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ACRONYMS AND ABBREVIATIONS

Bq	becquerel
CEDE	committed effective dose equivalent
CFR	Code of Federal Regulations
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
ft	foot
Ge(Li)	lithium-drifted germanium
GSD	geometric standard deviation
hr	hour
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
MFP	mixed fission 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 (1×10^{15} Bq)
POC	probability of causation
SRDB Ref ID	Site Research Database Reference Identification (number)
TBD	technical basis document
TBq	terabecquerel (1×10^{12} Bq)
TLD	thermoluminescent dosimeter

U.S.C.	United States Code
wk	week
yr	year
§	section or sections

4.1 INTRODUCTION

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

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

For employees of DOE or its contractors with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision [42 U.S.C. § 7384n(b)] says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer … was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the POC [probability of causation[1]] guidelines established under subsection (c) …" [42 U.S.C. § 7384n(b)]. Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation, 42 C.F.R. Pt. 82) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work (NIOSH 2007).

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

- Background radiation, including radiation from naturally occurring radon present in conventional structures

- Radiation from X-rays received in the diagnosis of injuries or illnesses or for therapeutic reasons

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

4.1.1 Purpose

Lawrence Livermore National Laboratory (LLNL) in Livermore, California, 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 products, and accelerator-produced isotopes. This TBD discusses two sites: the main site, designated as 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, designated as Site 100, provided LLNL with beta/photon film dosimeters and processing, and PICs (Thompson 1953). The practice of providing film and pocket ionization dosimeters to all workers has been in effect since March 1953 (Thompson 1953) and in 1958 film badges became part of the security badge (Nolan 1958), which effectively mandated that all workers wear their film dosimeters at all times. There is ongoing collaboration between these two laboratories. To provide the basis for estimating the environmental dose for years when monitoring did not occur, this TBD provides annual intakes and ambient external doses from 1952 (1955 for Site 300) to 2005 (the last year with publicly available data).

4.1.2 Scope

This TBD documents historical practices at LLNL and provides information for the evaluation of environmental radiation data. It can serve as a supplement to, or substitute for, individual monitoring data.

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 resuspension of radioactive materials (Section 4.2) and for 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 Petition Information for LLNL

Classes Added to the SEC

NIOSH has determined, with concurrence from the Secretary of Health and Human Services (HHS), that MFP doses at LLNL cannot be reconstructed with sufficient accuracy between 1950 and 1973, inclusive. For this reason, the following class of LLNL employees has been added to the Special Exposure Cohort (SEC) (Leavitt 2008).

> *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.*

Class Recommended by NIOSH for addition to the SEC

NIOSH has subsequently determined that MFP doses for workers not monitored for radiation exposure at LLNL also cannot be reconstructed with sufficient accuracy between 1950 and 1973, inclusive. For this reason, NIOSH has recommended that the following class of LLNL employees be added to the SEC (NIOSH 2010).

> 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.

The NIOSH-recommended SEC class includes all workers during the SEC period, regardless of whether an individual worker was monitored for radiation exposure. Because of the identified dose reconstruction infeasibility, all dose reconstructions for all workers having employment during the SEC period are considered partial dose reconstructions. If monitoring data are available for workers included in the SEC, dose is to be assigned as appropriate based on such data; however, such dose reconstructions are still considered partial dose reconstructions because the HHS has determined that exposure to mixed fission and activation products during the SEC period cannot be bounded.

4.2 INTERNAL DOSE FROM ONSITE ATMOSPHERIC RADIONUCLIDE CONCENTRATIONS

4.2.1 Ambient Air Sample Collection Network

4.2.1.1 Before 1959

A review of records indicates that the earliest available environmental moniotoring data for the LLNL main site was 1959 (Beaufait 1961), and for Site 300 was 1961 (LRL 1962). Before the 1980s LLNL's almost exclusive mission was weapons research. Therefore, it was assumed that the radionuclide emission rate would be proportional to the weapons-related activity at the site from design and

fabrication to 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 coworker data might be more appropriate than environmental data.

4.2.1.2 1959 through 1971

Air samples were collected in 7 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	E
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.

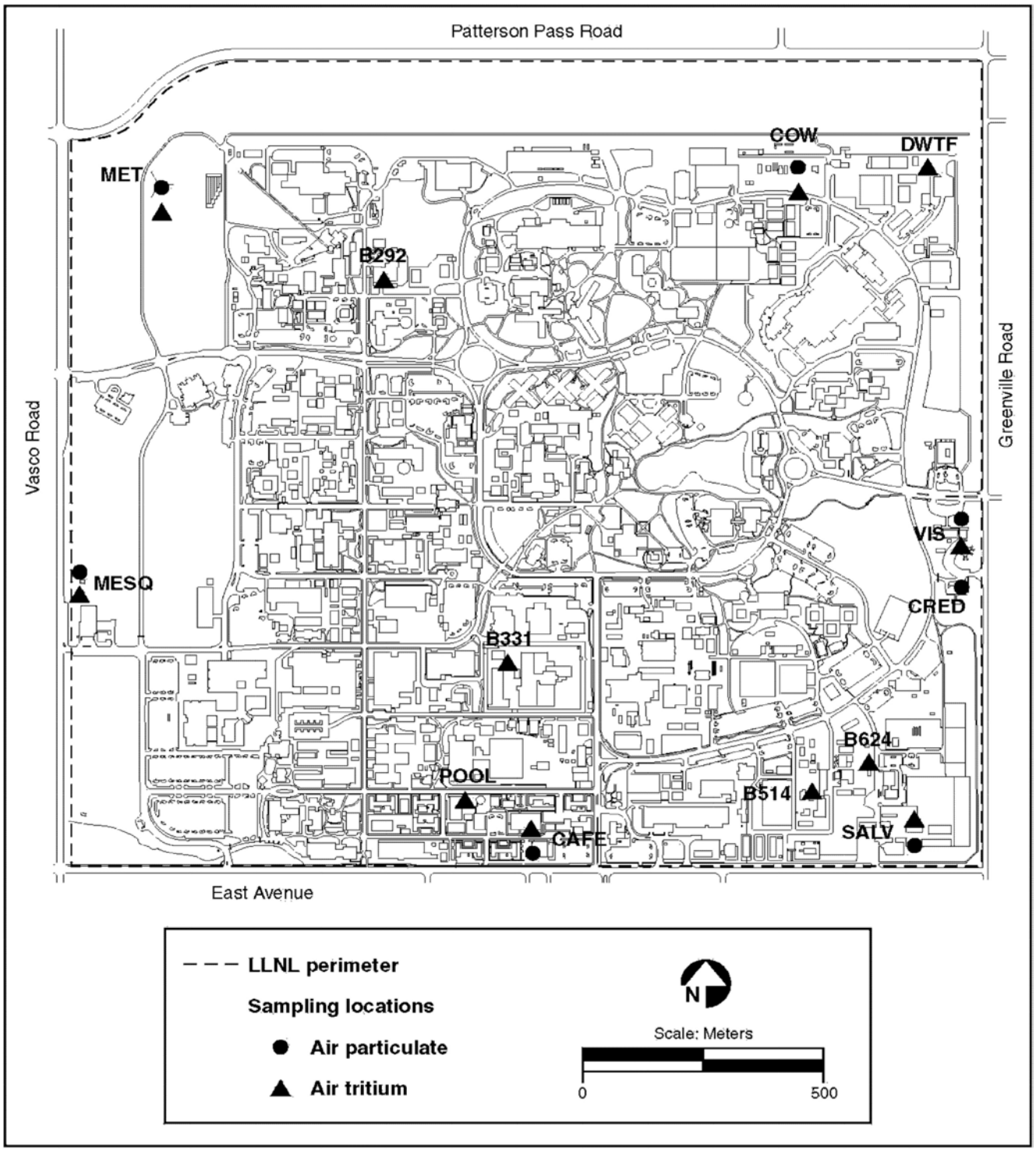

Figure 4-1. Main site air particulate and tritium sampling locations (Gallegos et al. 2002).

