

ORAU TEAM Dose Reconstruction Project for NIOSH

Oak Ridge Associated Universities I NV5|Dade Moeller I MJW Technical Services

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06/09/2020	02	Revised to address the cancellation of ORAUT-PROC-0060. Incorporates onsite ambient dose data and air concentration data through 2018. Revised 1952 to 1972 tritium air concentration data. Revised to update environmental data tables and references before second formal internal review. Incorporates formal internal and NIOSH review comments. Constitutes a total rewrite of the document. Training required: As determined by the Objective Manager. Initiated by Stephen Spanos.

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ACRONYMS AND ABBREVIATIONS

aCi attocurie (1 \times 10⁻¹⁸ Ci)

AEC U.S. Atomic Energy Commission

AWE atomic weapons employer

Bq becquerel

CEDE committed effective dose equivalent

Ci curie

d day

DCG derived concentration guideline
DOE U.S. Department of Energy
DOL U.S. Department of Labor

EEOICPA Energy Employees Occupational Illness Compensation Program Act of 2000

EPA U.S. Environmental Protection Agency

Ge(Li) lithium-drifted germanium

hr hour

HT tritiated hydrogen gas HTO tritiated water vapor

ICT Insulating Core Transformer

in. inch

kCi kilocurie

L liter

LBNL Lawrence Berkeley National Laboratory
LLNL Lawrence Livermore National Laboratory

m meter

MeV megaelectron-volt, 1 million electron-volts

MEI maximally exposed individual

MFAPs mixed fission and activation products

mi mile
min minute
mrem millirem
mSv millisievert

nBq nanobecquerel

NESHAPs National Emissions Standards for Hazardous Air Pollutants

NIOSH National Institute for Occupational Safety and Health

ORAU Oak Ridge Associated Universities

PBq petabecquerel (1 x 10¹⁵ Bq)

s second

SEC Special Exposure Cohort

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SRDB Ref ID Site Research Database Reference Identification (number)

TBD technical basis document TBq terabecquerel (1 \times 10¹² Bq) TLD thermoluminescent dosimeter

U.S.C. United States Code

wk week

yr year

χ/Q atmospheric dispersion coefficient

§ section or sections

4.1 INTRODUCTION

Technical basis documents (TBDs) and site profile documents are not official determinations made by the National Institute for Occupational Safety and Health (NIOSH) but are rather general working documents that provide historical background information and guidance to assist in the preparation of dose reconstructions at particular U.S. Department of Energy (DOE) or Atomic Weapons Employer (AWE) facilities or categories of DOE or AWE facilities. They will be revised in the event additional relevant information is obtained about the affected DOE or AWE facility(ies), such as changing scientific understanding of operations, processes, or procedures involving radioactive materials. These documents may be used to assist NIOSH staff in the evaluation of Special Exposure Cohort (SEC) petitions and the completion of individual dose reconstructions under Part B of the Energy Employees Occupational Illness Compensation Program Act of 2000 (EEOICPA).

In this document the word "facility" is used to refer to an area, building, or group of buildings that served a specific purpose at a DOE or AWE facility. It does not mean nor should it be equated to an "AWE facility" or a "DOE facility." The term "AWE facility" is defined in EEOICPA to mean "a facility, owned by an atomic weapons employer, that is or was used to process or produce, for use by the United States, material that emitted radiation and was used in the production of an atomic weapon, excluding uranium mining or milling" [42 *United States Code* (USC) 7384I(5)]. On the other hand, a DOE facility is defined as "any building, structure, or premise, including the grounds upon which such building, structure, or premise is located—(A) in which operations are, or have been, conducted by, or on behalf of, the [DOE] (except for buildings, structures, premises, grounds, or operations ... pertaining to the Naval Nuclear Propulsion Program); and (B) with regard to which the [DOE] has or had—(i) a proprietary interest; or (ii) entered into a contract with an entity to provide management and operation, management and integration, environmental remediation services, construction, or maintenance services" [42 USC 7384I(12)]. The DOE determines whether a site meets the statutory definition of an AWE facility and the U.S. Department of Labor (DOL) determines if a site is a DOE facility and, if it is, designates it as such.

Under EEOICPA, a Part B cancer claim for benefits must be based on an energy employee's eligible employment and occupational radiation exposure at a DOE or AWE facility during the facility's designated time period and location (i.e., a "covered employee with cancer"). After DOL determines that a claim meets the eligibility requirements under Part B of EEOICPA, DOL transmits the claim to NIOSH for a dose reconstruction. EEOICPA provides, among other things, guidance on eligible employment and the types of radiation exposure to be included in an individual dose reconstruction. Under EEOICPA, eligible employment at a DOE facility includes individuals who are or were employed by DOE and its predecessor agencies, as well as their contractors and subcontractors at the facility [42 USC 7384I(11)]. Also under EEOICPA, the types of exposure to be included in dose reconstructions for DOE employees are those radiation exposures incurred in the performance of duty. As such, NIOSH includes all radiation exposures received as a condition of employment at DOE facilities in its dose reconstructions for covered employees, which may include radiation exposures related to the Naval Nuclear Propulsion Program at DOE facilities, if applicable. This is because NIOSH does not determine the fraction of total measured radiation exposure at a DOE facility that is contributed by the Naval Nuclear Propulsion Program at the DOE facility during a specified period of time for inclusion in dose reconstruction.

NIOSH does not consider the following types of exposure as those incurred in the performance of duty as a condition of employment at a DOE facility. Therefore these exposures are not included in dose reconstructions for covered employees [NIOSH 2010]:

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

4.1.1 Purpose

This technical basis document (TBD) documents historical practices at Lawrence Livermore National Laboratory (LLNL) in Livermore, California, and provides information for the evaluation of environmental radiation exposure data. It can serve as a supplement to, or substitute for, individual monitoring data.

LLNL began operations in 1952 as the University of California Radiation Laboratory, which was a branch of what is now the Lawrence Berkeley National Laboratory (LBNL). It was later known as the Lawrence Radiation Laboratory and the Lawrence Livermore Laboratory before assuming its current name. The University of California operated LLNL for DOE and its predecessor agencies from 1952 through September 2007, when a partnership of the University of California, Bechtel Corporation, Babcock and Wilcox, the URS Corporation, and Battelle Memorial Institute assumed this role. Lawrence Livermore National Security has operated the site since then. Throughout its history, LLNL has processed and handled a variety of radionuclides, including uranium and transuranic elements, mixed fission and activation products (MFAPs), and accelerator-produced isotopes. This TBD discusses two sites: the main site (Site 200) where research and development activities have taken place since 1952 and Site 300 where explosive testing experiments have occurred. Site 300 explosive testing included depleted and natural uranium, natural thorium, and tritium triggers. Site 300 began operations in 1955. This TBD addresses potential for internal dose from the breathing of airborne concentrations of radionuclides that have been released on the LLNL site.

The receptors of concern in this TBD are unmonitored workers, namely LLNL employees who did not wear external dosimetry or who were not monitored for internal exposures. Initially, LBNL (Site 100) provided LLNL with beta/gamma film dosimeters including processing and pocket ionization chambers (Thompson 1953). The practice of providing film and pocket dosimeters to all workers has been in effect since March 1953 (Thompson 1953). In 1958, film badges became part of the security badge (Nolan 1958), which effectively mandated that all workers wear film dosimeters at all times. There is ongoing collaboration between LLNL and LBNL. To provide the basis for estimating the environmental dose for years when monitoring did not occur, this TBD provides annual intakes to 2018 and ambient external doses from 1952 (1955 for Site 300) to 2018 (the last year with publicly available ambient external data for the revision to address cancellation of ORAUT-PROC-0060).

4.1.2 <u>Scope</u>

Occupational environmental dose refers to dose from exposures workers received while on the site but outside the facilities at LLNL from elevated ambient radiation, facility discharges to the environment, and resuspension of radionuclides in soils. Effluents can result in internal and external exposures by inhalation of airborne radionuclides, ingestion of radionuclides, and exposure to external radiation. This TBD describes the estimated annual intakes for inhalation exposure and the estimated radiation doses as a result of ambient exposures at LLNL. Environmental measurements do not distinguish sources of emissions and, therefore, reflect air concentrations from nearby as well as distant sources. The estimates of emissions were useful in filling some gaps in measurement data and were critical to estimating exposures before the start of comprehensive and routine measurement data reports.

Section 4.2 of this TBD contains detailed information about the collection and analysis of air samples and the use of this information for dose reconstruction. There was a potential for ingestion of radioactive materials from ingestion of water that might have contained tritium (Section 4.3). Section 4.4 describes the potential external doses from sources of radiation outside the process buildings.

Attributions and annotations, indicated by bracketed callouts and used to identify the source, justification, or clarification of the associated information, are presented in Section 4.5.

4.1.3 Special Exposure Cohort

The Secretary of the U.S. Department of Health and Human Services has designated the following classes of LLNL employees as additions to the SEC:

Employees of the DOE, its predecessor agencies, and DOE contractors or subcontractors who were monitored for radiation exposure while working at the Lawrence Livermore National Laboratory from January 1, 1950, through December 31, 1973, for a number of workdays aggregating at least 250 workdays or in combination with workdays within the parameters established for one or more other classes of employees in the Special Exposure Cohort (Leavitt 2008).

All employees of the Department of Energy, its predecessor agencies, and their contractors and subcontractors who worked at the Lawrence Livermore National Laboratory in Livermore, California from January 1, 1950 through December 31, 1973, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees in the Special Exposure Cohort (Sebelius 2010).

