



ORAU TEAM Dose Reconstruction Project for NIOSH

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ACRONYMS AND ABBREVIATIONS

AEC	U.S. Atomic Energy Commission
AGHCF	Alpha Gamma Hot Cell Facility
AMAD	activity median aerodynamic diameter
ANL-E	Argonne National Laboratory-East
APS	Advanced Photon Source
ATLAS	Argonne Tandem Linear Accelerator System
ATSR	Argonne Thermal Source Reactor
BRC	Body Radioactivity Counter
CEDE	committed effective dose equivalent (50-year)
cm	centimeter
CP-1	Chicago Pile 1
CP-2	Chicago Pile 2
CP-3	Chicago Pile 3
CP-3'	Chicago Pile 3 Prime
CP-5	Chicago Pile 5
d	day
D&D	decontamination and decommissioning
DL	detection limit
DOE	U.S. Department of Energy
dpm	disintegrations per minute
EBR-II	Experimental Breeder Reactor No. 2
EBWR	Experimental Boiling Water Reactor
EEOICPA	Energy Employees Occupational Illness Compensation Program Act
hr	hour
HVEM	High-Voltage Electron Microscope
ICRP	International Commission on Radiological Protection
in.	inch
IPNS	Intense Pulsed Neutron Source
IVEM	Intermediate-Voltage Electron Microscope
keV	kilo electron volt, 1,000 electron volts
kW	kilowatt
L	liter
LINAC	linear accelerator
MARS	Microcosm for Acid Rain Studies
MDA	minimum detectable activity
MDC	minimum detectable concentration
MeV	mega electron volt, 1 million electron volts
min	minute
mL	milliliter
MPC	maximum permissible concentration

MPL	maximum permissible limit
mrem	millirem
nCi	nanocurie
NIOSH	National Institute for Occupational Safety and Health
NRTS	National Reactor Testing Station
SNM	special nuclear materials
TBD	technical basis document
U.S.C.	United States Code
WBC	whole-body counter
ZGS	Zero Gradient Synchrotron
ZPR-I	Zero Power Reactor No. 1
ZPR-II	Zero Power Reactor No. 2
ZPR-IV	Zero Power Reactor No. 4
ZPR-IV'	Zero Power Reactor No. 4 prime
ZPR-IX	Zero Power Reactor No. 9
ZPR-V	Zero Power Reactor No. 5
ZPR-VI	Zero Power Reactor No. 6
ZPR-VII	Zero Power Reactor No. 7
μ Ci	microcurie
μ g	microgram
μ m	micrometer
§	section

5.1 INTRODUCTION

Technical basis documents (TBDs) 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 [DOE] facility” as defined in the Energy Employees Occupational Illness Compensation Program Act [EEOICPA; 42 U.S.C. § 7384l(5) and (12)]. EEOICPA defines a DOE facility as “any building, structure, or premise, including the grounds upon which such building, structure, or premise is located ... in which operations are, or have been, conducted by, or on behalf of, the Department of Energy (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program)” [42 U.S.C. § 7384l(12)]. Accordingly, except for the exclusion for the Naval Nuclear Propulsion Program noted above, any facility that performs or performed DOE operations of any nature whatsoever is a DOE facility encompassed by EEOICPA.

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

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

The purpose of this TBD is to provide information to assist in the evaluation of occupational internal radiation dose associated with operations at Argonne National Laboratory-East (ANL-E) from 1946 to the present. The information includes discussion of monitored and unmonitored worker exposure, as well as missed dose, using the methodology in *Internal Dose Reconstruction implementation Guideline* (NIOSH 2002b). This TBD provides guidance for assigning radionuclide intakes based on

individual dosimetry records and for situations where individual personnel monitoring data is unavailable or inadequate.

Argonne National Laboratory was established on July 1, 1946 and this TBD covers activities since that date. Work done at ANL-E was a continuation of that done by the Metallurgical Laboratory (Met Lab) of the University of Chicago beginning in 1941. The Met Lab is classified as an Atomic Weapons Employer under EEOICPA. Job locations for workers who transferred from the Met Lab did not change until land and buildings were acquired for ANL-E.

5.2 SUMMARY OF ACTIVITIES AND PROCESSES

The ANL-E site is described in detail in the *Technical Basis Document for the Argonne National Laboratory East (ANL-E) – Site Description* (ORAUT 2006a). The following paragraphs summarize that information to the extent necessary as background for the internal dosimetry programs that existed over time at ANL-E.

5.2.1 Site History and Facility Description

The ANL-E was established as the first national laboratory on July 1, 1946; the University of Chicago has operated ANL-E since its creation. The research that ANL-E carried out in the early years as a national laboratory began under the university's Metallurgical Laboratory, which built the first nuclear reactor, Chicago Pile 1 (CP-1), under the West Stands of the university's Stagg Field. Before 1946, three reactors (CP-1, CP-2, and CP-3) were built, the latter two 25 miles southwest of Chicago at the Palos Park Site A in the Argonne Woods. The 7.9-hectare (19-acre) Site A became known as Argonne Laboratory in 1943. The CP-1 reactor was rebuilt at Site A and renamed CP-2. The natural uranium fuel for the CP-3 reactor was replaced with enriched uranium during its period of operation, and the reactor was renamed CP-3'. The CP-2 and CP-3' research reactors were operated until 1954 (Wescott and O'Rourke 2001). The present site of ANL-E is about 5 miles west of Site A (Wescott and O'Rourke 2001). This site was acquired in 1947, and was called Site D (*D* for Dupage County). Table 2-1 in ORAUT-TKBS 0036-2, Technical Basis Document for the Argonne National Laboratory East (ANL-E) provides a summary of ANL-E buildings and operations (ORAUT 2006a).

Military projects were of top priority (Wescott and O'Rourke 2001) during the early years at ANL-E, but the mission has encompassed research on power reactor design and development including breeder and boiling water reactors. Reactors for producing plutonium have been studied at ANL-E since its inception. Reactors were also used at ANL-E for basic research. The CP-5 reactor, which was the first research reactor at ANL-E Site D, went on line in 1954 and operated until 1979. Research and training for commercial power plants occurred at CP-5. The Zero Power Reactor No. 6 (ZPR-VI) and ZPR-IX were constructed in Building 315 in the 1960s to relieve the workload of the ZPR-III at the National Reactor Testing Station (NRTS) in Idaho. Argonne had a demonstration reactor during the 1950s called the Argonaut (Argonne Nuclear Assembly for University Training). In 1962, the Juggernaut reactor was built in Building 335 based on the design of the Argonaut reactor. The Janus reactor, brought on line in 1964 and shut down in 1992, was built specifically for biological research into the effects of ionizing radiation.

During the 1960s, high-energy physics research requiring particle accelerators was one of the most important areas of investigation at ANL-E. Before the 1960s, the laboratory had four accelerators. The 60-Inch Cyclotron built in 1951 in Building 211 was used for nuclear solid-state physics, chemistry, isotope production, and biological studies. Two Van de Graaff accelerators in Buildings 208 and 203 were used in basic research similar to that conducted with the Cyclotron. A Dynamitron accelerator in Building 203 was used in basic physics experiments. The first linear accelerator

(LINAC) was installed in 1962 in Building 211. The most elaborate accelerators are those associated with the Argonne Tandem Linear Accelerator System (ATLAS), which was the world's first superconducting heavy-ion accelerator. The Zero Gradient Synchrotron (ZGS) operated from 1963 to 1979. Together with bubble chambers, the ZGS put ANL-E at the forefront of subatomic structure and neutrino research in the 1960s and 1970s. The Intense Pulsed Neutron Source (IPNS) went on line in 1981 and continues to be used in materials research.

Basic research in the 1950s at ANL-E largely involved reactor projects but included research into other areas of radiation science including waste management, radiation sterilization, and the basic properties of the atom. Containment facilities allowed research involving hazardous radioactive materials to be performed safely.

Biological studies on the effects of radiation on animals, plants, and humans were initiated in the 1950s. Most of these studies focused on occupational or medical exposures to radium. The Biology Division focused on whole-body experiments using gamma irradiation facilities and the Janus reactor during the 1960s to early 1980s. Following that time, the focus shifted more to studies on the cellular and molecular level.

Other activities at ANL-E have included research on superconductors and fuel fabrication and production. Environmental studies became an important mission beginning in the late 1960s. Decontamination and decommissioning (D&D) of ANL-E facilities has been ongoing since the 1950s. ORAUT (2006a) provides detailed information on these activities.

Over the course of its operations, ANL-E has measured radionuclide effluents released to the environment. ORAUT (2006a) summarizes the air and water effluent releases as well as incidents involving the release of radioactive materials or exposure to workers. ORAUT (2006b) details potential occupational environmental doses, which are not part of this internal dosimetry TBD.

5.2.2 Nuclear Materials on Site

Descriptions of nuclear materials present at the ANLE Site can be found in ORAUT-TKBS-0036-2 (ORAUT 2006a).

5.3 BIOASSAY PROGRAMS

Urine and fecal analyses were conducted at ANL-E for various radionuclides, as was whole-body counting (WBC) to evaluate radionuclide intakes. The ANL-E bioassay program is described in a collection of unrelated documents from 1946 to 1995, the most comprehensive of which is *Technical Basis Document for Internal Dosimetry* (Bertelli 2003). This document was first developed in 1995 based on a similar Hanford document. A section for uranium dosimetry based on ANL-W procedures was added later in Revision 1¹. Bertelli (2003) can be assumed to represent the bioassay program from 1995 to 2005.

5.3.1 Radionuclides of Concern

The bioassay programs evolved over time. Table 5-1 lists the radionuclides of concern for internal dosimetry (Bertelli 2003) and the type of bioassay used to assess worker intakes.

¹ The title page of the version of Bertelli (2003) reviewed for this analysis indicates Revision 00, but the document is Revision 1 as noted in the Acknowledgments.

Table 5-1. Radionuclides of concern for internal dosimetry (Bertelli 2003)

Primary Nuclides of Concern	Type of Analysis
^3H	Urine
Corrosion Products (^{58}Co , ^{60}Co , ^{54}Mn and ^{59}Fe)	In vivo measurements (whole body counting)
^{90}Sr	Urine and feces In vivo ^{137}Cs measurements as an indicator of ^{90}Sr .
^{125}I , ^{131}I	In vivo measurements (whole body counter or thyroid counter using NaI or phoswich detector)
^{134}Cs , ^{137}Cs	In vivo measurements (whole body counting) Urine
Uranium	Urine
^{239}Pu , ^{240}Pu , and ^{241}Am	Urine and fecal sampling In vivo measurements (lung counting)
^7Be , ^{14}C , ^{22}Na , ^{32}P , ^{33}P , ^{45}Ca , ^{99}Tc , ^{147}Pr , ^{226}Ra , ^{230}Th , ^{232}Th ,	Other radionuclides, bioassay by urinalysis
^{147}Pr , ^{226}Ra , ^{22}Na ,	Other radionuclides, whole body counting

5.3.1.1 Solubility Classes

The solubility classes for particular radionuclides on the site are generally not specified. Unless the solubility class is clearly defined for a particular radionuclide, the dose reconstructor should use the class that is most claimant-favorable for the intake indicated by the bioassay data.

