



# ORAU TEAM Dose Reconstruction Project for NIOSH

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**PUBLICATION RECORD**

<b>EFFECTIVE DATE</b>	<b>REVISION NUMBER</b>	<b>DESCRIPTION</b>
12/21/2004	00	New Technical Basis Document for the Los Alamos National Laboratory – Occupational Internal Dose. Incorporates formal internal and NIOSH review comments. Initiated by Jack E. Buddenbaum.
10/15/2009	01	Incorporates information from documents not available at the time the original document was published. The current information available for uranium has been incorporated into Section 5.2.4. Clarification of MDAs, UNAA/DNAA analysis results, guidance for selection of enrichment, and a description of the new LANL Bioassay Repository (LABDR) content have been included. Appropriate references have been added. For claims involving uranium UNAA bioassay results: Because of the ambiguity in the interpretation of the uranium bioassay results when results were transferred into the LABDR, the guidance in the TBD might not have been interpreted consistently by DRs and PRs. UNAA/DNAA data provided by LANL is for DU and EU rather than U-238 and U-235. A table providing an aged “pure” <sup>238</sup> Pu mixture has been provided for use when exposures to heat source technology plutonium is suspected. Changes to the mixture might affect the total dose for plutonium intakes evaluated as “pure” <sup>238</sup> Pu. In addition, the Tiger Team Assessment (DOE 1991), the Human Experiments and the details of the RaLa SEC Report (NIOSH 2006) have been included in the revision. Incorporates formal internal and NIOSH review comments. Adds Attributions and Annotations section. A description of the class of employees added to the SEC for SEC Petition-00051 was added to the text describing the class of employees added under SEC Petition-00061 (employees associated with RaLa operations). A new Section 5.2.4.6 was added describing the limitations on internal dose reconstruction for employees in the non-excepted TAs (as listed in the petition). Added statement about only partial dose reconstructions are possible for most TAs. A new bullet was added in Section 5.7.2, General Guidance, indicating that only partial internal dose reconstructions are possible. Incorporates methods for estimating maximum doses in accordance with the evaluation report for Petition SEC-00109, changes to address comments by OCAS commenters, updated dose reconstruction methodologies and references, and addition of an algorithm to guide dose reconstructors for dose calculation and dose assignment. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Donald N. Stewart.
04/18/2013	02	Revision initiated to incorporate SEC-00109. Removed provisions to estimate unmonitored intakes consistent with the wording in the Advisory Board recommendation (Melius 2012). Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Donald N. Stewart.

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## ACRONYMS AND ABBREVIATIONS

25	<sup>235</sup> U or the standard ~93% enrichment (LANL designation)
28	<sup>238</sup> U or depleted uranium (LANL designation)
49	<sup>239</sup> Pu (LANL designation)
aCi	attocurie, 10 <sup>-18</sup> Ci
AEC	U.S. Atomic Energy Commission
AL	action level
AMAD	activity median aerodynamic diameter
Bq	becquerel
CAM	continuous air monitor
CFR	Code of Federal Regulations
Ci	curie
cm	centimeter
CMR	Chemistry and Metallurgy Research
cpm	counts per minute
d	day
D-38	depleted uranium mixture
DA	delayed action
DAC	derived air concentration
DL	detection limit
DNAA	delayed neutron activation analysis
DOE	U.S. Department of Energy
DOL	U.S. Department of Labor
DR	dose reconstructor
DTPA	diethylene triamine pentaacetic acid
dpm	disintegrations per minute
dps	disintegrations per second
DU	depleted uranium (less than 0.72% <sup>235</sup> U)
EEOICPA	Energy Employees Occupational Illness Compensation Program Act of 2000
ESH	Environmental, Safety, and Health
ET2	extrathoracic airways (posterior nasal passage, larynx, pharynx, and mouth)
EU	enriched uranium (more than 0.72% <sup>235</sup> U)
g	gram
GeLi	lithium-drifted germanium
GM	Geiger-Müller
GSD	geometric standard deviation
HPGe	hyper-pure germanium
hr	hour
HRL	Health Research Laboratory
HT	elemental tritium (tritiated gas)
HTO	tritium oxide (water or water vapor)
IBF	Ion Beam Facility
ICRP	International Commission on Radiological Protection
IMBA	Integrated Modules for Bioassay Analysis
in.	inch



IVML	In Vivo Measurements Laboratory (system)
keV	kiloelectron-volt, 1,000 electron-volts
L	liter
LABDR	LANL Bioassay Data Repository
LAMPF	Los Alamos Meson Physics Facility
LANL	Los Alamos National Laboratory (1981 to present)
LASL	Los Alamos Scientific Laboratory (until January 1981)
L <sub>c</sub>	critical level
LN(TH)	lymph nodes, thoracic
LSC	liquid scintillation counting
L X-ray	low-energy X-ray
MAC	maximum allowable concentration
MAP	mixed activation product
mBq	millibecquerel
mCi	millicurie
MDA	minimum detectable activity
MDTA	minimum detectable true activity
MeV	megaelectron-volt, 1 million electron-volts
MFP	mixed fission product
mg	milligram
min	minute
mL	milliliter
mm	millimeter
mo	month
MPBB	maximum permissible body burden
MPC	maximum permissible concentration
MPL	maximum permissible level
mR	milliroentgen
mrem	millirem
mrep	millirep
MSMA	minimum significant measured activity
MT	metal tritide
nCi	nanocurie
NCRP	National Council on Radiation Protection and Measurements
NDA	no detectable activity
NIOSH	National Institute for Occupational Safety and Health
NRS	nonroutinely sampled
NTA	neutron track analysis
NU	natural uranium
OBT	organically bound tritium
Or-93	oralloy (Oak Ridge alloy); uranium enriched to 40% or 93%
ORAU	Oak Ridge Associated Universities
ORNL	Oak Ridge National Laboratory
OTLBD	ORAU Team LANL Bioassay Database
OWR	Omega West Reactor
PA	prompt action
pCi	picocurie

PDF	Portable Document Format
PF	Plutonium Facility
PHA	pulse height analysis
POC	probability of causation
PR	peer reviewer
PUQFUA	plutonium body burden (Q) from urine analysis
RaLa	radioactive lanthanum
RAS	radiometric alpha spectroscopy
RBM	red bone marrow
RIR	Radiological Incident Report
RTG	radioisotopic thermoelectric generator
s	second
SEC	Special Exposure Cohort
SRDB Ref ID	Site Research Database Reference Identification (number)
TA	Technical Area
TBD	technical basis document
TIMS	thermal ionization mass spectroscopy
TRU	transuranic
U.S.C.	United States Code
UF	uranium fluorophotometric
UNAA	uranium analysis using DNAA
UPPU	You Pee Plutonium (Club)
UR	uranium radiometric (extraction chemistry with radiometric alpha proportional counting)
VFP	volatile fission product
WGPu	weapons-grade plutonium
wk	week
X10	plutonium (LANL designation)
yr	year
μBq	microbecquerel
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 historical background information and guidance to assist in the preparation of dose reconstructions at particular sites or categories of sites. They will be revised in the event additional relevant information is obtained about the affected site(s). These documents may be used to assist NIOSH staff in the completion of the individual work required for each dose reconstruction.

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

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

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

- 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

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

### 5.1.1 Purpose

The purpose of this technical basis document (TBD) is to provide information about occupational internal dose at the Los Alamos National Laboratory (LANL).<sup>2</sup> Occupational internal dose is the dose that was received by an individual from an intake of radioactive material while performing tasks in LANL buildings and structures or from activities outside buildings, such as burial of waste and monitoring of tests, where intakes of radioactive material could occur.

### 5.1.2 Scope

This document contains information for reconstruction of occupational internal doses at LANL facilities throughout its history. LANL has a long history of research and production missions, and there is potential for exposures to a diverse set of radioactive materials. Early controls on the use, handling, and storage of radioactive material were based on maintenance of intakes below an assumed safe exposure or tolerance level, so measurements and recordkeeping practices might not support accurate dose reconstructions in all cases. For these reasons and others (see section 5.1.3), three classes of LANL employees have been added to the Special Exposure Cohort (SEC) as described in Section 5.1.3. The addition of these classes of employees has limited the scope of potential internal dose reconstructions at LANL to the following.

- Internal dose from tritium from 1950 to the present;
- Internal dose from polonium from 1944 to 1956;
- Internal dose from plutonium from 1944 to the present;
- Internal dose from uranium from 1943 to the present;
- Internal dose from other radionuclides present at LANL may be reconstructed only in cases where internal dose monitoring data may be used for any individual claim, in accordance with the NIOSH dose reconstruction processes described in this TBD.

The remainder of this section describes the SEC, certain historical events that are important to internal dose reconstruction, and the overall approach to internal dose reconstruction for LANL workers. Sections 5.2 and 5.3 discuss *in vitro* and *in vivo* bioassay, respectively. Section 5.4 describes interferences and uncertainties, and Section 5.5 discusses the treatment of unmonitored intakes. Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 5.6.

Attachment A provides materials for reconstruction of occupational internal dose for monitored workers. Attachment B discusses bioassay for radionuclides other than primary radionuclides, and Attachment C describes the respiratory protection program. Attachment D contains descriptions of some incidents that resulted in internal doses, and Attachment E describes scaling of coworker dose intakes consistent with type and duration of exposures. Attachment F discusses a bounding dose estimate for potential intakes of radioactive material for onsite responders to the Cerro Grande Fire of 2000.

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<sup>2</sup> The facility was known as the Los Alamos Scientific Laboratory (LASL) until January 1981, when the name was changed to Los Alamos National Laboratory. For convenience, this TBD uses LANL for all years of operation.

### 5.1.3 Special Exposure Cohort

The Secretary of the U.S. Department of Health and Human Services has designated four classes of LANL employees as additions to the SEC based on the findings and recommendations of NIOSH and the Advisory Board on Radiation and Worker Health Board:

- Employees of DOE predecessor agencies and their contractors or subcontractors who were monitored or should have been monitored for exposure to ionizing radiation during radioactive lanthanum (RaLa) operations at Technical Area (TA)-10 (Bayo Canyon Site), TA-35 (Ten Site), and Buildings H, Sigma, and U (in TA-1) for a number of workdays aggregating at least 250 workdays during the period from September 1, 1944, through July 18, 1963, or in combination with workdays within the parameters that have been established for one or more other classes of employees in the SEC (Leavitt 2006; NIOSH 2006).

Through the course of ongoing dose reconstruction and research, NIOSH determined that, due to undocumented worker movements across the site and limited claimant-specific information pertaining to work locations, it was unable to eliminate any specific worker from potential exposure scenarios based on assigned work location. NIOSH has found that a determination cannot always be made as to whether or not an employee worked in Technical Areas with a history of radioactive material use, or whether an employee should have been monitored for radiological exposures. Accordingly, NIOSH determined that it was necessary to remove the area-specific and monitoring criteria from the class description above, and to expand the SEC class definition to include all areas of LANL, and all employees of the DOE, its predecessor agencies, and their contractors and subcontractors who worked at LANL during the specified time period, regardless of monitoring. For this reason, the class of employees listed below was added.

- All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Los Alamos National Laboratory in Los Alamos, New Mexico from March 15, 1943 through December 31, 1975, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort (Sebelius 2010).
- Employees of DOE, its predecessor agencies, and DOE contractors or subcontractors who were monitored or should have been monitored for radiological exposures while working in operational TAs with a history of radioactive material use at LANL for an aggregate of at least 250 workdays during the period from March 15, 1943, through December 31, 1975, or in combination with workdays within the parameters that have been established for one or more other classes of employees in the SEC (Leavitt 2007; NIOSH 2007).
- Employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at LANL from January 1, 1976, through December 31, 1995, 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 (Sebelius 2012; NIOSH 2012).

NIOSH has determined that it does not have access to sufficient information – including internal personnel dosimetry, workplace monitoring data, or sufficient process and radiological source information – that would allow it to estimate with sufficient accuracy the potential internal doses workers might have received. NIOSH has determined that there is insufficient information either to estimate the maximum radiation dose for every type of cancer for which radiation doses are reconstructed that could have been incurred under plausible circumstances by any member of the classes or to estimate the radiation doses of members of the classes more precisely than a maximum

dose estimate. NIOSH has determined that it is possible to reconstruct or bound external doses, occupational medical doses, and the occupational internal doses listed in Section 5.1.2.

#### **5.1.4 Background of Internal Exposures**

When operations began at LANL in 1943, the only method of monitoring intake was through loose contamination swipes. Swipes with a lightly oiled filter paper were taken of surface areas likely to be contaminated. Any swiped area with an activity of more than 500 cpm (1,000 dpm) alpha required decontamination. (The efficiency of the stationary counters used to count the swipes approached 50%.) In addition, nasal swipes (also called nose counts) were used to indicate potential intakes. Nasal swipes with alpha activity higher than 50 cpm indicated the need for follow-up bioassay. The first air samplers became available in the fall of 1944. However, the swipe technique continued as the primary method of detection in many areas until the early 1950s. Respiratory protection equipment (e.g., assault gas masks or respirators) was used as early as 1944 (Oppenheimer 1944).

In 1943, radiation hazards of the project were limited to external radiation from the cyclotron, the Van de Graaff accelerator, radium sources, and a few micrograms of plutonium that arrived in the summer of 1943. In the spring of 1944, the first milligram quantities of plutonium arrived at LANL. Until that time, the laboratory was in the construction phase. There were also internal and external radiation hazards from uranium (ENSR 2002).

In 1944, the radiological hazards of plutonium had been recognized, although it was not yet realized that plutonium was more hazardous than radium. Safety regulations based on experience with radium dial paint plants were established. Measures to control personnel exposures included multiple changes of clothing, showers before leaving the building, use of surgical gloves and respirators, and use of closed systems whenever possible. These measures were primitive by current standards. Most workers cooperated with safety rules to the best of their ability, but the potential for contamination and intakes was present. During the tension and feverish activity of the development of the first atomic bomb, it was difficult to avoid some shortcuts in the observation and enforcement of safety rules (Hempelmann, Richmond, and Voelz 1973). The Laboratory went from handling a few micrograms of plutonium in 1943 to kilogram quantities in 1945. This provided little time for the usual development of safe methods of handling and safety equipment design (Schulte and Meyer 1957). As research determined that plutonium was more hazardous than radium, tolerance levels and maximum permissible body burdens (MPBBs) were reduced significantly. The MPBB for plutonium began at 5  $\mu\text{g}$  (0.33  $\mu\text{Ci}$ ) in 1943. This was reduced to 1  $\mu\text{g}$  (0.07  $\mu\text{Ci}$ ) in October 1944 and further to 0.6  $\mu\text{g}$  (0.04  $\mu\text{Ci}$ ) in 1951 (Langham et al. 1962).

##### **5.1.4.1 Early Research with Humans**

The Plutonium Experiment, which involved plutonium tracers, was undertaken between April 1945 and July 1947. This was a joint project between LANL and the Atomic Energy Project of the University of Rochester School of Medicine and Dentistry. If there are references to involvement in human experimentation in the dosimetry records or the telephone interview, and they are determined to be relevant to the dose reconstruction, then details for this experimentation are available in Section A.15.

##### **5.1.4.2 Early Safety Efforts**

Early safety efforts were based on working in safe contamination levels (LASL 1944a). In 1952, the Health-Safety Rules for Building 52 included restriction of access only through locker rooms and release of clothing with levels less than 100,000 cpm (LASL 1952a). Smoking was permitted in various contaminated areas at least through 1952 (LASL 1952b). In 1954, contamination measurements were based on contact with a shield-open Geiger-Müller (GM) tube for beta/gamma

and a Pee Wee probe of a 55-cm<sup>2</sup> area for alpha. The efficiency of these portable probes is approximately 10% to 15%. Swipes that were counted on a fixed proportional counter had an efficiency of approximately 40% to 50%, although the oil on the swipes that increased the collection efficiency might have decreased the counting efficiency slightly. A total alpha count rate of 500 cpm on a swipe corresponded to 0.007 µg or 0.0004 µCi of the plutonium isotopic mixture of the times (Hempelmann, Richmond, and Voelz 1973).

Tolerance limits were established as the level below which the risk of health effects was considered acceptable (safe) to continue work or to not take immediate action to correct the condition (LASL 1945). The tolerance limit for wounds was 10 cpm for alpha and 0.15 mrep/hr for beta/gamma, except wounds contaminated with <sup>90</sup>Sr, for which the tolerance limit was 0.05 mrep/hr. The skin contamination tolerance limit was 1 mrep/hr for beta/gamma, except for <sup>90</sup>Sr, which was 0.05 mrep/hr. The skin contamination tolerance limit for alpha emitters was 1,000 cpm for polonium, 500 cpm for Tuballoy or oralloy, and 250 cpm for plutonium. Decontamination was required when swipe results were greater than 500 cpm. Floors in laboratories were mopped once or twice a day to maintain safe contamination levels (Hempelmann, Richmond, and Voelz 1973). Corrective actions appropriate to the situation (e.g., decontamination of an area, release of personnel with skin contamination) were initiated if tolerance limits were exceeded. Workers exceeding bioassay tolerance limits, as discussed in later sections, might have been restricted temporarily or permanently from working with radioactive materials (Kolodney 1946). Early Health Group reports indicate contamination inside many respirators, which indicates improper storage and handling and poor fit during use (LASL 1944b).

Kilogram quantities of plutonium began to arrive at LANL in April 1945. At that time, portable alpha counters, continuously operating air samplers, supplied air lines, and specially made positive pressure masks were available. Procedures were performed in open hoods and wooden dry boxes, which were the precursor to the modern glovebox. Research indicated that there was potential for work and casual encounters with plutonium and other radionuclides at various air concentrations or surface contamination levels, including levels that exceeded radiation exposure or control limits. Given that these operations were, in many cases, the first of their kind and that health physics practices were being developed and implemented at the same time plutonium processes were being brought on line, some level of chronic or episodic intake during this early period would be a reasonable assumption for any worker in the Laboratory (Hempelmann, Richmond, and Voelz 1973). The *Health Safety Report, Chemical and Metallurgical Division, May, 1944* (LASL 1944c), discussed the contamination surveys from D-118 and D-119 as an example. "These data are further proof that both wet and dry material are airborne. They emphasize the necessity for assuming that every uncovered item in the laboratory is contaminated" (LASL 1944c). Therefore, exposure to some amount of airborne contamination is a reasonable assumption for all workers during this early era, regardless of job title, even construction trade workers and janitors. Consideration of this fact is a part of the basis for the additions of classes of LANL workers to the SEC as discussed above.

From 1943, the Health Group was responsible for the establishment of health standards, specifically for safe levels of exposure to radiation and to radioactive and chemical materials. The Health Group's primary concern was to protect the health of Laboratory employees. Formalized Health Rules were established for various areas (Burke 1946; Tribby 1946a). Until mid-1951, for want of adequate staff, the Group accepted help for monitoring radiation-related activities from staff members in the Chemistry and Metallurgy Research (CMR) organization. Over the years, the Health Group evolved into the Health Division [and its successors the Environmental, Safety, and Health (ESH) Division and the Health, Safety, and Environment Division], with groups in each to address health physics, medical, (industrial) safety, biomedical research, industrial hygiene, industrial waste treatment, and environmental studies. While several division and group name changes have occurred since 1943, the generic Health Physics Group has existed since 1951. Throughout this period, that group has had the responsibility for assigning and scheduling bioassay analyses for intakes of all radioactive materials. Until the late 1990s, the Industrial Hygiene Group performed all bioassay analyses. Since

the late 1990s, bioassays have been performed by one of the chemistry groups. The Medical Group has treated individuals accidentally exposed to radiation and radioactive materials, performed physical examinations, and treated industrial accidents.

Over the years, many improvements have been made in monitoring, bioassay techniques, safety equipment, and safety procedures (Schulte and Meyer 1957). Nevertheless, the potential for monitored and unmonitored intakes has existed throughout the history of the site.

Nuclides with the widest historical and current application throughout the LANL facilities are:

- Tritium ( $^3\text{H}$ ),
- Uranium ( $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ ),
- Plutonium ( $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$  and, to a lesser exposure significance,  $^{242}\text{Pu}$  and  $^{244}\text{Pu}$ ),
- Polonium ( $^{210}\text{Po}$ ) through the late 1950s, and
- Americium ( $^{241}\text{Am}$ ).

These radionuclides of primary internal dosimetric concern are listed in Laboratory reports from 1943 to the present. (These are referred to as *primary* radionuclides throughout the remainder of this TBD.) Other radionuclides, such as the mixed fission products (MFPs) and activation products (mixed activation products, or MAPs)  $^{14}\text{C}$ ,  $^{11}\text{C}$ ,  $^{13}\text{N}$ , and  $^{75}\text{Se}$ , are associated with work areas and years of operation in Attachment A, Table A-8 (Inkret et al. 1998a). The fission products  $^{140}\text{La}$ ,  $^{140}\text{Ba}$ ,  $^{90}\text{Sr}$ , and  $^{89}\text{Sr}$  that are associated with RaLa operations (September 1, 1944, through July 18, 1963) are also of significance. Dose reconstruction has been determined to be infeasible for radionuclides other than primary radionuclides in the period before January 1, 1976. This includes doses from RaLa operations, as the doses from these operations did not result from primary radionuclides. From this date through December 31, 1995, NIOSH has determined that it is feasible to reconstruct doses for which internal dose monitoring information is included in individual claim files, or in cases in which doses from primary radionuclides may be assigned using coworker intakes (NIOSH 2012).

Work areas include plutonium facilities ( $^{238}\text{Pu}$  or  $^{239}\text{Pu}$ ), uranium facilities, polonium facilities, tritium facilities, laboratory facilities, reactors, accelerators, and others. Case files might not have specific information about the assigned work areas of individuals, but detailed work histories, dose monitoring information, and incident reports might be found in the documents from DOL. When information about the work location is available, Table A-8 in Attachment A can be used to determine the probable nuclides. Section 5.2 discusses *in vitro* methods for specific radionuclides. Excreta bioassay methods for determining internal exposures were developed for plutonium in late 1944 (fully implemented in April 1945), for polonium in 1944, for uranium in 1949, and for tritium in 1950. Only workers with a significant potential for exposure were monitored. A survey in 1986 estimated that approximately 350 persons had known burdens of plutonium.

Starting in 1944, blood tests were performed after potential exposures. These were typically performed for blood count parameters in relation to external radiation exposure or the probability of poisoning rather than the concentration of radioactive material in the blood (Kolodney 1946); the results are not generally applicable to internal dose calculations.

Air samples, which might be identifiable in relation to an individual's record, were collected and analyzed beginning in 1944 but are not routinely provided with the LANL records.

As the state of the art of radiation detection progressed, whole-body counting for fission products began in 1955, wound counting began before 1967, and chest counting began in 1970. Section 5.3 discusses historical and current *in vivo* bioassay methods.



### 5.1.4.3 Bioassay Program

Before the 1970s, individuals were assigned to a bioassay program as determined by the area health physics monitors. LANL deemed this program sufficient to ensure that all workers who might require monitoring were monitored. However, instances might have occurred, especially in the early history, in which a person not normally assigned to radiation work was asked to participate as a substitute in a task that involved radiation or radioactive materials. These persons were not likely to have regularly, or possibly ever, participated in the bioassay program. It is possible that their participation in these tasks was never recorded. Indications of this type of exposure might come from claimant interviews or work history statements. For this reason, potential unmonitored exposures should be considered in many cases for LANL workers. However, NIOSH has determined that it is infeasible to estimate internal dose other than those based on case-specific bioassay monitoring information or those that can be based on coworker intakes for primary radionuclides for all workers before January 1, 1996.

In the 1970s, LANL initiated the use of an Employee Health Physics Checklist. This checklist allowed the evaluation of each individual for potential internal and external exposure. Individuals were placed on a monitoring schedule based on this checklist. The checklist is still used and was computerized in 1998 as the Dosimetry Enrollment System.

#### 5.1.4.3.1 Individual Bioassay Results

The Oak Ridge Associated Universities (ORAU) Team has generated a database from the LANL Bioassay Data Repository (LABDR in this document) to consolidate dosimetry records and make these data available for use in the dose reconstructions. The repository consists of several applications including the Bioassay Enrollment Scheduling and Tracking, In Vivo Measurements Laboratory (IVML), and Radiological Incident Reports (RIR) systems, as applicable. Electronic data from many other sources were collected and uploaded as part of the project. The new database is referred to as the ORAU Team LANL Bioassay Database in this document (OTLBD). The OTLBD database consists of the *in vitro* records for only the five major analytes of dosimetric significance – uranium, plutonium, americium, polonium, and tritium – and the results of *in vivo* bioassay. Other bioassay might have been performed for a worker but the results have not been captured in the database. The results will most likely not be supplied by LANL without special requests. Autopsy results are clearly labeled (ORAUT 2009a).

Bioassay results were supplied to the ORAU Team in both Portable Document Format (PDF) and Microsoft Excel formats; however, dose reconstructors are to use the PDF files in the case records system. In both types of file, the information is divided into five sections: the demographics of the worker, Current *In Vitro* Analysis Data (1990 to the present), Historical *In Vitro* Analysis Data (pre-1990), *In Vivo* Analysis Data, and Incident Data. Specific details about each section of the report are discussed below in the sections on the relevant radionuclides and in Attachment A.

A description of the bioassay data that were reviewed and used to populate the OTLBD, along with the detailed validation and verification that were performed as part of that process, is being compiled in ORAUT-OTIB-0063, *Los Alamos National Laboratory Bioassay Repository Database* (ORAUT 2009a). As a result of this project, LANL management and the NIOSH Project Manager have determined that the level of verification and validation is acceptable given the financial constraints of the Bioassay Data Repository Project. The validation and verification process is described in ORAUT (2009a).

Each PDF file has a page with the description of the database and a summary of the validation process of the data. A section on Data Use Instructions is included. The information in these sections is not repeated here because it might be modified if the format of the files is revised in the future. These sections in the PDF file should be reviewed for each energy employee in the event that

certain parameters might have been modified or updated in future files. Units and codes are clearly identified. Units might not be consistent within a sample result. For example, the sample result might be listed in picocuries per 24 hours, and the minimum detectable activity (MDA) might be listed in picocuries per sample or aliquot. Units of activity per day and activity per sample or aliquot are not necessarily equivalent.

Descriptive codes and format descriptions are listed in each PDF file. Current codes are described in Attachment A. Information on documented incidents might be reported in the PDF file. This section might have captured recorded incidents that were listed in the original *in vitro* bioassay plutonium and americium database. Other incident information might not have been captured in the database.

Records associated with individual claims may indicate that chelation therapy was administered. Approximately 35 workers are known to have received chelation therapy for plutonium and americium intakes at LANL, and notes about chelation therapy might be found in the comments section of the PDF files. The comment "URINEfix" indicates a bioassay sample that is associated with chelation. Chelation therapy was listed as a code in the LABDR, but only the comments (not the codes) were transferred to the current OTLBD. It is assumed that every chelation event in the original LABDR included comments, but this is not ensured. Records of administration of chelating agents, associated with individual worker names, exist in the project database.

#### **5.1.4.3.2 UPPU Club**

One small but significant group of workers is the UPPU Club (intended to be pronounced as "You Pee Pu"). This group consists of individuals who accumulated a significant plutonium body burden and who agreed to be monitored periodically and continue to be monitored even past the end of their employment at LANL. Only two members were added to the group after the initial 1951 startup. Membership in this group can be noted in claimant interviews. Bioassay results can be found in the individual's record many years past the end of employment or past the time of potential exposure. This group has typically been monitored at 5-year intervals (Hempelmann, Richmond, and Voelz 1973).

#### **5.1.5 Unmonitored Workers and Unreported Bioassay Results**

The following sections describe several types of employees for which potential unmonitored internal doses must be considered when reconstructing internal doses. In each case, a review must be performed for potential unmonitored internal doses using case information. Programmatic problems identified in the Tiger Team assessment (DOE 1991) indicate that the potential exists for unmonitored intakes. Unmonitored intakes may be estimated using ORAUT-OTIB-0062, *Internal Dosimetry Coworker Data for Los Alamos National Laboratory* (ORAUT 2009b). However, doses from potential intakes of other radionuclides may only be based on dose monitoring information in claim files. Because of the potential for internal dose that is not reflected in internal dose monitoring information, classes of LANL employees have been added to the SEC that include all workers in the period before January 1, 1996.

##### **5.1.5.1 Workers Before 1949**

The potential effects of exposure to plutonium were recognized early by its discoverer, Glenn Seaborg (Hempelmann, Richmond, and Voelz 1973). Therefore, when the first few micrograms of plutonium arrived at LANL in 1943, there was awareness of some of the radiological and biological effects and hazards. Stringent safety measures were put in place immediately, including the use of homemade dry boxes when practical. "However, because of the urgency of the times, work with plutonium had to proceed, and improvised methods of monitoring and decontamination were unbelievably primitive by today's standards" (Hempelmann, Richmond, and Voelz 1973). In 1944, bioassay techniques

consisted of swipes of both nostrils at the end of the working day. Monitoring results were described as whether maximum permissible concentrations (MPCs) were exceeded, not whether there was an intake. When kilogram quantities of plutonium arrived at LANL in 1945, the surface swipes and nasal swipe counting (as primary monitoring) had been replaced in certain areas by continuously operating air samplers and portable alpha counters. Then, by March 1945, a urine assay method for plutonium had been developed. Body burden estimates were made from urine results as early as 1953.

Working conditions were described as “deplorable by present-day standards” (Hempelmann, Richmond, and Voelz 1973) until September 1946, when the new facility at DP Site (TA-21) was constructed. Before the move to TA-21, suspension of work and decontamination of the area were required when alpha contamination exceeded 10,000 cpm (assume about 50% counting efficiency). Decontamination was recommended between 2,000 and 10,000 cpm but was not required. Even hallways and other unrestricted areas had contamination. In 1944, shoe covers that were worn by secretaries and others working throughout the buildings had count rates of 2,500 to 7,500 cpm (LASL 1944b). Contamination above 500 cpm (0.007  $\mu\text{g}$  or 0.0004  $\mu\text{Ci}$  of the plutonium isotopic mixture) was the reporting level for plutonium areas. In June and July 1945, over 50% of the laboratories had areas that routinely exceeded the maximum removable contamination level. The potential for unmonitored intakes was significant in the early years (1944 to 1946) for any site worker, but bioassay was provided only for the most exposed workers. Because the excretion of plutonium is continuous after an intake, significant intakes might have been identified on later routine bioassay samples with improved sensitivity for workers who remained at LANL.

As bioassay sensitivities and respiratory protection equipment improved, the potential for intakes decreased. Due to rigorous workplace monitoring, the probability that a worker could have received a large intake of radioactive material that was unmonitored and unnoticed was less after 1946, although the probability of unmonitored small intakes was larger. Periodic reports from H Division of air samples, contamination incidents, and hot spots continued to identify a significant number of over-tolerance occurrences throughout the 1960s. Respiratory protection was available but, except in a few locations, was only donned when a continuous air monitor (CAM) alarmed or when airborne levels approached tolerance or action levels (ALs). Therefore, a potential existed for an intake before the alarm. Review of *Summary of Radiological Incident Reports (RIR), January 1993 to June 1998* indicates that, while the number of RIRs has decreased significantly, the potential for unmonitored small intakes continues to exist (Bates 1998).

In addition, instances might have occurred, especially in the early history, in which a person who was not normally assigned to radiation work was asked to participate as a substitute in a task that involved radiation or radioactive materials. These workers were not likely to have regularly, or perhaps ever, participated in the bioassay program or potentially had their participation in the task recorded. Indications of this type of exposure might come from claimant interviews.

#### **5.1.5.2 Short-Term Workers**

Short-term workers, such as summer students, persons engaged in postdoctoral work, contractors, and teachers, might not have fully participated in routine bioassay programs. These workers were monitored for internal exposure only in unusual circumstances. Near their termination dates, workers might have received *in vivo* counts. Workers might have been required to submit an initial urine sample or have an initial *in vivo* count.