All employees of the Department of Energy, its predecessor agencies, and its contractors and subcontractors who worked in any area at the Lawrence Livermore National Laboratory in Livermore, California, during the period from January 1, 1974 through December 31, 1989, for a number of work days aggregating at least 250 work days, occurring either solely under this employment or in combination with work days within the parameters established for one or more other classes of employees in the SEC (Burwell 2016).

NIOSH determined that doses to monitored workers from MFAPs at LLNL cannot be reconstructed with sufficient accuracy from 1950 through 1973 (NIOSH 2007). In addition, NIOSH determined that doses to unmonitored workers from MFAPs at LLNL cannot be reconstructed with sufficient accuracy between 1950 and 1973, inclusive (NIOSH 2010b). Further, NIOSH determined that internal doses from ²³³U at LLNL cannot be reconstructed with sufficient accuracy from 1974 through 1989 (NIOSH 2016).

Each SEC class includes all workers during the SEC period. Because of the identified dose reconstruction infeasibility, all dose reconstructions for all workers having employment during an SEC period are considered partial dose reconstructions. If monitoring data are available for workers in the SEC, dose is to be assigned as appropriate based on such data. However, such dose reconstructions are considered partial dose reconstructions because exposure to MFAPs and ²³³U during the respective LLNL SEC periods cannot be reconstructed.

4.2 INTERNAL DOSE FROM ONSITE ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS

4.2.1 <u>Ambient Air Sample Collection Network</u>

4.2.1.1 Before 1959

A review of records indicates that the earliest available environmental monitoring data for the LLNL main site was 1959 (Beaufait 1961) and that for Site 300 was 1961 (LRL 1962). Before the 1980s, the LLNL mission was almost exclusively weapons research. Therefore, it was assumed that the radionuclide emission rate would be proportional to the weapons-related activity at the site from design, fabrication, and analysis of samples that were retrieved after a test. The peak for testing at LLNL was from 1962 through 1971 with an average of 31 tests per year (DOE 2000). From 1953

through 1961, LLNL sponsored an average of 7 tests per year (DOE 2000). During the entire period of LLNL-sponsored weapons testing from 1953 through 1992 the average was 14.5 per year.

It was assumed that all activity that was detected at the monitoring stations was from LLNL activities. This assumption is conservative because this was during a time of high fallout due to atmospheric testing, and in many cases offsite locations had higher activity than onsite locations due to the fallout. Therefore, for the early years before monitoring, dose reconstruction should use the highest measured results from the later years in which monitoring occurred. Section 4.2.3.1 provides estimated intakes based on the highest measured activity, despite location, as being favorable to claimants. For workers that should have been monitored, intakes based on co-exposure data might be more appropriate than environmental data.

4.2.1.2 1959 through 1971

Air samples were collected in seven locations around the main site, including two perimeter locations (LRL 1962). The two perimeter locations, southeast perimeter and west perimeter, were collected continuously for a 7-day period on HV-70 paper 4 in. wide by 9 in. long (LRL 1962). The other air sampling stations were at a variety of distances from along the perimeter to as far as 5 mi from the perimeter (LRL 1962). Table 4-1 lists the seven onsite sampling stations this analysis used for estimates of occupational environmental exposures, and Figure 4-1 shows their locations as black dots. The SALV and MESQ locations were established as early as 1961 for perimeter air monitoring (LRL 1962). An air sampling station was added on the east side of LLNL in 2000. This station, CRED, was near the area representative of the maximally exposed individual (MEI) for LLNL.

Table 4-1. Main site ambient air sampling station locations (Gallegos et al. 2002).

Designation	Location	Direction from center of LLNL
COW	At security fence north of outer loop road	N
MET	Meteorological station	NW
VIS	At security fence near east entrance	Е
CAFÉ	At security fence near south entrance	S
SALV	At security fence near southeast perimeter fence	SE
MESQ	At security fence near west entrance	W
CRED	Solar evaporators for plutonium waste processing; representative of MEI	SE

At Site 300, six air sampling stations were established in 1962, each within the site perimeter. The air sampling equipment at Site 300 was equivalent to the equipment at the main site. Table 4-2 lists these stations, and Figure 4-2 shows their locations. The Bunker 801E, ECP, EOBS, GOLF, WCP, and WOBS locations were established in 1962. As stated above, this was during a time of high fallout due to atmospheric testing, and in many cases offsite locations had higher activity than onsite locations due to the fallout. Therefore, Section 4.2.3.1 presents intakes estimated from the highest measured activity, despite location, as being favorable to claimants.

4.2.1.3 After 1971

In 1971, LLNL established a network of permanent stations to collect air samples from the site perimeter and other locations inside the site (Gallegos et al. 1992). LLNL collected and analyzed air samples for gross alpha and gross beta radiations, such as ⁹⁰Sr, ²³⁸Pu, and ²³⁹⁺²⁴⁰Pu, and ²³⁵U and ²³⁸U. The existing monitoring networks were established in 1992 for surveillance of air particulates and tritium in and around the Livermore site and Site 300, as well as in the surrounding Livermore Valley and the City of Tracy (Gallegos et al. 1992). Tables 4-1 and 4-2 list the stations, and Figures 4-1 and 4-2 show their locations; black triangles represent tritium monitoring stations. As stated above, this was during a time of high fallout due to atmospheric testing and in many cases

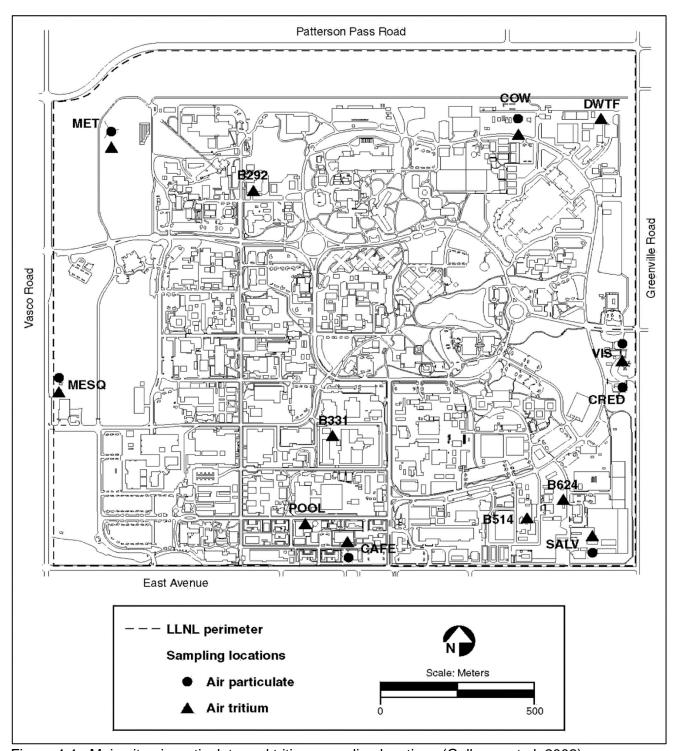


Figure 4-1. Main site air particulate and tritium sampling locations (Gallegos et al. 2002).

offsite locations had higher activity than onsite locations due to the fallout. The highest measured activity based on all onsite locations is presented, as being favorable to claimants.

4.2.1.4 Purpose and Placement of Sampling Locations

The principal purpose of the ambient air monitoring network was to assess if air emissions from LLNL affected the air quality in the surrounding area. Air samples were collected from locations where a significant concentration of effluents from LLNL operations could reasonably be detected regardless

Table 4-2. Site 300 ambient air sampling station locations (Gallegos et al. 2002).

Designation	Location	Direction from center of Site 300
СОНО	Near south perimeter of facility; representative of MEI	S
EOBS	Northeast of Bunker 801E	NE
ECP	Center of Site 300	Е
WCP	Test Area	W
GOLF	South perimeter near fence	S
TFIR	Offsite location in City of Tracy	E
NPS	North of Bunker 801E	N
WOBS	West of Bunker 850	W
801E	East of Bunker 801E	E

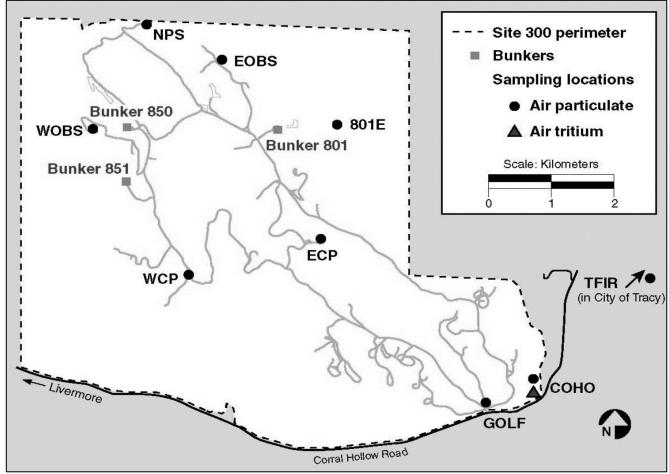


Figure 4-2. Site 300 and downtown Tracy air particulate and tritium sampling locations (Gallegos et al. 2002).

of local meteorology (Griggs and Buddemeier 1986). This demonstrated compliance with DOE derived concentration guidelines (DCGs) or U.S. Environmental Protection Agency (EPA), State of California, and (as of 1997) U.S. Nuclear Regulatory Commission regulations for airborne releases to the general public around LLNL. There were seven onsite monitoring stations (Griggs and Buddemeier 1986). This TBD considers only those ambient air monitoring locations inside the LLNL security fence for worker intake. The largest air concentration value was reported based on all onsite monitoring locations, to be favorable to claimants. Table 4-1 lists these locations.

