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## ACRONYMS AND ABBREVIATIONS

| | |
|---|---|
| 25 | LASL reference to $^{235}U$ or the standard ~93% enrichment |
| 28 | LASL reference to $^{238}U$ or depleted uranium |
| 49 | LASL reference to plutonium-239 |
| AL | (Los Alamos) Action Level |
| ALI | Annual Limit on Intake (ICRP 30); Annual Limit of Intake (ICRP 60) |
| AMAD | Activity Median Aerodynamic Diameter |
| Bq | Becquerel = 1 disintegration per second |
| BZA | Breathing Zone Air Sampler |
| CAM | Continuous Air Monitor |
| CDE | Committed Dose Equivalent |
| cpm | cpm – counts per minute |
| CMR | Chemistry and Metallurgy Research |
| D-38 | depleted uranium mixture |
| DAC | Derived Air Concentration |
| DIL | Derived Investigation Level |
| DNAA | delayed neutron activation analysis |
| DOE | U.S. Department of Energy |
| DOP | dioctyl phthalate |
| dpm | disintegrations per minute |
| $dpm/m^3$ | disintegrations per minute/ cubic meter |
| DTPA | diethylenetriaminepentaacetic acid |
| DU | depleted uranium (less $^{235}U$ than natural uranium, nominally 0.72%) |
| EDE | Effective Dose Equivalent |
| ESH | Environmental, Safety, and Health Division |
| EU | enriched uranium (more $^{235}U$ than natural uranium, nominally 0.72%) |
| GeLi | Germanium Lithium drifted semiconductor detector used for gamma spectroscopy. |
| GM | Geiger-Mueller detector |
| HPGe | Hyper Pure Germanium Detector |
| HRL | Health Research Laboratory |
| HSE | Health, Safety, and Environment Division |
| HT | elemental tritium (tritiated gas) |
| HTO | tritium oxide (water or water vapor) |
| HUMCO | “Human Counter”, whole-body counter used between1955-1970 |
| IA | Induced activity |
| ICRP | International Commission on Radiological Protection |
| IDLH | immediately dangerous to life and health |
| IMBA | Integrated Modules for Bioassay Analysis (computer program) |
| ionium | $^{230}Th$ –the decay product of $^{234}U$ |
| keV | kilo electron volt |
| Kr | krypton |

| | |
|---|---|
| kW | kilowatt |
| | |
| $L_c$ | Critical Level |
| LAMPF | Los Alamos Meson Physics Facility |
| LASL | Los Alamos Scientific Laboratory (until January 1981) |
| LANL | Los Alamos National Laboratory (1981 to present) |
| LSC | liquid scintillation counter used for tritium urinalysis starting in 1970 |
| | |
| MAC | Maximum Allowable Concentration |
| MDA | Material Disposal Area |
| MDA | Minimum Detectable Activity |
| MDTA | Minimum Detectable True Activity |
| MeV | million (mega) electron volts |
| MPBB | maximum permissible body burden: limit used from startup until adoption of ICRP 30 methods, approximately 1990 |
| MPC | Maximum Permissible Concentration |
| MPL | Maximum Permissible Level |
| mR | milli (1/1000) R |
| mrem | milli (1/1000) rem |
| MSMA | Minimum Significant Measured Activity |
| MT | metal tritide (tritium bound to metallic compounds, such as hafnium) |
| MW | megawatt |
| | |
| NaI | Sodium iodide gamma/ photon scintillation detector, a.k.a. NaI(Tl) |
| nCi | nanocurie |
| NCRP | National Council on Radiation Protection and Measurements |
| NDA | No Detectable Activity |
| NIOSH | National Institute for Occupational Safety and Health |
| NTA | Neutron Track Analysis |
| | |
| OBT | organically bound tritium |
| OR-93 | Oralloy, enriched uranium (Oak Ridge Alloy); either 40% or 93% enrichment |
| OU | Operable Unit |
| OWR | Omega West Reactor |
| | |
| PF | Plutonium Facility |
| PHA | Pulse Height Analysis, used in gamma and alpha spectroscopy analysis |
| | |
| R | roentgen (unit of exposure) |
| rad | unit of absorbed dose |
| RaLa | Radioactive Lanthanum Project (radioactive barium and lanthanum implosion tests) 1944 through 1962 |
| RAS | radiometric alpha spectroscopy, a.k.a. pulse height analysis (PHA) |
| rep | roentgen equivalent, physical (former unit of absorbed dose) |
| rem | roentgen equivalent, man (dose equivalent) |
| | |
| Sv | sievert; the SI unit for dose equivalent. (1 Sv = 100 rem.) |
| | |
| $T_3O_8$ | uranium oxide based on tuballoy (natural uranium) |
| TA | Technical Area |
| TIMS | Thermal Ionization Mass Spectroscopy |

| | |
|---|---|
| U.S.C. | United States Code |
| $U_{nat}$ | Natural uranium |
| VFP | volatile fission products |
| WGPu | Weapons Grade plutonium |
| X10 | LANL reference to plutonium |
| ZnS | zinc sulfide scintillation detector for alpha counting |
| µCi | microcurie |
| µg | microgram |
| α | alpha radiation |
| β | beta radiation |
| γ | gamma radiation |

## 5.0 OCCUPATIONAL INTERNAL DOSE

Technical Basis Documents and Site Profile Documents are general working documents that provide guidance concerning 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 the National Institute for Occupational Safety and Health (NIOSH) 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 facility" as defined in the Energy Employee Occupational Illness Compensation Program Act of 2000 (42 U.S.C. § 7384l (5) and (12)).

Occupational internal dose is the dose received by an individual from an intake of radioactive material while performing tasks within buildings and structures of the Los Alamos National Laboratory (LANL), which was called the Los Alamos Scientific Laboratory (LASL) until January 1981, or from activities outside buildings, such as burial of waste and monitoring of tests, where intakes of radioactive material could occur. This document contains information for reconstruction of occupational internal doses at LANL facilities throughout its history.

## 5.1 INTRODUCTION

When operations began at LASL in 1943, the only method of monitoring intake was through loose contamination swipes. Swipes with a lightly oiled filter paper were made 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 more 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.

Radiation hazards were minimal during 1943. In the spring of 1944, the first few milligrams of plutonium arrived at LANL. Until that time, the laboratory was in the construction phase. In 1943, hazards of the project were limited to external radiation from the cyclotron, the Van de Graaff, radium sources, and a few micrograms of plutonium that arrived in the summer of 1943. In addition, there were some 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, showering 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 developing 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 burdens were reduced significantly. Maximum permissible body burdens (MPBBs) began

at 5 µg (0.33 µCi) of plutonium in 1943. These were reduced to 1 µg (0.07 µCi) in October 1944 and reduced again to 0.6 µg (0.04 µCi) in 1951 (Langham et al. 1962).

Early safety efforts were based on working in safe contamination levels. In 1954, contamination-level measurements were based on contact with a shield open (Geiger-Mueller) GM tube for beta/gamma and a Pee Wee probe of 55-$cm^2$ area for alpha. The efficiency of these portable probes is approximately 10% to 15%. Swipes counted on a fixed proportional counter had an efficiency of approximately 40% to 50%, although the oil on the swipes, used to increase 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).

The tolerance for wounds was 10 cpm for alpha and 0.15 mrep/hr beta/gamma, except wounds contaminated with $^{90}Sr$ for which the tolerance was 0.05 mrep/hr. The skin contamination tolerance was 1 mrep/hr beta/gamma, except $^{90}Sr$, which was 0.05 mrep/hr. The skin contamination tolerance for alpha emitters was 1,000 cpm for polonium, 500 cpm for Tuballoy or Oralloy, and 250 cpm for plutonium. The tolerance for floor counts was 100 cpm/swipe alpha but definitely less than 500 cpm/swipe alpha, and bench tops were usually 50 cpm/swipe alpha or less. Floors in laboratories were mopped once or twice a day to maintain safe contamination levels (Hempelmann, Richmond, and Voelz 1973). Early Health Group reports indicate contamination inside many of the respirators indicating improper storage and handling and poor fit during use (LASL 1944a).

Kilogram quantities of plutonium began to arrive at LASL 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 online, some level of chronic or episodic intake during this period would be a reasonable assumption.

From 1943, the Health Group was responsible for establishing 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. 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, Environmental, Safety, and Health (ESH) and Health, Safety, and Environment (HSE) Divisions], with multiple groups therein 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, as well as performed physical examinations and treated industrial accidents.

Over the years, many improvements have been made in monitoring, bioassay techniques, safety equipment, and safety procedures. 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 ($^{3}H$)
- Uranium ($^{238}U$, $^{234}U$, $^{235}U$)
- Plutonium ($^{238}Pu$, $^{239}Pu$, $^{240}Pu$, $^{241}Pu$ and, to a lesser exposure significance, $^{242}Pu$ and $^{244}Pu$)
- Polonium ($^{210}Po$)
- Americium ($^{241}Am$)

These radionuclides of primary concern are listed in Laboratory reports from 1943 through 2004. They have been associated with work areas and years of operation in Attachment 5A, Table 5A-10. Information applicable to both monitored workers (e.g., units, tolerance, reporting levels, and frequencies) and unmonitored workers (e.g., establishing maximizing and plausible intakes) is also listed in Attachment 5A.

Work areas include plutonium facilities ($^{238}Pu$ or $^{239}Pu$), uranium facilities, polonium facilities, tritium facilities, laboratory facilities, reactors, accelerators, and others. Many of the exposure histories and work records are not specific about the assigned work areas of individuals. However, when information concerning the work location is available, Table 5A-10 in Attachment 5A can be used to determine the probable nuclide(s) and inhalation absorption type. Section 5.2 discusses *in vitro* methods for specific radionuclides. Excreta bioassay methods for determining internal exposures were developed in late 1944 for plutonium (fully implemented in April 1945) and polonium, in 1949 for uranium, and in 1950 for tritium. Only workers with a significant potential for exposure were monitored. Although the number of individuals monitored has increased, not all individuals working at LANL are currently monitored. A survey taken in 1986 estimated that approximately 350 persons had known burdens of plutonium.

Blood tests were performed following potential exposures starting in 1944. However, these blood tests were performed for blood count levels related to external radiation exposure rather than the radioactive content of the blood. Therefore, any records of blood counts performed on an individual or mentioned in the claimant interviews will not be directly applicable to internal dose calculations. Air samples, identifiable with an individual's record, were performed beginning in 1944. However, it is not apparent how the records of these samples might be available in the individual's dose records. Records of chelation therapy for plutonium and americium are noted in the bioassay record.

As the state of the art of radiation detection progressed, whole-body counting for fission products was begun in 1955, chest counting was begun in 1970, and wound counting was performed beginning before 1967. Section 5.3 discusses *in vivo* bioassay methods used currently and historically at LANL.

### 5.1.1 Bioassay Results of Individuals

At the time of this technical basis document, LANL had submitted bioassay data for only a few individuals. Only summary dose reports have been submitted for all claims. The submitted bioassay results were for plutonium only and in an interim format excerpted from the current database. The format of these results is not in the final format expected for the data. All descriptive information provided in this document is based on speculation of the final format of the data that will eventually be supplied. The descriptions listed in Attachment 5A are based on the current structure of data in databases. These records are expected to be supplied as text or spreadsheet files. No information is currently available on any hardcopy reports that might be supplied for *in vitro* analysis results. *In vivo* reports are available and are described.

Codes and text that describe the assigned intake scenario are often listed in the database of plutonium and americium bioassay results. Descriptive codes and format descriptions are also addressed in Attachment 5A, Section 5A.1.

When a dose has been assigned to an individual, the dose summary report might contain the intake or incident date used in the calculation. An assumed intake date might also be available on the incident line in the current data base. An assumed intake date might be available on the "white card" that accompanies the *in vivo* reports.

If the report of an individual indicates that chelation therapy has been administered, then initially the excretion patterns cannot be correlated with the International Commission on Radiological Protection (ICRP) or other standard methodology. Historically, approximately 35 workers are known to have received chelation therapy for plutonium and americium intakes at LANL. Methods of handling these data are discussed in Section 5.4. Contact the internal dosimetry supervisor for guidance in interpreting bioassay data following chelation therapy.

### 5.1.2 Bioassay Program

Prior to 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, where a person not normally assigned to radiation work was asked to participate as a substitute in a task involving 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.

Short-term workers, such as summer students, persons participating in postdoctorate work, and teachers, might not have fully participated in routine bioassay programs. These workers were monitored for internal exposure only in unusual circumstances. Near the termination date, the worker might have received an *in vivo* count. The worker might have been required to submit an initial urine sample or have an initial *in vivo* count.

The Zia Company was the service workers' contractor. Zia employees participated in a separate monitoring program from that used for laboratory employees. Zia employees had a special computer program that locked out access to plutonium areas if participation in a plutonium bioassay program was not recorded within 425 days. Attachment 5A contains a list of criteria and exempt job categories. In 1978, in an attempt to reduce the number of sampled Zia employees to 500 per year; supervisors who only performed inspections were eliminated from the schedule.

In the 1970s, LANL initiated 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*.

One small, but significant, group of workers is the UPPU club (translated as "You pee Pu"). This group, established by Wright Langham in 1951, consists of individuals who had 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.

## 5.2 *IN VITRO* BIOASSAY

Historically, the *in vitro* bioassay program included the nuclides and techniques listed in Table 5-1. This section contains a detailed discussion of the techniques that were used for selected radionuclides. Table 5A-26, Attachment 5A, contains a summary of *in vitro* bioassay sensitivities listed by technique and year.

Table 5-1. Historical *in vitro* bioassay.

| Material | Start year | Comments |
|---|---|---|
| Tritium | 1950 | |
| Uranium | 1949 | In practice, 1951 |
| Plutonium | 1944 | |
| Polonium-210 | 1944 | |
| Americium-241 | 1954 | |
| 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 | Pending data from LANL | Not done regularly |

### 5.2.1 Plutonium

The most serious intakes at LANL have involved isotopes of plutonium and $^{241}$Am. The first urinalysis performed for evidence of plutonium uptake was performed on July 18, 1944, using a chemical procedure developed at the University of Chicago Metallurgical Laboratory in June 1944. However, Health Group personnel soon discovered that the procedure, using a 50-cm$^3$ aliquot of a 24-hour urine sample, did not have the sensitivity to meet the tolerance level of a 5-µg (0.33-µCi) burden of plutonium. The first recorded accident in which a human was subjected to a possible intake of plutonium occurred on August 19, 1944. This accident resulted in LASL being authorized to proceed, under the direction of Wright Langham, to develop a more sensitive procedure. The resulting 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 $^{239}$Pu. (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 $^{239}$Pu.)

The chemical forms of plutonium currently and historically encountered at LANL are oxide, nitrate, fluoride, and metal. Plutonium oxide is the most common form encountered and should be assumed if other information is not available.

**Respiratory Tract Absorption Type**
The absorption type assigned for plutonium isotopes is dependent on the chemical process. Table 5-2 lists absorption types as related to selected processes (Hempelmann, Richmond, and Voelz 1973). Table 5A-10 provides information on absorption types associated with locations.

**Sample Collection Procedures**
At the onset of the LASL 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

Table 5-2. Plutonium respiratory tract absorption type.

| Process/area | Isotope | Compound | Type | Comments |
|---|---|---|---|---|
| Recovery area | Pu-239 | Plutonium nitrate | M | In addition, the area might have oxalates, oxides, hydroxide and others based on source of input |
| Purification | Pu-239 | Nitrate, oxalate, sodium plutonyl acetate | M | |
| Fluorination | Pu-239 | Oxalate, fluoride, oxide | M-S | Powder dust, respirators worn |
| Reduction | Pu-239 | Metallic, fluoride, chloride | S | Welding |
| Heat source production | Pu-238 | High fired oxides | S | Possible extended retention and delayed excretion |

established at the hospital to ensure collection of contamination-free samples (Nickson 1945). This procedure was later deemed extremely expensive and was eliminated in 1952. A procedure that allowed collection of an equivalent 24-hour sample while the employee was off the site was initiated. A modified procedure to collect a 24-hour equivalent (two morning and two evening voidings) sample 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 dpm/day results. All sample results are expected to be reported in units per 24-hour day.

LASL instituted a sample validation procedure (Lawrence 1978). Attachment 5A, Section 5A.3, lists the validation procedure used for plutonium urine bioassay samples. This procedure is more rigorous than can be implemented without the computer software. However, certain samples in the database have been marked invalid based on these criteria. The dose reconstructor should not attempt to implement the validation procedure but should use best judgment when evaluating samples for inclusion in the intake calculations.

**Missed Intakes**

Intakes of plutonium could 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 LASL workers with high and low potential for exposure to plutonium has shown that measurable amounts of plutonium, above that which is expected from global fallout, could occur in low exposure potential individuals (Foreman, Moss, and Langham 1960).

The long-term excretion pattern of plutonium isotopes can permit 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 be directly related to the last bioassay result below the detection level.** The intake might have occurred many years earlier. Illustrating the magnitude of the potentially missed intake is one case cited in Hempelmann, Richmond, and Voelz (1973) that discusses an individual following a potential intake in 1944. A vial containing 10 mg of plutonium chloride exploded in a young chemist's face. The face was thoroughly scrubbed and the mouth washed out. Heavy contamination of the face persisted for several days. From the ionization of expired air, it was estimated that the level of contamination of the mouth was approximately 10 µg. The individual's urine analysis was less than detection levels in 1944, but more recent urine analysis determined that the individual did have a plutonium body burden. In this situation, the intake date would be 1944 and the dose should be assessed from that date.

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

**Routine Sample Frequencies**
Tables 5A-17 through 5A-20 in Attachment 5A list the routine sampling frequencies by historical period. The routine sampling frequencies can provide an upper bound of potential exposure for an individual, allow the appropriate grouping of samples into intake regimes, and provide a better estimate of intake dates. In 1978, sampling frequencies were reevaluated for LASL 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.

**Sample Analysis Procedures**
Initially the urine bioassay analysis procedure could not distinguish adequately between plutonium and polonium. 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 was in use between October 1949 and January 1957. No corrections were made for chemical blanks, recovery factors or counting geometry until 1957. 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) 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.

Use of a ZnS counter began in 1966. All of these methods measured total alpha activity from plutonium isotopes. Prior to 1967, when alpha spectroscopy was begun, isotopic fractions can be selected based on process knowledge, as described below in Plutonium Isotopic Ratios (Mixtures). Predominantly pure $^{238}$Pu was not encountered at LASL prior to 1968. Plutonium-241 and $^{241}$Am were not measured by total plutonium alpha procedures and the dose contribution from these nuclides might need to be accounted for using the mixtures in Tables 5-3, 5-4, and 5-7, unless the plutonium in the intake was known to be a pure isotope.

From 1967 to present-day operations, counting has been performed by alpha spectroscopy for specific plutonium isotopes. In 1997, thermal ionization mass spectroscopy (TIMS) and application of the class-100 clean room were added to the analytical technique. Samples are now routinely analyzed for $^{239}$Pu with TIMS and for $^{238}$Pu with radiometric alpha-spectroscopy (RAS). The sensitivity of the analysis for $^{239}$Pu has been reduced 40-fold by the addition of TIMS (Inkret et al. 1999a). TIMS does not measure $^{240}$Pu. Therefore, the amount of $^{240}$Pu associated with the mixture should be accounted for using Table 5-3, 5-4, or 5-7.

Minimum Detectable Activities (MDAs) reported in procedures and reports for the various radiochemical and counting methods are listed in Table 5-5.

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

**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 reported results for all periods should be interpreted as

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

| Mixture designation: | Fresh | 5-year | 10-year | 15-year | 20-year | 25-year | 30-year |
|---|---|---|---|---|---|---|---|
| Years of aging[a]: | 0 | 5 | 10 | 15 | 20 | 25 | 30 |
| 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 | 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 |
| Activity ratios | | | | | | | |
| Pu-239+240:Am-241 | NA | 66 | 37 | 27 | 23 | 20 | 19 |
| Pu-239+240:Pu-238 | 68 | 77 | 80 | 83 | 86 | 90 | 93 |
| Pu-239: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 | NA | 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.

Table 5-4. Activity composition of reference weapons-grade 6% plutonium mixture.

| Mixture designation: | Fresh | 5-year | 10-year | 15-year | 20-year | 25-year | 30-year |
|---|---|---|---|---|---|---|---|
| Years of aging[a]: | 0 | 5 | 10 | 15 | 20 | 25 | 30 |
| 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 |
| Activity ratios | | | | | | | |
| Pu-239+240:Am-241 | NA | 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 | NA | 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.

units/24-hr sample unless otherwise specified in the record. Some samples taken in response to suspected acute intakes might have been "spot" samples of less than 24-hr excretion. These sample results are expected to be either noted in the database or the results normalized to 24-hr excretion.

**Detection Sensitivities, Reporting Limits, Tolerance Limits**

Detection sensitivity was not a primary concern in the early years of operation of LASL. The concern was whether a tolerance limit was exceeded. Results below MDA were originally reported as "less-than" values ("LX.XX") or 0 until the 1980s, when actual results, positive and negative, were listed in the database. In the current database, all results, from all periods, have been listed as actual values,

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

| Nuclide | Year(s) | Sample type[c] | Technique ("era") | MDA level Unit/24-h sample[a] | Reporting limit Unit/24-h sample[a] | Tolerance limit[j] Unit/24-h sample[a] |
|---|---|---|---|---|---|---|
| Pu–Total alpha | 1944–1949 | U | Cupferron[h] | 0.8 pCi[e] (1 dpm DL (Moss 1990)) | >0.8 pCi | 7 cpm/24 hr or 6.3 pCi |
| | 1949–1/1957 | U | Bi-phosphate/alpha counting | 0.4 pCi [e] | 2 dpm or 0.9 pCi | 7 dpm/24 hr or 3 pCi |
| | 1/1957–1965 | U | Aluminum nitrate/NTA | 0.05 dpm at 99% confidence or 0.03 pCi[f] | 0.2 dpm or 0.09 pCi | 7 dpm/24 hr or 3 pCi |
| | 1966 | U | ZnS | 0.07 dpm or 0.03 pCi[g] | | |
| Pu-239 | 1967–1997 | U | RAS (PHA) only starting in 1971 | 0.03 pCi[i] (1mBq) | | |
| | 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<br>Oxalate<br>Rapid | Alkaline earth- 0.015 pCi/800 cc<br>Oxalate – 0.015 pCi/700 cc<br>Rapid – 1.6 pCi/L | | |
| | 1997–2002 | U | TIMS/RAS (alpha spec[b] ) | 7.6 µBq/24-hr[d]<br>1.03E-4 pCi/ 24-hr sensitivity | | |
| | 1997–present | U | RAS (alpha spec) | 8.1E-3 pCi or 300 µBq | | |
| | 2003–present | U | TIMS[k] | 3E-4 pCi | | |
| Pu-238 | 1967–1971 | U | RAS (alpha PHA) | 0.03 pCi[i] | 0.2 dpm/24hr investigate | |
| | 1971–1976 | U | RAS (alpha PHA) | 0.07 dpm or 0.03 pCi[i] | | |
| | 1977–1997 | U | RAS (PHA) | 0.03 pCi[i] | | |
| | 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. Unless otherwise noted.
b. Source: Inkret et al. (1999a). 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.
c. U=urine, F=fecal
d. 2x sensitivity
e. Source: Lawrence (1978).
f. Source: McInroy et al. (1991).
g. Source: Moss et al. (1969).
h. "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 1945)
i. Source: LASL (1978a)
j. Tolerance Limit – Considered the level under which it was 'safe'.
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.

positive or negative. When not specified, two times the detection level (or uncertainty of the blank) is considered MDA. Average chemical recoveries and matrix blanks were not reported prior to 1957. Therefore, the values reported in the tables for MDAs prior to 1957 are actually the results that would be reported if an analyzed personal 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 on whether the value was positive; therefore, these are being treated as MDAs in this document (Lawrence 1978).

