988.

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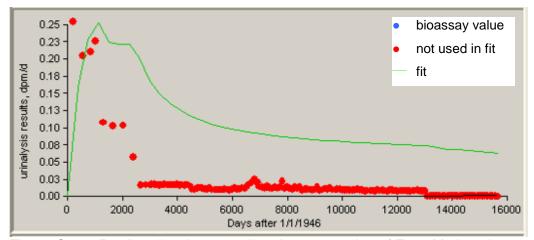


Figure C-11. Predicted 50th-percentile urinary excretion of Type M plutonium from 1946 to 1988 based on four independent intakes, compared to bioassay data.

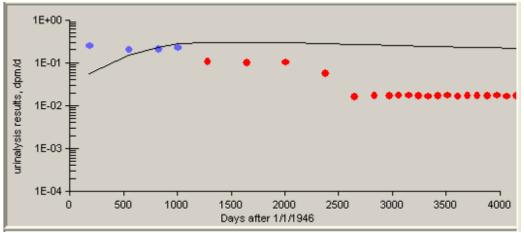


Figure C-12. 50th-percentile plutonium urinalysis data for intakes of Type S material, 1946 to 1948.

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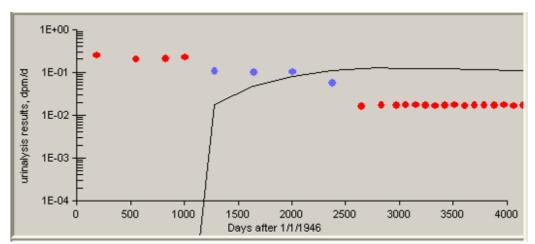


Figure C-13. 50th-percentile plutonium urinalysis data for intakes of Type S material, 1949 to 1952.

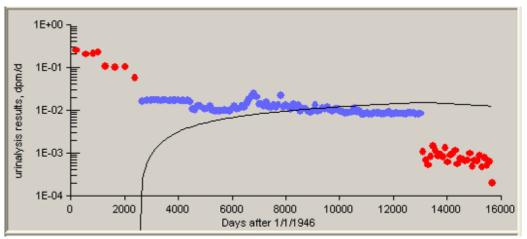


Figure C-14. 50th-percentile plutonium urinalysis data for intakes of Type S material, 1953 to 1981.

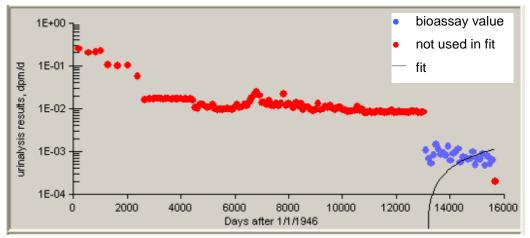


Figure C-15. 50th-percentile plutonium urinalysis data for intakes of Type S material, 1982 to 1988.

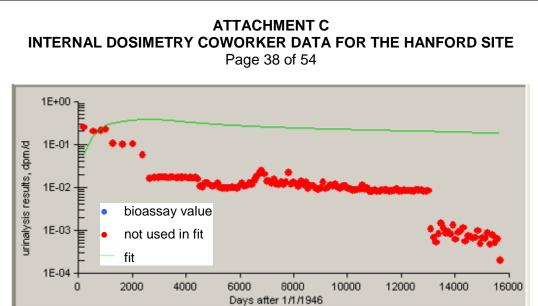


Figure C-16. Predicted 50th-percentile urinary excretion of Type S plutonium from 1946 to 1988 based on four independent intakes, compared to bioassay data.

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Table C-33.	Statistical sum	mary of uranium	24-hour urinary	excretion rates (µg	/d), 1948 to 1988. ^a