#### **5.1.5.3 Zia Company and Other Contract Service Workers**

The Zia Company was the service worker contractor. Zia employees participated in a separate monitoring program from that for Laboratory employees. As early as 1946, directives restricted the assignment of maintenance workers to either DP East or West (not both) because of the difficulty of

separating the radionuclides in urine bioassay samples and monitoring DP East workers for plutonium when they might have been exposed in both areas (Burke 1946; Tribby 1946a). In 1954, that restriction was removed because of the improvement in the bioassay techniques that would permit the specific radionuclides to be identified. However, although listed as a monthly requirement for hazardous areas as early as 1946, submission of urine samples was dependent on determination of a "high nose count" or an "extremely hazardous job" or at the request of the DP East Section Leader (Meyer 1954).

In 1975, it was determined that the H-1 and H-5 efforts to schedule plutonium urine samples for Zia employees were inadequate. Only Zia employees who were permanently assigned to DP West and the CMR Building were being sampled; other Zia employees in plutonium areas were not. To ensure adequate coverage of Zia employees, the scheduling of plutonium samples for Zia was delegated to the Zia Safety Office. The requirement was annual samples for those employees "required to work in Pu areas" (Lawrence 1976). A quarterly report of the H-1 Division stated that Zia management had interpreted this to mean employees "required to enter Pu areas for any reason," (University of California 1978a p. 208), whereas a number of individuals in these areas would not have had a potential for intake of plutonium. "By July 1976 it became apparent that the number of Zia employees being sampled for Pu was growing without bounds". A program review was conducted in 1978, in which it was shown that the number of bioassay samples had "exceeded the level of effort agreed upon by Zia management," (University of California 1978a). In 1978, in another attempt to reduce the number of sampled Zia employees to 500 per year; supervisors who only performed inspections were eliminated from the schedule. In later years, some jobs were exempt from monitoring. Attachment A, Table A-9, contains a list of criteria and exempt job categories (University of California 1978a). In more recent years, other service contractors have participated in site activities.

#### **5.1.5.4 RaLa Project Operations, 1944 to 1963**

Workers on the RaLa Project from September 1, 1944, through July 18, 1963, might or might not have been monitored for potential intakes of  $^{90}\text{Sr}$ ,  $^{89}\text{Sr}$ ,  $^{140}\text{La}$ , and  $^{140}\text{Ba}$ ; although it appears that RaLa workers at TA-10 (Bayo Canyon Site), TA-35 (Ten Site), and Buildings H, Sigma, and U (in TA-1) were monitored for external radiation. A total of about 250,000 Ci of RaLa was released to the atmosphere from these experiments. The highest exposures at LANL from RaLa operations were to the chemists who prepared the sources.

*In vitro* bioassay results related to RaLa operations have not been found in claim files to date. The whole-body counting program was not well established until 1970, although there are reports as early as 1957 that identified specific nuclides for workers at the cyclotron (Buckland 1959). Workplace air monitoring data for the RaLa areas also have not been found. Therefore, estimates of intakes and assessment of internal dose are not possible for RaLa operations workers (NIOSH 2006).

#### **5.1.5.5 Responders to the Cerro Grande Wildlands Fire of 2000**

In May of 2000, areas of Los Alamos were overrun by the 48,000-acre wildlands fire that came to be known as the Cerro Grande fire. Some LANL structures were destroyed and damaged, but there was no damage or destruction of stored special nuclear material. According to meetings conducted with personnel employed by LANL, responders to that incident included LANL staff and members of outside emergency response organizations who were not monitored for potential intakes of radioactive material.

During the fire, sampling of airborne particulate was conducted using LANL's AIRNET system. The most abundant detected radioactive materials resulted from resuspension of radon progeny that had accumulated on vegetation and on the forest floor, but concentrations of plutonium, americium, and uranium were also measured. These results were, in general, consistent with measurements that

were made outside the fire period (Eberhart 2010). For estimation of doses from potential intakes of anthropogenic radionuclides, the results of the monitoring were evaluated as described in Attachment F. Doses to a responder in the area of the highest measured concentration, assuming a 60-hour exposure, are listed in Tables F-2 and F-3, in Attachment F.

Because no organ dose was greater than 0.001 rem, no additional unmonitored dose, beyond doses from environmental concentrations of radioactive material, are necessary for potential exposure during the Cerro Grande fire.

### 5.1.6 Overview of Internal Dose Reconstruction Process for Los Alamos National Laboratory

As described in the preceding portions of this introduction, doses inferred from internal dose monitoring records might not completely represent the total dose that was received by a LANL employee. NIOSH has determined that it is not feasible to reconstruct internal doses when bioassay information is not present in claim files. Two types of internal exposures must be addressed in dose reconstruction for LANL workers: doses based on available internal dose monitoring records (measured and missed doses) and unmonitored intakes, which may be estimated using coworker intakes based on bioassay data for primary radionuclides for monitored workers.

The dose reconstructor must review the records in the case with special care to identify all potential internal doses, including potential unmonitored intakes. This evaluation will result in two possible methods for estimating internal dose: (1) calculating the dose from bioassay results (missed and fitted internal doses); and (2) assigning the dose based on coworker dose intakes in ORAUT-OTIB-0062, *Internal Dosimetry Coworker Data for Los Alamos National Laboratory* (ORAUT 2009b).

Calculation of internal doses is discussed in Sections 5.2 to 5.4, which address *in vitro* bioassay, *in vivo* bioassay, and interferences and uncertainties, respectively. In addition, a methodology is given from ORAUT-OTIB-0062 (ORAUT 2009b) to allow the assignment of a dose using coworker dose intakes.

### 5.2 **IN VITRO BIOASSAY MINIMUM DETECTABLE AMOUNTS, COUNTING METHODS, AND REPORTING PROTOCOLS FOR PRIMARY RADIONUCLIDES**

Historically, the *in vitro* bioassay program included the nuclides and techniques listed in Tables 5-1 and 5-2. This section contains a detailed discussion of the analysis techniques that were used for selected radionuclides. Table A-24, Attachment A, summarizes *in vitro* bioassay sensitivities by technique and year. However, without a special request, only the results for the primary radionuclides (tritium, uranium, plutonium, <sup>210</sup>Po, and <sup>241</sup>Am as listed in Table 5-1) will be supplied for the worker. If an exposure to primary radionuclides that is not monitored by *in vitro* bioassay is suspected, dose is assigned based on the coworker intakes in ORAUT (2009b). In some cases, special inquiries can be made to determine if additional bioassay results exist for the worker (e.g., MFPs or <sup>90</sup>Sr).

Table 5-1. Historical *in vitro* bioassay, primary radionuclides.

Material	Start year	Comments
Tritium	1950	
Uranium	1949	Significant numbers of results begin in 1950.
Plutonium	1944	
Polonium-210	1944	
Americium-241	1954	

Table 5-2. Historical *in vitro* bioassay not available in claim records.

Material	Start year	Comments
Gross beta	1952 (maybe 1947)	Not done regularly
Protactinium-231	1958	Not done regularly
Radium-226	1958	Not done regularly
Thorium-230	1958	Not done regularly
Actinium-227	1954	Not done regularly
Strontium-90	Unknown	Not done regularly

### 5.2.1 Plutonium

The most serious intakes at LANL have involved isotopes of plutonium and  $^{241}\text{Am}$ . Operations with small quantities of plutonium and curie quantities of  $^{210}\text{Po}$  were conducted primarily in D Building in 1944. The first urinalysis for evidence of plutonium uptake was performed on July 18, 1944, using a chemical procedure that was developed at the University of Chicago Metallurgical Laboratory in June 1944. However, Health Group personnel soon discovered that the procedure, which used a 50-cm<sup>3</sup> aliquot of a 24-hour urine sample, did not have the sensitivity to meet the tolerance level of a 5- $\mu\text{g}$  (0.33- $\mu\text{Ci}$ ) burden of plutonium. One of the first recorded accidents in which a human was subjected to a possible intake of plutonium occurred in May 1944 (Hempelmann, Richmond, and Voelz 1973). A minor chemical exposure on August 19, 1944, was the first recorded accident in which a human was subject to a possible intake of more than the 5- $\mu\text{g}$  plutonium tolerance limit with the release of 10 g of plutonium into the face of a chemist. This accident resulted in LANL being authorized to proceed to develop a more sensitive procedure. The resultant cupferron extraction procedure and subsequent procedures are discussed later in this section (Moss 1990).

References to products called X10 and 49 indicate plutonium or isotopic mixtures of plutonium that are predominantly  $^{239}\text{Pu}$  (Lawrence 2004). (The “49” is a shorthand reference that is a combination of the last digit of the atomic mass number, 94 for plutonium, and the last digit of the atomic weight, 239 for  $^{239}\text{Pu}$ .)

The chemical forms of plutonium that have been currently and historically encountered at LANL include oxide, nitrate, fluoride, and metal. Plutonium oxide is the most common form. However, in lieu of detailed information specific to the case, dose reconstructors typically assign the absorption type resulting in the highest dose. When such information is not present, types M and S are evaluated, and, in accordance with ORAUT-OTIB-0049, *Estimating Doses for Plutonium Strongly Retained in the Lung* (ORAUT 2010), correction factors for certain organs could result in additional dose.

#### 5.2.1.1 **Special Considerations for $^{238}\text{Pu}$ Dosimetry Related to a Wing 9 Incident**

A special case of excretion kinetics has been observed at LANL. The 1971 Wing 9 incident involved intakes of high-fired  $^{238}\text{Pu}$  oxide that exhibited a very low excretion rate for the first 100 days after intake. A gradual rise in excretion rate was observed after that period, with no additional suspected intake (Miller et al. 1999). This has been characterized as a “nonmonotonic” excretion rate, and this phenomenon observed at Los Alamos, along with a similar event observed at the Mound site, are described by ORAUT (2013).

In 1971, an accidental release of  $^{238}\text{Pu}$  occurred in Wing 9 during examination of a radioisotope thermoelectric generator (RTG) powered by a ceramic  $^{238}\text{PuO}_2$ . Intakes from this accident were characterized by nonmonotonic excretion rates by the exposed individuals (Guilmette, Griffith, and Hickman 1994). Parameters for the Integrated Modules for Bioassay Analysis (IMBA) program were developed that mimic these excretion rates, as documented in ORAUT (2013). The parameters are

reproduced here for applications to dose estimates that could be associated with this accident. These parameters, listed as “type J,” are compared with types M and S in Table 5-3.

Table 5-3. IMBA input parameters for types M, S, and nonmonotonic type J.

Rate constant	Type M	Type S	Type J
$k_{p,t}$	90	100	0.00189
$k_{t,blood}$	0.005	0.0001	0.000257
$k_{p,blood}$	10	0.1	1.00 E-06
$k_r$	100	100.1	0.001891
$k_s$	0.005	0.0001	0.000257
$f_r$	0.09995	0.001	-0.156671

Dose reconstructors are cautioned that  $^{238}\text{PuO}_2$  typically exhibits excretion rates somewhere between types M and S (Cheng et al. 2004), and either M or S should normally be assumed. However, the dose from type J should be calculated for individuals who were involved in the 1971 Wing 9 incident. It is not intended that Type J should be applied as an overestimating assumption; only that it is an available assumption for fitting bioassay data.

### 5.2.1.2 Sample Collection Procedures

At the onset of the LANL bioassay program, samples were collected in a clean area after a decontamination shower. However, these samples had a potential for contamination. Occasional high values were assumed to be an artifact of the sample and not evidence of internal exposure (Hempelmann, Richmond, and Voelz 1973). By the spring of 1945, a Health Pass Ward was established at the hospital to ensure collection of contamination-free samples (Nickson 1945). This procedure allowed for 2 days off the job, 1 day to clean up, and one 24-hour period spent at Los Alamos Hospital where the urine sample was collected. The procedure was modified in 1948 to allow only 1 day off the job during the sample collection period. While a contamination-free sample was ensured, this procedure was later deemed extremely expensive and was eliminated in 1952 (Clark 2005). A procedure that allowed collection of an equivalent 24-hour sample while the employee was off the site was initiated. Between 1952 and 1958, the samples were collected in three disposable bottles that were carried in a kit. The employee was to collect four voidings in the three bottles, which some found objectionable and led them to collect only three voidings. The kits were rarely washed and therefore were considered a source of potential contamination. In January 1958 the kit provided four disposable bottles (Clark 2005). A modified procedure to collect a 24-hour equivalent sample (two morning and two evening voidings) with a four-bottle disposable kit continues to be used.

Studies of special, timed spot samples have been performed to provide data that are used to apply correction by volume and specific gravity to provide realistic results in disintegrations per minute per day. All sample results, except thermal ionization mass spectroscopy (TIMS) results, are expected to be reported in units (typically picocuries) per 24-hour day in the database. TIMS results are reported in both activity per 24 hours and activity per sample. No sample volume is usually provided. The current database provides a clear description of units for each result. If MDAs are listed for an analysis, these values might be listed in units of activity per sample with a sample volume. Note that activity per sample and activity per 24 hours are not equivalent. It appears that the 24-hour sample concentrations have been normalized to a 1,400-mL volume.

LANL instituted a sample validation procedure (Lawrence 1978). The Plutonium Body Burden (Q) from Urine Analysis (PUQFUA) programs were revised through PUQFUA4. Therefore, other validation protocols might have been in effect after 1978. The purpose of these programs was to perform a statistical test of high values to determine if these results should be invalidated.

Attachment A, Section A.4, lists the validation procedure for plutonium urine bioassay samples. This procedure is more rigorous than can be implemented without computer software. However, certain samples in the original database were marked invalid based on these criteria. Because the codes were not carried over to the current database, it is expected that these designations have been removed. However, this practice is mentioned in the event that comparison of the results in the current database is made to the original database or logbooks. The dose reconstructor should not attempt to implement this validation procedure but should use best judgment in the evaluation of samples for inclusion in the intake calculations. At present, Bayesian statistics are used by LANL to determine the statistical validity of results. Notes on these determinations are included in the comments in the database. Indication that LANL considered the sample as invalid and the reason for the status might be included in the notes that follow the results in the PDF file. These comments might or might not be relevant to the dose reconstructors determination of the validity of the result.

### 5.2.1.3 Missed Intakes

Intakes of plutonium can occur from both acute and chronic exposures. Chronic exposures might not be identified as incidents but can still result in a measurable burden of plutonium. A body burden can result from chronic inhalation exposure to a low-level plutonium-contaminated atmosphere. A study of autopsy tissues from LANL workers with high and low potential for exposure to plutonium has shown that measurable amounts of plutonium, above that expected from global fallout, could occur in individuals with low exposure potential (Foreman, Moss, and Langham 1960). As noted above, smoking was permitted in various contaminated areas at least through 1952 (LASL 1952b).

The long-term excretion pattern of plutonium isotopes permits plutonium intakes that produced bioassay results below the detection threshold in the early years to become detectable as the sensitivity of the analysis technique improved. The date of the intake might not relate directly to the last bioassay result below the detection level. The intake might have occurred many years earlier (Hempelmann, Richmond, and Voelz 1973).

The first urine tests for plutonium were developed in 1944 and 1945. Urinalysis was difficult and time consuming. Therefore, personnel with the most positive nasal swipes had the most urine bioassays (Hempelmann 1946; ENSR 2002).

### 5.2.1.4 Routine Sample Frequencies

Table A-15 in Attachment A lists the routine sampling frequencies by period. In 1978, sampling frequencies were reevaluated for LANL and Zia employees. To attempt to reduce the number of sampled Zia employees to 500 per year, supervisors who only performed inspections were eliminated from the schedule (University of California 1978a). Special and emergency sample frequencies are listed in Tables A-19 to A-21 of Attachment A.

### 5.2.1.5 Sample Analysis Procedures

At first, the urine bioassay analysis procedure could not distinguish adequately between plutonium and polonium. Plutonium samples in 1944 were occasionally contaminated with polonium (Clark 2005). During this period, the total alpha results were assigned to either plutonium or polonium based on the individual's work history. The procedure was modified in the fall of 1944 to extract the plutonium. The count time was 30 to 60 minutes (Moss 1990).

The cupferron procedure was in use through late 1949. The bismuth phosphate-lanthanum fluoride serial coprecipitation procedure was in use between October 1949 and January 1957. No corrections were made for chemical blanks or counting geometry; average chemical recovery factors (82.3%



±19.4%) might have been used to interpret the data until 1957 (Clark 2005). The alpha proportional counters were in use until 1957.

Improvements were made in the coprecipitation procedure and counting techniques until the procedure was replaced by an aluminum nitrate extraction with neutron track analysis (NTA) (Clark 2005) counting in 1957. Urine samples were radiochemically processed and electroplated, and activities were determined by exposure to NTA emulsions. The exposure time for this method was 10,000 minutes with a background of 0.005 dpm. An ion-exchange technique replaced the aluminum nitrate in 1963 (Clark 2005). Use of a ZnS counter began in 1966. All of these methods measured total alpha activity from plutonium isotopes.

Radiometric alpha spectroscopy (RAS) was available for use beginning in 1967 (Clark 2005). The use of RAS in the analysis is indicated when both  $^{238}\text{Pu}$  and  $^{239}\text{Pu}$  are reported for the same sample. Plutonium-241 and  $^{241}\text{Am}$  were not measured by total plutonium alpha procedures. The dose contribution from these nuclides should be accounted for using the mixtures listed in Tables 5-4, 5-5, and 5-6, unless the plutonium in the intake was known to be a "pure" isotope.

From 1967 to present-day operations, counting has been performed by RAS for specific plutonium isotopes. Alpha spectroscopy cannot distinguish between  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  because of the similarity of the alpha energies. Therefore, the results for RAS that are labeled as  $^{239}\text{Pu}$  are actually  $^{239+240}\text{Pu}$ .

In 1997, TIMS and application of the class 100 clean room were added to the analytical technique. Samples are now routinely analyzed for  $^{239}\text{Pu}$  with TIMS and for  $^{238}\text{Pu}$  with RAS. The sensitivity of the analysis for  $^{239}\text{Pu}$  has been reduced 40-fold by the addition of TIMS (Inkret et al. 1999). TIMS does not measure  $^{240}\text{Pu}$  unless the activity of  $^{239}\text{Pu}$  is above an unspecified threshold, in which case the results for  $^{240}\text{Pu}$  would be reported separately (Lewis 2006a). Therefore, the amount of  $^{240}\text{Pu}$  in the mixture should be accounted for with the use of Table 5-4, 5-5, or 5-6 when  $^{239}\text{Pu}$  results are listed for TIMS.

Dose calculations performed in dose reconstructions, including those with IMBA, typically assume a combination of  $^{239+240}\text{Pu}$  that is treated as  $^{239}\text{Pu}$ . Therefore, if the results of TIMS analysis are used for calculation of intake, the contribution of  $^{240}\text{Pu}$  must be accounted for in the calculation.

In the current LABDR, if TIMS results are listed for a sample the  $^{239}\text{Pu}$  RAS analysis for that sample could be footnoted with an asterisk (\*). The footnote states that  $^{240}\text{Pu}$  activity is not measured in the result. The comment is an artifact of the logic in the database and could be misleading to the dose reconstructor. The flag is applied to all results listed for the sample when TIMS analysis is included. RAS cannot distinguish between the energies of  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ ; therefore the activity of  $^{240}\text{Pu}$ , if any, is included in the RAS activity listed as  $^{239}\text{Pu}$  (Lewis 2006a).

MDAs from procedures and reports for the various radiochemical and counting methods are listed in Table 5-7. The MDAs that are reported with the results in the LABDR are frequently in units of picocuries per sample. The results of the analysis are listed as actual values (positive, negative, or zero) but in units of picocuries per 24-hour sample. These are not equivalent units. The sample size is usually listed. The dose reconstructor should normalize the units before determining if the reported results are above or below the MDA. TIMS results are listed in picocuries per 24-hour sample and picocuries per sample, but no sample volume is typically listed and the listed results are not identical. Therefore, the results in picocuries per 24 hours should be used for the dose reconstruction. No MDA value is typically listed with the TIMS results, so the MDA in Table 5-7 should be used to determine if results are above the MDA.

Table 5-4. Activity composition of nominal 3% plutonium mixture (Kinderman et al. 1953).

Mixture designation:	Fresh	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr
Years of aging: <sup>a</sup>	0	5	10	15	20	25	30
Component	Specific activity in mixture (Ci/g)						
Pu-238	9.85E-04	8.72E-04	8.39E-04	8.06E-04	7.75E-04	7.45E-04	7.16E-04
Pu-239 <sup>b</sup>	6.01E-02	6.01E-02	6.01E-02	6.01E-02	6.00E-02	6.00E-02	6.00E-02
Pu-240	6.74E-03	6.73E-03	6.73E-03	6.73E-03	6.72E-03	6.72E-03	6.72E-03
Pu-241	1.44E-01	1.13E-01	8.92E-02	7.01E-02	5.51E-02	4.33E-02	3.40E-02
Pu-242	<1E-06	<1E-06	<1E-06	<1E-06	<1E-06	<1E-06	<1E-06
Am-241	0	1.02E-03	1.82E-03	2.44E-03	2.92E-03	3.28E-03	3.56E-03
Pu-239+240	6.7E-02	6.7E-02	6.7E-02	6.7E-02	6.7E-02	6.7E-02	6.7E-02
Pu-alpha	6.8E-02	6.8E-02	6.8E-02	6.8E-02	6.8E-02	6.7E-02	6.7E-02
Total alpha	6.8E-02	6.9E-02	6.9E-02	7.0E-02	7.0E-02	7.1E-02	7.1E-02
Component	Activity ratios						
Pu-239+240:Am-241	N/A <sup>c</sup>	66	37	27	23	20	19
Pu-239+240:Pu-238	68	77	80	83	86	90	93
Pu-239 <sup>b</sup> :Pu-240	8.92	8.92	8.92	8.92	8.92	8.92	8.92
Pu-241:Pu-239+240	2.2	1.7	1.3	1.0	0.83	0.65	0.51
Pu alpha:Pu-238	69	78	81	84	87	91	94
Pu alpha:Am-241	N/A <sup>c</sup>	66	37	28	23	21	19
Pu-alpha:Pu-239+240	1.01	1.01	1.01	1.01	1.01	1.00	1.00
Pu-241:Pu alpha	2.1	1.7	1.3	1.0	0.82	0.64	0.50

- a. Time since separation of Am-241 from the plutonium mix.  
b. Use Pu-239 only for TIMS results; otherwise use Pu-239+240 for RAS or total plutonium alpha.  
c. N/A = not applicable.

Table 5-5. Activity composition of reference weapons-grade 6% plutonium mixture (Battelle 2003).

Mixture designation:	Fresh	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr
Years of aging: <sup>a</sup>	0	5	10	15	20	25	30
Component	Specific activity in mixture (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	0	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
Component	Activity ratios						
Pu-239+240:Am-241	N/A <sup>b</sup>	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-239:Pu-240	4.24	4.24	4.24	4.24	4.24	4.24	4.24
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	N/A <sup>b</sup>	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

- a. Time since separation of Am-241 from the plutonium mix.  
b. N/A = not applicable.

Urine, fecal, and tissue samples have been analyzed for plutonium. Most *in vitro* bioassay samples have been urine. Fecal and tissue samples have been performed only if requested in special circumstances.

Table 5-6. Activity composition of reference fuel-grade (12%) plutonium mixture (Battelle 2003).

Mixture designation:	Fresh	5-yr	10-yr	15-yr	20-yr	25-yr	30-yr
Years of aging: <sup>a</sup>	0	5	10	15	20	25	30
Component	Specific activity in mixture (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	0	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
Component	Activity ratios						
Pu-239+240:Am-241	N/A <sup>b</sup>	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-239:Pu-240	1.93	1.93	1.93	1.93	1.93	1.93	1.93
Pu-241:Pu-239+240	3.87E+1	3.05E+1	2.40E+1	1.88E+1	1.48E+1	1.17E+1	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	N/A <sup>b</sup>	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

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

b. N/A = not applicable.

### 5.2.1.6 Plutonium Isotopic Ratios (Mixtures)

Dose assessments at LANL were historically based solely on the isotopes identified in the analysis. At present LANL dose assessments are based on a 6% plutonium mixture. The amount of <sup>241</sup>Am present (i.e., the aging) in a sample with positive <sup>239</sup>Pu or <sup>238</sup>Pu is determined by analysis for <sup>241</sup>Am or by process knowledge. If the determination of the mixture is based on process knowledge, the amount of <sup>241</sup>Am could be noted in the comments. The dose reconstructor should evaluate <sup>241</sup>Am results for the intake. There could be a short interval between the plutonium and the americium samples (Lewis 2006b).

Most plutonium mixtures at LANL before 1957 should be assumed to be nominally 3% weapons-grade plutonium (WGPu) and after 1956 to be nominally 6% WGPu (Lewis 2006b). Tables 5-4 and 5-5 list activity and weight ratios as referenced for DOE sites for 3% and 6% WGPu, respectively.

In the early years, when total alpha measurements were made for plutonium, <sup>239</sup>Pu was the isotope chosen based on process knowledge. Results of these total plutonium alpha measurements before 1967 are found in the database and listed as <sup>239</sup>Pu. Since 1967, plutonium analysis has been performed with RAS. RAS cannot distinguish between <sup>239</sup>Pu and <sup>240</sup>Pu because the alpha energies are similar and overlapping. However, <sup>239+240</sup>Pu and <sup>238</sup>Pu can be resolved using RAS. Therefore, when quantities of pure <sup>238</sup>Pu arrived at LANL in 1968, the analysis was capable of distinguishing between the isotopes of plutonium. If the results of both isotopes are not reported as positive, then exposure to a mixture of predominantly pure <sup>238</sup>Pu is usually indicated when the <sup>238</sup>Pu results are positive and the <sup>239</sup>Pu results are below the MDA.

Based on the isotopic composition of the material that was released to the atmosphere in 1970 during an accident in TA-21 (DP West) involving "pure" <sup>238</sup>Pu (by weight, with a specific activity of 14.1 μCi/μg), a typical mixture consists of 80% <sup>238</sup>Pu, 16.9% <sup>239</sup>Pu, 2.8% <sup>240</sup>Pu, 0.2% <sup>241</sup>Pu, and 0.1% <sup>242</sup>Pu (Meyer 1970). "Pure" <sup>238</sup>Pu was typically present in areas involving heat source technology. A typical pure <sup>238</sup>Pu mixture is described in Table 5-8. It is assumed that pure <sup>238</sup>Pu would be "fresh"

Table 5-7. Plutonium bioassay sensitivity as listed in procedures and reports.

Nuclide	Period	Sample type <sup>a</sup>	Technique (era)	MDA level Unit/24-hr sample <sup>b</sup>	Reporting limit Unit/24-hr sample <sup>b</sup>	Tolerance limit <sup>c</sup> Unit/24-hr sample <sup>b</sup>
Pu-total alpha	1944	U	Cupferron <sup>d</sup>	0.7 pCi (Clark 2005) <sup>e</sup>	>0.8 <sup>e</sup> pCi	7 cpm/24 hr or 6.3 pCi
	1945–1949	U	Cupferron <sup>d</sup>	0.16 to 0.05 pCi <sup>f</sup> (1 dpm [0.45 pCi] DL [Moss 1990])	>0.8 <sup>e</sup> pCi	7 cpm/24 hr or 6.3 pCi
	1949–01/1957	U	Biphosphate/alpha counting	0.20 to 0.07 pCi <sup>f</sup>	2 dpm or 0.9 pCi 0.4 pCi <sup>g</sup>	7 dpm/24 hr or 3 pCi
	01/1957–1965	U	Aluminum nitrate/NTA	0.03 pCi <sup>h</sup> (0.05 dpm at 99% confidence) <sup>i</sup>	0.2 dpm or 0.09 pCi	7 dpm/24 hr or 3 pCi
	1966	U	ZnS	0.03 pCi <sup>j</sup> (0.07 dpm)		
Pu-239	1967–1997	U	RAS (PHA) only starting in 1971	0.03 pCi <sup>j</sup> (1 mBq)		
	1977–1981	F	RAS (PHA)	1 nCi/sample (less if Am-241 ratio known)		
	1981–1983	F	Phoswich detector, 4-cm sample thickness	0.4 nCi/sample or 400 pCi/sample (17-keV X-rays)		
	1982–1986	U	Alkaline earth <sup>i</sup> Oxalate <sup>i</sup> Rapid <sup>i</sup>	Alkaline earth - 0.015 pCi/800 cm <sup>3</sup> Oxalate - 0.015 pCi/700 cm <sup>3</sup> Rapid - 1.6 pCi/L		
	1997–2002	U	TIMS/RAS (alpha spec <sup>k</sup> )	1.03E-4 pCi/ 24-hr sensitivity 7.6 μBq/24-hr <sup>l</sup>		
	1997–present	U	RAS (alpha spec)	8.1E-3 pCi		
	2003–present	U	TIMS <sup>k,m</sup>	3E-4 pCi		
Pu-238	1967–1971	U	RAS (alpha PHA)	0.03 pCi <sup>m</sup>	0.2 dpm/24 hr investigate	
	1971–1976	U	RAS (alpha PHA)	0.03 pCi <sup>m</sup>		
	1977–1997	U	RAS (PHA)	0.03 pCi <sup>m</sup>		
	1977–1981	F	RAS (PHA)	0.4 nCi/sample		
	1981–1983	F	Phoswich detector, 4-cm sample thickness	0.2 nCi/sample (17-keV X-rays)		
	1997–present	U	RAS (alpha spec)	8.1E-3 pCi or 300 μBq		

a. U = urine; F = fecal.

b. Unless otherwise noted.

c. The tolerance limit is considered the level under which it was safe.

d. "A successful method of analyzing urine was developed in Jan. 1945 but could not be used as a routine test until a contamination free laboratory (ML Building) was ready for use in Feb. 1945" (Hempelmann 1946).

e. Not adjusted for potential chemical recovery (Clark 2005). Assumes "0.05 dpm/sample" refers to the entire 24-hour sample.

f. Background count rate 1 cpm (changed to 0.1 cpm at some unknown time before 1957), 1,000-minute count time, 50% efficiency, average recovery 82.3% ±19.4% (1945 to 1949) and 67% ±21% (November 1949 to January 1957).

g. Results above these values were considered high (i.e., positive) and subject to statistical investigation. Source: Lawrence (1978).

h. Campbell et al. (1972); McInroy et al. (1991).

i. Procedures on record, not considered the default analysis method. Verify method used from dosimetry monitoring record.

j. Moss et al. (1969).

k. TIMS results are for Pu-239 only; the results do not include Pu-240, which is indistinguishable from Pu-239 by other types of analysis.

l. 2x sensitivity. University of California (1978a).

m. Inkret et al. (1999). TIMS with ultra-trace chemistry and class-100 clean room and alpha spectroscopy methods. Use of alpha spectroscopy allows direct measure of chemical efficiency and detection of Pu-238. TIMS quantifies Pu-239 only, RAS quantifies Pu-239+240.

during processing in the heat source technologies. However, intakes of aged pure  $^{238}\text{Pu}$  could have been possible during decontamination or decommissioning activities in areas where pure  $^{238}\text{Pu}$  was processed. Table 5-8 includes the activity ratios necessary to calculate the appropriate mixture for aged "pure"  $^{238}\text{Pu}$ . Potential ingrowth of  $^{241}\text{Am}$  can be determined from the ratios in Table 5-8.

Table 5-8. "Pure"  $^{238}\text{Pu}$  mixture (Meyer 1970).

Mixture designation:	Weight fraction (fresh)	Fresh	5 yr	10 yr	15 yr	20 yr	30 yr
Years of aging: <sup>a</sup>		0	5	10	15	20	30
<b>Component</b>		<b>Specific activity in mixture (Ci/g)</b>					
Pu-238	0.80	1.370E+07	1.317E+07	1.266E+07	1.217E+07	1.170E+07	1.081E+07
Pu-239	0.169	1.048E+04	1.048E+04	1.048E+04	1.048E+04	1.047E+04	1.047E+04
Pu-240	0.028	6.352E+03	6.349E+03	6.345E+03	6.342E+03	6.339E+03	6.332E+03
Pu-241	0.002	2.061E+05	1.620E+05	1.274E+05	1.001E+05	7.871E+04	4.864E+04
Pu-242	0.001	3.933E+00	3.933E+00	3.933E+00	3.932E+00	3.932E+00	3.932E+00
Am-241	N/A <sup>b</sup>	0	1.463E+03	2.601E+03	3.484E+03	4.167E+03	5.094E+03
<b>Component</b>		<b>Activity ratios<sup>c,d</sup></b>					
Pu-238/Pu-239	N/A	1,307.3	1,256.7	1,208.0	1,161.3	1,117.5	1,032.5
Pu-238/Pu-240	N/A	2,156.8	2,074.3	1,995.3	1,919.0	1,845.7	1,707.2
Pu-238/Pu-241	N/A	66.5	81.3	99.4	121.6	148.6	222.2
Pu-238/Pu-242	N/A	3.483E+06	3.349E+06	3.219E+06	3.095E+06	2.976E+06	2.749E+06
Pu-238/Am-241	N/A	0	9002.1	4867.4	3493.1	2807.8	2122.1

- Time since separation of Am-241 from the plutonium mix.
- N/A = not applicable.
- Calculate dose for Pu-239 and Pu-240 separately using these ratios.
- "Pure" Pu-238 was not on site at LANL until after the introduction of RAS for analysis of bioassay samples. Therefore, the activity ratios for total alpha plutonium are not applicable.