**Prior to the start of RAS analysis in 1967, plutonium results were analyzed as total alpha plutonium. These results are found in the $^{239}$Pu database, but should be treated as total alpha plutonium, representative of the combination of $^{238}$Pu and $^{239+240}$Pu. Results from TIMS after 1997 are only $^{239}$Pu, without the contribution from $^{240}$Pu. Before using the results in dose calculations, the $^{240}$Pu contribution can be added to the $^{239}$Pu and entered as $^{239}$Pu, or the dose reconstructor can make separate calculations for $^{239}$Pu and $^{240}$Pu.**

Tables of plutonium bioassay sensitivities, as listed in procedures and reports, are found in Table 5-5. Also listed in Table 5-5 are Reporting Limits and Tolerance Limits for certain years. Tolerance Limits were defined as the level that could be accepted as "safe." Reporting Levels are those values above the MDA that are considered significant for recording or follow-up. However, the database, when completed, should have the actual bioassay results even if they are below the reporting level.

**Calculating pCi/24-hr for Urine Bioassay Results**

All plutonium bioassay results records are processed through the current database prior to reporting. Therefore, all results, from all periods, are expected to be reported in pCi/24 hr, with associated uncertainties reported as one standard deviation. However, should the need arise to calculate plutonium urine bioassay analysis results that are not fully processed, equations in Lawrence (1978) can be used to convert the listed units to pCi/24 hr. Select the method based on the "era" of the samples.

**Plutonium Isotopic Ratios (Mixtures)**

Dose assessments performed at LANL were and are currently based solely on the isotope(s) identified in the analysis. When total alpha measurements were made for plutonium, $^{239}$Pu was chosen based on process knowledge. **Results of these total alpha measurements prior to 1967 are found in the database and listed as $^{239}$Pu.** Since 1967, plutonium analysis is performed with alpha spectroscopy. RAS cannot distinguish between $^{239}$Pu and $^{240}$Pu because the alpha energies are similar and overlapping. However, $^{239+240}$Pu and $^{238}$Pu can be resolved using RAS. Thus, when quantities of $^{238}$Pu arrived at LASL in 1968, the analysis was capable of distinguishing between the isotopes of plutonium. Since 1997, TIMS analysis is available and is capable of distinguishing between $^{239}$Pu and $^{240}$Pu. **Results listed for TIMS analysis will be only $^{239}$Pu.** Typically, dose calculations performed in dose reconstructions, including those performed with the Integrated Modules for Bioassay Analysis (IMBA) computer program, assume a combination of $^{239+240}$Pu, treated as $^{239}$Pu. Therefore, if the results of TIMS analysis are used for calculation of intake, the contribution of $^{240}$Pu must be accounted for in the calculation.

No definitive historical information on the $^{240}$Pu:$^{239}$Pu atom ratios of LASL/LANL sources exists, nor is there any information on how the ratios vary with time and location. However, prior to 1970, plutonium at LASL was Hanford-derived (Gallaher and Efurd 2002). According to environmental impact studies, atom ratios from LASL plutonium released prior to 1970 would have been 0.01 to 0.03 $^{240}$Pu:$^{239}$Pu. LASL plutonium was Savannah River Site-derived starting in 1970. The atom ratio of LASL plutonium release after 1970 was expected to be 0.05 to 0.07, although ratios up to 0.13

indicated the sediment was affected by LASL plutonium rather than purely global fallout. Ratios above 0.07 in environmental samples were indicative of a mixture of global fallout and LASL plutonium. However, Table 5-6 describes the plutonium mixture found in 1960 in dirt used to sandblast plutonium parts during the demolition of a plutonium filter facility. Thus, although the $^{240}$Pu:$^{239}$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, pre-1970 mixtures of 3% listed in Table 5-3 are suggested when isotopic ratios are not known (Gallaher and Efurd 2002).

Table 5-6. Fresh plutonium mixture (Christensen, Garde, Valentine 1975)

| Isotope | % by weight |
|---|---|
| Pu-239 | 93.5% |
| Pu-240 | 6.0% |
| Pu-241 | 0.5% |

Table 5-4 lists activity and weight ratios as referenced for U.S. Department of Energy (DOE) sites for 6% weapons-grade plutonium (WGPu). These ratios are based on the presumed age of the material for DOE sites. There was practically no americium present in the plutonium handled at LASL in the1944-1945 period (Voelz, Grier, and Hempelmann 1985). Throughout the history of the site, plutonium mixtures are usually considered to be fresh and, therefore, do not initially contain a significant amount of americium.

Although trace quantities (by weight) of $^{238}$Pu have always been present in WGPu, relatively pure $^{238}$Pu did not arrive at LANL until 1968. At LANL, the relatively pure $^{238}$Pu has always been processed in separate dry boxes and gloveboxes, in later years, from WGPu to prevent cross-contamination of the two forms. Tables 5-3, 5-4, and 5-7 list the relative activities of plutonium isotopes and $^{241}$Am, which grows in from $^{241}$Pu, for 3% $^{240}$Pu (Gallaher and Efurd 2002), 6% $^{240}$Pu and 12% $^{240}$Pu mixtures (Carbaugh 2003). In these tables, "aging" refers to the time since the $^{241}$Am was separated from the plutonium.

Table 5-7. Activity composition of reference fuel-grade (12%) plutonium mixture.

| **Mixture designation:** | **Fresh** | **5-year** | **10-year** | **15-year** | **20-year** | **25-year** | **30-year** |
|---|---|---|---|---|---|---|---|
| **Years of aging[a]:** | **0** | **5** | **10** | **15** | **20** | **25** | **30** |
| 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 |
| Activity ratios | | | | | | | |
| Pu-239+240:Am-241 | NA | 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 | NA | 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.

Since 1967, $^{238}$Pu and $^{239+240}$Pu were measured separately; therefore, if positive results are identified in the sample, the ratio of one to the other can be used to estimate (1) the category of the plutonium mixture and (2) the activities of $^{241}$Pu and $^{241}$Am.

Prior to 1967, the measured quantity was total alpha from plutonium, which means the total of $^{238}$Pu and $^{239+240}$Pu. Total plutonium results are treated as $^{239}$Pu in the database. Most plutonium mixtures handled at LANL should be assumed to be nominally weapons-grade (6%) or, prior to 1957, 3%.

The nature of the process work at LANL, however, involved research-grade plutonium mixtures usually considered to be fresh. While the plutonium mixture encountered in processes should be assumed to be fresh, aged plutonium might have been encountered during building demolitions and in other areas where plutonium or plutonium contamination might have been in place for many years. Any $^{241}$Am observed in lung counts performed years after the intake can be assumed to be the result of the in-growth of $^{241}$Am from the $^{241}$Pu in the mixture over time or the $^{241}$Am in the initial plutonium mixture unless the incident report specifically indicates potential exposure to pure $^{241}$Am or urine bioassay for $^{241}$Am is found for the intake period. Assume 3% or 6% plutonium mixture unless other information is available in the dosimetry records.

**In addition, pure $^{238}$Pu might be encountered. A typical plutonium isotopic composition, by weight, with a specific activity of 14.1 µCi/µg, of the material released to the atmosphere in 1970 during an accident in Technical Area (TA)-21 (DP West) involving pure $^{238}$Pu is $^{238}$Pu (80%), $^{239}$Pu (16.9%), $^{240}$Pu (2.8%), $^{241}$Pu (0.2%), and $^{242}$Pu (0.1%) (Meyer 1970). Pure $^{238}$Pu is typically found in areas involving heat source technology.**

Fuel-grade plutonium (12%) was not routinely encountered at LANL. However, for maximizing dose, 10-year-old, 12% fuel-grade mixture can often be claimant-favorable. For intakes since about 1996, a 20-year-old mixture could be assumed. See Table 5-7.

**Validation of Samples**

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. The dose reconstructor should review the sample information carefully and make a decision on the relevance of the sample result(s). Section 5A.3 in Attachment 5A lists the protocol followed by the LASL computer software to validate a sample result. This section is provided for **information only**, to allow the dose reconstructor to possibly understand the criteria that were used to invalidate the sample result(s). This method **should not be used** by the dose reconstructor in the validation process.

The excretion rate following an intake 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 2001. Attachment 5A, Section 5A.9, contains actual bioassay results (urine bioassay and nasal swipes) 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. These results can be useful to the dose reconstructor when judging the variability of results to determine sample validity.

**Excretion Rate**

Pulmonary absorption type should be assumed according to work location or material composition, if known.

Special cases of excretion kinetics have been observed at LANL. The 1971 Wing 9 incident involving intakes of high-fired $^{238}$Pu oxide exhibited a very low excretion rate for the first 100 days after intake. A gradual rise in excretion rate was observed following that time, with no additional intake suspected (Miller et al. 1999). In addition, infrequent cases of plutonium inhalations at other DOE sites, probably associated with calcined or high-fired oxides, have been known to exhibit a low urinary excretion rate and a long retention time in the lung, consistent with the "bound state" in the ICRP (1994a) lung

model. The dose reconstructor should be aware of this possible type of behavior when evaluating a series of bioassay results. This is especially important when establishing the intake date because there might be several negative bioassay samples following the incident before the first positive sample. In these situations, the plutonium-to-americium ratio of the bioassay results is significantly different than the ratio of the inhaled product. Further, results of lung counts are significantly different from the lung burden predicted by the urine bioassay results (Hammond, Lagerquist, and Mann 1968). If chest count data show a long retention time, the dose reconstructor should use their best judgment on forcing the data to fit a type-S model or decreasing the absorption rate constants in the lung model.

**Sampling Protocol for Special Accidental Exposure**

Sampling protocols for special and accidental exposures are listed in Attachment 5A. The information might be useful to the dose reconstructor in estimating the date of intake if no date is apparent in the records. The incident line(s) in the plutonium database might list information concerning any incident related to the results. These records might provide more precise information on known or suspected intake dates. In addition, the record of nasal swipes, positive or negative, can aid the dose reconstructor in determining a plausible intake date. However, absence of a positive [above the 50-dpm Maximum Permissible Level (MPL)] nasal swipe should not invalidate an intake. In practice, a negative nasal swipe might have precluded further bioassay. The sampling protocol can provide guidance to the dose reconstructor for grouping bioassay samples into an intake regime. Tables 5A-21 to 5A-23 in Attachment 5A list the historical and current protocols for sampling for plutonium exposures.

**Nasal Swipes**

Until the first urine bioassay analysis was perfected and available in February 1945, “nose counts” were relied on to be a qualitative indicator of plutonium intake (Hempelmann 1945). After the development of urine bioassay techniques, nasal swipes were used to indicate the need for follow-up bioassay, although bioassay was not always performed immediately following a positive nasal swipe (McInroy et al. 1991). The MPL was 50 dpm, alpha, per nostril. 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 follow-up bioassay being performed by LANL. In addition, in 1944 LASL began the practice that if the two nasal counts varied significantly, the higher nasal count was considered spurious (LASL1944b). Information concerning a positive nasal swipe can often be found in the incident line of the database entry 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.

**Computer-Based Calculations and Sorting**

Beginning in 1959, LASL routinely employed 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.

Of relevance to dose reconstruction, these computer codes also 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 5A.4 of Attachment 5A.

All plutonium and americium data currently reside in the Los Alamos Bioassay Data Repository database. Data results, current and historical, have been converted to units of activity (typically pCi) per 24 hours. All results are reported in actual values (e.g., positive, negative, or zero). Uncertainty values are listed for each measurement. Codes for the interpretation of the records are in Section 5A.1 of Attachment 5A.

### 5.2.2 Americium

At LANL $^{241}$Am is usually encountered as a trace contaminant in plutonium. However, there is potential for exposure to pure $^{241}$Am at LANL. No exposure to pure $^{241}$Am was likely prior to the beginning of the americium bioassay program in 1954, although a procedure for determining $^{241}$Am in urine was in development in 1948 (LASL 1948).

Current and historical bioassay results are stored in the Los Alamos Bioassay Data Repository database with the plutonium results. Codes and formats of these results are likely to be identical to those described in Attachment 5A, Section 5A.1, for plutonium bioassay results.

There is an 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. Conversely, 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 mixture.

Prior to the mid-1990s, very few individuals submitted urine samples that were analyzed for the presence of $^{241}$Am. Samples were submitted in response to incidents involving exposures to sources of $^{241}$Am not contained in the plutonium mixture. Estimates of internal doses are based on the results of chest counts and urine bioassay (Inkret et al. 1998a). Urinalysis is the principal bioassay method used for the assessment of intakes. ICRP (1994b) lists the respiratory absorption type for americium as type M. However, if $^{241}$Am is determined to be part of a plutonium mixture or in-growth from the intake of a plutonium mixture, the absorption type assigned to the plutonium mixture should be assumed for the $^{241}$Am.

The current routine and special sampling programs have protocols similar to those for the plutonium program. Table 5A-18 in Attachment 5A describes the routine sampling frequency. Table 5A-24 describes the current program of special sampling frequencies. The sampling protocols might aid the grouping of samples into intake regimes or identification of an intake date. 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. Lack of a positive result on a nasal swipe should not eliminate the possibility of an intake.

**Minimum Detectable Activities**
Historically, emphasis was not on individuals being below the MDA, but rather being below the tolerance level. All bioassay results are expected to be reported in pCi/24-hr sample; therefore, the listed urine bioassay MDAs have been normalized to pCi/24-hr sample.

Historically, results below MDA were reported as less-than values (“LX.XX”) or 0 until the 1980s, when actual results, positive and negative, are listed in the database. In the current database all results from all periods have been listed as actual values, positive or negative. When not specified, two times the detection level (or uncertainty of the blank) is considered MDA. Table 5-8 lists MDAs. MDAs are based on MDAs listed in procedures and reports.

Table 5-8. $^{241}$Am bioassay techniques and sensitivities.

| Sample type | Year | Method | MDA |
|---|---|---|---|
| Urine | 1954 – 1957 | Unknown | 9.E-01 pCi/24hr[a] |
| Urine | 1958 – 1982 | Chemical extraction/ proportional counting | 2.E-01 pCi/24hr[b] |
| Fecal | 1977 | Phoswich | 4.E-02 nCi/sample |
| Urine | 1983[c] – 2002 | Co-precipitation/alpha spec | 1.5E-02 pCi/24hr |
| Fecal | 1983[c] | Am/Pu screening/Phoswich | 1.E-02 nCi/sample |
| Urine | 2003 – present | Alpha spec | 1.0E-02 pCi/24hr |

a. No MDA available, use DIL; Tolerance level 3.1 pCi/sample.
b. Source: Milligan et al. (1958); method can carry over thorium, plutonium, curium, actinium, and neptunium. Exact end date to the start of this MDA is not known.
c. Source: Gautier (1983); exact start date of the MDA is not known.
d. Source: Inkret et al. (1998a).
e. Sent to an offsite laboratory.

When no MDA value is provided with the sample, dose reconstructors should use the values in Table 5-8 to determine whether a listed bioassay result is positive or to calculate missed dose using detection levels. It is not expected that MDAs will be provided with results because all results are now reported in actual values. MDAs in Table 5-8 are listed in pCi/24-hour sample. At present, the electronic database contains results in units of pCi/24-hour sample.

### 5.2.3 Tritium

Tritium was encountered in several forms: tritiated water (HTO), tritiated gas (HT), organically bound tritium (OBT), and metal tritide (MT). Each form has unique characteristics. Each year approximately 100 individuals are monitored for tritium intakes at LANL. If the form is not indicated or the work area is not specific, then, with the exception of metal tritides, assume HTO, which is the form generally encountered. However, in the HRL and IBF Buildings, the predominant form of tritium was OBT.

Tritium incidents have been recorded for accelerator areas.

**Organically Bound Tritium**
The first approximation of the dose from OBT (labeled) compounds is the tritium in body water dose. However, the absorption, distribution, and excretion of tritium-labeled compounds are specific to the chemical and physiological behavior of the particular compound. Specific guidance is available in OCAS-TIB-002 (OCAS 2003a) when the dose records indicate an exposure to tritium-labeled compounds.

Locations such as the HRL and IBF list labeled compounds as the principal type of radionuclides encountered at these facilities. Intakes of labeled compounds do not follow the same biokinetics as the ICRP Publication 68 defaults (ICRP 1994b). OCAS-TIB-002 (OCAS 2003a) discusses the IMBA model for OBT.

**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. If urine bioassay results are interpreted as HTO, when the actual exposure was to MT, the calculated dose to the lung and the whole body will be an underestimate. Inkret et al. (1999b) shows that the particle size at LANL is approximately 2 to 10 μm. In simulated lung fluids, the dissolution rate of $4.7 \times 10^{-6}$/day and a corresponding biological half-time in the lungs of $1.5 \times 10^{5}$ days were observed. Long-term excretion of the dissolved tritium component can be observed for periods up to 50 days after an acute

intake. The derived investigation levels (DILs) range from 0.005 - 5 µCi/L compared with 5 µCi/L for HTO.

OCAS-TIB-002 (OCAS 2003a) provides guidance on the evaluation of MT intakes. Metal tritides are referred to as *Tritium Particulates*. Information on building locations and years of operation when MT might have been encountered is not currently available. The claimant telephone interview can provide indications that a person was exposed to MT. If the dose reconstructor encounters a case involving exposure to MT, discussion among the task leadership might be needed to determine the appropriate biokinetic model.

Interpretation of codes and units of the bioassay data for tritium is discussed in Attachment 5A, Section 5A.1.

**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 (McClelland and Milligan 1954), the tolerance for tritium in urine was 250 µCi/L. The analytical range was 1 to 250 µCi/L ($5.18 \times 10^4$ to $1.30 \times 10^7$ Bq/24 hr). In 1958, an internal GM counting technique was used. The reported MDA for this method was 1 µCi/L ($5.18 \times 10^4$ Bq/24 hr). In 1970, liquid scintillation counting began and is still in use today. With the introduction of liquid scintillation counting, the detection limit was 1 µCi/L ($5.18 \times 10^4$ Bq/24 hr). The MDA consistently improved.

For tritium results, the denominator used for reporting purposes was *per liter* (/L) of urine. All data from 1950 through 1992 were converted to µCi/L in 1992. However, results in the current database are listed as Bq/24-hr sample. At present, all results are likely to be reported in units of Bq/24-hr sample. Tritium bioassay results can be found as early as 1946.

Results for samples can be listed as "0" or coded as "LX.XX" 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 5-9 and summarized in Attachment 5A, Table 5A-26. All results in the current database are likely to be reported in actual values, positive, negative, or zero.

Table 5-9. Tritium urine bioassay sensitivity levels.

| Time | Detection level | Reporting level | Counting method |
|---|---|---|---|
| 1950 – 1951 | 5.18E4 Bq/24 h[a] (1 µCi/L) | | Electroscope |
| 1952 – 1953 | 5.18E4 Bq/24 h[a] (1 µCi/L) | | GM counter |
| 1954 – 1957 | 5.18E4 Bq/24 h (1 µCi/L) | | |
| 1958 – 1968 | 5.18E4 Bq/24 h (1 µCi/L) | | Internal GM |
| 1969[b] – 1987 | 1.04E3 Bq/24 h (0.02 µCi/L)[b] | 5.18E4 Bq/24 h (1 µCi/L) | LSC |
| 1988 – 1998 | 5.18E2 Bq/24 h (0.01 µCi/L) | 5.18E3 Bq/24 h (0.1 µCi/L) | LSC and 1 ml raw urine |
| 1999 – present | 2.59E2 Bq/24 h(0.005 µCi/L) | | LSC |

a. Expected to be the same as 1954
b. Source: Gautier (1983).

**Routine Sampling**

The routine sampling protocols for tritium bioassay throughout the history of the program are listed in Attachment 5A, Table 5A-19. Routine sampling protocols can provide an upper bound to intake and assist in grouping of samples into intake regimes.

It should be noted that the Kanne Chamber used to monitor tritium releases at the accelerator was recalibrated in 1977 and found to be calibrated high by a factor of x10. The previous calibration had been done 15 years earlier.

### 5.2.4 Uranium

Historically, uranium was primarily either depleted (D-38, 28) or enriched (Or-93, 25) at LANL with a variety of isotopic ratios (Little, Miller, and Guilmette 2003a). Table 5-10 provides generic uranium conversion factors derived from Rich et al. (1988). Tables 5-11 and 5-12 describe the nominal weight composition and fractional activity for each mixture (Lawrence 1990b). However, if specific isotopic information is not available the default values in IMBA can be used in the dose reconstruction.