The Livermore site radiological air particulate sampling network consisted of seven samplers at the perimeter with one (CRED) serving as the sitewide MEI as reported for National Emissions Standards

for Hazardous Air Pollutants (NESHAPs) monitoring. CRED was in the southeast quadrant in an area of known plutonium contamination from historical operations that included solar evaporators for plutonium-containing liquid waste.

The Site 300 air particulate monitoring network included eight sampling units that were placed around the site and near firing tables and one in downtown Tracy. Site 300 is in a remote area and access is limited. LLNL based the selection of the monitoring sites on safety, power, and access considerations. The COHO location served as the sitewide MEI location for NESHAPs reporting purposes. LLNL added two sampling systems in the Livermore Valley in July 1997 as part of the new low-volume radiological air particulate sampling network (Althouse et al. 2001). These samplers were generally upwind of the Livermore site. LLNL used the results to establish background levels of gross alpha and beta activity for direct comparison to emissions from the air effluent samplers.

In addition, LLNL maintained 12 continuously operating airborne tritium samplers on the main site to assess current activities that influence environmental impacts. These stations were deployed in 1973; specific stacks were sampled for tritium as early as 1971 (Gede and Gildea 1980; SAIC 1993).

4.2.1.5 Radionuclides of Significance

The LLNL environmental monitoring program identified radionuclides of significance. The program analyzed air samples for the presence of gross alpha and gross beta radiations from 1959 through 1970 and for specific isotopes afterward, which represented more than 90% of the LLNL radioactive materials inventory. It analyzed air samples specifically for the presence of tritium, ²³⁸Pu, ²³⁹⁺²⁴⁰Pu, and isotopes of uranium. From 1959 through 1970, dose reconstructors should select the assigned dose from the higher of two calculated doses: one using the gross alpha concentration to calculate the presence of ²³⁹Pu, and a second using ²³⁴U. The gross beta concentration should be assumed to result from ⁹⁰Sr [1].

For Site 300, radionuclides of significance were selected according to the inventory of radioactive materials and the type of work that was performed at that site. Specifically, the presence of depleted uranium was indicated in the analysis of ²³⁸U and ²³⁵U. Based on a review of the ratios of ²³⁵U to ²³⁸U air sample concentration data in the environmental reports, depleted uranium was predominant in air samples through about 1985 at Site 300. After 1985, the Site 300 uranium air sample concentration data indicated that natural uranium was most prevalent. The main site uranium air sample concentration data indicated that natural uranium was predominant for all years of available data. For estimates based on this TBD, the dose reconstructor should assign the dose that is associated with ²³⁴U for natural uranium, which is favorable to claimants. Through 1970, dose reconstruction should use the gross alpha concentration for Site 300 to calculate the presence of ²³⁴U; it is assumed that the maximum radiation dose is assigned as 100% ²³⁴U versus a varying percentage of ²³⁴U and ²³⁵U [2]. Data are listed in the tables in Section 4.2.3.2. Assume the gross beta concentration is ²³⁴Th [3]. After 1970, the ²³⁸U and ²³⁵U mass-based air concentration data were converted to activity concentration based on the specific activities in the environmental reports. The activity of ²³⁴U was determined by multiplying the ²³⁸U activity by the ratio of the activity fraction of ²³⁴U to ²³⁸U for natural uranium in ORAUT-TKBS-0035-5. Lawrence Livermore National Laboratory - Occupational Internal Dose (ORAUT 2010). The ²³⁴U, ²³⁸U, and ²³⁵U activities were then summed and reported as ²³⁴U. These values are presented later in Tables 4-7 and 4-8.

4.2.2 Sampling and Analysis Methods

LLNL used several different networks, each representing a general location and type of analysis, to perform environmental air sampling. There were separate networks for sampling radiological particulates and beryllium particulates at the Livermore site and Site 300 as well as a low-volume radiological air sampling network, a tritium-sampling network in Livermore, and one tritium-sampling location at Site 300. Four collection media were employed: glass-fiber filters for radiological

particulates, cellulose filters for beryllium particulates, membrane filters for low-volume radiological particulates, and silica gel for tritium. All monitoring networks used continuously operating samplers (Gallegos et al. 1992).

As outlined in the *Environmental Regulatory Guide for Radiological Effluent Monitoring and Environmental Surveillance* (DOE 1991), data for gross alpha, gross beta, and gamma isotopes on air filters were used only as trend indicators; specific radionuclide analysis occurred for plutonium and uranium dependent on location. All analytical results were reported as a measured concentration per volume of air. When activity was less than the minimum detectable concentration, the calculated value was reported. Particle size distributions were not determined because the estimated effective dose equivalent to the MEI is well below the 0.01-mSv (1-mrem) allowable limit (DOE 1991). The analytical laboratory used ²³⁰Th and ⁹⁰Sr as calibration sources to determine alpha and beta counting efficiencies, respectively. Annual counting efficiency measurements were made for each detector. Periodic crosschecks used EPA-certified standards. Background and efficiency checks occurred daily, and a matrix and method blank were run with every batch of 20 samples. LLNL kept records of background and counting efficiency variations in the counting equipment. The analytical laboratory reported the actual instrumentation values, which included negative results when background measurements were higher than those for the filters (DOE 1991).

Concentrations of various airborne radionuclides were measured at Livermore site perimeters, at offsite locations near the Livermore site, and at Site 300. From 1959 to 1964, air samples were collected from two perimeter locations and nine offsite locations on 4- by 9-in. HV-70 paper at a sample flow rate of about 4 ft³/min of air (LRL 1964). As discussed in Section 4.2.1.1, no monitoring data were found for the years before 1959 for the main site or before 1961 for Site 300. The samples were collected for 7 days. After a 4-day delay for decay of radon and thoron progeny, gross alpha and beta activities on the filters were determined with an automatic gas-flow proportional counter. Monthly composites of perimeter filters were counted for specific gamma-emitting radionuclides using a Ge(Li) detector equipped with Compton suppression.

After gamma counting, the perimeter filters were grouped by sampling location. The individual samples were analyzed for the presence of ²³⁹Pu, ²³⁸Pu, ¹³⁷Cs, ²³⁵U, and ²³⁸U. In July 1964, four sample locations were added on the perimeter; six locations around the perimeter were sampled (Hughey 1965). The six samplers on the Laboratory perimeter used 0.052 m² Whatman-41 cellulose filters. The particulates were collected on these filters using an average air flow rate of 700 L/min (Lindeken et al. 1978). Tritium was collected on columns packed with silica gel using an air flow rate of 0.5 L/min. Particulate filters were changed each week at all locations, and tritium samplers were changed every 2 weeks. From 1973 to 1976, area samples for tritium were exchanged each week. After 1976, tritium samples were exchanged every 2 weeks (SAIC 1993). Duplicate quality assurance samplers operated in parallel with the permanent sampler at each site; samples from these duplicates were analyzed to confirm results (Lindeken et al. 1978).

In April 1997, the radiological air particulate sampling filter media changed from cellulose to glass fiber. However, blank glass-fiber filters contain nontrivial amounts of naturally occurring radiological isotopes including ²³⁴U, ²³⁵U, ²³⁸U, ⁴⁰K, ²²⁶Ra, ²²⁸Ra, and ²²⁸Th (Althouse 1998). In fact, the amount of these naturally occurring isotopes in the filters is often greater than the amount the filter captures from the air. The filters are cut in half, and half of each filter is retained for beryllium analysis.

Gross alpha and gross beta activities were determined by gas-flow proportional counting, plutonium and uranium isotopes by alpha spectrometry, gamma emitters by gamma spectroscopy, and tritium by liquid scintillation. In 2001, a correction factor was applied to tritium concentrations to account for dilution of the collected tritium from air moisture by an unknown quantity of water in supposedly dry silica gel (Althouse et al. 2001). On average, the corrected concentrations were 1.6 times higher than uncorrected concentrations.

Gross alpha, gross beta, and gamma emitters on air filters were used as trend indicators; specific radionuclide analyses were completed for plutonium and uranium after 1971. Radiological analytical results were reported as a measured activity per volume of air. Particle size distributions on air samples were not determined because the estimated effective dose equivalent to the MEI (from the total particulate) was below the 10-mrem committed effective dose equivalent (CEDE) (Althouse et al. 2001).

Portions of the glass-fiber filters from the Livermore locations were analyzed for the presence of ²³⁹⁺²⁴⁰Pu. Similarly, portions of the glass-fiber filters from Site 300 were analyzed for the presence of ²³⁵U and ²³⁸U. The filters were placed in a muffle furnace to reduce organic content and then dissolved in a mixture of nitric and hydrochloric and/or hydrofluoric acids. Plutonium and uranium were separated by an ion exchange process. Each separated element was purified further by ion exchange. It was then electroplated onto a stainless-steel disk and analyzed by alpha spectrometry.

For gamma scanning, a site composite was created using all of the weekly glass fiber filters for Site 300 perimeter locations (801E, ECP, EOBS, GOLF, NPS, WCP, and WQBS). This composite was prepared for analysis in the same manner as that for plutonium and uranium samples. After it was muffled and digested, it was counted for more than 40 gamma-emitting radionuclides using Ge(Li) detectors. In addition to gamma scanning, the Site 300 composite was analyzed for plutonium.

Duplicate radiological quality assurance samples were processed to confirm the precision of the analytical results obtained from the samplers. A duplicate sampler was operated for 2 months in parallel with the permanent sampler at a given site. In addition, a trip blank was collected during each collection trip. The trip blanks and duplicates were processed in the same manner as that for the routine samples and analyzed for the same radiological parameters.