Tritium can be assumed to be in the form of tritiated water or water vapor because that is most typical at ANL-E (Bertelli 2003), and it is claimant-favorable because the dose from elemental tritium is significantly lower than the dose from tritiated water or water vapor. While the dose from organic forms of tritium can be significantly higher than for tritium in water, organic tritium compounds were not in widespread use at ANL-E (Bertelli 2003).

The corrosion or activation products (e.g. ^{60}Co) are generally in the oxide forms. The ANL-E *Technical Basis Document for Internal Dosimetry* recommends use of the least soluble form of the radionuclide established for the element in ICRP 30 unless sufficient data are available to derive individual-specific retention (Bertelli 2003). For the purpose of dose reconstruction, in the absence of specific data, the ICRP 66 inhalation type that is consistent with ICRP 30 Class Y is Type S (ICRP 1994).

According to Bertelli (2003) strontium, cesium and iodine intakes at ANL-E are considered to be ICRP 30 Class D, consistent with ICRP 66 Class F. Plutonium oxides are assumed to be ICRP Class Y (ICRP 66 Inhalation Type S). All other chemical forms are assigned ICRP 30 Class W, consistent with ICRP 66 Class M. Uranium compounds are assigned different inhalation types based on solubility with UF_6 , UO_2F_2 , and $\text{UO}_2(\text{NO}_3)_2$ assigned to ICRP 30 Class D (ICRP Type F); UO_3 , UF_4 , and UCl_4 , considered ICRP 30 Class W (ICRP 66 Type M); and UO_2 and U_3O_8 , ICRP 30 Class Y (ICRP 66 Type S).

5.3.1.2 Route of Intake

In general, the most reasonable routine route of intake for the radionuclides at ANL-E would have been inhalation. Under accident conditions, intake via ingestion or through breaks in the skin could have occurred. In such cases the accident or incident reports should provide the dose reconstructor with information on which to base a judgment as to route of intake.

5.3.1.3 Particle Size

The dose reconstructor should assume that the route of intake for radionuclides was inhalation and a particle size distribution of 5 μm activity median aerodynamic diameter (AMAD), the default ICRP 66 particle size. If specific information is available to the contrary, that information should be used.

5.3.2 Current Bioassay Program

The *Technical Basis Document for Internal Dosimetry* (Bertelli 2003) describes the existing bioassay program in detail. As of 2003, 400 of the 5,000 ANL-E workers participated in special and routine bioassay programs. The bioassay programs were developed in accordance with 10 C.F.R pt. 835, "Occupational Radiation Protection," which was based on the concepts in ICRP Publications 26 and 30 (ICRP 1977, 1979). ANL-E also used the DOE Standard *Internal Dosimetry* (DOE 1998) and *Internal Dosimetry Program Guide* (DOE 1999) as guidance for the bioassay programs. Current policy requires internal radiation monitoring for all workers exposed to surface or airborne radioactive contamination where the worker could receive 100 mrem committed effective dose equivalent (CEDE) or a committed organ dose in excess of 5 rem.

The bioassay program includes urine and fecal analysis as well as *in vivo* measurements. The type of bioassay depends on the physical and biological characteristics of the radionuclides to which a worker could have been exposed.

In addition to those requirements for bioassay, there are other objectives of the program including providing assurance for the radiation protection program. Bertelli (2003) notes that workplace monitoring is the primary means of controlling and limiting worker exposure. However, the internal dose to an individual worker was assessed only if *in vivo* or *in vitro* measurements confirmed intakes resulting in a CEDE of 1 mrem or greater.

5.3.2.1 Scheduling of Bioassay

The current policy for selection and frequency of bioassay is stated in an appendix to Bertelli (2003). In general, individuals have been selected based on several specific conditions. The primary criterion that dictated scheduling and type of routine bioassay samples was the goal of maintaining potential missed dose at less than 100 mrem in a year. The following conditions required bioassay measurements:

- Unanticipated radiological incidents – Bioassay was required for unanticipated incidents in which radioactive materials had the potential to be inhaled, ingested, absorbed through the skin, or introduced into the body by a puncture or wound. For such incidents, a sufficient number of samples were taken to permit a reasonable estimate of the intake, and therefore the CEDE, for each radionuclide that could have entered the body.
- Elevated Air Samples – Work in areas where exposures could exceed 4 derived air concentration-hr in any 1-week period required bioassay.
- Routine Respirator Use – Routine respirator use required bioassay sampling.
- New Operations with Dispersible Radioactive Materials – New operations for which there were no previous monitoring data required bioassay in addition to air monitoring.

- Confirmatory Bioassay Samples – Bioassay was required for approximately 10% of individuals who did not meet the other criteria as a check to ensure that these individuals did not need routine bioassay.
- Initial Work With Dispersible Radioactive Materials – Bioassay was required for initial work with dispersible radioactive materials as baseline samples.
- Termination Samples – All individuals who worked with dispersible radioactive materials were required to submit termination samples.

5.3.2.2 Specific Radionuclide Bioassay Methods

5.3.2.2.1 Tritium Urine Bioassay

Because tritiated water is uniformly distributed in the body, tritium bioassay is relatively simple. A spot sample is sufficient to determine tritium activity as long as the tritium has had sufficient time to equilibrate in the body. Bertelli (2003) recommends sample collection at home or through multiple voiding. The samples were analyzed in the liquid scintillation counter. The minimum detectable activity (MDA) for the procedure was 1,000 dpm/L.

5.3.2.2.2 Cobalt-60 and Other Activation Products

Neutron activation products like ^{60}Co are present in the reactor areas and are most likely to become airborne during decontamination. Shorter-lived activation products such as ^{54}Mn , ^{59}Fe , and ^{58}Co are also found in workers in and around nuclear reactors. Because they are generally gamma emitters, body burdens can be evaluated through WBC and with fecal and urine bioassay. The primary methods for bioassay for activation products were urine and fecal analyses, but *in vivo* analyses were also employed. Table 5-2 lists the MDAs for urine and fecal bioassay for activation products.

Table 5-2. MDAs for activation product bioassay (Bertelli 2003).

Nuclide	WBC (nCi)	Urine sample (dpm/L)	Fecal sample (dpm/d)
Co-58	1	20	10
Co-60	1	20	10
Mn-54	1	20	10
Fe-59	2	40	20

5.3.2.2.3 Strontium

Potential intakes of ^{90}Sr are often assessed using ^{137}Cs as an indicator because both nuclides have comparable yields from the fission of ^{235}U , but Bertelli (2003) describes specific bioassay methods for ^{90}Sr . ^{89}Sr is initially present after release of fission products, but it is not likely to be a significant factor in the dose to workers because of its relatively short half-life (50.5 d).

Bioassay techniques for strontium were primarily urine analyses. For urine bioassay, a stable strontium carrier is added to the sample, and the strontium is separated chemically. The samples are counted overnight, and the results are corrected for the ingrowth of ^{90}Y . Bertelli (2003) lists the MDA for urine as approximately 0.3 dpm/L. Bertelli (2003) indicates that fecal analysis for ^{90}Sr was not routinely used.

5.3.2.2.4 Cesium

Cesium-137 is present at ANL-E as one of the most significant fission products from a worker dose perspective. Because it is more dispersible than ⁹⁰Sr, ¹³⁷Cs is the major component of fission product intake. It is easily detected by both *in vivo* and *in vitro* bioassay methods, and it is a good indicator of fission product exposures.

In vivo measurements were made using the 8- by 4-in. NaI(Tl) detector and counting for 20 min. The WBC technique is capable of detecting 1 nCi of ¹³⁷Cs. Annual *in vivo* measurements were recommended for workers exposed to ¹³⁷Cs.

The MDA for urine bioassay is not specified, but Bertelli (2003) notes that it is comparable to the MDA for *in vivo* measurements. Urine bioassay was recommended for instances where the worker could have been exposed to ⁹⁰Sr as well as ¹³⁷Cs.

5.3.2.2.5 Iodine

The iodine isotopes of primary interest at ANL-E are ¹³¹I and ¹²⁵I. Iodine-131 is a fission product and ¹²⁵I was used in biological research at the site. Other iodine isotopes that could have been present as a result of fission or produced in accelerators by activation have short half-lives, except ¹²⁹I. However, ¹²⁹I may have been present but unless concentrated, in “negligibly small quantities” (Bertelli 2003). (Iodine-129 is included in Table 5-3 for completeness although there is no indication in the available documents that bioassay for this isotope was routinely performed.) Iodine exposures are assumed to occur primarily as a result of accident situations because use of containment, ventilation, and respiratory protection should prevent routine inhalation exposures.

Bioassay measurements are primarily *in vivo* methods. Iodine-131 can be readily detected using a NaI or phoswich detector system. Table 5-3 lists the MDAs for specific types of detection systems.

Table 5-3. MDAs for iodine isotopes (adapted from Bertelli 2003).

Type of measurement	MDA (nCi)		
	I-125	I-131	I-129
WBC Log counter (8" x 4" NaI system)	Not detectable	1.0 (based on a 20-min count over the chest region)	Not detectable
5-in.-diameter phoswich detector (for thyroid counts)	0.03	0.03	0.03

5.3.2.2.6 Plutonium and Americium Compounds

Transuranic nuclides are present in research areas, standards laboratory, reactor fuel areas, and reactor D&D areas. The plutonium is a mixture of isotopes with ²³⁹Pu and ²⁴⁰Pu as the primary nuclides of concern. The composition of the plutonium mixture “depends on the length of time the fuel was irradiated, the time since irradiation, and the time since processing of the fuel or purification of the plutonium” (Bertelli 2003). Plutonium-241 is also present at ANL-E along with its decay product ²⁴¹Am. Some facilities employ ²³⁶Pu and ²³⁸Pu as tracers.

The ANL “Draft Environmental Impact Statement” (ANL 1979) presents the isotopic compositions of two plutonium mixtures. For the scenario of a glovebox explosion in the New Brunswick Laboratory, the isotopic composition considered to be “representative of the most hazardous material expected to be received (i.e., from LWR recycle) and is not typical of present operations” was 2% ²³⁸Pu, 58% ²³⁹Pu, 23% ²⁴⁰Pu, 12% ²⁴¹Pu, and 5% ²⁴²Pu by mass fractions. For the scenario of a plutonium fire in one of the Building 350 vaults, the plutonium isotopic considered to “bound all similar risks at ANL”

was 0.1% ^{238}Pu , 70.9% ^{239}Pu , 24% ^{240}Pu , 4% ^{241}Pu , and 1% ^{242}Pu by mass fractions. Values for the ^{241}Am component were not given. Dose assessments were performed either by assuming that all of the ^{241}Pu had converted to ^{241}Am or none of the ^{241}Pu had converted to ^{241}Am , and the higher of the two assessments was used.

Unless case-specific information is available, the DR may consider the LWR recycle isotopic composition for maximizing assessments and the second scenario isotopic composition for best estimates. In lieu of ^{241}Am data, the DR may consider the approach used by ANL in the draft impact statement.