At LANL, the predominantly pure  $^{238}\text{Pu}$  has always been processed in dry boxes and gloveboxes separate from those for processing WGPu to prevent cross-contamination of the two forms.

There is no definitive historical information on the  $^{240}\text{Pu}$ : $^{239}\text{Pu}$  atom ratios of LANL sources, and there is no information on how the ratios vary with time and location. However, before 1970, plutonium at LANL came from the Hanford Site (Gallaher and Efurd 2002). According to environmental impact studies, atom ratios from LANL plutonium that was released before 1970 would have ranged from 0.01 to 0.03  $^{240}\text{Pu}$ : $^{239}\text{Pu}$ . LANL plutonium came from the Savannah River Site starting in 1970. The atom ratio of LANL plutonium that was released after 1970 was expected to range from 0.05 to 0.07, although ratios up to 0.13 indicated the sediment was affected by LANL plutonium (rather than deriving purely from global fallout). Ratios above 0.07 in environmental samples were indicative of a mixture of global fallout and LANL plutonium.

However, Table 5-9 describes the plutonium mixture that was found in 1960 in dirt that was used to sandblast plutonium parts during the demolition of a plutonium filter facility. Thus, although the  $^{240}\text{Pu}$ : $^{239}\text{Pu}$  ratios in pre-1970 plutonium mixtures might have been nominally 0.01 to 0.03, use of a 6% fresh mixture for the pre-1970 years is not unreasonable. However, in general the pre-1970 mixtures of 3% listed in Table 5-4 are suggested when isotopic ratios are not known (Gallaher and Efurd 2002). Mixtures of less than 6% (i.e., 3%) are appropriate to assume during the Manhattan Project era (Lewis 2006a). Intakes of plutonium in the early history of the site should be considered fresh. Aging of plutonium was addressed with continual reprocessing, and although the nature of the process work at LANL involved research-grade plutonium mixtures that are usually considered fresh, there is indication that some americium refining took place at LANL. For this reason, aged plutonium mixtures should be assumed when plutonium intakes resulted from decommissioning, other sources where long-term residual contamination might have been expected (Lewis 2006b), and when the source term is unknown. Assume 3% or 6% fresh plutonium mixture when specific information is available in

Table 5-9. Fresh plutonium mixture  
(Christensen, Garde, and Valentine 1975).

Isotope	Weight percent
Pu-239	93.5%
Pu-240	6.0%
Pu-241	0.5%

the case records (Lewis 2006a,b) to support this assumption. There was practically no americium present in the plutonium at LANL in 1944 or 1945 (Voelz, Grier, and Hempelmann 1985).

Any  $^{241}\text{Am}$  results that are observed in lung counts that were performed years after the intake can be assumed to be the result of (1) the ingrowth of  $^{241}\text{Am}$  from the  $^{241}\text{Pu}$  in the mixture over time or (2) the  $^{241}\text{Am}$  in the initial plutonium mixture unless the incident report specifically indicates potential exposure to pure  $^{241}\text{Am}$  or positive urine bioassay for  $^{241}\text{Am}$  is found for the intake period.

Fuel-grade plutonium (12%) was not known to be routinely encountered at LANL. However, the Clementine reactor (which operated from 1949 to 1952) and the LAMPRE I reactor (1961 to 1963) used plutonium fuel. Plutonium doses for individuals who worked in these facilities should be based on the application of a 12% fuel-grade mixture in accordance with Table 5-6.

To summarize, dose reconstructors should make the following assumptions for plutonium isotopic mixtures.

- 3% fresh plutonium mixture 1944-1956 for most plutonium operations. No aging should be assumed for weapons research;
- 6% fresh plutonium mixture from 1957 onward for weapons-related activities. 6% 10 aged plutonium should be assumed for activities not related to weapons, including maintenance, decontamination, and exposures to plutonium source terms of an unknown nature; and
- Fuel grade (12% Pu-240) for reactor operations personnel with plutonium bioassay in the years 1949-1952 and 1961-1963.

#### 5.2.1.7 Correcting for Urinalysis Volume

Urine samples were corrected for estimated 24-hour excretion of plutonium based on sample volume and/or specific gravity (Gautier 1983). All results in the OTLBD have clearly listed units with associated uncertainties that are reported as 1 standard deviation unless otherwise specified in the record. Some samples that were taken in response to suspected acute intakes might have been spot samples of less than 24-hour excretion. These sample results are either noted in the database or, usually, the results are normalized to 24-hour excretion. In addition, the MDAs might not be in the same units as the activity results. As stated above, activity per 24-hr sample and activity per sample are not equivalent units. TIMS results in the OTLBD are typically listed in picocuries per 24 hours and picocuries per sample, with no listed sample size.

#### 5.2.1.8 Calculating Picocuries per 24 Hours for Urine Bioassay Results

All plutonium bioassay results records have been processed through the current database before reporting. However, should the need arise to calculate plutonium urine bioassay analysis results that are not fully processed (for results outside those supplied with claim files), equations in Lawrence (1978) can be used to convert the listed units to picocuries per 24 hours. Method selection should be based on the era of the samples. These calculations could be necessary to resolve differences if claimant-supplied results differ from results in the database.

### 5.2.1.9 Detection Sensitivities, Reporting Limits, and Tolerance Limits

Detection sensitivity was not a primary concern in the early years of operation of LANL. The concern was whether a tolerance limit was exceeded. Results below the MDA were originally reported as less-than values [listed as "LX.XX," where X.XX is the MDA or detection limit (DL)] or "0" until the 1980s; after which actual positive and negative results were listed in the database.

In the current LABDR, all results from all periods have been listed as actual positive or negative values. When the MDA is not specified in the original result, it is considered to be 2 times the detection level (or uncertainty of the blank). Average chemical recoveries and matrix blanks were not routinely reported before 1957. Therefore, the values in the original dosimetry records for MDAs before 1957 were actually the results that would be reported if an analyzed urine sample had results that were in the upper 12% of the background data, assuming subtraction of the standard background count rate, application of average counting efficiencies, and chemical recoveries. These are the values that would have triggered evaluations of whether the value was positive; therefore, these are being treated as MDAs in this document (Lawrence 1978). However, values in Clark (2005) for average background count rates, recoveries, and count times, and the assumption that the entire 24-hour sample was analyzed, would result in lower MDA values. Values were calculated from the referenced data using the MDA equation (HPS 1996). Both values are listed in Table 5-7.

Bioassay results in the database might have accompanying MDAs if the sample volume is also listed. The MDAs might be listed as activity per sample rather than as activity per 24 hours; this might also be true for the sample results. If the listed MDA values are used, the units must be normalized to activity per 24 hours. In some cases in the historical data the Activity field value might be null or zero. In these cases the MDA value should be used. This MDA value is either a sample MDA, if known, or an *a priori* MDA for the counting system of that era. The Uncertainty field value is valid only when an Activity value is present. Zero values rather than MDA values can exist as real values, although in historical times zero values were often reported when no activity was counted. If the MDA column is null, an MDA value for that era is unknown. All MDA values in the LABDR are true MDAs and not critical levels ( $L_{CS}$ ).

$$MDA = \frac{3 + 4.66S_b}{ETY} \quad (5-1)$$

where:

$S_b$  = standard deviation of the background count

$E$  = efficiency of the counter

$T$  = count time

$Y$  = chemical recovery

$MDA$  = the MDA at 95% confidence

Before the start of RAS analysis in 1967, plutonium results were analyzed as total alpha plutonium. These results are listed as  $^{239}\text{Pu}$  in the LABDR, but should be treated as total alpha plutonium, representative of the combination of  $^{238}\text{Pu}$  and  $^{239+240}\text{Pu}$ . Results from TIMS after 1997 are only  $^{239}\text{Pu}$ ; there is no contribution from  $^{240}\text{Pu}$ . Before use of the results in dose calculations, the  $^{240}\text{Pu}$  contribution can be added to the  $^{239}\text{Pu}$  and entered as  $^{239}\text{Pu}$ , or the dose reconstructor can make separate calculations for  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$ .

Plutonium bioassay sensitivities as listed in procedures and reports are in Table 5-7. In addition, Table 5-7 lists reporting and tolerance limits for certain years. Tolerance limits were defined as the level that could be accepted as safe. Reporting limits are values above the MDA that are considered

significant for recording or follow-up. However, the database contains the actual bioassay results even if the results are below the reporting level.

When MDA values are available in the database, these values are typically listed in units of activity per sample. Except for TIMS analysis, the sample mass is also listed. It is assumed that the per-sample results use the listed sample mass. While this mass is usually a mass that is approximately the 1,400-mL volume, which is the default excretion per day, these are not exactly equivalent. Appropriate normalization to 1,400 mL should be made.

To summarize comparison of sample results to MDAs,

- When results are in activity/24 hours and no MDA is listed, the dose reconstructor should use the applicable default MDA from Table 5-7;
- When MDAs are listed in activity/sample, the results are usually listed as activity/sample as well on a separate line, which makes the determination straightforward;
- When MDAs are listed in activity per sample, in some cases, the sample mass is listed as well, and instructions in the paragraph above should be followed to normalize to daily excretion;
- When MDAs are listed in activity per sample, and the result is not, the result may be neglected in the case of a non-detect, when the MDA is assumed instead; but
- If none of these options are applicable, the site lead for dose reconstruction should be contacted.

#### **5.2.1.10 Validation of Samples**

In the original database, sample results in a worker's dose record might be marked as invalid because the results did not meet the statistical criteria for a valid sample. If results in the LABDR are being compared to original data (or to bioassay results in files in the original database format before June 2005, which might still be in claim folders) and comments on the invalidity of the sample are noted, the dose reconstructor should review the sample information carefully and make a decision on the relevance of sample results. Section A.4 in Attachment A lists the protocol followed by the historical LANL computer software to validate a sample result. This section is provided for information only to enable the dose reconstructor to understand the criteria that were used to cause a sample result to be marked as invalid. This method should not be used by the dose reconstructor in the validation process. The current LABDR might list comments taken from the original database. These comments frequently relate to the Bayesian statistical methods currently in use at LANL and are not necessarily relevant to the dose reconstruction.

The excretion rate after an intake of plutonium typically remains consistent or gradually rises for many years after intake. An example is the original group of 26 individuals who received intakes in 1944 and 1945 and still had measurable urine bioassay results in 1991 (Voelz and Lawrence 1991). Attachment A, Section A.10, contains urine bioassay and nasal swipe results) from an individual with intakes confirmed by autopsy. These results are an example of the possible variability of results for an individual with an actual burden of plutonium.

#### **5.2.1.11 Sampling Protocol for Special Accidental Exposure**

Sampling protocols for special and accidental exposures are listed in Attachment A, Section A.12. However, review of the current LABDR does not completely support the bioassay frequencies that are listed in the internal dosimetry program. An example is a sample marked as involving chelation. The



previous sample, taken 7 months earlier, appeared to be part of the routine program. The next sample after the chelation was 7 months later. There was no sampling in between, no initial sample to confirm the intake, and no information on the date or details of the intake, except a notation of "URINEFIX" on the sample, which is assumed to be the code for chelation.

The incident section in the internal dose file provided to the dose reconstructor might list information on an incident that relates to the results. These records can provide more precise information on known or suspected intake dates. In addition, the record of nasal swipes, positive or negative, might be available in the incident section. Tables A-19 to A-21 in Attachment A list the historical and current protocols for sampling for suspected plutonium exposures.

#### **5.2.1.12 Nasal Swipes**

Until the first urine bioassay analysis was perfected and available in February 1945, nasal swipes (sometimes called nose counts at LANL) were relied on to be a qualitative indicator of plutonium intake (Hempelmann 1946). After the development of urine bioassay techniques, nasal swipes were used to indicate the need for follow-up bioassay, although it was not always performed immediately after a positive nasal swipe (McInroy et al. 1991). The maximum permissible level (MPL) was 50 dpm alpha per nostril. Positive nasal swipes can aid the dose reconstructor in determination of a plausible intake date. Dose reconstructors should not consider the absence of activity above 50 dpm a reason to invalidate bioassay results because many circumstances can contribute to a negative nasal swipe if an intake has occurred. A less-than-MPL nasal swipe, however, might have precluded a follow-up bioassay. In addition, in 1944 LANL began the practice that if the two nasal counts varied significantly the higher nasal count was considered spurious (Hempelmann 1944) and the lower of the two counts was considered the true count (LANL 1994). Information about a positive nasal swipe can often be found in the incident section of the PDF file for the individual.

Intakes are not calculated from nasal swipe results, but the results of nasal swipes are used as indicators of possible intakes. Nasal swipes continue to be used in the present bioassay program as an indicator of possible intakes.

#### **5.2.1.13 Computer-Based Calculations and Sorting**

Beginning in 1959, LANL routinely used several computer-based systems to track plutonium bioassay and intakes. The first IBM-704 plutonium body burden report was made in September 1959. This program estimated the body burden of the employee and the amount of increase in body burden at 6-month intervals (Lawrence 1978).

Of relevance to dose reconstruction, these computer programs allowed the tracking of individuals to ensure that those working with plutonium or frequenting plutonium areas were on the appropriate routine or special monitoring programs.

Details of these historical computer programs are in Section A.4 of Attachment A.

#### **5.2.1.14 LANL Bioassay Data Repository**

All plutonium data reside in the LABDR. Plutonium results, current and historical, have been converted to units of activity (typically picocuries) per 24 hours. All results are reported in actual values (i.e., positive, negative, or zero). Uncertainty values are listed for each measurement. Codes for the interpretation of the records are in the PDF file that is supplied for each individual's results. Records of chelation therapy for plutonium might be noted in the comment field. However, the code in the original database that indicated that the sample was affected by chelation was not captured when the records were transferred from the original repository to the LABDR. Only comments with the

results were transferred. It is assumed that all chelation events would have comments, but that might not be the case. The standard comment notation is "URINEfix" with no information on intake date, etc. The dose reconstructor should carefully evaluate any grouping of high bioassay results. However, only approximately 35 workers have undergone chelation therapy in the history of the site. Therefore, if chelation therapy cannot be validated, chelation should not be assumed for positive bioassay results.

### 5.2.2 Americium

At LANL  $^{241}\text{Am}$  is usually encountered as a trace contaminant in plutonium mixtures. However, there is also potential for exposure to pure  $^{241}\text{Am}$  at LANL during actinide research activities and processing of plutonium mixtures. A procedure for determining  $^{241}\text{Am}$  in urine was in development in 1948 (LASL 1949a). No exposure to pure  $^{241}\text{Am}$  was likely before the beginning of the americium bioassay program in 1954.

All americium data reside in the LABDR. Americium-241 results, current and historical, have been converted to units of activity (typically picocuries) per 24 hours. All results are reported in actual values (i.e., positive, negative, or zero). Uncertainty values might be listed for each measurement. Codes for the interpretation of the records are in the PDF file that is supplied for each individual's results. Records of chelation therapy for americium might be noted in the comment field, usually with the notation "URINEfix." However, as with the plutonium data, the code in the original database that indicated that the sample was affected by chelation was not captured when the records were transferred from the original repository to the LABDR. Only comments with the results were transferred. It is assumed that all chelation events would have comments, but that might not be the case.

There is no historical indication that workers participated in the americium bioassay program only if there was a potential for exposure to pure americium. Therefore, plutonium mixtures should not be inferred from americium bioassay results if no plutonium bioassay results are found for that period. At present, *in vivo* measurements might be performed to confirm the plutonium aging if plutonium bioassay results are above the MDA (Lewis 2006b). However, if the selected plutonium mixture (based on plutonium bioassay results) indicates the presence of americium, the absence of americium bioassay should not preclude the calculation of the dose from the americium contribution to the plutonium mixture.

Before the mid-1990s, very few individuals submitted urine samples that were analyzed for  $^{241}\text{Am}$ . Samples were submitted in response to incidents that involved exposures to sources of  $^{241}\text{Am}$  that are not contained in the plutonium mixture. Estimates of internal doses are based on the results of chest counts and urine bioassay (Inkret et al. 1998b). Urinalysis is the principal bioassay method for assessment of intakes. Publication 68 from the International Commission on Radiological Protection (ICRP) lists the respiratory absorption type for americium as type M (ICRP 1995). However, if  $^{241}\text{Am}$  is determined to be part of a plutonium mixture or ingrowth from the intake of a plutonium mixture, the absorption type for the plutonium mixture should be assumed for the  $^{241}\text{Am}$  (ORAUT 2007b).

The current routine and special sampling programs have protocols similar to those for the plutonium program (Inkret et al. 1998c). Table A-16 in Attachment A describes the routine sampling frequency. Table A-22 describes the current program of special sampling frequencies. The potential for intake can be indicated by a positive nasal swipe. A positive nasal swipe is indicated by 50 dpm alpha in either nostril. However, lack of a positive result on a nasal swipe does not eliminate the possibility of an intake.

Dose from <sup>241</sup>Am intakes associated with plutonium mixtures can be reconstructed when associated plutonium data are available. Dose from pure sources may be estimated by bioassay data when they are present in claim records.

### Minimum Detectable Activities

The historical practice emphasized being below the tolerance level rather than being below the MDA. All bioassay results are reported in picocuries per 24-hour sample; therefore, the listed urine bioassay MDAs in Table 5-10 have been normalized to picocuries per 24-hour sample. MDAs in the LABDR are listed in units of picocuries per aliquot. However, if no volume or mass for the aliquot is given, the sample volume should not be considered the aliquot volume. Results below the MDA were historically reported as less-than values (“LX.XX”, where X.XX is the MDA) or “0” until the 1980s. In the current LABDR all results from all periods have been listed as actual values, positive or negative.

When not specified, 2 times the detection level (or uncertainty of the blank) is considered the MDA. Table 5-10 lists MDAs. MDAs are based on MDAs listed in procedures and reports.

If no MDA value is provided with the sample or the MDA provided with the results cannot be normalized to a 24-hour volume, dose reconstructors should use the values in Table 5-10 to determine if a listed bioassay result is above the MDA or to calculate missed dose using detection levels.

Table 5-10. Americium-241 bioassay techniques and sensitivities.

Sample type	Year	Method	MDA
Urine	1954–1957	Unknown	9.E-01 pCi/24 hr <sup>a</sup>
Urine	1958–1982	Chemical extraction/ proportional counting	2.E-01 pCi/24 hr <sup>b</sup>
Fecal	1977	Phoswich	4.E-02 nCi/sample
Urine	1983 <sup>c</sup> –2002 <sup>d</sup>	Coprecipitation/alpha spec <sup>e</sup>	1.5E-02 pCi/24 hr
Fecal	1983 <sup>c</sup>	Am/Pu screening/phoswich	1.E-02 nCi/sample
Urine	2003–present	RAS	1.0E-02 pCi/24 hr

- No MDA available, use derived investigation level; tolerance level 3.1 pCi/sample (LASL 1954).
- McClelland (1958); method can carry over thorium, plutonium, curium, actinium, and neptunium. Exact start or end date of this MDA is not known.
- Gautier (1983); exact start date of the MDA is not known.
- Sent to an offsite laboratory from 2000 to 2002.
- Inkret et al. (1998d).

### 5.2.3 Tritium

Tritium bioassay started in 1950. The program was formalized in 1952 (Clark 2005). Tritium was encountered in several forms: tritium oxide as water or gas (HTO), elemental tritium or tritiated gas (HT), organically bound tritium (OBT), and metal tritides (MTs). Each form has unique characteristics. Each year approximately 100 individuals are monitored for tritium intakes at LANL. If the work location was in the Health Research Laboratory (HRL) and the Ion Beam Facility (IBF) Buildings, the predominant form of tritium was OBT. Stable MTs were handled in TA-35 (Building 1) and the TSF (TA-21-209, Room 179). Outside these facilities, in cases where exposures to OBT or MT are not implied by case-specific information, assume HTO. Guidance for estimating doses for OBT and SMT is available in ORAUT-OTIB-0066, *Calculation of Dose from Intakes of Special Tritium Compounds* (ORAUT 2007a).

The potential for tritium intakes at the cyclotron and Van de Graaff accelerators might have existed before the beginning of the bioassay program in 1950. Site locations for potential exposure to tritium are in Table A-8 in Attachment A. Bioassay results for tritium are usually available in the LABDR. However, if only whole-body dose is listed in the dosimetry records and the bioassay results are not

available, note that all dose results from 1950 to 1988 were reevaluated by J. N. P. Lawrence in 1989 (Clark 2005) using ICRP Publication 30 parameters (ICRP 1979).

### 5.2.3.1 Organically Bound Tritium

Information on potential exposures to OBT is not complete, but locations such as the HRL and IBF Buildings used labeled compounds as the principal type of radionuclides in these facilities. Guidance on the calculation of doses from OBT is given in ORAUT-OTIB-0066, *Calculation of Doses from Intakes of Special Tritium Compounds* (ORAUT 2007a).

### 5.2.3.2 Metal Tritides

Tritium exposures in the form of MT aerosols were possible. The compounds include the chemical hydrides and dihydrides of hafnium, erbium, titanium, zirconium, and other metals. Uranium beds were used to capture tritium, so uranium tritide was also present (Nasise 1988, p. 4). Information on building locations and years of operation when MTs might have been encountered is not complete, but two facilities, TA-35 (Building 1) and the TSF (TA-21-209, Room 179), were specially designed to handle highly reactive tritides. Glovebox lines were used to contain the tritides, and a recycling system was used to capture escaping tritium (Nasise 1988). Experimentation with MTs required dry, inert environments in order to prevent contamination of the material under experimentation with water from the environment.

The High-Pressure Tritium Laboratory (HPTL), was an early tritium facility (TA-33-86) that used open-faced hoods, rather than gloveboxes (LANL 1988, Horak 1995). The sealed apparatus in the hoods was the primary confinement, and the protection provided by the hoods was secondary. In contrast, the facility for handling MT built in 1953 (TA-35-2) did use a dry glovebox system (Harper and Garde 1982, p. 3). No specific information on the form of the tritium processed in the Weapon Subsystem Laboratory, which used hoods that could be completely enclosed, has yet been located.

ORAUT (2007a) provides guidance on the evaluation of MT intakes. MTs are referred to as *tritium particulates*. In addition to potential exposure in the facilities listed above, claimant telephone interviews can provide indications that a person was exposed to MTs.

Interpretation of codes and units of the bioassay data for tritium is discussed in Attachment A, Section A.2.

### 5.2.3.3 Analytical Techniques

From startup of monitoring in 1950 until 1969, HTO in urine was analyzed by hydrogen evolved by dropping the urine sample on calcium carbide, collecting the hydrogen in a vacuum, and drawing the hydrogen into a glass electrometer chamber. In 1952, a gross GM-type system replaced the glass electrometer. In 1954, the tolerance for tritium in urine was 250  $\mu\text{Ci/L}$  (McClelland and Milligan 1954). The analytical range was 1 to 250  $\mu\text{Ci/L}$ . In 1958, an internal GM counting technique was used. The reported MDA for this method was 1  $\mu\text{Ci/L}$ . In 1970, liquid scintillation counting (LSC) began and is still in use today. With the introduction of LSC, the DL was 1  $\mu\text{Ci/L}$ . The MDA consistently improved.

For tritium results, the denominator used for reporting purposes was per liter of urine. In 1992, all data from 1950 through 1992 were converted to microcuries per liter. Tritium bioassay results can be found as early as 1946.

Results of samples were initially listed as "0" or coded as "LX.XX" (where X.XX is the MDA) to indicate that the value was less than the sensitivity or reporting level of the analysis. In the late 1980s, the practice of recording the result (positive or negative) was begun, although as late as 1990 "0" was still

being entered into bioassay results when the value was below minimum detection levels (Lawrence 1990a). MDA values are listed in Table 5-11 and summarized in Attachment A, Table A-24. All results in the LABDR are likely to be reported in actual values (positive, negative, or zero).

Table 5-11. Tritium urine bioassay sensitivity levels ( $\mu\text{Ci/L}$ ).

Time	Detection level	Reporting level <sup>a</sup>	Counting method
1950–1951	1 <sup>b</sup>		Electroscope
1952–1953	1 <sup>b</sup>		GM counter
1954–1957	1		
1958–1968	1		Internal GM
1969 <sup>c</sup> –1987	0.02 <sup>a,c</sup>	1	LSC
1988–1998	0.01 <sup>a</sup>	0.1	LSC and 1 mL raw urine
1999–present	0.005		LSC

- Use the reporting level as the MDA because results below these values were not reported (Clark 2005).
- Expected to be the same as 1954.
- Gautier (1983).

#### 5.2.3.4 Routine Sampling

The routine sampling protocols for tritium bioassay throughout the history of the program are listed in Attachment A, Table A-17. Historically, persons working with at least 1 Ci of tritium were required to participate in a bioassay program with sampling every 2 weeks. In 1971, the program was changed to determine the frequency of sampling based on the level of tritium in the previous urine sample (Clark 2005).

In 1998 the required turnaround times for tritium samples of all priorities was final completion within 10 days. The preliminary report was due on priority 1 and 2 samples within 3 hours or by Monday morning (Inkret et al. 1998c). Inkret et al. did not specify the definitions of priorities 1 and 2.

#### 5.2.4 Uranium

Bioassays were initiated in 1949 with analyses for uranium mass as an indicator of exposure (Clark 2005). Historical uranium at LANL was primarily either depleted uranium (DU; often referred to at LANL as D-38 or 28) or enriched uranium (EU; referred to as Or-93 or 25) with a variety of isotopic ratios (Little, Miller, and Guilmette 2003a). Uranium exposures from 1950 to 1957 were expected to be primarily from DU because lathing and milling operations took place in large rooms with no exhaust systems. EU work was conducted in dry boxes, so the potential for intakes was lower (Clark 2005). Welding on DU was introduced in 1951. However, ventilation and other measures prevented serious overexposures (Shipman et al. 1952). Engineering controls, especially ventilation, were improved in the late 1960s and reduced the potential of intakes of uranium (Lewis 2006c).

Conversion factors differ among DOE sites because the fractional activity of the  $^{234}\text{U}$  differs in the enrichments. Therefore, 0.17% enriched DU at LANL might have a different amount of  $^{234}\text{U}$  than the same enrichment at a different DOE site, dependent on the source of the DU and the processing. Tables 5-12 and 5-13 describe the nominal weight composition and fractional activity for typical DU and EU mixtures, respectively (Lawrence 1990b). These are the typical enrichments said to be used at LANL. These tables should be used unless there is specific information that states that the enrichments differ significantly from the enrichments listed in Tables 5-12 and 5-13. It should also be noted that the PDF files that were provided with worker bioassay data list a factor of  $3.33 \times 10^{-7}$  Ci/g for  $^{238}\text{U}$ . This is the conversion factor for pure  $^{238}\text{U}$  only; it does not apply to the total alpha uranium results that are reported, especially for the uranium analysis using delayed neutron activation analysis (DNAA; UNAA for uranium analysis) as discussed below. The  $3.33 \times 10^{-7}$  Ci/g factor should not be used for conversion of the results to DU or EU.

Table 5-12. Nominal weight composition and fractional activities for D-38 uranium (Lawrence 1990b).

Measure	U-234	U-235	U-236	U-238	Total
Weight fraction	0.00002	0.003	0.000003	0.997	
Curies in 1 g D-38	1.251E-07	6.489E-09	1.942E-10	3.355E-07	4.673E-07
Isotopic specific activity (pCi/μg)	6.25E+03	2.16E+00	6.47E+01	3.37E-01	4.673E-01
Fraction of total activity	0.2677	0.0139	0.0004	0.7180	

Table 5-13. Nominal weight composition and fractional activities for OR-93 uranium (Lawrence 1990b).

Measure	U-234	U-235	U-236	U-238	Total
Weight fraction	0.011	0.933	0.002	0.054	
Curies in 1 g Or-93	6.879E-05	2.018E-06	1.295E-07	1.817E-08	7.096E-05
Isotopic specific activity (pCi/μg)	6.25E+03	2.16E+00	6.47E+01	3.37E-01	7.096E+01
Fraction of total activity	0.9695	0.0284	0.0018	0.0003	

DU (D-38) is the most common form of uranium that has been encountered at LANL (Inkret et al. 1998a), although natural uranium (NU, also called Tuballoy) was used in conventional weapons testing from 1949 to 1970. The most common chemical forms are oxides and metal. ( $T_3O_8$  can refer to an oxide of NU.) However, LANL has always treated uranium as either solubility class D or W. Urine assay data suggested that “all known LANL exposures to uranium were to a relatively soluble form (not class Y)” (Lawrence 1984, 1990b). The partition between classes D and W could not be determined. Class W was historically used for reporting results (Lawrence 1992a) because it produced larger doses.

In addition, intake was calculated by LANL based on class D mass of uranium, for assaying in relation to the nephrotoxic limit for uranium. The nephrotoxic limit is not relevant to dose reconstruction. However, the dose reconstructor should evaluate the data to determine the absorption type that is most appropriate and favorable to the claimant. Statements about welding DU and eventually EU, which would have been a potential for more insoluble forms of uranium, are in Shipman et al. (1952).

The LANL program calculated intakes for individuals with at least one positive urine analysis result. The potential for intake was indicated by a positive nasal swipe (50 dpm in either nostril). Lack of a positive result on a nasal swipe should not eliminate the possibility of an intake, but follow-up bioassay was not likely to have been performed as a result of a nasal count of less than 50 dpm.

Otherwise, dose reconstructors can use the nominal compositions in Tables 5-12 and 5-13 as default compositions when no other information is available on the enrichment of the uranium (listed as either mass or weight fraction of  $^{235}\text{U}$  or as a weight percent when multiplied by 100). The values in Tables 5-12 and 5-13 are the compositions at LANL at least from 1970 through 1990 (Lawrence 1990b). In addition, in Lawrence (1990b) the presence of  $^{236}\text{U}$  in the isotopic mixture is indicated. The  $^{236}\text{U}$  is indicative of recycled uranium (RU). However, LANL did not process uranium except to extrude or machine it. Oak Ridge National Laboratory (ORNL) was the source of the uranium that was processed at LANL.

#### 5.2.4.1 Sampling Protocol

Historical sampling protocols are listed in Attachment A, Table A-18. The program was initiated in 1949 with analysis for uranium mass as an indicator of exposure. Samples were collected on Fridays just before workers left the site to maximize the sensitivity of bioassay detection (Lawrence 1992a). Since 1983, spot samples have been collected as far removed from the potential exposure time as possible, usually Monday mornings before workers entered the work area. The sample might have contained the last voiding on a Sunday evening and/or the first voiding on a Monday morning if extra

volume was necessary. This protocol ensured that the large fraction (approximately 80%) of rapidly excreted uranium, experienced on the first day after an intake, was excluded from the sample (Lawrence 1984). Beginning in July 1993, the larger volume of sample that was required for the alpha spectroscopy method of analysis made the Sunday evening/Monday morning collections standard protocol (Lawrence 1992b).

In 1992, retroactive calculations were made by Lawrence of the data beginning in 1949. Data for 1951 were not included in these calculations because the bound Los Alamos Notebooks could not be found. Clark (2005) suggests that these notebooks were never found and, therefore, 1951 data are missing from the records. However, review of the current database indicates a number of uranium bioassay records for 1951 similar to the number of records in adjacent years. Therefore, it can be assumed that no data are missing (Buddenbaum 2006). For enriched  $^{235}\text{U}$ , the conversion of data into the computer files and retroactive calculations started with the 1955 data. Assays that were identified as  $^{238}\text{U}$  were converted to micrograms per day with values less than the MDA or zero set to the *minimis* MDA. Assays that were identified as  $^{235}\text{U}$  were converted to picocuries per day with values less than the MDA or zero set to the *minimis* MDA. In the LABDR, the micrograms-per-day and picocuries-per-day results have been converted to micrograms per liter and picocuries per liter, respectively. These results should be normalized to a 24-hour day using an assumed volume of 1,400 mL/d of urine.

