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Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL). Part 3. Routine Releases, 1973 - 2005

S-R. Peterson

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ABSTRACT

Annual mean concentrations of tritium in air moisture, calculated from data obtained from an air tritium sampler near the LLNL Discovery Center, were compared with annual mean air moisture concentrations predicted from atmospheric releases of tritium for the years 1973 through 2005. The 95% confidence intervals on the predictions and observations usually overlapped. When the distributions of predictions and observations were different, predictions were higher. Using both the observed and predicted air concentrations as input to the tritium dose model, DCART, annual doses to a hypothetical adult, child (age 10) and infant (age 6 months to 1 year) assumed to be living at LLNL's Discovery Center were calculated. Although the doses based on predicted air concentrations tended to be higher, they were nevertheless indistinguishable from doses based on observed air concentrations when uncertainties were taken into account. Annual doses, calculated by DCART and based on observed and predicted air concentrations, were compared with historical tritium doses reported annually by LLNL. Although the historical doses were calculated using various assumptions over the years, their agreement with the DCART predictions is remarkable. The Discovery Center was not the location of the site-wide maximally exposed individual (SW-MEI) from 1974 through 1978. However, doses at the location of the SW-MEI for those years were indistinguishable from those at the Discovery Center when uncertainties were taken into account. The upper confidence limits for all doses were always well below the current regulatory limit for dose to a member of the public (100 μSv or 10 mrem per year) from atmospheric releases (40 CFR Part 61, Subpart H). Based on observed air concentrations, the 97.5% confidence limit on the cumulative dose to the hypothetical person born in 1973 and living through 2005 at the Discovery Center was 150 μSv (15 mrem), while that of the hypothetical adult who spent his entire life at the Discovery Center was 100 μSv (10 mrem). Comparable doses calculated from predicted air concentrations were about 35% greater. The comparison of predicted and observed air concentrations demonstrate that some reasonable confidence can be placed in the concentrations that will be predicted from tritium releases to the atmosphere in Part IV (1953 – 1972) of the Tritium Dose Reconstruction; because dose estimations are linearly related to air concentrations, reasonable confidence can also be placed in the doses that will be predicted.

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INTRODUCTION

This Tritium Dose Reconstruction (TDR) has been undertaken to account for dose to the public from tritium releases to the atmosphere from facilities at the Livermore site of Lawrence Livermore National Laboratory (LLNL) over its lifetime; the probability that the predicted doses could not have been exceeded is greater than 97.5%. This report, which is Part 3 of "Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Releases to the Atmosphere from Lawrence Livermore National Laboratory (LLNL)[1]" presents radiological doses to the public from routine releases for 1973 through 2005[2]. For all of these years, except 1974 through 1978, the location of the site-wide maximally exposed individual (SW-MEI[3]) was essentially at the Discovery Center (formerly Visitors Center) LLNL. Doses were predicted using annual mean air tritium concentrations measured at the Discovery Center (see Location VIS, Figure 1) as input to the tritium dose model, DCART (Peterson 2006). Air concentrations at the Discovery Center were also calculated from annual release rates and dilution factors[4] (see Part 2 of the TDR), and dose predictions were calculated using these air concentrations in DCART. From 1974 through 1978, doses were similarly calculated at the location of the SW-MEI to the south of the Laboratory (Location Q, Figure 1).

Tritium has been released as part of normal operations since operations began at LLNL. Between 1973 and 2005, an estimated 60,700 Ci (2,250 TBq) of tritium (either as tritiated water vapor (HTO[5]) or tritiated hydrogen gas (HT[6]) were released to the atmosphere from routine operations at LLNL (Table 1a). This estimate, based on data compiled in Part 2 of the TDR, has a 2.5% confidence limit of 56,700 Ci (2,100 TBq) and a 97.5% confidence limit of 64,700 Ci (2,400 TBq). The quantity of tritium released routinely between 1973 and 2005 is less than 10% of the total estimated amount of tritium released from the Livermore site (including accidental releases) over LLNL's lifetime. Tritium was also released from the Tritium Research Laboratory (TRL) of neighboring Sandia National Laboratories/California (SNL/CA) between 1979 and 1995. An estimated 6,120 Ci (227 TBq) were released routinely from the TRL, with a 2.5% confidence limit of

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[1] TDR Part 1. Description of Tritium Dose Model (DCART) for Chronic Releases from LLNL (Peterson 2006).
TDR Part 2. LLNL Annual Site-Specific Data 1953 – 2005 (Peterson 2007)
TDR Part 4. Routine Releases, 1953 – 1972. In draft.
TDR Part 5. Accidental Releases. In draft.
TDR Part 6. Summary. In draft.
TDR Part 7. Tritium Dose Reconstruction using Organically Bound Tritium in Wood of Trees. Proposed

[2] Routine ambient air sampling for tritium began in May 1973, and the first complete year of sampling was 1974. Because samples taken for the last eight months of the year at LLNL are usually representative of the mean for an entire year, 1973 has been included in this part of the TDR.

[3] A hypothetical member of the public who receives the greatest LLNL-induced effective dose equivalent (summed over all pathways) from all sources of radionuclides released at a site.

[4] The term "dilution factor" will be used throughout this report to refer to the air concentration for unit source strength (or χ/Q); units are actually Bq m^{-3} / Bq s^{-1} or Ci m^{-3} / Ci s^{-1}. The term, although standard for χ/Q, can be misleading because the higher the dilution factor, the higher the air concentration.

[5] Throughout the report, aqueous tritium in any form (e.g., T_2O, DTO and HTO) will be referred to as HTO.

[6] Throughout the report, gaseous tritium in any form (e.g., T_2, DT, and HT) will be referred to as HT.

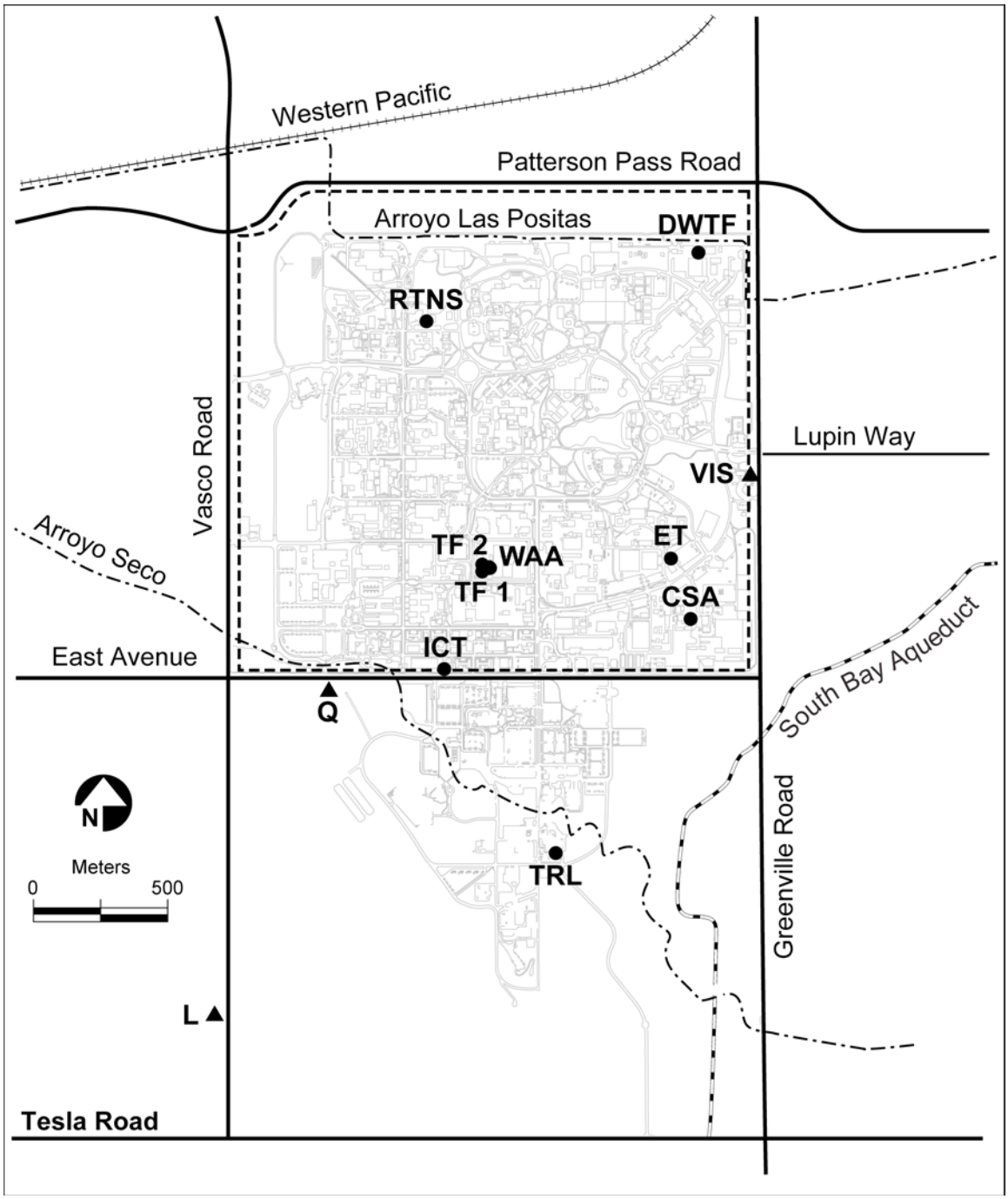

Figure 1. Sources of tritium (•) at LLNL and SNL/CA relative to the location of the potential site-wide maximally exposed individual (▲). ▲VIS is the Discovery Center; Q was an automotive garage. Modeled sources include TF1 and TF2 (the south and north stacks of the Tritium Facility), WAA diffuse source at Building 331, ICT (Building 212), RTNS (Building 292). CSA (Building 612 Yard and the Building 624 incinerator) ET (evaporation trays), TRL (Tritium Research Laboratory at SNL/CA), and DWTF (Decontamination and Waste Treatment Facility).