4.2.3 <u>Estimation of Potential Annual Intakes from Airborne Radionuclides</u>

This section discusses estimation of annual intakes from the air monitoring results for all air surveillance locations at the Livermore site and Site 300. This TBD relies on several data sources for estimating ambient airborne radionuclide concentrations and for annual airborne releases by radionuclide. The principal sources were the annual environmental reports for LLNL from 1959 to 2018.¹ The analysis for this TBD collected measurements from the air monitoring locations in Tables 4-1 and 4-2 from these documents and analyzed them in accordance with the collection and analysis methods Section 4.2.2 describes. If data were not available or additional information was necessary, these documents were supplemented by information from DOE or the University of California.

Factors other than transport and release rates have influenced the gradual reduction in the measured air concentrations over the history of LLNL. Over the past 40 years of monitoring, concentrations of airborne radioactive materials have decreased by more than 3 orders of magnitude. These factors include:

- Improvements in monitoring methods that reduced minimum detectable concentrations;
- Reductions in releases of naturally occurring radioactive material from a nearby fossil-fuel generating plant, which has improved its environmental controls over the years; and
- Reductions in the effects of atmospheric weapons testing including radioactive decay of fallout.

1 The References section lists all directly cited references, then separately lists the environmental reports in date order.

The method for calculation of the annual intakes from onsite atmospheric radionuclide concentrations was applied directly to available air sampling data. Earlier LLNL environmental reports provided annual average air concentrations through 1993. After this time, annual median air concentrations were reported. For simplicity, they are referred to as "medians" for each year. The largest annual median for the year, over all locations, is referred to as "maximum annual median air concentration."

The following factors limited the estimates of airborne concentrations at specific locations around the LLNL site using traditional transport modeling approaches:

- Number of release points;
- Characteristics of the release points, which include stacks, vents, and other emission sources;
- Limited number of air sampling locations;
- Relatively short distances between the release points and the onsite receptor locations; and
- Density and configurations of buildings at the site.

Sections 4.2.3.1 and 4.2.3.2 provide information on gross alpha and gross beta inhalation exposures for the periods from 1952 to 1958 for the main site, and from 1955 to 1960 for Site 300 and from 1959 to 1970 for the main site, and from 1961 to 1970 for Site 300, respectively; the estimates for the earlier period are based on the data from the later period. Section 4.2.3.3 discusses tritium exposures for the period from 1952 to 1972, before tritium monitoring began. Section 4.2.3.4 provides estimated inhalation exposures based on air monitoring for tritium starting in 1973 and for plutonium and uranium for 1971 to 2018.

All inhalation intakes in the tables in the following sections are based on a 50-hour workweek, which was chosen based on a review of telephone interviews for LLNL claims (ORAUT 2020c). Over half of the telephone interviews indicated working more than 40 hr/wk. The inhalation exposure of 50 hr/wk is favorable to claimants and applicable for most cases.

The 1952 to 1970 gross alpha and gross beta air concentration data in Sections 4.2.3.1 and 4.2.3.2, the 1973 to 2018 tritium air concentration data, and the 1971 to 2018 plutonium and uranium air concentration data in Section 4.2.3.4 are supported by ORAUT (2020a). The 1952 to 1972 tritium air concentration data in Section 4.2.3.3 are supported by ORAUT (2020b).

4.2.3.1 Gross Alpha and Gross Beta, before 1959

For the main site before 1959, and for Site 300 before 1961, environmental air monitoring samples were not available. The estimated air concentrations and potential intakes of gross alpha and gross beta activity before 1959 for the main site and before 1961 for Site 300 are listed in Tables 4-3 and 4-4. The annual intakes assumed a breathing rate of 3,000 m³/yr based on 20 L/min for 50 hr/wk and 50 wk/yr with a geometric standard deviation for these data of 3 [4]. These data are based on using the highest annual average of the measured data for years during which environmental monitoring occurred. All assigned environmental internal doses for the main site before 1959, and for Site 300 before 1961, are considered partial dose assessments due to the LLNL SECs for MFAPs and ²³³U. The intakes in Tables 4-3 and 4-4 may be adjusted to reflect work other than 50 hr/wk. The intake rates in Tables 4-3 and 4-4 would be multiplied by the ratio of the hours worked per week to 50 hr/wk.

4.2.3.2 Gross Alpha and Gross Beta, 1959 to 1970

Data from the environmental reports were available for 1959 to 1970 for the main site and for 1961 to 1970 for Site 300. The air monitoring data provided no isotopic analysis; the data were reported only for the gross alpha and gross beta radiations. Table 4-5 lists air concentrations and estimated intakes for the LLNL site, and Table 4-6 lists the values for Site 300. All assigned environmental internal doses for 1959 to 1970 are considered partial dose assessments due to the LLNL SEC classes for MFAPs and ²³³U. The intakes in Tables 4-5 and 4-6 may be adjusted to reflect work other than

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Table 4-3. Main site estimated air concentrations and annual median intakes via inhalation, 1952 to 1958.^a

Year	Gross alpha (Pu-239 or U-234) air concentration (Bq/m³)	Gross alpha (Pu-239 or U-234) annual inhalation intake (Bq/yr)	Gross beta (Sr-90) air concentration (Bq/m³)	Gross beta (Sr-90) annual inhalation intake (Bq/yr)
1952	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1953	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1954	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1955	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1956	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1957	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1958	8.510E-04	2.553E+00	9.620E-01	2.886E+03

a. All intakes are based on a 50-hour workweek. The inhalation doses calculated may be adjusted based on the guidance in Section 4.2.3.1.

Table 4-4. Site 300 estimated air concentrations and annual median intakes via inhalation, 1955 to 1960.^a

Year	Gross alpha (U-234) air concentration (Bq/m³)	Gross alpha (U-234) annual inhalation intake (Bq/yr)	Gross beta (Th-234) air concentration (Bq/m³)	Gross beta (Th-234) annual inhalation intake (Bq/yr)
1955	4.810E-04	1.443E+00	1.554E-01	4.662E+02
1956	4.810E-04	1.443E+00	1.554E-01	4.662E+02
1957	4.810E-04	1.443E+00	1.554E-01	4.662E+02
1958	4.810E-04	1.443E+00	1.554E-01	4.662E+02
1959	4.810E-04	1.443E+00	1.554E-01	4.662E+02
1960	4.810E-04	1.443E+00	1.554E-01	4.662E+02

a. All intakes are based on a 50-hour workweek. The inhalation doses calculated may be adjusted based on the guidance in Section 4.2.3.1.

Table 4-5. Main site maximum annual median air concentrations and estimated annual intakes via inhalation, 1959 to 1970.^a

Vaar	Gross alpha (Pu-239 or U-234) air concentration	Gross alpha (Pu-239 or U-234) annual inhalation	Gross beta (Sr-90) air concentration	Gross beta (Sr-90) annual inhalation
Year	(Bq/m³)	intake (Bq/yr)	(Bq/m³)	intake (Bq/yr)
1959	7.400E-04	2.220E+00	2.701E-01	8.103E+02
1960	8.510E-04	2.553E+00	9.620E-01	2.886E+03
1961	3.700E-04	1.110E+00	3.293E-02	9.879E+01
1962	2.331E-04	6.993E-01	9.250E-02	2.775E+02
1963	1.073E-04	3.219E-01	1.221E-01	3.663E+02
1964	1.554E-04	4.662E-01	2.738E-02	8.214E+01
1965	4.070E-05	1.221E-01	5.476E-03	1.643E+01
1966	3.700E-05	1.110E-01	2.627E-03	7.881E+00
1967	1.480E-05	4.440E-02	4.662E-03	1.399E+01
1968	3.700E-05	1.110E-01	4.366E-03	1.310E+01
1969	1.480E-05	4.440E-02	4.070E-03	1.221E+01
1970	1.480E-05	4.440E-02	7.844E-03	2.353E+01

All intakes are based on a 50-hour workweek. The inhalation doses calculated may be adjusted based on the guidance in Section 4.2.3.2.

50 hr/wk. The intake rates in Tables 4-5 and 4-6 would be multiplied by the ratio of the hours worked per week to 50 hr/wk.

The gross alpha and gross beta inhalation intakes in Tables 4-5 and 4-6 should be assigned as a lognormal distribution with a geometric standard deviation of 3.

The isotopes of significance at Site 300 are ²³⁸U and ²³⁵U, both of which decay by alpha radiation.

Table 4-6. Site 300 maximum annual median air concentrations and annual intakes via inhalation, 1961 to 1970.^a

Year	Gross alpha (U-234) air concentration (Bq/m³)	Gross alpha (U-234) annual inhalation intake (Bq/yr)	Gross beta (Th-234) air concentration (Bq/m³)	Gross beta (Th-234) annual inhalation intake (Bq/yr)
1961	4.810E-04	1.443E+00	2.960E-03	8.880E+00
1962	3.700E-04	1.110E+00	1.369E-01	4.107E+02
1963	1.073E-04	3.219E-01	1.554E-01	4.662E+02
1964	1.665E-04	4.995E-01	2.220E-02	6.660E+01
1965	3.700E-05	1.110E-01	6.290E-03	1.887E+01
1966	4.440E-05	1.332E-01	3.182E-03	9.546E+00
1967	3.700E-05	1.110E-01	5.550E-03	1.665E+01
1968	7.400E-06	2.220E-02	5.069E-03	1.521E+01
1969	3.700E-05	1.110E-01	5.735E-03	1.721E+01
1970	3.700E-05	1.110E-01	8.880E-03	2.664E+01

a. All intakes are based on a 50-hour workweek. The inhalation doses calculated may be adjusted based on the guidance in Section 4.2.3.2.