Table 5-10. Uranium conversion tables.

| Enrichment percent | Fraction by activity | | | Fraction by mass | | | Total |
|---|---|---|---|---|---|---|---|
| | U-234 | U-235 | U-238 | U-234 | U-235 | U-238 | U pCi/µg |
| 0.1 | 0.225 | 0.010 | 0.765 | 0.00001597 | 0.001 | 0.99898 | 0.438 |
| 0.17 | 0.260 | 0.010 | 0.730 | 0.00002 | 0.0017 | 0.999 | 0.465 |
| 0.2 | 0.285 | 0.010 | 0.705 | 0.00002178 | 0.002 | 0.997978 | 0.476 |
| 0.3 | 0.340 | 0.010 | 0.650 | 0.00003 | 0.003 | 0.997 | 0.514 |
| 0.5 | 0.410 | 0.020 | 0.570 | 0.00004 | 0.005 | 0.995 | 0.591 |
| 0.7 | 0.480 | 0.020 | 0.500 | 0.00005 | 0.007 | 0.993 | 0.668 |
| 0.711 | 0.486 | 0.022 | 0.492 | 0.00005 | 0.007 | 0.993 | 0.672 |
| 0.72 | 0.486 | 0.022 | 0.492 | 0.00005 | 0.007 | 0.993 | 0.675 |
| 3 | 0.745 | 0.045 | 0.210 | 0.00019 | 0.030 | 0.970 | 1.57 |
| 90 | 0.970 | 0.029 | 0.001 | 0.010 | 0.900 | 0.090 | 62.1 |
| 93 | 0.972 | 0.027 | 0.001 | 0.010 | 0.930 | 0.060 | 65.1 |
| 95 | 0.973 | 0.026 | 0.001 | 0.010 | 0.950 | 0.040 | 67.2 |

Table 5-11. D-38 uranium – nominal weight composition and fractional activities (Lawrence 1990b).

| Isotope | U-234 | U-235 | U-236 | U-238 | Total |
|---|---|---|---|---|---|
| Weight fraction | 0.00002 | 0.003 | 0.000003 | 0.997 | |
| Curies in 1g 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 5-12. Or-93 uranium – nominal weight composition and fractional activities (Lawrence 1990b).

| Isotope | U-234 | U-235 | U-236 | U-238 | Total |
|---|---|---|---|---|---|
| Weight fraction | 0.011 | 0.933 | 0.002 | 0.054 | |
| Curies in 1g 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 | |

Depleted (D-38) is the most common form of uranium encountered at LANL (Inkret et al. 1998b), although natural uranium (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 natural uranium.) 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). However, the partition between Class D and W could not be determined. Historically, Class W was used for reporting results (Lawrence 1992a) because it produced larger

doses. The equivalent absorption type M is recommended unless other forms are specified in the individual's dose record (Little, Miller, and Guilmette 2003a).

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, so type M should be used as the default. Therefore, absorption type F or M should be considered unless type S is indicated by the fitting of the bioassay or by process knowledge. Statements about welding on uranium, both depleted and eventually enriched, which would have been a potential for more insoluble forms of uranium are found in LASL (1951). However, if specific isotopic information is not available, the default values in IMBA can be used in the dose reconstruction.

If the specific compound of uranium is known, Table 5A-29 in Attachment 5A can be used to determine the solubility class and/or absorption type.

The LANL program calculated intakes for individuals having at least one positive urine analysis result. The potential for intake was indicated by a positive nasal swipe. A positive nasal swipe was 50 dpm in either nostril. Lack of a positive result on a nasal swipe should not eliminate the possibility of an intake.

Table 5-10 lists activity and weight fractions for several typical enrichments (Rich et al. 1988). Otherwise, dose reconstructors can use the nominal compositions in Tables 5-11 and 5-12 as default compositions when no other information is available on the enrichment (mass or weight fraction of $^{235}U$ multiplied by 100%) of the uranium, although IMBA defaults are also appropriate. These are the compositions found at LASL from 1970 through 1990, at least (Lawrence 1990b).

**Sampling Protocol**

The routine and non-routine sampling protocols provide information that is helpful in estimating intake dates and grouping samples into intake regimes. Historical sampling protocols are listed in Attachment 5A, Table 5A-20. The program was initiated in 1949 with analysis for uranium mass, purely as an indicator of exposure. Samples were collected on Fridays just before leaving work 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 going into the work area. The sample might also have consisted of 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 following an intake was excluded from the sample (Lawrence 1984). Beginning in July 1993, the larger volume of sample required by the alpha spectroscopy method of analysis made the Sunday evening/ Monday morning collections standard protocol (Lawrence 1992b).

Retroactive calculations were made by Lawrence of the data beginning in 1949. Data for 1951 was not included in the retroactive calculations performed in 1992 by Lawrence because the bound Los Alamos Notebooks (LANB) could not be found. These data might now be available. For $^{235}U$, enriched uranium, the conversion of data into the computer files and retroactive calculations started with the 1955 data. Assays identified as $^{238}U$ were converted to µg/day with values less than MDA or 0 set to the "minimis" MDA. Assays identified as $^{235}U$ were converted to pCi/day with values less than MDA or 0 set to the "minimis" MDA.

### Uranium Analysis Techniques

Several analysis techniques were employed over the history of LANL. Techniques include fluorophotometric, ion exchange, or extraction chemistry with radiometric alpha proportional counting, delayed neutron activation analysis (DNAA), and radiometric alpha spectroscopy (RAS).

Table 5-13 summarizes the routine urinalysis detection levels for various periods. The results of urine bioassay for uranium have been entered into a database. However, no examples are currently available of the bioassay data format that will be provided for the claimant from this database. Therefore, units, codes, normalization of sample volume, and other information cannot be described for the dose reconstructor. The results in 1991 or before might be reported in actual values, positive or negative (Lawrence 1992a). There was discussion of starting the practice of establishing a minimum detectable activity and reporting results less than the minimum detectable activity as "0" after 1992.

Table 5-13. Routine uranium urinalysis detection levels.

| Period | Method[a] | MDA | Decision level | Reporting level[b] |
|---|---|---|---|---|
| 1949 – 1967 | Fluorophotometric (DU or Unat) | None listed[c] | 50 µg/L | >100 µg/L |
| 1968 – 2/1976 | Fluorophotometric (DU or Unat) | 4 µg/L[c,d] U | | |
| 3/1976 – 1978 | Fluorophotometric (DU or Unat) | 1 µg/L[d] 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) | ? dpm/L[c,e] | 50 dpm/L | >100 dpm/L |
| 3/1971 – 1976 | Extraction/alpha proportional counting | 15 dpm/L[c] | | |
| 6/1976 – 1/1977 | Extraction/alpha proportional counting | 10 dpm/L[c] | | |
| 2/1977-12/1977 | Extraction/alpha proportional counting | 4 pCi/L | | |
| 1978 – 7/1982 | DNAA/delayed neutron counting[f] Only U-235 counted | 1 µg/L U-238 0.17% enriched | | |
| 7/1982 – 6/1991 | DNAA/delayed neutron counting[f] Only U-235 counted | 4 pCi/L U-235[f] 93% enriched | | |
| 1982 – 6/1991 | DNAA/delayed neutron counting[f] Only U-235 counted | 4 µg/L U-238 0.17% enriched | | |
| 1982 – 1992 | Anion exchange/colorimetric (used to confirm DNAA >30 µg/L | 1 µg/50 $cm^3$ | | |
| 7/1991[g] – 1998 | RAS[h] for each isotope | 0.1 pCi/L | 0.05 pCi/L | |
| 1998 – present | RAS[h] for each isotope | 0.008 pCi/L | 0.004 pCi/L (1 standard deviation) | |

a. Method listed.
b. Exceeding Reporting Levels required investigation and evaluation (Lawrence 1984).
c. Lawrence (1984).
d. 50 µg/L considered positive indication of natural uranium material in the body (Dummer 1958).
e. Specific for U-235 and U-233, 50 dpm/24-hr sample considered positive indication of enriched uranium in the body (Dummer 1958).
f. **U-235 results should be divided by 0.0284 to calculate the $^{234}U$ activity. See Section on Delayed Neutron Counting below for full explanation of the DNAA results including minimum reporting levels.**
g. To maintain consistency during the transition from DNAA to RAS, urine results for 1991 were reported as DNAA results. For samples analyzed starting July 1991, summation of the U-234, U-235, and U-238 results was made. If the sum was less than 4 pCi/L, a single result was produced with the value "L4.00 pCi/L" and labeled U235, simulating the DNAA results. In addition, each result was converted to µg/L and summed to obtain the total uranium mass. If the sum was less than 4.0 µg/L, a single result was produced with the value of "L4.00 µg/L" and labeled U238. RAS values were used directly in 1992 (Lawrence 1992b).
h. Can measure U-234, U-235, and U-238. Alpha spectrometry cannot differentiate between U-233 and U-234.

### Delayed Neutron Counting

DNAA was the analytical technique used between 1978 and 1992. Interpretation of the results during this time was dependent on identifying 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 results reported by the DNAA method for enriched uranium analyses are labeled U235 in the records, but have always included the alpha activity of $^{234}$U that is about 34 times that of $^{235}$U. Similarly, results for depleted uranium analyses are labeled U238, but have always been the total uranium mass based on the assumed isotopic mass fractions of 0.00002 $^{234}$U, 0.0017 $^{235}$U, and 0.9983 $^{238}$U.**

By the DNAA method, the minimum reporting value was 4 µg/L for depleted uranium ($^{238}$U at 0.17% enrichment) and 4 pCi/L for enriched uranium (93% enriched). For D-38 (depleted uranium), a 4-µg/L sample would contain 0.0147 pCi/L of $^{235}$U [(4µg/L * 0.0017 * 2.163 × $10^{-6}$ Ci/g * 1 × $10^{12}$ pCi/Ci)/1 × $10^{6}$ µg/g]. The enrichment of $^{235}$U assumed for depleted uranium at this period was 0.17%. A 4-pCi/L sample of enriched uranium (93%) would contain approximately 0.116 pCi/L of $^{235}$U [0.116 pCi/L divided by 0.0284 from Table 5-12 equals 4 pCi/L]. If the person's actual urine sample contained 0.116 pCi/L of $^{235}$U, the reported value for enriched uranium would be 4 pCi/L. If the analysis was designated for both enriched and depleted uranium, the D-38 value would be reported as approximately 32 µg/L [(0.116/0.0147) times 4 µg/L] and 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 person's dose (Lawrence 1992b).

Uncertainty for this method was considered to be 96% ±6% at 5 – 26 pCi/L for enriched uranium and 93% ±8% at 4 -17 µg/L for depleted uranium.

This method was eventually deemed operationally unacceptable because of longer-than-promised 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 5-13. More guidance on interpreting the results will be available when examples of the uranium database are available.

**Radiochemical, Alpha Spectroscopy (RAS Technique)**

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

Several problems were initially encountered with this method. No chemical or synthetic urine blanks were used to establish 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 analysis indicated isotopic ratios outside the realm of probability. The computer program converted all negative values to zero. All 1991 data yielded negative values for $^{235}$U, rather than the expected negative and positive values. In early 1992, the computer program was still not modified to correct for chemical blanks.

**Excretion of Environmental Levels of Uranium**

Natural uranium from nonoccupational intakes (primarily food and water) is excreted in urine at levels above the analytical MDAs for either the elemental uranium analysis or the alpha spectrometry analysis. The $^{234}$U:$^{238}$U ratio can be used to distinguish depleted uranium from natural uranium. Table 5-14 lists activity ratios for natural uranium.

Table 5-14. Natural uranium (Tuballoy).

| Isotope | U-234 | U-235 | U-236 | U-238 | Total |
|---|---|---|---|---|---|
| Weight fraction | 0.0000537 | 0.0072 | 0 | 0.99274 | |
| Specific activity pCi/µg | 0.33367 | 0.01557 | | 0.33367 | 0.68291[a] |
| Fraction of total activity | 0.4886 | 0.0228 | 0 | 0.4886 | |

a. As listed in IMBA NIOSH Phase I database USDOE Version 1.0.42.

A 1992 study **(Little, Miller, and Guilmette 2003a)** listed the average drinking water concentrations for the Los Alamos/ White Rock/Santa Fe area as 0.015 µg/L (0.01 pCi/L or 0.00037 Bq/L). **Therefore, using 0.01 as the factor of drinking water concentration to excretion concentration (Little, Miller, and Guilmette 2003a,b),** it is reasonable to use the urinary excretion values of 0.00015 µg/L for elemental analyses, 0.0001 pCi/L for $^{234}$U and $^{238}$U, and essentially anything detected for $^{235}$U, to distinguish between natural background and potential occupational exposure for uranium, unless the worker's file indicates that the excretion was from natural sources

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

Beginning in 1992, all workers participating 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 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, the values should be multiplied by 0.01 to calculate the concentrations of uranium isotopes to be subtracted from the individual's bioassay results prior to calculating an occupational intake.** The factor is based on consuming 1.1 L of water a day (Little, Miller, and Guilmette 2003b). The concentrations were found to vary widely even for individuals. The concentration of the most closely associated water sample should be subtracted from the bioassay sample rather than an average value for all the samples listed for the individual. Other variables, such as drinking habits, dietary components, and individual physiological differences, can influence the individual's baseline excretion rate of environmental uranium. However, the use of the factor is the best approximation available to the dose reconstruction.

However, when other environmental data are not available, the default environmental decision levels should apply to the entire history of LANL.

Background excretion of uranium in feces probably varies 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 Fission and Activation Product Analysis

The first gross beta urine count was devised in 1947 (LASL 1947). Fission product urinalysis was the method used to monitor for intakes of fission products until whole-body counting was fully implemented in 1970. A procedure dated 1958 lists gross beta urine analysis from $^{90}$Sr/Y, $^{140}$Ba/La, $^{144}$Ce/Pr, $^{89}$Sr, and gross fission products (Milligan et al. 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 >200 dpm/L. A similar procedure of oxalate coprecipitation and beta counting, effective in 1974 (Gautier 1983), lists a sensitivity of 1-2 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 $^{137}$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 $^{131}$I and other fission products. Room air concentrations and urine bioassay results have been located in office memoranda (LASL 1959b). 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 $^{140}$Ba/La and $^{90}$Sr/Y exists for those participating in the RaLa chemistry and testing, 1944 through 1962, 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 5 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, $^{140}$Ba was recorded; for an 8-day half-life, $^{131}$I was recorded. Results had a background of $^{40}$K subtracted (typically 20 cpm/1500 mL). (LASL 1959b)

Gas leaks were detected with filter papers and charcoal. Filter papers and charcoal were analyzed by gamma spectroscopy. Personnel potentially exposed were analyzed in the human spectrometer [9.5-in. x 6-in. NaI(Tl) crystal]. Iodine-135 (6.57-hour half-life)/$^{135}$Xe (9-hour half-life), $^{131}$I, and $^{133}$I were observed in individuals exposed to gas leaks at reactors (LASL 1959b). Through the late 1940s and as late as 1961, prior to the use of charcoal cartridges, iodine was analyzed from the paper filter assuming a collection efficiency of 0.1%. Radioiodine and noble gases are released from facilities performing 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 $^{131}$I release (environmental release <1 mCi/y) from TA-48 operations, and the OWR Facility (TA-2) is not a source of radioiodine.

Interpretation of the fission/activation product urinalysis in a way that is meaningful, as representative of all the possible fission products 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 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 interpreting 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, the environmental background radiation in Los Alamos was as high as 5 mR/hr from fresh fission products. These radionuclides were found in urine bioassays for gross fission products. The background levels, which were variable, provide a complicating factor when using 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/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.

**Strontium**

Records of $^{90}$Sr urinalyses, routine or special, are very sparse. The historical compilations of procedures do not list a specific $^{90}$Sr urinalysis procedure. It appears that any record of $^{90}$Sr analysis actually indicates that LANL performed a gross beta analysis or sent a sample to an outside laboratory. Strontium-90 dose currently can be reconstructed only when $^{90}$Sr results are actually listed for an individual. No information is available on MDA for $^{90}$Sr analysis. Section 5.3 discusses the early HUMCO whole-body counters as being capable of counting the *bremsstrahlung* from $^{90}$Sr. It appears that, at most, several hundred bioassay analyses might have occurred during the history of LANL.

Exposure to $^{90}$Sr can be expected for persons working in the RaLa program because $^{90}$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 $^{90}$Sr. Persons from TA-1, Sigma, H, and U Buildings, where RaLa operations were conducted, would also have the potential for intakes of $^{140}$Ba and $^{140}$La. The RaLa sources were prepared by Group G-7 or Group 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 some time between April and December 1950. The Idaho Chemical Processing Plant became the source of purified $^{140}$Ba in 1956, and a typical shipment was about 40,000 Ci of $^{140}$Ba. The $^{140}$La sources prepared at Ten Site were usually in the range of 2,000 to 4,000 Ci. Almost 2 million curies of $^{140}$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 $^{140}$La would have been detectable in the HUMCO screening and quantified in the NaI follow-up, although no MDA is currently available for those nuclides. Savannah River Site lists an MDA of 9.3 nCi for that period.

### 5.2.6 Polonium

Polonium was used in initiators in quantities of 100 Ci/month or more starting in February 1945 and increasing to 500 Ci/month by December 1945. Prior to that time, smaller quantities were handled at TA-1, 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 $^{210}$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.

Work with $^{210}$Po was of a limited scope at LANL. Therefore, missed dose for polonium should be assessed only if actual polonium bioassay has been 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, LASL (1977a) states, “During the 1940s and early 1950s, $^{210}$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) states that the possibility of exposure should be investigated at greater than 10 dpm/L in a urine bioassay sample.

Hemplemann (1945) states that, because of the ease of the analysis, polonium urine bioassay was performed routinely and frequently on all individuals working with polonium. However, records of routine polonium bioassays might not have been transferred to the current LANL database and might not be available in individual dose records.

Initially the urine bioassay analysis procedure could not distinguish adequately 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.

Detection limits for routine urinalysis are listed in Table 5-15. A procedure is listed for $^{210}$Po in urine in Gautier (1983). MDA continues to later periods unless changed.

Table 5-15. Routine $^{210}$Po urinalysis detection levels.

| Period | MDA | Recheck | Tolerance |
|---|---|---|---|
| 1943-1952 | | | 440 dpm/24h[a] |
| 1953 | | | 50 dpm/24h[a] |
| 1954 | 10 dpm/L | 100 dpm/L | 500 dpm/L |
| 1955 | 0.1 pCi/L | | |
| 1958 | | 100 dpm/L | 500 dpm/L |

a. Source: LASL (1979).

Because $^{210}$Po is a naturally occurring radionuclide from the $^{238}$U decay chain, $^{210}$Po exists naturally in urine and feces. Nothing was found in the records indicating that a study on natural excretion levels for persons living around LANL had been conducted. ICRP Publication 23 indicates that excretion levels differ between smokers and nonsmokers (ICRP 1975). The following estimated excretion values are provided: urine, smokers - 0.065 pCi/d, nonsmokers - 0.011 pCi/d; feces, smokers - 3.3 pCi/d, nonsmokers - 3.2 pCi/d.

If there are person-specific baseline values for urine or fecal excretion of $^{210}$Po, those should be used to subtract from later results. If not, the ICRP (1975) values above can be used.

### 5.2.7 Other Limited-Exposure Radionuclides

LANL has always been a center for research. As such, small-scale (in terms of either the number of persons or activity of the source) use of various radionuclides not addressed above has occurred throughout the history of LANL. Little or no documentation has been found on bioassay for these nuclides.

These nuclides should be assessed only when there is an indication that the worker had a potential for interaction with that nuclide.

**Actinium-227**

Notebook 6489 containing $^{227}$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 $^{227}$Ac bioassay have been found. Results listed were 0 to 0.6 dpm/sample.

Potential worker encounter with $^{227}$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).

No other information is currently available.

**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 $^{32}$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 is found for $^{14}$C.

**Thorium**
A procedure for a coprecipitation technique and a colorimetric final determination is listed as effective January 1963. The MDA for this analysis is listed as 20 µg/L. Dummer (1958) lists the MDA for the colorimetric procedure as 0.01 µg/L. While plutonium can coprecipitate with thorium, the colorimetric determination prevented any interference from the plutonium. An earlier procedure titled "The Determination of Thorium$^{230}$ in Urine" is listed in Milligan et al. (1958). This procedure uses low background alpha proportional counting and lists a sensitivity of 0.05 dpm (no volume units) of $^{230}$Th. The procedure states that natural thorium, plutonium, some americium, curium, actinium, and neptunium are carried over with the precipitate.

Only the results of radiometric analysis of 44 urine samples for $^{230}$Th performed in 1958 have been found thus far. A sensitivity of 0.2 dpm (no volume units) is listed for these analyses. Dummer (1958) suggests the use of alpha proportional counting for $^{230}$Th bioassay samples. Thorium-230 is also referred to as ionium.

LASL (1951) discusses monitoring of thorium daughters in air. Daughter products would be produced from the decay of $^{232}$Th. Casting, machining, and other operations with thorium metal were undertaken in 1951. This suggests that $^{232}$Th was in use during this period. Thorium-232 has been in environmental emissions from waste sites at LANL. After 1980, some work with thorium-impregnated cloth was performed. The thorium in the cloth was probably $^{232}$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, storing ingot and oxide forms, which would suggest $^{232}$Th and daughters. Therefore, the dose reconstructor should use best judgment when selecting the isotope or mixture. Thorium-230 is the maximizing conversion from mass units.

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 $^{230}$Th incidental to uranium exposure).

If radiometric alpha spectroscopy analysis, rather than total alpha, results of $^{232}$Th are encountered, the contribution from decay product nuclides should be included. Thorium-232 decays with a 6.7-year half-life to $^{228}$Ra, which decays with a 5.75-year half-life to 6.13-hour $^{228}$Ac, which decays to 1.91-year $^{228}$Th. See Table 5-16.

Table 5-16. 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 |

**Protactinium-231**

A procedure for a coprecipitation technique and proportional alpha counting is documented in Milligan et al. (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.

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

*In vivo* counting equipment and techniques were developed in the late 1950s and have been in routine use for measuring X-ray and gamma-ray-emitting radionuclides since 1970, and possibly as early as 1960. There is some indication that some of the counts recorded between the beginning of the program in 1955 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.

### 5.3.1 Whole-Body Counters

The first whole-body counter to be used at LASL was the HUMCO I. This "human counter" became operational in 1955. The counter consisted of a large double cylinder with a liquid scintillation fluid (possibly trichloroethylene) filling 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 $^{40}$K (1-2 MeV) and one for $^{137}$Cs + $^{40}$K Compton counts between 0.5 and 0.8 MeV. The result was obtained by subtracting out the contribution of $^{40}$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 $^{90}$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) crystal were approximately the same as those for the HUMCO, except 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. of pre-World War II steel. The resolution was improved, but it remained a screening counter.

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 $^{241}$Am, and 200- to 2,000-keV photons (CsI) for a qualitative assessment of a variety of fission and activation nuclides. A planar Hyper Pure 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. Finally, a HPGe (formerly a GeLi) 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.

The twin Phoswich detectors were replaced by twin three-detector arrays of HPGe detectors in 1999. The Phoswich and germanium systems were operated concurrently during the 1998-1999 period. Two of the six HPGe detectors are used when a thyroid count is required.

