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ACRONYMS AND ABBREVIATIONS

CEDE	committed effective dose equivalent
conc	concentration
DOE	U.S. Department of Energy
DU	depleted uranium (less than 0.7 percent ^{235}U)
EPA	U.S. Environmental Protection Agency
LLNL	Lawrence Livermore National Laboratory
LRL	Lawrence Radiation Laboratory
MEI	maximally exposed individual
NESHAPS	National Emissions Standards for Hazardous Air Pollutants
QA	quality assurance
SW-MEI	site-wide maximally exposed individual
TBD	technical basis document
TLD	thermoluminescent dosimeter
U.S.C.	United States Code

4.0 OCCUPATIONAL ENVIRONMENTAL DOSE

Site Profiles, which include Technical basis documents (TBDs), 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 when additional relevant information is obtained about the affected site(s). These documents may be used to assist the 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 facility" as defined in the Energy Employees Occupational Illness Compensation Program Act of 2000 (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 DOE employees with cancer, the DOE facility definition only determines eligibility for a dose reconstruction, which is a prerequisite to a compensation decision (except for members of the Special Exposure Cohort). The compensation decision for cancer claimants is based on a section of the statute entitled "Exposure in the Performance of Duty." That provision (42 U.S.C. § 7384n(b)) says that an individual with cancer "shall be determined to have sustained that cancer in the performance of duty for purposes of the compensation program if, and only if, the cancer … was at least as likely as not related to employment at the facility [where the employee worked], as determined in accordance with the [probability of causation] guidelines established under subsection (c) …." 42 U.S.C. § 7384n(b). Neither the statute nor the probability of causation guidelines (nor the dose reconstruction regulation) define "performance of duty" for DOE employees with a covered cancer or restrict the "duty" to nuclear weapons work.

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

This Site Profile documents historical practices at the Lawrence Livermore National Laboratory (LLNL) and provides information for the evaluation of internal and external dosimetry data for unmonitored and monitored workers; it can serve as a supplement to, or substitute for, individual monitoring data.

Occupational environmental dose refers to exposures received by workers outside the facilities at the Lawrence Livermore National Laboratory (LLNL) from elevated ambient radiation, from facility discharges to the environment, and from 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 estimated radiation dose as a result of ambient exposures at LLNL. Environmental measurements do not distinguish the source of emissions and, therefore, will reflect air concentrations from nearby as well as distant sources. The estimates of emissions were useful in filling in some gaps in measurement data and were critical to estimating exposures before the start of comprehensive and routine measurement data reports.

4.1 INTRODUCTION

LLNL began operations in 1952, when it was known as the University of California Radiation Laboratory, a branch of what is now the Lawrence Berkeley National Laboratory, or Lawrence Radiation Laboratory (LRL) at Livermore, California. 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, which conducts research and development activities, and Site 300, where explosive testing experiments occur. Site 300 began operations in 1955. There was a potential for internal dose from the breathing of airborne concentrations of radionuclides released on the LLNL site; Section 4.2 of this TBD contains detailed information about the collection and analysis of air samples and how this information is used for dose reconstruction. There was a potential for ingestion of radioactive materials from resuspension of radioactive materials (Section 4.3) and the ingestion of water potentially containing tritium (Section 4.4). Section 4.5 describes the potential external dose from sources of radiation outside and inside the process buildings and Section 4.6 discusses possible sources of uncertainty associated with each of these sources of environmental dose.

The receptors of concern addressed in this TBD were unmonitored workers, namely LLNL employees who did not wear external dosimetry or who were not monitored for internal exposures. At LLNL, everyone, including administrative personnel, wore some form of external radiation dosimetry as of March 1958 (Nolan 1958). To provide the basis for estimating the environmental dose for years when monitoring did not occur or was not sufficient to apply to coworkers, this TBD provides annual intakes and ambient external dose from 1952 (1955 for Site 300) to 2001 (the last year with publicly available data).

4.2 INTERNAL DOSE FROM ONSITE ATMOSPHERIC RADIONUCLIDE CONCENTRATION

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

4.2.1.1 Pre 1971

Air samples were collected in 1961 in eleven 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 seven day period on HV-70 paper, measuring 4x9 inches. The other air sampling stations were located at a variety of distances ranging from the perimeter to as far as five miles from the perimeter. As shown on Table 4-1, sample locations SALV and MESQ were established as early as 1961, for the purposes of perimeter air monitoring. At Site 300, six air sampling locations were established in 1962; each of the locations were located within the boundary of the Site 300 perimeter. The air sampling equipment located at Site 300 was equivalent to the equipment used at the main site. As shown in Table 4-2, sample locations Bunker 801E, ECP, EOBS, GOLF, WCP and WOBS were established in 1962.

4.2.1.2 Post 1971

In 1971, LLNL established a network of permanent stations to collect air samples from the perimeter of the facility and other locations inside the facility (Gallegos and Surano 1992). LLNL collected and analyzed air samples for gross alpha and beta radiations, ^{3}H, ^{239}Pu and ^{240}Pu, and ^{235}U and ^{238}U. The existing monitoring networks were established in 1992 for surveillance of air particulates and tritium in the environs of the Livermore site and Site 300, as well as in the surrounding Livermore Valley and the City of Tracy (Gallegos and Surano 1992). Tables 4-1 and 4-2 list the sampling locations for the monitoring network at LLNL and Site 300, respectively, and Figures 4-1 and 4-2 show these locations.

Table 4-1. Ambient air sampling station locations within LLNL boundary.

Designation	Location	Direction from center of LLNL
COW	Located 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 maximum exposed individual	SE

Table 4-2. Ambient air sampling station locations at Site 300.

Designation	Location	Direction from center of Site 300
COHO	Near south perimeter of facility; representative of maximum exposed individual	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

4.2.1.3 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 be reasonably detected regardless of local meteorology (LLNL 1986). This demonstrated compliance with U.S. Department of Energy (DOE) Derived Concentration Guidelines 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.[1] This TBD considers only those ambient air monitoring locations inside the LLNL security fence for worker intake. Table 4-1 lists these monitors and their locations.

[1] 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 for LLNL.