5,440 Ci (201 TBq) and a 97.5% confidence limit of 6,810 Ci (252 TBq); about 74% of the total tritium released during normal operations was HTO (Table 1b). During the same period of time (i.e., 1979 – 1995), LLNL released about 33,000 Ci (1,200 TBq) from normal operations; 90% of this was released from the Tritium Facility (Building 331).

Table 1a. Quantities of HT and HTO, in curies, released routinely from the Livermore site between 1973 and 2005. CL is the confidence limit on the uncertainty.

Type of release	Best estimate	2.5% CL	97.5% CL
HT	29,900	26,600	35,200
HTO	30,600	28,600	32,800
Sources of Routine Releases			
HT from Tritium Facility	23,200	21,200	25,300
HTO from Tritium Facility	25,900	23,900	28,000
HT from other stacks	6,620	4,130	9,420
HTO from other stacks	2,200	1,700	2,730
HTO from area sources	2,450	2,060	2,840

Note: An additional 6,200 Ci of HT were released accidentally.

Table 1b. Total HT or HTO in curies released routinely from the Tritium Research Laboratory of Sandia National Laboratories/CA between 1979 and 1995. CL is the confidence limit on the uncertainty.

Type of release	Best estimate	2.5% CL	97.5% CL
HT routine	1,580	1,400	1,780
HTO routine	4,540	3,880	5,200

Note: An additional 200 Ci of HTO and 1100 Ci of HT were released accidentally.

In addition to the routine annual releases of tritium discussed in this report, between 1973 and 2005 LLNL and SNL each had two releases that will be modeled as accidental. LLNL released 5,200 Ci of HT on June 8, 1984 and 1,000 Ci of HT on January 24, 1985; SNL released 200 Ci of HTO on one Saturday in January 1986 and 1,100 Ci of HT on August 18, 1987. These accidental releases and their estimated dose impacts are being included in Part 5 of the TDR.

Release rates of HT and HTO from normal operations at the Livermore site and SNL/CA for 1973 through 2005 are summarized in Table 2. A detailed accounting of release rates from each facility, including estimated uncertainty, may be found in Part 2 of the TDR; the tabular summaries of the data are repeated in Appendix A, Tables A1 – A7, of this document.

Table 2. Estimated annual release rates of HT and HTO (Ci) from stack and area sources.

	HTO from stacks					HTO from area sources			HT from stacks		
	B331	B212	B624 Incinerator	B292	SNL/CA or DWTF*	B331 WAA	B612 Yard	Evaporation Trays	B331	B212	SNL/CA or DWTF*
1973	1380	45.0				63.3	46.4	~4.30	1180	330	
1974	657	66.8				43.2	19.8	~4.30	1090	493	
1975	1120	145				58.7	36.7	~4.30	1260	1070	
1976	1810	140				75.8	62.2	~4.30	1250	1030	
1977	2120	246	0.80			83.8	77.6		1270	1810	
1978	2130	133	0.80			110	78.3		2310	984	
1979	1630	52.0	0.80	18.7	4.64	101	59.7		2450	384	1.08
1980	1420	8.35	0.80	27.7	20.5	54.3	52.0		779	61.6	4.77
1981	1530	2.79	0.80	70.7	34.8	63.5	56.2		1040	20.6	8.11
1982	1310	4.06	0.80	89.6	164	47.3	47.9		605	29.9	38.1
1983	1210	16.7	0.80	130	74.2	73.6	44.4		1770	123	21.2
1984	1040	1.31	0.80	229	146	47.7	38.2		887[7]	9.69	18.5
1985	652	0.600	0.80	336	389	24.5	23.6		338[8]	4.40	128
1986	630	2.00	4.43	178	415[9]	26.9	23.1		457	13.0	129
1987	1250	4.00	0.216	126	573	65.3	45.8		1390	34.0	159[10]
1988	1670		0.343	8.46	1040	98.3	61.2		2310		543
1989	1620			3.84	659	73.4	59.4		1350		180
1990	694				244	31.1	35.5		562		50.8
1991	696				352	28.4	25.5		455		113

[7] An additional 5,200 Ci of HT were released accidentally.
[8] An additional 1,000 Ci of HT were released accidentally.
[9] An additional 200 Ci of HTO were released accidentally.
[10] An additional 1,100 Ci of HT were released accidentally.

Table 2 continued

	HTO from stacks					HTO from area sources			HT from stacks		
	B331	B212	B624 Incinerator	B292	SNL/CA or DWTF*	B331 WAA	B612 Yard	Evaporation Trays	B331	B212	SNL/CA or DWTF*
1992	104				134	4.42	3.79		79.6		130
1993	114				132	9.22	4.24		124		55.3
1994	76.1				91.2	3.01	1.92		60.9		4.10
1995	62.8				73.0	31.4	4.17		29.0		1.06
1996	181					10.1	12.4		33.5		
1997	267					8.04	7.19		31.9		
1998	84.5					14.4	8.47		25.0		
1999	213					15.8	7.61		66.8		
2000	35.2					2.32	5.47		4.91		
2001	18.3					0.679	2.27		1.71		
2002	32.8					0.755	2.33		3.48		
2003	103					8.70	3.43		6.17		
2004	12.1				19.0*	0.695	3.18		4.42		1.0*
2005	30.2				2.62*	4.76	1.48		1.57		0.107*

Since May 1973, LLNL has been continuously monitoring concentrations of HTO in air (per m^3) and air moisture (per L) at the sampling location, VIS, near the Discovery Center on the eastern perimeter of the Livermore Site. Location VIS is within 200 m of the UNCLE Credit Union[11], which, based on dispersion modeling, has served since 1992 as the location of the SW-MEI for compliance with 40 CFR Part 61, Subpart H (National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities – NESHAPs) (US EPA 1989). Given, in addition, that about 54% of the time winds blow from the south-southwest through west directions and show very little change from year to year, and given that CAP88-PC (Parks 1992), the regulatory dispersion and dose model used for NESHAPs compliance, has been shown not to underestimate air concentrations at VIS (Peterson 2003, 2004), the Discovery Center (VIS location) was a reasonable choice for the hypothetical residence of the hypothetical individuals (adult, child [age 10] and infant [age 6 months to 1 year]) in this TDR[12]. Furthermore, from about the time of its dedication (July 31, 1976), the public had access to this location[13] (then called the LLNL Visitors Center).

As long as the LLNL Tritium Facility dominates releases and no other major sources exist, the SW-MEI will be in the vicinity of the UNCLE Credit Union or the Discovery Center[14]. However, because Building 212 was an important source of HT between 1974 and 1978, because releases from SNL/CA between 1986 and 1989 were nearly as great as those from the Tritium Facility, and because HTO was released from Building 292 between 1981 and 1987 (Table 2), alternate potential locations for the SW-MEI were investigated. Using a method described in EMP-R-DA, the location of the SW-MEI was determined for 1975, 1985, and 1992, years when the Tritium Facility and associated activities did not dominate releases. In 1985 and 1992, no location other than the Discovery Center would have had higher doses. In 1975, however, the results showed clearly that three privately-owned buildings south of the Laboratory to the west of the SNL/CA property would have received slightly higher doses than those at the Discovery Center. Dilution factors were calculated for the three locations as they had been for the Discovery Center[15]. Air concentrations and dose at those three locations were calculated for 1973 to 1980. For the years 1974 through 1978, the highest dose of any location was

[11] An air tritium monitor (CRED) was placed near the UNCLE Credit Union in July 2003. The VIS and CRED air samplers are 150 m apart.

[12] Air concentrations were calculated at VIS because of the long, continuous history of measured tritium concentrations at that location.

[13] In keeping with LLNL's practice, the SW-MEI need not be a residence. If a building to which the public has access for even a few hours each day is closer to the site perimeter than any residence, it serves conservatively as the location of the SW-MEI. For consistency (and conservatism) in the TDR, the Discovery Center was assumed the location of the SW-MEI even before the Discovery Center opened.

[14] Because the UNCLE Credit Union is slightly closer in the same direction to the Tritium Facility, predicted air concentrations are higher than those at the Discovery Center. Measured air concentrations at VIS and CRED, however, are statistically indistinguishable based on counting errors alone, although the long-term mean air concentration at CRED is 17% higher than at VIS.

[15] By using CAP88-PC with a site-specific wind file for 2000 – 2003 that accounts for mean annual wind speeds and stability classes in each of 16 sectors, and distance and direction from source to receptor.

calculated at Location Q, which was an automotive garage, not a residence. Dilution factors for Location Q are shown in Table 3.

Table 3. Directions, distances, and dilution factors from each facility to Location Q.