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

Designation	Location	Direction from center of Site 300
COHO	Near south perimeter of facility; representative of MEI	S
EOBS	Northeast of Bunker 801E	NE
ECP	Center of Site 300	E
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

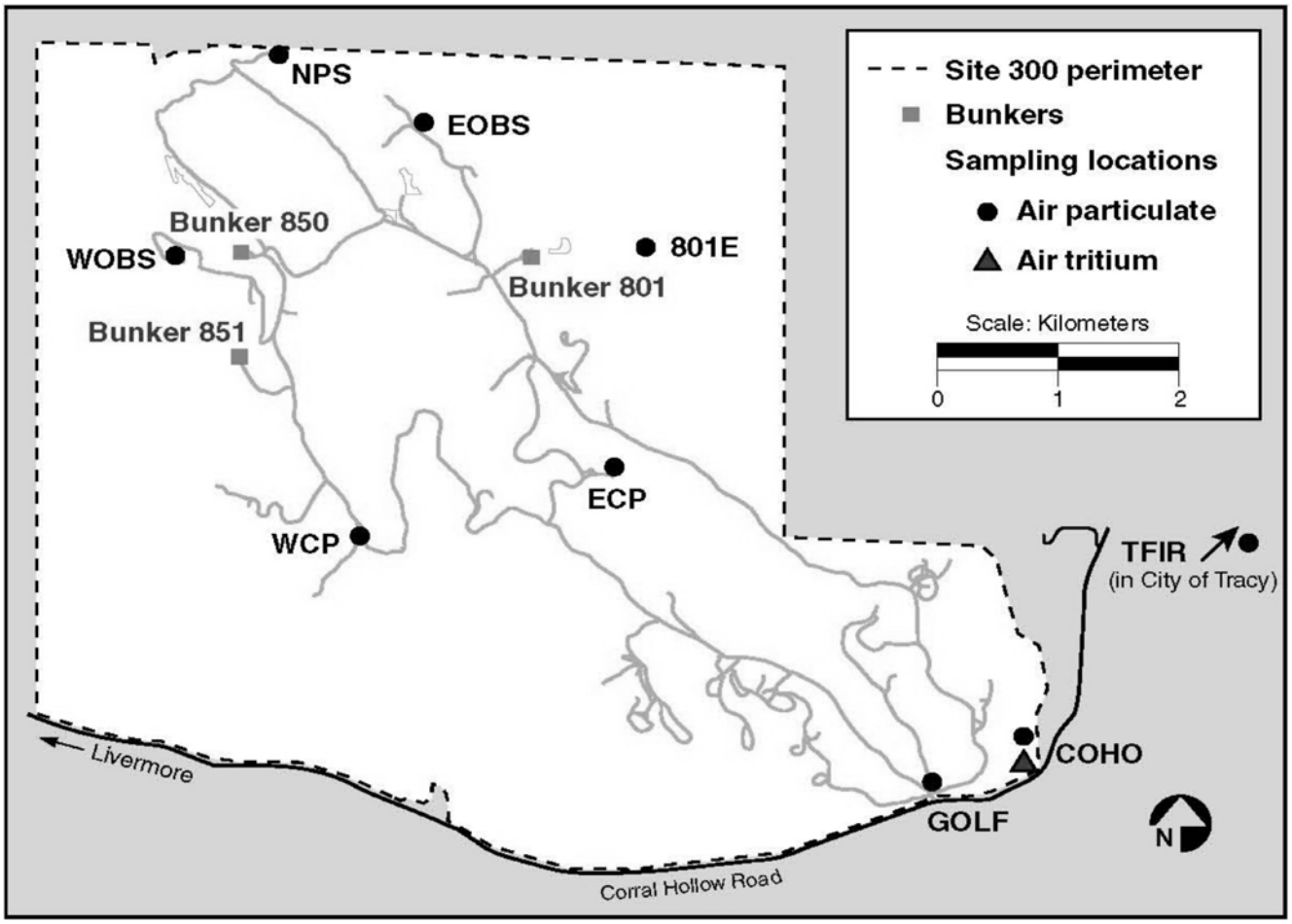

Figure 4-2. Site 300 and downtown Tracy air particulate and tritium sampling locations (Gallegos et al. 2002).

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 beta radiations, such as ^{3}H, ^{238}Pu and $^{239+240}Pu$, and ^{235}U and ^{238}U. 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 offsite locations had higher activity than onsite locations due to the fallout. The highest activity is presented, despite location, 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 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 unless a higher value was reported for an offsite monitoring location, in which case that data is reported to be favorable to claimants. Table 4-1 lists these locations.

Air samplers were positioned to provide reasonable probability that any significant concentration of radioactive effluents from LLNL operations would be detected. 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 that represented more than 90% of the LLNL radioactive materials inventory. It analyzed air samples specifically for the presence of tritium, ^{238}Pu, and $^{239+240}$Pu, 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 ^{239}Pu, and a second using ^{234}U. The gross beta concentration should be assumed to result from ^{90}Sr [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 indicated the analysis of ^{238}U and ^{235}U. For estimates based on this TBD, the dose reconstructor should assign the dose that is associated with ^{234}U for depleted uranium. Through

1971, dose reconstruction should use the gross alpha concentration for Site 300 to calculate the presence of ^{234}U; it is assumed that the maximum radiation dose is assigned as 100% ^{234}U versus a varying percentage of ^{234}U and ^{235}U [2]. Data are listed in the tables in Section 4.2.3.2. Assume the gross beta concentration is ^{234}Th [3].

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 ^{230}Th and ^{90}Sr as calibration sources to determine alpha and beta counting efficiencies, respectively. Annual counting efficiency measurements were made for each detector. Periodic cross-checks 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 ft^3/min of air (LRL 1964). As discussed in Section 4.2.1.1, no monitoring data was found for the years before 1959 for the main site, and 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 ^{239}Pu, ^{238}Pu, ^{137}Cs, ^{235}U, and ^{238}U. 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 m^2 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 with 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 ^{235}U, ^{238}U, ^{40}K, ^{226}Ra, ^{228}Ra, and ^{228}Th (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 $^{239+240}Pu$. Similarly, portions of the glass-fiber filters from Site 300 were analyzed for the presence of ^{235}U and ^{238}U. The filters were placed in a muffled 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 Estimation of Potential Annual Intakes from Airborne Radionuclides

This section discusses estimation of annual intakes from the air monitoring results from 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 2005.[2] 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.