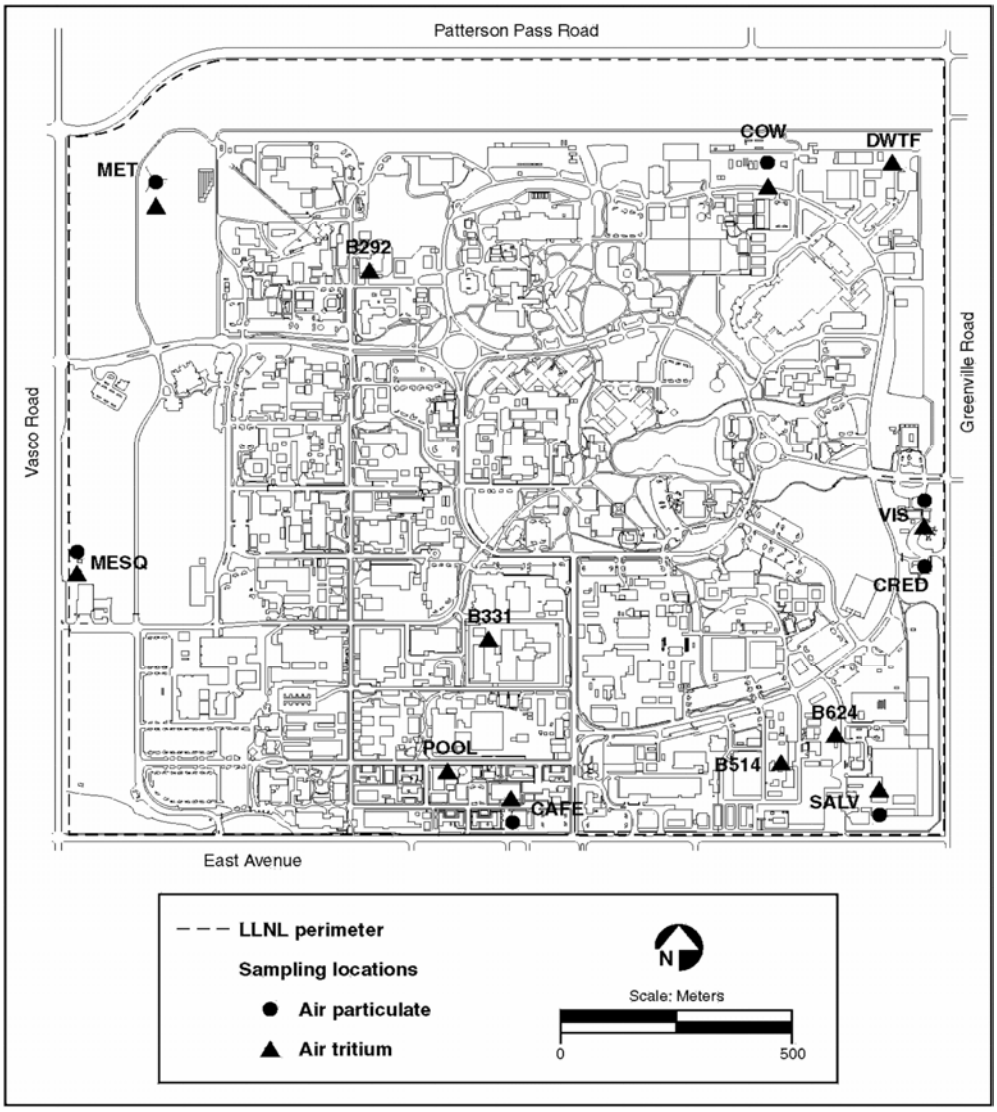

Figure 4-1. Air particulate and tritium sampling locations on Livermore site (Source: LLNL 2002).

Air samplers were positioned to provide reasonable probability that any significant concentration of radioactive effluents from LLNL operations was detected. The Livermore site radiological air particulate sampling network consisted of seven samplers at the perimeter with one (CRED) serving as the site-wide maximally exposed individual (SW-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 attributed to historical operations, which included the operation of solar evaporators for plutonium-containing liquid waste.

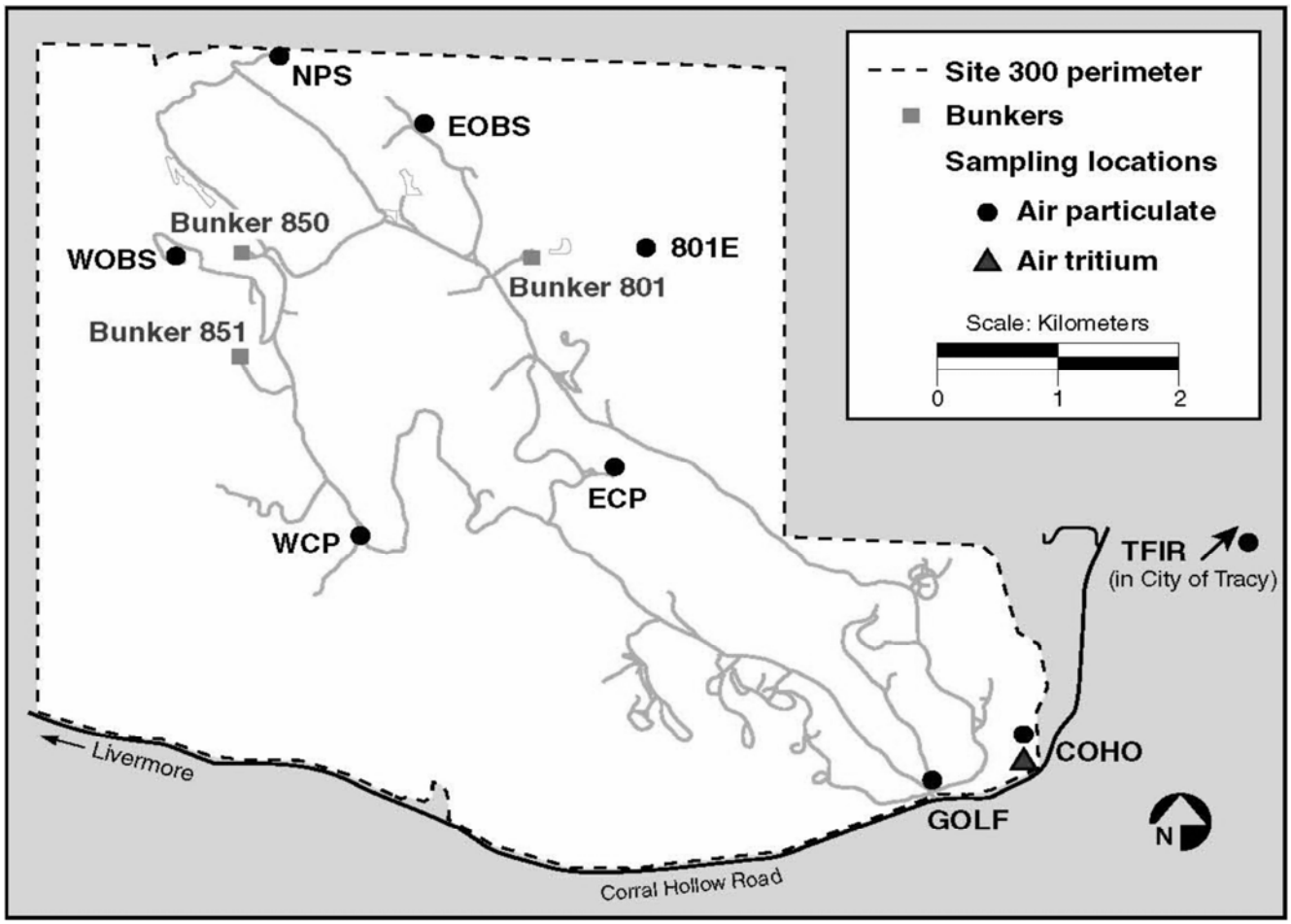

Figure 4-2. Air particulate and tritium sampling locations at Site 300 and off site (Source, LLNL, 2002).

The Site 300 air particulate monitoring network included eight sampling units 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 SW-MEI 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 (LLNL 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 Livermore 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).