4.2.3.3 Tritium, 1952 to 1972

Environmental air monitoring samples were not analyzed for tritium before 1973. A tritium dose reconstruction was performed in 2007 by LLNL (Peterson 2007). According to this study, although not reported to the public, the releases of tritium from significant sources (e.g., both the original Tritium Facility that opened in 1953 in what is now Building 231 and the present Tritium Facility [Building 331] that began operations in 1959) were estimated or measured during these years and, starting at least in 1956, were reported quarterly to the U.S. Atomic Energy Commission (AEC). Before 1956, tritium releases were estimated (Peterson 2007).

The sources of tritium were the stacks for Buildings 331 (north and south), 231, and 212; Building 212 room air; and area sources such as Buildings 231, 331, 514, and 612 along with the Evaporation Trays. Routine tritiated water vapor (HTO), and routine tritiated hydrogen gas (HT) releases were reported in the study. The majority of tritium released was from the Building 231 and Building 331 stacks. A comparison of the HTO and HT Buildings 231 and 331 stack releases indicate that from 1953 through 1960 approximately 37% of the tritium released was HTO. From 1961 through 1972, approximately 54% of the released tritium was HTO. While the HTO and HT releases were within the same order of magnitude for 1953 through 1972, HT releases were not considered in this TBD because HTO has a dose coefficient 10,000 times that of HT (ICRP 1995).

The LLNL Discovery Center (Location VIS) location was assessed for the sitewide maximum exposed individual (Peterson 2007). Atmospheric dispersion coefficients (χ /Q) in s/m³ and annual HTO releases for 1953 through 1972 were determined for each LLNL tritium release. The annual tritium air concentrations were determined by the product of the annual tritium release multiplied by the appropriate χ /Q and summed. The χ /Q coefficients for Building 331 south stack were used as the basis for determining the Building 331 stack release concentrations because they were higher than those of the north stack. There were no tritium releases estimated or reported in 1952. LLNL began working with tritium late in 1952, very shortly after the laboratory opened, when a demand for a source of 14-MeV neutrons arose as a consequence of the nascent laboratory's entry into thermonuclear weapons research (Gede and Gildea 1980). Routine releases from significant sources of tritium at LLNL did not start until 1953 when the original Tritium Facility began operations (Peterson 2007). Any

unreported tritium releases in late 1952 would likely have been small in comparison with routine releases that began in 1953 and later. The largest tritium intake was 10,090 Bq/yr for 1957 (ORAUT 2020b). This results in an annual tritium dose of less than 0.001 rem. Therefore, the 1952 through 1972 annual tritium doses are all less than 0.001 rem. Radionuclides contributing doses less than 0.001 rem are not considered significant. The main site 1952 through 1972 tritium air concentrations and annual intakes are not listed for these years. All assigned environmental internal doses from 1952 to 1972 are considered partial dose assessments due to the LLNL SEC classes for MFAPs and 233U.

No tritium release data are available for Site 300 for 1955 to 1972. The Site 300 tritium doses are assumed to be the same as those for the main site. This is based on Section 4.3.2.4, which indicates that the Site 300 tritium values from 1973 and after are consistently less than 10% of the main site values during this time. Since the main site 1955 through 1972 annual tritium doses are all less than 0.001 rem, the Site 300 1955 through 1972 tritium air concentrations and annual intakes are not listed for these years.

4.2.3.4 Tritium, Uranium, and Plutonium, after 1970

Estimates of offsite doses to members of the public from the 1971 to 2018 environmental reports (see the References section) indicated that the potential internal dose from airborne releases to LLNL workers should be relatively low (10-mrem CEDE or less). Air data from nearby locations and at the security fence were consistent from location to location but dependent on year. This provided reasonable approximation of general airborne radioactivity and establishment of trends as a function of time.

Samples were collected and analyzed for airborne radionuclides at the perimeter of the facility. Air sample measurements were available from onsite diffuse source locations such as tritium for 1991 through 2007 for Buildings 292, 331, 514, and 624 and plutonium for 1991 through 1999 for Building 531 and air sampling location CRED. The higher air concentrations between the diffuse sources and perimeter locations were assigned for these years. Table 4-7 lists main site concentrations for $^{239+240}$ Pu, and 234 U. This TBD assumed that a worker inhaled 20 L of air per minute for 50 hr/wk or approximately 3.0 × 106 L of air per year (3,000 m³/yr). For tritium, the total uptake by the body was increased 50% to account for absorption by the skin in addition to the inhalation pathway (Gede and Gildea 1980). The largest main site tritium intake was 7,470 Bq/yr for 1998 (ORAUT 2020a). This results in an annual tritium dose of less than 0.001 rem. Therefore, the main site annual tritium doses after 1970 are all less than 0.001 rem. Radionuclides contributing doses less than 0.001 rem are not considered significant. The tritium air concentrations and annual intakes after 1970 are not listed for these years. Figure 4-3 shows how the concentration of $^{239+240}$ Pu decreased from 1986 through 2005. The concentration dropped by more than 20 times over the 20 years of sampling.

As Section 4.2.1.5 describes, the isotopes of significance at LLNL were ²³⁹⁺²⁴⁰Pu and ²³⁴U, which decay by alpha radiation. Isotopes that decay by beta radiation were determined to be progeny of these two parent isotopes.

Due to the LLNL SEC classes, all assigned environmental internal doses for 1971 to 1973 from MFAPs, and those from ²³³U from 1971 to 1989, are considered partial dose assessments.

Tables 4-7 and 4-8 indicate the potential for exposure to inhalation of airborne radionuclides at the main site and Site 300 respectively. The Site 300 tritium doses are not listed because the Site 300 values are consistently less than 10% of the main site values, and all main site annual tritium doses after 1970 are less than 0.001 rem. After 1970, only alpha-emitting radionuclides are listed. This is because the gross beta annual intakes after 1970 trend down to where the resulting internal doses are less than 0.001 rem. Radionuclides contributing doses less than 0.001 rem are not considered significant. In years for which air concentration data were unavailable, release data and maximum air

Table 4-7. Main site maximum annual median air concentrations and annual intakes via inhalation, 1971 to 2018. a,b,c

	Pu-239+240	Pu-239+240	U-234 ^d	U-234 ^d
	air concentration	annual inhalation	air concentration	annual inhalation
Year	(Bq/m³)	intake (Bq/yr)	(Bq/m³)	intake (Bq/yr)
1971	2.627E-06	7.881E-03	9.544E-06	2.863E-02
1972	1.184E-06	3.552E-03	1.646E-05	4.937E-02
1973	2.664E-04	7.992E-01	1.645E-05	4.936E-02
1974	3.108E-06	9.324E-03	1.333E-05	3.999E-02
1975	1.258E-06	3.774E-03	1.267E-05	3.801E-02
1976	1.036E-06	3.108E-03	1.481E-05	4.443E-02
1977	1.554E-06	4.662E-03	2.139E-05	6.417E-02
1978	1.961E-06	5.883E-03	2.698E-05	8.095E-02
1979	8.510E-07	2.553E-03	1.432E-05	4.295E-02
1980	5.180E-07	1.554E-03	1.267E-05	3.801E-02
1981	6.660E-07	1.998E-03	1.974E-05	5.923E-02
1982	2.220E-07	6.660E-04	1.777E-05	5.332E-02
1983	8.880E-07	2.664E-03	2.452E-05	7.356E-02
1984	1.480E-07	4.440E-04	1.580E-05	4.739E-02
1985	2.220E-07	6.660E-04	1.465E-05	4.394E-02
1986	1.517E-07	4.551E-04	1.445E-05	4.335E-02
1987	1.850E-07	5.550E-04	1.688E-05	5.065E-02
1988	1.184E-07	3.552E-04	3.624E-05	1.087E-01
1989	1.425E-07	4.274E-04	1.442E-05	4.325E-02
1990	1.517E-07	4.551E-04	1.070E-05	3.211E-02
1991	2.490E-07	7.470E-04	2.572E-05	7.716E-02
1992	2.740E-07	8.220E-04	1.386E-05	4.157E-02
1993	2.700E-07	8.100E-04	1.149E-05	3.446E-02
1994	1.660E-07	4.980E-04	6.388E-06	1.916E-02
1995	1.356E-07	4.068E-04	6.750E-06	2.025E-02
1996	1.259E-07	3.777E-04	7.442E-06	2.233E-02
1997	4.740E-08	1.422E-04	1.092E-05	3.275E-02
1998	1.550E-08	4.650E-05	3.639E-06	1.092E-02
1999	2.690E-08	8.070E-05	1.173E-05	3.520E-02
2000	1.520E-08	4.560E-05	1.173E-05	3.520E-02
2001	1.260E-08	3.780E-05	1.173E-05	3.520E-02
2002	8.360E-09	2.508E-05	3.109E-06	9.326E-03
2003	4.830E-09	1.449E-05	4.148E-06	1.245E-02
2004	4.610E-09	1.383E-05	4.788E-06	1.436E-02
2005	5.240E-09	1.572E-05	2.508E-05	7.524E-02
2006	1.440E-09	4.320E-06	1.023E-05	3.069E-02
2007	1.780E-09	5.340E-06	8.953E-06	2.686E-02
2008	5.880E-09	1.764E-05	8.395E-06	2.519E-02
2009	7.660E-09	2.298E-05	9.117E-06	2.735E-02
2010	5.310E-09	1.593E-05	8.067E-06	2.420E-02
2011	4.940E-09	1.482E-05	5.411E-06	1.623E-02
2012	3.320E-09	9.960E-06	8.166E-06	2.450E-02
2013	1.150E-09	3.450E-06	5.903E-06	1.771E-02
2014	1.980E-09	5.940E-06	6.985E-06	2.096E-02
2015	4.740E-09	1.422E-05	6.034E-06	1.810E-02
2016	1.540E-09	4.620E-06	5.411E-06	1.623E-02
2017	3.620E-09	1.086E-05	5.345E-06	1.604E-02
2018	3.380E-09	1.014E-05	4.181E-06	1.254E-02
2010	0.0001 00	1.0112 00	1.1012 00	1.2012 02

a. The data are the maximum of the median measured value for a specific year from the available stationary monitoring locations.

b. All intakes are based on a 50-hour workweek. The inhalation doses calculated may be adjusted based on the guidance in Section 4.2.3.4.

c. The annual tritium doses are all less than 0.001 rem. The tritium air concentrations and annual intakes are not listed for these years.

d. The actual concentration for this isotope was not reported. Isotopes of uranium were summed per Section 4.2.1.5 and reported as U-234.