**Minimum Detectable Activity and Decision Levels**

Operationally an observed signal must exceed the Decision Level ($L_C$), formerly referred to as the Maximum Significant Measured Activity (MSMA), to result in the decision "detected" (decision level) that there is some activity in the lung, body, or other organ (Vasilik et al. 1984). The MDA, formerly the Minimum Detectable True Activity (MDTA), is the smallest amount of activity required to be in the lungs or organ so that a measurement of an individual can be expected to imply, correctly, that presence of activity with a predetermined degree of confidence. MDA (or MDTA) and $L_C$ (or MSMA) values are listed in Table 5-17 for various years of operation. The MDA and $L_C$ values for lung counting are summarized in Attachment 5A, Table 5A-27. The values of MDA and $L_C$ are calculated according to the theoretical developments of Currie and of Altshuler and Pasternack (Vasilik et al. 1984). No information is currently available on MDAs for the thyroid detector or for $^{131}I$ or $^{125}I$ in the whole-body count.

Whole-body and lung counting are the primary methods for determining intakes of fission and activation products. When both whole-body (*in vivo*) and *in vitro* results are available, whole-body count results are generally considered more sensitive and dose reconstructors should use them to determine intake.

Results found in bioassay records are generally reported in nCi unless otherwise indicated. Results listed as NULL indicate NDA (No Detectable Activity), not MDA. Results less than the $L_C$ are marked as NDA in the database. The actual decision-level values and counting errors might be available on the report of the individual but are not available in the database until 2003.

An *in vivo* count spectrum is not analyzed for a fission/activation product radionuclide unless a peak 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 correlated to MDA or critical level calculations, especially with the broad peaks associated with scintillation-type 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.

**Cesium-137 and Other Intakes from Fallout**

Most workers in the early days of whole-body counting had detectable activities of $^{137}Cs$. Most of this was attributed to fallout. Some workers had even higher levels of $^{137}Cs$ from consumption of wild game. A decision level used to establish the difference between occupational and non-occupational sources of $^{137}Cs$ and other fallout radionuclide intakes has not been discovered in the records. In lieu of other information, the guidance in Table 5-18 can be used.

Table 5-17. Routine whole-body counting detection levels.[a]

| Period | Nuclide | $L_C$ nCi | MDA[b,c] nCi |
|---|---|---|---|
| 1955-1958[d] | Cs-137 | | 8 |
| | Sr-90[e] | | 30 |
| 1959-1970[d] | Cs-137 | | 4 |
| | Sr-90[e] | | 30 |
| 1971-1984 | Be-7 | 0.9 | 1.8 |
| | Cs-134 | 0.9 | 1.8 |
| | Cs-137 | 0.9 | 2.1 |
| | Co-57 | 2.1 | 4.8 |
| | Co-60 | 0.78 | 1.8 |
| 1985 to 1998 | Tl-202 | 0.5 | 0.9 |
| | C-11 (based on 511 keV)[f] | 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 | 6.0 |
| | 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 | 2.2 | 4.4 |
| | Ce-144 | 12.1 | 24.2 |
| | Cr-51 | 6.4 | 12.8 |
| | Co-57 | 1.4 | 2.8 |
| | Cu-67 | 1.5 | 3.0 |
| | 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 to present[g] | 511 keV | 1.15 | 2.3 |
| | Be-7 | 4.2 | 8.4 |
| | Ce-141 | 1.35 | 2.7 |
| | Ce-144 | 6.5 | 13.0 |
| | Co-56 | 0.55 | 1.1 |
| | Co-58 | 0.5 | 1.0 |
| | Co-60 | 0.45 | 0.9 |
| | Cr-51 | 4.45 | 8.9 |
| | Cs-134 | 0.5 | 1.0 |
| | Cs-137 | 0.6 | 1.2 |
| | Cu-67 | 1.5 | 3.0 |

Table 5-17 (Continued). Routine whole-body counting detection levels.[a]

| | | | |
|---|---|---|---|
| 1999 to present[g] (continued) | 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 | 41.0 |
| | Sb-124 | 0.45 | 0.9 |
| | Sc-46 | 0.7 | 1.4 |
| | Se-75 | 0.85 | 1.7 |
| | Tl-202 | 0.5 | 1.0 |
| | U-235 | 1.35 | 2.7 |
| | V-48 | 0.4 | 0.8 |
| | Zn-65 | 1.1 | 2.2 |
| | Zr-95 | 0.8 | 1.6 |

a. 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.
b. Based on 95% confidence of detection.
c. MDA = $L_C * 2$, unless otherwise specified
d. 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.
e. By *bremsstrahlung.*
f. 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.
g. Lower sensitivities might be available using the lung counter for certain nuclides if lung counting is appropriate to the dose reconstruction.

- The $^{137}$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 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 non-occupational source, the $^{137}$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 $^{137}$Cs for the United States for the years most likely to produce interference with occupational whole-body count results (NCRP 1987). Those values are listed in Table 5-18. If no other fission or activation products are linked to the intake and the $^{137}$Cs result is less than the values in Table 5-18, the $^{137}$Cs result can be assumed to be due to fallout.

Table 5-18 Mean body burdens of $^{137}$Cs from fallout in the United States (nCi).[a]

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

a. Source: NCRP (1987).

### 5.3.2 Lung Burdens

Lung burdens of $^{239}$Pu, $^{238}$ Pu, and $^{241}$Am were monitored using the Phoswich lung detectors beginning in 1970. The 59.5-keV gamma line of $^{241}$Am is used to determine the $^{241}$Am burden (50- to 70-keV region). If the isotopic ratio for a given intake is known, the $^{239}$Pu and $^{238}$ Pu can be determined from the $^{241}$Am. Otherwise, the plutonium is determined from the U-L X-ray region. When $^{241}$Am, $^{239}$Pu, and $^{238}$ Pu are present, corrections for the contribution of the Np-L X-rays, from the decay of $^{241}$Am, to the 14- to 25-keV $^{239}$Pu and $^{238}$Pu region must be considered. The Phoswich detectors were eventually replaced by arrays of HPGe detectors, greatly improving the energy resolution. Improved energy resolution permits the system to distinguish between gamma and X-ray lines that are closer together. However, because the U-L X-ray energies for the decay of $^{239}$Pu and $^{238}$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 the sensitivity level, varies for every count due to the effects of chest wall thickness on the attenuation of the 17-keV X-rays and the 59.5-keV gamma ray. Therefore, the MDA listed with the count, when available, should be used. The MDA and $L_C$ values listed in Table 5-19 are nominal and are based on the calibration chest wall thickness (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 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. **LASL (1977b) suggests that for large individuals, MDAs should be increased 50%**. 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 Attachment 5A, Table 5A-28.

The minimum intake detectable using *in vivo* counting for americium, plutonium, or uranium is much larger than the minimum intake detectable through *in vitro* methods. Results or detection limits for *in vivo* methods should typically be used only to bound intakes determined from *in vitro* results of detection levels and not as the primary source of determination of intake or missed dose for these nuclides. 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

Table 5-19. MDA $L_C$ values for lung counting (nCi).

| Period | Radionuclide | $L_C$ | MDA[a] |
|---|---|---|---|
| Extended count time 1977[b] | Am-241 | | 0.3 |
| | Pu-238 | | 10 |
| | Pu-239 | | 21 |
| 1980 -1984[c] (Ennis 2003)[d] (1970 – 1979)[e] | Am-241 | 0.155 | 0.31 |
| | Pu-238 | 11 | 22 |
| | Pu-239 | 24 | 48 |
| 1984 (Vasilik et al. 1984)[f] | Am-241 | 0.16 | 0.32 |
| | Pu-238 | 14 | 28 |
| | Pu-239 | 30 | 60 |
| 1998[g] to present (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 to 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 (LASL 1977b).
c. Might be applicable to the startup of the Phoswich system in 1970, no other information available.
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 = $L_C * 2$
g. Lung counter has 10-300 keV and 80-3000 keV ranges, so a lower sensitivity for certain fission and activation products can be obtained.

more than 50 years. None of these individuals has had activity above background using *in vivo* techniques (Voelz et al. 1979; Voelz, Grier, and Hempelmann 1985) except one individual with a positive lung count for $^{241}$Am 37 years later. This individual was suspected of an additional intake of plutonium containing $^{241}$Am in 1957.

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

Lung counts for follow-up on members of the UPPU Club were typically longer (60 minutes rather than 15 or 20 minutes) 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. (Rule of thumb is quadruple the count time half the MDA.)

Lung counting can be useful in determining actual lung burdens when high-fired oxides of plutonium are involved. The extreme insolubility of these oxides has been described at several DOE sites including LANL (Hammond, Lagerquist, and Mann 1968). These oxides typically exhibit a lower than expected urine excretion rate, and the total lung burden might not be adequately predicted using standard excretion equations. The dose reconstructor should use best judgment on forcing the data to fit a type S model or decreasing the absorption rate constants in the lung model. High-fired oxides involving $^{238}$Pu from heat source production have been encountered at LANL (Miller et al. 1999).

Results in bioassay records should be assumed to be reported in nCi 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 decision-level 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 $^{239}$Pu and $^{241}$Am. In recent years, routine $^{235}$U and $^{234}$Th (as $^{238}$U) have been 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 contaminated with plutonium. This probe was capable of detecting soft plutonium X-rays. The sensitivity of this probe was $1 \times 10^{-9}$ Ci of plutonium unshielded. This was equivalent to detection of one-tenth of a permissible body burden of plutonium embedded in tissue to a depth of 1 cm (LASL 1959a). In 1977, a new NaI detector (12 mm x 2 mm) was being evaluated. This produced an MDA of 0.07 nCi based on WGPu (LASL 1977c).

Wound counting was used primarily as a tool for surgeons to locate plutonium in the wound, not as results used to calculate internal dose. Wound monitoring continues to be performed. In most cases, intake and dose will not be assessed directly from the wound count but rather from the resultant urine bioassay data. Follow-up studies of wounds found that plutonium does not readily migrate from the wound site to uptake in the majority of the incidents (Voelz and Lawrence 1991). No other information on instrumentation or sensitivities is available.

## 5.4 INTERFERENCES AND UNCERTAINTIES

### 5.4.1 Contamination

Until the Health Pass Ward was established in 1945, the potential existed for sample contamination. It is likely that a contaminated sample will show up as an obvious outlier in the dataset for a given worker. The use of a result from a contaminated sample could result in an overestimate of the workers dose; however, the sample result should be considered real if other data do not demonstrate the sample to be a false positive result. Some variability is expected in any set of results. See Section 5A.9, Attachment 5A, for an example of a valid series of bioassay results for an individual whose autopsy results verified plutonium intake(s).

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 no detectable activity on the same day or within a few days, external contamination can be suspected. Radon progeny and medical diagnostic or therapeutic procedures involving radionuclides can cause interference to *in vivo* measurements, especially for sodium iodide (NaI) detectors. However, unless the count was invalidated or noted as being influenced by such interferences, the results should be used as recorded.

### 5.4.2 Uncertainties

**_In Vivo_ Counting**

Uncertainties for bioassay measurements might be included with the results for excreta or *in vivo* measurements. For *in vivo* results, uncertainties were not reported in the database until 2003; however, uncertainties might be listed on individual reports prior to 2003. The listed uncertainties are typically reported as one standard deviation.

Uncertainties associated with chest counting are reduced by use of different calibrations for different chest wall thicknesses and use of ultrasound to measure chest wall thickness. A 1-sigma uncertainty of about 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 and/or liver. The uncertainty in lung activity estimates affected by contributions from activity in the liver and skeleton would likely 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%. The mathematical correction was made for routine counts. Special counts and counts with positive results were typically corrected using ultrasound chest wall thickness measurements.

Based on the above discussion, the assumption provided in the *Internal Dose Reconstruction Implementation Guidelines* (NIOSH 2002) is adequate and should be used; namely, the standard deviation is 0.3 times the MDA or reporting level, with the exception of chest counts, for which 0.5 times the MDA should be used. For results greater than 3 times the MDA or reporting level, the standard deviation can be assumed to be 0.1 times the result, based on Currie's quantification level (Currie 1968). Actual tests for *in vivo* counts of phantoms show even smaller uncertainty, but 0.1 is appropriate for broad applications. If actual standard deviations or other indications of uncertainty are reported with a bioassay measurement result, the reported value should be used.

**_In Vitro_ Measurements**

Uncertainties used for both normal and lognormal distributions are listed in the database for plutonium and americium samples. The measurement uncertainty at one standard deviation and the

biovariability are listed with the results. 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. A study of the variations found in plutonium urinary data (Moss et al. 1969) determined that the LASL employee results exhibited as lognormal distribution with a geometric standard deviation of 1.9 calculated from the ratio of the 84th percentile to the 50th percentile.

The tritium database lists the measurement uncertainty at one standard deviation with the bioassay results. It is likely that the uranium database will also list the uncertainty of each measurement at one standard deviation.

**Chelation Therapy**
Chelation therapy is used only in circumstances where significant doses are expected if the material is allowed to follow natural bioelimination kinetics. Fewer than 35 individuals have undergone chelation therapy at LANL for americium or plutonium intakes. The urine bioassay results directly associated with the chelation therapy period are coded with a "C" in the database (see Attachment 5A, Section 5A.1.1). The chelation process causes the rate of bioelimination of the plutonium or americium to increase. Therefore, the results of urine bioassay samples taken during treatment and for at least 100 days following the last treatment, which could last up to 100 days, should not be used to determine the intake. The intake should be evaluated based on later urine bioassay results, if available. Other considerations might have to be applied. The dose reconstructor should thoroughly investigate the dose and medical records when chelation therapy is noted.

## 5.5 AIR CONCENTRATION IN SELECTED BUILDINGS

**Average Airborne Concentration Levels**
Chronic intakes, or frequent intermittent intakes, can be modeled as chronic. Very low-level, frequent, intermittent intakes might have occurred for the highest risk workers, but the intakes were below detectability at the time. For radionuclides with long residence times in the body, chronic intakes lead to a slow buildup of activity in the body and a concomitant increase in urinary excretion. Studies have shown that room air concentration is not necessarily a reliable quantitative predictor of intake because of the variations in respiratory conditions and particle dispersion within an area. Even breathing zone (BZA) samples are not always an accurate predictor of the amount of intake (Whicker 2004). However, maximum and average airborne concentrations can be helpful in establishing boundary conditions for intakes. Maximum and average airborne contamination levels in buildings with high exposure potential are listed in Table 5-20. Airborne concentrations were not available for all years of operation, especially for fission products. Average concentrations are calculated from general air samples as simple averages or averages obtained directly from LASL reports. Blanks in the table indicate that no information is currently available for that period.

### 5.5.1 Respiratory Protection Program

The first reference to the need for respiratory protection for LASL workers is in a memo requesting "assault-type gas masks for testing in connection with toxic effects of X10 (plutonium) dust" (Oppenheimer 1944). The May 31, 1944, "Health Safety Rules" for Buildings H and V-8 specify 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 1944c). Positive pressure masks were being introduced into the program at that time. Dry boxes and respirators were a satisfactory method of handling dry material; however, individuals working with solutions consistently received the highest exposure despite all precautions.

Table 5-20. Summary of airborne concentrations from selected buildings.

| Building | Nuclide[a,b] | Year | Maximum airborne concentration[c] dpm/m$^3$ | Average airborne concentration[c] dpm/m$^3$ |
|---|---|---|---|---|
| D | Alpha | 1945 | 674.0 | |
| D | Alpha U | 1945 | 2,564.400 | |
| H | Alpha Po | 1945 | 48.00 | |
| DP East | Alpha (EU) | 1945 | 2,458.0 | 68.2 |
| DP West | Alpha | 1945 | 254.0 | |
| D | Alpha (Pu) | 1945 | 480.0 | 151.0 |
| D | Alpha (Pu) | 1946 | 2,590.0 | 46.86 |
| DP East | Alpha (EU) | 1946 | 147,400.0 (Po) | 271.27 |
| TU Building | Alpha | 1946 | 1,958.400 | 43.400 |
| DP West | Alpha | 1946 | 2,400.0 | |
| TU Building | Alpha | 1947 | 515.000 | |
| H | Alpha Po | 1947 | | 8.80 |
| Sigma | Total alpha U | 1947 | 187.20 | |
| D | U | 1947 | 55.00 | |
| D | Pu | 1947 | 134.00 | |
| D | Cold lab alpha | 1947 | 15.0 | |
| Sigma | U | 1948 | 1,393.80 | 124.60 |
| U | Pu | 1948 | 379.40 | 66.67 |
| H | Po | 1948 | 52.80 | 4.50 |
| D | U | 1948 | 2,564.40 | 21.23 |
| D | Pu | 1948 | 860.80 | 11.53 |
| D | Cold lab alpha | 1948 | 882.4 (normally 35.8) | 5.675 |
| Am Lab | Am alpha | 1948 | 6.2 | 1.6 |
| Sigma | Total alpha U | 1948 | 1,393.80 | 124.60 |
| DP West | Alpha | 1949 | 1,694.0 | |
| Am Lab | Am alpha | 1949 | 18.0 | |
| Press Rm | Alpha | 1949 | | 198.0 |
| TU Bldg Furnace | Alpha | 1949 | | 4,095.0 |
| Waste Treatment Lab | Alpha | 1949 | 13.8 | |
| M | Alpha | 1949 | 580.8 | 84.0 |
| DP East | Polonium | 1949 | 2,344.0 | |
| U | Alpha | 1949 | 37.2 | |
| D | Pu | 1952 | | 2.4 |
| CMR | Pu | 1952 | | 1.9 |
| M | U | 1952 | | 4.6 |
| D | Pu | 1953 | 600.4 | 3.2 |
| CMR | Pu | 1953 | 388.7 | 1.0 |
| U | Pu | 1953 | 4.5 | 1.4 |
| M | U | 1953 | 210.6 | 2.4 |
| D[d] | U | 1953 | 1,969.3 | 4.3 |
| CMR | U | 1953 | 21.1 | 1.2 |
| CMR | Pu | 1954 | 2,851.3 | 1.3 |
| CMR | Pu | 1955 | 527.2 | 0.7 |
| CMR | Pu | 1956 | 162.7 | 0.7 |
| CMR | Pu | 1957 | 351.0 | 0.8 |
| CMR | Pu | 1958 | 1,370.2 | 1.0 |
| CMR | Pu | 1959 | 6,712.9 | 1.0 |
| CMR | Pu | 1960 | 518.1 | 0.9 |
| CMR | Pu | 1961 | 426.0 | 1.0 |
| CMR | Pu | 1962 | 4,680.0 | 1.0 |
| CMR | Pu | 1963 | 166.0 | 1.0 |
| CMR | Pu | 1963 | 4.0 | 0.0 |

Table 5-20 (Continued). Summary of airborne concentrations from selected buildings.

| Building | Nuclide[a,b] | Year | Maximum airborne concentration[c] dpm/m$^3$ | Average airborne concentration[c] dpm/m$^3$ |
|---|---|---|---|---|
| CMR | Pu | 1967 | 285.0 | 1.0 |
| CMR | Pu | 1968 | 5,370.0 | 1.0 |
| CMR | Pu | 1968 | 11.0 | 0.0 |
| CMR | Pu | 1969 | 685.0 | 1.0 |
| CMR | Pu | 1969 | 11.0 | 0.3 |
| CMR | U | 1954 | 77.4 | 0.8 |
| CMR | U | 1955 | 112.1 | 0.7 |
| CMR | U | 1956 | 67.8 | 1.0 |
| CMR | U | 1957 | 2,231.1 | 1.4 |
| CMR | U | 1958 | 51.3 | 1.3 |
| CMR | U | 1959 | 24.0 | 1.2 |
| CMR | U | 1960 | 94.1 | 1.2 |
| CMR | U | 1961 | 35.0 | 1.0 |
| CMR | U | 1962 | 43.0 | 1.0 |
| CMR | U | 1963 | 65.0 | 1.0 |
| CMR | U | 1963 | 11.0 | 0.0 |
| CMR | U | 1967 | 80.0 | 1.0 |
| CMR | U | 1968 | 53.0 | 1.0 |
| CMR | U | 1968 | 14.0 | 0.6 |
| CMR | U | 1969 | 65.0 | 1.0 |
| CMR | U | 1969 | 258.0 | 2.4 |
| Laundry | Total alpha | 1948 | 844 | 450.533 |
| Laundry | Total alpha | 1949 | 2,268 | 583.067 |
| Laundry | Total alpha | 1950 | 78 | 42.2857 |
| TA-21[e] | I-131 | 1961 | 1.4E4 | NA |
| DP West | I-131 | 1964 | 302 | 5.5 |
| Sigma | Beta -gamma[f] | 1947 | 1.82E+06 | |
| H | Beta -gamma[f] | 1947 | | 0.0 |
| U | Beta -gamma[f] | 1947 | | 5.95E+05 |
| Sigma | Beta -gamma[f] | 1948 | 1.86E+09 | 8.21E+07 |
| H | Beta -gamma[f] | 1948 | 3.33E+08 | 7.39E+07 |
| U | Beta -gamma[f] | 1948 | 1.47E+09 | 1.95E+08 |
| CMR | Mixed fission[f] | 1961 | 78.0 | 12.8 |
| CMR | Mixed fission[f] | 1962 | 11,627.0[g] | 19.7[g] |
| CMR | Mixed fission[f] | 1963 | 518.0 | 15.0 |
| CMR | Mixed fission[f] | 1964 | 19,256.0[h] | 39.0[h] |
| CMR | Mixed fission[f] | 1965 | 13,404.0[i] | 22.0[i] |
| CMR | Mixed fission[f] | 1966 | 93,887.0[j] | 366.7[j] |
| CMR | Mixed fission[f] | 1967 | 19,256.0 | 39.0 |
| CMR | Mixed fission[f] | 1968 | 13,404.0 | 22.0 |
| CMR | Mixed fission[f] | 1969 | 93,887.0 | 366.7 |
| CMR | Mixed fission[f] | 1970 | 14,163 | |
| CMR | Mixed fission[f] | 1971 | 18,104 | |

a. Guidance on adjusting for different enrichments of uranium or mixtures of plutonium is in Section 5.2.
b. Information on specific mixtures of fission products is not readily available from LANL Guidance.
c. Results prior to January 1, 1953, were reported as cpm/L. Listed results have been converted to dpm/m$^3$ assuming a nominal counting efficiency of 50%.
d. D Building demolished in 1954.
e. Dummer (1961).
f. Mixed Fission Products (assume gross beta/gamma) tolerance is $1 \times 10^{-7}$ µCi/cc ($2.22 \times 10^5$ dpm/m$^3$) through 1948; MAC is $6.7 \times 10^3$ dpm/m$^3$ from 1961 to 1969.
g. Highest concentration of 11,627 dpm/m$^3$ is in "uranium cell corridor - no personnel exposure." Next highest concentration is 207 dpm/m$^3$. Average concentration would be 11.8 dpm/m$^3$.
h. "No personnel exposure" to highest concentration of 19,256 dpm/m$^3$. Next highest concentration is 2,551dpm/m$^3$.