4.2.1.4 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, and for specific isotopes that represented more than 90% of the LLNL radioactive materials inventory. Specifically, it analyzed air samples for the presence of tritium, ^{239}Pu, and isotopes of uranium. Tables 4-3 (a and b)

and 4-4 (a and b) list the results for ^{234}U because the radiation dose conversion factor was greater for that isotope than for ^{238}U and ^{235}U. For the periods of 1961 through 1971, the gross alpha concentration should be used to calculate the presence of ^{239}Pu and a second calculation using ^{234}U; the assigned dose should be selected from the higher of the two calculations. The gross beta concentration should be assumed to be thorium 234.

For Site 300, radionuclides of significance were selected according to the inventory of radioactive materials and the type of work performed at that site. Specifically, the presence of depleted uranium (DU) indicated the analysis of ^{238}U and ^{235}U. For the purpose of this TBD, the dose reconstructor should assign the dose associated with ^{235}U and ^{234}U for depleted uranium. For the periods of 1961 through 1971, the gross alpha concentration for Site 300 should be used to calculate the presence of ^{235}U and a second calculation using ^{234}U; the assigned dose should be selected from the higher of the two calculations. The gross beta concentration should be assumed to be thorium 234.

4.2.2 Methodology

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 and 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 (LLNL 1992a).

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 detected on air filters were used only as trend indicators; specific radionuclide analysis occurred for plutonium and uranium, depending on the location. All analytical results were reported as a measured concentration per volume of air. When activity was less than the minimum detection 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 standards certified by the EPA. 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 that occurred in the counting equipment. The analytical laboratory reported the actual instrumentation values, including negative results that occurred when background measurements were higher than those for the filters.

Concentrations of various airborne radionuclides were measured at Livermore site perimeters, at offsite locations near the Livermore site, and at Site 300. Prior to 1964, air samples were collected from two perimeter locations and nine offsite locations on 4-in. by 9-in. HV-70 paper at a sample flow rate of approximately 4 ft^3/min of air (LRL, 1964). The samples were collected for a period of 7 days. After a 4-day delay for decay of the radon/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. Following gamma counting, the perimeter filters were grouped by sampling locations. 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 site

perimeter were sampled (LRL, 1964). The six samplers on the Laboratory perimeter used 5.2×10^{-2} m^2 Whatman-41 cellulose filters. The particulates were collected on Whatman filters using an average air flow rate of 700 L/min (Lindeken and Wang, 1978). Tritium was collected with columns packed with silica gel, with 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 control samplers operated in parallel with the permanent sampler at a given site, and samples from these duplicates were analyzed to confirm results (Lindeken and Wang, 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 (Althouse 1998) including ^{235}U, ^{238}U, ^{40}K, ^{226}Ra, ^{228}Ra, and ^{228}Th. In fact, the amount of these naturally occurring isotopes in the filters was often greater than the amount of the isotopes being filtered from the air. The filters were 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 (LLNL 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 analysis was 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 maximally exposed individual (from the total particulate) was below 10 mrem committed effective dose equivalent (CEDE; LLNL 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 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 used 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 (QA) samples were processed to confirm the precision of the analytical results obtained from the samplers. A duplicate QA 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 route. The QA trip blanks and QA duplicates were processed in the same manner as that used for routine samples and analyzed for the same radiological parameters.

Table 4-3a. Maximum site-wide annual median intakes (Bq/yr) via inhalation.[a]

Year	Gross Alpha		Gross Beta	
	Conc (Bq/m^3)	Intake (Bq/yr)	Conc (Bq/m^3)	Intake (Bq/yr)
1961	3.1E-4	0.9	9.3E-3	28
1962	1.7E-4	0.5	9.0E-3	27
1963	1.2E-4	0.4	1.3E-1	390
1964	1.2E-4	0.4	2.4E-2	72
1965	3.7E-5	0.1	6.5E-3	20
1966	3.7E-5	0.1	2.6E-3	7.8
1967	2.9E-5	0.09	4.7E-3	14
1968	3.7E-5	0.1	4.4E-3	13
1969	3.7E-5	0.1	3.8E-3	11
1970	3.7E-5	0.1	5.6E-3	17
1971	7.5E-5	0.2	1.5E-2	45

a. Assume an inhalation rate of 20 L/min for 2,500 hr per year.

Table 4-3b. Maximum site-wide annual median intakes (Bq/yr) via inhalation.[a]

Year	Tritium		Pu-239+240		U-234	
	Conc (Bq/m^3)	Intake[b] (Bq/yr)	Conc (Bq/m^3)	Intake (Bq/yr)	Conc (Bq/m^3)	Intake (Bq/yr)
1972	1.4[c]	6.4E3	1.3E-6[c]	3.9E-3	1.3E-6[c]	3.9E-3
1973	1.4	6.4E3	1.3E-6[c]	3.9E-3	1.3E-6[c]	3.9E-3
1974	1.7	7.8E3	1.3E-6[c]	3.9E-3	1.3E-6[c]	3.9E-3
1975	1.7	7.8E3	1.3E-6[c]	3.9E-3	1.3E-6[c]	3.9E-3
1976	3.0	1.4E4	1.0E-6	3.0E-3	1.1E-6	3.3E-3
1977	2.2	1.0E4	1.3E-6[c]	3.9E-3	1.1E-6[c]	3.3E-3
1978	2.0	9.0E3	1.3E-6[c]	3.9E-3	1.1E-6[c]	3.3E-3
1979	1.6	7.1E3	8.5E-7	2.5E-3	1.1E-6	3.3E-3
1980	1.4	6.4E3	5.2E-7	1.5E-3	9.2E-7	2.8E-3
1981	1.6	7.1E3	5.0E-7[c]	1.5E-3	1.1E-6[c]	3.3E-3
1982	1.4	6.4E3	5.0E-7[c]	1.5E-3	1.1E-6[c]	3.3E-3
1983	1.1	4.9E3	5.0E-7[c]	1.5E-3	1.1E-6[c]	3.3E-3
1984	1.3	5.9E3	5.0E-7[c]	1.5E-3	1.1E-6[c]	3.3E-3
1985	1.0	4.5E3	2.2E-7	6.6E-4	1.1E-6	3.3E-3
1986	0.9	4.1E3	1.5E-7	5.6E-4	1.1E-6	3.3E-3
1987	1.7	7.8E3	1.9E-7	5.8E-4	1.3E-6	3.9E-3
1988	1.3	5.9E3	3.3E-8	9.9E-5	2.7E-6	8.1E-3
1989	0.9	4.1E3	7.0E-8[c]	2.1E-4	2.7E-6[c]	8.1E-3
1990	0.5	2.3E3	7.0E-8[c]	2.1E-4	2.7E-6[c]	8.1E-3
1991	0.7	3.3E3	7.0E-8	2.1E-4	1.9E-6	5.8E-3
1992	0.2	9.0E2	6.8E-8	2.0E-4	1.1E-6	3.3E-3
1993	0.2	9.0E2	2.7E-8	8.1E-5	8.7E-7	2.6E-3
1994	0.1	4.5E2	3.4E-8	1.0E-4	6.8E-7	2.0E-3
1995	0.07	3.3E2	3.5E-8	1.0E-4	6.9E-7	2.1E-3
1996	0.2	9.0E2	2.4E-8	7.3E-5	7.7E-7	2.4E-3
1997	0.1	4.5E2	8.8E-9	2.6E-5	6.6E-7	2.0E-3
1998	0.09	4.1E2	8.6E-9	2.6E-5	4.6E-7	1.4E-3
1999	0.09	4.1E2	5.5E-9	1.6E-5	8.9E-7	2.6E-3
2000	0.05	2.3E2	9.1E-9	2.8E-5	1.3E-8	3.9E-5
2001	0.04	1.8E2	9.1E-9	2.8E-5	1.3E-8	3.9E-5

a. Assume an inhalation rate of 20 L/min for 2,500 hr per year.
b. Assume total intake is the sum of inhalation and skin absorption. Total intake is 1.5 × the inhalation intake.
c. The actual concentration for this isotope was not reported. The result is a maximum value reported for a different year. See Section 4.2.3 for additional information.