Facility	Direction	Distance in m	χ/Q (s m^{-3})
B212	WSW/W	427	$1.349 \times 10^{-5} \pm 4.046 \times 10^{-6}$
B331 south stack	SW	707	$7.417 \times 10^{-7} \pm 2.225 \times 10^{-7}$
B331 north stack	SW	732	$5.259 \times 10^{-7} \pm 1.887 \times 10^{-7}$
B624 incinerator	W/WSW	1320	$1.899 \times 10^{-6} \pm 6.647 \times 10^{-7}$
B331 area	SW	744	$9.314 \times 10^{-6} \pm 2.794 \times 10^{-6}$
B612 area	WSW/W	1370	$5.399 \times 10^{-6} \pm 1.620 \times 10^{-6}$
Evaporation Trays	WSW	1330	$4.797 \times 10^{-6} \pm 2.399 \times 10^{-6}$

Although DCART is probabilistic[16] and all parameters are assigned distributed values, the extremely conservative assumption was made that the hypothetical SW-MEI lived at the location 24 hours a day, 365 days a year. To further assure that the calculated dose consequences were conservative, all food (i.e., all vegetables, animal fodder, and animal products) was assumed to have been grown at that location at the same mean annual air concentration measured by the air tritium sampler, even though this was physically impossible. Drinking water for animals was assumed to come from a small (non-existent) pond; drinking water for people was assumed to contain no tritium attributable to LLNL (Moran et al. 2002; Beller et al. 2005; Moran 2005).

In this report, predicted concentrations of tritium in air moisture at location VIS are first compared with annual mean observed concentrations to check that the selected release rates were not underestimated. Secondly, annual doses to adults calculated from annual mean observed air concentrations are compared with annual doses to adults at the Discovery Center calculated from the quantities of tritium released during a year and dispersion modeling. Thirdly, doses at Location Q between 1974 and 1978 are compared with those at the Discovery Center. Fourthly, doses predicted to the SW-MEI, whether at Location Q or Location VIS (i.e., the Discovery Center), are compared with annual doses historically reported by LLNL in the Site Annual Environmental Reports (SAER). Finally, doses to children and infants are compared with those to adults, doses from releases of HT are compared with those from releases of HTO of equivalent magnitude, and other such comparisons are made.

Appendix A contains all input data and associated uncertainties used for this part of the TDR. Most of these data were published in Part 2 of the TDR but are provided here for the convenience of the reader. In Appendix B, predictions of tritium in air moisture from this TDR are compared with previous predictions (Peterson 2003; Peterson 2004), made

[16] Each calculation involved 25,000 runs using Latin Hypercube Sampling in the Crystal Ball 2000® software (Decisioneering, Inc. 1515 Arapahoe Street, Suite 1311, Denver Colorado USA 80202).

with different assumptions, using the dispersion model in CAP88-PC (Parks 1992), and an attempt is made to explain predictions that are greater than observations by more than a factor of two. Appendix C summarizes tritium doses reported in the SAERs (1973 – 2005) and the changes in models and assumptions through time that may help explain differences seen between doses reported in SAERs and doses predicted for this TDR.

PREDICTIONS OF TRITIUM IN AIR MOISTURE

Results

Predicted-to-observed (P/O) ratios of the means of the predicted distributions for tritium concentrations in air moisture and the mean annual observed tritium concentrations in air moisture at VIS for 1973 to 2005 are shown in Figure 2. The annual mean predicted and observed concentrations and their 95% confidence intervals for 1973 to 2005 are compared in Figure 3.

Mean air moisture concentrations were overestimated (P/O > 1) at VIS for all years except 1974 (Figure 2). The upper confidence limits for the predictions barely exceeded the upper confidence limits for the observations for 1986 (Figure 3), however. The high uncertainty about the 1986 observations is due to elevated tritium concentrations in air that were observed between September 1986 and April 1987[17]. Only seven years were over-predicted by more than a factor of 2.0, with the greatest P/O ratio being 2.6 in 1995. Excepting 2004, it was only those years with P/O > 2 (1981, 1989, 1991, 1995, 1998, and 1999) that show no or very little overlap between confidence limits of predictions and observations (Figure 3).

[17] The source of these elevated concentrations was investigated at the time without success (Godwin 1987; Holland and Carlsen 1987).

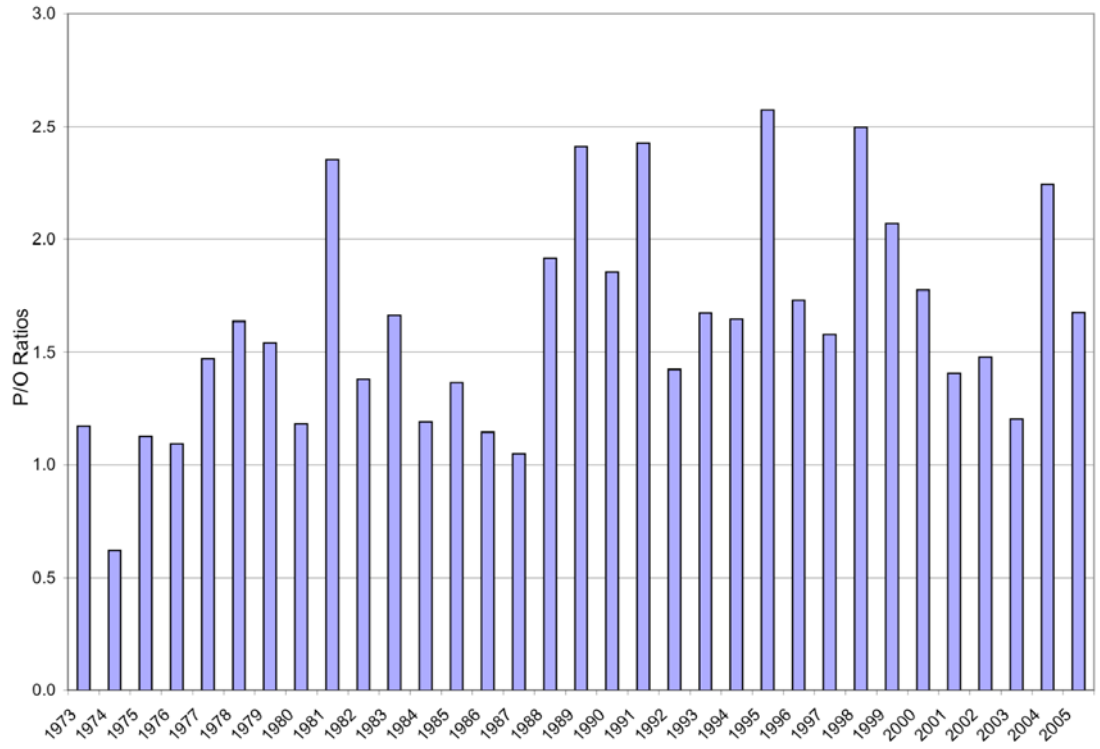

Figure 2. Predicted-to-observed ratios of mean annual tritium concentrations in air moisture at VIS.

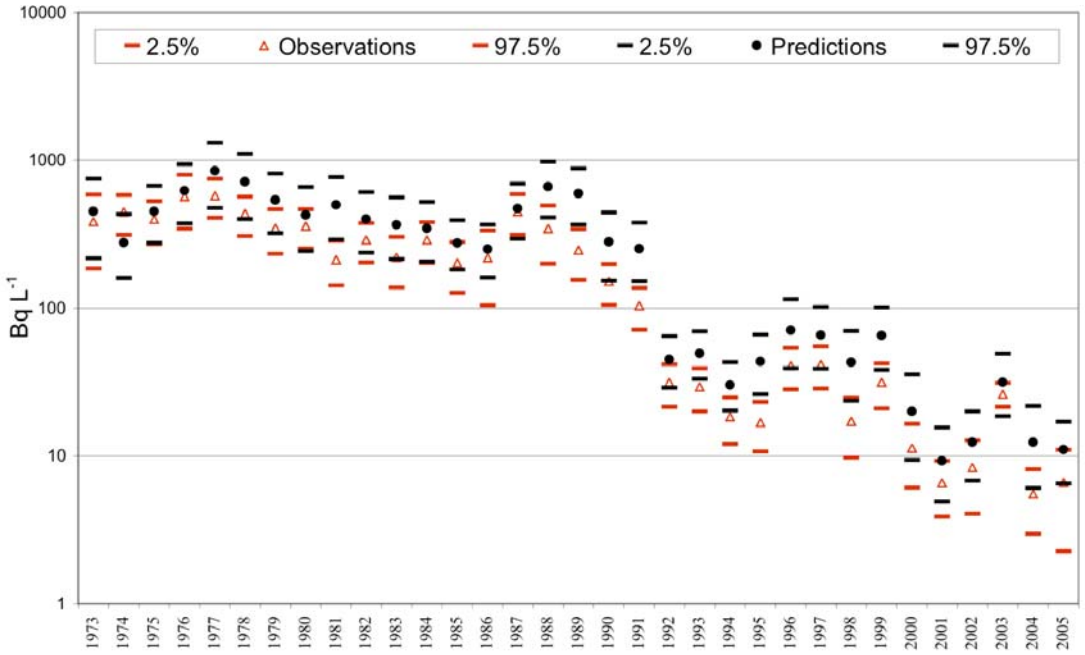

Figure 3. Mean predicted and observed tritium concentrations in air moisture at VIS with 95% confidence intervals on both. Predictions were calculated using CAP88-PC and observed mean annual absolute humidity

The size of the confidence intervals, defined as the upper confidence limit divided by the lower confidence limit, is reasonably small for both observations and predictions (Table 4). The mean interval on the predictions is a factor of 2.7, slightly greater than the factor of 2.3 for the observations; the maximum interval on the observations was a factor of 4.8 in 2005 due to seven of the biweekly samples being below the detection limit; the maximum on the predictions was a factor of 3.8 in 2000.

Table 4. Mean, maximum, and minimum ratios (upper confidence limit value divided by lower confidence limit value) for observed and predicted air concentrations at VIS.