[2] The References section lists all directly cited references, and then separately lists all environmental reports in date order.

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.

The method for calculation of the annual intakes from onsite atmospheric radionuclide concentrations was applied directly to available air sampling data. Air sampling measurement error and uncertainty were evaluated using the maximum reported value for the year at any location. Available data were limited to annual averages for each location; statistical analysis was not possible due to the limited amount of data.

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.

Section 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, plutonium, and uranium for 1971 to 2005.

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 1961 for Site 300 are listed in Tables 4-3 and 4.4. The analysis assumed a breathing rate of 2,400 m^3/yr based on 20 L/min for 2,000 hr/yr, and a geometric standard deviation (GSD) for these data of 3 [4]. This data is based on using the highest of the measured data for years during which environmental monitoring occurred. All environmental internal doses assigned prior to 1959 are considered partial dose assessments due to the LLNL SEC for MFP.

4.2.3.2 Gross Alpha and Gross Beta from 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 list air concentrations and estimated intakes for the LLNL site, and Table 4-6 lists the values for Site 300.

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

	Gross alpha (Pu-239 or U-234)		Gross beta (Sr-90)	
Year	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)
1952	1.6E-3	3.8	6.7E-1	1,610
1953	1.6E-3	3.8	6.7E-1	1,610
1954	1.6E-3	3.8	6.7E-1	1,610
1955	1.6E-3	3.8	6.7E-1	1,610
1956	1.6E-3	3.8	6.7E-1	1,610
1957	1.6E-3	3.8	6.7E-1	1,610
1958	1.6E-3	3.8	6.7E-1	1,610

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

	Gross alpha (U-234)		Gross beta (Th-234)	
Year	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)
1955	6.7E-3	16	2.4E+0	5760
1956	6.7E-3	16	2.4E+0	5760
1957	6.7E-3	16	2.4E+0	5760
1958	6.7E-3	16	2.4E+0	5760
1959	6.7E-3	16	2.4E+0	5760
1960	6.7E-3	16	2.4E+0	5760

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

	Gross alpha (Pu-239 or U-234)		Gross beta (Sr-90)	
Year	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)
1959	7.4E-4	1.8	2.7E-1	648
1960	8.5E-4	2.0	6.7E-1	1,610
1961	1.6E-3	3.8	4.1E-1	984
1962	1.7E-4	0.4	9.0E-3	22
1963	1.2E-4	0.3	1.3E-1	312
1964	1.2E-4	0.3	2.4E-2	58
1965	3.7E-5	0.1	6.5E-3	16
1966	3.7E-5	0.1	2.6E-3	6
1967	2.9E-5	0.07	4.7E-3	11
1968	3.7E-5	0.1	4.4E-3	10
1969	3.7E-5	0.1	3.8E-3	9
1970	3.7E-5	0.1	5.6E-3	14

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

Year	Gross alpha (U-234)		Gross beta (Th-234)	
	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)
1961	6.7E-3	16	2.6E-3	6.2
1962	1.9E-2	44	1.4E-1	336
1963	3.4E-3	8	2.4E+0	5760
1964	1.3E-3	3	1.9E-1	456
1965	1.3E-4	0.3	2.4E-2	58
1966	2.0E-4	0.5	2.9E-2	70
1967	1.9E-4	0.5	1.6E-1	384
1968	3.7E-5	0.09	2.5E-2	60
1969	7.4E-5	0.2	3.9E-2	94
1970	4.1E-4	1	3.4E-2	82

The isotopes of significance at Site 300 are ^{238}U and ^{235}U, both of which decay by alpha radiation. The presence of beta radiation indicates the progeny of uranium.

All environmental internal doses assigned from 1952 to 1970 are considered partial dose assessments due to the LLNL SEC for MFP.

4.2.3.3 Tritium from 1952 to 1972

Environmental air monitoring samples were not analyzed for tritium before 1972. Most of the released tritium was airborne with the dominant source being the Tritium Facility (Building 331, previously Building 172), which began operation in 1952. The total activity of tritium released was reported to be approximately 28 PBq (760 kCi) over the more than 40 years of LLNL history (Tate et al. 1999). Most of this activity was released in two events: 13 PBq (350 kCi) on January 21, 1965, beginning at 3:30 p.m., and 11 PBq (300 kCi) on August 6, 1970, beginning at 6:14 a.m. Both releases were in the form of hydrogen gas and were released from a 30-m-tall stack over a period of less than 1 hr (DOE 1982). Dispersion modeling estimated air concentrations of tritium to range from 3.2×10^2 to 5.2×10^8 Bq/m^3 over the period of the release within 0.5 mi of the stack. Potential intakes of tritium were estimated for the release to range from 300 to 6×10^8 Bq with a corresponding estimated radiation dose of less than 8 mrem; the 95th-percentile dose was calculated to be less than 19 mrem.

Assuming the remaining 4 PBq was released over the 40-year history as routine discharges (exclusive of the releases in 1965 and 1970), an estimate of potential exposure can be derived for the period for which no environmental data are available. To be favorable to the claimant, it is assumed that the 4 PBq of tritium was released before 1972 and distributed equally for each year. That is, it is assumed that 200 TBq were discharged each year [5]. A study that reviewed the release of tritium reported a range of 40 to 280 TBq/yr (1 to 7.5 kCi) before 1992 and between 3 and 12 TBq (80 and 320 Ci) since 1992 (Tate et al. 1999).

Using the dispersion calculations from 1970, the estimated annual release of 200 TBq/yr, and the same weather conditions that existed during the 1970 release, the estimated concentration of tritium ranges from 5.8 to 2×10^6 Bq/m^3 for 1952 to 1971. It is unreasonable to assume that a worker stayed in the exact locations of maximum exposure (immediately downwind of the Building 331 stack and at the points of maximum impact) for the entire exposure period of 20 years. Therefore, it is not reasonable to assume that the maximum concentration represents the likely potential exposure. The

estimated concentration of 1.7×10^4 Bq/m^3 was derived for the maximum site boundary location and is more likely to represent the average potential exposure to a single worker at LLNL for the exposure period. The value of 1.7×10^4 Bq/m^3 was calculated using the concentration at the site boundary 0.5 mi northeast (about 40°) from the Building 331 release point for the 1970 release (ATSDR 2001). The concentration at this point was divided by the total 1970 11 PBq (300 kCi) release; the resultant value was then multiplied by the estimated annual release of 200 TBq/yr. The 40° direction was chosen because it closely reflects (within 5°) to the predominant southwest wind direction for LLNL (Althouse et al. 2001).

For purposes of an estimated radiation dose that is favorable to claimants, an intake of 3×10^7 Bq (1.7×10^4 Bq/m^3) of tritium as HTO should be used for each year from 1952 to 1971. This value is considered favorable to claimants because only approximately one-half of the total activity of tritium releases occurred during this period (excluding the 1965 and 1970 events). In addition, the airborne concentration of 3.14 in becquerel per cubic meter per released curie in a year was compared with the onsite measurements (rather than the perimeter measurements) that were made in the immediate vicinity of the diffuse sources of tritium beginning in 1991, such as Building 331 (Gallegos et al. 1992). The value of 1.7×10^4 Bq/m^3 represents 3.14 Bq/m^3 per released curie during the year. The highest single measured value adjacent to an onsite source for 1991 through 2000 was 0.46 Bq/m^3 per released curie, which occurred in 1995; this value is approximately 20 times higher than the maximum median onsite measurement for that year (Harrach et al. 1996a,b). The highest maximum tritium median value was 0.15 Bq/m^3 per released curie, which occurred in 1998. Further favorability to claimants arises because all the tritium is considered to be HTO even though a large fraction of the emissions were in the form of hydrogen (tritium) gas, and HTO has a higher dose conversion factor than tritium gas [6].