Figure 4-3. Calculated annual median concentrations of ²³⁹⁺²⁴⁰Pu, 1986 to 2005 (Peterson et al. 2005).

concentration data were evaluated and estimates of the air concentration were generated. Tables 4-3 through 4-8 list these data and demonstrate that the chosen values are reasonable and favorable to claimants. Using the maximum airborne radionuclide concentration in a year (either from actual data or an estimated value), annual intakes that are favorable to claimants for the radionuclides of concern were derived using an assumed individual annual respiration rate of 3,000 m³/yr. The intakes in Tables 4-7 and 4-8 may be adjusted to reflect work other than 50 hr/wk. The intake rates in Tables 4-7 and 4-8 would be multiplied by the ratio of the hours worked per week to 50 hr/wk. The plutonium and uranium inhalation intakes in Tables 4-7 and 4-8 should be assigned as a lognormal distribution with a geometric standard deviation of 3.

4.3 INTAKES FROM TRITIUM IN DRINKING WATER, 1952 TO 2018

The annual environmental reports discuss concentrations of radionuclides in the drinking water at LLNL (see the References section). Several water sources were sampled and analyzed; five were identified as sources of drinking water and are designated BELL, GAS, PALM, ORCH, and TAP. The water from these locations was sampled at least once per year. In addition, LLNL sampled the onsite swimming pool (POOL). The median activity in the drinking water was estimated from calculated values and found to be below the minimum analytical detectable activity.

All assigned environmental internal doses for 1971 through 1973 are considered partial dose assessments due to the LLNL SEC for MFAPs, and 1971 through 1989 for ²³³U.

Table 4-8. Site 300 maximum annual median air concentrations and annual intakes via inhalation, 1971 to 2018.^{a,b}

	Pu-239+240 ^c	Pu-239+240 ^c	U-234 ^d	U-234 ^d
	air concentration	annual inhalation	air concentration	annual inhalation
Year	(Bq/m³)	intake (Bq/yr)	(Bq/m³)	intake (Bq/yr)
1971	2.109E-06	6.327E-03	1.530E-04	4.589E-01
1972	1.073E-06	3.219E-03	7.567E-05	2.270E-01
1973	6.290E-07	1.887E-03	3.948E-05	1.184E-01
1974	1.591E-06	4.773E-03	3.126E-05	9.377E-02
1975	1.036E-06	3.108E-03	2.139E-05	6.416E-02
1976	1.073E-06	3.219E-03	2.648E-05	7.945E-02
1977	8.880E-07	2.664E-03	3.011E-05	9.033E-02
1978	1.332E-06	3.996E-03	5.264E-05	1.579E-01
1979	5.180E-07	1.554E-03	1.596E-05	4.788E-02
1980	4.070E-07	1.221E-03	2.089E-05	6.268E-02
1981	7.030E-07	2.109E-03	1.290E-04	3.869E-01
1982	1.850E-07	5.550E-04	1.793E-05	5.380E-02
1983	3.700E-08	1.110E-04	3.093E-05	9.278E-02
1984	1.110E-07	3.330E-04	2.106E-05	6.317E-02
1985	7.400E-08	2.220E-04	9.050E-06	2.715E-02
1986	2.442E-08	7.326E-05	7.405E-06	2.221E-02
1987	2.960E-08	8.880E-05	1.218E-05	3.653E-02
1988	3.071E-08	9.213E-05	1.519E-05	4.556E-02
1989	1.110E-08	3.330E-05	8.062E-06	2.418E-02
1990	7.400E-09	2.220E-05	1.070E-05	3.209E-02
1991	2.300E-08	6.900E-05	1.288E-05	3.865E-02
1992	1.000E-08	3.000E-05	1.195E-05	3.584E-02
1993	3.900E-09	1.170E-05	1.185E-05	3.554E-02
1994	4.310E-09	1.293E-05	4.776E-06	1.433E-02
1995	4.800E-09	1.440E-05	4.446E-06	1.334E-02
1996	3.700E-09	1.110E-05	4.644E-06	1.393E-02
1997	3.600E-09	1.080E-05	5.614E-06	1.684E-02
1998	1.600E-09	4.800E-06	2.438E-06	7.314E-03
1999	1.790E-09	5.370E-06	7.487E-06	2.246E-02
2000	1.340E-09	4.020E-06	6.548E-06	1.964E-02
2001	2.070E-09	6.210E-06	5.778E-06	1.733E-02
2002	3.440E-09	1.032E-05	6.232E-06	1.870E-02
2003	1.810E-09	5.430E-06	3.788E-06	1.136E-02
2004	1.600E-09	4.800E-06	3.607E-06	1.082E-02
2005	1.500E-09	4.500E-06	2.509E-06	7.526E-03
2006	2.490E-09	7.470E-06	2.706E-06	8.117E-03
2007	1.200E-10	3.600E-07	2.804E-06	8.412E-03
2008	3.240E-09	9.720E-06	3.411E-06	1.023E-02
2009	2.670E-09	8.010E-06	2.935E-06	8.805E-03
2010	3.970E-10	1.191E-06	1.705E-06	5.116E-03
2011	9.320E-10	2.796E-06	2.263E-06	6.788E-03
2012	9.320E-10	2.796E-06	3.263E-06	9.789E-03
2013	3.980E-11	1.194E-07	2.656E-06	7.969E-03
2013	1.090E-10	3.270E-07	2.656E-06	7.969E-03
2015	1.090E-10	3.270E-07	2.132E-06	6.395E-03
2016	1.400E-10	4.200E-07	2.246E-06	6.739E-03
2017	7.480E-10	2.244E-06	2.460E-06	7.379E-03
2017	7.480E-10	2.244E-06	2.082E-06	6.247E-03
2010	7.400E-10	Z.Z44E-00	Z.UUZE-UU	0.247E-03

a. All intakes are based on a 50-hour workweek. The inhalation doses calculated may be adjusted based on the guidance in Section 4.2.3.4.

b. The annual tritium doses are all less than 0.001 rem. The tritium air concentrations and annual intakes are not listed for these years.

c. Originating from fallout (Gallegos 1998).

d. Isotopes of uranium were summed per Section 4.2.1.5 and reported as U-234.

Concentrations of tritium in the POOL location ranged from 0.8 to 200 Bg/L; the maximum concentration was reported in 1988. The LLNL swimming pool is close to the main sources of tritium at LLNL (Gallegos et al. 2002). This TBD assumed that a worker at LLNL ingested 1.4 L of water per day for 5 days per week and 50 hr/wk or 350 L/yr. The 1.4 L of water per day intake was based on the maximum tap water intake of approximately 1.75 L/d (ICRP 2002). Assuming that the daily tap water intake of 1.75 L/d occurs over a 16-hour period, adjusting for 10 hours per workday yields approximately 1.1 L/d. This daily water intake is rounded up to 1.4 L/d, as being favorable to claimants. The intake of 1.4 L/d is 80 percent of the maximum tap water intake of 1.75 L/d. It is reasonable to assume that most of the daily water intake occurs during work hours because of higher physical activity. The daily water intake assumed is also consistent with the daily urine excretion rate of 1.4 L/d. The values before 1966 were selected from the maximum concentration that was reported for the period from 1966 to 2018. No data were available for drinking water at Site 300; this TBD assumed that the concentration of tritium was the same as that at LLNL. The largest tritium intake was 36.626 Bg/yr for 1975 (ORAUT 2020a). This results in an annual tritium dose of less than 0.001 rem. Therefore, the 1952 through 2018 annual tritium doses from drinking water are all less than 0.001 rem. Radionuclides contributing doses less than 0.001 rem are not considered significant. The tritium drinking water concentrations and annual intakes for 1952 through 2018 are not listed for these vears.

It should also be noted that the annual tritium dose from both air concentration and drinking water data combined are less than 0.001 rem. This is based on combining the largest tritium inhalation intake of 10,090 Bq/yr for 1957 (ORAUT 2020b), and the largest drinking water intake of 36.626 Bq/yr for 1975 (ORAUT 2020a).

4.4 AMBIENT RADIATION

4.4.1 Gamma Radiation

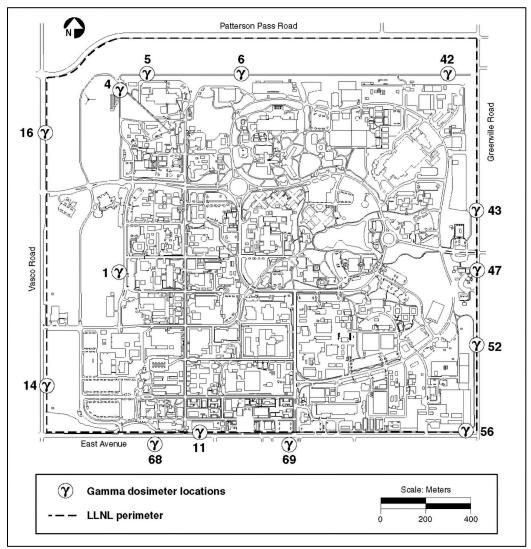
From 1967 to the present, thermoluminescent dosimeters (TLDs) have been used on the main site to determine ambient external radiation levels, including natural background (terrestrial and cosmic) radiation. TLDs were deployed at Site 300 in July 1988. Figures 4-4 and 4-5 show the locations of the monitoring stations at the main site and Site 300, respectively. The data for this analysis were summarized from the annual environmental reports for 1989 to 2018 (see the References list).