Effective			Effective			Effective		
bioassay	GM	GM*GSD	bioassay	GM	GM*GSD	bioassay	GM	GM*GSD
date	(50th)	(84th)	date	(50th)	(84th)	date	(50th)	(84th)
04/01/1948	3.88	12.48	08/15/1958	4.34	14.70	08/15/1968	2.96	8.82
10/1/1948	5.50	21.41	11/15/1958	3.96	12.54	11/15/1968	3.21	9.10
02/15/1949	4.20	15.22	02/15/1959	3.94	12.97	02/15/1969	2.38	5.98
05/15/1949	3.32	10.87	05/15/1959	4.59	15.29	05/15/1969	2.73	7.06
08/15/1949	7.02	20.71	08/15/1959	5.54	19.18	08/15/1969	2.48	6.26
11/15/1949	4.65	15.47	11/15/1959	4.48	16.23	11/15/1969	2.27	5.67
02/15/1950	6.97	23.35	02/15/1960	4.20	14.36	07/01/1970	2.24	5.65
05/15/1950	4.57	15.20	05/15/1960	5.82	20.97	07/01/1971	2.61	6.88
08/15/1950	4.71	13.26	08/15/1960	4.47	14.79	07/01/1972	3.10	8.19
11/15/1950	3.62	11.81	11/15/1960	4.34	15.45	07/01/1973	2.27	5.34
02/15/1951	3.97	13.61	02/15/1961	4.76	18.96	07/01/1974	2.86	8.96
05/15/1951	4.15	14.93	05/15/1961	4.85	17.54	07/01/1975	0.97	5.99
08/15/1951	4.51	16.53	08/15/1961	3.76	11.26	07/01/1976	0.79	4.84
11/15/1951	4.15	13.99	11/15/1961	3.86	11.32	07/01/1977	0.39	1.42
02/15/1952	5.71	20.24	02/15/1962	3.68	11.21	07/01/1978	0.25	0.76
05/15/1952	4.84	16.17	05/15/1962	3.05	8.70	07/01/1979	0.29	0.96
08/15/1952	4.48	14.71	08/15/1962	2.78	7.73	07/01/1980	0.25	0.74
11/15/1952	5.15	16.08	11/15/1962	2.74	7.69	07/01/1981	0.41	2.02
02/15/1953	4.89	15.49	02/15/1963	3.77	10.83	07/01/1982	0.36	2.11
05/15/1953	4.07	12.44	05/15/1963	3.19	8.55	07/01/1983	0.13	0.64
08/15/1953	3.39	10.92	08/15/1963	2.73	7.43	02/15/1984	0.14	0.82
11/15/1953	3.62	12.79	11/15/1963	2.76	7.37	05/15/1984	0.20	0.78
02/15/1954	3.02	9.77	02/15/1964	2.39	6.25	08/15/1984	0.25	1.15
05/15/1954	3.02	9.26	05/15/1964	2.54	6.79	11/15/1984	0.32	1.34
08/15/1954	3.16	10.11	08/15/1964	2.89	9.30	02/15/1985	0.31	1.25
11/15/1954	3.78	12.74	11/15/1964	2.66	7.54	05/15/1985	0.28	0.89
2/14/1955	3.17	9.87	02/15/1965	3.56	11.55	08/15/1985	0.23	0.71
05/15/1955	3.44	11.06	05/15/1965	3.82	13.26	11/15/1985	0.15	0.45
08/15/1955	2.77	8.14	08/15/1965	4.39	16.82	02/15/1986	0.13	0.38
11/15/1955	3.21	10.35	11/15/1965	3.72	11.94	05/15/1986	0.27	1.36
2/14/1956	2.61	7.45	02/15/1966	3.09	10.22	08/15/1986	0.21	0.78
05/15/1956	3.55	11.48	05/15/1966	3.02	9.28	11/15/1986	0.18	0.62
08/15/1956	4.20	14.94	08/15/1966	3.56	12.10	02/15/1987	0.12	0.43
11/15/1956	2.88	8.92	11/15/1966	3.13	9.35	05/15/1987	0.15	0.50
02/15/1957	4.28	15.51	02/15/1967	2.85	8.54	08/15/1987	0.14	0.42
05/15/1957	5.70	21.26	05/15/1967	2.97	8.84	11/15/1987	0.15	0.43
08/15/1957	6.23	23.63	08/15/1967	2.71	7.71	02/15/1988	0.14	0.41
11/51/1957	6.88	24.31	11/15/1967	2.77	8.13	05/15/1988	0.09	0.23
02/15/1958	6.65	22.18	02/15/1968	3.06	9.14	08/15/1988	0.09	0.29
05/15/1958	5.34	18.07	05/15/1968	3.73	11.62	11/15/1988	0.02	0.30

a. Through 1981, excretion was converted to µg/d from µg/L assuming 1.4L/d excretion.

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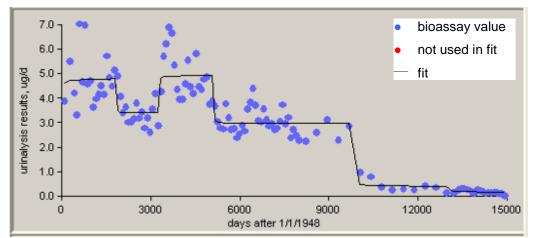


Figure C-17. 50th-percentile uranium urinalysis data for intakes of Type F material, 1948 to 1988.

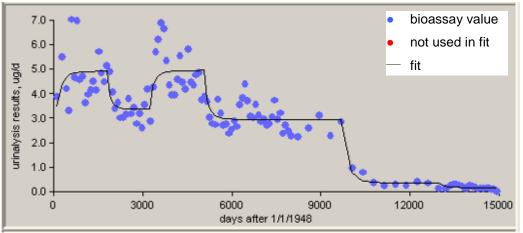


Figure C-18. 50th-percentile uranium urinalysis data for intakes of Type M material, 1948 to 1988.

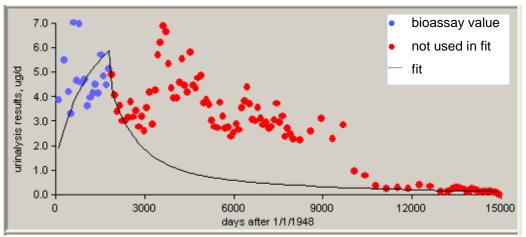


Figure C-19. 50th-percentile uranium urinalysis data for intakes of Type S material, 1948 to 1952.