Bioassay methods at ANL-E include *in vivo* measurements using phoswich detectors, urinalysis, and fecal sampling. The nominal MDA for ^{239}Pu , for a chest-wall thickness of 2.5 cm is approximately 80 nCi for a 20 min count, but it is dependent on chest-wall thickness. This MDA does not meet the criterion that the bioassay be able to detect an intake that would result in an annual dose of 100 mrem. Therefore, *in vivo* bioassay under these conditions is not adequate for routine measurements.

Urine and fecal bioassay are necessary for determining intakes of plutonium. Urine bioassay detects soluble and moderately soluble forms of plutonium, but fecal bioassay is necessary to assess intakes of insoluble plutonium. Urine bioassay requires an overnight or 16-hr urine sample. The document does not describe how the 16-hour sample volume is normalized to represent a 24-hour excretion. All of the actinides are analyzed as a group from a single urine sample. Fecal samples are ashed, dissolved, and electrodeposited onto planchets. Presumably, the planchets are counted in an alpha spectrometry system that allows separation of individual nuclides. The MDAs for fecal analysis are approximately 0.05 dpm/sample.

Workers with known internal depositions of plutonium can be required to participate in a long-term bioassay monitoring schedule. The urine bioassay detection limits (DL) for $^{239,240}\text{Pu}$ and ^{241}Am are approximately 0.02 dpm/L (Bertelli 2003).

5.3.2.2.7 Uranium

Uranium is present at the ANL-E site in hot cells, hoods, glove boxes, radioactive waste, irradiated reactor fuel, and in research areas generally mixed with other radionuclides. Uranium bioassay is not currently part of the routine bioassay program (Bertelli 2003). However, should it be necessary, urine bioassay and *in vivo* methods could be used.

5.3.2.3 Bioassay Schedule

The scheduling of bioassay measurements, described in a 1997 Health Physics procedure, was based on frequency of exposure with the general protocol given in Table 5-4 (Marchetti, Holtzman, Keane 1997). Table 5-5 lists the types of bioassay required for specific types of exposures, and Table 5-6 lists the codes for those analysis types.

Table 5-4. Scheduling of bioassay samples.

Class	Work assignment	Sample frequency (d)
1	Persons who work directly with radioactive materials with a frequency of at least: Daily, in situations such as plutonium glove box decontamination and volume reduction or similar work 4 d/month 1–2 d/month	30 91 182
2	Persons who work in the same area as Class 1 but do not work with radioactive materials	182
3	Persons who regularly provide a service to radioactive material work areas	182
4	Persons who provide a service as needed or visit radioactive material work areas 1 to 2 hr per week	182
	Nonscheduled personnel	Only when they have been in or near an area where a known or suspected release has occurred

Table 5-5. Required bioassays by exposure.

Radionuclides	Types of bioassay required	Types of analyses
Irradiated reactor fuel (including mixed fission and activation products)	Urine, WBC	Gamma spectrometry, actinides, Sr-90, beta liquid spectrometry
Iodine	Urine, WBC, thyroid	Gamma spectrometry, beta liquid spectrometry
Tritium (HTO or organic)	Urine	beta liquid spectrometry
Uranium	Urine, WBC	Gamma spectrometry, actinides
C-14, S-35, P-32, P-33	Urine	Beta liquid spectrometry, S-35, P-32
Sr-90	Urine	Sr-90
Actinides – transportable	Urine	Gamma spectrometry, actinides
Actinides – nontransportable	Fecal	Gamma spectrometry, plutonium, uranium, americium, thorium
	Lung count	Gamma spectrometry, X-rays

Table 5-6. Analysis codes.

Analysis code	Description
ACT	All alpha-emitting actinides (actinium, thorium, protactinium, uranium, neptunium, plutonium, americium, curium, berkelium, californium, einsteinium)
ALP	Alpha counting – heavy elements precipitated from urine
AM	Americium, alpha-emitting isotopes
BLS	Beta emitters by liquid scintillation counting
CA	Ca-45
GAM	Gamma spectrometry
NI	Ni-63
P	P-32
PM	Pm-147 and beta-emitting actinides
PU	Plutonium, alpha-emitting isotopes of Pu and Np
RA	Ra-226
S	S-35
SR	Sr-89, Sr-90
T	Tritiated water
TC	Tc-99
U	Uranium, alpha-emitting isotopes
TH	Thorium, alpha-emitting isotopes
WBC	Whole-body count, organ count, lung count
Lung	Lung count, low-energy photon spectrometry <i>in vivo</i>
THY	Thyroid count, low-energy photon spectrometry <i>in vivo</i>

5.3.3 Bioassay Before 1995

The bioassay methods and frequency before the development of Bertelli (2003) are described in various monthly bioassay reports and other documents, including memoranda, reports, and responses to inquiries.

5.3.3.1 Bioassay from 1946 to 1972

Nicksen (1946) cites maximum permissible levels (MPLs) of radionuclides in urine and refers to fecal and sputum analyses but notes that no MPL had been established. The MPL for plutonium in urine was given as 13.3 dpm/24-hr sample based on a body burden of 1 µg. This implies that bioassay programs were carried out but does not specifically describe these programs.

A 1947 letter from the Associate Director of ANL to the U.S. Atomic Energy Commission (AEC), notes that blood, urine, and fecal analyses were performed to form a record of the body content of toxic materials such as plutonium, uranium and beryllium for each individual exposed to radiation or toxic materials (Hilberly 1947). No details of the program were provided.

The Procedures of the Bioassay Group at the Argonne National Laboratory (Myers et al. 1952) describes the scheduling, recordkeeping, and analysis procedures for internal dose assessment in 1952. The document notes that urine and feces were analyzed on a regular schedule. The samples could be analyzed for tritium, ¹⁴C, gross beta (nonvolatiles in boiling HNO₃ with beta activities of energies greater than 0.3 MeV), radium, protactinium, polonium, and gross alpha (thorium, plutonium, neptunium, americium, and curium).

The scheduling of bioassay was dependent on the amount of radioactive material handled, frequency of exposure, chemical and physical nature of the material, apparent biological dangers of the material, working conditions, recommendations from the supervisors, and other factors. For example, a worker with sufficient activity to take in more than the MPL in a single event would have monthly analyses at a minimum. If a worker was handling tracer levels of a low-energy beta emitter so that intake of the entire amount used over the year would not exceed the MPL, that worker would be scheduled for annual bioassay. Individuals falling between these two extremes would be scheduled for quarterly or semiannual bioassay. Employees who worked in technical areas were requested to submit both urine and fecal samples at termination. Urine samples were collected in 1-quart glass bottles and fecal samples in 1-pint cardboard cartons.

The laboratory kept three types of notebook records: A workbook listed the types of analyses required, volume of the sample and volume analyzed, date of analysis, and analyst identification; a counter notebook listed the results of the analysis; and a record book contained a permanent record of all of the information about the analysis. If an individual bioassay showed concentrations greater than the pre-set MPLs, an immediate resample was requested. Table 5-7 lists the 1952 MPLs, which were somewhat different from Bioassay Operational Levels from an unidentified document and included in the materials used in an epidemiologic study of workers who received doses in excess of 5 rem in any year (Strom 1982). According to interviews at ANL-E in 1982, the revised values were in effect around 1960. Table 5-7 lists the later values in parentheses. The table for 1960 included an entry for beta activity with a normal range of 2,000 to 10,000 dpm/1,500 mL with a resample activity level of 12,000 dpm/1,500 mL. The reported beta activities for routine urine samples typically included naturally occurring ⁴⁰K.

Table 5-7. MPLs, 1952 (adapted from Myers et al. 1952) and after 1959.^a

Nuclide units	MPL in body		MPL in urine (1960 and later) [dpm/24-hr or /1,500 mL]	MPL in feces (dpm/24-hr sample)	Amount in urine requiring resample (1960 and later) [dpm/1,500 mL]
	μCi	μg			
Ac-227	0.04		8.8	175	0.7
Am-241	0.04	0.01	4.4 (4)	65	0.7 (0.4)
Cf-242	0.04	8E-06	350	3,500	40
Pa-231	0.04	0.8	8.8	0.9	0.7
Po-210	0.005	N/A	300 (25)	4,500	40 (2.5)
Pu-239	0.04	0.6	4.4 (4)	4.4	0.7 (0.4)
Ra-226	0.1	0.1	22 (6)	220	12 (1.0)
Th-230	0.04	1.4	8.8	90	0.7
Th-234	0.8		Not reported	Not reported	Not reported
Np-237	0.04	57	8.8	90	0.7
H-3	1E04	N/A	6E08 (8E07)	6E07	1.5E07 (7.5E06)
Sr-90/Y-90	1	N/A	1000 (400)	2000	1000 (40)

a. N/A = not applicable.

Monthly bioassay reports were submitted to Industrial Hygiene and Safety (Pingel, 1953; Robinson 1955) detailing the detectable amounts of radioactivity in urine and feces. The 1953, 1954, 1961, and 1962 monthly reports were reviewed (Pingel 1953 Robinson 1955, 1962, 1963).

The 1953 reports for the first 10 months of the year listed all results whether the activity was detectable or not. The vast majority of gross alpha results in urine were listed as zero dpm/mL. However, the DL was not defined in these reports. Repeat samples were requested for individuals whose gross alpha activity was 1.0 dpm/mL or greater. No repeat samples were requested for concentrations less than 1.0 dpm/mL. Carbon-14 and ³H in urine were reported; they were nearly all shown as “L 25” dpm/mg for ¹⁴C and “L1000” dpm/cm³ for ³H. Presumably these values were the DLs.

The minimum positive fecal sample gross alpha concentration for 1953 was 0.2 dpm for a 15.4-g sample. In contrast to the urine samples, more than half of the fecal samples were positive.

In 1954 the lowest reported activity in urine for a group of radionuclides presumably analyzed together (plutonium, neptunium, thorium, actinium, americium, and curium) was 0.3 dpm/1500 mL. The lowest reported activity for those radionuclides in feces was 0.3 dpm for a total sample (in that case, 28.4 g). Several of the monthly reports listed fluoride-insoluble alpha activity. “Fluoride insoluble” is another term for gross alpha analysis and reported as “Alpha.” According to Procedure ANL-4509 (1951) the process was co-precipitation with bismuth phosphate and lanthanum fluoride. In 1975 the gross alpha procedure used cerium fluoride instead of lanthanum fluoride. During the first part of the year, gross alpha in urine and feces was given in the report. The values above are the lowest values given either for the specific radionuclides or for gross alpha (fluoride-insoluble) activity (or both).

Radium-226 and ²³⁵U were reported separately. One positive tritium urine sample was reported in the January 1954 Bioassay Report (collection date 12/2/53) (Sedlet 1954). This indicates that either tritium analysis in urine was not routine or that there were very few positive samples.

Blood and sputum analyses were reported for several months during the first half of 1954.

Several individual bioassay records for 1951 to 1957 were reviewed to determine the recordkeeping method. The records were kept on cards that included the date, bioassay type, element for analysis, excretion per 1,500 mL (not explained on the card but which might be the ratio of 1,500 cm³ to the

actual volume of the sample), type of exposure, hours worked, air analysis of the area, and remarks. The column for type of exposure recorded the pH of the urine in many cases rather than type of exposure. A bioassay monthly report from 1958 (author unknown) for Great Lakes Carbon Co. reported results as less than 5 mg per 1,500 cm³ (This is most likely a typographical error with “m” indicating the prefix “micro” instead of “milli”).