#### 5.2.4.2 Uranium Analysis Techniques

Several analysis techniques were employed over the history of LANL. Techniques have included uranium fluorophotometric (UF), ion exchange, or extraction chemistry with radiometric alpha proportional counting (UR), DNAA (or UNAA), and RAS. UF and UR were total uranium techniques with the enrichment selected because of workplace knowledge. The uranium enrichment for DNAA/UNAA results was also determined by workplace knowledge.

Table 5-14 summarizes the routine urinalysis detection levels for various periods. The results of urine bioassay for uranium have been entered in the database. The results in 1991 or before are likely to be reported in actual values (positive, zero, or negative) (Lawrence 1992a). Results below the MDA might be listed as zero or blank.

Table 5-14. Routine uranium urinalysis detection levels.

Period	Method	MDA	DL	Reporting level <sup>a</sup>
1949–1967	UF (DU or NU)	None listed <sup>b</sup>	50 µg/L <sup>c</sup>	>100 µg/L
1968–02/1976	UF (DU or NU)	4 µg/L <sup>b,d</sup> U		
03/1976–1978	UF (DU or NU)	1 µg/L <sup>d</sup> U		
1949–1954	Anion exchange/gross alpha counting (possibly used)	25 dpm/L		>100 dpm/L
1955–1971	Extraction/alpha proportional counting (U-234 alphas measured)	Unknown <sup>b,e</sup>	50 dpm/L <sup>c</sup>	>100 dpm/L
03/1971–1976	Extraction/alpha proportional counting	15 dpm/L <sup>b</sup>		>100 dpm/L
06/1976–01/1977	Extraction/alpha proportional counting	10 dpm/L <sup>b</sup>		
02/1977–12/1977	Extraction/alpha proportional counting	4 pCi/L		
1978–07/1982	UNAA/delayed neutron counting <sup>f</sup> Only U-235 counted	1 µg/L DU 0.17% enriched		
07/1982–06/1991	UNAA/delayed neutron counting <sup>f</sup> Only U-235 counted	4 pCi/L EU 93% enriched		
1982–06/1991	UNAA/delayed neutron counting <sup>f</sup> Only U-235 counted	4 µg/L DU 0.17% enriched		
1982–1992	Anion exchange/colorimetric (used to confirm UNAA >30 µg/L (not a standard method of analysis))	1 µg/50 cm <sup>3</sup>		
07/1991 <sup>g</sup> –1998	RAS <sup>n</sup> for each isotope	0.1 pCi/L	0.05 pCi/L	
1998–present	RAS <sup>n</sup> for each isotope	0.008 pCi/L	0.004 pCi/L (1 standard deviation)	

- Exceeding reporting levels required investigation and evaluation (Lawrence 1984).
- Lawrence (1984).
- Use as detection level (MDA = 2 × DL) because this level would be used to determine positive results.
- 50 µg/L was the level at which LANL considered the result to be a positive indication of NU material in the body; at 100 µg/L, LANL identified the need for a workplace evaluation (Dummer 1958).
- Specific for U-235 and U-233, 50 dpm/24-hour sample considered positive indication of EU in the body (Dummer 1958).
- See Section 5.2.4.3 for full explanation of the interpretation of UNAA results including minimum reporting levels.
- Results for July 1991 through December 1991 might be listed as UNAA results in the LABDR. RAS values were used directly in 1992 (Lawrence 1992b).
- Can measure U-234, U-235, and U-238. Alpha spectrometry cannot differentiate between U-233 and U-234.

### 5.2.4.3 Delayed Neutron Counting

DNAA (also known as UNAA in the LABDR) was the analytical technique LANL used between 1978 and 1992. The technique might have been implemented as early as 1962 (ORAUT 2009a), but UNAA was not in general use until 1978. Interpretation of the results during this time was dependent on identification of the radioisotopic mixture to which the person might have been exposed (Lawrence 1992b). Because the person's work history might put them with D-38 one day and Or-93 the next, uncertainty can be introduced into the results.

The DNAA method depends on counting the number of delayed neutrons produced by fission of <sup>235</sup>U after thermal neutron activation. This method also fissions <sup>239</sup>Pu and <sup>233</sup>U but, according to Gautier (1983), <sup>239</sup>Pu is not an interference unless the concentration in the sample is greater than 300 pCi/L. Uranium-233 was not a significant intake potential at LANL.

The reported results of the DNAA method for EU analyses are labeled <sup>235</sup>U in the records, but have always included the alpha activity of <sup>234</sup>U that is about 34 times that of <sup>235</sup>U. Results for DU analyses are similarly labeled <sup>238</sup>U, but have always been the total uranium mass based on the assumed isotopic mass fractions of 0.00002 <sup>234</sup>U, 0.0017 <sup>235</sup>U, and 0.9983 <sup>238</sup>U (Lawrence 1992a).



The DNAA/UNAA was designated as a screening procedure and not a quantitative measurement. (Although, except for the confirmatory procedure for results above 30 µg/L, there were no other listed quantitative procedures for that period). At the time when DNAA/UNAA was initiated, the uranium exposures had been reduced due to enhanced workplace ventilation units. The ventilation units drew off most of the airborne particulates from machining and grinding operations so the higher exposures in the 1950s and early 1960s were eliminated (Lewis 2006c).

In the DNAA measurement, calibration curves were made up of standards of EU (93%  $^{235}\text{U}$ ) and DU (0.17%  $^{235}\text{U}$ ) (Gautier 1983). These calibration curves were plotted (labeled) as delayed neutron counts versus  $^{235}\text{U}$  pCi/L or  $^{238}\text{U}$  µg/L. For the EU plot, concentration was in terms of total activity of EU. For the DU plot, concentration was in terms of total mass of DU. The calibration curve was selected based on the type of work, and then the concentration of uranium was interpolated.

When a worker was first entered in the system, it was noted on the health physics checklist whether the worker worked with EU, DU, or both. This information was transferred to the sample kit label as "235," "238," or "235/238." Absolutely no indication of the fraction of time or work location would have been recorded.

If the worker had a "235" on the sample kit, the EU calibration curve was used and an activity result that was labeled as  $^{235}\text{U}$  was calculated. If the worker had "238" on the sample kit, the DU calibration curve was used, and the mass result that was labeled  $^{238}\text{U}$  was calculated. If the worker had "235/238" on the sample kit, both curves were used to determine results and both a  $^{235}\text{U}$  by activity result and a  $^{238}\text{U}$  by mass result were calculated. No fractionation of the results is assumed when both isotopes are listed. The dose reconstructor must either use the result or the MDA most favorable to the claimant or make a judgment on the appropriateness of either DU or EU dependent on telephone interviews or DOE records.

The DNAA method actually measures the amount of  $^{235}\text{U}$  in the sample. Only if the ratios of  $^{238}\text{U}/^{235}\text{U}$  and  $^{234}\text{U}/^{235}\text{U}$  are well known can the uranium mass and activity in the sample be accurately determined (Lawrence 1992a). By the DNAA method, the minimum reporting value was 4 µg/L for DU (0.17% enrichment) and 4 pCi/L for EU (93% enriched). For D-38 (DU), a 4-µg/L sample would contain 0.0147 pCi/L of  $^{235}\text{U}$  [(4 µg/L × 0.0017 × 2.163 × 10<sup>-6</sup> Ci/g × 1 × 10<sup>12</sup> pCi/Ci)/(1 × 10<sup>6</sup> µg/g)]. The enrichment of  $^{235}\text{U}$  assumed for DU at this period was 0.17%. A 4-pCi/L sample of EU (93%) would contain approximately 0.116 pCi/L of  $^{235}\text{U}$  [0.116 pCi/L ÷ 0.0284 (from Table 5-13) = 4 pCi/L]. If the person's actual urine sample contained 0.116 pCi/L of  $^{235}\text{U}$ , the reported value for EU would be 4 pCi/L. If the analysis was designated for both EU and DU, the D-38 value would be reported as approximately 32 µg/L [(0.116 pCi/L ÷ 0.0147 pCi/L) × 4 µg/L] and was labeled U-238 (Lawrence 1992b). Uncertainty about the material of exposure, whether enriched or D-38, could introduce as much as a factor-of-4 error in the evaluation of a worker's dose (Lawrence 1992b).

Uncertainty for this method was considered to be 96% ±6% at 5 to 26 pCi/L for EU and 93% ±8% at 4 to 17 µg/L for DU.

This method was eventually deemed operationally unacceptable because of long turnaround times and the need to identify the mixture. LANL changed to RAS techniques between July 1991 and January 1992. Extreme care should be used in interpreting the results of bioassays between these dates. Normalization might have been done to RAS results to maintain consistency with the DNAA results, as discussed in footnote g to Table 5-14.

#### 5.2.4.4 Radiometric Alpha Spectroscopy

This method consisted of a full chemical extraction process for the uranium from the urine. A tracer of  $^{232}\text{U}$  was added to determine chemical recovery. The extracted and plated uranium was counted by RAS and the activities of  $^{232}\text{U}$ ,  $^{234}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  were determined (Lawrence 1992a).

Several problems were initially encountered with this method. No chemical or synthetic urine blanks were used to establish the MDA. The counting blank was a blank planchet rather than a reagent blank, which would produce a lower background. Chemical recoveries ranged from 10% to 60%. Some analyses indicated isotopic ratios outside the realm of probability. The computer program converted all negative values to zero. All 1991 data yielded negative values for  $^{235}\text{U}$  rather than the expected negative and positive values. In early 1992, the computer program was still not modified to correct for chemical blanks.

Results that are listed as RAS in the current database, which are isotopic, should be evaluated according to the type of enrichment that is determined for the intake. Table 5-15 provides the factors to be applied to the isotopic bioassay results to estimate the total uranium as either DU, NU, or EU.

Table 5-15. Factors to convert isotopic activities to total uranium.

Radionuclide	Activity factor <sup>a</sup>		
	DU	NU	EU
U-235	N/A	N/A	35.211
U-234	3.736	2.058	1.031
U-238	1.393	2.033	N/A

a. N/A = not applicable and indicates that the fraction of the isotope is too small to be used as an indicator for the given enrichment.

#### 5.2.4.5 Excretion of Environmental Levels of Uranium

NU from nonoccupational intakes (primarily food and water) can be excreted in urine at levels above the analytical MDAs for either the elemental uranium analysis or the alpha spectrometry analysis. The  $^{234}\text{U}$ : $^{238}\text{U}$  ratio can be used to distinguish DU from NU. For the purposes of dose reconstruction, environmental uranium is not subtracted from monitoring results, and all uranium is assumed to result from occupational exposure. Therefore, this section is for information only. Table 5-16 lists activity ratios for NU.

A 1992 study by Little, Miller, and Guilmette (2003a) listed the average drinking water concentrations for the Los Alamos/White Rock/Santa Fe area as 0.015  $\mu\text{g/L}$  (0.01 pCi/L or 0.00037 Bq/L). Therefore, with the use of 0.01 as the factor of drinking water concentration to excretion concentration (Little, Miller, and Guilmette 2003a,b), LANL used the urinary excretion values of 0.00015  $\mu\text{g/L}$  for elemental

Table 5-16. Characteristics of NU (Tuballoy).

Measure	U-234	U-235	U-236	U-238	Total
Weight fraction	0.0000537	0.0072	0	0.99274	
Specific activity (pCi/ $\mu\text{g}$ )	0.33367	0.01557		0.33367	0.68291 <sup>a</sup>
Fraction of total activity	0.4886	0.0228	0	0.4886	

a. As listed in IMBA Version 1.0.42.

analyses, 0.0001 pCi/L for  $^{234}\text{U}$  and  $^{238}\text{U}$ , and essentially anything that was detected for  $^{235}\text{U}$  to distinguish between natural background and potential occupational exposure for uranium. In some cases, specific information in the worker's file indicates that the excretion was from natural sources.

New Mexico is known for high levels of NU in the soil and groundwater. However, some LANL workers lived in areas of particularly high NU concentrations that ranged from 0 to 4 Bq/L (108 pCi/L) in 1992 and up to 6 Bq/L (162 pCi/L) in 2001 (Little, Miller, and Guilmette 2003b). These areas of high concentration are primarily in the Española area.

Beginning in 1992, all workers who participated in the uranium bioassay program were requested to bring samples of their drinking water to LANL for analysis with each urine bioassay sample (at least once each year). The concentrations were found to vary widely even for individuals. The results of the analyses of these water samples are listed with the bioassay data for the individual. When water sample results are noted in the record, LANL multiplied the results by 0.01 to calculate the concentrations of uranium isotopes to be subtracted from the individual's bioassay results before calculation of an occupational intake. The factor is based on the consumption of 1.1 L of water per day (Little, Miller, and Guilmette 2003b). When LANL assesses dose or potential occupational intakes, the concentration of the most closely associated water sample might be subtracted from the bioassay sample results rather than an average value for all the drinking water samples for the individual. (There is no evidence that the individual bioassay results for the individuals in the database have had the drinking water concentrations subtracted. Therefore, these results should be used as found.) Other variables, such as drinking habits, dietary components, and individual physiological differences, can influence the individual's baseline excretion rate of environmental uranium.

Background excretion of uranium in feces probably varied over an even larger range than urinary excretion. Fecal samples were rarely obtained for potential uranium intakes; when they were, the investigation report should discuss how the results were interpreted.

### 5.2.5 Polonium

Polonium for use in initiators was handled in quantities of 100 Ci/mo or more starting in February 1945 and increasing to 500 Ci/mo by December 1945. Before that time, smaller quantities were handled at in TA-1 at H and Gamma Buildings (ENSR 2002). DP East, Buildings 151, 152, 153, and later (1949) 155 were also used to process polonium for initiators.

Work at LANL with  $^{210}\text{Po}$  ceased in 1959. After that time, polonium was encountered only in the form of sealed Po-Be sources. After 1959, there were two incidents in which sealed Po-Be sources broke open; personnel involved in the incidents submitted urine samples for bioassay. Unless a worker was involved in the broken source incidents, no intakes should be calculated after 1959.

Work with  $^{210}\text{Po}$  was of a limited scope at LANL. Therefore, missed dose for polonium should be assessed only if actual polonium bioassay was performed for the individual or if there is indication, through claimant interview, that the employee was actually exposed to polonium. Potential missed dose should be assessed only during the interval covered by the bioassay or work period. In 1945, only two persons exceeded the 1,500-cpm/24-hr sample tolerance for polonium (ENSR 2002). However, University of California (1977a) stated, "During the 1940s and early 1950s,  $^{210}\text{Po}$  exposures at LASL occurred with some regularity. There was no method in routine usage to determine the actual exposure. (Exposure was controlled from raw urine assay data.)" Dummer (1958) stated that the possibility of exposure should be investigated at greater than 10 dpm/L in a urine bioassay sample.

Hempelmann (1946) stated that, because of the ease of the analysis, polonium urine bioassay was performed routinely and frequently on all individuals who worked with polonium. These records are expected to be in the current database.

The initial urine bioassay analysis procedure could not adequately distinguish between plutonium and polonium. During 1944, the total alpha results were assigned to either plutonium or polonium based on the individual's work history. The procedure was modified in the fall of 1944 to extract the plutonium. Hempelmann (1944) indicates that this modified procedure was originally developed at Monsanto. LASL (1945) lists the procedure and states that the recovery of polonium on the copper-plated disk was expected to be greater than 80%. Gautier (1983) contains a procedure for  $^{210}\text{Po}$  in urine with an effective date of May 1, 1955, that states that the average recovery of standards was  $90\% \pm 3\%$  at the 15-pCi concentration. Discussions with a retiree indicate that no correction for recovery was included in the calculations [1]. Despite the assumption of 100% recovery by LASL personnel, based on the recovery factor that has been determined to apply to samples analyzed by the Monsanto method (ORAUT 2013a), the 10% recovery correction is applicable to LANL results as well.

Detection limits for routine urinalysis are listed in Table 5-17. A procedure is listed for  $^{210}\text{Po}$  in urine in LASL (1954) and Gautier (1983). The MDA continues to later periods unless changed.

Table 5-17. Routine  $^{210}\text{Po}$  urinalysis detection levels.

Period	MDA	Recheck	Tolerance limit <sup>b</sup>
1943–1952			440 dpm/24 hr <sup>a</sup>
1953			50 dpm/24 hr <sup>a</sup>
1954	10 dpm/L	100 dpm/L	500 dpm/L
1955	0.1 pCi/L		
1958		100 dpm/L	500 dpm/L

a. LASL (1979).

b. Workers exceeding the tolerance limit were removed from exposure potential until the levels dropped below the tolerance limit.

## 5.2.6 Other Limited-Exposure Radionuclides

LANL has always been a center for research. As such, small-scale use of various radionuclides (in terms of either the number of persons or activity of the source) that are not addressed above has occurred throughout the history of LANL. Although some documentation has been found on bioassay for these nuclides, the records are not provided to the dose reconstructor as part of the case-specific information.

## 5.3 ***IN VIVO* BIOASSAY 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 1970, and possibly as early as 1960. There is some indication that some of the counts between the beginning of the program in 1955 (Van Dilla 1959) and the 1960s were performed for development of the program rather than actual suspected intakes. Counts during this period should be evaluated as closely as possible for validity in the dose reconstruction. No *in vivo* results are expected to be found in the LABDR before 1969.

### 5.3.1 Whole-Body Counters

The first whole-body counter to be used at LANL was the HUMCO I (Ennis 2003), which became operational in 1955 (the name stood for human counter). The counter consisted of a large double cylinder with a liquid scintillation fluid (possibly trichloroethylene) that filled the annular space between the cylinders. The scintillation fluid was viewed with an array of 5-in. photomultiplier tubes on the outside wall of the cylinder. The individual was placed inside the count chamber. The count rate was compared to the background count rate. The system typically used two energy windows: one for  $^{40}\text{K}$

(1 to 2 MeV) and one for  $^{137}\text{Cs} + ^{40}\text{K}$  Compton counts between 0.5 and 0.8 MeV. The result was obtained by subtracting the contribution of  $^{40}\text{K}$ . The system was not used for photons below 100 keV. This system was used to screen individuals who might have been exposed to fission products at the reactors or in flyovers during weapons testing. It was also used to detect the bremsstrahlung from  $^{90}\text{Sr}$  intakes. The energy resolution of these counters was poor. When an elevation of the background in a region of interest was observed, the individual was referred for screening with either the shadow-shield or full-shield 4- by 8-in. NaI(Tl) crystals (Healy 1970). The sensitivities of the NaI(Tl) crystals were approximately the same as those for the HUMCO, but the count time was significantly longer.

The HUMCO II became operational in 1958. The HUMCO II was housed in a count room (SB-16) made of 7-in. pre-World War II steel. The resolution was improved, but it remained a screening counter. Evaluations were made using the shadow shield or NaI(Tl) crystals. The 1958 annual progress report for the H-1 Group discusses "significant" measurable  $^{65}\text{Zn}$  in cyclotron workers (Buckland 1959).

In 1970, an *in vivo* counter capable of measuring four separate regions of the body began operation (Vasilik and Aikin 1983). Twin phoswich (CsI and NaI) detectors were placed over the lungs. The two layers of the detector were capable of simultaneously, yet separately, monitoring chest burdens for 10- to 250-keV photons (NaI), for plutonium and uranium isotopes and  $^{241}\text{Am}$ , and for 200- to 2,000-keV photons (CsI) for a qualitative assessment of a variety of fission and activation nuclides. A planar hyperpure germanium (HPGe) detector monitored the region between 10 and 250 keV with excellent energy resolution and could be positioned over the liver or thyroid as needed. Last, a lithium-drifted germanium (GeLi) detector (which was later replaced by an HPGe detector) was positioned under the prone subject. This detector was primarily for whole-body assessment. This system could both identify radionuclides and quantify the burdens. In 1990, the Tiger Team Assessment report indicated that the phoswich detectors were not state-of-the-art and could not accurately measure plutonium and americium in the presence of interfering gamma emitters, such as  $^{137}\text{Cs}$  (DOE 1991). The phoswich and germanium systems were operated concurrently during the period from 1998 to 1999. The twin phoswich detectors were replaced by twin three-detector arrays of HPGe detectors in 1999. Two of the six HPGe detectors were used when a thyroid count was required.

### 5.3.1.1 Minimum Detectable Activities and Decision Levels

Operationally, an observed signal must exceed the  $L_C$ , formerly referred to as the minimum significant measured activity (MSMA), to result in the decision that some activity has been detected in the lung, body, or other organ; this is also called the decision level (DL) (Vasilik et al. 1984). The MDA, formerly the minimum detectable true activity (MDTA), is the smallest amount of activity that can be in the lungs or organ such that a measurement of an individual can be expected to imply, correctly, the presence of activity with a predetermined degree of confidence. The MDA (or MDTA) and  $L_C$  (or MSMA) values are listed in Table 5-18 for various years of operation. The MDA and  $L_C$  values for

Table 5-18. Routine whole-body counting detection levels (Vasilik et al. 1984; Ennis 2003).<sup>a</sup>

Period	Nuclide	$L_C$ (nCi)	MDA (nCi) <sup>b,c</sup>
1955–1958 <sup>d</sup>	Cs-137		8
	Sr-90 <sup>e</sup>		30
1959–1970 <sup>d</sup>	Cs-137		4
	Sr-90 <sup>e</sup>		30
1971–1984	Be-7	0.9	1.8
	Cs-134	0.9	1.8
	Cs-137	0.9	2.1

Period	Nuclide	L <sub>c</sub> (nCi)	MDA (nCi) <sup>b,c</sup>	
	Co-57	2.1	4.8	
	Co-60	0.78	1.8	
1985–1998	Tl-202	0.5	0.9	
	C-11/N-13 <sup>f</sup>	0.3	0.5	
	Eu-152	2.2	3.3	
	Co-58	0.5	0.9	
	Co-56	0.5	0.9	
	Hg-197	3.1	4.6	
	Hg-195	2.5	3.7	
	Hg-195m	1.8	3.2	
	Hg-197m	3.8	6	
	Hg-203	0.8	1.2	
	Hg-193m	0.7	1.5	
	Cs-134	0.5	1.1	
	Os-185	0.6	1.1	
	V-45	0.5	0.8	
	Be-7	3.4	8.7	
	Sc-46	0.5	0.9	
	Mn-54	0.5	0.9	
	Cs-137	0.6	1.1	
	Co-60	0.5	0.8	
	Br-77	1.7	3.4	
	Sb-124	0.4	0.8	
	Ce-141 <sup>g</sup>	2.2	4.4	
	Ce-144 <sup>g</sup>	12.1	24.2	
	Cr-51	6.4	12.8	
	Co-57	1.4	2.8	
	Cu-67	1.5	3	
	Fe-59	1.2	2.4	
	Se-75	1.1	2.2	
	Se-73	0.4	0.8	
	Na-22	0.4	0.8	
	Zn-65	0.8	1.6	
	1999–present <sup>g</sup>	511 keV	1.15	2.3
		Be-7	4.2	8.4
Ce-141 <sup>g</sup>		1.35	2.7	
Ce-144 <sup>g</sup>		6.5	13	
Co-56		0.55	1.1	
Co-58		0.5	1	
Co-60		0.45	0.9	
Cr-51		4.45	8.9	
Cs-134		0.5	1	
Cs-137		0.6	1.2	
Cu-67		1.5	3	
Eu-152		1.3	2.6	
Hg-203		0.6	1.2	
Mn-54		0.4	0.8	
Na-22		0.45	0.9	
Os-185		0.45	0.9	
Ra-226		20.5	41	
Sb-124		0.45	0.9	
Sc-46		0.7	1.4	
Se-75		0.85	1.7	
Tl-202		0.5	1	
U-235	1.35	2.7		

Period	Nuclide	L <sub>C</sub> (nCi)	MDA (nCi) <sup>b,c</sup>
	V-48	0.4	0.8
	Zn-65	1.1	2.2
	Zr-95	0.8	1.6

- Listing of an MDA for a radionuclide does not necessarily mean that that radionuclide was frequently encountered. The MDAs listed in the individual's results for a given count should be used if available.
- Based on 95% confidence of detection.
- MDA = L<sub>C</sub> × 2 unless otherwise specified.
- The HUMCO I and II systems were designed for screening subjects. Subjects found to have contamination levels above background were referred to the 8- by 4-in. NaI detector, which had the same sensitivities with an extended count time.
- By bremsstrahlung.
- Based on 511 keV. C-11 is a positron emitter with no photons. However, the 511-keV peak should always be present due to positron annihilation. The 511-keV peak can have interference contributions from other sources including pair production interactions from nuclides with photon energies greater than 1,022 keV.
- Lower sensitivities might be available using the lung counter for certain nuclides if lung counting is appropriate to the dose reconstruction.

lung counting are summarized in Table 5-20. The values of MDA and L<sub>C</sub> are calculated in accordance with the theoretical developments of Currie and of Altshuler and Pasternack (Vasilik et al. 1984). No information is available on MDAs for the thyroid detector or for <sup>131</sup>I or <sup>125</sup>I in the whole-body counter. If available, the MDAs that are listed in the records of the worker should be used rather than the MDAs listed in the tables.

Whole-body and lung counting are the primary methods for determining intakes of fission and activation products. However, both *in vitro* and *in vivo* results might be available.

Results in the bioassay records are generally reported in nanocuries unless otherwise indicated. Results that are listed as NULL indicate no detectable activity (NDA) rather than the MDA. Results less than the L<sub>C</sub> are marked as NDA in the database. The actual DL values and counting errors might be available on the report for the individual, but they are not available in the IVML database until 2003. However, MDAs are listed with *in vivo* count results in the LABDR for <sup>241</sup>Am and <sup>239</sup>Pu. Since 2004 MDAs are also listed for <sup>235</sup>U and <sup>234</sup>Th. In addition, every count performed since 1969 when the program was formalized has been recorded in the database and has a "White Card" on file. Counts without positive results for any analyte are marked NDA. However, the results of counts from before 1969 were only performed in conjunction with significant radiological events. Records of these measurements are in logbooks, but they might only indicate that measurements were made, without associated results. Early *in vivo* measurements were made in "go/no-go" fashion using instruments with little or no energy discrimination capability. Negative results might not have been recorded but only verbally communicated to the subject. Logbooks with early *in vivo* measurements results are in LANL archives on the site (Hoover 2007).

An *in vivo* count spectrum is not analyzed for a fission or activation product radionuclide unless a peak that is associated with that nuclide is visible in the spectrum. When that peak is visible, the suspected nuclide is added to the library and the spectrum is reanalyzed. Visual or non-library-driven software recognition of a peak can be subjective and not directly correlate to MDA or L<sub>C</sub> calculations, especially with the broad peaks that are associated with scintillation detectors. For whole-body counts, it is not reasonable to assume that a worker was exposed to or is being monitored for all radionuclides potentially reportable simply because an MDA was determined and listed on the report.

In general, no information is available in the reports on the assignment of respiratory absorption type for specific fission and activation product nuclides. Individual guidance might be available in the narrative on the White Card that accompanies a worker's *in vivo* bioassay report. It is unlikely that the

information on the White Card has been transferred to the remarks section of the LABDR. However, if the dose reconstructor deems it appropriate, copies of the White or Beige Cards can be requested.

RaLa operations occurred at LANL from September 1, 1944, through July 18, 1963. The internal dose from intakes or potential intakes of  $^{140}\text{La}$  and  $^{140}\text{Ba}$  from RaLa operations at TA-10 (Bayo Canyon Site), TA-35 (Ten Site), and Buildings H, Sigma, and U in TA-1 for chemists and other maintenance and operations workers and security personnel cannot be assessed due to the lack of a well-established *in vivo* bioassay program during that period. Some whole-body counts might have been performed beginning in 1955. *In vivo* results might be available in the LABDR.

### 5.3.1.2 Cesium-137 and Other Intakes from Fallout

Many workers in the early days of whole-body counting had detectable activities of  $^{137}\text{Cs}$ . Most of this was attributed to fallout. Some workers had even higher levels of  $^{137}\text{Cs}$  from consumption of wild game. A DL used to establish the difference between occupational and nonoccupational sources of  $^{137}\text{Cs}$  and other fallout radionuclide intakes has not been discovered in the records. In lieu of other information, the following guidance can be used:

- The  $^{137}\text{Cs}$  intake should be considered occupational if the same whole-body count detected other fission or activation products. It should also be considered occupational if a fission or activation product or radiostrontium urinalysis showed detectable activity and the sample was obtained in a reasonable time before or after the whole-body count or within the period between the previous and next whole-body counts. The reasonable time is based on the biological retention pattern of the radionuclide in the body.
- All other fission or activation products that are identified in the whole-body or lung count should be considered occupational unless specifically stated on the White Card and the reasons for invalidating the results are acceptable to dose reconstruction practices.
- If an investigation occurred and the record clearly shows that the intake was due to a nonoccupational source, the  $^{137}\text{Cs}$  can be disregarded. The results of the investigation would be noted on the White Card.
- National Council on Radiation Protection and Measurements (NCRP) Report 94 provides mean body burdens of  $^{137}\text{Cs}$  for the United States for the years most likely to produce interference with occupational whole-body count results (NCRP 1988). Those values are listed in Table 5-19. If no other fission or activation products are linked to the intake and the  $^{137}\text{Cs}$  result is less than the values in Table 5-19, the  $^{137}\text{Cs}$  result can be assumed to be due to fallout.



Table 5-19. Mean body burdens of  $^{137}\text{Cs}$  from fallout in the United States (nCi).<sup>a</sup>

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	1975	1.1
1963	11	1976	1.6
1964	19	1977	1.1
1965	16		

a. NCRP (1988).

### 5.3.2 Lung Burdens

Lung burdens of  $^{239}\text{Pu}$ ,  $^{238}\text{Pu}$ , and  $^{241}\text{Am}$  were monitored using the phoswich lung detectors beginning in 1970. The 59.5-keV gamma line of  $^{241}\text{Am}$  is used to determine the  $^{241}\text{Am}$  burden (50- to 70-keV region). If the isotopic ratio for a given intake is known, the  $^{239}\text{Pu}$  and  $^{238}\text{Pu}$  can be determined from the  $^{241}\text{Am}$ . Otherwise, the plutonium is determined from the uranium low-energy X-ray (L X-ray) region. When  $^{241}\text{Am}$ ,  $^{239}\text{Pu}$ , and  $^{238}\text{Pu}$  are present, corrections for the contribution of the neptunium L X-rays, from the decay of  $^{241}\text{Am}$ , to the 14- to 25-keV  $^{239}\text{Pu}$  and  $^{238}\text{Pu}$  regions must be considered. The phoswich detectors were eventually replaced by arrays of HPGe detectors, which greatly improved the energy resolution. Improved energy resolution permitted the system to distinguish between gamma and X-ray lines that are closer together. However, because the uranium L X-ray energies for the decay of  $^{239}\text{Pu}$  and  $^{238}\text{Pu}$  are the same, there is no way to differentiate between these two isotopes in an actual measurement. Isotopic information about the exposure is used to determine the appropriate calibration factor.

Efficiency, and therefore sensitivity level, varies for every count due to the effects of chest wall thickness on the attenuation of the 17-keV X-ray and the 59.5-keV gamma ray. Therefore, the MDA that is listed with the count should be used when available. The MDA and  $L_C$  values in Table 5-20 are nominal and based on the calibration chest wall thickness of 2.5 cm. These can be used for correlation with projected bioassay results. Chest wall thickness for the individual can typically be found on the White Card that is associated with the *in vivo* counting record. Chest wall thickness is estimated by weight:height ratios for routine counting and by ultrasound for special or positive counts. For lung (chest) counts, increases in chest wall thickness can increase the MDA for the individual count. University of California (1977b) suggests that MDAs should be increased 50% for large individuals. The dose reconstructor should use best judgment in determining the applicability of the listed MDAs for bounding missed dose projections. The MDA and  $L_C$  values for lung counting are summarized in Table 5-20.