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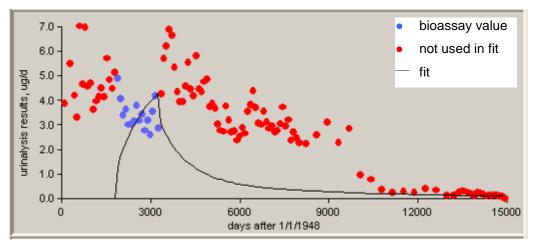


Figure C-20. 50th-percentile uranium urinalysis data for intakes of Type S material, 1953 to 1956.

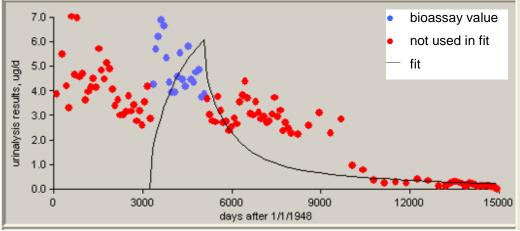


Figure C-21. 50th-percentile uranium urinalysis data for intakes of Type S material, 1957 to 1961.

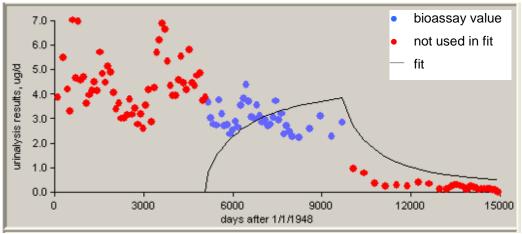


Figure C-22. 50th-percentile uranium urinalysis data for intakes of Type S material, 1962 to 1974.

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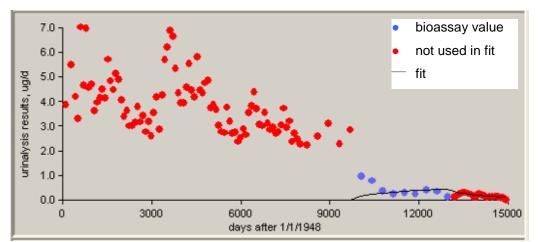


Figure C-23. 50th-percentile uranium urinalysis data for intakes of Type S material, 1975 to 1983.

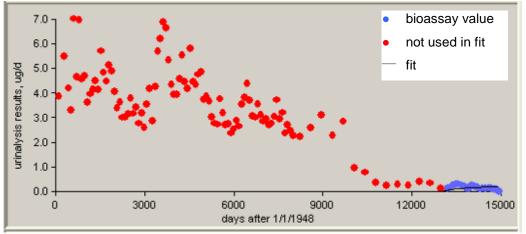


Figure C-24. 50th-percentile uranium urinalysis data for intakes of Type S material, 1984 to 1988.

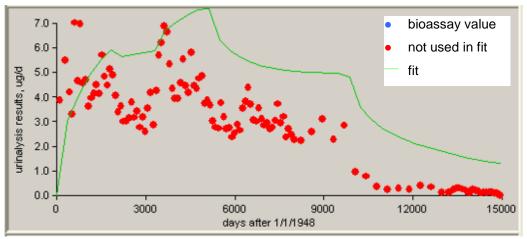


Figure C-25. Predicted 50th-percentile urinary excretion of Type S uranium from 1946 to 1988 based on six independent intakes, compared to bioassay data.

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Table C-34. Statistical summary of ⁹⁰ S	Sr 24-hour urinary	excretion rates	(dpm/d), '	1966 to 1988.
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Effective			Effective		
bioassay date	GM (50th)	GM*GSD (84th)	bioassay date	GM (50th)	GM*GSD (84th)
07/01/1966	20.74	56.46	11/15/1983	0.410	2.73
07/01/1969	14.53	55.88	02/15/1984	0.807	1.76
07/01/1971	1.76	4.56	05/15/1984	1.04	2.70
07/01/1972	2.33	12.27	08/15/1984	1.00	1.66
07/01/1973	1.64	4.51	11/15/1984	1.06	1.87
07/01/1974	1.38	3.82	02/15/1985	1.30	2.88
07/01/1975	0.923	2.57	05/15/1985	1.18	2.38
07/01/1976	1.20	3.40	08/15/1985	1.06	1.69
07/01/1977	0.835	2.12	11/15/1985	1.11	2.20
07/01/1978	1.009	3.82	02/15/1986	1.12	2.26
02/15/1979	0.882	2.55	05/15/1986	1.05	1.88
08/15/1979	2.00	5.58	08/15/1986	1.14	2.05
07/01/1980	1.92	4.92	11/15/1986	1.27	2.29
07/01/1981	2.33	7.60	02/15/1987	1.28	2.02
02/15/1982	0.289	1.43	05/15/1987	1.43	2.28
05/15/1982	0.322	1.58	08/15/1987	1.43	2.18
08/15/1982	0.350	1.72	11/15/1987	1.47	2.21
11/15/1982	0.802	7.91	02/15/1988	1.79	2.79
02/15/1983	0.665	2.60	05/15/1988	1.53	2.57
05/15/1983	0.387	3.13	08/15/1988	1.32	2.02
08/15/1983	0.743	3.78	11/15/1988	0.327	0.993

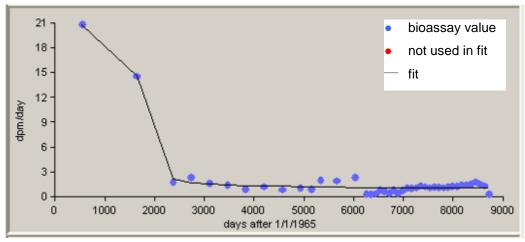


Figure C-26. 50th-percentile strontium urinalysis data for intakes of Type F material, 1965 to 1988.