A 1959 bioassay report for Great Lakes Carbon Company showed worker concentrations for enriched uranium down to 0.2 dpm per 1,500 cm³ (Sedlet 1959). The MPL for enriched uranium was given as 50 dpm per 1,500 cm³.

The bioassay reports for 1961 and 1962 were more descriptive than for 1953 and 1954 (Pingel 1953, Robinson 1955); however, they included only the results for nonroutine and special analyses. The analyses performed during that period included alpha (plutonium, neptunium, thorium, actinium, americium, and curium), beta activity, iodine, plutonium, polonium, radium, tritium, uranium, and other miscellaneous nuclides (e.g., ⁹⁹Tc, ⁶⁵Zn). Gross alpha data were reported in some cases. The reports did not generally specify the type of bioassay but, because the data were reported as disintegrations per minute per specified volume, it can be assumed that they refer to urine bioassay. Several reports from that period indicated activity in fecal samples and nasal swabs. In one case, a blood sample was indicated with a value of 0 dpm/1,500 mL. Because it is unlikely that a 1,500-mL blood sample was analyzed, this probably was a typographical error in the report. In another case, skin scrapings were analyzed. Because only special and nonroutine sample data were included in the monthly reports, it is unclear whether routine fecal analyses occurred during that period.

For most radionuclides, the concentrations were reported in disintegrations per minute per 1,500 cm³. The ³H concentration was reported in disintegrations per minute per cm³. The DL for ³H was 1,000 dpm/cm³ in the 1950s and early 1960s. Bioassay program reports for 1967 indicated a DL of 100 dpm/mL for ³H (Sedlet 1968). The DLs for other radionuclides were not easily discernable from the annual reports. Beta concentrations were often reported as “normal range” rather than a specific value, but the normal range was not defined. The lowest alpha emitter data was given as zero with no minimum detectable concentration (MDC) value provided. Uranium concentrations were reported in disintegrations per minute per 1,500 cm³ without reference to isotopic ratios or an MDC.

At least one of the monthly reports referred to requests for resampling and follow-up bioassay for individuals until the tritium concentration was less than 10,000 dpm/cm³. There is no statement in the report that this was standard practice.

A DL for polonium was given as 2 dpm/1,500 cm³ for a special analysis (volume of 100 mL) and 4 dpm/1,500 mL for a routine volume of 50 mL in one of the monthly reports (Sedlet 1962).

Monthly reports for 1968 were reviewed (Sedlet 1969). These reports contained information similar to that included in the 1961 to 1962 reports. It is unlikely that significant changes were made in the program in the intervening years. Results were given only for special and nonroutine samples. The list of analytes remained generally the same with the addition of gamma activity. The gamma activities specified radionuclides and listed a result of “normal” for individuals whose spectra were presumably in the range of background. A DL for plutonium of 0.2 dpm was given for a fecal sample.

A sample page of bioassay data from 1969, presumably a page from a bioassay log, showed quarterly normal (sic) uranium bioassay data. The log provided the data by location and worker. Quarterly results were recorded for most of the individuals with only one or two records for 1969 for some individuals. All of the results for those sample pages were less than 5 with no units indicated. There was no indication on the log page regarding who made the log entries.

Estimated decision levels, discussed in Section 5.3.4 were derived from the “less than” values in the Bioassay Program Reports and, in a few cases, the lowest reported positive value.

5.3.3.2 Urine and Fecal Analysis from 1973 to 1995

ANL-E developed a Health and Safety Manual before 1973 (ANL 1973). That document indicates that industrial safety and health physics policies had previously been issued through the Laboratory’s *Policy and Practice Guide*. The Health and Safety Manual was subject to revisions over time.

Chapter V-13 of the Health and Safety Manual dated March 1, 1975, describes bioassay procedures (ANL 1975). The document notes that urine samples were requested from all new employees, returning employees, and temporary employees with previous occupational exposure. Urine samples were requested from all terminating employees. The document did not exclude employees who did not work in areas with dispersible radionuclides, which indicates that the program likely applied to all employees.

Specific requirements for periodic urine and fecal analyses or *in vivo* measurements included the potential to receive a dose commitment in any calendar quarter in excess of 10% of the standard. The document does not specify bioassay methods or DLs, and it does not provide information about selection of employees for *in vivo* bioassay.

The 1976 monthly Bioassay Program Reports include the results of special and nonroutine bioassay samples (Fairman 1976). The records show less-than values for plutonium in fecal samples at 0.05 dpm/sample and 0.05 dpm/1,500 mL for urine bioassay. The basis for this presumed detection limit is not provided in the documents. The less-than values for tritium in urine was 25 dpm/mL. A reported value of 0.25-dpm/mL was a typographical error because all other less-than values for tritium for 1976 were given as 25 dpm/mL. The minimum value given for uranium in urine was simply zero, so no MDA could be discerned from the data. The lowest positive value for uranium was 0.1 dpm/1,500 mL.

Gamma activities were reported as “normal,” again with no specified MDA. Values for beta oxalate analysis were listed simply as “below detection limit.” Strontium and ¹⁴C minimum values were also listed as “below detection limit.” The apparent DL for ²⁴⁹Cf in feces was 0.1 dpm. The minimum values for radium and polonium and +3 and +4 actinides in urine were 0 dpm/1,500 mL.

Table 5-8 lists various draft and final laboratory procedures used from 1970 to 1989 for urine and fecal analysis of analytes of concern. In most cases the specific nuclide procedures described in Table 5-8 were from a laboratory manual identified in the header on the procedure as IDOP:IDCL (presumably Internal Dosimetry Operational Procedures: Internal Dosimetry Chemistry Laboratory) Revision 00. The headers on the procedures were generally undated. However, several of the individual procedures had handwritten dates on the first page. In some cases, the procedures had identifying headers, e.g. ALP-1 for the gross alpha procedure. It is reasonable to assume that the urine and fecal bioassay requirements did not change significantly during that period.

The ANL-E internal dosimetry program was reviewed in 1982 in conjunction with an epidemiologic study of workers who had received doses greater than 5 rem in any year (the 5-rem cohort) (Strom 1982). The investigators noted that all but one of the 54 individuals in the 5-rem cohort had internal dose monitoring records in comparison to the ANL-E average of 10% of workers. This indicates that most of the individuals likely to be exposed to high levels of radionuclide concentrations and high external exposure rates were monitored.

Table 5-8. Summary of bioassay analysis methods, 1970 to 1989.

Analyte	Medium	Description of method	Date
S-35	Urine	Low-background beta counting – Sulfate from ashed urine sample precipitated as barium sulfate and counted.	Undated
Ra-226	Urine and feces	Alpha counting – Coprecipitation of radium with lead sulfate, barium chloride, and barium sulfate separates radium from other activities and provides a mount sufficiently thin for alpha counting.	4/1/75 and 12/1/88
Sr and Ba	Urine and feces	Beta counting – Carriers added and carbonates precipitated from solution made basic with NaOH. Sample scavenged with ferric hydroxide after a series of nitrate precipitations from concentrated nitric acid solutions. Ba separated from Sr as the chromate and Sr is precipitated and weighed as the oxalate. Ba precipitated and weighed as the chloride. Precipitates mounted on stainless-steel planchets for beta counting.	6/12/79 and undated IDOP:IDCL* Rev. 00 *(See text)
Ni-63	Urine	Liquid scintillation counting – Aliquot wet-ashed and residue dissolved in dilute nitric acid. Ni precipitated as the hydroxide, dissolved in HCl and passed through anion exchange column to remove Fe. Ammonium citrate is added if rare earths or Zr-Nb are potential interferences. Ni precipitated with dimethylglyoxime. Precipitate dissolved in concentrated nitric acid, converted to the oxide then to the chloride. Chloride dissolved in water and incorporated into a gel scintillation cocktail for liquid scintillation counting. Recoveries are >95% and the counting efficiency is approximately 40%. MDA at the 95% probability level is <33 dpm/1000 mL.	12/1/88
Tritium	Urine	Liquid scintillation counting – Urine vacuum-distilled and scintillation solution added to an aliquot of the distillate. Sample, blanks, and spikes counted.	4/2/75, 8/17/87 and IDOP: IDCL Rev. 00
Tc-99	Urine	Low-background Geiger-Mueller or proportional counter – Technetium (IV) carried on ferric hydroxide precipitate, then oxidized to Tc (VII) with concentrated nitric acid. Technetium coprecipitated with cupric sulfide, filtered, and mounted for counting.	12/1/88
Gamma emitters	Urine	Gamma spectrometer – Samples counted by gamma spectrometry using a Ge detector.	8/17/87 and Undated IDOP:IDCL Rev 00
C-14	Urine	Liquid scintillation counting – Aliquot of urine sample added directly to gel scintillation cocktail.	7/1/68, 4/3/75, and 11/30/88
Pu, Np, Am, Cm, U, Th	Feces	Alpha spectrometry – Sample ashed, yield monitors added, and leached with nitric acid in microwave oven. Pu (+Np) and Th purified using anion exchange column. Am (+Cm) and U purified using carbamoyl methylphosphine oxide dissolved in tributyl phosphate. Counting plates made for Pu and U fractions using hexone extraction; for AM and Th fractions by electrodeposition.	4/8/75, 5/27/87 and IDOP:IDCL Rev 00 8/04/89
Po	Urine	Low-background alpha spectrometer – Po electrochemically plated onto Ag planchet from dilute HCl solution of ashed urine salts.	Undated IDOP:IDCL Rev 00
Ca-45	Urine	Liquid scintillation counting - Aliquot of raw or ashed urine sample mixed directly with gel scintillation cocktail.	8/25/75
Beta activity	Urine	Low-background beta counter – Coprecipitation with strontium oxalate from urine.	4/9/75 and 12/1/88
Actinide and lanthanides	Urine	Alpha spectrometer and low-background beta counter – Actinides concentrated from urine using alkaline phosphate precipitation, separated from interfering elements using a column loaded with octyl(phenyl)-N-N-diisobutyl CMPO/TBP. Electrodeposited and counted in an alpha spectrometer then low-background beta counter for determination of Pr-147 or other lanthanides.	IDOP:IDCL* Rev 00 6/12/89 *(See text)
Gross alpha	Urine	Alpha counter – Heavy elements precipitated from the urine in alkaline phosphate then coprecipitated with bismuth phosphate then cerium fluoride. Cerium fluoride mounted for counting. Uranium does not come through the procedure.	ALP-1 4/1/75 and 11/16/84 Rev.

Strom (1982) reports the investigators used the Bioassay Laboratory Routine Sampling Scheduling Criteria dated 1972 to extract dose information. Scheduling criteria were based on the amount and

frequency of radionuclides handled. The schedules ranged from bimonthly to annual. The schedule did not specify the type of bioassay, but most of the routine bioassay measurements during this period were urine analysis.