Observations from earlier environmental reports, which were based on surveys with portable instruments, state, "0.02 mrem/h at all locations." The instrumentation might be considered primitive by today's standards, and the site did not employ continuous monitoring, so these results have been overshadowed by recent and more reliable observations. The external gamma dose rate of 0.02 mrem/hr corresponds to 175 mrem/yr (0.02 mrem/hr \times 8,760 hr/yr). This dose value is bounded by the assignment of the main site highest average and maximum gamma doses based on the reported data in Table 4-9 for 1972 and 1975, respectively, for 1952 through 1967 when no doses were reported.

Over the years, various combinations of TLD chip configurations and monitoring locations were used. From 1988 to the present, minimal changes in external radiation levels were observed at the perimeter. The analysis for this TBD developed the environmental radiological profile for LLNL for use by dose reconstructors when personal dosimetry or bioassay program participation was not required.

Site annual environmental reports, health physics surveys, and other reports were reviewed for data that would be useful in reconstructing ambient radiation levels.

The ambient radiation that was measured by TLDs near the security fence included natural background radiation, nuclear weapons testing fallout, and cosmic radiation. The TLDs provided an indication of worker exposure levels in the general proximity of the security fence but not inside



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Figure 4-4. Main site gamma dosimeter locations, 2001 (Gallegos et al. 2002).

buildings. LLNL compared these data annually with TLD data from offsite locations and values for State of California and regional exposure levels. The determination has always been that onsite ambient radiological conditions as measured at the security fence were not significantly different from offsite, state, and regional annual exposure levels. This was attributed to the geology of the region around LLNL.

The locations of the monitoring points from which data were summarized in this section add uncertainty to the results. The monitoring points, as stated above, have been located around the LLNL perimeter and off the site to monitor public exposures.

The estimated accuracy of the area monitors for external radiation is ±20%. However, subtracting background from these measurements adds more uncertainty because of the variability and selection of background information. The environmental reports gave the results as cumulative exposures in millirem per year based on 8,760 hours of exposure, which is the total number of hours in 365 days. External dose for some years was not available. For the main site, the maximum external dose was reported in 1975; the maximum site average was reported in 1972; therefore, the 1972 and 1975 dose values were used respectively for the years noted. For Site 300, the maximum value was noted in 1994; this value was used for the average for years before 1989 when monitoring began. These maximum values of environmental exposure were recommended for years when data were

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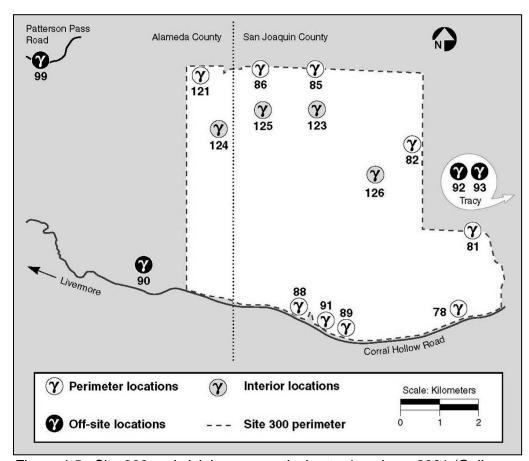


Figure 4-5. Site 300 and vicinity gamma dosimeter locations, 2001 (Gallegos et al. 2002).

unavailable for the main site and Site 300. Tables 4-9 and 4-10 provide, for the main site and Site 300, respectively, average and maximum annual exposures for the 8,760-hr/yr exposure period. The yearly exposure values provided in Tables 4-9 and 4-10 should be adjusted in accordance with ORAUT-OTIB-0088, External Dose Reconstruction (ORAUT 2018). In cases where annual work hours and areas are uncertain or otherwise not specified, environmental doses are based on a bestestimate approach that assumes 2,500 hours at the site average dose values. Partial years of employment, or work other than 50 hr/wk may be adjusted in accordance with guidance in ORAUT-OTIB-0088, External Dose Reconstruction (ORAUT 2018). Best estimate onsite ambient photon doses should be assigned as a normal distribution with an error of 20%.

4.4.2 **Neutron Radiation**

Data that describe ambient neutron measurements on the main site were found in site environmental radiation reports beginning in 1972 (Gudiksen et al. 1973). From 1972 through 1987, elevated ambient neutron dose was measured at the perimeter fence near Building 212. The source of these elevated readings was the Insulating Core Transformer (ICT) accelerator, which began operations in 1966 in Building 212 and produced 14-MeV neutrons (Olson 1974). The ICT accelerator ceased operations in 1987 (LLNL 1990). The ICT accelerator was also referred to as the Rotating Target Neutron Source I. This distinguished it from the Rotating Target Neutron Source II, which began operations in 1978 in Building 292. Figure 4-6 shows the locations of the environmental neutron dosimeters. Stations 5 and 3 were typically the maximum value locations.

Table 4-11 lists the maximum and available average ambient neutron values for the main site. Only maximum values were available from the reports for 1972 to 1980. Average and maximum values

Table 4-9. Main site external gamma radiation (mrem/yr) [5].a,b

Table 4-9.	. Main site exterr	nal gamma radiat	
	Average,	Maximum,	
Year	8,760 hr/yr	8,760 hr/yr	
1952	89 ^c	317 ^d	
1953	89 ^c	317 ^d	
1954	89°	317 ^d	
1955	89°	317 ^d	
1956	89°	317 ^d	
1957	89°	317 ^d	
1958	89°	317 ^d	
1959	50	317 ^d	
1960	89°	317 ^d	
1961	88	317 ^d	
1962	44	317 ^d	
1963	50	317 ^d	
1964	50	317 ^d	
1965	50	317 ^d	
1966	89°	317 ^d	
1967	20	317 ^d	
1968	20	163	
1969	34	83	
1970	15	105	
1971			
1972	89	122	
1973	80	96	
1974	74	137	
1975	88	317	
1976	84	293	
1977	79	227	
1978	64	93	
1979	61	96	
1980	63	81	
1981	54	61	
1982	49	57	
1983	51	73	
1984	48	53	
1985	59	80	

Year	Average, 8,760 hr/yr	Maximum, 8,760 hr/yr
1986	61	70
1987	65	72
1988	63	70
1989	63	75
1990	65	72
1991	65	78
1992	66	76
1993	65	74
1994	72	80
1995	56	61
1996	55	60
1997	60	64
1998	60	66
1999	58	63
2000	57	65
2001	56	100
2002	65	65
2003	56	62
2004	57	64
2005	58	65
2006	58	64
2007	58	68
2008	57	64
2009	59	64
2010	55	60
2011	57	63
2012	56	63
2013	58	64
2014	57	63
2015	56	63
2016	57	63
2017	57	61
2018	58	63

- a. The results in this table reflect LLNL data for an exposure period of 8,760 hr/yr. In cases where annual work hours and areas are uncertain or otherwise not specified, environmental doses are based on a best-estimate approach that assumes 2,500 hours at the site average dose values in accordance with ORAUT (2018).
- b. The 2018 main site onsite ambient gamma dose can be extended to later years.
- c. External dose for these years was not available. The maximum average external dose was reported in 1972. Therefore, 1972 dose values are used for the noted years.
- d. External dose for these years was not available. The maximum external dose was reported in 1975. Therefore, the 1975 dose values are used for the years noted.

were available for 1981 to 1993. Again, the values from the original data were for an 8,760-hr/yr exposure period. The yearly exposure values provided in Table 4-11 should be adjusted in accordance with ORAUT-OTIB-0088, *External Dose Reconstruction* (ORAUT 2018). In cases where annual work hours and areas are uncertain or otherwise not specified, environmental doses are based on a best-estimate approach that assumes 2,500 hours at the site average dose values. Partial years of employment, or work other than 50 hr/wk may be adjusted in accordance with guidance in ORAUT-OTIB-0088, *External Dose Reconstruction* (ORAUT 2018). Best estimate onsite ambient neutron doses should be assigned as a normal distribution with an error of 20%.