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Table C-35. Statistical summary of ¹⁴⁷Pm 24-hour urinary excretion rates (dpm/d), 1967 to 1977.

Effective bioassay date	GM (50th)	GM*GSD (84th)
01/01/1967	25.7	82.9
2/12/1969	28.6	88.5
02/15/1971	13.7	35.6
07/01/1977 ^a	10.5	27.4

a. Very few bioassays were obtained during 1972 through 1974 so no statistics were developed. A single intake rate was fit for 1970 through 1979, using the 1971 and 1977 statistical values.

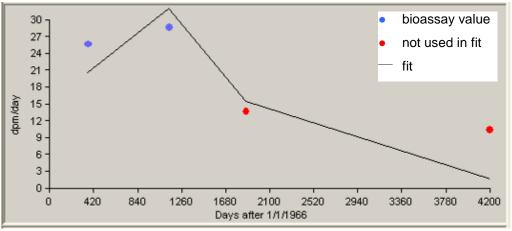


Figure C-27. 50th-percentile promethium urinalysis data for intakes of Type M material, 1966 to 1969.

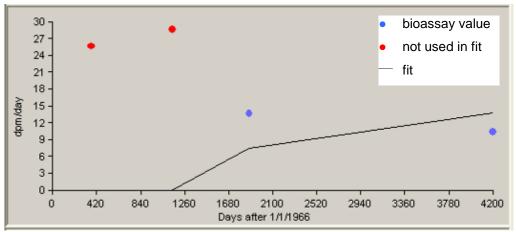


Figure C-28. 50th-percentile promethium urinalysis data for intakes of Type M material, 1970 to 1979.

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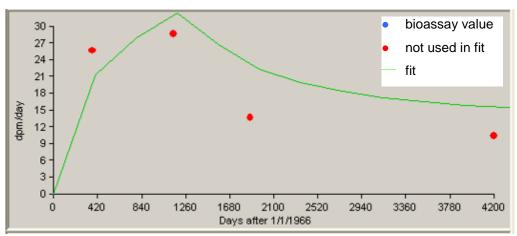


Figure C-29. Predicted 50th-percentile urinary excretion of Type M promethium from 1966 to 1979 based on two independent intakes, compared to bioassay data.

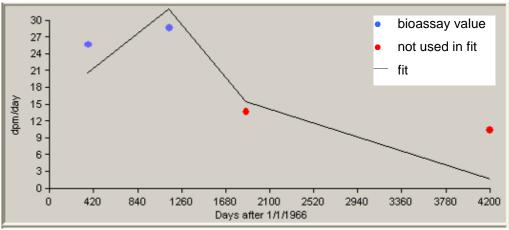


Figure C-30. 50th-percentile promethium urinalysis data for intakes of Type S material, 1966 to 1969.

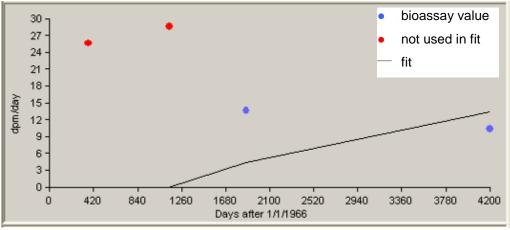


Figure C-31 50th-percentile promethium urinalysis data for intakes of Type S material, 1970 to 1979.

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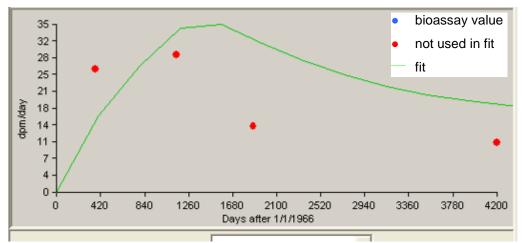


Figure C-32. Predicted 50th-percentile urinary excretion of Type S promethium from 1966 to 1979 based on two independent intakes, compared to bioassay data.