	Observations	**Predictions**
Mean	2.3	2.7
Maximum	4.8	3.8
Minimum	1.4	2.1

Table 5 shows the fraction of total predicted HTO in air moisture from each potential source of tritium for each year at location VIS. Although HT does contribute a small fraction to the HTO measured in ambient air (Figure 4), for the comparison shown in Table 5, it can be omitted as not significantly affecting the relative contributions of the HTO from each facility to the HTO concentrations at VIS. From 1973 through 1994, the LLNL Tritium Facility (Building 331) was the most significant contributor to HTO at VIS. As the release rate dropped from the Tritium Facility after 1994 and the SNL/CA TRL closed, the importance of the relatively small diffuse sources (the Building 331 Waste Accumulation Area[18] [WAA] and the Building 612 Yard) became more important; in the last eleven years, the Building 612 Yard (six times) and the Building 331 WAA (once) were the most significant contributors to tritium in air moisture at VIS.

Data are presented graphically in Figure 5 to demonstrate not only the relative contributions of each facility to the annual concentrations of tritium in air at VIS but also the actual estimated concentrations of tritium in air from each facility over time. Concentrations (Bq m^{-3} HTO) shown in Figure 5 account for the contribution from HT converted to HTO (the fractions in Table 5 do not account for the contribution of HT). The sum of mean predictions from each facility for each year ("total predicted) may be compared with the mean observed tritium concentration in air ("observed"; see Table A10).

[18] A Waste Accumulation Area (WAA) is an officially designated area that meets current environmental standards and guidelines for temporary (less than 90 days) storage of hazardous waste before pickup by the Radioactive and Hazardous Waste Management Division for off-site disposal. The Building 331 WAA was closed in February/March 2003, but tritiated waste continued to be stored at the location of the WAA (radioactive waste does not need to be stored in a WAA) (Chase 2005).

Table 5. Fraction of total HTO predicted at location VIS contributed by each tritium source at LLNL each year. Highest fraction for each year is in bold italics.

	B331	B212	B292	SNL/CA or DWTF*	B624 Incin	B331 WAA	B612 Yard	Evap. Trays
1973	***0.56***	0.040				0.078	0.29	0.031
1974	***0.50***	0.11				0.10	0.23	0.058
1975	***0.49***	0.14				0.080	0.25	0.034
1976	***0.54***	0.090				0.068	0.28	0.022
1977	***0.52***	0.13			< 0.01	0.061	0.29	
1978	***0.54***	0.073			< 0.01	0.084	0.30	
1979	***0.55***	0.038	< 0.01	< 0.01	< 0.01	0.10	0.31	
1980	***0.58***	< 0.01	< 0.01	< 0.01	< 0.01	0.068	0.33	
1981	***0.59***	< 0.01	0.010	< 0.01	< 0.01	0.071	0.32	
1982	***0.56***	< 0.01	0.015	0.041	< 0.01	0.062	0.32	
1983	***0.53***	0.016	0.023	0.019	< 0.01	0.10	0.31	
1984	***0.53***	< 0.01	0.045	0.043	< 0.01	0.074	0.30	
1985	***0.45***	< 0.01	0.089	0.15	< 0.01	0.051	0.25	
1986	***0.44***	< 0.01	0.049	0.17	0.022	0.058	0.25	
1987	***0.49***	< 0.01	0.019	0.13	< 0.01	0.078	0.28	
1988	***0.47***		< 0.01	0.17	< 0.01	0.086	0.27	
1989	***0.52***		< 0.01	0.12		0.071	0.29	
1990	***0.47***			0.096		0.064	0.37	
1991	***0.49***			0.15		0.064	0.29	
1992	***0.41***			0.31		0.053	0.23	
1993	***0.40***			0.27		0.099	0.23	
1994	***0.45***			0.32		0.055	0.18	
1995	0.23			0.16		***0.36***	0.24	
1996	0.44					0.077	***0.48***	
1997	***0.66***					0.062	0.28	
1998	0.32					0.17	***0.51***	
1999	***0.55***					0.13	0.32	
2000	0.28					0.056	***0.67***	
2001	0.33					0.037	***0.63***	
2002	0.46					0.033	***0.51***	
2003	***0.56***					0.15	0.29	
2004	0.17			0.084*		0.031	***0.71***	
2005	***0.44***			0.012*		0.21	0.33	

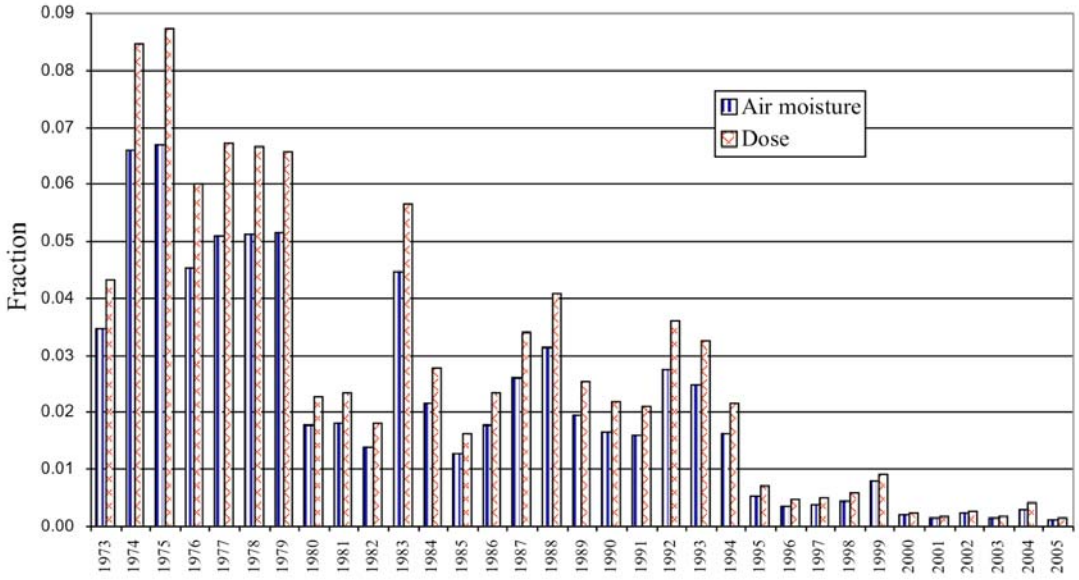

Figure 4. Fractional contribution of HT [HT converted to HTO/total tritium] to air moisture (predicted using CAP88-PC and observed mean annual absolute humidity) and to dose to an adult (predicted using DCART).

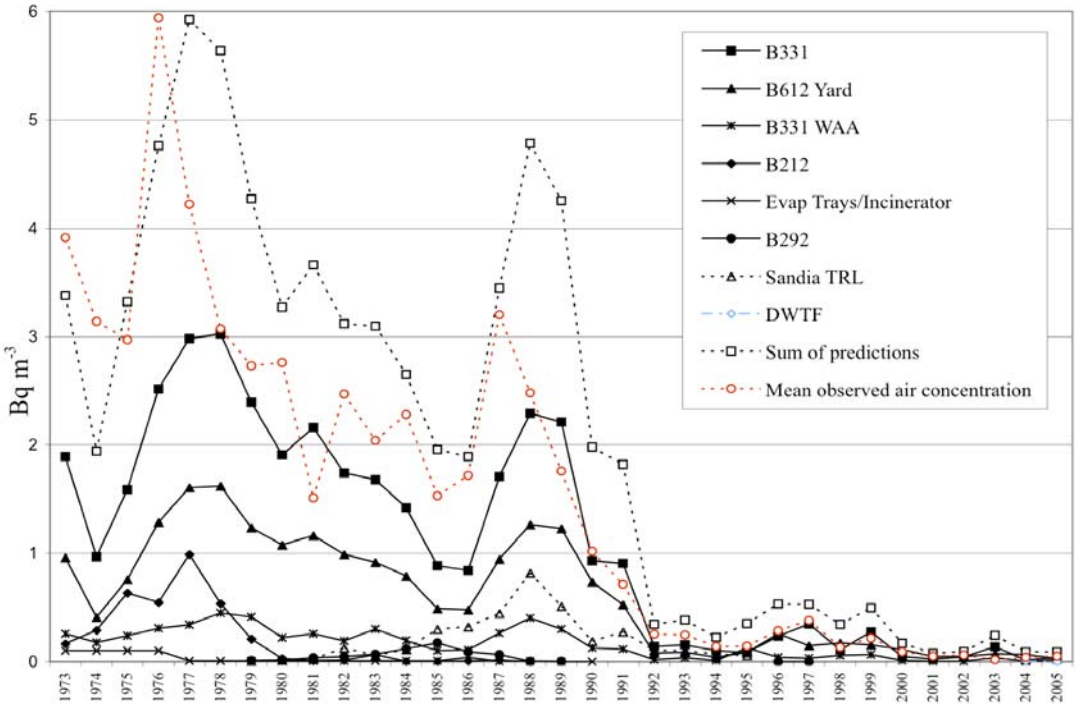

Figure 5. Annual predicted concentrations of tritium in air contributed by the various facilities, the annual sums of the predicted concentrations of tritium in air, and the mean annual observed concentrations of HTO in air.

Discussion

VIS was chosen as the location at which to calculate air moisture concentrations and doses for this TDR because of its proximity to the location of the Livermore site's SW-MEI for NESHAPs compliance, because air tritium sampling has been carried out at VIS since 1973, and because CAP88-PC was known to have a tendency to over-estimate air concentrations there (Peterson 2004) (see Appendix B). Although it may be preferable that predictions of air concentrations be accurate (e.g., P/O = 1 ± 5%), in practice this is a highly unlikely occurrence given the large uncertainty in dispersion calculations[19]. Thus, to err on the side of over-prediction adds a margin of health-protectiveness to a dose reconstruction. At VIS, the long-term mean P/O ratio is 1.6, a comfortable and not excessive degree of over-estimation.