MDAs are typically reported for  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  for lung counts. However,  $^{241}\text{Am}$  is not always detectable even if a plutonium lung burden exists. Much of the plutonium at LANL, especially in the early years, was fresh [2]. Plutonium burdens from the 26 individuals from the original Manhattan Project have been calculated by LANL to be 6 to 80 nCi since before 1946. These individuals have been followed for more than 50 years. Twenty-one of them received their first lung counts in 1971. At that time the  $^{239}\text{Pu}$  MDA was 7 nCi for a 2,000-second count. Positive counts were obtained for 14 of the 21 measured persons. The lung burdens ranged from 3 to 10  $\mu\text{Ci}$   $^{239}\text{Pu}$  (Hempelmann, Richmond, and Voelz 1973). None of these individuals has had  $^{241}\text{Am}$  activity above background

Table 5-20.  $L_C$  and MDA values for lung counting (nCi).

Period	Radionuclide	$L_C$	MDA <sup>a</sup>
1970–1973 (2,000-s count time) (Hempelmann, Richmond, and Voelz 1973)	Pu-239	3.5	7
Extended count time 1977 <sup>b</sup>	Am-241		0.3
	Pu-238		10
	Pu-239		21
1980–1984 <sup>c</sup> (Ennis 2003) <sup>d</sup> (1970–1979) <sup>e</sup>	Am-241	0.155	0.31
	Pu-238	11	22
	Pu-239	24	48
1984 (Vasilik et al. 1984) <sup>f</sup>	Am-241	0.16	0.32
	Pu-238	14	28
	Pu-239	30	60
1998 <sup>g</sup> –present <sup>h</sup> (Ennis 2003)	Am-241	0.1	0.2
	Am-243	0.1	0.2
	Pu-238	10	20
	Pu-239	31	62
	Th-234	0.85	1.7
	U-235	0.1	0.2
	Np-237	0.2	0.4
	Np-239	0.1	0.2
1998–present Fission/activation products (Ennis 2003)	511 keV	0.1	0.2
	Be-7	0.35	0.7
	Ce-141	0.1	0.2
	Ce-144	0.25	0.5
	Co-56	0.1	0.2
	Co-58	0.05	0.1
	Co-60	0.1	0.2
	Cr-51	0.35	0.7
	Cs-134	0.05	0.1
	Cs-137	0.1	0.2
	Cu-67	0.1	0.2
	Eu-152	0.1	0.2
	Hg-203	0.05	0.1
	Mn-54	0.1	0.2
	Na-22	0.1	0.2
	Nd-147	0.1	0.2
	Os-185	0.05	0.1
	Ra-226	0.9	1.8
	Sb-124	0.05	0.1
	Sc-46	0.1	0.2
Se-75	0.05	0.1	
Tl-202	0.05	0.1	

a. Assume chest wall thickness of 2.3 cm.

b. As listed in 1977 quarterly progress report based on a 60-minute count time and a person of average build for a UPPU Club member (University of California 1977b).

c. Might be applicable to the startup of the phoswich system in 1970; no other information available. For a 2,000-s count time.

d. Assume chest wall thickness of 2.5 cm.

e. There is a reasonable correlation between the expected MDA for a 15- to 20-minute count time, typical of standard *in vivo* counting beginning in 1970, and the 60-minute count MDAs listed in 1977.

f.  $MDA = L_C \times 2$ ; recounts were performed if the results were greater than the  $L_C$ .

g. Lung counter has 10- to 300-keV and 80- to 3,000-keV ranges, so a lower sensitivity for certain fission and activation products can be obtained.

h. The phoswich detectors were replaced by an array of six planar HPGe detectors around 1998 (Ennis 2003).

using *in vivo* techniques (Voelz et al. 1979; Voelz, Grier, and Hempelmann 1985) except one individual with a positive lung count for  $^{241}\text{Am}$  37 years later. This individual was suspected of an additional intake of plutonium that contained  $^{241}\text{Am}$  in [year redacted].

Based on the above discussion, ingrowth of  $^{241}\text{Am}$  from early intakes (1944 to 1945) should be expected to be negligible. Intakes in later years might result in  $^{241}\text{Am}$  ingrowth or might have contained  $^{241}\text{Am}$  in the original intake.

Follow-up lung counts for on members of the UPPU Club were typically longer (60 minutes rather than 15 or 20) than those for routine or investigational lung counting. Therefore, the MDAs for these counts should be expected to be between 50% and 70% of the standard MDAs listed. (The rule of thumb is to quadruple the count time and halve the MDA.)

Results in bioassay records should be assumed to be in nanocuries unless otherwise stated. Results listed as NULL indicate NDA (not MDA). Results less than the  $L_C$  are marked as NDA in the database. The actual DL values and counting errors might be available on the report of the individual but are not available in the database until 2003.

All individuals who receive lung counts are monitored for  $^{239}\text{Pu}$  and  $^{241}\text{Am}$ . In 2004,  $^{235}\text{U}$  and  $^{234}\text{Th}$  (as  $^{238}\text{U}$ ) were added to the routine *in vivo* analysis library.

### **5.3.3 Wound Monitoring**

In August 1959, the H-6 Group acquired a probe to be used to monitor wounds that were contaminated with plutonium. This probe was capable of detecting soft plutonium X-rays. The sensitivity of this probe was 1 nCi of plutonium when it was unshielded. This was equivalent to detection of one-tenth of a permissible body burden of embedded plutonium in tissue to a depth of 1 cm (LASL 1959). In 1977, a new NaI detector (12 by 2 mm) was being evaluated. This produced an MDA of 0.07 nCi based on WGPu (University of California 1977c).

Wound counting was used primarily as a tool for surgeons to locate plutonium in the wound, not as results for calculation of internal dose. Wound monitoring continues to be performed. In most cases, intake and dose are not assessed directly from the wound count but rather from urine bioassay data in accordance with the guidance in ORAUT-OTIB-0022, *Guidance on Wound Modeling for Internal Dose Reconstruction* (ORAUT 2005a). No other information on instrumentation or sensitivities is available.

## **5.4 INTERFERENCES AND UNCERTAINTIES**

### **5.4.1 Discrepancies in Results**

In the early years of the bioassay program, analysis techniques and protocols did not use chemical blanks and recoveries in the manner that became standard protocol in later years. When the databases were being constructed, beginning in the late 1960s, an attempt was made to incorporate parameters in relation to current analysis procedures (Glover 2006). Therefore, the results initially reported to the worker might not be the same as the results that finally appear in the database that was submitted to NIOSH. A project to validate these results is described in ORAUT-OTIB-0063, *Los Alamos National Laboratory Bioassay Repository Database* (ORAUT 2009a). If there are discrepancies between results in the current database and the results supplied by the worker, the dose reconstructor should refer to the results of the validation project to resolve any discrepancies.

## 5.4.2 Contamination of Samples

Until the Health Pass Ward was established in 1945, the potential existed for sample contamination. It is likely that a contaminated sample will appear as an obvious outlier in the dataset for a worker. The use of a result from a contaminated sample could result in an overestimate of the worker's dose, but the sample result should be considered real if other data do not demonstrate it to be a false positive result. Some variability is expected in any set of results. See Section A.10, Attachment A, for an example of a valid series of bioassay results for an individual whose autopsy results verified plutonium intakes. The Health Pass Ward was eliminated in 1952 because of expense. However, protocols to allow the samples to be collected in a contamination-free environment were established.

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 shows a dramatic decrease in activity or NDA on the same day or within a few days, external contamination can be suspected. Radon progeny and medical diagnostic or therapeutic procedures that involve radionuclides can cause interference 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.

## 5.4.3 Uncertainties

### 5.4.3.1 *In Vivo* Counting

Uncertainties for bioassay measurements might be included with the results for excreta or *in vivo* measurements. For *in vivo* results, uncertainties are not reported in the database until 2003, but uncertainties might be listed on individual reports before 2003. The listed uncertainties are typically reported as 1 standard deviation.

Uncertainties that are associated with chest counting are reduced by the use of different calibrations for different chest wall thicknesses and the use of ultrasound to measure chest wall thickness. A 1-sigma uncertainty of approximately 20% for americium and uranium values in chest counting, not including correction for interferences from bone and liver, is assumed. 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. The uncertainty in the estimate of chest thickness using the height:weight correction was at least 50% (University of California 1977b). The mathematical correction was made for routine counts. Special counts and counts with positive results were typically corrected using ultrasound chest wall thickness measurements.

### 5.4.3.2 *In Vitro* Measurements

Uncertainties for normal distributions are often listed in the LABDR for samples. The measurement uncertainty is typically listed at 1 standard deviation with the results (Lewis 2006a). When results are listed as negative, positive, or zero, use of a normal distribution is recommended because a lognormal distribution would not be applicable to negative results. However, a study of the variations in plutonium urinary data (Moss et al. 1969) determined that the LANL employee results exhibited as a lognormal distribution with a geometric standard deviation (GSD) of 1.9 (from the ratio of the 84th percentile to the 50th percentile).

## 5.5 UNMONITORED INTAKES

Exposure circumstances discussed throughout this TBD indicate that for Los Alamos National Laboratory employees, there is a potential for unmonitored intakes of secondary radionuclides. Partially as a result of this potential, NIOSH has added classes of LANL employees to the SEC that include all workers through December 31, 1995. For workers for whom partial dose reconstructions are performed, internal doses that may be assigned are limited to those based on bioassay monitoring information in the claim records and to potential intakes (implied by case-specific information) that may be assigned using coworker intakes. For determining when unmonitored intakes may have occurred, the most useful sources of information for this review (beyond internal dose monitoring records) are listed below.

### 5.5.1.1 Energy Employee or Survivor Interview

The interview record offers the energy employee the opportunity to choose from a list of radionuclides to which he or she might have been exposed, and offers the opportunity to include others that are not listed. In addition, the employee is asked about potential exposures and to provide details of exposures in relation to job assignments or incidents. The quality of information about exposures is much lower when the interviewee is a survivor rather than the energy employee. Even many energy employees did not have a detailed knowledge of what they might have been exposed to, or if they had, they might not remember. Some employees check every radionuclide in the list; this is not typically a credible set of exposures because not all of the radionuclides in the interview list were present at LANL.

Often LANL interviewees have excellent recollections of their activities. In dose reconstructions that have been completed to date, this has been shown to be particularly true of researchers and scientists, who typically listed the radionuclides to which they were most exposed. What is more, the individuals most likely to be exposed to LANL's less common radionuclides are researchers and scientists because routine operations for processing did not generally occur on these materials.

### 5.5.1.2 Initial Case File

Another excellent resource for identifying work assignments that could result in unmonitored potential exposures is the initial case file supplied by the DOL. This file often has records that might list detailed job assignments. This information can be used to define a potential unmonitored exposure. In some cases, occupational medical records not included in the DOE files are in the DOL file. These can include records that contain important detail on potential exposures including extremity dose information, nasal count data, airborne contamination reports, and incident reports. Dose reconstructors estimating doses for LANL workers should always, therefore, review the DOL files.

Once the potential for an unmonitored intake is identified, a variety of techniques can be applied to estimate doses from unmonitored intakes as described in this section.

Assignment of coworker dose is done in accordance with the sections below, which discuss specific radionuclides, and with ORAUT-OTIB-0060, *Internal Dose Reconstruction* (ORAUT 2007b). Evaluation of the incidents in attachment E should be performed before assignment of dose from coworker intakes to see if any correspond with a potential intake.

## 5.5.2 Unmonitored Intakes of Primary Radionuclides

Monitoring for primary radionuclides has been performed in most years at LANL, although bioassay results for americium are sparse before the common use of *in vivo* measurements from the 1970s. As described in Section 5.1.5, some work groups or individuals might not have been monitored or might

have been incompletely monitored, even for primary radionuclides. For unmonitored intakes of primary radionuclides that are implied in case records, except americium, dose may be assigned for using the coworker dose intakes in ORAUT-OTIB-0062, *Internal Dosimetry Coworker Data for Los Alamos National Laboratory* (ORAUT 2009b). Reconstruction of internal dose from intakes of americium has been determined to be infeasible for periods before January 1, 1976 (NIOSH 2007); americium dose assignment from this date forward is discussed in the next section.

From 1943 through 1946 there was a significant potential for unmonitored intakes of plutonium (nominally 3% fresh mixture),  $^{210}\text{Po}$  in designated areas, and uranium (nominally either DU or EU even though NU was used extensively in conventional weapons testing).

### 5.5.2.1 Plutonium

In accordance with the designation of the class of employees added to the SEC, reconstruction of internal dose from intakes of plutonium has been determined to be infeasible for periods before 1944 (NIOSH 2007). Bioassay for plutonium was not available before late 1944 and was not completely established until 1945. If a worker had a potential for plutonium exposure from 1943 through 1946, with no bioassay during that period but plutonium bioassay during later years, dose reconstructors may, in some cases, use the later plutonium bioassay with assumptions that are favorable to claimants to bound intakes (Schulte and Meyer 1957). However, if the worker did not participate in a plutonium bioassay program after this period, coworker dose intakes from ORAUT-OTIB-0062 (ORAUT 2009b) should be assigned from January 1, 1944, onward for  $^{239}\text{Pu}$  mixtures.

### 5.5.2.2 Polonium

Polonium was used in nuclear weapons initiators, and the primary manufacture of the parts took place at the Monsanto Chemical Company. In February 1945, the schedule for polonium delivery from Monsanto to TA-1 was increased from a few curies to 100 Ci/mo by June and 500 Ci/mo by December. At TA-1, polonium and radium were handled in H and Gamma Buildings. H Building was used for preparation of neutron and alpha sources for initiators and isotopic experiments. Workers who were involved in these operations were part of Group CM-15. Operations in H Building were carried out between 1943 and July 1945 when operations were moved to the DP Site. Polonium-210 was processed through various operations that included (1) solution chemistry, (2) electrodeposition, (3) high-vacuum distillation, (4) metal plating, and (5) counting and assay of polonium.

Intakes of polonium might be type F or M, and unless bioassay data are precisely fit, the favorable to claimant absorption type should be assumed. Intakes of polonium do not exhibit a long-term excretion in the same manner as plutonium intakes; therefore, dose assessment for early intakes using bioassay data from a later time is not possible.

For polonium, existing bioassay results were used to estimate coworker dose intakes for 1944 to 1956. Before 1944, the reconstruction of dose from intakes of polonium was determined to be infeasible (NIOSH 2007).

### 5.5.2.3 Uranium

Coworker dose intakes for uranium are available beginning in 1947. For exposure before 1947, the uranium intakes assigned for atomic weapons employers conducting uranium machining activities should be assigned to those workers who might have been involved with uranium machining (Battelle 2011) as given in Table 5-21. These intake rates are applicable to machining work involving NU. Potential internal dose from exposure due to machining activities involving highly enriched uranium cannot be reconstructed.

Table 5-21. Hypothetical chronic intakes for NU, 1943 to 1946.

Job category	Intake (pCi/d)
Machinist	19,654
General laborer	9,827
Supervisor	4,914
Clerical	491

#### 5.5.2.4 Tritium

Coworker dose intakes have been calculated for LANL as described in ORAUT (2009b) and cover 1950 and onward. Reconstruction of tritium doses has been determined to be infeasible before the year 1950 (NIOSH 2007).

#### 5.5.2.5 Americium-241.

A large number of bioassay results exist for this radionuclide in the post-1975 period. Although reconstruction of dose from americium before 1976 has been determined to be infeasible (NIOSH 2007), dose reconstruction for the period after 1975 is straightforward due to the availability of personnel monitoring results, which are primarily from *in vivo* monitoring

### 5.5.3 Unmonitored Intakes of LANL Radionuclides other than Primary Radionuclides

Since the mid-1970s, LANL has maintained consistency in its approach to internal dose monitoring. As described by the LANL program (Hoover 2008):

*...internal (and external) dose monitoring is prescribed based on the likelihood of exposure. Routine internal dosimetry programs ... are established and implemented based on worker locations and activities, tailored to the types, quantities, and potential for intake of radioactive materials. Special internal dosimetry measurements are made under unique circumstances – typically radiological events – where additional or unique measurements are warranted, including when unusual results are obtained.*

As listed in the reference, LANL maintains the following bioassay capabilities:

- Actinium-227. LANL has the ability to detect <sup>227</sup>Ac with target *in vivo* measurements. Targeted dosimetry was performed in the past, but no intakes were detected.
- Neptunium-237. A routine threshold for internal dose monitoring exists in LANL TBDs. A contract exists to process *in vitro* samples, and the ready ability to perform *in vivo* measurements exists.
- Protactinium-231. LANL can detect <sup>231</sup>Pa with *in vivo* measurements if warranted.
- Curium-244. LANL maintains *in vitro* capability with targeted bioassay and analysis of samples at ORNL.
- Thorium-230 and -232. LANL maintains an accredited *in vitro* bioassay program under the DOE Laboratory Accreditation Program using a contract laboratory for sample analysis. *In vivo* bioassay might be conducted on the site, and <sup>232</sup>Th has been detected using *in vivo* analysis.
- Strontium-90. LANL is able to detect this radionuclide using *in vitro* analysis. Targeted bioassay has been conducted in the past.

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

#### 5.5.3.1 Actinium-227

Several buildings in TA-21 were used for work with  $^{227}\text{Ac}$  that was used as a substitute for the  $^{210}\text{Po}$  used in weapons initiators. Weapons that used initiators are likely to have been retired by about 1963, and it is not known whether actinium was used as part of a weapon that was actually deployed. The actinium production program was cancelled in 1955 (DOE 1993), and processing activities at LANL would have ended by 1955 or 1956.

Actinium-227 bioassay results exist for LANL workers only for 1954, but have not to date been available to the dose reconstructor. A memorandum from the U.S. Atomic Energy Commission (AEC) that described LANL releases indicates that 2.5 Ci of  $^{227}\text{Ac}$  had been disposed in covered absorption beds, with a decay-corrected inventory of 1.4 Ci as of November 11, 1973 (Wingfield 1974). The originally disposed amount had decayed to approximately 56% of its original value, which implies a residence time of approximately 18 years in the burial ground. This would mean that the material was disposed of in 1955 or 1956.

A filter building was constructed in the late 1940s to process exhaust air from Buildings 146 and 152 in TA-21 (Harper and Garde 1981). The likely primary source of any intake of actinium in the period after 1975 was the decontamination and decommissioning of this facility. Health physics techniques that were applied to the decommissioning and decontamination operation likely controlled the potential intakes of workers, and potential intakes to individuals outside this group would be negligible.

#### 5.5.3.2 Curium-244

The effort to isolate actinides, along with investigation into their chemistry, was conducted partly at LANL, and it appears that use of this radionuclide related primarily to actinide research (Penneman and Keenan 1960). Research was conducted in the Medical Laboratory in TA-1 and in the CMR-4 laboratory at DP West in TA-21. A number of papers were published on the radiochemistry of americium and curium that span a period from 1958 to 1967 (these were identified in searches of the Office of Science and Technical Information electronic archive using keywords *americium* and *curium*). A 1974 AEC memorandum that identified airborne releases in the period before 1967 lists a one-time release of  $^{244}\text{Cm}$  from Building 42 in TA-1 (the medical laboratory or ML Building) over the period from 1944 to 1956 (Wingfield 1974). Curium bioassay results are available for 1955 in a bioassay logbook (LANL 1954–1957) but are not known to be in claimant records.

In 2003, a worker's DOE badge was found to be contaminated with  $^{244}\text{Cm}$ . However, rather than suggesting a generalized exposure hazard, the material was traced back to legacy contamination of objects that had been disposed as part of a remediation activity for the hot cell area of TA-48-1 (the Radiochemistry Laboratory). The material had been stored for tens of years and a sample from the box was labeled in the 1970s (DOE 2008). In this case, *in vivo* and *in vitro* bioassay using the RAS technique were performed; the individual's dose was estimated to be approximately 0.01 rem (Little, Miller, and Guillmette 2003c).

#### 5.5.3.3 Neptunium-237

Limited information was found about neptunium at LANL, but it appears that  $^{237}\text{Np}$  activities have primarily been associated with the Nuclear Fuels Group. The  $^{237}\text{Np}$  metal for the bare criticality experiment in 2002 (Roark 2003) was prepared at the CMR Building (Bodenstein et al. 1999). At that time, LANL maintained a routine dosimetry threshold for  $^{237}\text{Np}$  in internal dosimetry TBDs (Hoover 2008); however, no workers at that time were on routine bioassay programs. This is likely due to the



isolation that is afforded by the hot cells facilities in Wing 9 of the CMR Building. These facilities had been available at LANL since their dedication in 1961 (Bodenstein et al. 1999).

#### 5.5.3.4 Protactinium-231

Papers on the chemistry of protactinium were published from 1959 to 1979 (Kirby 1959; Asprey and Penneman 1964; Smith, Spirlet, and Muller 1979). The foundation work is documented in *Radiochemistry of Protactinium*, which is based on work performed at the Mound site (Kirby 1959).

Another Mound document (Eppley and Valleé 1979) shows that LANL had requested  $^{231}\text{Pa}$  in the amounts of 0.1, 30.1, and 11.0 g in fiscal years 1979, 1980, and 1981, respectively, but the pilot plant producing the material was shut down in 1979 (DOE 1993). Total production for the plant was less than 0.9 g (DOE 1993), so the amounts requested for years 1980 and 1981 could not have been delivered. No production-scale operations were identified at LANL, and there is no mention of the material in airborne radioactivity records or in the waste inventories in the 1974 AEC memorandum (Wingfield 1974). *In vitro* bioassay methodologies are described for  $^{231}\text{Pa}$  in Attachment C of this TBD, and LANL has maintained the ability to detect this radionuclide since the mid-1970s (Hoover 2008).

#### 5.5.3.5 Thorium

Two distinct source terms are associated with  $^{230}\text{Th}$  and  $^{232}\text{Th}$  at LANL.

#### 5.5.3.6 Thorium-230

In the 1950s an effort was underway within the AEC complex to build a production capacity for  $^{230}\text{Th}$  (which was also called ionium). A projected use of the material was for radioisotope heat sources. This work seems to be associated with a number of sites as part of a general effort sponsored by the AEC. No records were found among the available literature that indicated the presence of isolated material other than a number of bioassays for  $^{230}\text{Th}$  in 1958 found in LANL bioassay records. In December 1956, the Mound plant shipped 49 g of  $^{230}\text{Th}$  to the University of California Radiation Laboratory in Berkeley (DOE 1993). The material does not compose a part of the airborne release or waste disposal inventories that accumulated through 1973 in the 1974 AEC memorandum (Wingfield 1974).

#### 5.5.3.7 Thorium-232

It appears that there might have been two periods when  $^{232}\text{Th}$  work was conducted. The first period might be associated with a potential airborne exposure from the Sigma complex over the period from 1944 to 1963, and  $^{232}\text{Th}$  was one of the identified radionuclides (Wingfield 1974). Casting, machining, and powder metallurgy were conducted at the Sigma complex, which housed the Material Science and Technology Division. This facility was built in the 1950s and 1960s (LANL 2007). The material was in the form of ingots and oxides in the building designated as a Thorium Storage Building (Building 159). Building 66 in TA-3 was a warehouse where thorium was stored, and air emissions results from several years indicate airborne releases of  $^{232}\text{Th}$  from this location (Wingfield 1974).

A 1958 reference describes safety measures for uranium and thorium (Stout 1958). The procedures in the reference apply to both elements due to the similarity of the hazards (radioactivity, toxicity, explosion, and fire). The comment is made in this publication that this manual was directing a changed policy toward the handling of thorium, and that it was to be considered in the future an active alpha emitter (Stout 1958). The implication is that  $^{232}\text{Th}$ , with its very long half-life, had been considered essentially stable. It is true that the activity is low for this reason, but the ingrowth of its alpha-emitting progeny occurs within a short period based on their short half-lives. Because the

material had not been assumed to be a hazard before this statement, the material is assumed to consist of  $^{232}\text{Th}$  rather than the more active  $^{230}\text{Th}$ , which with its higher specific activity would have required controls with other active alpha emitters.

#### 5.5.3.8 Mixed Fission and Activation Products

MFPs are typically associated with reactors or with facilities where irradiated reactor fuel is processed. Mixed activation products (MAPs) are the result of neutron irradiation in a reactor core or an accelerator. Before the advent of whole-body counting, radiation protection policy for MFPs was often to collect bioassay samples for gross beta and assign the dose to the most limiting radionuclide. For this reason, bioassay results for a spectrum of radionuclides from this source term are not expected, and gross beta or isotopic samples for  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  are typical. The *in vivo* monitoring program is capable of detecting a large range of fission and activation products. Guidance for estimating dose from gross beta urine samples is given in ORAUT (2007c).

### 5.6 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).

- [1] William Moss. University of California (retired). Radiochemist. 2007. Discussions with William Moss indicate that no correction for recovery was included in the calculation of the polonium activity. There is no indication that any further recalculations were performed on the bioassay results before loading these results in the current bioassay database.
- [2] James Lawrence. University of California (retired). Health Physicist. 2006. Discussions with James Lawrence indicate that most of the early work with plutonium was with what he described as fresh plutonium.

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## GLOSSARY

### activation

Creation of a different isotope by bombarding the parent isotope with neutrons, protons, or other types of radiation that results in the formation of a different isotope or element.

### activity fraction

Proportion of the total activity due to a particular radionuclide.

### 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.

### BiPO<sub>4</sub> era

Period at LANL from October 1949 to January 1957 when plutonium samples were analyzed with the BiPO<sub>4</sub> technique.

### concentration guide

Average concentration of a radionuclide in air or water to which a worker can be continuously exposed without exceeding acceptable radiation dose standards.

### cupferron era

Period at LANL from March 1944 to October 1949 when plutonium samples were analyzed with the cupferron technique.

### diethylene triamine pentaacetic acid (DTPA)

Chelating agents in the form of calcium salts (CaDTPA) or zinc salts (ZnDTPA).

### dry box

Predecessor to the modern-day glovebox made of wood with ports for attached rubber gloves, often filled with inert gas to further contain dust. High-efficiency filters on the exhaust and pressure differentials controlled the spread of contamination to the outside. The operator could perform tasks while completely shielded from exposure to contaminated dust or other airborne material.

### implosion

Sudden inward compression and reduction in volume.

### ionium

Thorium-230, the decay product of <sup>234</sup>U.

### kiva

One of the remotely controlled critical assembly buildings associated with the Critical Experiment Facility at LANL. From Hopi, *ki-* means house, but the meaning of *-va* is unknown.

### nanocurie-year (nCi-yr)

Product of the current incremental body burden times the number of years from the estimated date of the incremental uptake to the date of the calculation. For a deceased person, it is the date of death; for a living person, it is the first date of the month after the latest sample result. Incremental nanocurie-years can be summed to provide total nanocurie-years.

### oralloy (OR-93)

Uranium enriched to 40% or 93% <sup>235</sup>U. The name derives from Oak Ridge alloy.

**rad**

Traditional unit for expressing absorbed radiation dose, which is the amount of energy from any type of ionizing radiation deposited in any medium. A dose of 1 rad is equivalent to the absorption of 100 ergs per gram (0.01 joules per kilogram) of absorbing tissue. The rad has been replaced by the gray in the International System of Units (100 rads = 1 gray). The word derives from radiation absorbed dose.

**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.

**tolerance values**

Value above which the concentration of a radionuclide in a bioassay sample indicates an unacceptable intake or an unacceptable body burden in the individual.

**T<sub>3</sub>O<sub>8</sub>**

Uranium oxide based on Tuballoy (natural uranium).

**Tuballoy**

Natural uranium.



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**A.1 SCOPE**

The purpose of Attachment A is to summarize the data available in Section 5 and to provide supporting information to discussions in the text.

**A.2 CODES USED IN BIOASSAY AND INTERNAL DOSE RECORDS**

**A.2.1 Los Alamos Bioassay Data Repository Codes**

The information for the worker in the current LABDR is provided to the dose reconstructor in PDF format. Each of these PDF files contains pages that define the codes that were used in reporting the data. These include codes that relate to the verification of the identification match and codes that relate to the bioassay results. These codes, as defined at the date of this document revision, are listed in Tables A-1 through A-7. However, the dose reconstructor should review the codes in the file to ensure they are interpreted in accordance with current status.

**A.3 LOCATIONS AND TYPICAL RADIONUCLIDES**

The historical and current typical locations for radionuclides are listed in Table A-8 (Inkret et al. 1998c; ENSR 2002).

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Table A-1. Identification match.

Match type	Match level	Description
s	1	Social Security Number was matched.
n1b	2	First name and entire birth date (month, day, and year) were matched.
nb	2	Name and birth date were matched.
n	3	Full name match.
n1	4	The first letter of the first name was matched (for example, J. J. Smith for John J. Smith) and the last name was matched.
nmd	4	Last name and birth month and birthday were matched.
nmy	4	Name and birth month and birth year were matched.
none	5	No match was found in the LANL personnel tables.

Table A-2. Assessment method codes for bioassay reports.

Code	Description
POR	Polonium by alpha spectroscopy (May 1, 1955, to present)
LS	Liquid scintillation (1981 to present)
UNAA	Uranium by DNAA (January 1, 1978, to January 1, 1992)
GB	Gross beta
T3R	Tritium by radiometric (1950 to 1981)
UF	Uranium by fluorophotometric determination (1950 to 1986)
UR	Uranium by radiometric (gross alpha) (January 1, 1982, to present)
TIMS	Thermal ionization mass spectroscopy (January 1, 1998, to present)
Ref. LA-UR-05-1942	Analysis type not available. Refer to LA-UR-05-1942 (Clark 2005).

Table A-3. *In vitro* collection requirements.

Code	Description
Routine	Collected on a regular schedule
Special	One-time-only collection for incident or follow-up
Prompt action samples	Collected or count performed after incident
Termination sample	Collected or count performed due to termination from program or LANL

Table A-4. *In vitro* collection protocols.

Type	Description
Spot sample	Single void collection
Simulated 24-hr	Morning and evening collection for 2-d period
True 24-hr	A 24-hr urine collection
500-mL sample	Two single voids collected for analysis
Home drinking water	A 500-mL sample of individual's home drinking water
Spot fecal	A single fecal voiding
Autopsy tissue sample	Collected at autopsy – tissue type listed as sample type
Unknown	Sample collection protocol not available. Refer to LA-UR-05-1942 (Clark 2005).
Blood	An intravenous sample of blood; see sample size and units for additional details

Table A-5. Sample units.

Code	Description
G	grams
ML	milliliters
L	liters

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Table A-6. *In vivo* detector systems.

Code	Description
GELI-WHOLE BODY	Whole-body counting system used in SB-14 from (approximately) 08/31/1978, to 09/06/1979, when Ge(Li) and HPGe were moved to SB-16 and new phoswich system was installed in SB-16.
HUMCO II-WHOLE BODY	These systems were in use before 1969 for contamination screening. There are no numerical results in the database for counts made with either of these systems.
HPGE-THYROID	HPGe detector used for thyroid counting.
ORTEC-WHOLE BODY ORTEC WBC	Single ORTEC coaxial HPGe detector over abdomen used as whole-body counter.
PGT-5-DET-CHEST SML	(Sum Low; 300 channels) five-detector array, used 01/01/2004 to 07/22/2004.
PGT-CHEST SMH	(Sum High; 1024 channels) six-detector array, amplification set to capture 0–3 MeV.
PGT-SKULL SML	(Sum Low; 300 channels) six-detector PGT array, on or before 06/20/1996 to after 01/01/2004
PHOS-BACK	Phoswich NaI/CsI.
PHOS-CHEST	Phoswich NaI/CsI.
PHOS-HAND	Phoswich NaI/CsI.
PHOS-HAND WOUND	Phoswich NaI/CsI.
PHOS-LIVER	Phoswich NaI/CsI.
PHOS-SKULL	Phoswich NaI/CsI.
SB14 9X5 NAI-THYROID	9 × 5 NaI(Tl) in SB-14 in SB-14 was the whole-body counting system from about 01/01/1969, to (approximately) 08/31/1978. $L_C$ is estimated as 0.5 × MDA.
SB14 9X5 NAI-WHOLE BODY	The 9 × 5 NaI(Tl) in SB-14 was the whole-body counting system from about 01/01/1969, to (approximately) 08/31/1978. $L_C$ is estimated as 0.5 × MDA.