Table 5-20 (Continued). Summary of airborne concentrations from selected buildings.

i. "No personnel exposure" to highest concentration of 13,404 dpm/$m^3$. Next highest concentration is 2,233 dpm/$m^3$.
j. "No personnel exposure" to highest/next highest concentrations of 93,887 and 45,997 dpm/$m^3$. Next highest concentration is 21,553 dpm/$m^3$.

Respiratory protective devices were provided during the Manhattan Project by the U.S. Army Chemical Warfare Laboratories. The M-9 mask with either the M-11 canister for gases and particulates or the M-14 canister for particulates was used beginning in 1946. The efficiency of the canister was 1 in 100,000 particles of dioctyl phthalate (DOP; AIHA 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 wherever 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.

The Industrial Hygiene group at LASL continued to be at the forefront of development of respiratory devices and programs. Respiratory protection programs are documented beginning with Dummer (1958). The programs include 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 5-21. The protection factors appear to remain consistent throughout the history of the program within the exceptions noted.

Table 5-21. Respiratory protection apparatus

| Equipment | Protection factor | Remarks |
|---|---|---|
| Half-mask | Not more than 10x MPC[a] | b, c |
| Full-face mask | Not more than 50x MPC (Dummer 1958)[d] | b |
| Full-face mask | Not more than 100x MPC (Healy 1970) (LANL 1984)[e] | b |
| Full-face mask with fit field tested | Not more than 1000x 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 (IDLH)
c. Half-face respirators were not permitted to be used in airborne highly toxic dust such as plutonium (LASL 1964). LANL (2004a) states 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 98-99% efficient.
e. Powered air-purifying respirators not more than 3,000 times with high-efficiency 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 (CAM) in the area.

**Continuous Air Monitors**

CAM alarms have been historically used as an indication that room air concentration had changed and that respiratory protection was required. Whicker et al. (1997) found that the selection of the location of the CAM was critical to the reliability of the response. The typical location of CAMs at the ventilation exhaust point of a room was not considered optimal. Results also suggest that when a worker causes the release and is at or near the release point, the worker could be exposed for a significant period before a radioactive cloud reaches the CAM.

The above limitations suggest that, without other interventions, the possibility exists that workers could be exposed to intakes that did not trigger alarms. However, after 1970 when the bioassay programs were well-established, the majority of workers with the potential for monitored and unmonitored intakes are expected to have participated in a bioassay program.

### 5.5.2 Maximizing and Best Estimate Intake Parameters

Intake parameters can be derived from airborne contamination levels for buildings with the highest exposure potential or highest intakes for various periods. Maximum and average airborne contamination levels for selected buildings are listed in Table 5-20. Incidents and intakes are listed in Table 5-22. Average airborne contamination levels either are derived as simple averages or are reported as averages listed in LASL reports. Simple averaging is assumed for LASL reports, although no information on the methods used to obtain these reported averages is available.

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 5-22; Table 5-23 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.

Table 5-24 lists maximum airborne concentrations for selected buildings during various intervals. In practice, individuals in areas of maximum airborne concentration would most likely have been wearing appropriate respiratory protection.

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 maximum permissible body burden 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 exposure of personnel had to be capable of detecting from 2 to 14 dpm plutonium in a 24-hr 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. The alpha MAC values used for selected buildings are listed in Table 5-24. Assume that the period for the listed MAC continues until changed. The MACs are dpm for total gross alpha with the primary nuclide for the area listed, except as noted. MACs were eventually replaced by Maximum Permissible Concentrations (MPCs) and Derived Air Concentrations (DACs) as controlling values. Los Alamos Action Levels (ALs) are also listed where applicable. It appears that at some time prior to 1964 the MAC values were changed to Action Levels with larger, more specific MAC values established for nuclides, enrichments, and mixtures. Table 5-25 includes an example of the Action Levels and MACs for selected buildings in 1964.

In 1948, the tolerance level for beta-gamma emitters in air was $1 \times 10^{-7}$ µCi/cc ($2.22 \times 10^{5}$ dpm/m$^3$). In December 1961, the MAC for mixed fission products was established in CMR Building, Wing 9 at 6.7

Table 5-22. Reported exposure incidents and results .

| Date | Incident |
|---|---|
| 1944–1946 | 26 workers received intakes estimated to be 6 to 80 nCi while involved in Pu 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 Pu salts in 1945 (Hempelmann and Langham 1953). |
| 1944 | Periodic overexposure of individuals working on RaLa project occurred for 6 months at start of project until “bugs” were worked out of remote chemical handling procedures. |
| 1944 | Highest nasal swipe for May was 11,372 cpm. (assume alpha) |
| 1945 | August - Building 52 handled large amounts of Po. Note at bottom of 52 air concentration data table stated: "Note: 0.75 c/m/L is used as 2-year tolerance value assuming 100% retention by lungs." |
| 1945 | 4 persons exceeded “safe” amount of Pu in body, 1 µg. |
| 1945 | Tolerance value for Po in urine samples 1,500 cpm/day (assume 50% efficiency for counter), exceeded by only 2 individuals. All persons working with Po were monitored. |
| 1945 | At the Water Boiler at Omega several instances of mild to moderate overexposure when gas exhaust lines leaked or during decontamination of active material. |
| 1948 | 11/2 - worker from GMX ingested sufficient amount of RaLa to give a reading on detector. |
| 1948 | 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 1949). |
| 1949-1951 | Number of airborne contamination results above Pu 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% of swipes in 1949 to 15% in 1951 for DP West and 60% to 15% for Tech Area (LASL 1951). |
| 1951 | 1,876 Pu urine samples below MAC and 492 Po urine samples from DP East were below tolerance. All routine tritium urine samples were below MAC (LASL 1951). |
| 1951 | DP West dismantle 408 and 413, high airborne plutonium exposures were kept to minimum. |
| 1951 | DP East experimental program with Ac. No overexposure; however, adequate urine analysis is not available, air samples difficult to evaluate.(LASL 1951) |
| 1951 | Pajarito Warehouse contaminated by leaking Po source. TA-33, Area 6 required cleanup from Po spill. |
| 1951 | Ruptured Pu slug at GT site vault. Contamination spread to main building. |
| 1951 | Airborne alpha activity in Room 513 DP West was consistently above MAC. Source could not be localized; however, cleaning attic and sealing openings into room decreased airborne levels to satisfactory (LASL 1951). MAC total alpha plutonium as of April 1951was 0.0044 cpm/L. |
| 1951 | Kilocurie quantities of tritium handled in CMR areas. “Health program for this work has been satisfactory.” |
| 1950’s late | Radioiodine Experiments: In late 1950s, Los Alamos biomedical researchers orally administered I-125 and -131 to group of 19 subjects. |
| 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.” |
| 8/1959 | Three employees > MPL, 11 > 50% MPL, 90% of people with ≤10% MPL for Pu (0.04 µCi) of 1,325 monitored (LASL 1959a) |
| 4/1959 | Radioactive gas leak at LAPRE II resulted in thyroid uptakes by at least 5 workers of I-131, I-133, and I-135 resulting in thyroid burdens equivalent to approximately 3 µCi I-131 (Van Dilla 1959). |
| 1959 | Laboratory air sample 3.8 dpm/, 16% of MPC for 1-week sample, highest value 2/1959 to 7/1959 (LASL 1959). |
| 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 d. (ICRP 1979; Miller et al. 1999), half-time for ICRP 30 model is 10,000 d, AMAD is 0.2µ. Intakes 2,150 to 210 nCi, found to fit other Wing 9 intakes. |
| 1972 - 1973 | Demolition of Bldg. 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 | February - one employee received Pu intake estimated at 1 MPBB (0.04 µCi) (LASL 1977a,d). |
| | January - oxide of Pu caught fire, airborne 4.5 to 11.65 dpm/$m^3$, no positive nasal swipes, no bioassay (LASL 1977b). |
| | H-3 urine bioassay following 1/77 incident at LAMPF < 1 µCi/L (LASL 1977a) |
| | February 10 incident resulted in <16 nCi lung burdens for five employees, one employee received DTPA chelation therapy. See Table 5-23 for details (LASL 1977b). Urine sample Pu intake estimates are 1 nCi (one worker), 4 nCi (two workers), 9 nCi (one worker). |
| | U-235 airborne concentrations of >1,500 dpm/$m^3$ and >25,000 dpm/$m^3$ for DP Bldg 4, Room 412 5/77 (LASL 1977b). |
| | June - Tritium in urine 101 µCi/L following airborne release DP, Bldg 5. |
| | June - Alpha contamination (up to 100,000 cpm) from damaged uranium target found on several persons at LAMPF switchyard (LASL 1977d). |
| | May - TA-35 and TA-41 alpha contamination escaped from pressure vessel, caused personnel contamination (LASL 1977d). |
| | 20,000 dpm alpha airborne (U-235 and U-238 operations). |
| | November - Tritium bioassay (confirmed) of P-9 employee 798 µCi/L. (LASL 1977c). |
| 1978 | $UF_6$ tube ruptured (5-10 g) spreading HF fumes and depleted U-238. (LASL 1978a). |
| | Operator replacing leaking hose at TA-33 stack had tritium bioassay of 4 µCi/L; 449 Ci HT released (LASL 1978b). |

Table 5-22 (Continued). Reported exposure incidents and results .

| Date | Incident |
|---|---|
| 1979 | Zia employee with nasal swipe of 553/70 dpm TA-21(LASL 1980a). |
| 1980 | TA-33 tritium exposure to employees. |
| | 54-µCi/L tritium bioassay TA-33 (LASL 1980b). |
| | 153-µCi/L tritium bioassay TA-33.(LASL 1980c). |
| 1983 | Pu spill in TA-55, 10 persons, no significant exposures (LASL 1983a). |
| | High airborne contamination results in high nasal swipes TA-55 Pu (LANL 1983b). |
| | 21 Pu 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; 1 wound above 20 nCi, numerous positive (>50 dpm) nasal wipes (LANL 1983b) |

Table 5-23. Summary of February 10, 1977, plutonium incident (LASL 1977d).

| Worker | Total systemic burden (increase) | | Nasal swipe (L/R) dpm | Chest burden[a] Pu nCi |
|---|---|---|---|---|
| | Pu-239 nCi | Pu-238 nCi | Total alpha | |
| Technician 1 | 8.2 (3.3) | 1.7 (0.1) | 12759/13385[b] | 7.5 |
| Technician 2 | 7.6 (5.8) | 0.7 (0.5) | 801/1849 | 13. |
| Supervisor 2 | 42.9 (7.5) | 3.2 (0.6) | 1091/1193 | 6. |
| Technician 3 | 21.8 (18.0) | 1.1 (0.4) | 611/1193 | 10. |
| HP Surveyor | 4.2 (0.3) | 0.7 (0.0) | 226/577 | 0 |

a. No worker had chest counts above minimum detectable activity (16 nCi)
b. Received DTPA chelation therapy

$\times 10^3$ dpm/m$^3$. In January 1964, the MAC (DP West, Room 401) for $^{131}$I was $2.9 \times 10^4$ dpm/m$^3$. In January 1968, a MAC for $^{131}$I was established at $6.7 \times 10^3$ dpm/m$^3$.

## 5.6 UNMONITORED WORKERS

The potential effects of exposure to plutonium were recognized early by its discoverer, Glen Seaborg (Hempelmann, Richmond, and Voelz 1973). Therefore, when the first few micrograms of plutonium arrived at LANL in 1943 there was an awareness 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). 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.

However, 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 were exceeded, not whether there was an intake. 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 was constructed. Even hallways and other non-restricted areas had contamination. In 1944, shoe covers worn by secretaries and others working throughout the buildings had significant count rates of 2,500 to 7,500 cpm (LASL 1944a). Contamination above 500 cpm (0.007 µg or 0.0004 µCi 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 -1946) for any site worker. Bioassay was provided only for those workers who were most exposed. However, because the

Table 5-24. Summary of maximum allowable concentrations (MAC) for selected areas (dpm/$m^3$).

| | TA-1 | | | | | | TA-3 | | TA-21 | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | (dpm/$m^3$) | | | | | | (dpm/$m^3$) | | (dpm/$m^3$) | | | |
| **Year** | **U Bldg U Labs** | **D Bldg EU Labs** | **D Bldg Pu/U Labs** | **TU Bldg normal U** | **Sigma Bldg U/EU Labs** | **M Bldg EU Labs** | **CMR U and EU** | **CMR Pu Labs and Waste Treatment/ Laundry** | **DP West EU** | **DP West Pu and Am CMR-4** | **DP East U Bldg 54** | **DP East Po** |
| 1945 | | | 70 | 1,500 Po | | | | | | | | |
| 1947 | | | 70 | | 400 | | | | | | | |
| 1950 | | | 70 | 400 | 400 | 400 | | | | | | |
| 8/1950 | | 68 | | 68 | 68 | 68 | | | | | | |
| 9/1950 | | | | | | | | 70; Pu, Am, Po, U | 68 | 70 | 68 | 1,400 Po |
| 4/1951 | | 8.8 | 8 | 66 | 66 | 66 | | 8.8 | 8.8 | 8.8 | | 1,400 Po |
| 5/1951 | | 8.8 | 8.8 | | | 8.8 | | | | | | |
| 10/1951 | | 66 | | | | 66 | | | | 8.8 | | |
| 4/1952 | 66 U<br>8.8 Pu | | | | | | 66 | 8.8 | | | | |
| 1961 | | | | | | | 66 $^{235}U$[a] | | | | | |
| 1963 | | | | | | | | 9.0 | | | | |
| 1964 | | | | | | | 66 (AL) | | | | | |
| 1967 | | | | | | | | 4.0 | | | | |

a. Wing 9.

Table 5-25. MAC and Action Levels for 1964 (Meyer 1964).

| Nuclide | Action level ($dpm/m^3$) | DP West ($dpm/m^3$) | CMR ($dpm/m^3$) | Sigma (TA-3) ($dpm/m^3$) | Shop 13 ($dpm/m^3$) |
|---|---|---|---|---|---|
| Natural U (sol) | 66 | | 154.0 | 154.0 | 154.0 |
| Natural U (insol) | 66 | | 132.0 | 132.0 | 132.0 |
| EU (sol) | 66 | | 1,100.0 | | |
| EU (insol) | 66 | | 220.0 | | |
| U-233 (sol) | 66 | | 1,100.0 | | |
| U-233 (insol) | 66 | | 220.0 | | |
| I-131 | NA | $2 \times 10^4$ | | | |
| Fission products | NA | | $6.7 \times 10^3$ | | |
| Am-241 (sol) | 8.8 | 13.2 | | | |
| Am-241 (insol) | 8.8 | 220.0 | | | |
| Pu-239 (sol) | 8.8 | 4.4 | | | |
| Pu-239 (insol) | 8.8 | 88.0 | | | |
| Cm -244 (sol) | 8.8 | 19.8 | | | |
| Cm-244 (insol) | 8.8 | 220.0 | | | |

excretion of plutonium is continuous following 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 received a large intake of radioactive material that was unmonitored and unnoticed is less after 1946. However, the probability of unmonitored small intakes is larger. Periodic reports from H Division of air samples, contamination incidents, and "hot spots" continue to report 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 CAM alarmed or airborne levels approached tolerance or action levels. Thus, a potential existed for an intake prior to the alarm. Review of a *Summary of Radiological Incident Reports (RIR) through June 1998* indicates, that while the number of RIRs had decreased significantly, the potential for unmonitored small intakes continues to exist. (LANL 1998)

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

As late as 1977, it is reported that releasable and non-releasable (contaminated) salvage materials were accessible to "scavenger personnel" who frequented these bins and possibly mixed the types of scrap while it was waiting for pickup (LASL 1977d). A fence was installed to prevent uncontrolled access when an incident resulted in contaminated scrap leaving the area on a truck.

The formal protocol for placing workers in bioassay programs was in place in 1970 with the Health Physics Checklist. Prior to the Health Physics Checklist, a person was placed in a bioassay program at the discretion of the health physics monitor in charge of the area. While these protocols ensured that most workers who required monitoring were monitored, some workers might not have been included. In addition, there is a possibility that the checklist might not have been updated when a worker's job title or responsibilities changed.

Over the history of the Laboratory, the health physics program has always been at the forefront of development of instruments, analysis procedures, and guidance for radiation protection.

No bioassay data, except plutonium and americium urine and fecal analysis, have been submitted to date. Therefore, the experiences of workers with monitored intakes cannot be used to develop a scenario for workers who were unmonitored. Potential intakes will be derived from removable contamination levels, tolerance and MAC air concentration levels, and airborne concentrations of significant radionuclides.

### 5.6.1 Workers Pre-1947

**Plutonium, Uranium, and Polonium**

Until the new DP laboratory facilities were built in 1946, contamination levels were significant. Stop work and decontamination of the area was required when alpha contamination exceeded 10,000 cpm (assume ~50% counting efficiency). Decontamination was recommended between 2,000 and 10,000 cpm, but was not required. Bioassay was not available before late 1944 and not completely established until 1945. If a worker had potential for plutonium exposure from 1943 through 1946, with no bioassay during that period but plutonium bioassay during later years, dose reconstructors could use the later plutonium bioassay with claimant-favorable assumptions to bound intakes. However, if the worker did not participate in a plutonium bioassay program after this period, Table 5-26 contains plutonium intakes that can be assigned. Although the potential of contamination in 1943 was much less than in the following years, an intake equal to 10% of the 1945 intakes is being assigned.

Table 5-26. Hypothetical chronic intakes for plutonium, 1943-1946.

| Period | Total Pu alpha | | Comments |
|---|---|---|---|
| | Worker dpm/day | Casual dpm/day | |
| 8/1943-12/31/1944 | 9.92 | 0.992 | 1% and 0.1% of 1945 intakes[a] |
| 1/1/1945 – 12/31/1945 | 992.0 | 99.2 | |
| 1/1/1946 – 12/31/1946 | 308.8 | 30.88 | |

a. Based on comparison of total plutonium material at the lab in 1943 (micrograms), 1944 (milligrams), and 1945 (kilograms).

For unmonitored workers potentially exposed to plutonium, uranium, or polonium from 1943 through 1946, hypothetical intakes were constructed based on the listed parameters. Nuclides should be selected according to available information on work locations or job descriptions. It is expected that most workers would not have worked with all nuclides simultaneously. Resultant dose should be reconstructed as a lognormal distribution with a GSD of 5. The GSD has been increased above the GSD of 3 attributed to biokinetics because of the additional uncertainty attributed to air sampling.

Intakes are derived assuming a breathing rate of 2,400 $m^3$ per working year for persons performing work in the area and 10% of the rate for persons casually entering the areas.

**Plutonium**

A limited number of air sample results were available for 1945 and 1946. These air sample data are consistent with normal resuspension factors (of $1 \times 10^{-6}$/cm to $1 \times 10^{-5}$/cm) if surfaces were slightly above or below the 0.004-nCi contamination criterion. Intakes are divided into three periods because of the amounts of plutonium present at the Laboratory. Plutonium mixtures during these years should be assumed as 3%, fresh. Intakes are based on average air sample results as listed in Table 5-20 for Building D, which was a Pu building. No respiratory protection is assumed, although respiratory protection was available for these years. Table 5-26 lists the hypothetical intakes for workers and casual encounters with plutonium 1943 through 1946.

For hypothetical acute intakes, a 4-hour exposure at the maximum air concentration for the applicable building can be assumed from Table 5-20.

**Polonium**

Average air sample data from DP East for 1945 and 1946 are used to estimate potential intakes for $^{210}$Po. No average concentration data are available for H Building. The average values are representative of the potential exposure if the individual was not involved in an incident. If the information indicates that the person might have been involved in an area of high airborne contamination, the maximum air concentrations listed in Table 5-20 for 1945 and 1946 would result in hypothetical acute intakes of $^{210}$Po for a hypothetical 4-hr period of exposure, without respiratory protection, of 12,000 dpm and 710,000 dpm, respectively.

For general application for unmonitored individuals the intakes listed in Table 5-27 can be assumed. Intakes of polonium do not exhibit a long-term excretion pattern compared to plutonium intakes; therefore, monitoring for these early intakes at a later time is not possible.

Table 5-27. Hypothetical intakes for polonium and uranium, 1943-1946.

| | Total alpha activity[a] | | |
|---|---|---|---|
| | Worker dpm/day | Casual dpm/day | Comments |
| 1943-12/31/1944 | 74.25 | 7.425 | Assumed from 1945 concentrations |
| 1/1/1945 – 12/31/1945 | 74.25 | 7.425 | |
| 1/1/1946 – 12/31/1946 | 149.16 | 14.92 | |
| 1/1/1947 – 12/31/1949 | 13.95 | 1.40 | For uranium only |

a. Can be interpreted as either Po-210 or U-234 as benefits the claimant, with the exception of 1947-1949.

Polonium was used in initiators, utilizing the ($\alpha$,n) reaction of $^{210}$Po and $^{9}$Be. In February 1945, the schedule for polonium delivery from Monsanto to TA-1 was increased from a few curies to 100 Ci per month by June and 500 Ci per month by December. At TA-1, polonium and radium were handled in H Building and Gamma Building. H Building was used for preparation of neutron and alpha sources for initiators and isotopic experiments. Workers involved in these operations were part of Group CM-15. Operations in H Building were carried out between 1943 to July 1945 when operations were moved to the DP Site. Polonium ($^{210}$Po) 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.

Absorption type F or M might be appropriate.

**Uranium**

Plutonium, uranium, and polonium were the most significant nuclides encountered during the 1943-1946 period. No bioassay was available until at least 1949; therefore, a hypothetical intake rate has been calculated for uranium until 1949. The average 1945 and 1946 air sample data from the DP East site, which was an enriched uranium facility as well as a polonium facility and D building, was used to construct the hypothetical intake for enriched uranium listed in Table 5-27. The average values are representative of the potential exposure if the individual was not involved in an incident. If the claimant interview indicated that the person might have been involved in an area of high airborne contamination, the maximum air concentrations listed in Table 5-20 for 1945 and 1948 would result in a hypothetical intake of $^{234}$U for a hypothetical 4-hour period of exposure, without respiratory protection, of 12,000 dpm and 670 dpm, respectively.

Absorption type F or M is appropriate for depleted or enriched uranium unless other specific information is available.