However, although P/O ratios are commonly used to compare results, a P/O ratio greater or less than one implies over- or under-prediction while, in actual fact, when confidence limits of predictions and observations overlap, predictions must be considered, if not equal to the observations, at least not significantly different from the observations. Confidence intervals fail to overlap even by a small margin in 1981, 1989, 1991 and 1995 (Figure 3). The results must thus be considered miss-predicted, although the error in these cases is health-protective. The lower confidence limit on predictions and the upper confidence limit on observations barely overlap for 1998 and 1999; overlap is minimal for 1988, 1993, and 1994. These results obviously tend towards over-prediction. The underestimation in 1974 is small enough that the predictions can be considered to agree with the observations when uncertainties are taken into account. Reasons for over-predictions are examined in Appendix B.

Confidence intervals on the observations (Figure 3) are greater than expected primarily due to the high uncertainty about the factor (1.6) used for all years prior to 2002 to adjust the analytical results to correct for dilution of the sample by "bound water" in the silica gel (Guthrie et al. 2002; Peterson 2007b). The uncertainty in recent years reflects the uncertainty that the annual mean concentration represents the entire year because of the presence of samples below the lower limit of detection.

The concentrations of HTO in air moisture derived from predicted HT concentrations in air by DCART are compared with total predicted HTO concentrations in air moisture (i.e., the sum of the contributions from HTO and HT released to the atmosphere) in Figure 4. The fraction for each year is small, peaking at about 6.6% during 1974 and 1975 when about 65% of the estimated total tritium released from the Livermore site was HT.

[19] When a Gaussian plume model has the proper parameters, annual average air concentrations over flat terrain can only be predicted within a factor of two to four, with accuracy decreasing as complexity of meteorological and terrain conditions increases (Miller and Hively 1987).

The relative contribution of tritium from various facilities to predicted air concentrations at VIS (Table 5 and Figure 5) depends on the magnitude of the release rate, the proximity of the release location to VIS, and whether or not the release was from a stack or an area source. LLNL's Tritium Facility releases were most important at VIS for many years because of their magnitude. SNL/CA's HTO releases, although higher than the Tritium Facility's between 1992 and 1995 (Table 2), were less important at VIS during that time period because of the distance (and direction) of the TRL from VIS. In 2004, the estimated release from the DWTF was slightly larger than that from the Tritium Facility, but the effect on VIS was less because of prevailing winds. Diffuse area sources close to VIS can have an impact out of proportion to the numbers of curies released. For example, in 2000 (Table 2), the release of an estimated 5.5 Ci from the Building 612 Yard contributed 67% of the predicted tritium in air moisture at VIS compared with 28% contributed by the 35 Ci HTO released from the Tritium Facility. Since 1993, as estimated by this TDR, the contributions of the Building 331 WAA and the Building 612 Yard to predicted tritium-in-air concentrations at the Discovery Center have averaged 11% and 41% respectively of the total.

The comparison in Figure 5 of predicted and observed concentrations of tritium in air volume (activity per m^3 of the air passed through the sampler) shows slightly different results compared with predicted-to-observed (Figure 2) and predicted and observed (Figure 3) concentrations of tritium in air moisture (activity per L of water collected from the volume of air passed through the sampler). In Figure 5, which is based on concentration in air volume rather than in air moisture, the predicted tritium concentration is lower than that observed in 1973, 1974, and 1976, while, based on air moisture, only the tritium concentration in 1974 was underestimated. This discrepancy is due to high values for 1973 and 1976 of tritium in air volume (see Table A10); although there should be a strong correlation between mean observed annual concentrations in air moisture and mean observed annual concentrations in air volume, for those years there was not.

The varying parameters that are needed to predict air moisture concentrations at VIS are release rates (Ci y^{-1}), dilution factors (s m^{-3}), and absolute humidity (kg m^3). The parameters to which the annual predicted air moisture concentrations are most sensitive will vary depending upon the relative release rates of tritium from the various facilities and the magnitude of the uncertainty about the release rate. Sensitivity analyses were carried out for 1974, 1980, 1985, 1990, 1994, 1998, and 2000, and the four parameters with the most impact on air moisture concentrations for each year were selected. The predictions were most sensitive to the uncertainty about the release rate from the Building 612 Yard. The parameters that ranked in the top four and the numbers of years out of the seven that they were in the top four are:

- Release rate of HTO from the Building 612 Yard; 7
- Dilution factor for Building 331 North Stack; 6

- Release rate of HTO from the Building 331 North Stack; 5
- Dilution factor for the Building 612 Yard; 5
- Release rate of HTO from the Building 331 WAA; 2
- Release rate of HTO from the Building 331 South Stack; 1
- Dilution factor for the SNL/CA TRL; 1
- Dilution factor for the Building 331 South Stack; 1

The Building 612 release rate ranked as the most important parameter for six out of the seven years because of its proximity to the Discovery Center and the relatively large uncertainty (± 30 - 50%) associated with the release rates. In 1994, air moisture at VIS was most sensitive to the dilution factor for the TRL, because, that year the TRL released more tritium than did the LLNL Tritium Facility. Uncertainty about the release rates of HTO from the Tritium Facility was relatively small (± 20%), but air moisture was nevertheless sensitive to these releases (particularly those from the north stack) because of their magnitude. Conversely, the uncertainty about the release rates from the Building 331 WAA was very large (up to ± 77%), and as a result, the prediction of tritium in air moisture was sensitive to the release rate even though the WAA was distant from the Discovery Center and the release rate was relatively low[20].

DOSE PREDICTIONS FOR ROUTINE RELEASES

Results

Dose Predictions

Dose predictions were made with two different sets of input. One set of calculations (see Figures 6 and 7) used the mean annual observed concentration of HTO in air and associated uncertainty measured at VIS (Table A10); the second set (Figures 8, 9, and 10) used the release rates (Tables A1 – A7) and dilution factors (Tables A8 and A9) and associated uncertainty as input. The predicted doses and 95% confidence intervals at the Discovery Center from calculations using both ambient air and release rates as input are compared in Figure 11.

Doses compared in the figures are to an adult member of the public, which is the normal receptor for regulatory purposes. Doses were two orders of magnitude higher in the early 1970s compared with the early 2000s. All doses were below the present 100 μSv (10 mrem) annual Environmental Protection Agency (EPA) dose limit for compliance with 10 CFR 61 Subpart H for dose from atmospheric releases.

[20] As mentioned[18], the WAA was closed in 2003. However, the name was kept for all years of the TDR to avoid confusion.

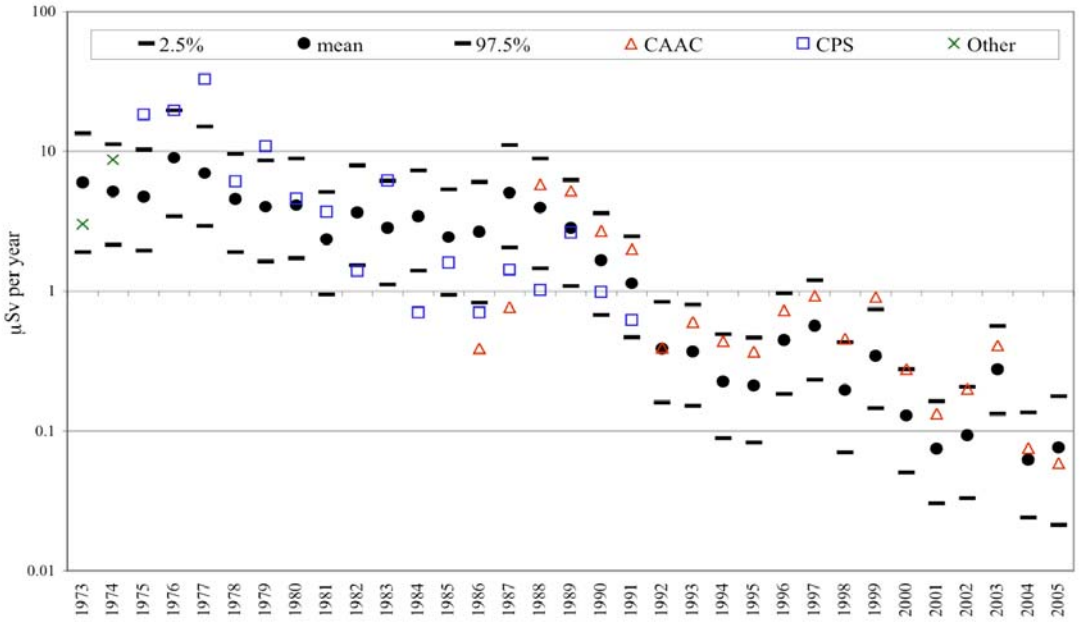

Figure 6. Annual mean doses with 95% confidence intervals predicted by DCART from observed air tritium concentrations at VIS compared with historic reported doses at LLNL calculated using AIRDOS-EPA or CAP88-PC (CAAC or Clean Air Act Code), the Continuous Point Source Code (CPS), and other methods.