Table A-7. *In vivo* collection protocols.

Type	Description
BASELINE	Initial count before work
NEW HIRE	Initial count
RECOUNT	Prior count indicates need for recount
REHIRE	Individual rehired and reenrolled in <i>in vivo</i> monitoring
REQUEST	Special count requested
ROUTINE	Count performed on a regular schedule
SPECIAL	One-time-only count for incident or follow-up
STANDARD	Count performed on a regular schedule
TERMINATION	Collected or count performed due to termination from program or LANL
TRANSFER	Final count performed due to individual transferring from radiological organization

#### A.4 COMPUTER CODE FOR VALIDATING PLUTONIUM BIOASSAY SAMPLES

This section is for information only. The dose reconstructor should not attempt to apply these criteria to datasets.

Plutonium samples can be marked as invalid in the database because the results did not fit statistical expectations. However, the dose reconstructor might find it useful to include these sample results.

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Table A-8. Current and historical locations.

Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-0			None	Original town site
TA-1, Original Main TA	1945	1965 active 1975 decommissioned	All	
TA-1, Building D, plutonium chemistry and metallurgy	1943	1954	Pu-239, Pu-238	
			U-238, DU	
			Am-241	Absorption depends on matrix or pure
			Po-210	
			Ba-140	
			La-140	Might indicate Sr-90
TA-1, Building D-2, contaminated laundry		1953	Pu-239; Pu-240; Pu-238; U-236; DU; Po-210; Ac-227; Ra-226	Depends on the compound
TA-1, Building D-5, Sigma vault - storage		1965	Pu-239, U-238	
TA-1, ML Building, medical laboratory			Cm Am	Processing
TA-1, Building C, uranium machining	1943	1964	Uranium	
TA-1, Building G, uranium and graphite sigma pile		1959	Uranium Ra-226	
TA-1, Building H and Gamma Building	1945	1957, 1959	Po-210 MFP <sup>b</sup>	Initiators Cs-137 contamination incident occurred
TA-1, Building HT, heat treatment and machining		1965	NU and EU	
TA-1, Building HT Barrel House, storage		1964	Pu-239, U-238	
TA-1, Building M, processing and recovery EU			EU	Processing, metallurgy, and recovery
TA-1, Building M-1, machining			U-238	
TA-1, Building O		1956	Radium Radon	Radon cooked off sources on a hot plate, Ra/RaBe calibration sources
TA-1, Building Q		1959	Radium Radon	A spill occurred Ra calibration sources
TA-1, Sigma Building		1965	NU, EU, thorium	Casting, machining, powder metallurgy
TA-1, Building TU, machining Tuballoy		1964	NU	
TA-1, Building TU-1, recovery of EU		1964	EU	Furnace for burning rags
TA-1, Building V, Machine Shop	1943	1959	Uranium	Unusual assignments

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Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-1, Building W, Van de Graaff accelerator			Uranium Po-210 Tritium Th-228	"Mesiothorium" Ra-228
TA-1, Building X, Cyclotron			Beryllium, uranium, lithium, tritium, strontium targets. Zn-65	Targets had induced beta activity.
TA-1, Building Y, Physics Laboratory			Tritium, uranium	
TA-1, Building Z, Cockcroft-Walton accelerator			None	
TA-2, Omega West Site				Housed critical experiments
TA-2, Water Boiler	1943	1974	U-235, I-131, I-125, Rb-88, Cs-137, Xe-131, Ar-41 tritium Pu-239	Enriched uranium fuel
TA-2, Clementine	1946	1952	NU, plutonium, I-131, I-125, Rb-88, Cs-137, Xe-131, Ar-41	Ruptured plutonium fuel rod, uranium reflectors
TA-2, OWR	1956	1995	U-235 I-131, I-125, Rb-88, Cs-137, Xe-131, Ar-41 Cr-51, Na-24 Tc-99m	Enriched uranium fuel I-125 production loop schedule – at times operated "around the clock"
TA-3, South Mesa Site, technical facilities	1953		All	Plutonium processing
TA-3, Building 29, CMR (SM-29)	1951		Pu-238	Wing 9 handled irradiated uranium and plutonium in hot cells. Small quantities of uranium and plutonium, MFPs including iodines, Pu-238
TA-3, Building FE-19			Plutonium	
TA-3, Building 34, Cryogenics			Tritium	3,000 Ci HTO released in 1979
TA-3, Building 16, Van de Graaff accelerator			Tritium	800 Ci HTO released in 1977
TA-3, Building 35, press building			NU, U-235	Normal metallic and oxides
TA-3, Building 39, Tech Shop			DU	

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Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-3 SM-40, Physics			All <sup>c</sup>	Incident contaminated large portion of building with Po-210 through ventilation (late 1950s or early 1960s)
TA-3, Building 66, Sigma Complex, metallurgy and fabrication	<1957		EU, DU powders, NU, plutonium, thorium	Plutonium processing Normal metallic and oxide uranium (McKown 1958)
TA-3, Building 102, Tech Shops			Uranium	
			Plutonium	
TA-3, Building 141, Rolling Mill			DU	
			Plutonium	
TA-3, CMR Building, Wings 3,5,7			H-3	HTO, HT
TA-3, CMR Building, Wing 9	1961		Cs-137	Potential for low-level chronic intake in hot cell work
			MFP <sup>d</sup> including I-131	
			Pu-238, -239, -240	0.1–10 µm activity median aerodynamic diameter (AMAD), oxide, nitrate, fluoride, and metal. Oxide is most common.
TA-3, IBF <sup>a</sup> , SM-16			I-125	Iodide, labeled organics
			H-3	Labeled DNA precursors (OBT), water (HTO), HT
			P-32	Labeled organics, phosphates
TA-3 Tritium Instrument Calibration Facility, SM-40			H-3	HTO, HT
TA-3-184, Occupational Health			Pu	
TA-3-216 Weapons Test Support			Pu	
TA-3-700, SM-700 Acid Neutralization and Pump Building			Pu	
TA-4, Alpha Site		1956	DU	Firing site until 1956, materials disposal site C

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Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-6, Two-Mile Mesa Site	1944	1950	DU	Detonator manufacturing
TA-8, Nondestructive testing	1984		Pu-239; Pu-238	Gun firing site
			U-235; DU	
			Co-60; Ir-192	
			Cs-137	
TA-10, Building, CMR-10, Bayo Canyon Site	1944	1950	Sr-90,	RaLa radiochemistry
			DU, NU	
			La-140	
			Ba-140	
TA-11, K Site	1947		Ra-226-Be	20-MeV betatron
TA-15, Electron accelerator	1962	Present	Pu-239	PHERMEX
			DU	
			H-3	
TA-16, S Site, WETF	1989	Present	Pu-239	Explosive casting and machining
			DU	
			H-3	
TA-16, WETF, Bldg 205		Present	H-3	Labeled DNA precursors (OBT), water (HTO), HT
TA-18			U-235; U-233 Pu-239; Pu-240;  MFP <sup>b</sup> ; I-131; polonium	0.1–10 μm AMAD, oxide, nitrate, fluoride, and metal. Oxide is most common. Ruptured polonium source, 1953
TA-18, Pajarito Laboratory, Rover reactor, criticality experiments	1946	1973	MFPs <sup>b</sup>	Betatron used from 1951 to 1954, EU metal sphere 1952; plutonium core added 1 year later. 1954 unreflected, delta phase plutonium.
TA-19, East Gate Laboratory		1962		None
TA-21, DP-West, plutonium facility	11/1945		WGpu	
TA-21, CMR, heat sources			Pu-238	Accident with glovebox breach, 1971
TA-21 DP West			Pu-238, -239, -240	0.1–10 μm AMAD, oxide, nitrate, fluoride, and metal. Oxide is most common.
TA-21, Buildings 2 and 3, wet chemistry		1982	Pu	1958 accident, separated phases in plutonium process tank, unshielded tank
TA-21, Buildings 4 and 5, dry chemistry		1981	Pu	
TA-21, Building 12, filter building		1975	Pu	Actinium-contaminated
TA-21, Building 3, oxalate precipitation operations			Pu-239 Pu-238 U-235	
TA-21, Building 4	1945	1948	EU hydride	



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Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-21, Building 4	1960		Pu-239	Hot cell examine irradiated plutonium and EU fuel elements
TA-21	1965		Pu-238 and Pu-239	
TA-21, Building 5, plutonium fabrication		Limited use in 1975	Pu-239, -238	Fire contaminated exhaust filter, 1959
TA-21, Building 150, plutonium fuels development, heat sources development	1963		Pu-238, -239	Sealed capillary broke, 2,800 x MPC 10/1970
TA-21, Building 210, plutonium research			Plutonium	
TA-21, Building DP-East			EU	
TA-21, Building 155, TSTA, deuterium and tritium fuels.	1984	1990	HT and HTO	>10 billion Ci. Equipment failure – H-3, 13.8 Ci released
TA-21, Buildings 151, 152, experimental program	1945		Po-210	Produced initiators
			Ac-227	Produced initiators
TA-21, Building 155	1949	1984	Po-210	Produced initiators
			Ac-227	Produced initiators
TA-21, Building 153	1945	In service until 1970–1973	Po-210	Produced initiators
TA-21 DP-East, TSTA Tritium Test Assembly Facility, Building 155 and the Salt Laboratory, Building 209			H-3	Labeled DNA precursors (OBT), water (HTO), HT
TA-21, Liquid Waste Reprocessing, Buildings 35 and 257	Late 1940s	1986	All <sup>c</sup>	Plutonium and transuranic liquid wastes
TA-22, TD Site			Ac-227	Produced initiators
TA-23 NU Site	1945	1950	Unknown	Firing site
TA-24 T Site	1944		DU	Facilities transfer to TA-16
TA-25, V Site	1944	1946	DU	Taken over by TA-16
TA-26, D Site	1946	1948	U-235, U-238 H-3	Storage vault
TA-27, Gamma Site (Far Point)	1945	1947	Pu-239 DU thorium	Plutonium gun assembly
TA-28, Magazine A	1979	Present	DU	Firing site
TA-29, Magazine B		1957	DU	Explosives storage area
TA-30 through TA-32				Unknown
TA-33, HP Site, High Pressure Tritium Laboratory, Building 86	1950s	1980s late	H-3	Tritium oxide (HTO), tritium gas (HT)
TA-35, LAMPRE	1955	1967	MFPs <sup>b</sup>	Molten plutonium fuel
			Sr-90	
			Co-60	
			VFPs <sup>d</sup>	
			MAP	
TA-35, LAPRE I, LAPRE II test reactors	1955	1960	MFPs <sup>b</sup>	Highly enriched U fuel
			Sr-90	
			Co-60	
			VFPs	
			MAPs <sup>e</sup>	
TA-35, Target Fabrication Facility (TFF), TSL-213			H-3	Labeled DNA precursors(OBT), water (HTO), HT

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Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-35, Ten Site, CMR-10	1950	1963	La-140, Ba-140	Sr-90 contamination suspected (F,S)
TA-35		1981	U-235 DU Np-237 Plutonium Polonium	General site
TA-35, Laser Fusion Research	1974		Unknown	
TA-36, Kappa Site	1950	Present	DU	
TA-37, Magazine Area C			DU	
TA-39, Ancho Canyon Site	1955	1960	NU, DU thorium	Firing points
TA-40, Detonator Firing Site			H-3	
TA-41, W Site, Weapons Group WX			H-3 plutonium uranium americium	Engineering of nuclear components Fabrication of test materials
TA-41, Ice House, Building 4			H-3	Tritium oxide (HTO), tritium gas (HT)
TA-42, Incinerator Site		1970	All <sup>c</sup>	Reduced low-level plutonium-contaminated waste
TA-43 HRL <sup>a</sup>	1953	1970	I-125 H-3 C-14 P-32	Iodide, labeled organics Labeled DNA precursors(OBT), water (HTO), HT Labeled DNA precursors, Labeled organics, phosphates
TA-43	1953	1970	All <sup>c</sup>	
TA-45, Radioactive Liquid Waste Treatment Plant, WD Site	1951 1960	1964–operations ended 1963 1967–decom.	All <sup>c</sup> MFPs <sup>b</sup>	Removed plutonium before discharging effluents
TA-46, WA Site	1950	1974		Rover batteries
TA-46, WA Site	1976	1980s	U-235, -238 Thorium	Uranium isotope separation
TA-48, Radiochemistry Site	1950s	Present	All MAPs <sup>e</sup> MFPs <sup>b</sup>	Actinide chemistry and hot cell isotope production
TA-48, Nuclear Chemistry	1950s	Present	Se-75 H-3 Cd-109 I-131	Spallation product, seen in hot chemistry on targets Tritium oxide (HTO), tritium gas (HT) CL/NO <sub>3</sub> <sup>-</sup> mixture loaded in SnPO <sub>4</sub> resin. Cd phosphate is most probable material of intake, very soluble, 1 μm AMAD Fission product chemistry
TA-49, Frijoles Mesa Site	1960	1961	H-3 plutonium uranium	

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Location	Start	Demolished or decommissioned	Nuclide <sup>a</sup>	Comment
TA-50, Waste Management Site	1963		Pu-238, -239, -240 All <sup>c</sup>	0.1–10 µm AMAD, oxide, nitrate, fluoride, and metal. Oxide is most common.
TA-51, Environmental Research Facility	1962	Present	Co-60 Sr-90	Animal exposure facility. Presently environmental impact research.
TA-52, Reactor Development Site	Late 1960s	1970	U-238, Pu-238 H-3 VFPs <sup>d</sup> krypton, xenon	High-temperature, gas-cooled, graphite reactor, UHTREX (Ultra-High Temperature Reactor Experiment)
TA-53, Los Alamos Neutron Science Center, largest accelerator facility, Los Alamos Meson Physics Facility (LAMPF)	1972		C-11, N-13, O-15, Ar-41 I-131	Short-lived air activation  Medical isotope production
			Induced activity in uranium targets, corrosion products	
TA-54, Waste Storage Facility			Pu-238, -239 -240 All <sup>c</sup>	0.1–10 µm AMAD, oxide, nitrate, fluoride, and metal. Oxide is most common.
TA-55, Plutonium Facility (PF-4)	1969		H-3	Labeled DNA precursors (OBT), water (HTO), HT
TA-55, Plutonium Facility (PF-4)	1969		Pu-238, -239, -240	0.1–10 µm AMAD, oxide, nitrate, fluoride, and metal. Oxide is most common.
LANSCE			Be-7	2A metal, metalloid behavior, very reactive, occurs in virtually massless quantities, typically seen when target cells are opened for maintenance, usually in oxide form
			C-11	Byproduct at LANSCE, seen in workers during beam cycle
			N-13	511 keV during beam cycle

- Intakes of labeled compounds do not follow the default ICRP Publication 68 models (ICRP 1995). However, doses based on bioassay results should be estimated using standard Project assumptions for the analyte.
- Cs-137.
- All = Pu-239, Pu-240, Pu-238 (type S-M); U-235, DU (type M); H-3 (SR-2,F); Po-210 (type F or M); Ac-227 (type S); Ra-226 (type S).
- VFPs = volatile fission products.
- C-11, N-13, O-15, Ar-41, Be-7, Na-22, Na-24, Co-58, Co-57, Mn-54, Mn-52, V-48.

Review of studies listing the bioassay results of the original 26 workers with intakes of plutonium shows considerable variability between samples in both the long and short term (Voelz et al. 1979). Section A.10 contains a complete listing of bioassay results for one individual. These results display typical variability seen in other individuals involved in the study. If the dose reconstructor chooses to maintain the result as invalid, the logic below will provide an explanation of how the data were evaluated. The information in the following sections is from Lawrence (1978).

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**A.4.1 Validations Techniques PUQFUA2**

The PUQFUA programs were revised through PUQFUA4. Therefore, other validation protocols might have been in effect after 1978.

1. The purpose of the validation is to determine high results to invalidate the sample.
2. Only samples of a single era could be used to validate samples from that era (e.g., samples analyzed by BiPO<sub>4</sub> or cupferron could be used to validate samples of the era).
3. Samples are considered positive if they exceed the value of LEAST. If the sample passes Test A, do not perform Test B. The tests are applied sequentially to the largest results first, then the second largest, etc., until all results exceeding LEAST are examined. If results are below LEAST, use Test C. LANL recognizes that unique situations occur when:
  - a. An individual urine result should be retained despite its being invalidated by the test procedures, and
  - b. An individual urine result should be invalidated despite its being validated by the test procedures.
4. Samples surviving Tests A, B, and C would be subjected to the PUQFUA1 primary validation technique.

Cupferron era		Other era	
LEAST	High	LEAST	High
0.10 pCi	0.8 pCi	0.075 pCi	0.4 pCi

**Test A**

1. If there are four or more positive samples (greater than LEAST)  $\pm 1$  year – continue.

Select the four closest positive samples,  $\pm 1$  year.

- a. Find the average of the four samples.
  - b. Find the average of the standard deviation.
2. If the sample result is less than the average plus one-fourth the standard deviation average, retain the sample.

**Test B – samples not passing the Test A criteria**

1. If there are not three or more positive samples after the sample being tested, within the same sample era, the sample is validated by default.
2. Determine if one or more retained potential accident dates occurred between the sample being tested and the next earlier positive sample.
3. Perform the calculation of the expected excretion levels on the next three positive samples.
4. Perform the calculation using the retained potential accident date or 15 days before the sample date if no potential accident date is available.

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$$RF = [I \div (E + I)]^{0.74} \quad (A-1)$$

where:

*RF* = reduction factor

*I* = number of days between assumed exposure and sample

*E* = number of days between the sample being tested and the date of the later sample

$$CL = RF \times UR \quad (A-2)$$

and

$$PS = RF \times U2S \quad (A-3)$$

where:

*CL* = calculated urine level

*UR* = result

*PS* = pseudo-standard deviation

*U2S* = standard deviation of the result

1. If the actual measured urine excretion level of two of the three later samples is greater than the calculated urine excretion rate minus one-third of the pseudo-standard deviation on the appropriate dates, retain the sample.
2. If two or more retained potential accident dates result in conflicting decisions, retain the sample.

### Test C

Test C tests all samples less than LEAST in any era. Test C eliminates those low or negative urine results that, if left valid, would cause subsequent invalidation of high results that preceded them.

All low and negative samples are retained until the first sample exceeding LEAST is encountered.

### Cupferron Era

1. If there is one sample >0.1 pCi in the set and the standard deviation is <0, the sample is rejected as too small.
2. If there are three or more samples >0.1 pCi in the set and the standard deviation is <0.1 pCi, the sample is rejected as too small.

### BiPO<sub>4</sub> Era

1. If there is one sample >0.075 pCi in the set and the standard deviation is <0, the sample is rejected as too small.
2. If there are three or more samples >0.075 pCi in the set and the standard deviation is <0.075 pCi, the sample is rejected as too small.

### NTA-ZnS or PHA Era

1. If there are at least two samples >0.075 pCi in the set and the standard deviation is ≤0, the sample is rejected as too small.

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2. If there are at least five samples >0.075 pCi in the set and the standard deviation is ≤0.04, the sample is rejected as too small.

**Surviving Samples**

Samples surviving the above tests are examined in relation to the earliest date of a validated HIGH sample. The date of the earliest sample that exceeds the high criteria is used to test the surviving samples. All surviving samples dated before the first high samples are kept with the added notation of no highs.

**Samples Occurring after the First High Sample**

1. The average of the four retained samples greater than LEAST and closest in time to the sample being tested is calculated. There is no time limit within the analysis era on dates of samples greater than LEAST.
2. If the standard deviation exceeds this average, the sample being tested is kept; otherwise, it is rejected.
3. The sample can be rejected if the result is <0 or the standard deviation is <LEAST.
4. If there are not four retained samples greater than LEAST, the sample being tested is kept by default.

**A.4.2 PUQFUA Primary Validation Technique**

1. Starting with the latest sample and working toward earlier samples, successive pairs are examined. The later sample of each pair is used to test the validity of the earlier sample.
2. If the earlier sample is validated, it is used as the later of the next pair of samples to be tested.
3. If the earlier sample is invalidated (i.e., set equal to zero for the calculations), the later sample of that pair remains the later of the next pair to be tested. The sample next earlier than the invalidated sample becomes the one to be tested.

$$U_c = U_e[(D_e - E_1)/(D_l - E_1)]^{0.74} \tag{A-4}$$

and

$$\sigma_c = \sigma_e[(D_e - E_1)/(D_l - E_1)]^{0.74} \tag{A-5}$$

and

$$(U_c - U_l) > 1.282Z_{1-\alpha} [(\sigma_c^2/n_c) + (\sigma_l^2/n_l)]^{1/2} \tag{A-6}$$

where:

- $D_e$  = integer date of  $U_e$
- $D_l$  = integer date of  $U_l$
- $E_1$  = estimated integer date of intake
- $U_c$  = calculated urine result expected from  $U_e$
- $U_e$  = earlier dated urine result

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$U_l$  = later dated urine result

$\sigma_c$  = calculated standard deviation of  $U_c$

$\sigma_e$  = standard deviation of  $U_e$

$\sigma_l$  = standard deviation of  $U_l$

1.282 $Z_{1-\alpha}$  = standard normal variable for  $(1 - \alpha)$  one-sided confidence interval;  $\alpha = 0.1$

### Hypothesis

4. Does the calculated urine result  $U_c$  exceed the measured result  $U_l$ ?
5. The  $U_l$  is permitted to exceed  $U_c$  because the larger value of  $U_l$  might have resulted from an additional intake between  $D_e$  and  $D_l$ .

The basic assumption is that, in the event of no additional intake, the urine level at any later time can be calculated from Langham's urinary elimination equation, provided the date of intake has been established. (PUQFUA4 did not use Langham's urinary equation, but rather the equations developed in the late 1980s or early 1990s, which take into account very-long-term elimination 20 or more years after uptake. Again, these are discussed fully in the description of PUQFUA4.)

6. Where  $n_c = n_l = 1$ ,  $U_c$  does exceed  $U_l$  at the chosen level of significance, and sample  $U_e$  on date  $D_e$  is rejected as invalid.

### A.5 SUMMARY OF COMPUTER CODES

The PUQSRT code provided a summary report from the PUQFUA database of:

1. Individuals with a potential accident recorded but no urine sample and individuals with a potential accident recorded after their latest urine sample
2. A list of individuals whose last urine sample indicated a body burden of >2 nCi
3. A list of individuals whose next-to-the-last urine sample indicated a body burden increase of >2 nCi
4. A list of individuals whose total body burden is >10 nCi

Tracking is available for  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{242}\text{Pu}$  body burdens. These reports were performed routinely.

The PUQFUA code provided calculation of  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ , and  $^{242}\text{Pu}$  body burdens and validation tests of the analytical results for the samples. Various versions were used over time as the code was upgraded to accommodate additional models and other features. The original version became operational in 1959. This program utilizes a set of power function elimination equations for the excretion of plutonium. Lawrence (1978) discusses the history and development of the program including its use to track accidents and potential accidents and validate urine samples. PUQFUA1 and PUQFUA2 are revisions to the original code that address recognized deficiencies. When possible, results of the calculations have been compared with autopsy data. PUQFUA1 tended to overestimate by a factor of 2 to 8; this was corrected in PUQFUA2. The latest version was PUQFUA4.

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Internal monitoring of individuals with a potential for exposure to plutonium was controlled using a computer code.

The Z1YRPUU code provided a method to mark Zia employees who were permitted access to plutonium areas based on the submission of their annual plutonium bioassay samples. This program was in use from 1976. Table A-9 lists the areas and exempt areas for access.

Table A-9. Zia employee access to plutonium areas (University of California 1978a).

Area	Urine sample within 425 d of entry	Exempt
Job requiring respiratory protection	X	
Modifications or repairs on dry boxes or other highly contaminated equipment	X	
Replacement of plutonium-contaminated filters at all sites	X	
Janitorial (long-term) work in plutonium operation areas	X	
Long-term operations (weeks) in areas of low levels of plutonium contamination (>1,000 dpm-60 cm <sup>2</sup> and <10,000 dpm-60 cm <sup>2</sup> )	X	
Decontamination of plutonium spills with >10,000 dpm-60 cm <sup>2</sup>	X	
Work in burial pits at TA-54 when personnel contamination potential is moderate to high	X	
Short-term jobs (2–3 d) when sizable quantities of plutonium (grams of Pu-238 or kilograms of Pu-239) are present in dry boxes (even when work is being done outside dry box)	X	
Supervisory personnel		X (base urine sample on record)
Short-term jobs (2-3 d) in areas of CMR Building, Ten Site, TA-50, TA-55, TA-54, TA-18, TA-48, or TA-21, where there is little plutonium contamination (<1,000 dpm-60 cm <sup>2</sup> )		X
Jobs in other minimum exposure potential areas when respiratory protection is not required and possibility of plutonium contamination is minimal		X

PUANUD (Plutonium Urine, Accident, Lung, and Wound Data) was a code to permit entry of urine assay, potential accident, lung count, and wound counting data into a single file that was used as input for the PUQFUA calculations. The software was used beginning in 1974 for <sup>238</sup>Pu and <sup>239</sup>Pu. Flags were available for the following categories.

1. Unspecified type of accident;
2. Wound case with excision;
3. Wound count equal to or greater than 0.2 nCi;
4. High room air count if next year's urine shows obvious increase;
5. High nasal count if next year's urine shows obvious increase;
6. Nose count over 1,000 dpm; and
7. Other accident if next year's urines show obvious increases.



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**A.6 CURRENT AND HISTORICAL *IN VITRO* BIOASSAY PROGRAMS**

Historically, the *in vitro* bioassay program included the nuclides listed in Table A-10. The current *in vitro* bioassay program includes the nuclides and techniques listed in Table A-11. The *in vitro* bioassay samples have been taken at LANL throughout the history of the bioassay program. Sensitivities vary with technique and period.

Table A-10. Historical *in vitro* bioassay.

Nuclide	Nuclide
Tritium	Iodine
Radium and daughters	Fission products
Uranium	Activation products
Plutonium	Other alpha emitters
Polonium	Other beta/gamma
Curium	Americium
Strontium	

Table A-11. Current *in vitro* bioassay performed, 1997 (Inkret et al. 1998a).

Material	Analytical technique	Number monitored
Tritium	Liquid scintillation	139
Uranium	Alpha spectroscopy	66
Plutonium	Alpha spectroscopy	1,467
Plutonium	TIMS	408
Americium	Alpha spectroscopy	75
Strontium	Liquid scintillation (gross beta counting–LSC sent to contract laboratory)	4

**A.7 ESTIMATION OF DATE OF INTAKE**

The following guidance from Lawrence (1978) might be relevant to an estimate of the date of plutonium intake, based on routine and special sampling protocol, when no intake date is recorded or to understand the LANL rationale for selection of an intake date listed with the bioassay results if no incident date is provided in the records. Otherwise, the dose reconstructor should use current models and best judgment in the determination of the dates for acute intakes.

1. Halfway between dates of consecutive pairs of samples if no potential accidents are recorded between samples.
2. One-half day before later sample of a pair if a potential accident is recorded on the date of the later sample.
3. The earliest potential accident date if several occur between the paired samples.
4. One-half day before the initial sample if the potential accident is recorded on the date of the initial sample.
5. The earliest potential accident date before the initial sample if any potential accidents occurred before the initial sample.
6. Fifteen days before the initial sample if no potential accidents occurred before the initial sample.

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7. Estimated date of intake of the later sample if a pair is reassigned to be the estimated date of intake of the earlier sample, if the earlier sample is invalidated and a potential accident date occurred on the same date as the earlier sample.

**A.8 ROUTINE SAMPLING PROCEDURES**

Contamination of bioassay samples was a concern throughout the history of the program. Various sampling protocols were followed to minimize the potential for contamination, to detect levels above the tolerance level, and to attempt to obtain an equivalent 24-hour collection. Table A-12 lists the sampling protocols.

Table A-12. Routine sampling procedure.

Years	Nuclide	Protocol
1944	Plutonium	Collected on 24-hr urine sample in clean areas after decontamination. Working in plutonium areas operations – weekly, daily nasal swabs. Blood counts every 6 wk (Hempelmann 1944).
1945–1952	Plutonium	Collected 24-hr urine sample away from working environment to ensure contamination-free urine (Clark 2005).
1953–1957	Plutonium	Metal kit for collection of last voiding of day and first in morning for 2 consecutive days (Clark 2005).
1958–1967	Plutonium	Four 1-pint bottle disposable kit. More closely approached volume of true 24-hr urine sample. Reduced potential contamination.
1944	Polonium	Collected on 24-hr urine sample in clean areas after decontamination. Working in polonium areas operations – weekly, daily nasal swabs. Blood counts every 6 wk (Hempelmann 1944).
1945–1952	Polonium	Collected 24-hr urine sample away from working environment to ensure contamination-free urine (Clark 2005).
1970–1975	Tritium	>150 µCi/L – 1/d; 150-75 µCi/L – every 2 wk; 74 -24 µCi/L – 1/wk (Healy 1970).
1975–present	Tritium	Routine – every 2 wk; or 2 hr after expected exposure then if <1 - no more samples, 1-10 µCi/L – 1 wk in next month, 10-100 µCi/L – weekly samples, >100 µCi/L – daily samples.
1944–1967	Uranium	Weekly urinalysis (Hempelmann 1944).
1967–present	Uranium	Annual, including drinking water sample since 1992 (Inkret et al. 1998c).

**A.9 SUMMARY OF ANALYTICAL TECHNIQUES FOR PLUTONIUM**

Various analysis techniques have been employed for plutonium over the history of the program. The techniques are summarized in Table A-13.

**A.10 EXAMPLE BIOASSAY RESULTS**

Table A-14 contains actual bioassay results from an individual whose 1959 autopsy results confirmed plutonium in the liver, lungs, bone, and lymph nodes. Plutonium-238/239 ratios indicative of early and late mixtures of plutonium were identified in the autopsy tissues. The late mixtures of plutonium were found in the lung. During his work at LANL, the individual had only 18 nasal swipes above 50 dpm (the level of significance). All the nasal swipes greater than 50 dpm were before 1948. Only one incident in 1955 showed a nasal swipe of approximately 28 dpm. Records of the work locations and results were carefully maintained (Foreman, Moss, and Langham 1960). No significant incidents or accidents were noted in the individual's records. The probable intake scenario for this individual is listed as long-term chronic or intermittent low-level inhalation. Review of the bioassay results should assist the dose reconstructor when reviewing the variability of other data sets.

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Table A-13. Plutonium urinalysis sensitivities and analytical techniques (Clark 2005).

Period	Urinalysis method
1944–1952	Plutonium extracted by cupferron chloroform, gas flow counter (1944), Simpson alpha counter efficiency = 50%, background = 1 cpm to 0.1 cpm (1945+). Could not differentiate polonium and plutonium. Selected based on potential exposure. 12% of blanks showed ~1.4 cpm or 0.8 pCi (1945). Assay results reflect no chemical blanks, recovery factor, or counting geometry corrections.
1949–1957	Bi-phosphate-La fluoride coprecipitation, Simpson alpha counter, efficiency = 50%, background = 0.1 cpm.
1957–1963	Bi-phosphate precipitation – alkaline earth-phosphate precipitation, plutonium plated on stainless-steel disk. NTA counting method, 1,000-min exposure to emulsion, background = 0.007 dpm.
1963–1965	Ion exchange/plutonium separated on an ion-exchange resin. Electroplated for NTA counting, 84% ±14% recovery.
1966	ZnS counter.
1967–1971	Either ZnS or alpha pulse height analysis (PHA) RAS permit measurement of Pu-238; background = 0.003 ±0.003 cpm.
1971–present	All alpha PHA, computerized spectrometry.
09/18/1980	Began Pu-242 analysis.
1982–1986	Coprecipitation (alkaline earth PO <sub>4</sub> or oxalate). Alpha spectrometry (60,000-s count time) or rapid Alpha Phosphor Scintillation. Counting (3,600 s).
1997	Alpha spectroscopy, based on class Y, 1 µm, Pu-239 (Inkret et al. 1999).
1997–present	TIMS with ultra-trace chemistry and class-100 clean room and alpha spectroscopy methods. Based on class Y, 1 µm, Pu-239, use of alpha spectroscopy allows direct measure of chemical efficiency and detection of Pu-238 (Inkret et al. 1999).