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Table C-36. Statistical summary of ⁶⁵Zn measured in whole-body counts (nCi), 1960 to 1984. Effective Effective Effective bioassay GM GM*GSD bioassay GM **GM*GSD** bioassay GM GM*GSD date (50th) (84th) date (50th) (84th) date (50th) (84th) 07/01/1960 2.45 7.80 05/15/1969 1.28 4.63 05/15/1977 0.788 2.83 04/01/1961 08/15/1969 2.16 9.58 1.05 3.66 08/15/1977 0.688 2.67 10/1/1961 2.13 7.78 11/15/1969 1.45 5.37 11/15/1977 0.388 1.27 04/01/1962 2.04 8.73 02/15/1970 1.45 5.56 02/15/1978 0.371 1.16 10/1/1962 2.15 12.08 05/15/1970 1.27 05/15/1978 4.31 0.369 1.15 02/15/1963 2.11 08/15/1970 8.44 1.56 5.11 08/15/1978 0.373 1.19 05/15/1963 1.60 6.25 11/15/1970 2.10 5.87 11/15/1978 0.396 1.28 08/15/1963 1.34 4.70 02/15/1971 1.52 5.06 02/15/1979 0.401 1.28 3.40 12.52 11/15/1963 05/15/1971 1.55 4.69 05/15/1979 0.415 1.37 1.02 02/15/1964 2.40 08/15/1971 1.29 08/15/1979 0.348 9.02 3.81 05/15/1964 2.12 7.22 11/15/1971 1.24 3.55 11/15/1979 0.406 1.31 08/15/1964 1.92 6.79 02/15/1972 1.18 3.92 02/15/1980 0.384 1.19 4.20 11/15/1964 16.06 05/15/1972 0.963 3.01 05/15/1980 0.403 1.32 02/15/1965 2.93 12.17 08/15/1972 1.17 3.74 08/15/1980 0.372 1.15 11/15/1972 05/15/1965 3.11 10.80 1.18 3.98 11/15/1980 0.348 1.01 08/15/1965 2.348 02/15/1973 1.18 4.21 1.09 9.40 02/15/1981 0.372 1.04 11/15/1965 2.56 10.10 05/15/1973 1.04 3.44 05/15/1981 0.354 08/15/1981 02/15/1966 2.82 9.32 08/15/1973 0.612 1.99 0.306 0.817 05/15/1966 2.87 10.10 11/15/1973 0.678 2.27 11/15/1981 0.316 0.854 0.497 08/15/1966 1.84 6.32 02/15/1974 1.61 02/15/1982 0.340 0.947 11/15/1966 1.86 05/15/1974 0.431 05/15/1982 0.944 8.03 1.33 0.338 02/15/1967 1.78 6.06 08/15/1974 0.456 1.41 08/15/1982 0.334 0.934 05/15/1967 3.35 12.02 11/15/1974 0.507 1.64 11/15/1982 0.331 0.909 08/15/1967 2.13 04/01/1975 0.405 1.23 0.323 8.26 02/15/1983 0.884 11/15/1967 1.50 5.91 10/1/1975 0.613 2.00 05/15/1983 0.334 0.922 02/15/1968 1.53 6.61 02/15/1976 0.647 2.19 08/15/1983 0.314 0.843 05/15/1968 1.38 5.45 05/15/1976 0.712 3.25 11/15/1983 0.337 0.937 08/15/1968 1.41 08/15/1976 0.673 0.354 1.03 6.18 2.46 04/01/1984 11/15/1968 1.83 6.78 11/15/1976 1.21 5.39 10/01/1984 0.391 1.09 02/15/1969 2.35 7.89 02/15/1977 0.964 3.74

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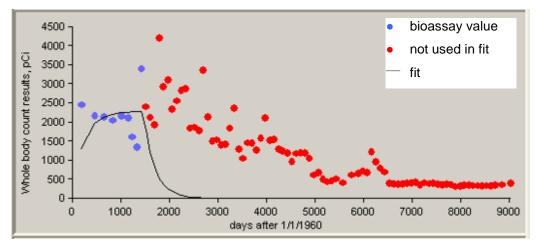


Figure C-33. 50th-percentile zinc whole-body counting data for inhalation intakes of Type S material, 1960 to 1963.

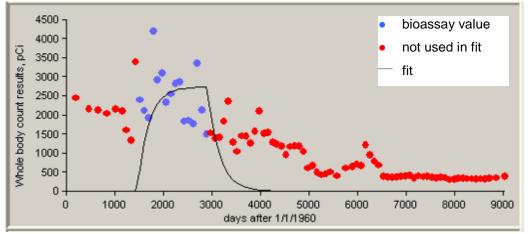


Figure C-34. 50th-percentile zinc whole-body counting data for inhalation intakes of Type S material, 1964 to 1967.

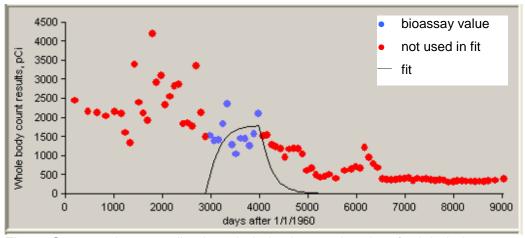


Figure C-35. 50th-percentile zinc whole-body counting data for inhalation intakes of Type S material, 1968 to 1970.

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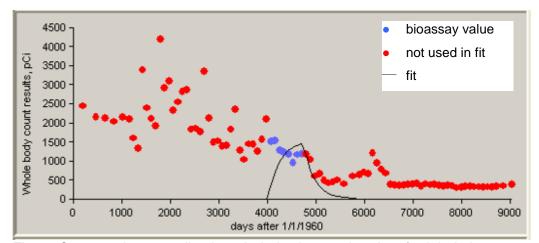


Figure C-36. 50th-percentile zinc whole-body counting data for inhalation intakes of Type S material, 1971 to 1972.

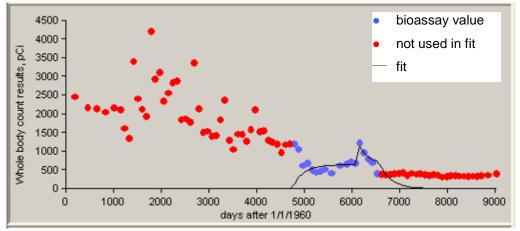


Figure C-37. 50th-percentile zinc whole-body counting data for inhalation intakes of Type S material, 1973 to 1977.