### 5.6.2 General Guidance

These considerations lead to the following reasonable assumptions for unmonitored workers at LANL:

- 1943 through 1946 – There was a significant potential for unmonitored intakes of plutonium, nominally 3%, fresh mixture; $^{210}$Po in designated areas; and uranium, nominally either depleted or enriched, although natural uranium was used extensively in conventional weapons testing.

- Claimant interviews can indicate that the worker might have participated in radiological assignments not typically part of the normal work functions without radiological records or monitoring. In the early years of operation substitution of non-radiological workers on radiological tasks was possible.

- Workers with potential exposure to fission or activation products prior to 1955 (possibly 1958) were not monitored. This includes exposures or potential exposures to $^{131}$I and $^{125}$I.

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## GLOSSARY

**acceptable MDA (AMDA)**
That MDA listed by nationally recognized standards as the minimum sensitivity that a laboratory should achieve. Actual detection levels are often lower than this AMDA.

**aging**
In the context of reactor fuel and mixtures of plutonium isotopes, the time since $^{241}Am$ was separated from the plutonium mixture.

**activation**
The induction of radioactivity in material by irradiation with neutrons.

**activity fraction**
The fraction of the total activity represented by a particular radionuclide.

**$BiPO_4$ era**
The period when plutonium samples were analyzed by the $BiPO_4$ technique; October 1949 to January 1957.

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

**cooling**
In the context of reactor fuel, the time since the fuel was removed from the reactor core.

**cupferron era**
The period when plutonium samples were analyzed using the cupferron analytical technique; March 1944 to October 1949.

**DTPA**
Chelating agents in the form of calcium salts (CaDTPA) or zinc salts (ZnDTPA) of diethylenetriaminepentaacetic acid.

**dry box**
The predecessor to the modern-day glovebox, the dry box was made of wood with ports for attached rubber gloves. The atmosphere often included inert gas to further contain the dust. High-efficiency filters on dry box exhaust and pressure differentials were used to control the spread of contamination to the outside. The operator could perform tasks completely separate from the material without exposure to the dusts, etc.

**fabrication**
Manufacturing

**hydrodynamic test**
High-explosive non-nuclear experiment to investigate hydrodynamic aspects of primary function up to mid to late stages of pit implosion.

**implosion**
A sudden inward compression and reduction in volume.

**Kiva**

A remotely controlled critical assembly building associated with the Critical Experiment Facility.

**nCi-years**

Nanocurie-years – The 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 nCi-years can be summed to provide total nCi-years

**NTA era**

The period when plutonium bioassay samples were analyzed by the NTA technique; January 1957 to April 1966.

**outfall**

The discharge point of a drain, sewer, or pipe as it empties into a body of water.

**rad**

unit of absorbed dose

**simulated**

In the context of urine sampling, collection of urine from about one-half hour before retiring to bed, through the sleep period, and for about one-half hour after rising for two consecutive nights, or other similar protocol, to simulate a 24-hour sample.

**Sv**

sievert -- the SI unit for dose equivalent. (1 Sv = 100 rem).

**tolerance values**

Refers to the concentration of a radionuclide in a bioassay sample, above which would indicate that an unacceptable intake had occurred or an unacceptable body burden existed in that individual.

**Tuballoy**

Natural uranium

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# ATTACHMENT 5A
# OCCUPATIONAL INTERNAL DOSE FOR MONITORED WORKERS

## TABLE OF CONTENTS

## LIST OF TABLES

## 5A.1 SCOPE

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

## 5A.2 CODES USED IN BIOASSAY AND INTERNAL DOSE RECORDS

### 5A.2.1 Los Alamos Bioassay Data Repository Codes

The codes in Tables 5A-1 to 5A-6 are in text files (.txt) from the Los Alamos Bioassay Data Repository. These files currently contain plutonium and americium bioassay data, both current and historical. Eventually, they are expected to contain uranium and thorium bioassay data. These codes and formats might not be applicable to bioassay data other than plutonium or americium. Autopsy results might be included in the database for certain individuals. This would be indicated by results after the date of death. Results coded as lung or liver would be for a tissue sample, not the *in situ* organ count. Autopsy results will not typically used for dose reconstruction.

The statistical approach currently in use at Los Alamos is the Bayesian Method. Therefore, certain parameters in the electronic text files are related to Bayesian statistics and are not applicable to dose reconstruction. These data are noted as such in the tables.

Table 5A-1. Pathway codes for current *in vitro* bioassay reports.

| Code | Pathway | Lung class/retention time[a] | Particle size, if applicable[a] | ICRP 66/68 retention type |
|---|---|---|---|---|
| iys | Inhalation | Y (D, W) | 0.2 µ AMAD (small) | S (F,M) |
| iym | Inhalation | Y (D, W) | 1.0 µ AMAD (medium) | S (F,M) |
| iyl | Inhalation | Y (D, W) | 5.0 µ AMAD (large) | S (F,M) |
| wnd | Wound[b] | 1/3 = half time 7d,2/3 =500 days | NA | NA |
| wdt | Wound[b] | 1/3 = half time 7d,2/3 =30 days | NA | NA |
| win | Wound[b] | Prompt, complete injection | NA | NA |
| wta | Wound[b] | Absorption | NA | NA |

a. Might not be specified in current Los Alamos Bioassay Data Repository; only pathway listed.
b. Use wound model in IMBA, if available. Otherwise, use injection model.

Table 5A-2. Assessment method codes for current bioassay summary reports.

| Code | Description | Code | Description |
|---|---|---|---|
| IVML | In vivo measurement laboratory | UF3.8 | Dose calculation method |
| UF3.5 | Dose calculation method | EVU15 | Enriched uranium urinalysis |
| ID1.3e | Dose calculation method | EVU18 | Depleted uranium |
| UF3.7e | Dose calculation method | Urine | Uranium urinalysis after 1992 |

Table 5A-3. *In vitro* bioassay type and incident verification codes from current dose reports.

| Code | Description | Comments |
|---|---|---|
| U | Urine | Assume 24-hr sample. |
| C | Urine affected by chelation | Ignore urine sample results for first 100 days following end of therapy. |
| L | Lung burden | *In vivo,* however, this can indicate lung tissue from autopsy. |
| F | Fecal | Assume per sample. |
| V | Liver burden | *In vivo*, however, this can indicate liver tissue from autopsy. |
| B | | This code is listed in manual.pdf file but not explained in software help file; might indicate bone from autopsy. |
| T | Total body burden | *In vivo.* |

Table 5A-4. Incident line.

| i and/or p | 02/28/1993[a] | i or iw or iwm[b] | 1.e1[b] | 2.e0 | 1.00[a] | Description of the incident |
|---|---|---|---|---|---|---|
| "i" -Incident related – indicates incident description line follows | Date of incident | Models beginning with i or w (inhalation or wound models), iw = models inhalation class W, etc. | Median of lognormal prior | Geometric standard deviation | Prior probability that intake is nonzero based on Bayesian statistics – not required for reconstruction | Optional |
| "p" – Date to modify prior probability (Bayesian statistical terminology) follows – not used in reconstruction. | Date to modify prior probability and (multiplier). | Primary codes are i or w. Other codes, e.g., Xwd, inhalation class (w), and particle diameter(d) – these parameters should be inferred from fitting of data. | | | | |

a. Prior probability is Bayesian Methodology
b. See Table 5A-1

Table 5A-5. Example urine data header[a] information.

| Data | Description |
|---|---|
| 123456M | IDsex – ID and gender |
| 12/31/1999 | ENDCALC – end of calculation period – used for dose calculations |
| 3.70E-02 | Bioassay unit (Bq) – factor; 3.70E-02 indicates that listed results are pCi/sample for urine and fecal, 3.70E+01 indicates nCi burden for chest and whole-body counts |
| 3.70E+01 | Intake unit (Bq) –factor 37 indicates that intake is expressed in nanocuries (not used for reconstruction) |
| 1.00E-2 | Dose unit (Sv) –factor .01 indicates that dose listed is in rem. (not used for reconstruction) |

a. Bioassay data is preceded by the following data.

Table 5A-6. Example bioassay data line.

| 03/01/1993 | 1174 | 0.0035 | N or I[a] | 0.30 | u | Kit number | Sim24h | RAS |
|---|---|---|---|---|---|---|---|---|
| Date of sample | Results in bioassay units listed above | 1 standard deviation in bioassay units – used with normal distributions | N= Not used by LANL in LANL dose assignment<br>I= Invalid<br>Blank = valid<br>C= chelation | Biovariability factor uncertainty used for lognormal distributions | Bioassay type[b] | -1, -2, -3 if not assigned in earlier years. Indicates separate collections | Kit type. Sim24h. Jnpl normalized to /24h. | Method[b,c,d,e] See note |

a. See Section 5A.3 for a description of plutonium sample results validation protocol.
b. See Table 5A-3.

Table 5A-6 (Continued).[a]

| Counts | Background counts | Calibration Factor | Sigma | Tb/T | Sigma | Additional Background | Sigma | Prior Background | Prior Teff/Tb |
|---|---|---|---|---|---|---|---|---|---|
| | | | | | | | | | |

a. Remainder of line contains raw data that might not be necessary to complete entries to IMBA.

### 5A.2.2 Other Database and Report Codes

The codes in Tables 5A-7 to 5A-9 are in the Safety First Data base, the *In Vivo* Database, and other records of bioassay at LANL.

A significant amount of editing and corrections were made to the original results in the tritium database by J.N.P. Lawrence. However, both databases still exist. Data from the original database, rather than the edited database, are expected to be transferred to the final database. In the interim, results can be submitted from either database.

Table 5A-7. Tritium bioassay in urine (edited database) .

| Z number | Collected | Nuclide | Result | Unit | Uncertainty | Unit |
|---|---|---|---|---|---|---|
| Might change if employment status changed. | Sample date | H-3 | "0" = < MDA or value | Bq/ 24 hr sample | 1 sigma (standard deviation) | Bq/24 hr sample |

As an alternative, if the unedited tritium results are supplied the format will be as follows.

| Name, last first | Employee number | Org. | Unk | Sample date | Sample type | Result | Units | Lab book | Pg | Initials | Analysis type |
|---|---|---|---|---|---|---|---|---|---|---|---|
| Energy E W | 00000 | AFB | LA | 72059 | U1 | 4.0 | UL | 9264 | 70 | OCR | T3R |

**Unk** This column has not been identified. However, contents of the database indicate that it might refer to a location code. This column is blank in the database for workers from Zia.

**Sample Type**; U1 is the only code currently identified in the results; others might apply.

**Results** can be positive or listed as L1.0 for less than 1.0. Zeros (0) should be interpreted as less than the current reporting limit.

**Units**: UL = µCi/L

**Analysis type:** T3R appears to be tritium, radiometric.

Table 5A-8. Uranium bioassay in urine. [Pending receipt of examples from LANL]

| | | | | |
|---|---|---|---|---|
| [Placeholder] | | | | |

The tabulated results of *in vivo* counts will be accompanied by a White Card for the individual. This card will contain a handwritten explanation of the evaluation of the count. In addition, information on chest wall thickness ($T_M$) and serial number is listed. The serial number is not relevant for the dose reconstruction at this time. The chest wall thickness might be useful in relating the thickness of the person's chest wall to a potential change in counting efficiency and, therefore, the MDA. The group with which the person was associated might be listed in the upper right-hand corner of the card. This can be helpful in determining the areas worked. Be aware that some groups worked site-wide. One day a machinist might be working with D-38 and the next day with Or-93. The record might also include a "Beige Card" that contains pertinent personal information, including chest wall thickness.

## 5A.3 LOCATIONS AND TYPICAL RADIONUCLIDES

The historical and current typical locations for radionuclides are listed in Table 5A-10 (Inkret et al. 1998c; ENSR 2002). When the compound of uranium is known, use Table 5A-29 in Section 5A-13 rather than the absorption type listed in Table 5A-10.

Table 5A-9. *In vivo* report codes.

| Field | Code | Description |
|---|---|---|
| Count type | Request | Request was sent by HRS-1 or equivalent to count individual. This usually occurs after potential intake. |
| | Routine | |
| | Special | Nonroutine count that was not initiated by incident. Example: follow-up count of retired employees who have had intake in past. Note in comment field should clarify result. Might not have been used this way in past. |
| | Termination | Termination count, performed for persons who have ever been monitored for specific radionuclide. Might not indicate potential exposure in that year. |
| | Recount | To confirm results of previous count or because of failed analysis. |
| Comments | See file | Check worker's file for notes |
| | MDTA | Results below MDTA |
| | Recount needed | Recommendation; look for recount in records. |
| | MDA | Results below MDA |
| Results | Null | When result is NDA = No detectable activity rather than MDA. Results are below reporting limit (probably decision level or MSMA) |

## 5A.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 data sets.**

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. Review of studies listing the bioassay results of the original 26 workers with intakes of Pu show considerable variability between samples both in the long and short term (Voelz et al. 1979). Section 5A.9 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.

**Validations Techniques PUQFUA2 (Lawrence 1978)**
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 BiPO4 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.

4. LANL recognizes that unique situations can and do occur when:

    a. An individual urine result should be retained despite its being invalidated by the test procedures, and