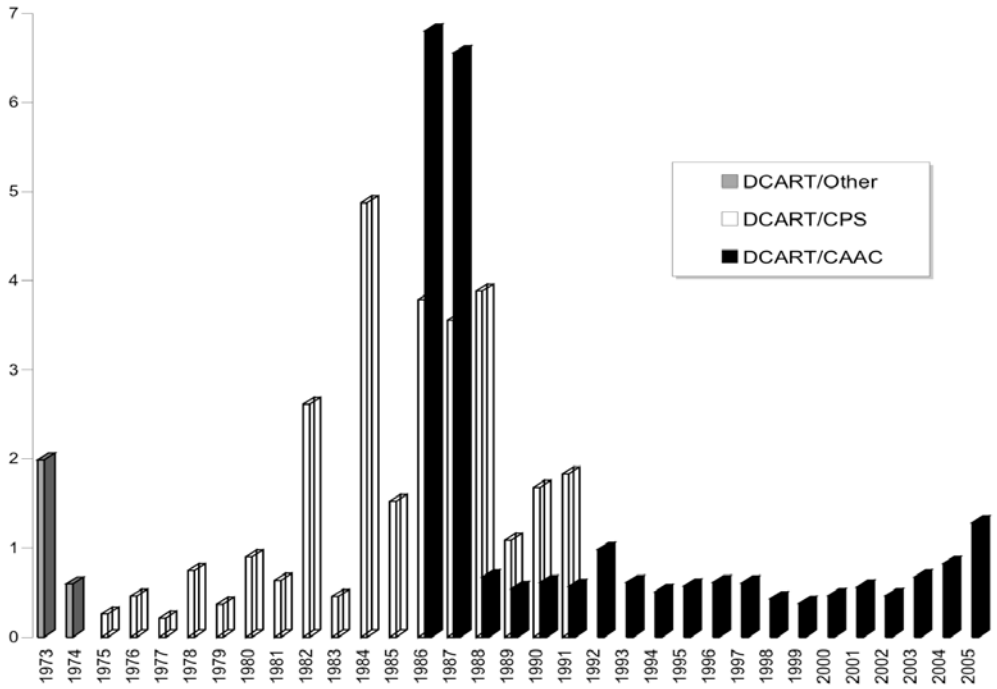

Figure 7. Ratios of annual doses predicted by DCART from mean annual ambient air concentrations at VIS to doses reported by LLNL calculated using AIRDOS-EPA or CAP88-PC (CAAC or Clean Air Act Code), the Continuous Point Source Code (CPS), and other methods. A ratio of 1 is perfect agreement.

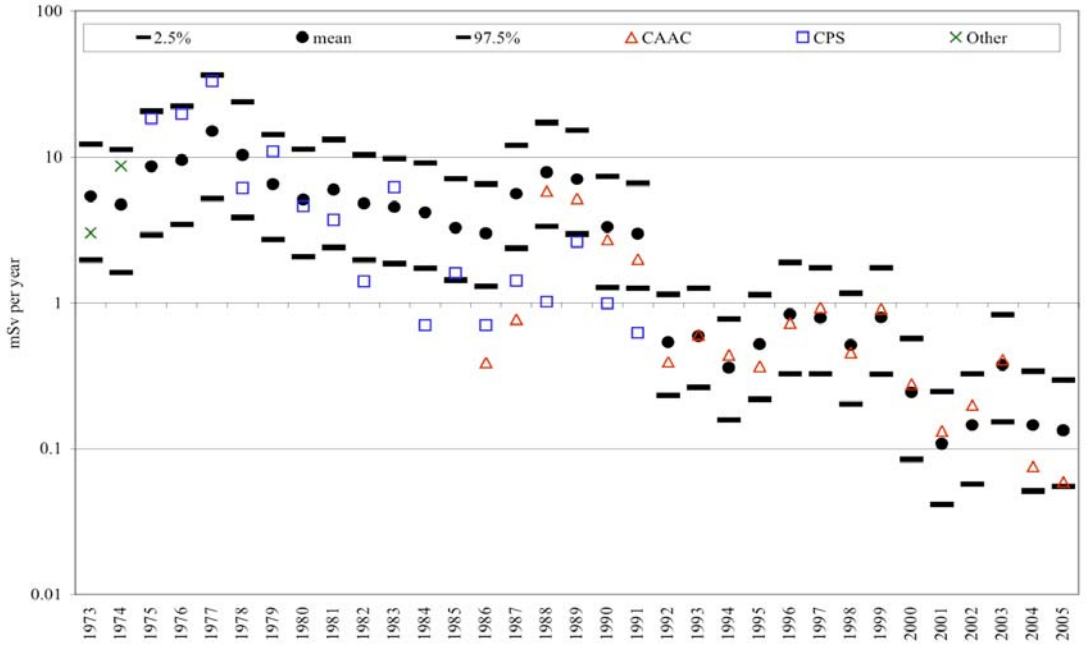

Figure 8. Annual mean doses with 95% confidence intervals, predicted by DCART to the SW-MEI (Location Q, 1974 – 1978; Location VIS, 1973, 1979 – 2005) from release rates and dilution factors, compared with historic tritium doses reported by LLNL. CAAC (Clean Air Act Code) represents AIRDOS-EPA and CAP88-PC; CPS is the Continuous Point Source Code.

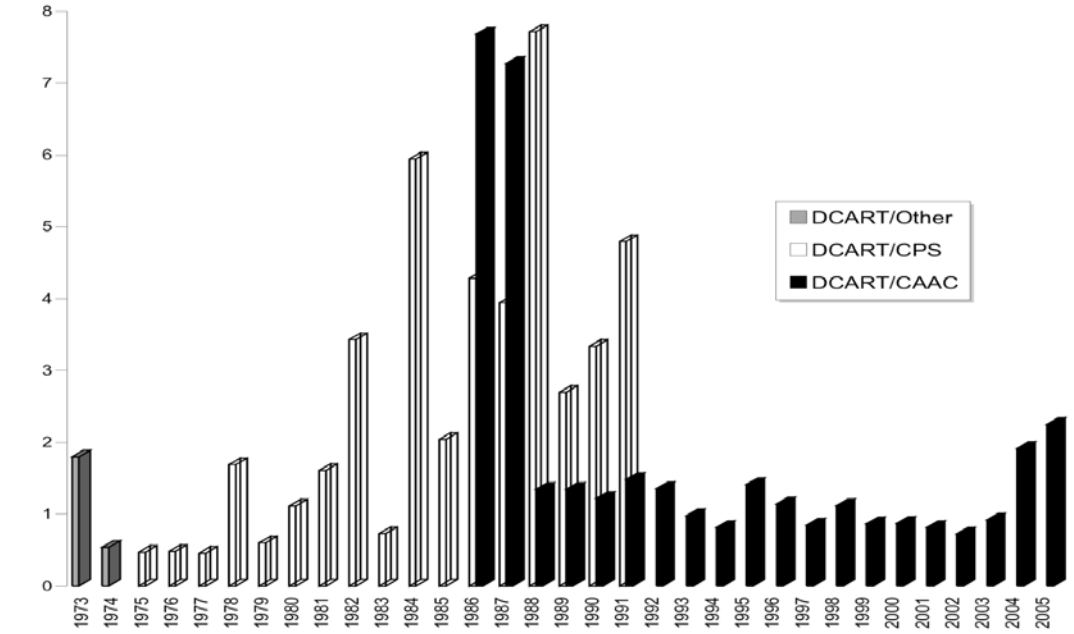

Figure 9. Ratios of annual doses predicted by DCART to the SW-MEI (Location Q, 1974 – 1978; Location VIS, 1973, 1979 – 2005) from release rates and dilution factors to historic tritium doses reported by LLNL. CAAC (Clean Air Act Code) represents AIRDOS-EPA and CAP88-PC; CPS is the Continuous Point Source Code. A ratio of 1 is perfect agreement.

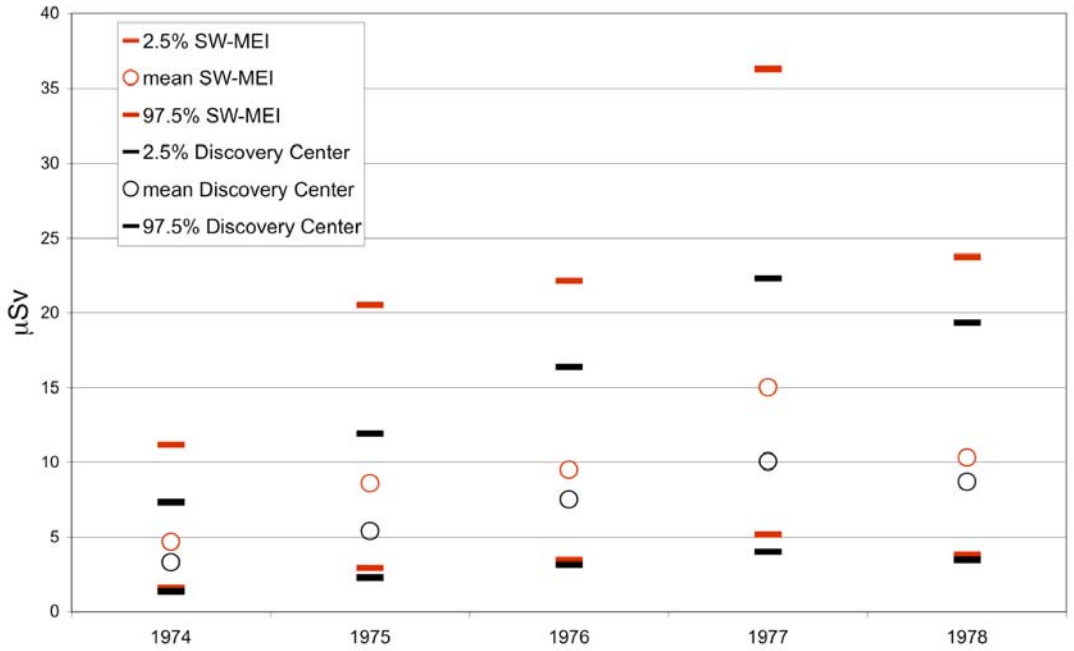

Figure 10. Doses predicted by DCART from release rates and dilution factors, with 95% confidence intervals, to the SW-MEI at Location Q and to the individual at the Discovery Center, 1974 through 1978.