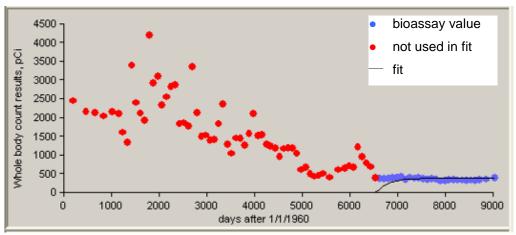


Figure C-38. 50th-percentile zinc whole-body counting data for inhalation intakes of Type S material, 1978 to 1984.

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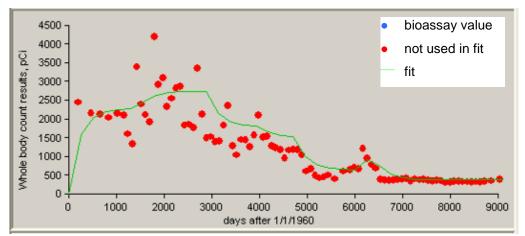


Figure C-39. Predicted 50th-percentile urinary excretion of inhaled Type S zinc from 1960 to 1984 based on six independent chronic intakes and one acute intake, compared to bioassay data.

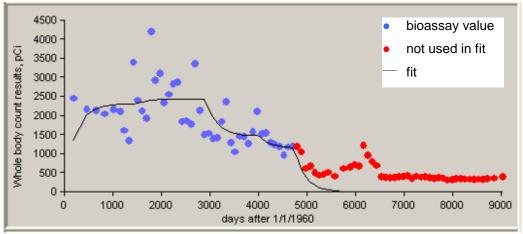


Figure C-40. 50th-percentile zinc whole-body counting data for ingestion intakes, 1960 to 1984.

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Table C-37. Statistical summary of ²⁴ Na measured in whole-body counts (nCi), 1960 to 1984.								
Effective			Effective			Effective		
bioassay	GM	GM*GSD	bioassay	GM	GM*GSD	bioassay	GM	GM*GSD
date	(50th)	(84th)	date	(50th)	(84th)	date	(50th)	(84th)
07/01/1960	0.221	0.587	02/15/1969	0.194	0.570	11/15/1976	0.158	0.434
04/01/1961	0.174	0.610	05/15/1969	0.204	0.649	02/15/1977	0.168	0.465
10/1/1961	0.187	0.616	08/15/1969	0.268	0.915	05/15/1977	0.166	0.466
02/15/1962	0.142	0.421	11/15/1969	0.285	0.979	08/15/1977	0.162	0.449
05/15/1962	0.182	0.609	02/15/1970	0.218	0.623	11/15/1977	0.162	0.458
08/15/1962	0.150	0.413	05/15/1970	0.228	0.605	02/15/1978	0.138	0.368
11/15/1962	0.145	0.414	08/15/1970	0.193	0.543	05/15/1978	0.148	0.401
02/15/1963	0.180	0.562	11/15/1970	0.196	0.559	08/15/1978	0.139	0.368
05/15/1963	0.185	0.515	02/15/1971	0.260	0.711	11/15/1978	0.150	0.414
08/15/1963	0.231	0.713	05/15/1971	0.234	0.591	02/15/1979	0.134	0.356
11/15/1963	1.11	4.02	08/15/1971	0.224	0.589	05/15/1979	0.155	0.421
02/15/1964	0.388	1.35	11/15/1971	0.220	0.591	08/15/1979	0.147	0.396
05/15/1964	0.355	1.16	02/15/1972	0.245	0.715	11/15/1979	0.145	0.387
08/15/1964	0.316	0.993	05/15/1972	0.255	0.760	02/15/1980	0.139	0.369
11/15/1964	0.647	2.06	08/15/1972	0.206	0.608	05/15/1980	0.140	0.372
02/15/1965	0.555	2.17	11/15/1972	0.243	0.673	08/15/1980	0.131	0.346
05/15/1965	0.295	0.983	02/15/1973	0.244	0.756	11/15/1980	0.140	0.370
08/15/1965	0.286	0.930	05/15/1973	0.196	0.556	02/15/1981	0.139	0.372
11/15/1965	0.292	0.891	08/15/1973	0.220	0.614	05/15/1981	0.138	0.366
02/15/1966	0.347	1.09	11/15/1973	0.176	0.501	08/15/1981	0.147	0.403
05/15/1966	0.233	0.650	02/15/1974	0.144	0.401	11/15/1981	0.133	0.348
08/15/1966	0.257	0.834	05/15/1974	0.147	0.431	02/15/1982	0.134	0.352
11/15/1966	0.218	0.687	08/15/1974	0.149	0.418	05/15/1982	0.149	0.407
02/15/1967	0.288	0.904	11/15/1974	0.139	0.369	08/15/1982	0.140	0.375
05/15/1967	0.428	1.740	02/15/1975	0.128	0.331	11/15/1982	0.143	0.387
08/15/1967	0.247	0.701	05/15/1975	0.131	0.340	02/15/1983	0.145	0.399
11/15/1967	0.346	0.998	08/15/1975	0.140	0.383	05/15/1983	0.142	0.385
02/15/1968	0.405	1.610	11/15/1975	0.131	0.342	08/15/1983	0.142	0.390
05/15/1968	0.269	0.887	02/15/1976	0.139	0.371	11/15/1983	0.150	0.414
08/15/1968	0.229	0.728	05/15/1976	0.148	0.404	04/01/1984	0.144	0.377
11/15/1968	0.256	0.805	08/15/1976	0.166	0.459	10/01/1984	0.171	0.495