Table 5A-10. Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| | | | | | ICRP 30 | ICRP 68 |
| TA-0 | | | None | Original town site | | |
| TA-1, Original Main Technical Area | 1945 | 1965 active<br>1975 decommissioned | All | | See footnote | See footnote |
| TA-1, Bldg. D, Pu chemistry and metallurgy | 1943 | 1954 | Pu-239, Pu-238,<br>U-238, DU,<br>Am-241,<br>Po-210,<br>Ba-140,<br>La-140 | Absorption depends on matrix or pure<br><br><br>May indicate Sr-90 | Y- W<br>Y<br>W, Y (Pu matrix)<br>D –W<br>D<br>D-W | M-S<br>M<br>M, S<br>F-M<br>F<br>F-M |
| TA-1, Bldg. D-2, Contaminated laundry | | 1953 | Pu-239; Pu-240; Pu-238; U-236; DU;<br>Po-210;<br>Ac-227;<br>Ra-226 | Depends on the compound | W - Y<br>W<br>D<br>Y<br>M | M - S<br>M<br>F-M;<br>M-S<br>M |
| TA-1, Bldg. D-5, Sigma vault - storage | | 1965 | Pu-239,<br>U-238 | | W<br>W | M<br>W |
| TA-1, Bldg. ML Bldg., Medical laboratory | | | Cm<br>Am | Processing | W<br>W | M<br>M |
| TA-1, Bldg. C, Uranium machining | 1943 | 1964 | Uranium | | W | M |
| TA-1, Bldg. G, Uranium and graphite sigma pile | | 1959 | Uranium<br>Ra-226 | | W<br>W | M<br>M |
| TA-1, Bldg. H, and Gamma Bldg. | 1945 | 1957, 1959 | Po-210<br>MFP | Initiators<br>Cs-137 contamination incident occurred | D<br>Various | F - M<br>Variable |
| TA-1, Bldg. HT, Heat treatment and machining | | 1965 | Unat and EU | | W | M |
| TA-1, Bldg. HT Barrel House, Storage | | 1964 | Pu-239,<br>U-238 | | D-W-Y<br>W | F-M – S<br>M |
| TA-1, Bldg. M, Processing and recovery EU | | | EU | Processing, metallurgy and recovery | W | M |
| TA-1, Bldg. M-1, Machining | | | U-238 | | W | M |
| TA-1, Bldg. O, | | 1956 | Radium<br>Radon | Radon cooked off sources on a hot plate, Ra /RaBe Calibration Sources | | |
| TA-1, Bldg. Q, | | 1959 | Radium<br>Radon | A spill occurred<br>Ra Calibration Sources | | |
| TA-1, Sigma Bldg, | | 1965 | Unat, EU,<br>Th | Casting, machining, powder metallurgy | W<br>W-Y | M<br>M-S |
| TA-1, Bldg. TU, Machining Tuballoy | | 1964 | Unat | | W | M |
| TA-1, Bldg. TU-1, Recovery of EU | | 1964 | EU | Furnace for burning rags | W | M |
| TA-1, Bldg. V, Machine Shop | 1943 | 1959 | Uranium | Unusual assignments | W | M |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| TA-1, Bldg. W, Van de Graaff accelerator | | | Uranium<br>Po-210<br>Tritium<br>Th-228 | <br><br><br>"Mesiothorium" | W<br>D<br>D<br>-- | M,<br>F M<br>FSR-2<br>M – S |
| TA-1, Bldg. X, Cyclotron | | | Be, uranium, lithium, tritium, strontium targets.<br>Zn-65 | Targets had induced beta activity. | Various | Various |
| TA-1, Bldg. Y, Physics Laboratory | | | Tritium,<br>Uranium | | | F<br>M |
| TA-1, Bldg. Z, Cockcroft-Walton accelerator | | | None | | | |
| TA-2, Omega West Site | | | | Housed critical experiments | | |
| TA-2, Water Boiler | 1943 | 1974 | U-235,<br>I-131, I-125,<br>Rb-88, Cs-137,<br>Xe-131, Ar-41<br>Tritium<br>Pu-239 | Enriched U fuel | W<br>D<br>D<br>NG | M<br>SR-1,F<br>F<br>SR-D |
| TA-2, Clementine | 1946 | 1952 | Unat, Pu,<br>I-131, I-125,<br>Rb-88, Cs-137,<br>Xe-131, Ar-41 | Ruptured Pu fuel rod, U reflectors | W<br>D<br>D | M<br>SR-1,F<br>F<br>SR-D |
| TA-2, Omega West Reactor (OWR) | 1956 | 1995 | U-235<br>I-131, I-125,<br><br>Rb-88, Cs-137,<br>Xe-131, Ar-41<br>Cr-51,<br>Na-24<br>Tc-99m | Enriched U fuel<br>I-125 production Loop Schedule – at times operated "around the clock" | | M<br>SR-1,F<br><br>F<br>SR-D<br>F,M,S<br>F<br>F,M |
| TA-3, South Mesa Site, Technical Facilities | 1953 | | All | Pu processing | See footnote | See footnote |
| TA-3, Bldg. 29, CMR – Chemical and Metallurgical Research (SM-29) | 1951 | | Pu-238 | Wing 9 handled irradiated U and Pu In hot cells. Small quantities of uranium and plutonium, mixed fission products including iodines, Pu-238 | Y | S–M |
| TA-3, Bldg. FE-19 | | | Plutonium | | W | M |
| TA-3, Bldg. 34, Cryogenics | | | Tritium | 3,000 Ci HTO released in 1979 | D | SR-2 |
| TA-3, Bldg. 16, Van de Graaff accelerator | | | Tritium | 800 Ci HTO released in 1977 | D | SR-2 |
| TA-3, Bldg. 35, Press building | | | U-235 | | W | M |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| TA-3, Bldg. 39, Tech Shop | | | DU | | W | M |
| TA-3 SM-40, Physics | | | All | Incident contaminated large portion of building with Po-210 through ventilation (late 1950s or early 1960s) | See footnote | See footnote |
| TA-3, Bldg. 66, Sigma Complex, metallurgy and fabrication | | | EU, DU powders, Pu | Pu processing | W | M |
| TA-3, Bldg. 102, Tech Shops | | | Uranium | | W | M |
| | | | Pu | | Y | S |
| TA-3, Bldg. 141, Rolling Mill | | | DU | | W | M |
| | | | Pu | | Y | S |
| TA-3 Chemistry and Metallurgy Bldg. (CMR), Wings 3,5,7 | | | H-3 | HTO, HT | D | SR-2, SR-D |
| TA-3 Chemistry and Metallurgy Bldg. (CMR), Wing 9 | 1961 | | Cs-137 | Potential for low-level chronic intake in hot cell work | D | F |
| | | | MFP including I-131 | | D | SR-1<br>F |
| | | | Pu-238, -239, -240 | 0.1-10µ AMAD, oxide, nitrate, fluoride and metal. Oxide is most common. | Usually Y, can be W | S -M |
| TA-3, Ion Beam Facility (IBF[a]), SM-16 | | | I-125 | Iodide, labeled organics | D | SR-1, F |
| | | | H-3 | Labeled DNA precursors(OBT), water (HTO), HT | D | SR-2(HTO, OBT),<br>SR-0 (HT) |
| | | | P-32 | Labeled organics, phosphates | W | F-M |
| TA-3 Tritium Instrument Calibration Facility, SM-40 | | | H-3 | HTO, HT | D | SR-2, SR-0 |
| TA-3-184, Occupational Health | | | Pu | | Y | S-M |
| TA-3-216 Weapons Test Support | | | Pu | | Y | S-M |
| TA-3-700,<br>SM-700<br>Acid Neutralization and Pump Bldg. | | | Pu | | Y | S-M |
| TA-4, Alpha Site | | 1956 | DU | Firing site until 1956, Materials disposal site C. | W | M |
| TA-6, Two-Mile Mesa Site | 1944 | 1950 | DU | Detonator manufacturing | W | M |
| TA-8, Nondestructive testing | 1984 | | Pu-239; Pu-238<br>U-235; DU | Gun firing site. | W-Y<br>D-W-Y | M – S |
| | | | Co-60; Ir-192 | | W | M |
| | | | Cs-137 | | Y | S |
| | | | | | D | F |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| TA-10, Bldg. CMR-10, Bayo Canyon Site | 1944 | 1950 | Sr-90, | RaLa radiochemistry | D or Y | F or S |
| | | | DU, Unat | | W | W |
| | | | La-140 | | D or W | F or M |
| | | | Ba-140 | | D | F |
| TA-11, K Site | 1947 | | Ra-226-Be | 20-MeV betatron | | |
| TA-15, Electron accelerator | 1962 | Present | Pu-239 | PHERMEX | W | M |
| | | | DU | | W | M |
| | | | H-3 | | D | SR-2 |
| TA-16, S Site, Weapons Engineering Tritium Facility (WETF) | 1989 | Present | Pu-239 | Explosive casting and machining | W | M |
| | | | DU | | W | M |
| | | | H-3 | | D | SR-2 |
| TA-16 WETF, Bldg 205 | | Present | H-3 | Labeled DNA precursors(OBT), water (HTO), HT | D | SR-2 (OBT, HTO)<br>SR-0 (HT) |
| TA-18 | | | U-235; U-233<br>Pu-239; Pu-240;<br><br>MFP;<br>I-131;<br>polonium | 0.1-10: AMAD, oxide, nitrate, fluoride and metal. Oxide is most common.<br>Ruptured Po source, 1953 | W<br>Usually Y, can be W<br>D-Y<br>D<br>D | M<br>M<br><br>F-S<br>SR-1, F<br>F |
| TA-18, Pajarito Laboratory, Rover reactor, criticality experiments | 1946 | 1973 | MFP | Betatron used from 1951 to 1954, Enriched uranium metal sphere 1952<br>Pu core added 1 year later.<br>1954 unreflected, delta phase Pu. | | |
| TA-19, East Gate Laboratory | | 1962 | | None | | |
| TA-21, DP-West, Plutonium facility | 11/45 | | WGPu | | Y | M-S |
| TA-21, CMR, Heat sources | | | Pu-238 | Accident with glovebox breached 1971 | Y | M-S |
| TA-21, Bldg. 2 and 3, Wet Chemistry | | 1982 | Pu | 1958 accident , separated phases in Pu process tank, unshielded tank | Y | M-S |
| TA-21, Bldg. 4 and 5, Dry chemistry | | 1981 | Pu | | Y | M-S |
| TA-21, Bldg. 12, Filter building | | 1975 | Pu | Ac contaminated | Y | M-S |
| TA-21, Bldg. 3, Oxalate precipitation operations | | | Pu-239<br>Pu-238<br>U-235 | | Y<br>Y<br>W | M-S<br>M-S<br>M |
| TA-21, Bldg. 4 | 1945 | 1948 | EU hydride | | | |
| TA-21, Bldg. 4 | 1960 | | Pu-239 | Hot cell examine irradiated Pu and EU fuel elements | Y | M-S |
| TA-21 | 1965 | | Pu-238 and Pu-239 | | Y | M-S |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| TA-21, Bldg. 5, Pu fabrication | | Limited use in 1975 | Pu-239, Pu-238 | Fire contaminated exhaust filter, 1959 | Y | M-S |
| TA-21, Bldg. 150, Pu fuels development, heat sources development | 1963 | | Pu-238, Pu-239 | Sealed capillary broke, 2800 x MPC 10/1970 | Y | M-S |
| TA-21, Bldg. 210, Pu research | | | Plutonium | | Y | M–S |
| TA-21, Bldg. DP-East | | | EU | | W | M |
| TA-21, Bldg. 155, Tritium Systems Test Assembly (TSTA), deuterium and tritium fuels. | 1984 | 1990 | HT and HTO | >10 billion Ci. Equipment failure -- H-3, 13.8 Ci released | D | SR-2, SR-0 |
| TA-21, Bldg. 151, 152, Experimental program | 1945 | | Po-210 | Produced initiators | D,W | F-M |
| | | | Ac-227 | Produced initiators | Y | S |
| TA-21, Bldg. 155 | 1949 | 1984 | Po-210 | Produced initiators | D,W | F-M |
| | | | Ac-227 | Produced initiators | Y | S |
| TA-21, Bldg. 153 | 1945 | In service until 1970-1973 | Po-210 | Produced initiators | D,W | F-M |
| | | | Ac-227 | Produced initiators | Y | S |
| TA-21 DP East TSTA Tritium Test Assembly Facility, Bldg 155 and the Salt Laboratory, Bldg 209 | | | H-3 | Labeled DNA precursors(OBT), water (HTO), HT | D | SR-2, SR-0 (HT) |
| TA-21, Liquid Waste Reprocessing, Bldg 35 and 257 | Late 1940s | 1986 | All | Pu and transuranic liquid wastes | See footnote | See footnote |
| TA-21 DP West | | | Pu-238, -239, -240 | 0.1-10µ AMAD, oxide, nitrate, fluoride and metal. Oxide is most common. | Usually Y, can be W | |
| TA-22, TD Site | | | DU | Detonator development | W | M |
| TA-23 NU Site | 1945 | 1950 | Unknown | Firing site | | |
| TA-24 T Site | 1944 | | DU | Facilities transfer to TA-16 | W | M |
| TA-25 V Site | 1944 | 1946 | DU | Taken over by TA-16 | | |
| TA-26 D Site | 1946 | 1948 | U-235, U-238<br>H-3 | Storage Vault | W<br>D | M<br>SR-2 |
| TA-27 Gamma Site (Far Point) | 1945 | 1947 | Pu-239<br>DU<br>Thorium | Pu gun assembly | Usually Y, can be W<br>W | M-S<br>M<br>M-S |
| TA-28 Magazine A | 1979 | Present | DU | Firing site | W | M |
| TA-29 Magazine B | | 1957 | DU | Explosives storage area | W | M |
| TA-30 through TA-32 | | | | unknown | | |
| TA-33, HP Site, High Pressure Tritium Laboratory, Bldg 86 | 1950s | 1980s late | H-3 | Tritium oxide (HTO), tritium gas (HT) | D | SR-2, SR-0 |
| TA-35, LAMPRE | 1955 | 1967 | MFP, | Molten Pu fuel | | |
| | | | Sr-90, | | | F,S |
| | | | Co-60 | | | M,S |
| | | | VFP | | | |
| | | | MAP | | | |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| TA-35, LAPRE I, LAPRE II test reactors | 1955 | 1960 | MFP | Highly enriched U fuel | | |
| | | | Sr-90 | | | F,S |
| | | | Co-60 | | | M,S |
| | | | VFP | | | |
| | | | MAP | | | |
| TA-35, Target Fabrication Facility (TFF), TSL-213 | | | H-3 | Labeled DNA precursors(OBT), water (HTO), HT | D | SR-2, SR-0 |
| TA-35, Ten Site, CMR-10 | 1950 | 1963 | La-140, | Sr-90 contamination suspected (F,S) | D –W | F-M |
| | | | Ba-140 | | D | F |
| TA-35 | | 1981 | U-235 | General site | W | M |
| | | | DU | | W | M |
| | | | Np-237 | | | M |
| | | | Pu | | | M-S |
| | | | Po | | | F-M |
| TA-35, Laser Fusion Research | 1974 | | unknown | | | |
| TA-36, Kappa Site | 1950 | Present | DU | | W | M |
| TA-37, Magazine Area C | | | DU | | W | M |
| TA-39, Ancho Canyon Site | 1955 | 1960 | Unat, DU<br>Thorium | Firing points | W | M |
| TA-40, Detonator Firing (DF) Site | | | H-3 | | D | SR-2, SR-0 |
| TA-41, W Site, Weapons Group WX | | | H-3<br>Pu<br>U<br>Am | Engineering of nuclear components<br>Fabrication of test materials | D<br>Y-D<br>W<br>W-Y | SR-2, SR-0<br>F-S<br>M<br>M-S |
| TA-41, Ice House, Bldg 4 | | | H-3 | Tritium oxide (HTO), tritium gas (HT) | | SR-2, SR-0 |
| TA-42, Incinerator Site | | 1970 | All | Reduced low-level Pu-contaminated waste | See footnote | See footnote |
| TA-43 HRL[a] Health Research Laboratory | 1953 | 1970 | I-125 | Iodide, labeled organics | D | SR-1, F |
| | | | H-3 | Labeled DNA precursors(OBT), water (HTO), HT | D | SR-2, SR-0 |
| | | | C-14 | Labeled DNA precursors, | D | SR-2 |
| | | | P-32 | Labeled organics, phosphates | W | F,M |
| TA-43 | 1953 | 1970 | All | | See footnote | See footnote |
| TA-45, Radioactive Liquid Waste Treatment Plant, WD Site | 1951 | 1964- operations ended | All | Removed Pu before discharging effluents | See footnote | See footnote |
| | 1960 | 1963 | MFP | | | |
| | | 1967– decom. | | | | |
| TA-46, WA Site | 1950 | 1974 | | Rover Batteries | | |
| TA-46, WA Site | 1976 | 1980s | U-235, 238 | U isotope separation | W | M |
| | | | Th | | | M-S |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| TA-48, Radiochemistry Site | 1950s | present | All<br>MAP<br>MFP | Actinide chemistry and hot cell isotope production | See footnote | See footnote |
| TA-48, Nuclear Chemistry | 1950s | Present | Se-75 | Spallation product, seen in hot chemistry on targets | | M or F |
| | | | H-3 | Tritium oxide (HTO), tritium gas (HT) | D | SR-2, SR-0 |
| | | | Cd-109 | $CL^-/NO_3^-$ mixture loaded in $SnPO_4$ resin. Cd phosphate is most probable material of intake, very soluble, 1: AMAD | D | F |
| | | | I-131 | Fission product chemistry | | F |
| TA-49, Frijoles Mesa Site | 1960 | 1961 | H-3<br>Pu<br>U | | D<br>Y-D<br>W | SR-2, SR-0<br>F-S<br>M |
| TA-50 Waste Management Site | 1963 | | Pu-238, -239, -240<br>All | 0.1-10: AMAD, oxide, nitrate, fluoride and metal. Oxide is most common. | Usually Y, can be W | S |
| TA-51, Environmental Research Facility | 1962 | Present | Co-60<br>Sr-90 | Animal exposure facility<br>Presently environmental impact research. | Y, possibly W<br>D, Y | S, possibly M<br>F, S |
| TA-52, Reactor Development Site | Late 1960s | 1970 | U-238,<br>Pu-238<br>H-3<br>VFP<br>Kr, Xe | High-temperature, gas-cooled, graphite reactor, UHTREX (Ultra-high Temperature Reactor Experiment) | W<br>Y<br>D | M<br>M-S<br>SR-2, SR-0 |
| TA-53, Los Alamos Neutron Science Center, largest accelerator facility, LAMPF | 1972 | | C-11, N-13, O-15, Ar-41<br>I-131<br>Induced activity in U targets, corrosion products | Short-lived air activation<br>Medical isotope production | | SR-2<br>SR-D<br>F<br>M,S |
| TA-54 Waste Storage Facility | | | Pu-238, -239 -240<br>All | 0.1-10: AMAD, oxide, nitrate, fluoride and metal. Oxide is most common. | Usually Y, can be W | M,S |
| TA-55 Plutonium Facility (PF-4) | 1969 | | H-3 | Labeled DNA precursors(OBT), water (HTO), HT | D | SR-2, F, SR-0<br>HT |
| TA-55 Plutonium Facility (PF-4) | 1969 | | Pu-238, -239, -240 | 0.1-10: AMAD, oxide, nitrate, fluoride and metal. Oxide is most common. | Usually Y, can be W | M, S |

Table 5A-10 (Continued). Current and historical locations and default respiratory absorption type.

| Location | Start | Demolished or decommissioned | Nuclide[a] | Comment | Inhalation class/type | |
|---|---|---|---|---|---|---|
| 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 | Y | S |
| | | | C-11 | Byproduct at LANSCE, seen in workers during beam cycle | D | SR-2, unless CO; SR-1 |
| | | | N-13 | 511 keV during beam cycle | D | F |

a. Intakes of labeled compounds do not follow the default ICRP 68 models. (ICRP 1994b)
b. See Table 5A-3.
c. 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)
d. MFP – mixed fission products; Cs-137
e. MAP – mixed activation 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
f. Delta phase plutonium = Pu-240 = 4.5%; also used sphere of 20.1% Pu-240 and 98.1% U-233
g. VFP – Volatile Fission Products

b. An individual urine result should be invalidated despite its being validated by the test procedures.

5. 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) ± 1 year – continue.

2. Select the four closest positive samples, ±1 year.

   a. Find the average of the four samples

   b. Find the average of the standard deviation

3. 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.

$$RF = [I/(E + I)]^{0.74}$$

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

$$CL = RF \times UR$$
$$PS = RF \times U2S$$

where:

CL = Calculated urine level
UR = result
PS = pseudo standard deviation
U2S = standard deviation of the result

5. 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.

6. 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 precede 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.

**$BiPO_4$ 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 < 0.075 pCi, the sample is rejected as too small.

**NTA-ZnS or PHA Eras**
1. If there at least two samples > 0.075 pCi in the set and the standard deviation is ≤ 0, the sample is rejected as too small.

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 exceeding 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. 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.

**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.

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

and

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

where:

$U_e$ = earlier dated urine result
$U_l$ = later dated urine result
$\sigma_e$ = standard deviation of $U_e$
$\sigma_l$ = standard deviation of $U_l$
$D_e$ = 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 $U_e$ and
$\sigma_c$ = calculated standard deviation of $U_{c,}$ and
$(1.282)Z_{1-\alpha}$ = standard normal variable for (1-α) one-sided confidence interval; α = 0.1

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

**Hypothesis**

1. Does the calculated urine result $U_c$ exceed the measured result $U_l$?

2. 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 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.)

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

## 5A.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 $^{238}$Pu, $^{239}$Pu, and $^{242}$Pu body burdens. These reports were performed routinely.

The PUQFUA code provided calculation of $^{238}$Pu, $^{239}$Pu, and $^{242}$Pu body burdens and validation tests of the analytical results for the samples. Various versions of the code 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.

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

The Z1YRPUU code provided a method of marking 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 5A-11 lists the areas and exempt areas for access.

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 various PUQFUA calculations. The software was used beginning in 1974 for $^{238}$Pu and $^{239}$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
7. Other accident if next year's urines show obvious increases

Table 5A-11. Zia employee access to plutonium areas.

| Area | Urine sample within 425 days of entry | Exempt |
|---|---|---|
| Job requiring respiratory protection | X | |
| Modifications or repairs on dry boxes or other highly contaminated | X | |
| Replacement of Pu-contaminated filters at all sites. | X | |
| Janitorial (long-term) work in Pu operation areas | X | |
| Long-term operations (weeks) in areas of low levels of Pu contamination (>1,000 dpm-60 $cm^2$ and <10,000 dpm-60 $cm^2$) | X | |
| Decontamination of Pu spills with more than >10,000 dpm-60 $cm^2$ | X | |
| Work in burial pits at TA-54 when personnel contamination potential is moderate to high | X | |
| Short-term jobs (2-3 days) whenever sizable quantities of Pu (grams of Pu-238 or kg 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 days) in areas of CMR Building, Ten Site, TA-50, TA-55, TA-54, TA-18, TA-48 or TA-21, where there is little Pu contamination (<1,000 dpm-60 $cm^2$) | | X |
| Jobs in other minimum exposure potential areas when respiratory protection is not required and possibility of Pu contamination is minimal. | | X |

## 5A.6 CURRENT AND HISTORICAL *IN VITRO* BIOASSAY PROGRAMS

Historically the *In Vitro* Bioassay Program included the nuclides listed in Table 5A-12. The current *in vitro* bioassay program includes the nuclides and techniques listed in Table 5A-13. The *in vitro* bioassay samples have been taken at LANL throughout the history of the bioassay program. Sensitivities vary with technique and period.

Table 5A-12. 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 5A-13. Current *in vitro* bioassay performed 1997 (Inkret et al. 1998b).

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

## 5A.7 ESTIMATION OF DATE OF INTAKE

The following guidance from Lawrence (1978) may be relevant to estimating the date of plutonium intake, based on routine and special sampling protocol, when no intake date is recorded or to understand LANL's rationale for selecting 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 determining the intake date 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 later sample.

3. The earliest potential accident date, if several occur between the paired samples.

4. One-half day before initial sample if potential accident is recorded on date of initial sample.

5. The earliest potential accident date before the initial sample if any potential accidents occurred before initial sample.

6. Fifteen days before the initial sample if no potential accidents occurred before initial sample.

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.

## 5A.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 5A-14 lists the sampling protocols.

## 5A.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 5A-15.

## 5A.10 EXAMPLE BIOASSAY RESULTS

Table 5A-16 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 of the nasal swipes greater than 50 dpm were prior to 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 are 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.

Table 5A-14. Routine sampling procedure.

| Years | Nuclide | Protocol |
|---|---|---|
| 1944 | Pu | Collected on 24-hr urine sample in clean areas following decontamination. Working in Pu areas operations – weekly, daily nasal swabs. Blood counts every 6 weeks (LASL 1944b). |
| 1945 – 1952 | Pu | Collected 24-hr urine sample away from working environment to ensure contamination-free urine. |
| 1953 – 1957 | Pu | Metal kit for collection of last voiding of day and first in morning for 2 consecutive days. |
| 1958 – 1967 | Pu | Four 1-pt bottle disposable kit. More closely approached volume of true 24-hr urine sample. Reduced potential contamination. |
| 1944 | Po | Collected on 24-hr urine sample in clean areas following decontamination. Working in Po areas operations – weekly, daily nasal swabs. Blood counts every 6 weeks (LASL 1944b). |
| 1945 –1952 | Po | Collected 24-hr urine sample away from working environment to ensure contamination-free urine. |
| 1970 – 1975 | Tritium | >150 µCi/L – 1 per day; 150-75 µCi/L – every 2 weeks; 74 -24 µCi/L – 1 per week (Healy 1970). |
| 1975 – present | Tritium | Routine – every 2 wks; or 2 hr after expected exposure then if <1- no more samples, 1-10 µCi/L – 1 w/in next month, 10-100 µCi/L – weekly samples, >100 µCi/L –daily samples. |
| 1944 — 1967 | Uranium | Weekly urinalysis (LASL 1944b). |
| 1967 – present | Uranium | Annual, including drinking water sample since 1992. |

Table 5A-15. Plutonium urinalysis—summary of sensitivities and analytical techniques.

| Period | Urine analysis method |
|---|---|
| 1944 – 1952 | Pu extracted by cupferron chloroform, gas flow counter (1944), Simpson alpha counter efficiency = 50%, background = 1 cpm to 0.1 cpm (1945+)<br>Could not differentiate Po and Pu. Selected based on potential exposure<br>12% of blanks showed ~1.4 cpm or 0.8 pCi<br>(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, Pu plated on stainless-steel disk. NTA counting method, 1,000-min exposure to emulsion, background 0.007 dpm. |
| 1963 – 1965 | Ion exchange/ Pu separated on anion-exchange resin. Electroplated for NTA counting, 84% ±14% recovery |
| 1966 | ZnS counter |
| 1967 – 1971 | Either ZnS or alpha pulse height analysis RAS (PHA) permit measurement of Pu-238; background= 0.003 ± 0.003 cpm |
| 1971 to present | All alpha PHA, computerized spectrometry |
| 9/18/80 | Began Pu-242 analysis |
| 1982 – 1986 | Coprecipitation (alkaline earth $PO_4$ or oxalate)<br>Alpha spectrometry (60,000-second count time) or rapid Alpha Phosphor Scintillation Counting (3,600 second) |
| 1997 | Alpha spectroscopy, Based on Class Y, 1 µm, Pu-239 (Inkret et al. 1999a) |
| 1997 to 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. 1999a). |

Table 5A-16. Example urine bioassay results.

| Date (L/R nasal count) | Average room air conc. dpm/m$^3$ | Dpm/24 h |
|---|---|---|
| 8/9/46 (189/320 dpm 7/29/46) | 6 – 188 | 1.2 |
| 9/19/46 (149/19 9/5/46) | | 4.7 |
| 12/18/46 (57/68 12/10/46) | | 1.7 |
| 4/18/47 (164/106 dpm 12/30/46); (102/61 dpm 1/21/47); (91/135 dpm 4/1/47) | 11 – 98 | 0.7 |
| 5/23/47 | | 0.7 |
| 6/26/47 | | 0.7 |
| 7/30/47 (144/40 dpm 7/7/47) | | 0.0 |
| 8/27/47 | | 1.0 |
| 10/2/47 | | 1.5 |
| 11/7/47 (120/78 dpm 10/3/47) | | 0.8 |
| 12/8/47 | | 4.0 |
| 1/13/48 | 24 – 69 | 1.0 |
| 2/13/48 (0/59 dpm 2/10/48) | | 0.0 |
| 3/19/48 | | 0.0 |
| 4/22/48 (86/3 dpm 4/26/48) | | 2.0 |
| 6/23/48 (244/72 dpm 6/10/48) | | 3.7 |
| 7/22/48 (72/1 dpm 7/2/48) | | 0.0 |
| 8/19/48 (65/0 dpm 8/2/48) | | 0.8 |
| 9/20/48 | | 3.0 |
| 10/26/48 | | 2.0 |
| 11/22/48 | | 2.5 |
| 12/21/48 (50/38 dpm 12/1/48) | | 0.0 |
| 1/24/49 | 19 – 72 | (Moved to uranium work) |
| 1/31/49 | | 2.0 |
| 7/14/49 | | 1.2 |
| 2/8/50 | | 0.8 |
| 9/1/50 | | 0.1 |
| 2/28/51 | | 0.8 |
| 9/4/51 | | 0.7 |
| 5/19/52 | | 0.3 |
| 12/14/52 | | 0.0 |
| 9/4/53 | | 0.0 |
| 6/4/54 | | Off uranium |
| 6/18/54 | | 0.0 |
| 6/8/55 | | (Moved back to plutonium) |
| 8/1/55 | 3 | 1.2 |
| 8/12/55 (22/28 dpm 8/9/55) | | 0.6 |
| 8/19/55 | | 0.0 |
| 9/30/55 | | 0.7 |
| 11/14/55 | | 0.7 |
| 12/27/55 | | 0.6 |
| 2/9/56 | 3 | 0.0 |
| 4/5/56 | | 0.7 |
| 4/30/56 | | 0.0 |
| 6/8/56 | | 0.4 |
| 7/20/56 | | 0.6 |
| 8/23/56 | | 0.5 |
| 9/25/56 | | 0.0 |
| 10/24/56 | | 0.0 |

Table 5A-16 (Continued). Example urine bioassay results.

| Date (L/R nasal count) | Average room air conc. dpm/m$^3$ | Dpm/24 h |
|---|---|---|
| 11/23/56 | | 0.0 |
| 12/17/56 | | 0.1 |
| 1/31/57 | 4 | 0.23 |
| 2/28/57 | | 0.68 |
| 4/12/57 | | 0.22 |
| 5/14/57 | | 0.12 |
| 6/14/57 | | 0.11 |
| 7/15/57 | | 0.03 |
| 8/19/57 | | 0.00 |
| 9/20/57 | | 0.20 |
| 10/22/57 | | 0.21 |
| 10/31/57 | | 0.39 |
| 11/14/57 | | 0.51 |
| 1/10/58 | 4 | 0.00 |
| 2/21/58 | | 0.65 |
| 3/25/58 | | 0.51 |
| 5/7/58 | | 0.025 |
| 6/19/58 | | 0.55 |
| 7/30/58 | | 0.49 |
| 9/15/58 | | 0.79 |
| 11/28/58 | | 0.47 |

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

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

## 5A.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 5A-17 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 currently available before Lawrence (1967). Table 5A-18 lists routine sampling frequencies for $^{241}$Am exposures; routine samples were not performed for $^{241}$Am prior to 1998. Routine and postexposure protocols are combined in Table 5A-19 for tritium. Table 5A-20 lists frequencies for uranium. Routine work or frequent entry in an area with beta/gamma emitting radionuclides currently requires annual whole-body count. (LANL 2004b)

## 5A.12 SAMPLING PROTOCOL FOR ACCIDENTAL EXPOSURES

Awareness of the sampling protocol associated with exposures provides guidance for associating 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 prior to Lawrence (1967). It should be assumed that the protocol continued until the year of the next dated reference unless otherwise stated. Tables 5A-21 to 5A-25 list protocols for accidental exposures.