4.2.3.4 Tritium, Uranium, and Plutonium after 1970

Estimates of offsite doses to members of the public from the 1971 to 2005 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. Table 4-7 lists main site concentrations for ^{3}H, $^{239+240}$Pu, and ^{234}U (^{3}H not reported for 1971). This TBD assumed that a worker inhaled 20 L of air per minute for 2,000 hr/yr or approximately 2.4×10^6 L of air per year (2,400 m^3/yr). The concentration of tritium ranged from 0.04 to 3 Bq/m^3; the potential for exposure to tritium ranged from 144 to 11,200 Bq/yr. For tritium, the total uptake by the body was increased by 50% to account for absorption by the skin in addition to the inhalation pathway (Gede and Gildea 1980). 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.

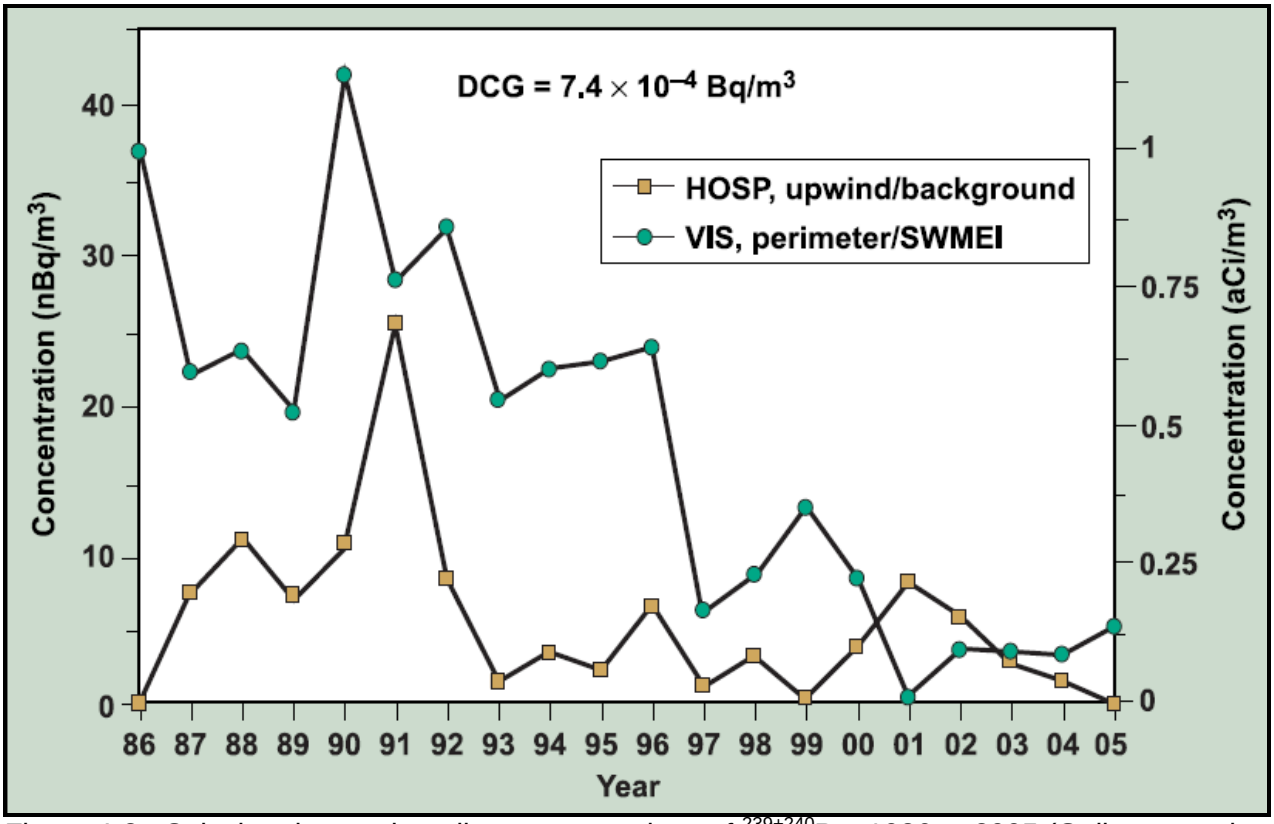

Figure 4-3. Calculated annual median concentrations of $^{239+240}$Pu, 1986 to 2005 (Gallegos et al. 2002).

Table 4-7. Main site maximum annual median air concentrations and intakes via inhalation, 1971 to 2005.[a]

Year	Tritium		Pu-239+240		U-234[b]	
	Air concentration (Bq/m^3)	Annual intake (Bq/yr)[c]	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)
1971	(d. e)	(d, e)	4.1E-6	9.8E-3	2.2E-6	5.3E-3
1972	1.4	5.0E+3	2.2E-6	5.3E-3	3.3E-6	7.8E-3
1973	1.4	5.0E+3	2.7E-4	6.5E-1	1.3E-6	3.1E-3
1974	1.7	6.2E+3	3.1E-6	7.4E-3	3.2E-6	7.6E-3
1975	1.7	6.2E+3	1.3E-6	3.1E-3	1.3E-6	3.1E-3
1976	3.0	1.1E+4	1.0E-6	2.4E-3	1.1E-6	2.6E-3
1977	2.2	8.0E+3	1.6E-6	3.8E-3	1.6E-6	3.8E-3
1978	2.0	7.2E+3	2.0E-6	4.8E-3	1.1E-6	2.6E-3
1979	1.6	5.7E+3	8.5E-7	2.0E-3	1.1E-6	2.6E-3
1980	1.4	5.0E+3	5.2E-7	1.2E-3	9.2E-7	2.2E-3
1981	1.6	5.7E+3	5.0E-7	1.2E-3	1.1E-6	2.6E-3
1982	1.4	5.0E+3	5.0E-7	1.2E-3	1.1E-6	2.6E-3
1983	1.1	3.9E+3	8.9E-7	2.1E-3	1.9E-6	4.7E-3
1984	1.3	4.7E+3	5.0E-7	1.2E-3	1.1E-6	2.6E-3
1985	1.0	3.6E+3	2.2E-7	5.3E-4	1.1E-6	2.6E-3
1986	0.9	3.3E+3	1.5E-7	3.6E-4	1.3E-6	2.8E-3
1987	1.7	6.2E+3	1.9E-7	4.6E-4	1.3E-6	3.1E-3
1988	1.3	4.7E+3	3.3E-8	7.9E-5	2.7E-6	6.5E-3
1989	0.9	3.3E+3	1.4E-7	3.4E-4	1.1E-6	2.8E-3
1990	0.5	1.8E+3	1.5E-7	3.6E-4	6.4E-6	1.5E-2
1991	0.7	2.6E+3	7.0E-8	1.7E-4	1.9E-6	4.6E-3
1992	0.2	7.2E+2	6.8E-8	1.6E-4	1.1E-6	2.6E-3
1993	0.2	7.2E+2	2.7E-8	6.5E-5	8.7E-7	2.1E-3
1994	0.1	3.6E+2	3.4E-8	8.2E-5	6.8E-7	1.6E-3
1995	0.07	2.6E+2	3.5E-8	8.4E-5	6.9E-7	1.7E-3
1996	0.2	7.2E+2	2.4E-8	5.8E-5	7.7E-7	1.9E-3
1997	0.1	3.6E+2	8.8E-9	2.1E-5	6.6E-7	1.6E-3
1998	0.09	3.3E+2	8.6E-9	2.1E-5	4.6E-7	2.1E-3
1999	0.09	3.3E+2	5.5E-9	1.3E-5	8.9E-7	2.1E-3
2000	0.05	1.8E+2	9.1E-9	2.2E-5	1.3E-8	3.1E-5
2001	0.04	1.4E+2	9.1E-9	2.2E-5	1.3E-8	3.9E-5
2002	0.64	1.5E+3	6.1E-9	1.5E-5	1.5E-6	5.3E-8
2003	2.8	6.7E+3	4.8E-9	1.2E-5	4.5E-7	1.1E-3
2004	0.06	1.4E+2	4.6E-9	1.1E-5	2.9E-7	7.0E-4
2005	0.5	1.2E+3	1.5E-9	3.6E-6	2.0E-7	4.8E-4