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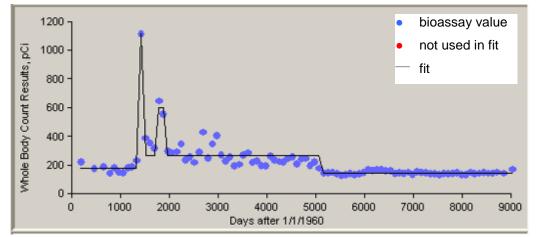


Figure C-41. 50th-percentile sodium whole-body counting data for inhalation intakes of Type F material, 1960 to 1984.

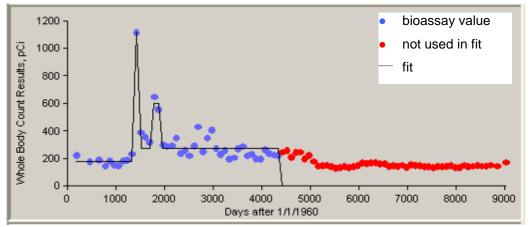


Figure C-42. 50th-percentile sodium whole-body counting data for ingestion intakes, 1960 to 1984.

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Table C-38.	Statistical	summary c	of ¹³⁷ Cs meas	ured in who	ole-body co	unts (nCi), 19	960 to 198	38.
Effective			Effective			Effective		
bioassay	GM	GM*GSD	bioassay	GM	GM*GSD	bioassay	GM	GM*GSD
date	(50th)	(84th)	date	(50th)	(84th)	date	(50th)	(84th)
02/15/1960	6.773	9.620	02/15/1969	11.964	18.931	02/15/1979	1.1775	2.9579
05/15/1960	6.689	9.347	05/15/1969	10.544	16.593	05/15/1979	0.8671	2.0518
08/15/1960	5.977	9.466	08/15/1969	8.526	13.837	08/15/1979	0.9219	2.1979
11/15/1960	5.770	8.302	11/15/1969	8.198	13.210	11/15/1979	1.2393	2.6536
02/15/1961	4.728	6.858	02/15/1970	6.417	12.039	02/15/1980	1.1519	2.8199
05/15/1961	4.482	6.764	05/15/1970	5.877	9.856	05/15/1980	0.8175	2.9124
08/15/1961	3.805	6.006	08/15/1970	5.417	9.242	08/15/1980	0.6695	1.8971
11/15/1961	3.491	5.184	11/15/1970	4.770	8.025	11/15/1980	0.7321	1.9398
02/15/1961	3.527	5.563	02/15/1971	4.049	7.580	02/15/1981	0.8278	2.1018
05/15/1961	3.457	5.055	05/15/1971	3.195	6.012	05/15/1981	0.4992	1.1927
08/15/1961	4.378	6.502	08/15/1971	2.574	4.939	08/15/1981	0.3353	0.9534
11/15/1961	5.253	7.678	11/15/1971	2.667	5.467	11/15/1981	0.4740	1.0964
02/15/1963	6.691	10.362	02/15/1972	2.824	5.511	02/15/1982	0.5655	1.3484
05/15/1963	6.773	9.620	05/15/1972	3.022	5.516	05/15/1982	0.4695	1.2468
08/15/1963	6.689	9.347	08/15/1972	2.176	4.728	08/15/1982	0.4364	1.0269
11/15/1963	5.977	9.466	11/15/1972	2.222	4.332	11/15/1982	0.5316	1.2952
02/15/1964	5.770	8.302	02/15/1973	2.0544	4.5035	02/15/1983	0.4257	1.1238
05/15/1964	4.728	6.858	05/15/1973	1.8482	3.4682	05/15/1983	0.3023	0.8154
08/15/1964	4.482	6.764	08/15/1973	1.8296	3.3453	08/15/1983	0.3705	0.9053
11/15/1964	3.805	6.006	11/15/1973	1.6404	3.3004	11/15/1983	0.4354	1.1195
02/15/1965	3.491	5.184	02/15/1974	1.2505	3.0204	02/15/1984	0.2487	0.6537
05/15/1965	3.527	5.563	05/15/1974	0.9186	2.1838	05/15/1984	0.2461	0.6285
08/15/1965	3.457	5.055	08/15/1974	1.0889	2.4621	08/15/1984	0.2430	0.6134
11/15/1965	4.378	6.502	11/15/1974	1.1091	2.7017	11/15/1984	0.2398	0.5927
02/15/1966	5.253	7.678	02/15/1976	1.0488	2.1653	02/15/1985	0.2450	0.6327
05/15/1966	6.691	10.362	05/15/1976	1.1197	2.6952	05/15/1985	1.1416	2.7971
08/15/1966	8.922	13.998	08/15/1976	0.9322	2.1818	08/15/1985	1.1220	2.8167
11/15/1966	11.218	16.663	11/15/1976	1.6176	3.9736	11/15/1985	1.1235	2.8170
02/15/1967	13.470	19.952	02/15/1977	1.3651	2.9877	04/01/1986	1.1169	2.7720
05/15/1967	15.374	23.543	05/15/1977	1.4688	3.4256	10/1/1986	1.1106	2.7455
08/15/1967	17.644	26.857	08/15/1977	1.1219	2.9244	04/01/1987	0.9186	2.1838
11/15/1967	18.129	26.767	11/15/1977	0.8683	2.2466	10/01/1987	1.0889	2.4621
02/15/1968	17.811	28.334	02/15/1978	1.0525	2.6430	04/01/1988	1.1091	2.7017
05/15/1968	15.208	23.287	05/15/1978	0.7876	2.3797	10/01/1988	1.0488	2.1653
08/15/1968	15.288	23.190	08/15/1978	0.7788	2.2337			
11/15/1968	13.341	19.881	11/15/1978	1.0824	2.6808			