Table 5A-17. Routine plutonium sampling frequency.

| Frequency | 1944 (Kolodney 1946) | 1967 (Lawrence 1967) | 1973 (Lawrence 1973) | 1998 (Inkret et al. 1998a,c; LANL 2004b) |
|---|---|---|---|---|
| Monthly | Great exposure | | | |
| Quarterly | Moderate exposure | 1. Persons working with ≥ 10 g Pu-239 in chemical or metallurgical operations, inside or outside glovebox<br>2. Persons with 50% body burden. (MPBB= 0.04 µCi Pu-239) | 1. Persons working w/ ≥ 10g Pu-239 or ≥ 0.04g Pu-238 (~0.6-0.7 Ci of either) inside or outside glovebox, or<br>2. Persons w/ body burden >20,000 pCi | |
| Semiannual (biannual) | Slight exposure | 1. Persons working with <10g of Pu-239 in chemical or metallurgical operations<br>2. Supervisors of quarterly sampled category<br>3. Persons with 25% burden. (MPBB=0.04 µCi Pu-239) | 1. Persons working w/ Pu but ≤ 10 g Pu-239 or ≤ 0.04 g Pu-238 in chemical or metallurgical operations,<br>2. Supervisors of the quarterly sampled category, or<br>3. Persons with > 10,000 pCi body burden but < 20,000 pCi | 1. Working with ≥0.04 g (0.7Ci) of Pu-238, analyzed by RAS<br>2. Performing chemical or metallurgical operations or maintenance on systems containing ≥10g of Pu-239 or Pu-240 and ≥0.04 g of Pu-238 (0.6-0.8 Ci) analyzed once by TIMS and RAS and once by RAS |
| Annual | | 1. Other supervisory personnel<br>2. Persons working with sealed containers of Pu<br>3. Other persons with casual encounters with plutonium who regularly work in areas where Pu is handled. | 1. Supervisors, persons working with sealed containers of Pu;<br>2. Casual encounters with Pu, or<br>3. Working with prepared counting foils containing >20 mg of Pu-239 or 0.08 mg of Pu-238 (~1.3 mCi of either) | 1. Routine work with operations of <10g (0.6Ci) of Pu-239 or Pu-242 (0.6-0.8 Ci)<br>2. Performing maintenance on systems containing ≥10g of Pu-239 or Pu-240 (0.6-0.8 Ci) analyzed by TIMS and RAS<br>3. Working with operations of or performing maintenance on systems with <0.04 g of Pu-238<br>4. Line supervisors of personnel in semiannual categories<br>5. TRU glovebox, bag outs, etc.<br>6. ESH-1 RCTs who frequently enter work areas<br>7. All personnel with confirmed, measurable intakes of Pu-238 and/or Pu-239. |

Table 5A-17 (Continued). Routine plutonium sampling frequency.

| | 1944 (Kolodney 1946) | 1967 (Lawrence 1967) | 1973 (Lawrence 1973) | 1998 (Inkret et al. 1998a,c; LANL 2004b) |
|---|---|---|---|---|
| Initial (baseline) | | | All new hires or assigned to work with LANL where other persons are routinely sampled | 1. If there has been risk of exposure to Pu or if exposure history is missing or inconclusive.<br>2. Entry requirement for unescorted access to some facilities<br>3. Occasional work in plutonium areas but do not handle plutonium. |
| Termination | | | 1. Persons terminating employment who have previously submitted urine samples, or<br>2. Have been working in major Pu areas, but never sampled | Workers who submitted routine samples |
| UPPU Club (see Section 5.1) | | Complete bioassay and physical examination at 5-year intervals. Bioassay continues after termination of employment. | Complete bioassay and physical examination at 5-year intervals. Bioassay continues after termination of employment. | Complete bioassay and physical examination at 5-year intervals. Bioassay continues after termination of employment. |

a. Samples analyzed by TIMS – thermal ionization mass spectroscopy.
b. Samples analyzed by RAS – radiometric, alpha-spectroscopy.
c. The frequencies above do not apply to working with sealed sources in TA-15 where bioassay is as needed.

Table 5A-18. Routine sampling frequency $^{241}$Am exposures.

| Frequency | 1998 (Inkret et al. 1998; LANL 2004b) |
|---|---|
| Quarterly | None |
| Semiannual (biannual) | 1. Performing operations in gloveboxes with ≥0.2 g of Am-241 (0.6 Ci)<br>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)<br>2. Frequent entry or performing maintenance on systems containing <0.2 g of Am-241 (0.6 Ci)<br>3. Line supervisors of personnel in semiannual categories<br>4. ESH-1 RCTs who frequently enter work areas<br>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 – alpha-spectroscopy.

Table 5A-19. Sampling protocol for tritium.

| Period | Program | Protocol |
|---|---|---|
| 1950 – 1970 | | Biweekly for significant quantities or more often if exposure confirmed, removed from tritium work at 124 µCi/L. |
| 1970 – 1975 | | >150 µCi/L – 1 per day; 150-75 µCi/L – every 2 weeks; 74 -24 µCi/L – 1 per week (Healy 1970) |
| 1975 – 1998 | | Routine – every 2 wks; or 2 hours after expected exposure then if <1 µCi/L - no more samples, 1-10 µCi/L – 1 w/in next month, 10.1-100 µCi/L – weekly samples, >100 µCi/L –daily samples (Inkret et al. 1998b) |
| 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 metal tritide or 0.1 Ci of organic tritium | Every 2 weeks |

Table 5A-20. Uranium routine sampling protocol.

| Period | Protocol |
|---|---|
| Pre-1983 | Biweekly samples collected.<br>Collected on Fridays, minimum annual |
| 8/1983-6/1993 | Spot sample collected on Monday mornings before entering work area |
| 7/1993 | Last voiding on Sunday night and second on Monday morning, first voiding. |
| 1998 | Spot samples every 2 weeks/persons performing hands-on work/ potential for 100 mrem; machining operations, polishing operations, foundry work, chemistry operations in which >10g of U, work with oxidized metal, >100g of bulk powder.[a] |

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

## 5A.13 SUMMARY OF *IN VITRO* AND *IN VIVO* SENSITIVITIES

Table 5A-26 lists sensitivity data for *in vitro* bioassay.

Tables 5A-27 and 5A-28 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 $^{235}$U and $^{241}$Am (LASL 1977b).

Table 5A-21. Sampling protocol for accidental plutonium exposures (Lawrence 1967).

| Severity class | Description | Sampling protocol |
|---|---|---|
| PA (Prompt Action) | Most serious accidents<br>1. Injection detectable by plutonium wound monitor (~0.01 µg)<br>2. Chemical burns from plutonium solutions<br>3. Facial contamination > 20,000 cpm<br>4. Nose swipes > 500 dpm | 1. Evening following accident<br>2. 10 days later<br>3. 1 month later<br>4. 14-monthly thereafter. |
| DA (Delayed Action) | 1. No known equipment failure, 10x MPC for 1 week or 50x MPC for 1 day<br>2. Skin contamination in excess of limits<br>3. Superficially contaminated cuts that are positive on surface monitoring only.<br>4. Equipment failure causes exposure for indeterminate period. | 1. 1 month after accident<br>11 more samples at monthly intervals. No entry is made in "No. of days accident prior to sample" column of PUQFUA |
| NRS (Nonroutinely Sampled) | 1. Small wounds occurring in major plutonium areas (DP-West, Ten Site, CMR Bldg)<br>2. Exposures without respirators at 10x MPC for period less than 1 week.<br>3. Nose counts > 50 dpm<br>4. Skin contamination > 500 cpm/60 $cm^2$ | 1. 1 urine sample collected 1 month after intake. |
| Automatic Rescheduling | 1. Detect unexpected high exposure and verify its existence<br>2. Detect contaminated urine sample<br>3. Previous sample is 1 dpm/sample | 1. Request another sample<br>2. Persons routinely excreting plutonium are exempt from special sampling but are maintained on routine sampling |

Table 5A-22. Sampling protocol for accidental plutonium exposures (Lawrence 1973).

| Severity class | Description | Sampling protocol |
|---|---|---|
| PA (Prompt Action) | Most serious accidents<br>1. Injection detectable by plutonium wound monitor >2000 pCi (dl 600 pCi Pu-239 and 200 pCi Pu-238)<br>2. Chemical burns from plutonium solutions<br>3. Facial contamination > 40,000 dpm<br>4. Nose swipes > 500 dpm | 1. Evening following accident<br>2. 10 days later<br>3. 1 month later<br>4. 12 monthly thereafter.<br>Fecal samples might be collected |
| DA (Delayed Action) | 1. No respiratory protection, 10x MPC for 1 week or 50x MPC for 1 day<br>2. Skin contamination after decontamination in excess of limits<br>3. Superficially contaminated cuts that are positive (500 dpm) on surface monitoring only. | 1. Fecal collected on day 2 for inhalation<br>2. 1 month after accident fecal and urine<br>3. 11 more urine samples at monthly intervals. Possible fecal<br>Potential accident entry is made in PUQFUA |
| NRS (Nonroutinely Sampled) | 1. Small wounds occurring in major plutonium areas (DP-West, Ten Site, CMR Bldg) no activity detected. < 500 dpm<br>2. Exposures without respirators at 10x MPC for a period less than 1 week.<br>3. Nose counts >50 dpm<br>4. Skin contamination >1,000 dpm/ 60 $cm^2$<br>5. Potential accident date is noted in PUQFUA and no sample has ever been submitted or routine is not scheduled for at least 3 months. | 1. 1 urine sample collected 1 month after intake.<br>2. Fecal samples for types 2 and 3.<br>3. Memorandum sent to H-1; potential accident entry in PUQFUA might be required |
| Automatic Rescheduling | 1. Detect unexpected high exposure and verify its existence<br>2. Detect contaminated urine sample<br>3. Previous sample is 1 dpm/sample | 1. Request another sample<br>2. Persons routinely excreting plutonium are exempt from special sampling but are maintained on routine sampling |

Table 5A-23. Sampling protocol for accidental plutonium exposures (Inkret et al. 1998c).

| Severity class | Description | Sampling protocol |
|---|---|---|
| PA (Prompt Action) | Most serious accidents<br>1. Injection detectable by plutonium wound monitor > 0.2 nCi<br>2. Chemical burns from plutonium solutions, skin contamination > 500 dpm, α by 60 $cm^2$ probe or >0.2 nCi in wound area<br>3. Facial contamination > 10,000 dpm α<br>4. Nasal > 100 dpm<br>5. Airborne > 200 DAC-h, without respiratory protection.<br>6. Skin after decontamination is > 1,000 dpm α by 60 $cm^2$ probe<br>7. As requested | Pu-239, Pu-240<br>1. Urine samples days 1, 3, 5 after intake<br>2. Fecal samples days 1, 3, 5–optional<br>3. *In vivo* chest count days 3, 5–optional<br>Pu-238<br>1. Urine samples on days, 1, 3, 5, 8, 16, 30, 60, 120, 240 after intake<br>2. Fecal samples on days 1, 3, 5–optional<br>3. *In vivo* chest count days on 3, 5–optional<br>Urine analyzed by RAS and TIMS |
| DA (Delayed Action) | EDE > 100 mrem; CDE >1 rem any tissue or organ<br>1. Present in room when CAM alarms<br>2. Other individuals on Prompt Action (PA)<br>3. Positive wound count<br>4. Chemical burns; after decon skin >100 dpm<br>5. Facial contamination, before decon, >1,000 dpm<br>6. Nasal swipes; 1 nostril >15 dpm, both sum = 35 dpm α<br>7. Average ≥ 40 DAC-hr, without respiratory<br>8. Skin contaminate >100 dpm after decontamination.<br>9. Wounds contaminated with >250 dpm (by α probe)<br>10. Request by manager, group leader, or medical | Typically 1 urine, by RAS and TIMS, if Pu-239, or<br>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. Prompt Action schedule followed by monthly urine samples[a].<br>Might include blood and fecal.<br>Or as directed. |
| Follow-up | Any positive routine urine | As directed. |

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

Table 5A-24. Sampling protocol for accidental $^{241}$Am exposures (Inkret et al. 1998d).

| Severity class | Description | Sampling protocol |
|---|---|---|
| Pa (Prompt Action) | Most serious accidents<br>1. Injection detectable by Am wound monitor > 0.2 nCi<br>2. Chemical burns from Am-bearing solutions, skin contamination > 500 dpm, α by 60 $cm^2$ probe or > 0.2 nCi in wound area<br>3. Facial contamination > 10,000 dpm α<br>4. Nasal > 100 dpm<br>5. Airborne > 200 DAC-h, without respiratory protection.<br>6. Skin after decontamination is > 1,000 dpm α by 60-$cm^2$ probe<br>7. As requested | 1. Urine samples days 1,3,5, 8,16,30,60,120,240 after intake<br>2. Fecal samples days 1,3,5 – optional<br>3. *In vivo* chest count days 3,5 – optional |
| DA (Delayed Action) | EDE >100 mrem; CDE >1 rem any tissue or organ<br>1. Present in room when CAM alarms<br>2. Other individuals on Prompt Action (PA)<br>3. Positive wound count<br>4. Chemical burns; after decon skin > 100 dpm<br>5. Facial contamination, before decon, > 1,000 dpm<br>Nasal swipes; 1 nostril >15 dpm, both sum= 35 dpm α<br>6. Average ≥ 40 DAC-hr, without respiratory<br>7. Skin contaminated > 100 dpm after decontamination.<br>8. Wounds contaminated with > 250 dpm (by α probe)<br>9. Request by manager, group leader, or medical | Typically 1 urine, by RAS<br>At direction of ESH-1 personnel. |
| Chelation Therapy | Accident scenario exceeds PA by 20-40 times.<br>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.<br>Prompt Action schedule followed by monthly urine samples.<br>Might include blood and fecal or as directed. |

Table 5A-25. Uranium nonroutine sampling protocol.

| Years | Nuclide | Protocol |
|---|---|---|
| | | Pending input from LANL |
| | | |
| | | |
| 1998 | Uranium | Days 1, 4, 8 following possible incidents [high airborne, high alpha skin contamination with >10,000 dpm or nasal swipes over 100 dpm (summed)].[a] |

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

Table 5A-26. Summary of *in vitro* bioassay (except plutonium and americium) sensitivity.

| Radionuclide | Period | Type[a] | MDA | Reporting level |
|---|---|---|---|---|
| Plutonium | | U/F | See Table 5-5 | |
| Americium | | U/F | See Table 5-8 | |
| H-3 | 1950 -- 1968 | U | 5.18E4 Bq/24 h[c] (1 µCi/L) | |
| | 1969[d] – 1987 | U | 1.04E3 Bq/24 h (0.02 µCi/L)[c] | 5.18E4 Bq/24 h (1 µCi/L) |
| | 1988 – 1998 | U | 5.18E2 Bq/24 h (0.01 µCi/L) | 5.18E3 Bq/24 h (0.1 µCi/L) |
| | 1999 – present | U | 2.59E2 Bq/24 h(0.005 µCi/L) | |
| Uranium | | | See Table 5-13 | |
| Isotopic uranium[e] | | | See Table 5-13 | |
| Fission product | 1950–1970 | U | 50 to 100 dpm/L | |
| Sr-90 | | | No information available[b.] | |
| 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. | |
| Th-230 as thorium | 1958–1963 | U | 0.01 µg/ | |
| | 1963–? - present | U | 20 µg/L | |
| Pa-231 | 1985 | U | 0.88 dpm/24 hour sample | |

a. U = urine; F = fecal.
b. Currently sent to an offsite laboratory.
c. Source: Gautier (1983).

Table 5A-27. Summary of whole-body counting detection levels (nCi).[a]

| Period | Nuclide | $L_C$ | MDA[b,c] |
|---|---|---|---|
| 1955-1958[d] | Cs-137 | | 8 |
| | Sr-90[e] | | 30 |
| 1958-1970[d] | Cs-137 | | 4 |
| | Sr-90[e] | | 30 |
| 1970-1984 | Be-7 | 0.9 | 1.8 |
| | Cs-134 | 0.9 | 1.8 |
| | Cs-137 | 0.9 | 2.1 |
| | Co-57 | 2.1 | 4.8 |
| | Co-60 | 0.78 | 1.8 |
| 1984 to 1998 | Tl-202 | 0.5 | 0.9 |
| | C-11 (based on 511 keV)[f] | 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 | 6.0 |
| | 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 |

Table 5A-27 (Continued). Summary of whole-body counting detection levels (nCi)[a]

| Period | Nuclide | $L_C$ | MDA[b,c] |
|---|---|---|---|
| 1984 to 1998 (continued) | Sb-124 | 0.4 | 0.8 |
| | Ce-141 | 2.2 | 4.4 |
| | Ce-144 | 12.1 | 24.2 |
| | Cr-51 | 6.4 | 12.8 |
| | Co-57 | 1.4 | 2.8 |
| | Cu-67 | 1.5 | 3.0 |
| | 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 |
| 1998 to present[g] | 511 keV | 1.15 | 2.3 |
| | Be-7 | 4.2 | 8.4 |
| | Ce-141 | 1.35 | 2.7 |
| | Ce-144 | 6.5 | 13.0 |
| | Co-56 | 0.55 | 1.1 |
| | Co-58 | 0.5 | 1.0 |
| | Co-60 | 0.45 | 0.9 |
| | Cr-51 | 4.45 | 8.9 |
| | Cs-134 | 0.5 | 1.0 |
| | Cs-137 | 0.6 | 1.2 |
| | Cu-67 | 1.5 | 3.0 |
| | 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 | 41.0 |
| | Sb-124 | 0.45 | 0.9 |
| | Sc-46 | 0.7 | 1.4 |
| | Se-75 | 0.85 | 1.7 |
| | Tl-202 | 0.5 | 1.0 |
| | U-235 | 1.35 | 2.7 |
| | V-48 | 0.4 | 0.8 |
| | Zn-65 | 1.1 | 2.2 |
| | Zr-95 | 0.8 | 1.6 |

a. Listing of an MDA for a given radionuclide does not necessarily mean that the radionuclide was frequently encountered. If smaller MDAs are listed in the results for a given count, use those.
b. Based on 95% confidence of detection.
c. MDA = $L_C * 2$, unless otherwise specified.
d. The HUMCO I and II systems were designed for screening subjects. Subjects found to have contamination levels above background were referred to the 8- × 4–in. NaI detector, which had the same sensitivities with an extended count time.
e. By *bremsstrahlung*.
f. 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 1022 keV.
g. Lower sensitivities might be available using the lung counter for certain nuclides if lung counting is appropriate to the dose reconstruction.

Table 5A-28. MDA, $L_C$ values for lung counting.

| Period | Radionuclide | $L_C$ | MDA[a] |
|---|---|---|---|
| 1977 – 60 min count special UPPU group (LASL 1977b) | Am-241 | | 0.3 |
| | Pu-238 | | 10 |
| | Pu-239 | | 21 |
| 1980 (Ennis 2003)[b] | Am-241 | 0.155 | 0.31 |
| | Pu-238 | 11 | 22 |
| | Pu-239 | 24 | 48 |
| 1984 (Vasilik et al. 1984)[c] | Am-241 | 0.16 | 0.32 |
| | Pu-238 | 14 | 28 |
| | Pu-239 | 30 | 60 |
| 1998[d] to present (Ennis 2003) | Am-241 | 0.1 | 0.2 |
| | Am-243 | 0.1 | 0.2 |
| | Pu-238 | 10 | 20.0 |
| | Pu-239 | 31 | 62.0 |
| | 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 to 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; MDA should be increased by at least 50% for very large individuals (LASL 1977b).
b. Assume chest wall thickness of 2.5 cm.
c. MDA = $L_C * 2$
d. Lung counter has 10-300 keV and 80-3,000 keV ranges, therefore a lower sensitivity for certain fission and activation products can be obtained.

## 5A.14 INHALATION ABSORPTION TYPES

Table 5A-29. Solubility class and absorption type assigned to uranium compounds (Rich et al. 1988).

| Class D/Type F | Class W/Type M | Class Y/Type S |
|---|---|---|
| Uranium hexafluoride $UF_6$ | Uranium tetrafluoride $UF_4$ | Uranium aluminide $UAl_x$ |
| Uranyl fluoride $UO_2F_2$ | Uranium oxide $U_3O_8$ | Uranium carbide $UC_2$ |
| Uranyl nitrate $UO_2 (NO_3)_2$ | Uranium dioxide $UO_2$ | Uranium-zirconium alloy UZr |
| Uranyl acetate $UO_2(C_2H_3O_2)_2$ | Uranium tetroxide $UO_4$ | High-fired uranium dioxide $UO_2$ |
| Uranyl chloride $UO_2Cl_2$ | Ammonium diuranate $(NH_4)_2+ U_2O_7$ | |
| Uranyl sulfate $UO_2SO_4$ | | |
| Uranium trioxide $UO_3$ | | |

## 5A.15 IN VIVO BIOASSAY ROUTINE FREQUENCY

Table 5A-30 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.

Table 5A-30 *In vivo* bioassay routine frequency

| 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.<br>Routine work or system maintenance with $\leq$ 0.2 g pure americium or frequent entry where work is performed with any pure Am-241.<br>Routine work or frequent entry in any area with beta/gamma emitting radionuclides. |
| Pu1 | Biennially | Works with plutonium<br>Routine work or frequent entry in any area with Pu-239 or -240, including material processing, system maintenance, supervision, or other support.<br>Routine work or frequent entry in any area with Pu-238.<br>Personnel who routinely work with plutonium and perform chemical or metallurgical operations with < 10g of Pu-239 or -242 or 0.04g of Pu-238.<br>TRU glove box/fume hood operations<br>TRU hot jobs<br>TRU bag outs<br>Glovebox operations<br>Fume hood operations<br>Hot jobs<br>Bag outs<br>Maintenance operations on process systems, and<br>RCT duties. |
| Pu | Semiannually | Special request |
| Uranium | Annual | Weekly access to areas where depleted uranium is machined or polished, casting and cleaning crucibles outside dry boxes, chemical operations, including purification and recovery. Handling $\geq$ 100g metal or bulk powder outside gloveboxes, or operations with uranium-hexafluoride in uncontained systems. |
| MFP | Semiannually | Special request |
| MAP/MFP | Annual | Works in high contamination areas and or works in airborne radioactivity area. |
| Th | Annual | Works with thorium |