Potential ambient neutron dose is captured by application of the neutron-to-photon ratio to gamma dose (applied through 1968) or by using measured and missed neutron dose data for the period from

Table 4-10. Site 300 external gamma radiation (mrem/yr) [6].a,b,c

Table 4-10. Site 300 external gamma rad		nal gamma radiati
	Average,	Maximum,
Year	8,760 hr/yr	8,760 hr/yr
1955	88 ^d	99 ^d
1956	88 ^d	99 ^d
1957	88 ^d	99 ^d
1958	88 ^d	99 ^d
1959	88 ^d	99 ^d
1960	88 ^d	99 ^d
1961	88 ^d	99 ^d
1962	88 ^d	99 ^d
1963	88 ^d	99 ^d
1964	88 ^d	99 ^d
1965	88 ^d	99 ^d
1966	88 ^d	99 ^d
1967	88 ^d	99 ^d
1968	88 ^d	99 ^d
1969	88 ^d	99 ^d
1970	88 ^d	99 ^d
1971	88 ^d	99 ^d
1972	88 ^d	99 ^d
1973	88 ^d	99 ^d
1974	88 ^d	99 ^d
1975	88 ^d	99 ^d
1976	88 ^d	99 ^d
1977	88 ^d	99 ^d
1978	88 ^d	99 ^d
1979	88 ^d	99 ^d
1980	88 ^d	99 ^d
1981	88 ^d	99 ^d
1982	88 ^d	99 ^d
1983	88 ^d	99 ^d
1984	88 ^d	99 ^d
1985	88 ^d	99 ^d
1986	88 ^d	99 ^d

	Average,	Maximum,
Year	8,760 hr/yr	8,760 hr/yr
1987	88 ^d	99 ^d
1988 ^e	88 ^d	99 ^d
1989	77	89
1990	78	94
1991	78	93
1992	77	87
1993	76	86
1994	88	99
1995	64	72
1996	66	72
1997	72	77
1998	72	78
1999	71	80
2000	64	70
2001	64	66
2002	67	76
2003	67	74
2004	69	77
2005	69	83
2006	68	77
2007	69	74
2008	69	75
2009	69	75
2010	66	72
2011	67	73
2012	63	69
2013	69	76
2014	69	75
2015	67	76
2016	66	75
2017	67	77
2018	69	73

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- a. The results in this table reflect LLNL data for an exposure period of 8,760 hr/yr. In cases where annual work hours and areas are uncertain or otherwise not specified, environmental doses are based on a best-estimate approach that assumes 2,500 hours at the site average dose values in accordance with ORAUT (2018).
- b. Site average (millirem per year) is based on the maximum average value from later years where monitoring was performed. Therefore, no maximum or uncertainty is provided.
- c. The 2018 Site 300 onsite ambient gamma dose can be extended to later years.
- d. External dose for these years was not available. The maximum average and maximum external doses were reported in 1994. Therefore, the 1994 dose values are used for the years noted.
- e. Environmental radiation monitoring at Site 300 began on July 1, 1988.

1969 to the present. Considering that all LLNL workers were using neutron sensitive TLDs beginning at least in 1969, any significant neutron dose should have been detected and measured by these dosimeters (Unruh ca. 1969). Therefore, it is not necessary to add additional ambient neutron dose for monitored workers.

If dosimetry data are not available for a worker from 1969 to the present, then the maximum ambient neutron dose value may be adjusted in accordance with ORAUT-OTIB-0088, *External Dose Reconstruction* (ORAUT 2018). To be favorable to claimants, a neutron energy range of 0.1 to 2.0 MeV should be chosen along with an International Commission on Radiological Protection Publication 60 weighting factor correction of 1.91 (ICRP 1991). The value for 1972 should be used for

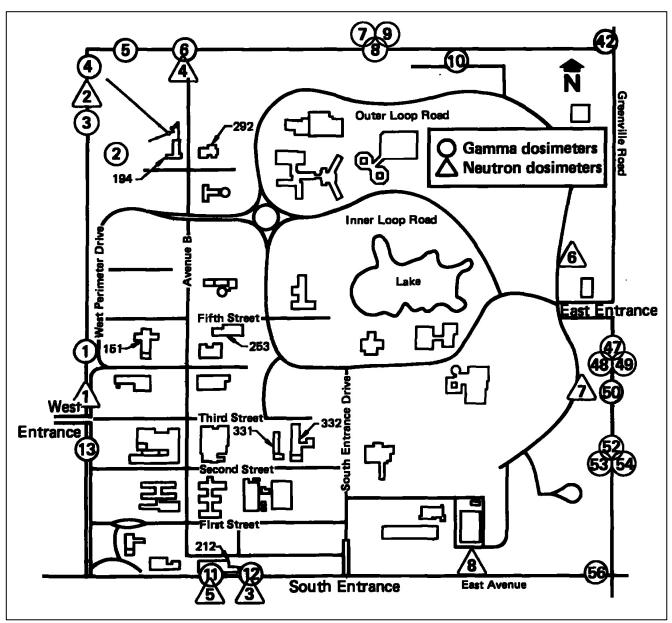


Figure 4-6. Main site neutron dosimeter locations, 1969 to present (Griggs and Buddemeier 1986).

1969 to 1971. If the worker was externally monitored from 1966 to 1969, the neutron-to-photon ratio can be assigned. If photon dosimetry data are not available for this period, then the neutron value for 1972 can be assigned for 1966 to 1969.

The neutron rem meters used by LLNL had aged and deteriorated by 1994 to the point where the neutron measurements did not meet the LLNL accuracy and precision requirements. Therefore, the neutron data collected during 1994 were not reported, and neutron monitoring was discontinued as of January 1995 (Harrach 1994). Table 4-11 indicates a significant drop in the onsite ambient neutron doses measured in later years after the ICT accelerator ceased operations in 1987. The measured onsite ambient neutron doses were a fraction of the measured onsite gamma doses after this time. The 1993 onsite ambient neutron dose can be extended to later years.

Table 4-11. Main site external neutron radiation (mrem/yr) [7] a,b

Year	Average, 8,760 hr/yr	Maximum, 8,760 hr/yr
1966	57	250
1967	57	250
1968	57	250
1969	57	250
1970	57	250
1971	57	250
1972	57	250
1973	57	250
1974	84	370
1975	160	700
1976	137	600
1977	126	550
1978	31	137
1979	18	80
1980	19	85
1981	8	33
1982	8	36
1983	26	113
1984	10	45
1985	2	7
1986	7	30
1987	2	9
1988	1	5
1989	1	6
1990	1	6
1991	2	7
1992	2	7
1993	2	7

- a. The results in this table reflect data provided by LLNL for an exposure period of 8,760 hr/yr. In cases where annual work hours and areas are uncertain or otherwise not specified, environmental doses are based on a best-estimate approach that assumes 2,500 hours at the site average dose values in accordance with ORAUT (2018)
- b. Neutron results were not reported in 1994 due to neutron detection equipment degradation not meeting accuracy and precision requirements. Neutron monitoring was discontinued as of January 1995 (Harrach 1994). The 1993 main site onsite ambient neutron dose can be extended to later years.

4.5 ATTRIBUTIONS AND ANNOTATIONS

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

[1] Szalinski, Paul A. Oak Ridge Associated Universities (ORAU) Team. Health Physicist. April 2007.

The surrogate radionuclides were derived from LLNL NESHAPs reports (Gallegos et al. 1998, 2000). The following is a quote from the reports for the Livermore site: "In addition, isotopic

analyses of mixtures of radionuclides are not always available, and radionuclide inventories are stated as 'gross alpha,' 'gross beta,' 'gross gamma,' or 'mixed fission products'." In these cases, ²³⁹Pu was used as the surrogate for gross alpha, ¹³⁷Cs was used as the surrogate for gross gamma, and ⁹⁰Sr was used as the surrogate for gross beta and mixed fission products to provide conservative dose estimates."

- [2] Szalinski, Paul A. ORAU Team. Health Physicist. April 2007.
 The surrogate radionuclides for gross alpha at Site 300 were derived from LLNL NESHAPs reports (Gallegos et al. 1998, 2000). For Site 300 the following applies for gross alpha analyses: "During Site 300 explosives experiments, the device containing depleted uranium is placed on an open air firing table and detonated."
- [3] Szalinski, Paul A. ORAU Team. Health Physicist. April 2007. The surrogate radionuclide for gross beta at Site 300 is ²³⁴Th. Only tritium and depleted uranium were reported as released into the environment at Site 300 (Gallegos et al. 1998, 2000). Thorium-234 is a progeny of the ²³⁸U.
- [4] Szalinski, Paul A. ORAU Team. Health Physicist. March 2007.
 A geometric standard deviation of 3 is used in ORAUT-OTIB-0018, *Internal Dose Overestimates for Facilities with Air Sampling Programs* (ORAUT 2005).
- [5] Szalinski, Paul A. ORAU Team. Health Physicist. April 2007.

 The external gamma radiation data are directly from site annual environmental reports, except where indicated by footnote. The site maximum and average values from the reports are listed in the table.
- [6] Szalinski, Paul A. ORAU Team. Health Physicist. April 2007.

 The external gamma radiation data are directly from site annual environmental reports, except where indicated by footnote. The site maximum and average values from the reports are listed in the table.
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 The external neutron radiation data are directly from site annual environmental reports. The site maximum and average values from the reports are listed in the table.

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GLOSSARY

alpha radiation

Positively charged particle emitted from the nuclei of some radioactive elements. An alpha particle consists of two neutrons and two protons (a helium nucleus) and has an electrostatic charge of +2.

background radiation

Radiation from cosmic sources, naturally occurring radioactive materials including naturally occurring radon, and global fallout from the testing of nuclear explosives. Background radiation does not include radiation from source, byproduct, or Special Nuclear Materials regulated by the U.S. Nuclear Regulatory Commission. The average individual exposure from background radiation is about 360 millirem per year.

becquerel (Bq)

International System unit of radioactivity equal to 1 disintegration per second; 1 curie equals 37 billion (3.7×10^{10}) Bq.

beta radiation

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

deep dose equivalent

Dose equivalent in units of rem or sievert for a 1-centimeter depth in tissue (1,000 milligrams per square centimeter).

dosimetry

Measurement and calculation of internal and external radiation doses.

radioactivity

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

rem

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

thermoluminescent dosimeter

Device for measuring radiation dose that consists of a holder containing solid chips of material that, when heated, release the stored energy as light. The measurement of this light provides a measurement of absorbed dose. Thermoluminescent dosimeters replaced film dosimeters at essentially all U.S. Department of Energy sites beginning in the 1960s.