5.3.3.3 *In Vivo* Bioassay

None of the bioassay monthly reports for the period provided data from lung counting or WBC. The 1971 Bioassay Program Reports described the Radioactivity Body Counter (RBC), later termed the Whole Body Counter (WBC), as it was constructed and noted that the instrument would need to be calibrated. No calibration records were found.

ANL (1990) described the WBC facility. The procedure did not indicate the date this facility went into service, but it was built in 1962 (Wescott & O-Rourke 2001). The facility consisted of three low-background counting rooms housed in an underground concrete vault designed to minimize background. The shielded counting rooms housed three different types of detection systems:

- Room 1 housed a system with two 11.5- by 4-in. NaI(Tl) detectors mounted above and below a bed that could be moved. The system simultaneously scanned a supine subject.
- Room 2 had a single 6-in.-diameter, 8-in.-long NaI(Tl) detector that was termed the “log” detector. The subject sat in a chair or reclined on a curved bed with the detector above the trunk of the subject.
- Room 3 was designed for counting low-energy photons. The system employed a pair of Phoswich detectors that consisted of thin 5-in.-diameter NaI(Tl)/CsI(Tl) detectors. The system was used for measuring actinides in the lung, and the phoswich detectors were also used for measuring ^{125}I in the thyroid and actinides in other organs. A gas proportional counter in this room distinguished between internally deposited actinides and external contamination.

The counting efficiencies for these systems were determined for various configurations using three principal methods:

- Mathematical analysis of counting data obtained from measurements of an internally contaminated subject and point source measurements of the specific gamma emitter of interest
- Measurements using an anthropomorphic phantom containing a known quantity of the radionuclide of interest
- Measurements of human volunteers after administration of tracer quantities of radionuclides

ANL (1990) did not provide the efficiencies and DLs.

Toohey (1980) described the detectors used in the WBC facility, initially called the Body Radioactivity Counter (BRC), for assessment of transuranic elements. Two detectors were used: A phoswich consisting of a thin NaI(Tl) crystal backed by a CsI(Tl) crystal for detection of low-energy gamma photons or X-rays, and a xenon-filled proportional counter for detecting external contamination. For mixed plutonium and americium contamination, the ^{241}Am 60-keV gamma ray was used to quantify the amount of ^{241}Am . The contribution from the ^{241}Am to the X-ray region (12 to 24 keV) was subtracted to determine the plutonium contribution. The alpha activity could be determined if the plutonium isotopic ratios were known. Toohey (1980) did not provide the DL for the method.

An April 2005 memorandum stated that the Phoswich detectors were no longer in service and that the MDAs for the instrument “could be taken as” three times the detection level (DL), which was not specified (Keane 2005). ANL-E documentation sometimes uses *decision* level and the code D_c to indicate DLs.

5.3.4 Estimated Decision Levels and Minimum Detectable Amounts or Concentrations

5.3.4.1 Minimum Decision Levels for 1946 to 1985

There has been no evidence found that MDAs were assessed or recorded before 1995. Instead, 10% of maximum permissible excretion levels, action (repeat sampling) levels, and reporting levels have been used. These levels, if found or discerned, are reported in previous sections. The information is significantly incomplete for certain periods and radionuclides.

The decision levels for years prior to 1985 can be inferred from the “less than” values for individuals reported in the Bioassay Program Monthly Reports. These values, where a positive value is assigned for an analyte result, can also be determined from the smallest positive measurement result. Tables 5-9 and 5-10 provide those values for urine bioassay and fecal bioassay, respectively (When similar “less than” values are given in the bioassay Monthly Reports only the earliest value is included in the table.)

Table 5-9. Summary of Detection Limits for Urine Bioassay Based on Reported “Less Than” Values .

Nuclide	“Less than” value	Earliest Year	Source of Information	Comments
Actinides	0.5 dpm/1500 mL	1958	Memo. J. J. Robinson to Novak. Oct 29, 1958.	Less than value, specified as Pu, Np, Th, Ac, Am and Cm
Actinides	0.1 dpm/1500 mL	1967	Sedlet. Bioassay Report. 1967	Unspecified nuclides
Alpha	0.4 dpm/1500 ml	1948	Individual bioassay record	
Alpha	0.5 dpm/1500 ml	1951	Individual radiation exposure record	
Alpha	0.6 dpm/1500 ml	1952	Sample log entry	
Alpha	0.3 dpm/1500 ml	1959	Individual bioassay record	
Alpha	0.05 dpm/L	1960	Memo. Keane to Dolacek. July 7, 1993	Denoted as minimum detectable urine alpha activity concentration using “fluoride insoluble method”
Alpha	0.1	1967	Memo Sedlet to Novak (11/3/67)	Reported as fluoride insoluble alpha (mostly due to Pu)
Alpha	0.133 dpm/1500 mL	1985	Individual Radiation Exposure Record	
Ca-45	0.5 nCi/1500 mL	1975	Fairman, Bioassay Program Reports- 1975	
C-14	25 dpm/mg C	1948	Individual bioassay record	
C-14	2 dpm/mL	1969	Sedlet. Bioassay Report. 1969	
Californium	0.1 dpm/1500 mL	1975	Fairman. Bioassay Report 1975	Unspecified isotope
H-3	100 dpm/cc	1948	Individual Bioassay Record	This value appears to be in error given the later much higher “less than” values
H-3	1000 dpm/cc	1953	Pringle 1953 Bioassay Report	
H-3	2000 dpm/cc	1960	Robinson 1960. Bioassay Report	“Less than” values prior to and after 1960 were 1000 dpm/mL
H-3	100 dpm/mL	1967	Sedlet. 1967-1968. Bioassay Report	

Nuclide	"Less than" value	Earliest Year	Source of Information	Comments
H-3	25 dpm/mL	1969	Sedlet. 1969 - 1973. Bioassay Reports, Fairman. 1976. Bioassay Report	
H-3	2.5E4 dpm/sample	1988	Individual bioassay record	
Iodine	25 dpm/1500 mL	1967	Sedlet. Bioassay Report. 1967	Unspecified isotope
Ni-65	33dpm/100 mL	1988	Bioassay Procedures 12/1/88	
Plutonium	0.1 dpm/1500 mL	1967	Bioassay Monthly Reports – 1967 – Sedlet	Unspecified isotope
Plutonium	0.05 dpm/1500 mL	1976	Fairman. Bioassay Reports 1976	Unspecified isotope
Plutonium	0.033 dpm/1500mL	1980	Individual Radiation Exposure Records	
Pu-238	0.05 dpm/1500 mL	1974	Bioassay Monthly Reports – 1974 – Fairman	
Pu-239	0.05 dpm/1500 mL	1974	Bioassay Monthly Reports – 1974 – Fairman	
Pm-147	300 dpm/1500 cc	1962	Robinson. Bioassay Report. 1962	
Pm-147	100 dpm/1500 cc	1967	Sedlet. Bioassay Report. 1967.	
Polonium	2 dpm/1500 mL	1961	Robinson, Bioassay Report.. 1961, Sedlet.	Unspecified isotope
Pr-147	50 dpm/1500 mL	1970	Sedlet. Bioassay Report. 1971	
Radium	0.5 –1 dpm/1500 mL	1967	Sedlet. Bioassay Report. 1967	Unspecified isotope
Radium	1.333E-1 dpm/mL	1985	Individual Radiation Exposure Record	"<"
Strontium	25 dpm/1500 mL	1970	Sedlet. Bioassay Report. 1970	Unspecified isotope
Uranium	1.65 (no units)	1951-57	Bioassay logs	The <1.65 in the logs was defined as <1.65 alpha/1500 ml ifor enriched uranium in a memo from DP ONeil to J. F. Ege Jr. (10/22/55)
Uranium	10 µg/1500 mL	1957	Individual bioassay cards	Unspecified isotopes
Uranium	5 µg/1500 mL	1958	Memo. Duffy to Novak. Oct 29, 1958	Unspecified isotopes
Uranium	0.2 dpm/1500 mL	1959	Memo Sedlet to Novak – Bioassay Results for Great Lakes Carbon	
Uranium	0.6 dpm/1500 mL	1961	Individual bioassay record	
Uranium	0.2 dpm/1500 mL	1967	Bioassay Monthly Reports from Sedlet	Unspecified isotopes
Uranium	0.2 dpm/mL	1985	Individual Radiation Exposure Record	
Zn-65	200 dpm/1500 mL	1975	Fairman. Bioassay Report. 1975	
Zr-95	200 dpm/1500 mL	1975	Fairman, Bioassay Program Report - 1975	

Table 5-10. Summary of Reported “Less Than” Values for Fecal Bioassay.

Nuclide	“Less than” Value	First Years	Source of Information	Comments
Actinides	1 dpm/sample (31.2 g)	1967	Sedlet. Bioassay Report. 1967	Unspecified nuclides
Actinides	0.1 dpm/sample (12 g)	1968	Sedlet. Bioassay Report. 1968	Unspecified isotope
C-14	25 dpm/mg	1960	Robinson. Bioassay Report. 1960	
Cf-249	10 dpm/sample	1971	Sedlet - Bioassay Report 1971	
Cf-249	0.5 dpm/sample	1971	Sedlet. Bioassay Report. 1972	
Cf-249	0.1 dpm/sample (12 g)	1976	Fairman. Bioassay Report. 1976	
Cm	0.1 dpm/12 g	1968	Sedlet. Bioassay Report. 1968	Unspecified isotope
Cs-137	10 dpm/sample (29 g)	1978	Fairman. Bioassay Report. 1978	Presumably 10 dpm per sample
Cs-134	10 dpm/sample	1978	Robinson – Bioassay Radioactivity Report -1978	
Co-57	10 dpm/sample	1978	Robinson – Bioassay Radioactivity Report – 1978	
Cr-51	50 dpm/sample	1978	Robinson – Bioassay Radioactivity Report - 1978	
Fe-59	400 dpm/sample	1968	Bioassay Monthly Reports 1967-1968 - Sedlet	
Fe-59	10 dpm/sample	1978	Robinson - Bioassay Radioactivity Report - 1978	
Np	0.1 dpm/12 g	1968	Sedlet. Bioassay Report. 1968	Unspecified isotope
Pu	0.1 dpm/sample (12g)	1968	Sedlet. Bioassay Report. 1968	Unspecified isotope
Pu	0.05 dpm/sample (20 g)	1978	Fairman. Bioassay Report. 1978	Presumably 0.05 dpm per sample
Pu-238	0.05 dpm/sample (67 g)	1975	Fairman. Bioassay Report. 1975	Presumably 0.05 dpm per sample
Pu-239	0.05 dpm/sample (33g)	1975	Fairman. Bioassay Report. 1975	Presumably 0.05 dpm per sample
Th	0.1 dpm/sample (12 g)	1968	Sedlet. Bioassay Report. 1968	Unspecified isotope
Zn-65	20 dpm/sample	1972	Sedlet - Bioassay Program Report – 1972	

In lieu of definitive statements regarding minimum detectable activities, and as a guide for dose reconstructors, Tables 5-11 and 5-12 for urine and fecal bioassay have been created from data displayed in Tables 5-9 and 5-10. These data represent the best available information regarding a suggested value for detection level in lieu of definitive information.