Table 4-4a. Site 300 maximum annual median intakes (Bq/yr) via inhalation.[a]

	Gross Alpha		Gross Beta	
	Conc (Bq/m^3)	Intake (Bq/yr)	Conc (Bq/m^3)	Intake (Bq/yr)
1961	4.8E-4	1.4	2.6E-3	7.8
1962	2.1E-4	0.6	1.4E-1	420
1963	1.0E-4	0.3	1.6E-1	480
1964	1.3E-4	0.4	2.2E-2	66
1965	2.6E-5	0.1	6.3E-3	19
1966	1.9E-5	0.06	3.2E-3	9.6
1967	1.5E-5	0.05	5.6E-3	17
1968	7.4E-5	0.2	5.6E-3	17
1969	7.4E-5	0.2	5.7E-3	17
1970	7.4E-6	0.02	8.6E-3	26
1971	3.7E-5	0.1	1.7E-2	51

a. Assume an inhalation rate of 20 L/min for 2,500 hr per year.

Table 4-4b. Site 300 maximum annual median intakes (Bq/yr) via inhalation.[a]

	U-238		U-235		Pu-239+240	
Year	Conc (Bq/m^3)	Intake (Bq/yr)	Conc (Bq/m^3)	Intake (Bq/yr)	Conc (Bq/m^3)	Intake (Bq/yr)
1972	2.0E-6[b]	6.0E-3	4.9E-8[b]	1.5E-4	6.2E-6[b]	1.9E-2
1973	2.0E-6[b]	6.0E-3	4.9E-8[b]	1.5E-4	6.2E-6[b]	1.9E-2
1974	2.0E-6[b]	6.0E-3	4.9E-8[b]	1.5E-4	6.2E-6[b]	1.9E-2
1975	2.0E-6[b]	6.0E-3	4.9E-8[b]	1.5E-4	6.2E-6[b]	1.9E-2
1976	2.0E-6	6.0E-3	4.8E-8	1.4E-4	6.2E-6	1.9E-2
1977	2.0E-6[b]	6.0E-3	4.9E-8[b]	1.5E-4	6.2E-6[b]	1.9E-2
1978	2.0E-6[b]	6.0E-3	4.9E-8[b]	1.5E-4	6.2E-6[b]	1.9E-2
1979	1.5E-6	4.5E-3	4.9E-8	1.5E-4	6.2E-6[b]	1.9E-2
1980	1.8E-6	5.4E-3	9.3E-8	2.8E-4	8.5E-7	2.6E-3
1981	1.8E-6[b]	5.4E-3	9.3E-8[b]	2.8E-4	8.5E-7[b]	2.6E-3
1982	1.8E-6[b]	5.4E-3	9.3E-8[b]	2.8E-4	8.5E-7[b]	2.6E-3
1983	1.8E-6[b]	5.4E-3	9.3E-8[b]	2.8E-4	8.5E-7[b]	2.6E-3
1984	1.8E-6[b]	5.4E-3	9.3E-8[b]	2.8E-4	8.5E-7[b]	2.6E-3
1985	1.0E-6	3.0E-3	3.8E-8	1.1E-4	3.0E-7	3.0E-4
1986	8.3E-7	2.5E-3	3.8E-8	1.1E-4	3.9E-8	1.2E-4
1987	2.8E-6	8.4E-3	4.7E-8	1.4E-4	5.9E-8	1.8E-4
1988	2.7E-6	8.1E-3	6.2E-8	1.9E-4	3.9E-7	1.2E-3
1989	3.4E-6[b]	1.0E-2	7.3E-8[b]	2.2E-4	3.9E-7[b]	1.2E-3
1990	3.4E-6[b]	1.0E-2	7.3E-8[b]	2.2E-4	3.9E-7[b]	1.2E-3
1991	3.4E-6	1.0E-2	7.3E-8	2.2E-4	7.8E-8	2.3E-4
1992	3.0E-6	9.0E-3	1.4E-7	4.2E-4	1.4E-8	4.2E-5
1993	1.9E-6	5.7E-3	6.1E-9	1.8E-5	7.3E-9	2.2E-5
1994	1.9E-6	5.7E-3	6.1E-8	1.8E-4	7.3E-9	2.2E-5
1995	1.7E-7	5.1E-4	8.2E-8	2.5E-4	1.2E-8	3.6E-5
1996	2.4E-6	7.2E-3	3.9E-8	1.1E-4	1.0E-8	3.0E-5
1997	2.2E-6	6.6E-3	1.0E-7	3.0E-4	1.7E-8	5.1E-5
1998	7.0E-6	2.1E-2	1.2E-7	3.6E-4	1.6E-8	4.8E-5
1999	2.2E-6	6.6E-3	7.3E-8	2.2E-4	8.7E-9	2.6E-5
2000	3.9E-6	1.7E-2	9.4E-8	2.8E-4	1.9E-8	5.7E-5
2001	8.8E-6	2.6E-2	4.6E-7	1.4E-3	9.8E-9	2.9E-5

a. Assume an inhalation rate of 20 L/min for 2,500 hr per year.
b. The actual concentration for this isotope was not reported. The result is a maximum value reported for a different year. See Section 4.2.3 for additional information.

4.2.3 Estimation of Annual Intake from Airborne Radionuclides

This section discusses 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 1976 and through 2001 (see the reference list at the end of this document). The analysis for this TBD collected measurements from the air monitoring locations listed in Tables 4-1 and 4-2 from these documents and applied them to the methodology described above. If data were not available or additional information was required, these documents were supplemented by information provided by DOE or the University of California (the DOE contractor at LLNL).

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

- Improved monitoring methods reducing the minimum detectable concentrations
- Naturally occurring radioactive material released from a nearby fossil generating plant that has reduced releases over the years through improved environmental controls
- The effect of atmospheric weapons testing and the radioactive decay of fallout

The methodology for the intake from onsite atmospheric radionuclide concentrations applied directly to available air sampling data. Air sampling measurement error and uncertainty were evaluated by using the maximum value reported 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 estimates of airborne concentrations at specific locations around the LLNL site using traditional transport modeling approaches:

- The numerous release points, which include stacks, vents, and other emission sources;
- The characteristics of the release points;
- The limited number of air sampling locations;
- The relatively short distances between the release points and the onsite receptor locations; and
- The density and configurations of buildings at the site.

4.2.3.1 Potential Intakes Prior to 1961

Prior to 1961, air monitoring data were not available for review. Estimated worker intakes in this revision of the TBD are restricted to post 1960 years, beginning in 1961. Efforts are currently in progress for developing intakes that occurred prior to 1961 and may be presented in a future revision of this TBD.