a. The data are the maximum of the median measured value for a specific year from the available stationary monitoring locations.
b. The actual concentration for this isotope was not reported. Isotopes of uranium were summed and reported as U-234.
c. Assume total intake is the sum of inhalation and skin absorption. Total intake is 1.5 times the inhalation intake.
d. Tritium was not reported for 1971.
e. Per Section 4.2.3 an intake of 3×10^7 Bq/yr (1.7×10^4 Bq/m^3) of tritium as HTO should be assumed for 1952 to 1971.

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

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 the same as those for the main site, which is favorable to claimants because the Site 300 values are consistently less than 10% of the main site values. Post 1970, only alpha emitting radionuclides are listed. This is because the gross beta annual intakes post 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 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 2,400 m^3/yr.

All environmental internal doses assigned from 1971 through 1973 are considered partial dose assessments due to the LLNL SEC for MFP.

Table 4-8. Site 300 maximum annual median air concentrations and intakes via inhalation, 1971 to 2005.

	Tritium[a]		Pu-239+240[b]		U-234[c]	
Year	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)	Air concentration (Bq/m^3)	Annual intake (Bq/yr)
1971	(d, e)	(d, e)	4.1E-6	9.8E-3	1.1E-5	2.6E-2
1972	1.4	5.0E+3	2.3E-6	5.5E-2	2.9E-5	7.0E-2
1973	1.4	5.0E+3	2.1E-6	5.0E-3	9.1E-6	2.2E-2
1974	1.7	6.2E+3	4.1E-6	9.8E-3	9.3E-6	2.2E-3
1975	1.7	6.2E+3	2.1E-6	5.0E-3	6.2E-6	1.5E-2
1976	3.0	1.1E+4	6.2E-6	1.5E-2	2.0E-6	4.8E-3
1977	2.2	8.0E+3	8.6E-6	2.1E-2	1.5E-6	3.6E-3
1978	2.0	7.2E+3	1.2E-5	3.0E-2	2.8E-6	6.7E-3
1979	1.6	5.7E+3	6.2E-6	1.5E-2	1.6E-6	3.7E-3
1980	1.4	5.1E+3	8.5E-7	2.1E-3	1.8E-6	4.3E-3
1981	1.6	5.7E+3	8.6E-5	2.1E-1	1.7E-6	4.0E-3
1982	1.4	5.1E+3	8.5E-7	2.1E-3	1.8E-6	4.3E-3
1983	1.1	3.9E+3	2.0E-5	4.9E-2	7.4E-8	1.8E-4
1984	1.3	4.7E+3	8.5E-7	2.1E-3	1.8E-6	4.3E-3
1985	1.0	3.6E+3	3.0E-7	7.2E-4	1.1E-6	2.1E-3
1986	0.9	3.3E+3	7.0E-8	1.7E-4	8.3E-7	2.0E-3
1987	1.7	6.2E+3	5.9E-8	1.4E-4	2.8E-6	6.7E-3
1988	1.3	4.7E+3	3.9E-7	9.6E-4	2.7E-6	6.5E-3
1989	0.9	3.3E+3	3.5E-8	8.4E-5	2.5E-6	5.9E-3
1990	0.5	1.8E+3	1.2E-8	2.8E-5	2.5E-6	5.8E-3
1991	0.7	2.6E+3	7.8E-8	1.8E-4	3.4E-6	8.0E-3
1992	0.2	7.2E+2	1.4E-8	3.4E-5	3.0E-6	7.2E-3
1993	0.2	7.2E+2	7.3E-9	1.8E-5	1.9E-6	4.6E-3
1994	0.1	3.6E+2	7.3E-9	1.8E-5	1.9E-6	4.6E-3
1995	0.07	2.6E+2	1.2E-8	2.9E-5	1.7E-7	4.1E-4
1996	0.2	7.2E+2	1.0E-8	2.4E-5	2.4E-6	5.8E-3
1997	0.1	3.6E+2	1.7E-8	4.1E-5	2.2E-6	5.3E-3
1998	0.09	3.3E+2	1.6E-8	3.8E-5	7.0E-6	1.7E-2
1999	0.09	3.3E+2	8.7E-9	2.1E-5	2.2E-6	5.3E-3
2000	0.05	1.8E+2	1.9E-8	4.6E-5	3.9E-6	1.4E-2
2001	0.04	1.4E+2	9.8E-9	2.3E-5	8.8E-6	2.1E-2
2002	0.64	1.5E+3	1.0E-8	2.4E-5	1.2E-6	2.9E-3
2003	2.8	6.7E+3	1.8E-9	4.3E-6	7.3E-7	1.8E-3
2004	0.06	1.4E+2	4.5E-9	1.1E-5	6.3E-7	1.5E-3
2005	0.5	1.2E+3	4.3E-9	1.0E-5	2.0E-6	4.8E-3

a. From Table 4-7.
b. Originating from fallout (Gallegos 1998).
c. Isotopes of uranium were summed and reported as U-234.
d. Tritium was not reported for 1971.
e. Per Section 4.2.3 an intake of 3×10^7 Bq/yr (1.7×10^4 Bq/m^3) of tritium as HTO should be assumed for 1955 to 1971.

4.3 INTAKES FROM TRITIUM IN DRINKING WATER FROM 1952 TO 1978

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 a site called POOL, which was the onsite swimming pool. The median activity in the drinking water was

estimated from calculated values and found to be below the minimum analytical detectable activity. Concentrations of tritium in the POOL location ranged from 0.8 to 200 Bq/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 3 L of water per day or approximately 1,100 L/yr. As a consequence, the estimated potential for ingestion of tritium ranged from 550 to 220,000 Bq/yr. Table 4-9 lists the potential for ingestion of tritium in drinking water over time. The values in the table before 1966 were selected from the maximum concentration that was reported for the period from 1966 to 2005. 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.