Table A-14. Example urine bioassay results (Foreman, Moss, and Langham 1960).

Date (left/right nasal count)	Average room air concentration (dpm/m <sup>3</sup> )	Activity (dpm/24 hr)
08/09/1946 (189/320 dpm 07/29/1946)	6–188	1.2
09/19/1946 (149/19 09/05/1946)		4.7
12/18/1946 (57/68 12/10/1946)		1.7
04/18/1947 (164/106 dpm 12/30/1946); (102/61 dpm 01/21/1947); (91/135 dpm 04/01/1947)	11–98	0.7
05/23/1947		0.7
06/26/1947		0.7
07/30/1947 (144/40 dpm 07/07/1947)		0.0
08/27/1947		1.0
10/02/1947		1.5
11/07/1947 (120/78 dpm 10/03/1947)		0.8
12/08/1947		4.0
01/13/1948	24–69	1.0
02/13/1948 (0/59 dpm 02/10/1948)		0.0
03/19/1948		0.0
04/22/1948 (86/3 dpm 04/26/1948)		2
06/23/1948 (244/72 dpm 06/10/1948)		3.7
07/22/1948 (72/1 dpm 07/02/1948)		0.0
08/19/1948 (65/0 dpm 08/02/1948)		0.8
09/20/1948		3.0
10/26/1948		2.0
11/22/1948		2.5
12/21/1948 (50/38 dpm 12/01/1948)		0.0

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Date (left/right nasal count)	Average room air concentration (dpm/m <sup>3</sup> )	Activity (dpm/24 hr)
01/24/1949	19-72	(Moved to uranium work)
01/31/1949		2.0
07/14/1949		1.2
02/08/1950		0.8
09/01/1950		0.1
02/28/1951		0.8
09/04/1951		0.7
05/19/1952		0.3
12/14/1952		0.0
09/04/1953		0.0
06/04/1954		Off uranium
06/18/1954		0.0
06/08/1955		(Moved back to plutonium)
08/01/1955	3	1.2
08/12/1955 (22/28 dpm 08/09/1955)		0.6
08/19/1955		0.0
09/30/1955		0.7
11/14/1955		0.7
12/27/1955		0.6
02/09/1956	3	0.0
04/05/1956		0.7
04/30/1956		0.0
06/08/1956		0.4
07/20/1956		0.6
08/23/1956		0.5
09/25/1956		0.0
10/24/1956		0.0
11/23/1956		0.0
12/17/1956		0.1
01/31/1957	4	0.23
02/28/1957		0.68
04/12/1957		0.22
05/14/1957		0.12
06/14/1957		0.11
07/15/1957		0.03
08/19/1957		0.00
09/20/1957		0.20
10/22/1957		0.21
10/31/1957		0.39
11/14/1957		0.51
01/10/1958	4	0.00
02/21/1958		0.65
03/25/1958		0.51
05/07/1958		0.025
06/19/1958		0.55
07/30/1958		0.49
09/15/1958		0.79
11/28/1958		0.47

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A body burden of plutonium was potentially maintained by this individual since 1946. The bioassay results varied as listed.

Average room air activity concentrations for each year are listed. When more than one room was involved, the range of average concentrations is listed. The period from 1946 to 1949 involved plutonium nitrate, plutonium oxalate, and plutonium fluorination. The period from 1955 to 1958 involved primarily liquid-liquid extraction of plutonium under greatly improved exposure conditions.

### A.11 ROUTINE SAMPLING FREQUENCY

Routine sampling frequencies place upper bounds on the potential exposure for monitored individuals with all results less than detection levels. Table A-15 lists routine sampling frequencies for plutonium. Although sampling of individuals with the highest potential for intakes was performed from the beginning of the program in 1944, no specific information on the nonincident sampling program is available before Lawrence (1967). Table A-16 lists routine sampling frequencies for  $^{241}\text{Am}$  exposures; routine samples were not performed for  $^{241}\text{Am}$  before 1998. Routine and postexposure protocols are combined in Table A-17 for tritium. Table A-18 lists frequencies for uranium. Routine work or frequent entry in an area with beta/gamma emitting radionuclides currently requires annual whole-body count (LANL 2004).

### A.12 SAMPLING PROTOCOL FOR ACCIDENTAL EXPOSURES

Awareness of the sampling protocol associated with exposures provides guidance for the association of results with intake regimes. Results corresponding to an incident can be expected to follow the appropriate protocols listed below. No information on the protocols is available before Lawrence (1967). It should be assumed that the protocol continued until the year of the next dated reference unless otherwise stated. Tables A-19 to A-23 list protocols for accidental exposures.

### A.13 SUMMARY OF *IN VITRO* AND *IN VIVO* SENSITIVITIES

Table A-24 lists sensitivity data for *in vitro* bioassay.

Tables A-25 and A-26 list sensitivity data for *in vivo* testing.

Monitoring was available for the liver and thyroid using the HPGe detector. In 1977 the MDA for the liver scan was reported to be less than 1 nCi for  $^{235}\text{U}$  and  $^{241}\text{Am}$  (University of California 1977b).

### A.14 *IN VIVO* BIOASSAY ROUTINE FREQUENCY

Table A-25 provides the standard protocol for scheduling *in vivo* counts. This is the current protocol. No information is available on when the protocol was first established; however, it has been in effect as long as anyone currently involved in the program can remember.

### A.15 HUMAN EXPERIMENTATION

The first research into the behavior and deposition of plutonium in the human body began by accident in August 1944 when a vial containing 10 mg of plutonium exploded in the face of a [title redacted], who swallowed a portion of the material. The chemist then used his own urine to develop a bioassay analysis to detect the early analysis technique for plutonium. However, this information did not

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Table A-15. Routine plutonium sampling frequency.

Frequency <sup>a</sup>	1944 (Kolodney 1946)	1967 (Lawrence 1967)	1973 (Lawrence 1973)	1998 (Inkret et al. 1998 <sup>b,c</sup> ; LANL 2004a)
Monthly	Great exposure			
Quarterly	Moderate exposure	<ol style="list-style-type: none"> <li>Persons working with <math>\geq 10</math> g Pu-239 in chemical or metallurgical operations, inside or outside glovebox</li> <li>Persons with 50% body burden (MPBB = 0.04 <math>\mu</math>Ci Pu-239)</li> </ol>	<ol style="list-style-type: none"> <li>Persons working with <math>\geq 10</math> g Pu-239 or <math>\geq 0.04</math> g Pu-238 (<math>\sim 0.6</math>–<math>0.7</math> Ci of either) inside or outside glovebox, or</li> <li>Persons with body burden <math>&gt; 20,000</math> pCi</li> </ol>	
Semiannual (biannual)	Slight exposure	<ol style="list-style-type: none"> <li>Persons working with <math>&lt; 10</math> g of Pu-239 in chemical or metallurgical operations</li> <li>Supervisors of quarterly sampled category</li> <li>Persons with 25% burden (MPBB = 0.04 <math>\mu</math>Ci Pu-239)</li> </ol>	<ol style="list-style-type: none"> <li>Persons working with Pu but <math>\leq 10</math> g Pu-239 or <math>\leq 0.04</math> g Pu-238 in chemical or metallurgical operations,</li> <li>Supervisors of the quarterly sampled category, or</li> <li>Persons with <math>&gt; 10,000</math> pCi body burden but <math>&lt; 20,000</math> pCi</li> </ol>	<ol style="list-style-type: none"> <li>Working with <math>\geq 0.04</math> g (0.7 Ci) of Pu-238, analyzed by RAS</li> <li>Performing chemical or metallurgical operations or maintenance on systems containing <math>\geq 10</math> g of Pu-239 or Pu-240 and <math>\geq 0.04</math> g of Pu-238 (0.6–0.8 Ci) analyzed once by TIMS and RAS and once by RAS</li> </ol>
Annual		<ol style="list-style-type: none"> <li>Other supervisory personnel</li> <li>Persons working with sealed containers of plutonium</li> <li>Other persons with casual encounters with plutonium who regularly work in areas where plutonium is handled</li> </ol>	<ol style="list-style-type: none"> <li>Supervisors, persons working with sealed containers of Pu,</li> <li>Casual encounters with Pu, or</li> <li>Working with prepared counting foils containing <math>&gt; 20</math> mg of Pu-239 or 0.08 mg of Pu-238 (<math>\sim 1.3</math> mCi of either)</li> </ol>	<ol style="list-style-type: none"> <li>Routine work with operations of <math>&lt; 10</math> g (0.6 Ci) of Pu-239 or Pu-242 (0.6–0.8 Ci)</li> <li>Performing maintenance on systems containing <math>\geq 10</math> g of Pu-239 or Pu-240 (0.6–0.8 Ci) analyzed by TIMS and RAS</li> <li>Working with operations of or performing maintenance on systems with <math>&lt; 0.04</math> g of Pu-238</li> <li>Line supervisors of personnel in semiannual categories</li> <li>Transuranic (TRU) glovebox, bag outs, etc.</li> <li>ESH-1 radiological control technicians (RCTs) who frequently enter work areas</li> <li>All personnel with confirmed, measurable intakes of Pu-238 and/or Pu-239</li> </ol>

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<b>Frequency<sup>a</sup></b>	<b>1944 (Kolodney 1946)</b>	<b>1967 (Lawrence 1967)</b>	<b>1973 (Lawrence 1973)</b>	<b>1998 (Inkret et al. 1998<sup>b,c</sup>; LANL 2004a)</b>
Initial (baseline)			All new hires or assigned to work with LANL where other persons are routinely sampled	<ol style="list-style-type: none"> <li>1. If there has been risk of exposure to plutonium or if exposure history is missing or inconclusive</li> <li>2. Entry requirement for unescorted access to some facilities</li> <li>3. Occasional work in plutonium areas but do not handle plutonium</li> </ol>
Termination			<ol style="list-style-type: none"> <li>1. Persons terminating employment who have previously submitted urine samples, or</li> <li>2. Have been working in major plutonium areas, but never sampled</li> </ol>	Workers who submitted routine samples
UPPU Club (see Section 5.1)		Complete bioassay and physical examination at 5-yr intervals. Bioassay continues after termination of employment.	Complete bioassay and physical examination at 5-yr intervals. Bioassay continues after termination of employment.	Complete bioassay and physical examination at 5-yr intervals. Bioassay continues after termination of employment.

a. The frequencies above do not apply to working with sealed sources in TA-15 where bioassay is as needed.  
b. Samples analyzed by TIMS.  
c. Samples analyzed by RAS.

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Table A-16. Routine sampling frequency <sup>241</sup>Am exposures.

Frequency	1998 (Inkret et al. 1998d; LANL 2004) <sup>a</sup>
Quarterly	None
Semiannual (biannual)	1. Performing operations in gloveboxes with ≥0.2 g of Am-241 (0.6 Ci) 2. Performing maintenance on systems containing ≥0.2 g of Am-241 (0.6 Ci)
Annual	1. Performing operations in gloveboxes with <0.2 g of Am-241 (0.6 Ci) 2. Frequent entry or performing maintenance on systems containing <0.2 g of Am-241 (0.6 Ci) 3. Line supervisors of personnel in semiannual categories 4. ESH-1 RCTs who frequently enter work areas 5. All personnel with confirmed, measurable intakes of Am-241
Initial (baseline)	If there has been risk of exposure to Am or if exposure history is missing or inconclusive
Termination	Workers who submitted routine samples

a. Samples analyzed by RAS.

Table A-17. Sampling protocol for tritium (Clark 2005).

Period	Program	Protocol
1950–1970		Biweekly for significant quantities or more often if exposure confirmed, removed from tritium work at 124 μCi/L.
1971–1975		>150 μCi/L – 1/d; 150–75 μCi/L – every 2 wk; 74–24 μCi/L – 1/wk (Healy 1970)
1975–1998		Routine – every 2 wk; or 2 hr after expected exposure then if <1 μCi/L - no more samples, 1–10 μCi/L – 1 wk in next month, 10.1–100 μCi/L – weekly samples, >100 μCi/L – daily samples (Inkret et al. 1998a)
1998–present	Work on regular or intermittent basis with or on systems that have contained 1 Ci in HT and any other form, or 0.1 Ci HTO, or MT, or 0.1 Ci of organic tritium	Every 2 wk

Table A-18. Uranium routine sampling protocol (Clark 2005).

Period	Protocol
Pre-1983	Biweekly samples collected Collected on Fridays, minimum annual
08/1983–06/1993	Spot sample collected on Monday mornings before entering work area
07/1993	Last voiding on Sunday night and second on Monday morning, first voiding
1998	Spot samples every 2 wk/persons performing hands-on work/potential for 100 mrem; machining operations, polishing operations, foundry work, chemistry operations in which >10 g of U, work with oxidized metal, >100 g of bulk powder. <sup>a</sup>

a. Source: Inkret et al. (1998c).

Table A-19. Sampling protocol for accidental plutonium exposures, 1967 (Lawrence 1967).

Severity class	Description	Sampling protocol
PA (prompt action)	Most serious accidents Injection detectable by plutonium wound monitor (~0.01 μg) Chemical burns from plutonium solutions Facial contamination >20,000 cpm Nose swipes >500 dpm	Evening after accident 10 d later 1 mo later Each 14 mo thereafter.



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<b>Severity class</b>	<b>Description</b>	<b>Sampling protocol</b>
DA (delayed action)	No known equipment failure, 10 × MPC for 1 wk or 50 × MPC for 1 d Skin contamination in excess of limits Superficially contaminated cuts that are positive on surface monitoring only. Equipment failure causes exposure for indeterminate period.	1 mo after accident 11 more samples at monthly intervals. No entry is made in "No. of days accident prior to sample" column of PUQFUA.
NRS (nonroutinely sampled)	Small wounds occurring in major plutonium areas (DP-West, Ten Site, CMR Building) Exposures without respirators at 10 × MPC for period less than 1 wk Nose counts >50 dpm Skin contamination >500 cpm/60 cm <sup>2</sup>	1 urine sample collected 1 mo after intake.
Automatic rescheduling	Detect unexpected high exposure and verify its existence Detect contaminated urine sample Previous sample is 1 dpm/sample	Request another sample Persons routinely excreting plutonium are exempt from special sampling but are maintained on routine sampling

Table A-20. Sampling protocol for accidental plutonium exposures, 1973 (Lawrence 1973).

<b>Severity class</b>	<b>Description</b>	<b>Sampling protocol</b>
PA (prompt action)	Most serious accidents Injection detectable by plutonium wound monitor >2,000 pCi (DL 600 pCi Pu-239 and 200 pCi Pu-238) Chemical burns from plutonium solutions Facial contamination >40,000 dpm Nose swipes >500 dpm	Evening after accident 10 d later 1 mo later Each 12 mo thereafter. Fecal samples might be collected
DA (delayed action)	No respiratory protection, 10 × MPC for 1 wk or 50 × MPC for 1 d Skin contamination after decontamination in excess of limits Superficially contaminated cuts that are positive (500 dpm) on surface monitoring only.	Fecal collected on day 2 for inhalation 1 mo after accident fecal and urine 11 more urine samples at monthly intervals. Possible fecal Potential accident entry is made in PUQFUA
NRS (nonroutinely sampled)	Small wounds occurring in major plutonium areas (DP-West, Ten Site, CMR Building) no activity detected. <500 dpm Exposures without respirators at 10 × MPC for a period less than 1 wk Nose counts >50 dpm Skin contamination >1,000 dpm/60 cm <sup>2</sup> Potential accident date is noted in PUQFUA and no sample has ever been submitted or routine is not scheduled for at least 3 mo.	1 urine sample collected 1 mo after intake. Fecal samples for types 2 and 3. Memorandum sent to H-1; potential accident entry in PUQFUA might be required
Automatic rescheduling	Detect unexpected high exposure and verify its existence Detect contaminated urine sample Previous sample is 1 dpm/sample	Request another sample Persons routinely excreting plutonium are exempt from special sampling but are maintained on routine sampling

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Table A-21. Sampling protocol for accidental plutonium exposures, 1998 (Inkret et al. 1998c).

<b>Severity class</b>	<b>Description</b>	<b>Sampling protocol</b>
PA (prompt action)	Most serious accidents Injection detectable by plutonium wound monitor >0.2 nCi Chemical burns from plutonium solutions, skin contamination >500 dpm alpha by 60-cm <sup>2</sup> probe or >0.2 nCi in wound area Facial contamination >10,000 dpm alpha Nasal >100 dpm Airborne >200 DAC-hr without respiratory protection. Skin after decontamination is >1,000 dpm alpha by 60-cm <sup>2</sup> probe As requested	Pu-239, Pu-240 Urine samples days 1, 3, 5 after intake Fecal samples days 1, 3, 5 –optional <i>In vivo</i> chest count days 3, 5 –optional Pu-238 Urine samples on days, 1, 3, 5, 8, 16, 30, 60, 120, 240 after intake Fecal samples on days 1, 3, 5 – optional <i>In vivo</i> chest count on days 3, 5 – optional Urine analyzed by RAS and TIMS
DA (delayed action)	Effective dose equivalent >100 mrem; committed dose equivalent >1 rem any tissue or organ Present in room when CAM alarms Other individuals on PA Positive wound count Chemical burns; after decontamination skin >100 dpm Facial contamination, before decontamination, >1,000 dpm Nasal swipes; 1 nostril >15 dpm, both sum = 35 dpm alpha Average ≥40 DAC-hr, without respiratory Skin contaminate >100 dpm after decontamination. Wounds contaminated with >250 dpm (by alpha probe) Request by manager, group leader, or medical	Typically 1 urine, by RAS and TIMS, if Pu-239, or At direction of ESH-1 personnel.
Chelation therapy	Accident scenario exceeds PA by 20–40 times. Medical procedure. Interpretation of early radiological results under advice of ESH-12 dose assessment.	Collection for duration of chelation therapy, up to 60–100 d after therapy. PA schedule followed by monthly urine samples. <sup>a</sup> Might include blood and fecal. Or as directed.
Follow-up	Any positive routine urine	As directed.

a. Samples taken for up to 100 d after termination of the therapy should not be used in the calculation of intake.

Table A-22. Sampling protocol for accidental <sup>241</sup>Am exposures (Inkret et al. 1998d).

<b>Severity class</b>	<b>Description</b>	<b>Sampling protocol</b>
PA (prompt action)	Most serious accidents Injection detectable by americium wound monitor >0.2 nCi Chemical burns from americium-bearing solutions, skin contamination >500 dpm alpha by 60 cm <sup>2</sup> probe or >0.2 nCi in wound area Facial contamination >10,000 dpm alpha	Urine samples days 1,3,5, 8,16,30,60,120,240 after intake Fecal samples days 1,3,5 – optional <i>In vivo</i> chest count days 3,5 – optional

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Severity class	Description	Sampling protocol
	Nasal >100 dpm Airborne >200 DAC-hr without respiratory protection. Skin after decontamination is >1,000 dpm alpha by 60-cm <sup>2</sup> probe As requested	
DA (delayed action)	Effective dose equivalent >100 mrem; committed dose equivalent >1 rem any tissue or organ Present in room when CAM alarms Other individuals on PA Positive wound count Chemical burns; after decontamination skin >100 dpm Facial contamination, before decontamination, >1,000 dpm Nasal swipes; 1 nostril >15 dpm, both sum = 35 dpm alpha Average ≥40 DAC-hr without respiratory Skin contaminated >100 dpm after decontamination Wounds contaminated with >250 dpm (by alpha probe) Request by manager, group leader, or medical	Typically 1 urine, by RAS At direction of ESH-1 personnel.
Chelation therapy	Accident scenario exceeds PA by 20–40 times. Medical procedure. Interpretation of early radiological results under advice of ESH-12 dose assessment.	Collection for duration of chelation therapy, up to 60–100 days after therapy. PA schedule followed by monthly urine samples. Might include blood and fecal or as directed.

Table A-23. Uranium nonroutine sampling protocol.

Years	Nuclide	Protocol
1998	Uranium	Days 1, 4, 8 after possible incidents [high airborne, high alpha skin contamination with >10,000 dpm or nasal swipes over 100 dpm (summed)](Inkret et al. 1998b)

Table A-24. *In vitro* bioassay (except plutonium and americium) sensitivity. See corresponding sections in the document for references.

Radionuclide	Period	Type <sup>a</sup>	MDA <sup>b</sup>
Plutonium		U/F	See Table 5-7
Americium		U/F	See Table 5-10
H-3	19	U	See Table 5-11
Uranium			See Table 5-14
Fission product	1950–1970	U	50 to 100 dpm/L
Sr-90			No information available <sup>d</sup>
Cs-137	1965–present	U	100 pCi/L
Po-210	1954	U	10 dpm/L
	1955–1960	U	0.1 pCi/L
P-32	1975	U	40 pCi/L

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Radionuclide	Period	Type <sup>a</sup>	MDA <sup>b</sup>
Th-230 as thorium	1958–1963	U	0.01 µg/L
	1963–?–present	U	20 µg/L
Pa-231	1985	U	0.88 dpm/24-hr sample

- a. U = urine; F = fecal.  
b. Source: Gautier (1983).  
c. Currently sent to an offsite laboratory.  
d. Use the reporting level because results were not reported below this level.

Table A-25. *In vivo* bioassay routine frequency (Inkret et al. 1998c).

Description	Frequency	Comments
Baseline	As requested	Unescorted access into radiological controlled areas, radiological buffer areas, or radiological areas. Personnel who occasionally work or visit areas where plutonium is handled but do not handle plutonium.
Pu2	Annually	Routine work or system maintenance with $\geq 0.2$ g pure Am-241. Routine work or system maintenance with $\leq 0.2$ g pure americium or frequent entry where work is performed with any pure Am-241. Routine work or frequent entry in any area with beta/gamma-emitting radionuclides.
Pu1	Biennially	Works with plutonium. Routine work or frequent entry in any area with Pu-239 or -240, including material processing, system maintenance, supervision, or other support. Routine work or frequent entry in any area with Pu-238. Personnel who routinely work with plutonium and perform chemical or metallurgical operations with <10 g of Pu-239 or -242 or 0.04 g of Pu-238. TRU glovebox/fume hood operations. TRU hot jobs. TRU bag outs. Glovebox operations. Fume hood operations. Hot jobs. Bag outs. Maintenance operations on process systems. Radiological control technician duties.
Pu	Semiannually	Special request.
Uranium	Annual	Weekly access to areas where DU is machined or polished, casting and cleaning crucibles outside dry boxes, chemical operations, including purification and recovery. Handling $\geq 100$ g metal or bulk powder outside gloveboxes, or operations with uranium-hexafluoride in uncontained systems.
MFPs	Semiannually	Special request.
MAPs/MFPs	Annual	Works in high contamination areas and/or in airborne radioactivity area.
Th	Annual	Works with thorium.

provide excretion rates and deposition patterns necessary to assess the health risks for other workers appropriately.

The Plutonium Experiment involving plutonium tracers was undertaken between April 1945 and July 1947. This was a joint project between LANL and the Atomic Energy Project of the University of Rochester School of Medicine and Dentistry. None of the subjects was administered the injections of plutonium at Los Alamos. However, the data were collected and analyzed by Wright Langham and

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others at Los Alamos. The subjects of the experiments were males over 45 years of age, an 18-year-old female, an 18-year-old male, and a 5-year-old Australian male child. The child was also injected

Table A-26. Amount of plutonium administered to subjects by intravenous injection<sup>a</sup> and the date of the administration (Langham et al. 1980).

Designation of the subject	Date of administration	Pu injected ( $\mu\text{g}$ ) <sup>b</sup>
HP-1	October 16, 1945	4.6
HP-2	October 23, 1945	5.1
HP-3	November 27, 1945	4.9
HP-4	November 27, 1945	4.9
HP-5	November 30, 1945	5.1
HP-6	February 1, 1946	5.3
HP-7	February 8, 1946	6.3
HP-8	March 9, 1946	6.5
HP-9	April 3, 1946	6.3
HP-10	July 16, 1946	6.1
HP-11	February 20, 1946	6.5
HP-12	April 10, 1945	4.7

a. Plutonium was administered as  $\text{Pu}^{+4}$ -citrate in 0.5 mL of 0.41% sodium citrate  $2\text{H}_2\text{O}$ .

b. Average standard deviation of determination of dose was 3%.

with radiostrontium and radiocerium. These individuals were selected because they suffered from terminal illness and were not likely to live beyond 10 years. Six of the individuals outlived the predictions including one person who died 44 years later at the age of 80 (Hughes 2000). Urine and fecal samples were collected from the living subjects and autopsy samples were collected from three patients who died within 30 days (Clark 2005). Intake results are available for 12 subjects as reported in Langham et al. (1980). [The original paper was published in 1950 but not declassified until 1971.] These intakes are listed in Table A-26. However, it appears that an additional six subjects were also involved. These are designated as Chi-X (University of Chicago) or Cal-X (University of California, San Francisco, where "X" is the subject number for that location (Hughes 2000).

It was determined that deposition was primarily in the trabecular bone, bone marrow, and liver (Clark 2005). The subject designation might be identified in the telephone interview.

Other human experimentation was conducted at Los Alamos in later years. Tritium experiments were conducted in the 1940s and 1950s. These experiments involved exposures of humans to HT, HTO, and HTO inhalation, ingestion, and submersion in vapor. Researchers used themselves and probably family members as subjects (Hughes 2000).

In the late 1950s, Los Alamos biomedical researchers orally administered  $^{125}\text{I}$  and  $^{131}\text{I}$  to a group of 19 subjects. The cohort included eight children between 4 and 10 years old, as well as three adolescents aged 12, 13, and 14. The remainder of the cohort consisted of eight adults whose ages ranged from 26 to 46. All of the subjects lived in Los Alamos and were either employed by the Laboratory or relatives of employees (Hughes 2000). Each subject was administered 0.01  $\mu\text{Ci}$  of iodine orally.

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**OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS**  
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In 1961, research on  $^{137}\text{Cs}$  was conducted with a cohort of four men aged 27, 33, 35, and 37. In the 1960s, four human subjects participated in a study involving  $^{65}\text{Zn}$  with a single oral administration. The subjects were the researchers, a coworker, and a family member of one of the scientists. There were 11 types of experiments involving approximately 130 subjects (Hughes 2000).

## ATTACHMENT B BIOASSAY FOR RADIONUCLIDES OTHER THAN PRIMARY RADIONUCLIDES

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### Fission and Activation Product Analysis

The first gross beta urine count was devised in 1947 (LASL 1947). Urinalysis was used to monitor for intakes of fission products until whole-body counting was fully implemented in 1970. A 1958 procedure lists gross beta urine analysis from  $^{90}\text{Sr}/\text{Y}$ ,  $^{140}\text{Ba}/\text{La}$ ,  $^{144}\text{Ce}/\text{Pr}$ ,  $^{89}\text{Sr}$ , and gross fission products (McClelland 1958). This might be the same as the procedure referenced in 1947. No sensitivity is listed for this procedure. According to the procedure, background was counted before and after each sample. Healy (1970) lists nonspecific sensitivities of 50 to 100 dpm/L with an investigation level of greater than 200 dpm/L. A similar procedure of oxalate coprecipitation and beta counting, effective in 1974 (Gautier 1983), lists a sensitivity of 1 to 2 dps/L and an MDA of 25 pCi/L. This procedure might have been effective as early as 1950. No further mention of a gross fission product procedure has been found. However, a procedure for  $^{137}\text{Cs}$  in urine using gamma spectroscopy lists an effective date of May 1965 and an MDA of 100 pCi/L (Gautier 1983).

Reactors operated from 1944 to 1992. Records of stack releases exist for  $^{131}\text{I}$  and other fission products. Room air concentrations and urine bioassay results have been found in office memoranda (LASL 1950–1960). Fission product air contamination around the Omega Water Boiler was noted as a “special problem” because ordinary filter paper failed to pick up the gas in the air. This air contamination was thought to have occurred because of residual  $^{235}\text{U}$  from an old spill that was lodged in the interstices of the reactor (Shipman et al. 1952).

Some individuals might have been exposed to fission products while off the Laboratory site or during weapons testing. In addition, a potential for exposure to  $^{140}\text{Ba}/\text{La}$  and  $^{90}\text{Sr}/\text{Y}$  exists for those participating in the RaLa chemistry and testing, 1944 through 1963, primarily at TA-10.

Urine bioassay samples were submitted for gross beta and gamma counting. Samples were analyzed by gamma spectroscopy (MDAs for specific geometry and nuclides are unknown.) The sample was then processed through alkaline phosphate precipitation and counted on a gas flow counter with a background of 530 cpm (40% efficiency). This would be an MDA of approximately 120 dpm/sample. Samples were counted at least five times over a 3-week interval to determine the half-life of the nuclides. For example, if the sample exhibited a 12-day half-life,  $^{140}\text{Ba}$  was recorded; for an 8-day half-life,  $^{131}\text{I}$  was recorded. Results had a background of  $^{40}\text{K}$  subtracted (typically 20 cpm/1,500 mL) (LASL 1950–1960).

Gas leaks were detected with filter papers and charcoal, which were analyzed by gamma spectroscopy. Potentially exposed personnel were analyzed in the human spectrometer (HUMCO). Individuals showing elevated background in the HUMCO were counted in the NaI(Tl) counter [9.5- by 6-in. NaI(Tl) crystal]. Iodine-135 (6.57-hour half-life)/ $^{135}\text{Xe}$  (9-hour half-life),  $^{131}\text{I}$ , and  $^{133}\text{I}$  were observed in individuals exposed to gas leaks at reactors (LASL 1950–1960; Van Dilla 1959). Through the late 1940s and as late as 1961, before the use of charcoal cartridges, iodine was analyzed from the paper filter with an assumed collection efficiency of 0.1%. Radioiodine and noble gases are released from facilities that performed fission product chemistry [Wing 9, CMR (TA-3) and TA-48], medical isotope preparation (TA-53), and research reactors (TA-35). There is a very small  $^{131}\text{I}$  release (environmental release of less than 1 mCi/yr) from TA-48 operations, and the Omega West Reactor (OWR) Facility (TA-2) is a source of radioiodine.

Interpretation of the fission and activation product urinalysis in a way that is meaningful, as representative of all the possible fission and activation products to which a worker might theoretically have been exposed, is a challenge. The gross beta procedure separated and counted radionuclides of alkaline earths and rare earths, such as strontium, yttrium, barium, lanthanum, and cerium. The

## ATTACHMENT B BIOASSAY FOR RADIONUCLIDES OTHER THAN PRIMARY RADIONUCLIDES

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procedure did not account for the radionuclides of ruthenium, cesium, zinc, cobalt, manganese, niobium, or zirconium. The abundances of all the fission products, in relation to each other, varied considerably. Certain reactors at LANL operated only briefly. Some exposure might have occurred during decommissioning operations and operations with weapons testing. No discussion is available for the interpretation of fission product mixtures. However, interviews with current and past LANL personnel involved with bioassay indicate that fission products were not considered a significant source term for intake among LANL workers.

During the late 1940s and early 1950s, atmospheric testing of nuclear weapons was still being done. After such tests in Nevada in the 1950s, these radionuclides were found in urine bioassays for gross fission products. The background levels, which were variable, provide a complicating factor for the use of gross fission product analyses for dose reconstruction. No specific guidance is available from LANL on nuclides or amounts to subtract from bioassay samples. Therefore, any activity detected in gross beta urine counts will have to be considered occupational.

The HUMCO whole-body counter was used for screening beginning in the 1950s with follow-up by a 4- by 8-in. NaI(Tl) detector. Body counting and thyroid counting in the current form were not available until 1970. Once whole-body counting was established, fission and activation products were more often evaluated by whole-body counting. If fission and activation product potential exposure is indicated, refer to Section 5.3 for *in vivo* bioassay sensitivities. The MDAs are listed for many of the fission and activation product nuclides (Vasilik and Aikin 1983).

At present, the LABDR does not contain MFP urine bioassay results, and there is no plan to supply results for workers unless specific requests are made for the results. Unless the telephone interview indicates that the worker might have received bioassay for MFPs, there is no way of knowing if these results exist for the worker. After the mid-1950s, when the HUMCO and the NaI(Tl) detectors were first placed in service, *in vivo* bioassay might have been used to assess the potential of intakes. The results of any *in vivo* bioassay performed before 1969 are not readily available (Hoover 2007).