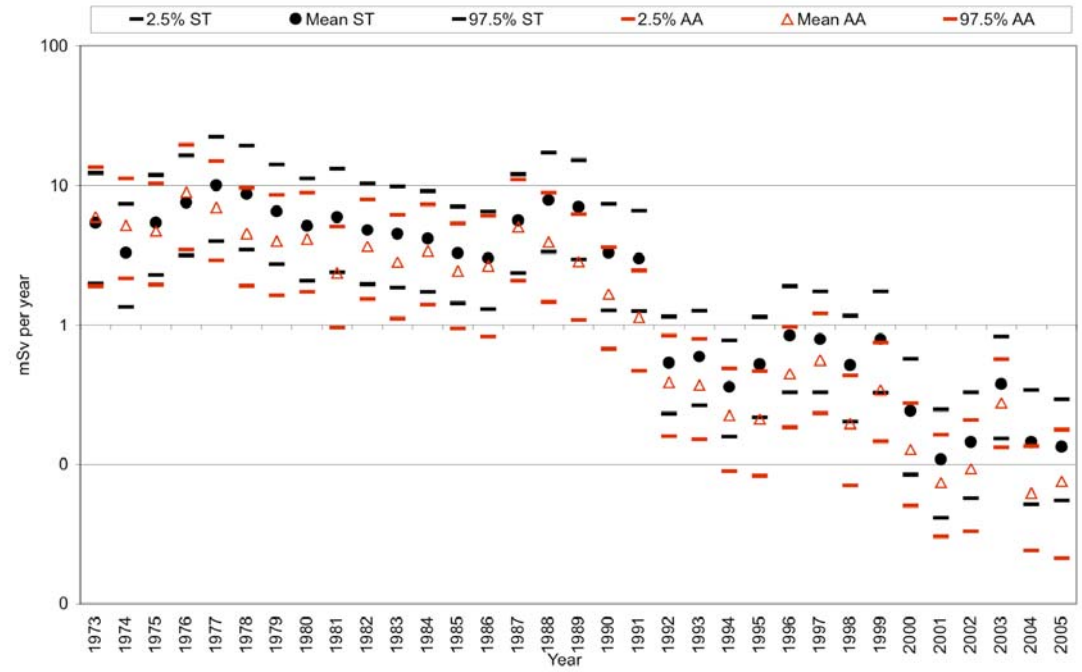

Figure 11. Doses (ST) predicted by DCART at the Discovery Center from release rates and dilution factors compared with doses (AA) predicted by DCART from observed ambient air concentrations; 95% confidence intervals are shown on both.

In Figures 6 and 8, mean doses and their 95% confidence intervals predicted by DCART are compared with tritium doses reported in SAERs. "CAAC" is the Clean Air Act Code (Moore et al. 1979), or AIRDOS-EPA, that became CAP88-PC (Parks 1992), the code used for LLNL's compliance with NESHAPs. "CPS" is the Continuous Point Source Code (Peterson et al. 1976). "Other" refers to the use of an observed air concentration to calculate dose in 1973 and the use of a dispersion equation in 1974 to calculate air concentration. Use of the CPS code and CAAC overlapped between 1986 and 1991. As noted previously, Appendix C summarizes the assumptions and models that LLNL has used to report annual doses.

Dose from observed air concentrations

In Figure 6, annual tritium doses reported in LLNL SAERs are compared with the doses and 95% confidence intervals predicted by DCART using observed mean annual ambient tritium in air concentrations at VIS. Thirteen reported doses fell outside the 95% confidence intervals of the DCART predictions: seven were below the lower confidence limit and six were above the upper confidence limit. The results of the CPS code varied from being higher than the upper confidence limit predicted by DCART to being below the lower confidence limit; the predictions of the CAAC were below the lower confidence limit for the first two years of its use but otherwise they fell within the confidence intervals predicted by DCART except for between 1998 and 2000, when the reported doses were slightly higher than the upper confidence limits predicted by DCART.

The size of the confidence intervals (upper limit value / lower limit value) for the dose predictions averages a factor of 5.6 with a high of 8.3 for 2005 and a low of 4.3 for 2003.

Ratios of the mean doses predicted from tritium concentrations in air to the doses reported between 1973 and 2005 are the same as the results presented in Figure 6 but viewed differently (Figure 7). In 12 out of 38 comparisons, the reported dose was lower than the mean dose predicted by DCART, and in 7 cases, the reported dose was lower by more than a factor of two. However, all results fell within a factor of 5 of each other, with the exception of the CAAC for 1986 and 1987. Between 1988 and 2005, the mean ratio (DCART/reported CAAC dose) was 0.63, with a minimum at 0.38 (1999) and a maximum at 1.3 (2005).

Doses from predicted air concentrations

In Figure 8, doses and the 95% confidence intervals predicted by DCART to the SW-MEI[21] from release rates and dilution factors are compared with tritium doses reported by LLNL in SAERs between 1973 and 2005. Ten reported doses fell outside the 95% confidence intervals predicted by DCART: all were under the lower confidence limits.

[21] The SW-MEI was at Location Q for the years 1974 – 1978; the rest of the time, the SW-MEI was at Location VIS (the Discovery Center).

The agreement between DCART dose predictions and those of CAAC is very good between 1988 and 2003. Agreement over the years with the CPS Code is variable, although in later years the CPS doses consistently fall below the lower confidence limits predicted using DCART.

The size of the confidence interval (upper limit value / lower limit value) for the predictions averages a factor of 5.6 with a high of 7.0 for 1975 and a low of 4.8 for 1993.

Annual mean dose estimates calculated using DCART are higher than those reported by LLNL in 25 out of 39 comparisons (Figure 9); in twelve cases, doses predicted by DCART are higher than those reported by more than a factor of two. Between 1988 and 2003[22], the mean ratio (DCART/reported CAAC dose) was 1.1, with a minimum at 0.73 (2002) and a maximum at 1.5 (1991).

Doses predicted to the adult at Location Q and to the adult at the Discovery Center are compared in Figure 10 for the years when the SW-MEI was at Location Q. Mean annual doses at Location Q are between 20% and 60% higher than doses at the Discovery Center for 1974 through 1978, but the uncertainty on the doses at Location Q is great enough to include the confidence intervals about the dose at the Discovery Center.

Dose methodologies compared

Predictions at the Discovery Center using DCART with the two inputs are compared in Figure 11. There is considerable overlap between the confidence intervals of doses predicted from observed air concentrations and doses predicted from release rates. However, the upper confidence limit for doses predicted from release rates is higher than that from doses predicted from observed air concentrations except for 1973, 1974, and 1976. Only for 1974 can there be concern that the predictions underestimated the observations.[23]

Averaged over thirty-three years, the size of the confidence interval about the predictions based on ambient air (5.6) is slightly greater than that from release rates (5.4). However, the size of the confidence interval on doses predicted from observed air tritium concentrations is more variable than that on doses predicted from release rates: the maximum uncertainty on a dose from observed concentrations is 8.3 compared with 6.8 for doses calculated from the quantity of tritium released annually, while the minimum uncertainty is less for doses from ambient air (4.3) compared with doses calculated from release rates (4.8).

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[22] The 2004 and 2005 comparisons between DCART doses and LLNL reported doses are not consistent with earlier years because of a change to the ingestion assumptions for the dose reported by LLNL (see Appendix C).

[23] The relationship between doses predicted from release rates or from ambient air concentrations should be similar to the relationship between predicted and observed air moisture concentrations (Figure 2), and it is except for 1973 and 1976. For those years (see Part 2 of the TDR), the linear relationship between tritium concentrations in air moisture (Bq L^{-1}) and in air volume (Bq m^{-3}) did not hold – the concentrations in air volume were higher than expected.

Relative Importance of Doses

Adult, child, infant

The mean dose estimated for a child of 10 is 23% higher than that of the adult; the infant's dose is 43% times higher than that of the adult. The uncertainty on the infant dose is greater than the uncertainty on either the adult or child dose, which are about the same. The upper confidence limit for the infant is about 70% higher than that of the adult, while the lower confidence limit is about the same as that of an adult. Thus the upper confidence limit for the infant will be the maximum estimated dose to a member of the public.

The highest dose predicted for an infant (97.5% confidence limit), calculated from the amount of tritium released, was 60.5 μSv (6.05 mrem) in 1977; the mean infant dose for 1977, calculated from the amount of tritium released, was 21.3 μSv (2.13 mrem). The infant dose (97.5% confidence limit) for 1977 obtained from observed air concentrations as input was 26.0 μSv (2.60 mrem).

Cumulative doses for routine releases, 1973 - 2005

Using the Crystal Ball probabilistic software, the 97.5% confidence interval on the sum of the annual doses obtained from observed air concentrations (i.e., the total probable maximum dose that could have been received by a hypothetical individual living at the Discovery Center between 1973 and 2005) was calculated. Doses were estimated for a hypothetical individual born in 1973 who grew up at the Discovery Center and lived there through 2005 and for a hypothetical adult who lived at the Discovery Center for the 33 years. Because DCART only calculates dose to three age groups (infant, child, and adult), it may be assumed conservatively that the infant born in 1973 received an annual dose based on an infant dose coefficient and infant intake until age 10, an annual dose based on a child's dose coefficient and child's intake until age 18, and an annual dose based on an adult's dose coefficient and intake for the next 15 years (the magnitude of the dose coefficient decreases with age, and infant doses are highest even with low intake).