The radionuclides represented in Tables 5-11 and 5-12 are those that were present more or less consistently throughout the history of the ANL-E facility. Other radionuclides (berkelium, californium, cobalt, iodine, mercury, polonium, promethium, protactinium, radium, strontium, technetium, and zinc) were present only sporadically depending on the particular research programs that were being conducted during a specific year.

Table 5-11. Estimated decision levels for urine bioassay by year.

Year	H-3	C-14	Gross Alpha	Actinides†	Pu	U*	Pu-238 Pu-239	Enr. U**
Units	dpm/cc dpm/mL	dpm/mg C	dpm/1500mL	dpm/ 1500 mL	dpm/ 1500mL	dpm/1500mL (µg/L)	dpm/1500mL	dpm/1500 mL
1946	1000	25						
1947	1000	25						
1948	1000	25	0.4					
1949	1000	25	0.4					
1950	1000	25	0.4					
1951-	1000	25	0.5			1.65		
1956	1000	25	0.6			1.65		
1957	1000	25	0.6			(10)		
1958	1000	25	0.6	0.5		(5)		
1959	1000	25	0.3	0.5		0.2		0.2
1960	1000	25	0.05	0.5		0.2		0.2
1961-	1000	25	0.05	0.5		0.6		0.2
1966	1000	25	0.05	0.5		0.6		0.2
1967	100	25	0.1	0.1	0.1	0.2		0.2
1968	100	25	0.1	0.1	0.1	0.2		0.2
1969-	25	25	0.1	0.1	0.1	0.2		0.2
1973	25	25	0.1	0.1	0.1	0.2		0.2
1974	25	25	0.1	0.1	0.1	0.2	0.05	0.2
1975	25	25	0.1	0.1	0.1	0.2	0.05	0.2
1976	25	25	0.1	0.1	0.05	0.1	0.05	0.2
1977	25	25	0.1	0.1	0.05	0.1	0.05	0.2
1978	25	25	0.1	0.1	0.05	0.1	0.05	
1979	25	25	0.1	0.1	0.05	0.1	0.05	
1980-	25	25	0.1	0.1	0.033	0.1	0.05	
1984	25	25	0.1	0.1	0.033	0.1	0.05	

† Sometimes reported as "Pu, Np, Th, Ac, Am, Cm"

*Values for 1976-1984 are based on lowest reported positive level (Robinson 1976)

** Based on lowest reported level (Sedlet 1959)

5.3.4.2 Estimated Minimum Detectable Activities for 1985-2001

The estimated MDAs for the years 1986 to 2000 were calculated based on reported standard deviations for an individual worker. There were 42 routine urine bioassay samples and 35 fecal samples analyzed for 13 radionuclides during that period. The reported standard deviation for each measurement was multiplied by a factor of 4.65 to obtain a MDA. The MDAs for each nuclide were averaged and the standard deviation calculated. The recommended MDA for each nuclide for the period is the average of the MDAs plus two standard deviations. The calculated values, given in Table 5-13, are consistent with current reported MDAs. The data showed no significant trends over time so the calculated values are applicable to the entire 1986-2000 time period. This method provides a reasonable but claimant favorable basis for estimating the MDA.

Table 5-12. Recommended Decision Levels for Fecal Bioassay by year (dpm) based on "Less Than" Values (All units are dpm per sample)

Year	Gross alpha ^a	Pu	U-235 ^a	U ^a	Actinides ^b	Pu-238	Pu-239
1946-							
1952							
1953	0.1						
1954	0.1		0.4		0.3		
1955-	0.1		0.03		0.3		
1957	0.1		0.03		0.3		
1958	0.1		0.03	16.3	0.3		
1959	0.1		0.03	16.3	0.3		
1960-	0.1		0.03	2.3	0.3		
1966	0.1		0.03	2.3	0.3		
1967	0.1		0.03	0.3	1.0		
1968	0.1	0.2	0.03	0.1	0.1		
1969-	0.1	0.1	0.03	0.1	0.1		
1974	0.1	0.1	0.03	0.1	0.1		
1975-	0.1	0.1	0.03	0.1	0.1	0.05	0.05
1977	0.1	0.1	0.03	0.1	0.1	0.05	0.05
1978-	0.1	0.05	0.03	0.1	0.1	0.05	0.05
1984	0.1	0.05	0.03	0.1	0.1	0.05	0.05

^a Based on Lowest Reported Value where there was an insufficient number of "less than" values

^b Based on Lowest Reported Value from 1953 to 1966.

Table 5-13. Estimated Bioassay MDAs for 1986 to 2000^a

Nuclide	Recommended MDA	
	Urine Analysis (dpm/liter)	Fecal Analysis (dpm/sample)
Am-241	0.036	1.77
Cm-242	0.059	0.55
Cm-244	0.030	NA
Cf-252	NA	0.10
Np-237	0.075	0.21
Pu-238	0.035	0.16
Pu-239	0.033	0.19
Pu-242	0.026	0.07
Th-228	0.052	2.82
Th-230	0.044	1.96
Th-232	0.031	1.00
U-234	0.068	1.82
U-235	0.032	0.60
U-238	0.059	1.49

^a NA means not analyzed

5.3.4.3 Current (2004-2005) Minimum Detectable Amounts or Concentrations

In contrast to the previous time periods, there is a considerable amount of information available with regard to current MDAs and methods for their determination. A set of intra-laboratory memos (Zhang

2005) lists the MDAs or MDCs for bioassay analyses for the analytes currently included in the routine monitoring program at ANL-E. The general equation used to calculate the MDA or MDC is:

$$MDA = (4.65 S_B + 3)/(K_{0.05} \times T), \quad (1)$$

where

- $K_{0.05}$ = Lower limit of the 90% confidence level of the calibration factor K
- S_B = Standard deviation of the net background count
- T = Count time

This method is consistent with the methodology recommended in the American National Standard *Performance Criteria for Radiobioassay*, especially the use of the lower limit of the 90% confidence level of the calibration factor (HPS 1996, Section A.5.4).

5.3.4.3.1 Unseparated Actinides in Urine

Zhang (2005) describes in detail the methods used to analyze urine bioassay samples for actinides. Unseparated actinides include plutonium, neptunium, americium, uranium, curium, and thorium. Table 5-14 lists the practical D_c 's and the a priori MDCs for calendar years 2004 and 2005. Results for this analysis might include test or analysis code "ACT."

Table 5-14. 2004-2005 Practical D_c 's and MDCs for unseparated actinides in urine (Zhang 2005).

Analyte	D_c (dpm/L)	MDC (dpm/L)
Pu-239,240	0.0070	0.044
Pu-238	0.0028	0.045
Pu-242	0.0021	0.040
Np-237	0.0103	0.049
Am-241	0.0028	0.045
U-234	0.0134	0.056
U-235	0.0036	0.048
U-238	0.0110	0.049
Cm-242	0.0254	0.095
Cm-244	0.0032	0.039
Th-232	0.0042	0.040
Th-230	0.0072	0.037
Th-228	0.0034	0.055

5.3.4.3.2 Gamma-Emitting Isotopes in Bioassay Samples

Gamma-emitting isotopes include ^7Be , ^{22}Na , ^{54}Mn , ^{58}Co , ^{60}Co , ^{134}Cs , ^{137}Cs , and ^{40}K . Table 5-15 lists the D_c 's and MDCs for urine and fecal bioassay samples. Results for this analysis might include test or analysis codes "GAM," "GAMMA," or a variation of these codes.

Table 5-15. D_c's and MDCs for gamma-emitting isotopes in urine and fecal samples (2004-2005).

Analyte	Urine samples		Fecal samples ^a	
	D _c (dpm/L)	MDC (dpm/L)	D _c (dpm)	MDC (dpm)
Be-7	87	199	na	Na
Na-22	13	30	na	Na
Mn-54	12	27	na	Na
Co-58	9	21	na	Na
Co-60	11.2	26	14	62
Cs-134	11	27	na	Na
Cs-137	11	25	15	64
K-40	208	473	294	1,244

a. na = not analyzed.

Bertelli (2003) provided the following MDAs for activation product bioassay.

Table 5-16. MDAs for activation product bioassay (Bertelli 2003).

Nuclide	WBC (nCi)	Urine sample (dpm/L)	Fecal sample (dpm/d)
Co-58	1	20	10
Co-60	1	20	10
Mn-54	1	20	10
Fe-59	2	40	20

5.3.4.3.3 Beta Liquid Scintillation Counting of Urine Samples

Liquid scintillation counting of urine samples involves counting a 0.003-L aliquot of the urine sample for 60 min. The beta energy spectrum is divided into three regions: Region A (Lowbeta) for low-energy beta emitters such as tritium; Region B (Midbeta) for medium-energy beta emitters such as ¹⁴C, ³⁵S, and ⁴⁵Ca; and Region C (Topbeta) for unspecified high-energy beta emitters other than ⁴⁰K, (Zhang, 2005). Table 5-17 lists the D_c's and MDCs. Results for this analysis might include test or analysis code "BLS."

Table 5-17. D_c's and MDCs for beta liquid scintillation counting of urine samples (2004-2005).

	D _c (dpm/L)	MDC (dpm/L)
Energy Region A: Lowbeta	1,324	3,242
Energy Region B: Midbeta	731	1,726
Energy Region C: Topbeta	743	2,120

5.3.4.3.4 Separated Actinides in Bioassay Samples

Separated actinides in bioassay samples include isotopes of uranium, plutonium, thorium, neptunium, americium, curium, and californium. Table 5-18 lists the D_c's and MDAs. Results for this analysis might include a test or analysis code of the element symbol in capital letters (e.g., PU for plutonium isotopes).

Table 5-18. D_c 's and MDAs for alpha spectrometry of separated actinides in urine and fecal samples (2004-2005).

Analyte	Urine samples		Fecal samples	
	D_c (dpm)	MDA (dpm)	D_c (dpm)	MDA (dpm)
U-234	0.017	0.045	0.022	0.082
U-235	0.010	0.042	0.016	0.081
U-238	0.016	0.045	0.021	0.079
Th-228	0.293	0.44	0.620	1.48
Th-230	0.273	0.31	0.324	0.55
h-232	0.019	0.055	0.066	0.28
Pu-238	0.032	0.10	0.039	0.17
Pu-239, 240	0.008	0.067	0.014	0.13
Pu-242	0.007	0.058	0.011	0.11
Np-237	0.005	0.059	0.010	0.12
Am-241	0.012	0.032	0.017	0.058
Am-243	0.009	0.033	0.016	0.063
Cm-244	0.004	0.023	0.008	0.046
Cf-252	0.006	0.023	0.009	0.043

5.3.4.3.5 Strontium-90 in Urine

The D_c is 0.23 dpm/L. The MDC is 0.66 dpm/L. Bertelli (2003) lists the MDC for urine as approximately 0.3 dpm/L and notes that fecal bioassay was not routinely used for strontium.

5.3.5 Estimation of Missed Dose for Unmonitored Individuals

The routine bioassay monitoring program at ANL-E has historically been conducted for workers (identified by self-reporting, by supervision, and by radiation protection staff) in areas involving possible internal exposure. Most workers at ANL-E were not identified to be at risk and were not monitored.