In addition to workers at the reactor areas, assessment of internal dose or potential internal dose for workers involved in exposure to ionizing radiation associated with RaLa operations at TA-10 (Bayo Canyon Site), TA-35 (Ten Site), and Buildings H, Sigma, and U (in TA-1) is not possible due to the lack of urine bioassay monitoring between September 1, 1944, and July 18, 1963, and the unavailability of *in vivo* monitoring until the mid-1950s (NIOSH 2006). These workers, primarily chemists but including other operations and maintenance workers and security personnel, had the potential for exposure to  $^{90}\text{Sr}$ ,  $^{89}\text{Sr}$ ,  $^{140}\text{Ba}$ , and  $^{140}\text{La}$ .

Similarly, sufficient records for radionuclides other than tritium, polonium, plutonium, and uranium have not been found that would allow the reconstruction of internal doses for most TAs with a history of radioactive material use. For this reason, only partial dose reconstructions are possible for LANL workers with employment before January 1, 1976 (see Section 5.1.3).

### Strontium

Records of  $^{90}\text{Sr}$  urinalyses, routine or special, are very sparse. Inkret et al. (1998c) stated that four bioassays were performed for  $^{90}\text{Sr}$  in 1997. It is estimated that approximately 200 bioassays were performed for  $^{90}\text{Sr}$  during the history of the site. The historical compilations of procedures do not list a specific  $^{90}\text{Sr}$  urinalysis procedure. It appears that any record of  $^{90}\text{Sr}$  analysis indicates that LANL performed a gross beta analysis (which was determined to be representative of  $^{90}\text{Sr}$ ) or sent a sample to an outside laboratory. Strontium-90 dose currently can be reconstructed only when  $^{90}\text{Sr}$  results are



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listed for an individual. No information is available on an MDA for  $^{90}\text{Sr}$  analysis. Section 5.3.1 discusses the early HUMCO whole-body counters as being capable of counting the bremsstrahlung from  $^{90}\text{Sr}$ .

Exposure to  $^{90}\text{Sr}$  can be expected for persons working in the RaLa Program because  $^{90}\text{Sr}$  contamination was present in the material used for the program. Persons frequenting the area of the shots and those involved with the extraction chemistry would have had a potential for exposure to and intake of  $^{90}\text{Sr}$ . Persons from TA-1, Sigma, H, and U Buildings, where RaLa operations were conducted, would also have the potential for intakes of  $^{140}\text{Ba}$  and  $^{140}\text{La}$ . The RaLa sources were prepared by Group G-7 or G-6 workers at the TA-10 Chemical Process Building from 1944 to 1950. This function moved to TA-35 (Ten Site) from 1951 to 1963. The name of the site is likely tied to the operating group, CMR-10. The CMR-10 group relocated to Ten Site sometime between April and December 1950. The Idaho Chemical Processing Plant became the source of purified  $^{140}\text{Ba}$  in 1956, and a typical shipment was about 40,000 Ci of  $^{140}\text{Ba}$ . The  $^{140}\text{La}$  sources prepared at Ten Site were usually in the range of 2,000 to 4,000 Ci. Almost 2 million Ci of  $^{140}\text{Ba}$  had been handled at Ten Site by 1963 when the RaLa Program was terminated. The TA-35 RaLa cell and control room have been completely dismantled. Barium-140 and  $^{140}\text{La}$  would have been detectable in the HUMCO screening and quantified in the NaI follow-up, although no MDA is available for those nuclides. The Savannah River Site lists an MDA of 9.3 nCi for that period (ORAUT 2005b). However, it is not possible to reconstruct internal dose or potential internal dose from  $^{90}\text{Sr}$  for workers with potential exposure to ionizing radiation associated with RaLa operations at TA-10 (Bayo Canyon Site), TA-35 (Ten Site), and Buildings H, Sigma, and U (in TA-1).

### Actinium

Notebook 6489, containing  $^{227}\text{Ac}$  *in vitro* bioassay results, has been found. Only 15 analyses are recorded in the notebook. All analyses were performed in 1954. No other records of  $^{227}\text{Ac}$  bioassay have been found. Results were 0 to 0.6 dpm/sample.

Potential worker encounters with  $^{227}\text{Ac}$  occurred during the decommissioning of TA-21-153 in 1978. However, all workers on the decommissioning wore full-face particulate respirators and anticontamination clothing. Workers were regularly monitored with nasal swipes and *in vivo* bioassay during the project. Results of the bioassay and nasal swipes were below detection levels (Harper and Garde 1981).

Shipman et al. (1952) mentioned the determination of  $^{226}\text{Ra}$  as the only practical method "at present" for estimating individual exposures to actinium.

No other information is available. Results for any bioassay that might have been performed for the individual will not be present in the LABDR. These results will be available only by special request.

### Phosphorus-32 and Carbon-14

Both of these radionuclides were encountered as labeled compounds. Standard biokinetics do not apply to labeled compounds. A bioassay procedure for  $^{32}\text{P}$  in urine lists an effective date of August 1975 and an MDA of 40 pCi/L. This is also the MDA listed in a procedure in 1983 (Gautier 1983). No bioassay procedure has been found for  $^{14}\text{C}$ .

Results for any bioassay that might have been performed for the individual will not be present in the LABDR. These results will be available only by special request.

**ATTACHMENT B**  
**BIOASSAY FOR RADIONUCLIDES OTHER THAN PRIMARY RADIONUCLIDES**

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**Thorium**

A procedure for a coprecipitation technique and a colorimetric final determination was listed as effective January 1963. The MDA for this analysis was listed as 20 µg/L. Dummer (1958) listed the MDA for the colorimetric procedure as 0.01 µg/L. While plutonium can coprecipitate with thorium, the colorimetric determination prevented interference from the plutonium. An earlier procedure titled "The Determination of Thorium<sup>230</sup> in Urine" was listed in McClelland (1958). This procedure used low-background alpha proportional counting and lists a sensitivity of 0.05 dpm (no volume units) of <sup>230</sup>Th. The procedure stated that natural thorium, plutonium, some americium, curium, actinium, and neptunium are carried over with the precipitate.

Only the results of the radiometric analysis of 44 urine samples for <sup>230</sup>Th in 1958 have been found thus far. A sensitivity of 0.2 dpm (no volume units) was listed for these analyses. Dummer (1958) suggested the use of alpha proportional counting for <sup>230</sup>Th bioassay samples. Thorium-230 is also referred to as ionium. Results for any bioassay that might have been performed for the individual will not be present in the LABDR. These results are available only by special request.

Shipman et al. (1952) discussed monitoring of thorium progeny in air from the decay of <sup>232</sup>Th. Casting, machining, and other operations with thorium metal were undertaken in 1951. This suggests that <sup>232</sup>Th was in use during this period. Thorium-232 has been identified in environmental emissions from waste sites at LANL. After 1980, some work with thorium-based materials was performed. The thorium was probably <sup>232</sup>Th.

The isotopic mixture should be based on the facility. Conversations with former LANL personnel suggest that most exposure to thorium would be incidental to the exposure to decay products of uranium. However, statements about work with thorium metal would seem to indicate otherwise. Building 159, Sigma Complex, is designated as a Thorium Storage Building for the storage of ingot and oxide forms, which would suggest <sup>232</sup>Th and progeny. Therefore, the dose reconstructor should use best judgment for selection of the isotope or mixture. Thorium-230 is the maximizing conversion from mass units. However, it is unlikely, given the current structure of the database, that results from thorium bioassays will ever be submitted for a worker unless specific requests are made for the results. In that situation, the isotope that was encountered would probably be known.

The Tiger Team Assessment report (DOE 1991) indicated that line managers were not aware that thorium and its decay products were internal radiological hazards and that workers handling gram quantities of dispersible thorium oxide powders and other thorium compounds should be identified by the checklist system for participation in the bioassay program.

Default absorption types for thorium are M and S. The absorption type should be selected based on the compound expected or the matrix (in the case of <sup>230</sup>Th incidental to uranium exposure).

If RAS rather than total alpha results of <sup>232</sup>Th are encountered, the contribution from decay product nuclides should be included. Thorium-232 decays with a 6.7-year half-life to <sup>228</sup>Ra, which decays with a 5.75-year half-life to 6.13-hour <sup>228</sup>Ac, which decays to 1.91-year <sup>228</sup>Th. See Table B-1.

Table B-1. Specific activity of thorium isotopes.

Isotope	Specific activity (pCi/µg)
Th-228	8.1946E+08
Th-230	2.0184E+04
Th-232	1.0966E-01

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**Protactinium-231**

A procedure for a coprecipitation technique and proportional alpha counting is documented in McClelland (1958). The MDA is listed as 0.88 dpm/24-hr sample. No other mention of this analysis is found in later procedure documents or reports.

Results for any bioassay that might have been performed for the individual will not be present in the LABDR. These results are available only by special request.

## ATTACHMENT C RESPIRATORY PROTECTION PROGRAM

### Respiratory Protection Program

The first reference to the need for respiratory protection for LANL workers was in a telex that requested “assault-type gas masks for testing in connection with toxic effects of X10 product [plutonium] dust” (Oppenheimer 1944). The May 31, 1944, *Health Safety Rules* for Buildings H and V-8 specified that “[a]n approved respirator or gas mask must be worn during operations requiring the exposure of the dry material to the air and during wet operations involving spray or splatter” (LASL 1944a). Positive pressure masks were being introduced into the program at that time (LASL 1944a). Dry boxes and respirators were a satisfactory method of handling dry material; however, individuals who worked with solutions consistently received the highest exposure despite all precautions.

Respiratory protective devices were provided during the Manhattan Project by the U.S. Army Chemical Warfare Laboratories. Use of the M-9 mask with either the M-11 canister for gases and particulates or the M-14 canister for particulates began in 1946. The efficiency of the canister was 1 in 100,000 particles of dioctyl phthalate (Adley et al. 1963). This mask continued to be used as a standard until commercially produced masks were available. By the time the handbook on radiation protection (Dummer 1958) was published, a variety of U.S. Bureau of Mines-approved commercial respirators and self-contained breathing apparatus was available. Approved respirators were used where possible. The Industrial Hygiene group maintained facilities for testing respiratory protection equipment and could make recommendations on equipment. The use of Wilson-type respirators was discussed in 1973 (Hempelmann, Richmond, and Voelz 1973).

The Industrial Hygiene group at LANL continued to be at the forefront of development of respiratory devices and programs. Respiratory protection programs are documented beginning with Dummer (1958). The programs included training in the use of and fit testing for air-purifying respirators and self-contained breathing apparatus. Protection factors assumed for respiratory protection equipment are listed in Table C-1. The protection factors appear to remain consistent throughout the history of the program within the exceptions noted.

Table C-1. Respiratory protection apparatus.

Equipment	Protection factor	Remarks
Half-mask	Not more than 10 x MPC <sup>a</sup>	b, c
Full-face mask	Not more than 50 x MPC (Dummer 1958) <sup>d</sup>	b
Full-face mask	Not more than 100 x, MPC (Healy 1970; LANL 1984) <sup>e</sup>	b
Full-face mask with fit field tested	Not more than 1,000 x MPC (Healy 1970)	b
Full-face mask with chemical canister	Used for organic gases and vapors up to 2%	b
Air-line equipment	Used above limits of air purifying respirators; no specific protection factors listed	
Self-contained breathing apparatus	Used above limits of air purifying respirators; no specific protection factors listed	

- a. Assumed 20% penetration because of fitting limitations and 50% workday usage.
- b. Air purifying respirators shall never be used in an atmosphere immediately dangerous to life and health.
- c. Half-face respirators were not permitted to be used in airborne highly toxic dust such as plutonium (Shipman 1964). LANL (2004) stated that half-face respirators are not approved for use with radiological materials or airborne radioactivity.
- d. Only one style was available at that time; masks were assumed to be only 98% to 99% efficient.
- e. Powered air-purifying respirators not more than 3,000 times with high-efficiency particulate air filters.

Respirator mask fit and proper usage remained a significant challenge throughout the development of the program. In addition, the decision to don a respirator is often based on the alarm of a continuous air monitor in the area.

**ATTACHMENT D**  
**DESCRIPTIONS OF SOME INCIDENTS RESULTING IN INTERNAL DOSES**

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**Maximizing and Best Estimate Intake Parameters**

Intake parameters can be derived from airborne contamination levels for buildings with the highest exposure potentials or highest intakes for various periods. Incidents and intakes are listed here in Table D-1.

Table D-1. Reported exposure incidents and results.

Year	Incident
1944–1946	26 workers received intakes estimated to be 6 to 80 nCi while involved in plutonium operations: purification, fluorination, metal reduction, and recovery. Average intake = 58 nCi (Voelz et al. 1979); 9 of 12 persons with highest exposures were working with water-soluble plutonium salts in 1945 (Hempelmann and Langham 1953).
1944	Periodic overexposure of individuals working on RaLa Project occurred for 6 mo at start of project until bugs were worked out of remote chemical handling procedures. Highest nasal swipe for May was 11,372 cpm (assume alpha). August – Worker opened a sealed container of active material without a respirator or face shield.
1945	August – Building 52 handled large amounts of polonium. Note at bottom of 52 air concentration data table stated: “0.75 cpm/L is used as 2-year tolerance value assuming 100% retention by lungs.” 4 persons exceeded safe amount of plutonium in body, 1 µg (Hempelmann 1946). Tolerance value for polonium in urine samples 1,500 cpm/d (assume 50% efficiency for counter), exceeded by only 2 individuals. All persons working with polonium were monitored (Hempelmann 1946). At the Water Boiler at Omega, several instances of mild to moderate overexposure when gas exhaust lines leaked or during decontamination of active material. Summer – 14 workers removed from plutonium production because urine assays indicated body burdens at or above the MPL of 7 cpm/24-hr sample (Hempelmann, Richmond, and Voelz 1973). Workers entered contaminated side of the air filters areas at DP East without approval (Cox 1945).
1946	April 15 – Tuballoy fire, Tech Area, firemen exposed without protective equipment (Tribby 1946b).
1948	November 2 – worker from GMX ingested sufficient amount of RaLa to give a reading on detector (LASL 1949a). Radioactive specks found near Bayo Canyon ranged from 0.1 to 10 µCi of RaLa at time of origin (LASL 1948).
1949	Difficulties with RaLa separations and extremely large quantities of material being worked with in Bayo Chemistry Area cause spread of contamination and few slight overexposures (LASL 1949b).
1949–1951	Number of airborne contamination results above plutonium MAC (0.0044 cpm/L) decreased from 40% of samples in 1949 to 7% in 1951 for DP West and 25% to 17% for Tech Area. Above-MPL nasal swipes decreased from 40% in 1949 to 15% in 1951 for DP West and 60% to 15% for Tech Area (Shipman et al. 1952).
1951	1,876 plutonium urine samples below MAC and 492 polonium urine samples from DP East were below tolerance. All routine tritium urine samples were below MAC (Shipman et al. 1952). DP West dismantle 408 and 413; high airborne plutonium exposures were kept to minimum (Shipman et al. 1952). DP East experimental program with actinium. No overexposure; however, adequate urine analysis is not available, air samples difficult to evaluate (Shipman et al. 1952). Pajarito Warehouse contaminated by leaking polonium source. TA-33, Area 6 required cleanup from polonium spill (Shipman et al. 1952).
	Ruptured plutonium slug at GT site vault. Contamination spread to main building. Air tolerance limits reached (Shipman et al. 1952).

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DESCRIPTIONS OF SOME INCIDENTS RESULTING IN INTERNAL DOSES**

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Year	Incident
	Airborne alpha activity in Room 513, DP West was consistently above MAC. Source could not be localized; however, cleaning attic and sealing openings to room decreased airborne levels to satisfactory (Shipman et al. 1952). MAC total alpha plutonium as of April 1951 was 0.0044 cpm/L. Kilocurie quantities of tritium handled in CMR areas. "Health program for this work has been satisfactory" (Shipman et al. 1952).
Late 1950s	Radiiodine Experiments: LANL biomedical researchers orally administered I-125 and I-131 to 19 subjects.
1955	A vial containing americium and curium (91%) exploded, contaminating 2 workers on the face and clothing (LASL 1955).
1956	Some high urine counts in uranium areas for employees working with incinerators without respirators: Sigma Building, TU Building, Tuballoy Shop, M Building, and Building 96 (McKown 1957).
1957	TA-18 air samples in vicinity of Honeycomb assembly were one-half of tolerance level, and in vicinity of Lady Godiva assembly air samples were "excessively high." February 12 – Godiva reactor went supercritical and contaminated the kiva (Buckland 1958). May 17 – Spill of Sr-90 within H-7 undetected. Contamination spread to homes (Buckland 1958). June 13 – Spill of Dy-165 resulted in airborne and personnel contamination (Buckland 1959). Monitoring of individual for urine, blood, and human counter showed only positive for the human counter. Suspected the human counter to have surface contamination. Worker's badge read 2 rep (Buckland 1958). Dismantling tritium equipment in TA-33 resulted in the contamination of three workers, urine concentrations up to 650 µCi/L (Buckland 1958)
1958	May 9 – Spill of Dy-165 resulted in airborne and personnel contamination, worker had 20 mR/hr at nostril (Buckland 1959). Total body counts performed on cyclotron personnel indicated measurable Zn-65 (LASL 1959).
1959	April – Radioactive gas leak at LAPRE II resulted in thyroid uptakes by at least five workers of I-131, I-133, and I-135 resulting in thyroid burdens equivalent to approximately 3 µCi I-131 (Van Dilla 1959). Laboratory air sample 3.8 dpm, 16% of MPC for 1-wk sample, highest value February to July (LASL 1950–1960). August – Three employees >MPL, 11 >50% MPL, 90% of people with ≤10% MPL for plutonium (0.04 µCi) of 1,325 monitored (LASL 1959)
1960	January 11 – Tritium exposure, 476 µCi/L-urine sample (Buckland 1961). March 2 – A power outage disabled exhaust ventilation resulting in the contamination of four laboratories (Buckland 1961). March 15 – Wind carried plutonium contamination from an assembly that was being identified to a member of H-3. Gross contamination and an intake of plutonium resulted. Contamination spread to the Administration building before being detected (Buckland 1961). December 1 – Activated Na-24 in glass wool contamination (Buckland 1961).
1971	Wing 9 of CMR – Inhalation of Pu-238, minimal urine excretion for 100 days, then rose to large values. ICRP 30 model modified to time constant of 10,000 days (ICRP 1979; Miller et al. 1999); half-time for ICRP 30 model is 10,000 days, AMAD is 0.2 µm. Intakes 2,150 to 210 nCi, found to fit other Wing 9 intakes.
1972–1973	Demolition of Building 12, DP Mesa, produced no bioassay above detectable and only four detectable nose swipes. Highest nose swipe was 85 dpm (Christensen, Garde, and Valentine 1975)
1977	January – Oxide of plutonium caught fire, airborne 4.5 to 11.65 dpm/m <sup>3</sup> , no positive nasal swipes, no bioassay (University of California 1977b).

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DESCRIPTIONS OF SOME INCIDENTS RESULTING IN INTERNAL DOSES**

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Year	Incident
	January – H-3 urine bioassay after January 1 incident at LAMPF <1 µCi/L (University of California 1977a).
	February – One employee received plutonium intake estimated at 1 MPBB (0.04 µCi) (University of California 1977a,d).
	February 10 – Incident resulted in <16-nCi lung burdens for five employees; one employee received diethylene triamine pentaacetic acid (DTPA) chelation therapy. Urine sample plutonium intake estimates are 1 nCi (one worker), 4 nCi (two workers), 9 nCi (one worker).
	May – U-235 airborne concentrations of >1,500 dpm/m <sup>3</sup> and >25,000 dpm/m <sup>3</sup> for DP Bldg 4, Room 412 (University of California 1977b).
	May – TA-35 and TA-41 alpha contamination escaped from pressure vessel, caused personnel contamination (University of California 1977d).
	June – Alpha contamination (up to 100,000 cpm) from damaged uranium target found on several persons at LAMPF switchyard (University of California 1977d).
	June – Tritium in urine 101 µCi/L after airborne release DP, Bldg 5. 20,000 dpm alpha airborne (U-235 and U-238 operations).
	November – Tritium bioassay (confirmed) of [redacted] employee 798 µCi/L (University of California 1977c).
1978	UF <sub>6</sub> tube ruptured (5-10 g) spreading HF fumes and depleted U-238 (University of California 1978a).
	[Title redacted] replacing leaking hose at TA-33 stack had tritium bioassay of 4 µCi/L; 449 Ci HT released (University of California 1978b).
1979	[Redacted] employee with nasal swipe of 553/70 dpm TA-21 (University of California 1980a).
1980	TA-33 tritium exposure to employees.
	54-µCi/L tritium bioassay TA-33 (University of California 1980b).
	153-µCi/L tritium bioassay TA-33 (University of California 1980c).
1983	Plutonium spill in TA-55, 10 persons, no significant exposures (LANL 1983a).
	High airborne contamination results in high nasal swipes TA-55 plutonium (LANL 1983b).
	21 plutonium wound counts, 3 = 0.1 nCi, 1 had 11.2 nCi Pu-239 and 0.4 nCi Am-241 and was surgically excised (LANL 1983c).
	Quarter 4 – One wound above 20 nCi, numerous positive (>50 dpm) nasal wipes (LANL 1983b).

Particulate filtering respirators were available and were used from the beginning of the program. Therefore, ambient air concentrations might not reflect the actual breathing air concentration of the workers.

Research continues to identify the intakes for both maximizing and general conditions. Examples of intakes identified thus far are listed in Table D-1; Table D-2 summarizes a particular incident. As the state of the respiratory protection program and engineering controls improved, the potential for chronic and acute intakes was reduced.

NOTE: While an attempt has been made to report only incidents with quantitative results, some incidents for which only qualitative comments were available in the records have been included in the list. Information is not available for every year of operation.

**ATTACHMENT D**  
**DESCRIPTIONS OF SOME INCIDENTS RESULTING IN INTERNAL DOSES**

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Table D-2. Summary of February 10, 1977, plutonium incident (University of California 1977d).

Worker	Total systemic burden (increase)		Nasal swipe (left/right) (dpm)	Chest burden <sup>a</sup>
	Pu-239 nCi	Pu-238 nCi	Total alpha	Pu nCi
[Redacted] 1	8.2 (3.3)	1.7 (0.1)	12,759/13,385 <sup>b</sup>	7.5
[Redacted] 2	7.6 (5.8)	0.7 (0.5)	801/1,849	13.
[Redacted] 2	42.9 (7.5)	3.2 (0.6)	1,091/1,193	6.
[Redacted] 3	21.8 (18.0)	1.1 (0.4)	611/1,193	10.
[Redacted]	4.2 (0.3)	0.7 (0.0)	226/577	0

a. No worker had chest counts above MDA (16 nCi).

b. Received DTPA chelation therapy.

Rather than preventing intakes completely, the focus of the respiratory protection program, especially in the early years of operation, was to not exceed allowable concentration limits. The MPBB for plutonium was 5 µg from 1943 to 1945. During 1945, when many of the exposures occurred, the tolerance dose of plutonium was assumed to be 1 µg. Therefore, a suitable method of measuring personnel exposure had to be able to detect from 2 to 14 dpm of plutonium in a 24-hour sample of urine.

The MACs for buildings with a high potential for exposure were established by 1947 and changed over the years as technology and understanding of the hazards increased. MACs were eventually replaced by MPCs and derived air concentrations (DACs) as controlling values. It appears that at some time before 1964, the MAC values were changed to ALs with larger, more specific MAC values established for nuclides, enrichments, and mixtures.



## ATTACHMENT E SCALING OF COWORKER DOSE INTAKES CONSISTENT WITH TYPE AND DURATION OF EXPOSURES

### Scaling of Assigned Intakes Using Case-Specific Information

The major presumptive exposures at LANL were made up of the primary radionuclides plutonium, uranium, tritium and, in early days, polonium. Other radionuclides may have resulted in intakes to personnel, but due to the small amounts of material, the limited operations, and the use of engineering controls such as dry boxes, potential exposures are relatively unlikely. NIOSH has determined that it is not feasible to reconstruct doses for these radionuclides before the year 1996. Work locations are another important consideration when evaluating for a potential unmonitored intake. In this attachment, a method is presented to scale assigned intakes based on job category and likely duration of exposure to limit the margin of overestimation.

### Level of Contact

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

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

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

### Dose Assignment

Fiftieth-percentile intakes are used to calculate organ doses based on the listed lognormal distribution of intakes, along with the listed GSD. Ninety-fifth-percentile intakes are assumed to be constant values (distribution-free). The actual intake values are calculated using the relationship below:

$$95th\text{-percentile intake} = 50th\text{-percentile intake} \times GSD^{1.645} \quad (E-1)$$

**ATTACHMENT F**  
**BOUNDING ESTIMATES OF ORGAN DOSES FROM POTENTIAL INHALATIONS OF**  
**RADIOACTIVE MATERIALS DURING THE CERRO GRANDE FIRE AT LOS ALAMOS**  
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**F.1 PURPOSE**

This attachment presents bounding estimates of potential organ doses for exposure to airborne radioactive materials on the Los Alamos site during the Cerro Grande fire in May 2000. Measurements of airborne radioactivity were conducted throughout the event as described in *Measurements of Air Contaminants during the Cerro Grande Fire at Los Alamos National Laboratory* (Eberhart 2010). Low values for measured concentrations (in attocuries per cubic meter) resulted in small intakes and low doses.

**F.2 INHALATION EXPOSURES**

Airborne concentrations of radioactive material from the fire are described in Eberhart (2010). The most abundant radioactive materials resulted from resuspension of radon progeny that had accumulated for many years on vegetation and on the forest floor. Concentrations of plutonium, americium, and uranium were also measured, but these results were, in general, consistent with measurements that were made outside the fire period (Eberhart 2010). Gamma spectroscopy identified only naturally occurring radionuclides. For the purpose of an overestimate of potential internal dose to Los Alamos workers, intakes for plutonium, americium, and uranium were calculated based on the highest measured values without considering likely background intakes. That is, material of natural origin was not rejected for the purpose of the maximum dose calculation described here.

**F.3 METHODOLOGY**

Intakes were estimated using the maximum measured concentrations of <sup>241</sup>Am, <sup>238</sup>Pu, <sup>239</sup>Pu, <sup>234</sup>U, and <sup>238</sup>U as follows:

$$Total\ intake = Concentration \left( \frac{dpm}{m^3} \right) \times Inhalation\ rate \left( \frac{m^3}{hr} \right) \times Exposure\ time\ (hr) \quad (F-1)$$

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### F.3.1 Breathing Rate

Standard breathing rate assumptions could underestimate actual rates for emergency workers who responded to the fire. For this reason, the breathing rate was adapted from ICRP (1975), Table 120 as follows:

$$V_m = f \times V_T \times \text{time (min)} \quad (\text{F-2})$$

where:

$V_m$  = volume of air per minute

$f$  = frequency of inhalation breaths per minute (1/min)

$V_T$  = tidal volume, which varies with activity level

In this case,  $f$  and  $V_T$  were selected for an adult performing heavy work (ICRP 1975, Table 120), with the following result:

$$V_h = 21 \left( \frac{1}{\text{min}} \right) \times 2,030 (\text{mL}) \times 60 \left( \frac{\text{min}}{\text{hr}} \right) = 2.56\text{E} + 06 \left( \frac{\text{mL}}{\text{hr}} \right) = 2.6 \left( \frac{\text{m}^3}{\text{hr}} \right) \quad (\text{F-3})$$

where:

$V_h$  = volume of air per hour

### F.3.2 Assumptions

Additional assumptions were:

- A bounding intake was used to overestimate worker internal dose consequences,
- A 60-hour exposure period was assumed,
- Individuals were assumed to spend the entire 60-hour period at the location of the highest potential intake,
- The highest measured values for each alpha emitter were identified from the data in Eberhart (2010) (these values are listed in Table F-1),
- The heavy work breathing rate was assumed for the entire 60-hour period, and
- Respiratory protection was not worn.

## F.4 INTAKES

Intakes calculated from the methodology described above are listed in Table F-1.

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Table F-1. Highest measured air concentrations of alpha emitters and consequent intakes.

Radionuclide	Date	Location	Concentration		Intake (dpm)	Concentration Source
			(aCi/m <sup>3</sup> )	(dpm/m <sup>3</sup> )		
Am-241	May 12	TA-54	125	2.78 E-04	0.043	Eberhart 2010, Table A-16
Pu-238	May 13	McDonald's	65	1.44 E-04	0.023	Eberhart 2010, Table A-17
Pu-239	May 13	Los Alamos Inn	472	1.05 E-03	0.17	Eberhart 2010, Table A-18
U-234	May 13	TA-5 (formerly TA-52, Beta Site)	1,709	3.79 E-03	0.59	Eberhart 2010, Table A-19
U-238	May 13	TA-5 (formerly TA-52, Beta Site)	4,188	9.30 E-03	1.45	Eberhart 2010, Table A-21

## F.5 RESULTS AND RECOMMENDATION

Organ doses that would result from the listed intakes are likely to bound doses due to conservative assumptions for location, exposure time, and breathing rate. Doses were estimated using IMBA and assuming an acute inhalation of each of the listed intakes, over a number of organs, as well as a cancer diagnosis in May 2010. For radionuclides with multiple absorption types, the type that resulted in the highest dose was selected.

Calculated doses to a several organs are listed by radionuclide in Table F-2. The highest total dose from a single radionuclide to any organ was  $2.92 \times 10^{-04}$  rem (0.29 mrem) to the thoracic lymph nodes (LN(TH)) for an assumed absorption type S intake of <sup>238</sup>U. Doses to all other organs, including the lung, thoracic lymph nodes, and red bone marrow, were less than this value. Two isotopes of uranium were measured, and both are present in the background and from anthropogenic sources.

Doses were totaled for each organ over the five radionuclides as shown in Table F-3. The largest total dose was  $4.75 \times 10^{-04}$  rem (0.48 mrem).

Because the dose results listed above are below the level regarded as significant for the calculation of probability of causation, 0.001 rem (1 mrem), and because dose reconstruction practice includes the assignment of a larger dose based on assumptions for environmental internal dose that are favorable to claimants, it is unnecessary to include dose estimated for exposure during the Cerro Grande fire.

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Table F-2. Calculated doses by radionuclide and organ.<sup>a</sup>

Radionuclide	Organ	Dose (rem) (absorption type)	Radionuclide	Organ	Dose (rem) (absorption type)
Pu-238	Lung	2.38 E-06 (S)	Pu-239	Lung	1.53 E-05 (S)
	RBM	8.09 E-07 (M)		RBM	5.80 E-06 (M)
	LN(TH)	5.79 E-06 (S)		LN(TH)	4.19 E-05 (S)
	Bone Surface	<i>1.05 E-05 (M)</i>		Bone Surface	<i>7.54 E-05 (M)</i>
	Liver	2.21 E-06 (M)		Liver	1.60 E-05 (M)
	ET2	3.09 E-06 (S)		ET2	2.25 E-05 (S)
U-234	Lung	4.51 E-05 (S)	Am-241	Lung	2.49 E-06 (M)
	RBM	8.00 E-07 (F)		RBM	1.33 E-06 (M)
	LN(TH)	<i>1.34 E-04 (S)</i>		LN(TH)	3.61 E-07 (M)
	Bone Surface	.11 E-06 (F)		Bone Surface	<i>2.18 E-05 (M)</i>
	Liver	8.43 E-07 (F)		Liver	2.87 E-06 (M)
	ET2	7.35 E-05 (S)		ET2	1.01 E-06 (M)
U-238	Lung	8.68 E-05 (S)			
	RBM	1.82 E-06 (F)			
	LN(TH)	<i>2.92 E-04 (S)</i>			
	Bone Surface	1.55 E-05 (F)			
	Liver	1.84 E-06 (F)			
	ET2	1.56 E-04 (S)			

a. Italics indicate the highest single-radionuclide dose to any organ.

Table F-3. Total organ doses for all evaluated radionuclides.

Organ	Total dose (rem)
Lung	1.52 E-04
RBM	1.06 E-05
<i>LN(TH)</i>	<i>4.75 E-04<sup>a</sup></i>
Bone Surface	1.30 E-04
Liver	2.37 E-05
ET2	2.56 E-04

a. Highest total dose to any organ from all five radionuclides.