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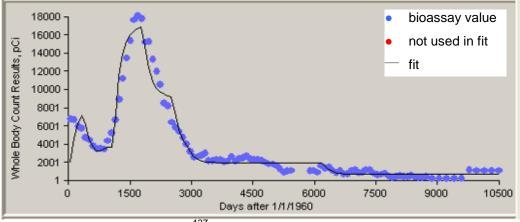


Figure C-43. 50th-percentile ¹³⁷Cs whole-body counting data for inhalation intakes of Type F material, 1960 to 1988.

SITE RESEARCH DATABASE REFERENCE NUMBERS

Table C-39. Hanford monthly reports.

358, 434, 442, 451, 5044, 5047, 5047, 5054, 5055, 5057, 5060, 5072, 5087, 5090, 5098, 5199, 5199, 33220, 33226, 33226, 33228, 33265, 33266, 33268, 33271, 34248, 34251, 34254, 34255, 34267, 34270, 34270, 34271, 34272, 34272, 34273, 34273, 34281, 34285, 34285, 34290, 34293, 34293, 34294, 34302, 34304, 34307, 34308, 34309, 34310, 34341, 34350, 34355, 34604, 35862, 35863, 35863, 35864, 35864, 35866, 35867, 36737, 36737, 36740, 36744, 36746, 36751, 36754, 36757, 36779, 36780, 36781, 36794, 36802, 36804, 36829, 36831, 36834, 36837, 36849, 36864, 36871, 36889, 36895, 36926, 36935, 36943, 37089, 37089, 37229, 37234, 37239, 37244, 37246, 37248, 37256, 37261, 37265, 37270, 37274, 37280, 37280, 37281, 37282, 37297, 37306, 37311, 37323, 37334, 37341, 37346, 37364, 37374, 37390, 37400, 37405, 37409, 37411, 37418, 37422, 37435, 37435, 37440, 37440, 37474, 37556, 37559, 37561, 37563, 37564, 37568, 37569, 37571, 37580, 37580, 37582, 37586, 37587, 37591, 40711, 40711, 40712, 40712, 40726, 40726, 40761, 40876, 40878, 40879, 40880, 40891, 40893, 40894, 40956, 40957, 40958, 40960, 40963, 40966, 40968, 40971, 40974, 40975, 40976, 40977, 40978, 40979, 40980, 40981, 40983, 40984, 40985, 40996, 40996, 41005, 41006, 41007, 41008, 41200, 41201, 41202, 41203, 41204, 41205, 41206, 41207, 41208, 41209, 41210, 41211, 41212, 41213, 41214, 41223, 41224, 41226, 41228, 41229, 41230, 41231, 41233, 41270, 41271, 41306, 41307, 41311, 41314, 41316, 41319, 41322, 41323, 41332, 41333, 41334, 41335, 41336, 41337, 41339, 41340, 41341, 41342, 41343, 41344, 41345, 41346, 41388, 51245, 52690, 52693, 53051, 53364, 53366, 53367

Table C-40. Tritium hard-copy bioassay data.

54902, 54903, 54905, 54906, 54917, 54924, 54925, 54927, 54928, 54929, 54930, 54932, 54937, 54943, 54944, 54947, 54948, 54957, 54960, 54961, 54963, 54964, 54967, 54970, 54971, 54972, 54973, 54974, 54975, 54976, 54977, 54978, 54979, 54980, 54981, 54982, 54983, 54984, 54986, 54987, 54988, 54989, 54990, 54991, 54992, 54993, 54994, 54995, 55036, 55038, 55039, 55040, 55041, 55043, 55049, 55053, 55060, 55071, 55073, 55076, 55079, 55083, 55094, 55095, 55096, 55097, 55098, 55099, 55100, 55101, 55102, 55103, 55104, 55105, 55106, 55107, 55108, 55109, 55110, 55111, 55112, 55113, 55114, 55115, 55116, 55117, 55118, 55119, 55139, 55140, 55143, 55145, 55146, 55147, 55152, 55167, 55168, 55169, 55170, 55172, 55175, 55178, 55184, 55185, 55186, 55187, 55188, 55189, 55190