4.2.3.2 Potential Intakes 1961 to 1971

Data from Environmental Reports were available for the years 1961 through 1971 (LRL 1961, 1963, 1964, 1965, 1966, 1967, 1968, 1969, 1970, 1971). The air monitoring data provided no isotopic analysis; the data were reported only for the gross alpha and gross beta radiations. These data are provided in Table 4.3a and 4.4a for the LLNL site and Site 300, respectively. As described in Section 4.2.1.2, the isotopes of significance at LLNL were $^{239+240}$Pu and ^{234}U, both decaying by alpha radiation. Isotopes decaying by beta radiation were determined to be progeny of these two parent isotopes.

Tritium environmental air monitoring data were not available for review. Estimated worker intakes of tritium in this revision of the TBD are restricted to the years beginning with 1972. Efforts are currently in progress for developing tritium intakes that occurred prior to 1972 and may be presented in a future revision of this TBD.

The isotopes of significance at Site 300 are ^{238}U and ^{235}U, both decaying by alpha radiation. The presence of beta radiation indicates the progeny of uranium. This TBD assumed that a worker inhaled 20 L of air per minute for 2,500 hr per year or approximately 3×10^6 L of air per year (3,000 m^3 of air per year). The air monitoring data indicated that potential inhalation exposures at the LLNL site to gross alpha radiation ranged from $3x10^{-5}$ to $3x10^{-4}$ Bq/m^3 or a potential inhalation exposure from 1 to 9 Bq/year. The air monitoring data indicated that potential inhalation exposures at Site 300 to gross alpha radiation ranged from $3x10^{-5}$ to $3x10^{-4}$ Bq/m^3 or a potential inhalation exposure from 1 to 9 Bq/year.

4.2.3.3 Potential Intakes post 1971

Past estimates of offsite doses to members of the public indicated that the potential internal dose from airborne releases to LLNL workers should be relatively low (10 mrem CEDE or less) (LLNL 1977, 1980, 1987, 1988, 1989, 1992a,b, 1993, 1994, 1995, 1996, 1997, 1998, 1999, 2000, 2001, 2002). 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 4-3a lists concentrations for ^{3}H, ^{239}Pu, and ^{234}U. This TBD assumed that a worker inhaled 20 L of air per minute for 2,500 hr per year or approximately 3×10^6 L of air per year (3,000 m^3 of air per year). The concentration of tritium ranged from 0.04 to 3 Bq/m^3; the potential for exposure to tritium ranged from 180 to 14,000 Bq/yr. For tritium, the total uptake by the body increased by 50% to account for absorption by the skin in addition to the inhalation pathway (Gede and Gildea, 1980). The concentration of ^{239}Pu ranged from 9.1×10^{-9} to 1×10^{-6} Bq/m^3; the potential for exposures to ^{239}Pu ranged from 0.00003 to 0.003 Bq/yr. Figure 4-3 shows how the concentration of $^{239+240}$Pu decreased from 1982 through 2001. The concentration dropped by more than 20 times over the 20 years of sampling. The concentration of ^{234}U ranged from 1.3×10^{-8} to 1×10^{-6} Bq/m^3; the potential for exposures to ^{234}U ranged from 0.00004 to 0.003 Bq/yr.

Tables 4-4a and b indicate the potential for exposure to inhalation of airborne radionuclides at Site 300. The concentration of ^{238}U ranged from 8.3×10^{-7} to 3×10^{-6} Bq/m^3; the potential for exposure to ^{238}U ranged from 0.009 to 0.01 Bq/yr. The concentration of ^{235}U ranged from 6×10^{-9} to 1.0×10^{-7} Bq/m^3; the potential for exposure to ^{235}U ranged from 1.8×10^{-5} to 0.001 Bq/yr. The concentration of ^{239}Pu ranged from 9.8×10^{-9} to 1×10^{-6} Bq/m^3; the potential for exposures to ^{239}Pu ranged from 2.6×10^{-5} to 0.003 Bq/yr.

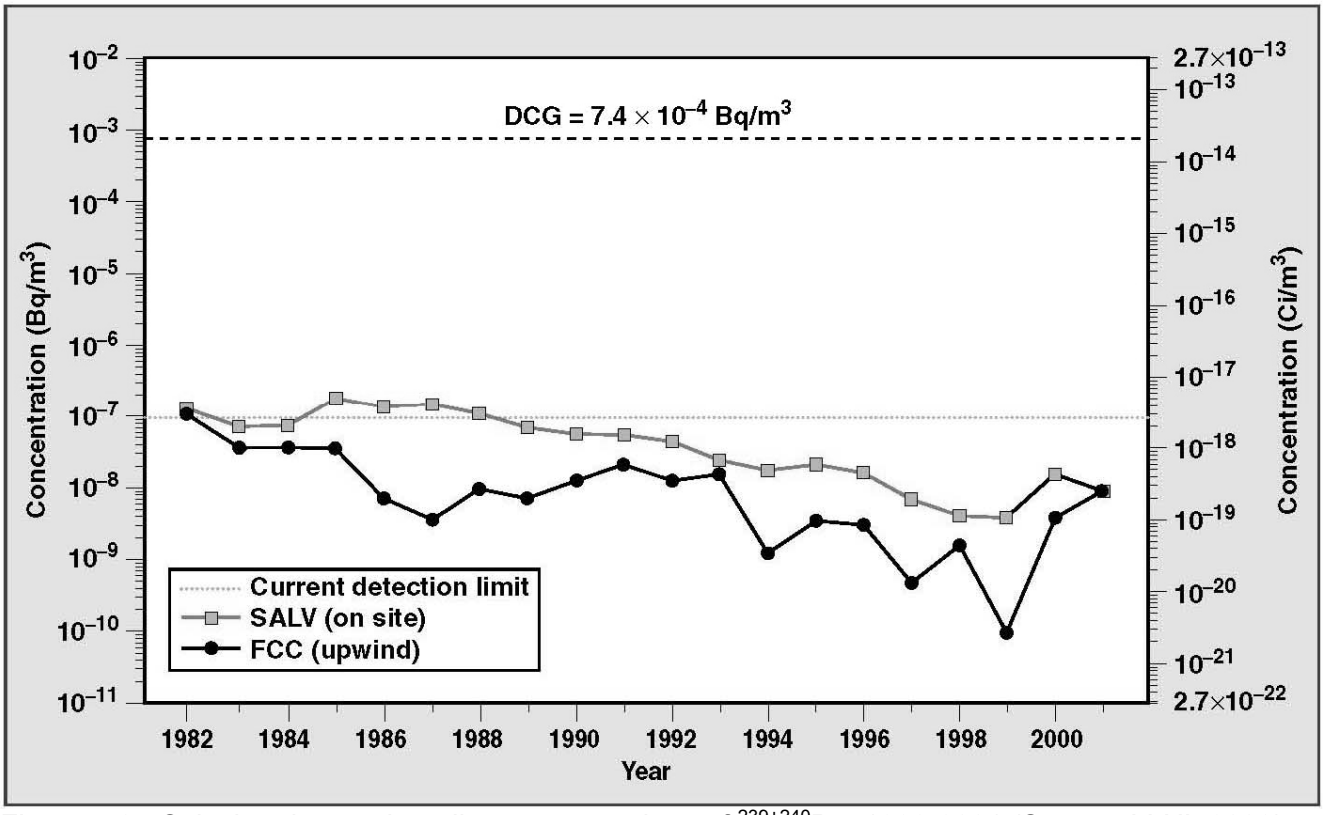

Figure 4-3. Calculated annual median concentrations of $^{239+240}$Pu, 1982-2001 (Source: LLNL 2002).