Table 4-9. Maximum sitewide annual intakes (Bq/yr) from tritium via drinking water for main site and Site 300[c].

Year	Water concentration (Bq/L)	Annual intake[a] (Bq/yr)	Year	Water concentration (Bq/L)	Annual intake[a] (Bq/yr)
1952	200[b]	2.2E+5	1979	11	1.2E+4
1953	200[b]	2.2E+5	1980	26[b]	2.9E+4
1954	200[b]	2.2E+5	1981	26[b]	2.9E+4
1955	200[b]	2.2E+5	1982	26[b]	2.9E+4
1956	200[b]	2.2E+5	1983	26[b]	2.9E+4
1957	200[b]	2.2E+5	1984	26[b]	2.9E+4
1958	200[b]	2.2E+5	1985	7.4	8.1E+3
1959	200[b]	2.2E+5	1986	7.4	8.1E+3
1960	200[b]	2.2E+5	1987	3.7	4.1E+3
1961	200[b]	2.2E+5	1988	6.6	7.2E+3
1962	200[b]	2.2E+5	1989	7[b]	7.7E+3
1963	200[b]	2.2E+5	1990	7[b]	7.7E+3
1964	200[b]	2.2E+5	1991	4.5	5.0E+3
1965	200[b]	2.2E+5	1992	4.4	4.8E+3
1966	185	2.0E+5	1993	1.0	1.1E+3
1967	185	2.0E+5	1994	2.8	3.1E+3
1968	185	2.0E+5	1995	0.8	8.8E+2
1969	185	2.0E+5	1996	2.1	2.3E+3
1970	200	2.2E+5	1997	2.4	2.6E+3
1971	200[b]	2.2E+5	1998	1.2	1.3E+3
1972	200[b]	2.2E+5	1999	2.2	2.4E+3
1973	200[b]	2.2E+5	2000	2.0	2.2E+3
1974	200[b]	2.2E+5	2001	3.0	3.3E+3
1975	200[b]	2.2E+5	2002	4.8	5.3E+3
1976	26	2.9E+4	2003	5.7	6.3E+3
1977	26[b]	2.9E+4	2004	3.1	3.4E+3
1978	26[b]	2.9E+4	2005	0.5	5.5E+2

a. Assume intake of 3 L of water per day; 1,100 L ingested per year.
b. The data for this year were not provided by LLNL. The maximum concentration was reported in 1970. Therefore the 1970 maximum annual tritium intake should be used for the years where no data was reported.
c. Site 300 started in 1955. Tritium doses shold be assigned from 1995 and beyond.

4.4 AMBIENT RADIATION

4.4.1 Gamma Radiation

From 1976 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 2005 (see the References list).

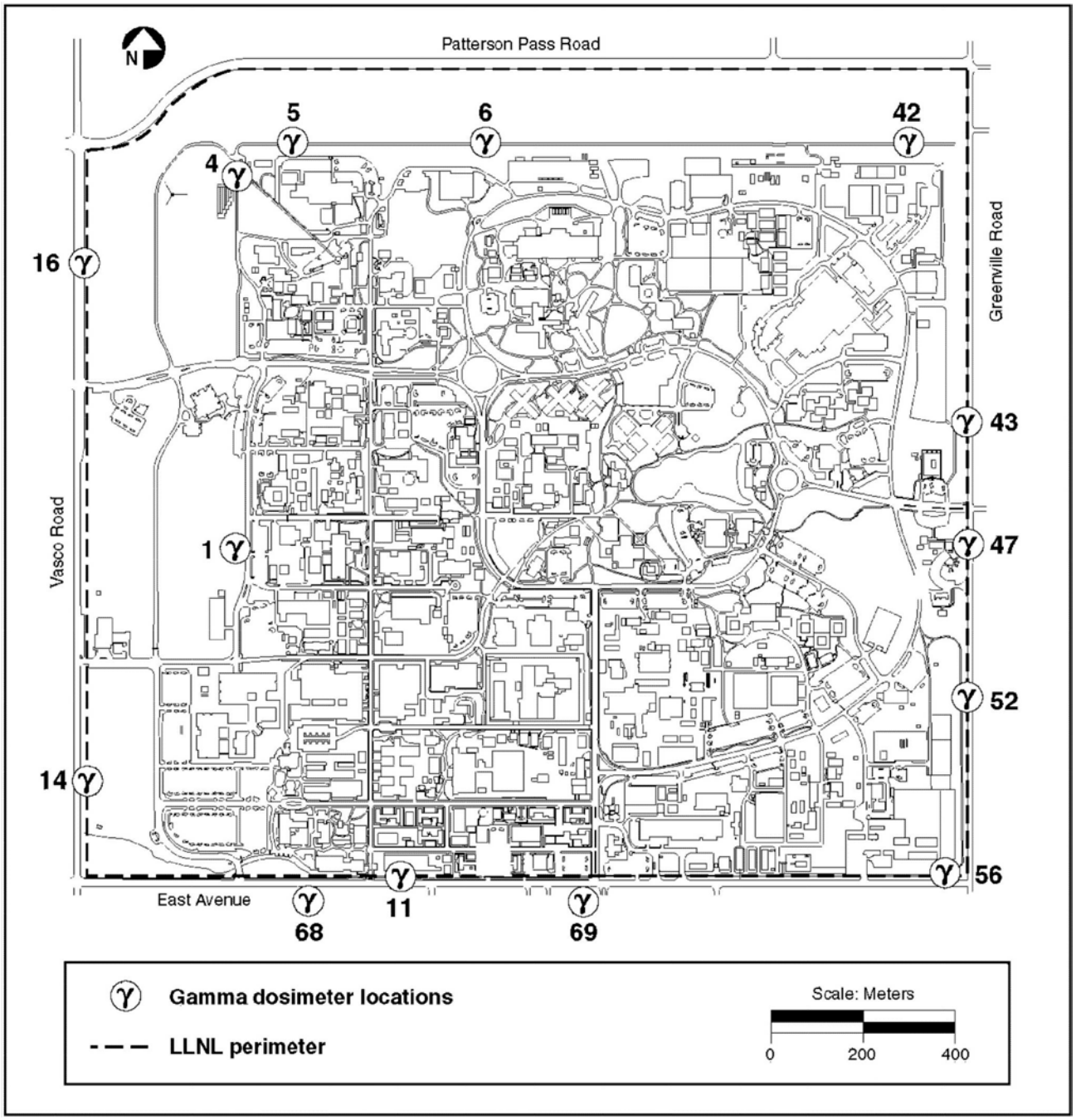

Figure 4-4. Main site gamma dosimeter locations, 2001 (Gallegos et al. 2002).