5.4 AIR SAMPLING

ANL-E monitored air in work areas with risk of airborne radionuclides from the onset of operations, and there are many archived boxes of air-sampling records. The Laboratory did not use air-sampling data to assign individual doses but only to provide a means for assessing the adequacy of control methods. Elevated air sample results might have been used to trigger requests for special bioassay (urine or fecal) samples from potentially affected workers. The extent of this practice was not readily discernable from the available documentation. In general, workers in areas with an active air-monitoring program would have been on a routine bioassay monitoring schedule.

ANL-E controlled the airborne radionuclides based on the maximum permissible concentration (MPC), also called "tolerance." (Nickson 1946) stated that, for alpha emitters, the MPC in air was 70 dpm/m³ (3×10^{-11} μ Ci/mL). No maximum permissible concentration was stated for beta emitters in 1946. Annotations on air sample worksheets in 1956 and 1959 indicate that the MPC for alpha emitters was still 70 dpm/m³, and the MPC value for beta emitters was 2200 dpm/m³ (1×10^{-9} μ Ci/mL). Analysis of air sampling work sheets from 1954 also indicate a beta MPC of 2200 dpm/m³.

The usefulness of air-monitoring data for dose reconstructions would be very limited because there is insufficient information available to correlate area air sample data with the location of a worker over time.

5.5 BIOASSAY REPORTS

This section shows and discusses examples of reports of bioassay data that the dose reconstructor might encounter in worker files. Personal identifiers have been removed from these examples.

Figure 5-1 is an example of the card used to record bioassay data starting in 1946 for monitored workers. The header information on the card is as follows. The F and M boxes indicated the gender of the worker. H and MO indicated the salary classification of the worker (hourly or monthly). A and T referred to the type of work (administrative or technical). The worker's job title, division, and location were recorded. When any of that information changed the old data was lined or crossed out and the new data were typed in, but the date of the change was not recorded. The Schedule codes W, Q, A, O, M, and S referred to the bioassay schedule frequency (weekly, quarterly, annually, other, monthly, or semiannually). If the schedule frequency changed the previous checked mark was crossed out, but the date of the change was not recorded. However, dose reconstructors should examine the recorded sample dates to determine the actual frequency. The other header information is self-explanatory.

SUBSTANCE ANALYZED		ELEMENT	SAMPLE		VOLUME ANALYZED	INSTRUMENT READING TOTAL COUNTS	ESTIMATED d/m/1,500 CC		REMARKS
DATE	NUMBER		DATE	VOLUME					
Urine	α		3-23-46	354	590	0	0		
"	"		5-3-46	434	350	4.1	0		
"	"		6-20-46	525	350	0	0		
"	"		8-24-46	630	250	0	0		
Feces	"		8-24-46	F-134		16.4	0	(total sample)	
Urine	"		8-27-46	634	600	0.7	0		
"	"		1-2-47	793	360	0.8	0		
Feces	"		1-2-47	F-111		2.0	0	(total sample)	
Urine	"		1-14-47	822	525	0	0		
"	"		6-26-47	1044	350	23.0	0	(total sample)	
"	"		12-17-47	1164	250	0	0		

Figure 5-1. Early bioassay data card.

The main body of the card contains the rows, generally one per analyte, for the results of the bioassay analysis. One sample could have had more than one analyte. The column headings are generally self-explanatory. The Element column could indicate an analysis type (e.g., α, β, or γ for gross alpha, gross beta, and gross gamma) or a specific radionuclide (e.g., Pu, ²³⁹Pu, U, and ³H). The result column headed by "Estimated d/m/1,500 cc" consists of two columns. The first column records the numerical value but not necessarily the magnitude; the second column is the exponent of the order of magnitude (e.g., 0 is 10⁰ or 1, 3 is 10³ or 1,000). The numerical value in the first column is multiplied by the magnitude indicated in the second column.

The Remarks field could contain supplemental information to help the dose reconstructor interpret the results especially in the case of special samples.

Figure 5-2 is another example of the card used to record bioassay data starting in 1946 for monitored workers with an accompanying handwritten record of count data for some of the samples. In this example, the count data are not presented for all samples.

The volume recorded as part of the count data is not the same as the volume recorded in the Volume Anal. (c.c.) column for these samples. The count data recorded under Net cts/min are all positive results, but the recorded results are 0 d/m/1,500 cm³. No conversion factor has been discerned yet to allow the dose reconstructor to calculate the activity in disintegrations per minute from the counts per minute data. The dose reconstructor might also note whether the data for all the samples were recorded on the card. In this case, the result for the September 28, 1953, sample 53 was not recorded on the card.

SUBSTANCE ANALYZED		ELEMENT	SAMPLE		VOLUME	INSTRUMENT	READING	NET	REMARKS	DATE
			DATE	NUMBER	VOLUME	TYPE	TOTAL	cts/min		
Urine	U		3-6-50	325			625	0	0	
"	"		10-20-50	1624			760	0	0	
"	"		1-4-51	5077			375	0	0	
"	"		4-4-51	5600			335	0	0	
"	"		5-10-51	5804			475	0	0	
"	"		6-10-51	6027			500	0	0	
"	"		8-16-51	6273			625	0.5	0	
"	"		2-4-52	7425			450	0	0	Working with hot sample - alpha emitter.
"	"		3-19-52	7900			450	0	0	
"	"		4-21-52	8343			390	0.5	0	
"	"		6-28-52	8859			500	3.8	0	
"	"		6-16-52	9289			165	0	0	
"	"		7-31-52	9762			325	0	0	
"	"		10-10-52	10850			425	0	0	
"	"		2-27-53	11976			500	0	0	
"	"		5-19-53	12277			500	0	0	
"	"		6-17-53	13305			215	0	0	
"	"		8-31-53	14071			365	0	0	
"	"		3-22-54	16950			500	0	0	
"	"		4-29-54	18649			300	0	0	
"	"	U235	9-29-54	18651			250	0	0	
Urine	U		11-25-55	1212			300	0	0	
"	"	U235	4-25-55	1244			275	0	0	
"	"		10-26-55	3521			375	0	0	
Feces	F		4-15-66	3385				0.4		(dpm per 47.2 g)
"	"		2-7-68	759				0.8		(dpm per 8.2 g)
"	"		2-7-68	740				"		
Urine	U		3-26-69	1867	225	1.0		1.1	3	70/ml
Feces	F		6-25-69	3852						Per 38 g
"	"		6-25-69	3853						Per 38 g

Sample #	Date	Volume	Net cts/min	Net cpm	Net dpm/ml	Net dpm/g
	8/31/53	115	35.0	13.0	22.0	22.0
	9/28/53	100	31.0	13.0	18.0	18.0
	3/22/54	125	53.0	13.0	40.0	36.4
	9/29/54	100	51.0	21.0	27.0	30.0
	4/25/55	100	59.0	21.0	38.0	40.8
	10/26/55	100	45.0	14.0	31.0	36.8

Figure 5-2. Early bioassay card with supplemental count data.

Figure 5-3 is an example of the data card in the 1960s; it includes examples of gross alpha, gross beta, and gross gamma analyses as well as results of special uranium analyses for urine and fecal samples. The gross alpha results for routine urine samples are all zero in this example. The gross beta results are all positive but with the note that the value includes ⁴⁰K. The dose reconstructor should remember the discussion preceding Table 5-7 that the normal range for beta bioassay measurements was 2,000 to 10,000 dpm/1,500 mL. In addition for the potassium measurements for some of the gross beta samples (all cases in this example), the ⁴⁰K result is greater than the gross beta result. The gross gamma results were all cited as "Normal." No DL for gross gamma measurements in this era has been discovered.

SUBSTANCE ANALYZED		ELEMENT	DATE	SAMPLE NUMBER	VOL.(ML)	VOLUME INFL.(CC)	RETENTION READING TOTAL CPM/HR	dpm/1500 CC	REMARKS
Urine	α	1-20-64	169	100	0	8.32	3	Inc. K-40	
"	β	5-25-64	1707	500	0	0			
"	β	5-25-64	1704	100	0	5.16	3	Inc. K-40	
"	β	12-15-64	9918	130	0	0			
"	β	12-15-64	9919	100	0	2.76	3	Inc. K-40	
"	β	12-15-64	9920	200	0	0			
"	β	7-7-65	5448	100	0	5.03	3	Inc. K-40	
"	γ	7-7-65	5449	100	0	--		Normal	
"	K	7-7-65	5450	0.1	0	8.37	3		
"	β	11-12-65	9790	100	0	4.95	3	Inc. K-40	
"	γ	11-12-65	9791	100	0	--		Normal	
"	K	11-12-65	9792	0.1	0	6.73	3		
"	α	3-23-66	2840	500	0	0			
"	β	3-23-66	2841	100	0	2.41	3	Inc. K-40	
"	γ	3-23-66	2842	300	0	--		Normal	
"	K	3-23-66	2843	0.1	0	3.34	3		
"	β	8-5-66	7343	750	100	3.18	3	Inc. K-40	
"	K	8-5-66	7344	0.1	0	3.99	3		
"	α	12-14-66	11329	950	500	0			
"	β	12-14-66	11330	100	0	2.16	3	Inc. K-40	
"	β	4-10-67	3189	700	100	4.35	3	Inc. K-40	
"	K	4-10-67	3190	0.1	0	8.23	3		
"	β	10-30-67	7458	850	100	4.25	3	Inc. K-40	
"	K	10-30-67	7459	0.1	0	4.64	3		
"	α	4-25-68	2281	450	345	0			
"	β	4-25-68	2282	100	0	4.40	3	Inc. K-40	
"	K	4-25-68	2283	0.1	0	6.38	3		
Feces	U	6-27-68	4588			311		Per 34 g - See Special Card No. 731	
Urine	U	6-27-68	4759	825	500	0		" " " "	
Feces	U	7-2-68	4761			42.5		Per total sample - See Spec. Card, 731	
Urine	U	6-26-68	4954	800	500	0		" " " "	
Feces	U	7-11-68	5095			15.5		Per total sample	
Urine	γ	9-10-68	6526	825	400	--		Normal	

Figure 5-3. Bioassay card from the 1960s.

For the special samples starting June 26, 1968, there was a Request for Bioassay Analysis card as shown in Figure 5-4. This card might contain information about the radionuclides and the circumstances leading to the request for a special or nonroutine bioassay sample.

REQUEST FOR BIOASSAY ANALYSIS			
			No. 731
			<input checked="" type="checkbox"/> Special
			<input type="checkbox"/> Non-routine
Date of Incident:	6/26/68	Time: ~1325	Building: 205
Isotopes Involved:	U (Normal - Depleted?)		
Estimated Amounts:	Small	(μ C)	mc c mg g Other
Chemical Form:	UC		
Physical State:	liquid	solid	gas powder solution Other
Names of People Involved (Include Division):	(CEN)		
Nature of Incident: Furnace disconnected from a glove box & then it was noticed that it was cherry red inside (burning).			
Remarks:			
Requested By:			
Deliver to Bioassay Group, D202 B333			(Urine) Analyzed by IH Group
IHS-41 (6-62) Special for neuranium -			

Figure 5-4. Request for Bioassay Analysis card.