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 (a and b) and 4-4 (a and b) list these data, and demonstrate that chosen values were reasonable and claimant-favorable. Using the maximum airborne radionuclide concentration in a year (either from actual data or an estimated value), claimant-favorable annual intakes for the radionuclides of concern were derived by using an assumed individual's annual respiration rate of 3,000 m^3/yr. Most years have only a potential, even by being claimant-favorable, for contributing approximately 10 mrem (CEDE) or less.

4.3 INTAKE FROM DRINKING WATER

The annual environmental reports discuss concentrations of radionuclides in the drinking water at LLNL (LRL 1966, 1967, 1968, 1969, 1970, 1970, 1971) (LLNL 1977, 1980, 1987, 1988, 1989, 1992a,b, 1993, 1994, 1995, 1996, 1997, 1998, 1999, 2000, 2001, 2002). Several water sources were sampled and analyzed; BELL, GAS, PALM, ORCH, and TAP were identified as sources of drinking water. The water from these locations was sampled at least once per year. The POOL sample location 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 (LLNL 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 880 to 220,000 Bq/yr. Table 4-5 lists the potential for ingestion of tritium over time. The values in the tables before 1966 were extrapolated and intended to be claimant favorable.

Table 4-5. Maximum site-wide annual intakes (Bq/yr) via drinking water.

Year	Tritium Conc (Bq/l)	Tritium Intake[a] (Bq/yr)	Year	Tritium Conc (Bq/l)	Tritium Intake[a] (Bq/yr)	Year	Tritium Conc (Bq/l)	Tritium Intake[a] (Bq/yr)
1952	100[b]	1.1E5	1969	185	2.0E5	1986	7.4	8.1E3
1953	100[b]	1.1E5	1970	200	2.2E5	1987	3.7	4.1E3
1954	100[b]	1.1E5	1971	100[b]	1.1E5	1988	6.6	7.2E3
1955	100[b]	1.1E5	1972	100[b]	1.1E5	1989	7[b]	7.7E3
1956	100[b]	1.1E5	1973	100[b]	1.1E5	1990	7[b]	7.7E3
1957	100[b]	1.1E5	1974	100[b]	1.1E5	1991	4.5	5.0E3
1958	100[b]	1.1E5	1975	100[b]	1.1E5	1992	4.4	4.8E3
1959	100[b]	1.1E5	1976	26	2.9E4	1993	1.0	1.1E3
1960	100[b]	1.1E5	1977	26[b]	2.9E4	1994	2.8	3.1E3
1961	100[b]	1.1E5	1978	26[b]	2.9E4	1995	0.8	8.8E2
1962	100[b]	1.1E5	1979	11	1.2E3	1996	2.1	2.3E3
1963	100[b]	1.1E5	1980	26[b]	2.9E4	1997	2.4	2.6E3
1964	100[b]	1.1E5	1981	26[b]	2.9E4	1998	1.2	1.3E3
1965	100[b]	1.1E5	1982	26[b]	2.9E4	1999	2.2	2.4E3
1966	185	2.0E5	1983	26[b]	2.9E4	2000	2.0	2.2E3
1967	185	2.0E5	1984	26[b]	2.9E4	2001	3.0	3.3E3
1968	185	2.0E5	1985	7.4	8.1E3			

a. Assume intake of 3 L of water per day; 1,100 L ingested per year.
b. Result was extrapolated from available annual environmental reports from other years.

No data were available for drinking water at Site 300; this TBD assumed that the concentration of tritium was the same as the source of drinking water at LLNL. Table 4-6 lists estimates for Site 300; the data were identical to the concentrations listed in Table 4-5.

Table 4-6. Site 300 maximum annual intakes (Bq/yr) via drinking water.[a]

Year	Conc (Bq/l)	Intake[b] (Bq/yr)	Year	Conc (Bq/l)	Intake[b] (Bq/yr)	Year	Conc (Bq/l)	Intake[b] (Bq/yr)
1955	100[b]	1.1E5	1971	100[b]	1.1E5	1987	3.7	4.1E3
1956	100[b]	1.1E5	1972	100[b]	1.1E5	1988	6.6	7.2E3
1957	100[b]	1.1E5	1973	100[b]	1.1E5	1989	7[b]	7.7E3
1958	100[b]	1.1E5	1974	100[b]	1.1E5	1990	7[b]	7.7E3
1959	100[b]	1.1E5	1975	100[b]	1.1E5	1991	4.5	5.0E3
1960	100[b]	1.1E5	1976	26	2.9E4	1992	4.4	4.8E3
1961	100[b]	1.1E5	1977	26[b]	2.9E4	1993	1.0	1.1E3
1962	100[b]	1.1E5	1978	26[b]	2.9E4	1994	2.8	3.1E3
1963	100[b]	1.1E5	1979	11	1.2E3	1995	0.8	8.8E2
1964	100[b]	1.1E5	1980	26[b]	2.9E4	1996	2.1	2.3E3
1965	100[b]	1.1E5	1981	26[b]	2.9E4	1997	2.4	2.6E3
1966	185	2.0E5	1982	26[b]	2.9E4	1998	1.2	1.3E3
1967	185	2.0E5	1983	26[b]	2.9E4	1999	2.2	2.4E3
1968	185	2.0E5	1984	26[b]	2.9E4	2000	2.0	2.2E3
1969	185	2.0E5	1985	7.4	8.1E3	2001	3.0	3.3E3
1970	200	2.2E5	1986	7.4	8.1E3			

a. These data were provided from LLNL site data. No data regarding radioactivity in drinking water were available specifically for Site 300.
b. Assume intake of 3 liters of water per day; 1,100 liters ingested per year.

4.4 AMBIENT RADIATION

From 1976 to the present, thermoluminescent dosimeters (TLDs) were used in determining ambient 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 Livermore site perimeter and Site 300, respectively. The data in Tables 4-7 and 4-8 were summarized from 1989 to 2001 in the annual environmental reports (see the reference list). Observations reported in earlier environmental reports, based on surveys with portable instruments, state "0.02 mrem/h at all locations." The instrumentation might have been primitive by today's standards and did not provide continuous monitoring, so these results were overshadowed by recent more reliable observations. The results were reported in the annual environmental reports as cumulative exposures in millirem per year based on 8,760 hr; Tables 4-7 and 4-8 reflect the data provided by LLNL.

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, as shown in Figure 4-6. Assuming a deep dose equivalent rate of 100 mrem/yr for all years would be claimant-favorable and reasonable, and deficiencies in earlier measurement techniques would become immaterial.

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 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 literature values for State 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.

4.5 UNCERTAINTY

As discussed in the previous sections, estimates of annual intakes employed conservative (i.e., claimant-favorable) assumptions when information was unavailable. For example, applying maximum annual exposures to all years was a claimant-favorable assumption, but not overly conservative. There was a lack of particle size and solubility. As a consequence, this TBD assumed that all effluents were in the respirable size range and selected solubility based on the highest radiation dose rate. Effluent data collected in 1976 and later have an estimated uncertainty of ±20%. Data from earlier years were less accurate because of less reliable measurement technology and less stringent reporting requirements.