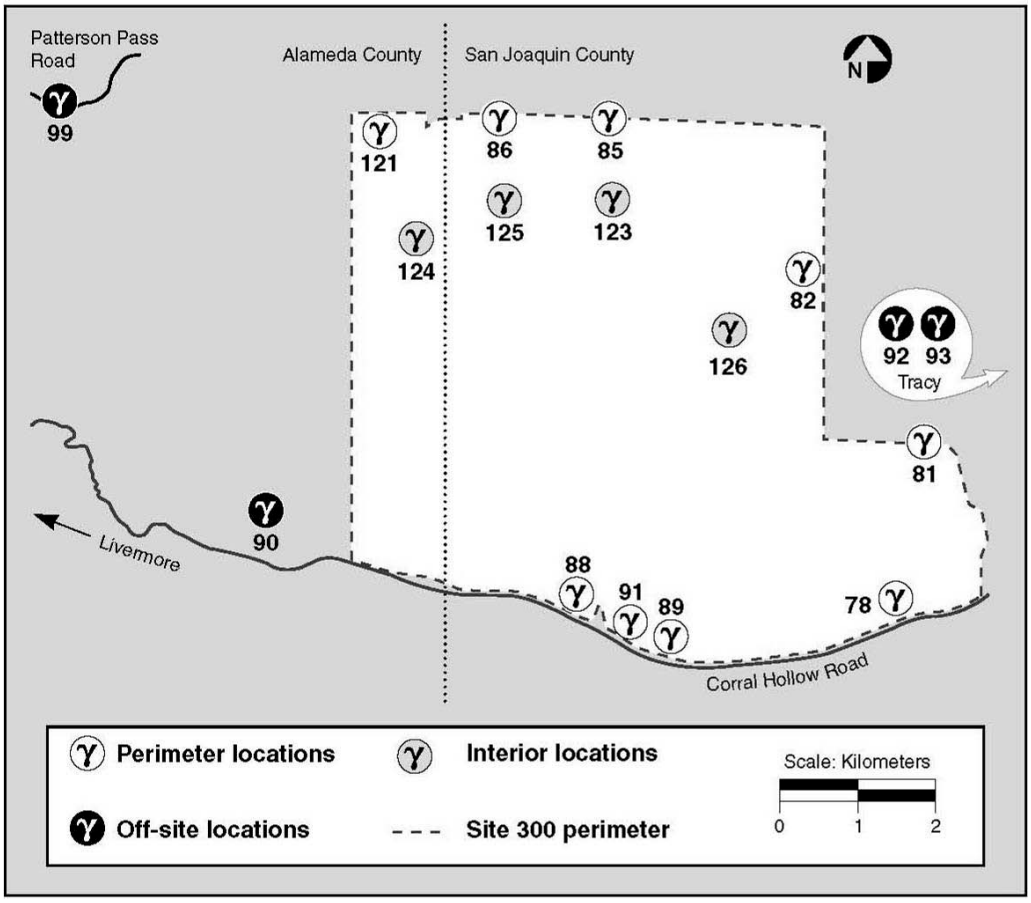

Figure 4-5. Site 300 and vicinity gamma dosimeter locations, 2001 (Gallegos et al. 2002).

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.

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 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. Before 1998, external environmental exposures were not monitored in relation to workers; that is, monitoring stations were not normally at the interior of the site among the process buildings. Because of data availability, therefore, public exposure information had to be used for worker environmental exposures. The maximum value of environmental exposure was recommended for years when data were unavailable to compensate for lack of worker-specific environmental dose information.

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. Tables 4-10 and 4-11 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 4-10, and 4-11 should be adjusted in accordance with ORAUT-PROC-0060, *Occupational Onsite Ambient Dose Reconstruction for DOE Sites* (ORAUT 2006).

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 accelerator, which began operations in 1966 in Building 212 and produced 14-MeV neutrons (Olson 1974). Figure 4-6 shows the locations of the environmental neutron dosimeters. Stations 5 and 3 were typically the maximum value locations.

Table 4-12 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 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 4-12 should be adjusted in accordance with ORAUT-PROC-0060, *Occupational Onsite Ambient Dose Reconstruction for DOE Sites* (ORAUT 2006).

Potential ambient neutron dose is captured by application of the neutron-to-photon ratio to gamma dose (applied pre-1969) or by using measured and missed neutron dose data for the period from 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 consider adding additional ambient neutron dose for monitored workers.

If dosimetry data are not available for a worker, and if the worker was likely located in the area of the LLNL campus near Building 212 from 1969 to 1987, then the maximum ambient neutron dose value may be adjusted in accordance with ORAUT-PROC-0060, *Occupational Onsite Ambient Dose Reconstruction for DOE Sites* (ORAUT 2006). 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

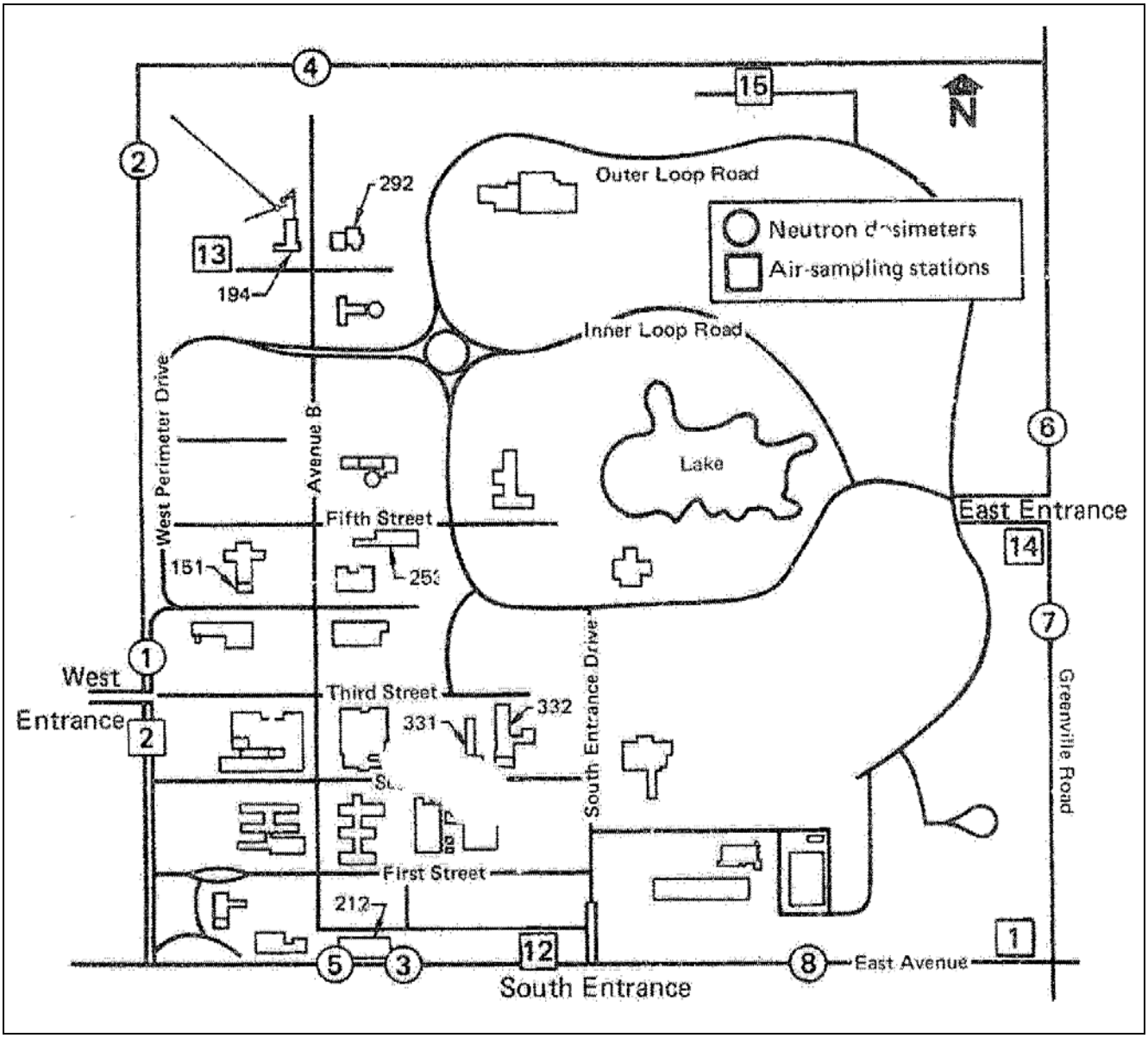

Figure 4-6. Main site neutron dosimeter locations (Griggs, Myers, and Buddemeier 1984).