Figure 5-5 is an example of the bioassay report starting in the mid-1970s. Unlike the previous cards, this report lists only the minimum result data without supplemental information. However, for radionuclide-specific analyses, the reporting (less-than) levels are stated and quantified. The Type is the type of sample (U for urine and F for fecal). The Size is the volume in milliliters of the urine sample or weight in grams of the fecal sample. The analyte ALP indicates gross alpha measurements with proportional counting, and GAM indicates gamma-emitting isotopes measured by gamma spectroscopy. The result is stated as DPM, but it is not clear whether it is per sample, per milliliter, per liter, or per 1,500 mL. Dose reconstructors should assume a continuation of the practice for the bioassay data cards of disintegrations per minute per 1,500 mL for urine samples (except for tritium) and disintegrations per minute per sample for fecal samples. For tritium, results in this era appear to be reported in disintegrations per minute per milliliter with a reporting level on a 1977 report of 2.5×10^4 dpm/mL.

Figure 5-6 shows the next evolution of the bioassay reports in about 1986. These reports show the radionuclide- or isotope-specific results along with the value of the standard deviation. It has not been determined from available documentation whether all analytical uncertainties were included in this value of the standard deviation. The units of the results are clearly stated as disintegrations per minute per liter for urine sample results and disintegrations per minute per sample for fecal results.

Some bioassay data reports starting in the 1990s list a standard set of radionuclides but show data only for some of the radionuclides; Figure 5-7 is an example. The key indicator is the zero value for the standard deviation. If the standard deviation is zero, the dose reconstructor should consider that no analysis was performed for that radionuclide.

Bioassay Results:				
Sample Number: 75-0943				
Date	Type	Size	Analyte	DPM
4/20/1975	U	375	ALP	0.000E+00
Sample Number: 76-1752				
Date	Type	Size	Analyte	DPM
5/3/1976	U	650	ALP	0.000E+00
5/3/1976	U	650	GAM	0.000E+00
5/3/1976	U	650	PU	<3.333E-02
Sample Number: 76-1755				
Date	Type	Size	Analyte	DPM
5/5/1976	F	54.1	ALP	0.000E+00
5/5/1976	F	54.1	GAM	5.200E+00
5/5/1976	F	54.1	PU-238	1.000E-01
5/5/1976	F	54.1	PU-239	1.800E+00
Sample Number: 76-3127				
Date	Type	Size	Analyte	DPM
8/9/1976	U	800	ALP	0.000E+00
8/9/1976	U	800	GAM	0.000E+00
8/9/1976	U	800	PU	<3.333E-02
Sample Number: 76-3054				
Date	Type	Size	Analyte	DPM
8/10/1976	F	26.0	ALP	2.500E+00
8/10/1976	F	26.0	GAM	0.000E+00
8/10/1976	F	26.0	PU-238	<5.000E-02
8/10/1976	F	26.0	PU-239	3.000E-01

Figure 5-5. Bioassay report from the mid-1970s.

Bioassay Results from Worker Protection System:**Sample Number: 1987-0384**

Date	Type (1)	Size (2)	Analyte	DPM (3)	± 1 S.D. (4)
2/24/1987	U	0.95	Am-241	0.003	0.004
2/24/1987	U	0.95	Cm-242	0.013	0.008
2/24/1987	U	0.95	Cm-244	0	0.004
2/24/1987	U	0.95	K-40	0	500
2/24/1987	U	0.95	Np-237	0.028	0.012
2/24/1987	U	0.95	Pu-238	0.003	0.004
2/24/1987	U	0.95	Pu-239	0.002	0.005
2/24/1987	U	0.95	Pu-242	0.004	0.004
2/24/1987	U	0.95	Th-228	0.006	0.006
2/24/1987	U	0.95	Th-230	0.002	0.005
2/24/1987	U	0.95	Th-232	0	0.004
2/24/1987	U	0.95	U-234	0.024	0.011
2/24/1987	U	0.95	U-235	0	0.006
2/24/1987	U	0.95	U-238	0.016	0.009

Note: (1) Type: U = Urine, F = Fecal**(2) Size: L for Type=U, g of ash for Type=F****(3) DPM/Litre for Type=U and DPM/sample for Type=F****(4) S.D. = Standard Deviation**

Figure 5-6. Bioassay data report starting about 1986.

Sample Number: 1995-0044

Date	Type (1)	Size (2)	Analyte	DPM (3)	± 1 S.D. (4)
1/8/1995	U	1	Am-241	0	0
1/8/1995	U	1	Cm-242	0	0
1/8/1995	U	1	Cm-244	0	0
1/8/1995	U	1	K-40	2335	523
1/8/1995	U	1	Lowbeta	1450	460
1/8/1995	U	1	Midbeta	250	230
1/8/1995	U	1	Np-237	0	0
1/8/1995	U	1	Pu-238	0	0
1/8/1995	U	1	Pu-239	0	0
1/8/1995	U	1	Pu-242	0	0
1/8/1995	U	1	Th-228	0	0
1/8/1995	U	1	Th-230	0	0
1/8/1995	U	1	Th-232	0	0
1/8/1995	U	1	Topbeta	670	230
1/8/1995	U	1	U-234	0	0
1/8/1995	U	1	U-235	0	0
1/8/1995	U	1	U-238	0	0

Figure 5-7. Bioassay report with partial data..

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GLOSSARY

accelerator

Device for imparting kinetic energy to charged particles.

activation products

Elements present in the structure of the reactor or the fuel rods that become activated by neutrons, as opposed to fission products that result from fission.

bioassay

Measurement of amount or concentration of material (usually radioactive material) in the body or in biological material excreted or removed from the body and analyzed for purposes of estimating the quantity of material in the body.

boiling-water reactor

Nuclear reactor in which the coolant, water, is permitted to boil as it absorbs the heat of the nuclear reaction.

breeder reactor

Nuclear reactor in which the operation produces a net increase in fissionable atoms.

committed

In relation to dose equivalent, refers to a total or time-integrated amount for 50 years after intake or onset of a chronic intake (or for a different period if so specified).

committed dose equivalent

Dose equivalent calculated to be received by an organ or tissue over a 50-year period following an acute intake of a radionuclide into the body or onset of chronic intake. It does not include contributions from radiation sources external to the body.

committed effective dose equivalent (CEDE)

Sum of the effective dose equivalents to various tissues or organs in the body each multiplied by the appropriate tissue weighting factor and committed for a 50-year period following an acute intake or onset of chronic intake. It does not include contributions from external dose.

control rod

Device manipulated within a nuclear reactor constructed of material to absorb neutrons for the purpose of slowing or increasing the nuclear reaction.

criticality

A sustained nuclear chain reaction (can be controlled or not).

curie

Special unit of radioactivity equal to 3.7×10^{10} disintegrations per second.

cyclotron

Accelerator capable of large beam currents where the beam is injected in the center of a circular magnet. A fixed radio frequency signal applied to two D-shaped electrodes accelerates the beam as it passes from one electrode to the other as the potential alternates. The radius of the beam increases as the energy increases.

decision level (Dc)

The value of a net observation (result) at or above which a decision is made that a positive quantity of analyte is present.

decommissioning

Removal of a facility from service, usually involving decontamination of radioactivity to specified levels and often involving demolition of the facility.

decontamination

Reduction or removal of radioactive material from a structure, area, object, or person. Decontamination can occur through (1) treating the surface to remove or decrease the contamination or (2) allowing natural radioactive decay to occur over time.

detection limit (DL)

A general term relating to the smallest amount of material detectable as a function of the measurement method and instrument background.

disintegrations per minute

Rate of radioactivity decay in a sample.

dose

Specific amount of energy from ionizing radiation or a toxic substance absorbed per unit of mass.

Dynamitron

Direct current accelerator manufactured by Radiation Dynamics. The terminal is charged with a many stage rectifier system excited by radio frequency.

enriched uranium

Uranium enhanced from its natural state to contain a higher abundance of the isotope ^{235}U .

exposure

(1) A measure of X- or gamma-ray radiation capable of ionizing air in units of roentgen. (2) Qualitatively used to describe the time and concentration of radionuclides to which a worker is exposed.

fission

Nuclear transformation characterized by the splitting of a nucleus into at least two other nuclei and the release of a relatively large amount of energy.

fission product

Elements or compounds resulting from fission.

hot cell

Specialized shielded laboratory in which radioactive materials can be handled with the aid of remotely operated manipulators. The walls and windows of the laboratory are made of materials designed to protect workers from radiation.

intake

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

in vitro

Of or relating to a process that takes place under artificial conditions or outside a living organism (e.g., in the laboratory). From Latin meaning *in glass*.

in vivo

Of or relating to a process that takes place within a living organism. From Latin meaning *in life*.

linear accelerator

Single-pass accelerator

minimum detectable amount or activity (MDA)

Smallest amount or activity of a radionuclide in a sample (or organ) that will yield a result above the detection level with a specific probability of a Type II (false negative) error while accepting a specific probability of a Type I (false positive) error.

minimum detectable concentration (MDC)

MDA expressed in units of concentration (e.g., dpm/L).

neutron

Basic nuclear particle that is electrically neutral with nearly the same mass as a hydrogen atom.

neutron, fast

Neutrons with energy equal or greater than 10 kilo electron volts.

Phoswich detector

A thin NaI detector combined with a thicker CsI detector which provides anticoincidence discrimination based on pulse rise time against higher energy photons which will excite both detectors. This reduces the low energy background enhancing detection of plutonium xrays.

positive level

Argonne Internal Dosimetry Program term for detection level: A level of a bioassay measurement at which the analyte to be detected (as opposed to being detectable).

radioactivity

Spontaneous emission of radiation, generally alpha or beta particles, gamma- and X-rays, and neutrons from unstable nuclei.

radioactive waste

Byproducts of nuclear processes that are radioactive and have no useful recyclable purpose (see *nuclear waste*).

radionuclide

Radioactive species of an atom characterized by the constitution of its nucleus specified by the number of protons, neutrons, atomic number, and mass number.

rem

Unit of dose equivalent equal to the product of the rad absorbed and the quality factor.

synchrotron

Roughly circular accelerator in which the particles travel in synchronized bunches at fixed radius.

tandem Van de Graaff or Dynamitron

Accelerator in which a charge exchange occurs in the terminal on either a thin foil or in a gas stripper tube.

tritium (H-3, ^3H)

Radioactive isotope of hydrogen having one proton and two neutrons. Tritium gas is produced in nuclear reactors and used to boost the explosive power of most modern nuclear weapons. It is also a constituent of irradiated water associated with reactor operations.

Van de Graaff accelerator

Accelerator in which constant high-voltage on a terminal in a pressurized tank accelerates the beam. The voltage is generated by charge carried on an insulating belt.

water-moderated reactor

Reactor in which water slows the speed of neutrons from fissioning atoms.

X-ray

(1) Ionizing electromagnetic radiation of external nuclear origin with energies less than 250 kilo electron volts. (2) Radiograph.

zero power

Operating a reactor to maintain a chain reaction at an extremely low power level producing very little heat. Zero power reactors are used as sensitive laboratory tools to pretest experimental loadings of test reactors and for other analytical purposes. Also called low power.