The location of the monitoring points from which data were summarized in this section adds 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

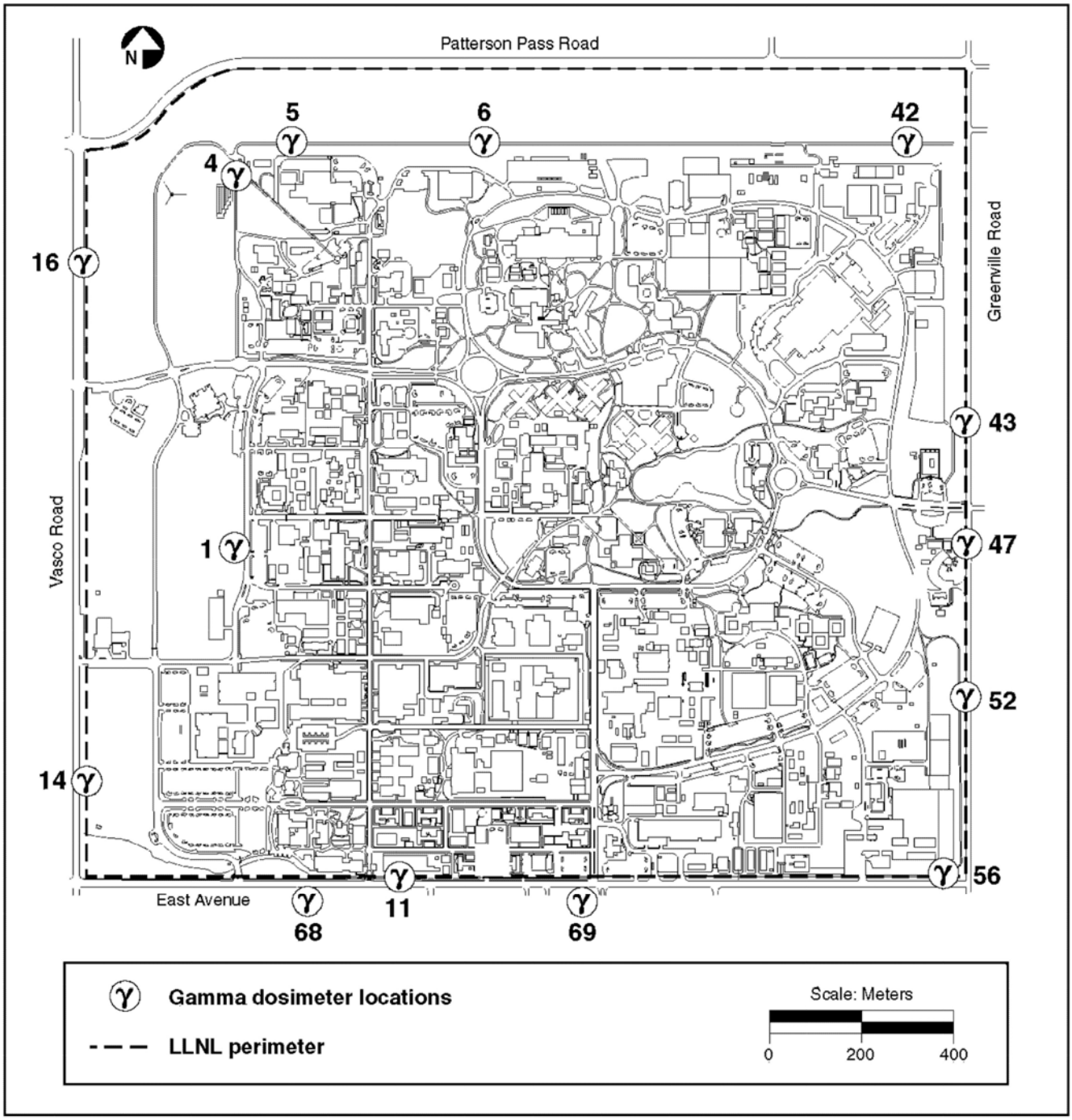

Figure 4-4. Gamma dosimeter locations, Livermore Site, 2001 (Source: LLNL 2002)

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. All external environmental dose data were adjusted to reflect a 2,000-hr work-year. The data were originally reported in site environmental reports as representative of an employee who worked at the site 24 hr a day, 365 days a year. Using a permanently located

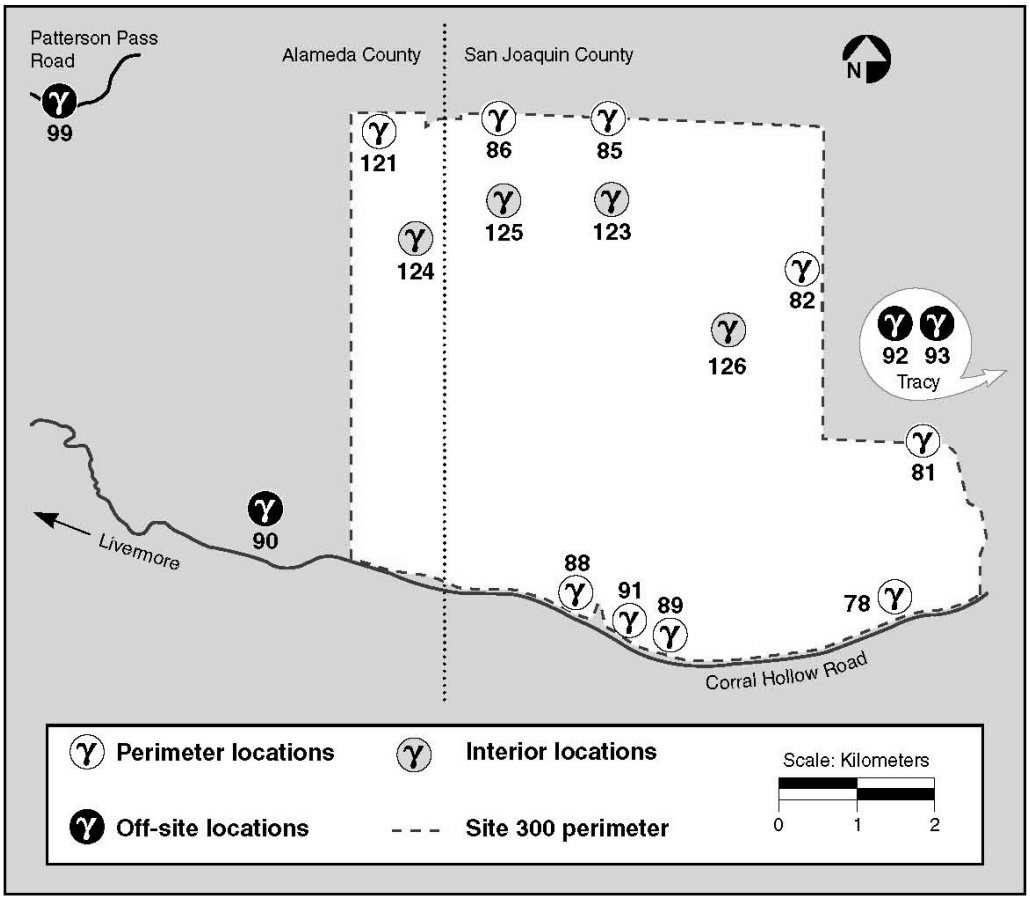

Figure 4-5. Gamma dosimeter locations, Site 300 and vicinity, 2001 (Source: LLNL 2002).

employee at the site, however, was an unrealistic assumption that would overstate onsite environmental exposures.