Protection Publication 60 weighting factor correction of 1.91 (ICRP 1991). The value for 1972 should be used for 1969 to 1971. If the worker was near Building 212 from 1966 to 1969, the neutron-to-photon ratio can be used. If photon dosimetry data is not available for this period, then the value for 1972 can be used for 1966 to 1969.

Table 4-10. Main site external gamma radiation (mrem/yr) [7].[a]

Year	Average, 8,760 hr/yr	Maximum, 8,760 hr/yr
1952	89[b]	317[c]
1953	89[b]	317[c]
1954	89[b]	317[c]
1955	89[b]	317[c]
1956	89[b]	317[c]
1957	89[b]	317[c]
1958	89[b]	317[c]

Year	Average, 8,760 hr/yr	Maximum, 8,760 hr/yr
1959	50	317[c]
1960	89[b]	317[c]
1961	88	317[c]
1962	44	317[c]
1963	50	317[c]
1964	50	317[c]
1965	50	317[c]
1966	89[b]	317[c]
1967	20	317[c]
1968	20	163
1969	34	83
1970	15	105
1971	77	257
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
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

a. The results in this table reflect data provided by LLNL for an exposure period of 8,760 hr/yr. The dose values based on 8760 hr/yr per year should be adjusted in accordance with ORAUT 2006.

b. External dose for these years was not available. The maximum average external

dose was reported in 1972. Therefore, the 1972 dose values are used for the noted years.

c. 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.

Table 4-11. Site 300 external gamma radiation (mrem/yr) [8].[a]

Year	Average, 8,760 hr/yr	Maximum, 8,760 hr/yr
1955	88[b]	99[b]
1956	88[b]	99[b]
1957	88[b]	99[b]
1958	88[b]	99[b]
1959	88[b]	99[b]
1960	88[b]	99[b]
1961	88[b]	99[b]
1962	88[b]	99[b]
1963	88[b]	99[b]
1964	88[b]	99[b]
1965	88[b]	99[b]
1966	88[b]	99[b]
1967	88[b]	99[b]
1968	88[b]	99[b]
1969	88[b]	99[b]
1970	88[b]	99[b]
1971	88[b]	99[b]
1972	88[b]	99[b]
1973	88[b]	99[b]
1974	88[b]	99[b]
1975	88[b]	99[b]
1976	88[b]	99[b]
1977	88[b]	99[b]
1978	88[b]	99[b]
1979	88[b]	99[b]
1980	88[b]	99[b]
1981	88[b]	99[b]
1982	88[b]	99[b]
1983	88[b]	99[b]
1984	88[b]	99[b]
1985	88[b]	99[b]
1986	88[b]	99[b]
1987	88[b]	99[b]
1988[d]	88[b]	99[b]
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

Year	Average, 8,760 hr/yr	Maximum, 8,760 hr/yr
2000	64	70
2001	64	66
2002	67	76
2003	67	74
2004	69	77
2005	69	83

a. The results in this table reflect data provided by LLNL for an exposure period of 8,760 hr/yr. The dose values based on 8760 hr/yr should be adjusted in accordance with ORAUT 2006.
b. External dose for these years was not available. The average and maximum external doses were reported in 1994. Therefore, the 1994 dose values are used for the years noted.
c. Site average (millirem per year) is based on the maximum value from later years where monitoring was performed. Therefore, no maximum or uncertainty is provided.
d. Environmental radiation monitoring at Site 300 began on July 1, 1988.

Table 4-12. Main site external neutron radiation (mrem/yr) [9].[a]

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

a. The results in this table reflect data provided by LLNL for an exposure period of 8,760 hr/yr. The dose values based on 8760 hr/yr should be adjusted in accordance with ORAUT 2006.

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. Integrated Environmental Management. 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, ^{239}Pu was used as the surrogate for gross alpha, ^{137}Cs was used as the surrogate for gross gamma, and ^{90}Sr was used as the surrogate for gross beta and mixed fission products to provide conservative dose estimates."

[2] Szalinski, Paul A. Integrated Environmental Management. 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. Integrated Environmental Management. Health Physicist. April 2007. The surrogate radionuclide for gross beta at Site 300 is ^{234}Th. Only tritium and depleted uranium were reported as released into the environment at Site 300 (Gallegos et al. 1998, 2000). ^{234}Th is a progeny of the ^{238}U.

[4] Szalinski, Paul A. Integrated Environmental Management. Health Physicist. March 2007. A GSD of 3 is used in ORAUT-OTIB-0018, *Internal Dose Overestimates for Facilities with Air Sampling Programs* (ORAUT 2005).

[5] Szalinski, Paul A. Integrated Environmental Management. Health Physicist. March 2007. 200 TBq was arrived at as an annual average by assuming that the 4 PBq that was not attributed to the two major release events was released over a 20-year period rather than the actual 40-year period. This results in an annual release that is overestimated and favorable to the claimant.

[6] Szalinski, Paul A. Integrated Environmental Management. Health Physicist. March 2007. The assumption that all the tritium is HTO is favorable to the claimant because, according to ICRP Publication 68, HTO has a much higher dose coefficient than tritium gas (ICRP 1995).

[7] Szalinski, Paul A. Integrated Environmental Management. 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.

[8] Szalinski, Paul A. Integrated Environmental Management. 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.

[9] Smith, Matthew H. Dade Moeller & Associates. Sr. Health Physicist. November 2009. 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. Alpha particles have low penetrating power and a short range (a few centimeters in air). Outside the body, the most energetic alpha particle generally fails to penetrate the dead layers of cells covering the skin or a sheet of paper. Alpha particles emitted by radionuclides inside the body are a more significant health risk.

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. Most of the direct fission products are (negative) beta emitters. Exposure to large amounts of beta radiation from external sources can cause skin burns (erythema), and beta emitters can be harmful inside the body. Thin sheets of metal or plastic can stop beta particles.

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
Disintegration of certain elements (e.g., radium, actinium, uranium, and thorium) accompanied by the emission of alpha, beta, gamma, and/or neutron radiation from unstable 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 average American receives 360 millirem a year from background radiation. 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 (TLD)
Device for measuring radiation dose that consists of a holder containing solid chips of material that, when heated by radiation, release the stored energy as light. The measurement of this light provides a measurement of absorbed dose.