Uncertainty related to internal exposures presents similar concerns. The highest internal exposures occurred during the early years, and lessened over time with increased controls and better equipment. Assumption of the maximum uptake for all years reduces the need to include an uncertainty factor for intakes.

Table 4-7. External gamma radiation (mrem/yr).

Year	Site average	Error	Maximum	Year	Site average	Error	Maximum
1952	88[a]	10[a]	100[a]	1977	88[a]	10[a]	100[a]
1953	88[a]	10[a]	100[a]	1978	88[a]	10[a]	100[a]
1954	88[a]	10[a]	100[a]	1979	61	3	96
1955	88[a]	10[a]	100[a]	1980	88[a]	10[a]	100[a]
1956	88[a]	10[a]	100[a]	1981	88[a]	10[a]	100[a]
1957	88[a]	10[a]	100[a]	1982	88[a]	10[a]	100[a]
1958	88[a]	10[a]	100[a]	1983	88[a]	10[a]	100[a]
1959	50	10	100[a]	1984	88[a]	10[a]	100[a]
1960	88[a]	10[a]	100[a]	1985	59	6	80
1961	88	10	100[a]	1986	61	5	69
1962	44	10	100[a]	1987	65	7	71
1963	50	10	100[a]	1988	62	7	69
1964	50	10	100[a]	1989	88[a]	10[a]	100[a]
1965	50	10	100[a]	1990	88[a]	10[a]	100[a]
1966	88[a]	10[a]	100[a]	1991	65	6	78
1967	20	5	100[a]	1992	66	5	76
1968	20	5	100[a]	1993	65	5	74
1969	20	5	100[a]	1994	72	9	79
1970	20	10	100[a]	1995	56	8	67
1971	52	5	90	1996	55	4	60
1972	88[a]	10[a]	100[a]	1997	60	1	64
1973	88[a]	10[a]	100[a]	1998	60	2	66
1974	88[a]	10[a]	100[a]	1999	58	3	63
1975	88[a]	10[a]	100[a]	2000	57	2	65
1976	84	2	293	2001	56	1	100

a. External dose for these years was not available. The maximum external dose was reported in 1961. Accordingly, the 1961 dose values are used for the years noted.

Table 4-8. Site 300 external gamma radiation (mrem/yr).

Year	Site average[a,b,c]	Error [c]	Maximum [c]
1955	100		
1956	100		
1957	100		
1958	100		
1959	100		
1960	100		
1961	100		
1962	100		
1963	100		
1964	100		
1965	100		
1966	100		
1967	100		
1968	100		
1969	100		
1970	100		
1971	100		
1972	100		
1973	100		
1974	100		
1975	100		
1976	100		
1977	100		
1978	100		
1979	100		
1980	100		
1981	100		
1982	100		
1983	100		
1984	100		
1985	100		
1986	100		
1987	100		
1988	100		
1989	78	7[a]	89[a]
1990	78	7[a]	89[a]
1991	78	7	89
1992	77	5	87
1993	76	6	85
1994	64	1	74
1995	64	1	74
1996	66	2	72
1997	72	4	77
1998	72	4	78
1999	71	1	80
2000	64	1	70
2001	63	1	66

a. External dose for these years was not available. The maximum external dose was reported in 1991. Accordingly, the 1991 dose values are used for the years noted.
b. Environmental radiation monitoring at Site 300 began on July 1, 1988.
c. Site average (mrem/yr) is based on 10% above maximum value from later years where monitoring was performed. Therefore, no maximum or error is provided.

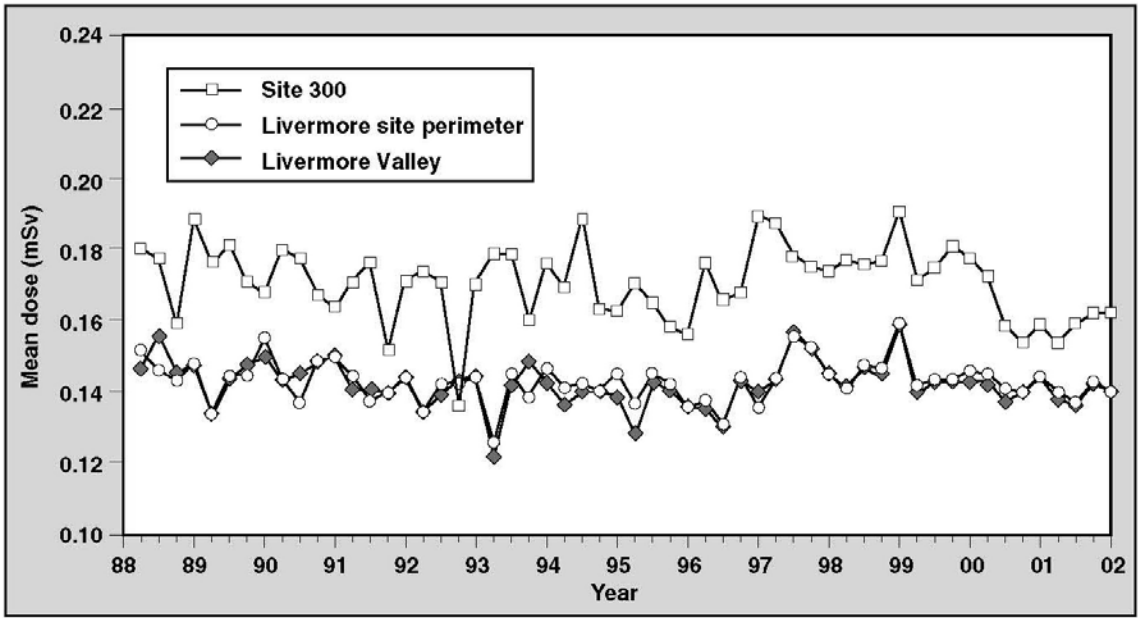

Figure 4-6. Quarterly mean gamma dose measurements at the Livermore site perimeter, 1988-2001 (Source: LLNL 2002)

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GLOSSARY

alpha radiation
Radiation consisting of charged particles identical with the isotope ^{4}He.

background radiation
Radiation received that is not associated with a worker's occupation. This includes cosmic and terrestrial sources.

becquerel (Bq)
The derived SI unit of radioactivity equal to one disintegration per second.

beta radiation
Radiation consisting of electrons emitted spontaneously from the nuclei of certain radioactive elements.

deep dose equivalent
Dose equivalent at a depth of 1.0 cm in soft tissue.

dosimetry
The science of assessing absorbed dose, dose equivalent, effective dose equivalent, etc., from external and/or internal sources of radiation.

radioactivity
The spontaneous emission of radiation, generally alpha or beta particles, gamma rays, and neutrons from unstable nuclei.

rem
A unit of dose equivalent.

thermoluminescent dosimeter
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.