The dose (1973 – 2005) at the 97.5% confidence limit from routine releases to the hypothetical person who grew up at the Discovery Center was 150 μSv (15 mrem) compared with the cumulative dose of (100 μSv [10 mrem]) to the hypothetical adult who spent his entire life at the Discovery Center. These doses are about 35% higher when estimated using predicted rather than observed air concentrations.

Dose contribution of HT

The fraction of dose to an adult due to releases of HT compared with the total tritium dose to an adult is shown for each year in Figure 4. The contribution of HT to dose is greater than the contribution of HT to ambient HTO in air concentrations, but it never exceeds 9% even for those years of high relative releases of HT. Because the effect of HT on dose is dependent upon diet, there is a small difference in the effect between an

adult and child and infant. The effect of a release of HT on the dose to a child is about 1% less than to an adult; for an infant, the effect is about 3% more than to an adult.

Dose from inhalation

In DCART, inhalation contributed about 20% of total dose for adults, 19% for children, and 11% for infants. Variability between years on these percentages is about ± 10%.

Dose from swimming

Dose from swimming in the LLNL pool was calculated for each year. Given the assumptions about hours spent swimming[24], the percentage that swimming contributed to total dose averaged 0.003% for an adult, 0.008% for a child, and 0.004% for an infant.

Sensitivity of Dose Predictions

With the exception of release rates, dilution factors, absolute humidity, and, for some years, relative humidity, pathways and parameter values (and associated uncertainty) leading to dose in DCART for the TDR do not vary from year-to-year. It thus might be expected that the parameters to which total dose is sensitive also might not vary from year to year. The sensitivity of total dose at the Discovery Center was examined (all parameter values varying) for 1980 (a year when the Tritium Facility release rates were a normal fraction of total releases [45%]), 1985 (the year when other stacks released more tritium than did the Tritium Facility), 1994 (the year that area sources were the least important and the SNL/CA TRL released more tritium than did the Tritium Facility), and 1998 (the year that area sources were the largest fraction of total tritium released). The values obtained from each sensitivity analysis were ranked by correlation coefficient to obtain the four parameters to which the endpoint, dose, was most sensitive. Results are presented in Table 6a. The parameters to which dose is sensitive, common to adult, child, and infant, are shown for each year in Table 6b.

With the exception of the release rate of HTO from the Building 612 Yard, none of the parameters, to which dose was consistently sensitive, is associated with dispersion. Dose is sensitive to the intake of leafy vegetables and fruit/fruit vegetables, and to the dose coefficient for HTO. In all, nine parameters were found in the top ranked lists for sensitivity. The Building 612 Yard was important as a source both when the release rate of the Yard was derived from the release rate of the Tritium Facility (see Part 2 of the TDR) and when the release rate was actually measured, as in 1998.

[24] Triangular distributions in hours per year for adult (0 – 2.5 – 150), child (0 – 12 – 42), and infant (0 – 12 – 42) (see Part 2 of the TDR).

Table 6a. Parameters to which dose at the Discovery Center is sensitive for representative years.

Importance	Dose to adult	Dose to child	Dose to infant
Highest	**Fruit/fruit vegetable intake** HTO dose coefficient	**HTO dose coefficient**	**Milk uptake** Fruit/fruit vegetable intake HTO dose coefficient
Higher	None	OBT dose coefficient Release from B612 Yard	Release from Building 612 Yard
High	Leafy vegetable intake Release from Building 612 Yard	Fruit/fruit vegetable intake	Leafy vegetable intake

Note: Parameters in bold are those that ranked first
"Highest" refers to those parameters that were ranked in the top four for all years.
"Higher" refers to those parameters that were ranked in the top four for three years.
"High" refers to those parameters ranked in the top four for two years.

Table 6b. Parameters common to adult, child, and infant to which dose at the Discovery Center is sensitive for each year analyzed.

Year	Parameters
1980	Release from Building 612 Yard HTO dose coefficient
1985	Fruit/fruit vegetable intake HTO dose coefficient
1994	Fruit/fruit vegetable intake HTO dose coefficient
1998	Release from Building 612 Yard HTO dose coefficient

Discussion

Given the very different assumptions of DCART compared with all other models used by LLNL to calculate reported doses, the agreement between doses predicted by DCART and those reported historically by LLNL is remarkable, particularly when ambient tritium concentrations were used as input to DCART (Figures 6 and 7). However, it is useful to understand the various compensatory factors that caused the resulting agreement (see below and Appendix C).

Starting in 1973, LLNL reported annual tritium doses to the public. That year the dose was calculated from the highest weekly observed air tritium concentration obtained from the newly established air tritium monitoring network. From 1974 on, dose was based on

dispersion modeling. The doses shown in Figures 6 through 9 were obtained by summing all doses reported by LLNL. Up until 1985, the dose from each facility was reported as "fence post" dose, which is the equivalent of the currently used maximally exposed individual (MEI[25]), and the doses from each facility were not summed in the SAERs[26]. Summing these doses, each of which is calculated for a different location, is not comparable to the dose to the SW-MEI predicted by DCART and will result in a higher total dose than would have occurred at any location of the SW-MEI. From 1986 through 1991, the doses were reported for each facility at a perimeter location (unspecified). These doses were added together in the SAERs, however, which implies that dose to a SW-MEI was being calculated. From 1992 onwards, for compliance with NESHAPs, LLNL, reported doses specifically to the SW-MEI from both monitored stacks and diffuse area sources. The SW-MEI was defined at that time as the UNCLE Credit Union (Surano et al. 1993), and it is that dose that is most directly comparable to the dose predicted by DCART at the Discovery Center.

There are other factors that account for differences between the doses reported by LLNL and those predicted from ambient or predicted air concentrations using DCART (Figures 6 through 9). For instance, prior to 1992, only doses from stack releases were reported; from 1992 onwards, doses from both stack and diffuse sources were reported. Also, between 1973 and 1988, only tritium inhalation doses to the MEI or SW-MEI from stack releases were calculated using dispersion modeling[27]. Inhalation has always been the most important pathway to tritium dose for workers, and that perspective probably was retained even for dose to the public. With the advent of the CAAC, it was recognized that ingestion dose from tritium has the potential to be much more important to a member of the public than dose from tritium inhalation. (In DCART and the CAAC, inhalation only contributes about 20% or 16%, respectively, of a total tritium dose that assumes all food but no drinking water is contaminated.) In addition, for some years, all tritium released was modeled as HTO, while in other years, the HT released was not included in dose because the inhalation dose from HT is about four orders of magnitude lower than that from HTO (Table C3).

Some differences in reported and predicted results will have been due to dispersion modeling. Gaussian models may differ in the way in which σ_y[28] and σ_z[29] are calculated,

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[25] A hypothetical member of the public who, over an entire year, receives the maximum effective dose equivalent (summed over all pathways) from a given source of radionuclides to air. Generally, the MEI is different for each source at a site.

[26] The fence post dose, although not always identified, was most probably along the Laboratory's southern boundary with East Avenue for the Tritium Facility and Building 212, at least. As demonstrated in this TDR, the location of the SW-MEI was along East Avenue from 1974 through 1978 (observed concentrations at air tritium sampling location CAFÉ [adjacent to East Avenue] were about 50% greater than concentrations at VIS during the early years).

[27] Ingestion doses were calculated in the SAERs from 1979 onwards using equations from the US Nuclear Regulatory Commission's Regulatory Guide 1.109 (US NRC 1977) and observed concentrations in sampled vegetation or foodstuff, e.g., milk; these doses were reported in the data tables with the measured concentrations in vegetation and foodstuff.

[28] Standard deviation of the plume width in a horizontal direction

[29] Standard deviation of the plume width in a vertical direction

the way plume rise is calculated, whether or not, or how, building wake effects are handled, and the ways in which deposition and plume depletion are taken into account. Differences such as these in the models will result in the prediction of different dilution factors. The quality of the meteorological data collected and the way the data are formatted as input to the model can also affect predicted dilution factors. The result of these differences is that each Gaussian dispersion model will calculate a different dilution factor for each different set of meteorological data. In the case of CAP88-PC, there is only about ± 10% difference between dilution factors calculated from annual or four- or five-year wind files for each of the historical tritium sources with VIS as the receptor. This small difference is due largely to the fact that the wind blows towards VIS from the tritium sources with high frequency (about 54% of the time winds blow from the south-southwest through west directions). In contrast, the wind blows towards Location Q from most major sources with a much lower frequency than it does towards the Discovery Center. As a result, the variability at Location Q of dilution factors calculated by CAP88-PC using different wind files for a single facility can be as much as ± 40%.

Between 1979 and 1995, the SNL/CA TRL was operational. The dose contribution of SNL/CA should not be included as part of LLNL's dose impact, but it has been included in the TDR so that a direct comparison between doses predicted from ambient air concentrations and from dispersion modeling can be made. The percentage of total dose at the Discovery Center predicted to have been contributed by the TRL ranges from 1.9% (1979 – 1984) to 30% (1992 – 1994); the mean contribution for the years not listed is 14%. Thus, if only dose to the SW-MEI at the Discovery Center from LLNL operations during the years that the SNL/CA TRL was operational were being calculated, it would be up to one-third lower than what is shown in the figures.

The most similar, or at least less variable, comparison between DCART predictions and doses reported by LLNL occurs after 1992, the first year LLNL predicted doses using the regulatory model CAP88-PC for NESHAPs compliance. For these years, inhalation and ingestion doses were predicted for similar locations (VIS is very close to the UNCLE Credit Union). Although inhalation and ingestion assumptions in DCART and CAP88-PC are very different and the dose coefficients are different, when HTO concentrations in air and absolute humidity are the same, when neither model includes any contaminated drinking water, and when both models assume all food is homegrown, the dose to an adult calculated using DCART is 43%[30] that predicted by CAP88-PC (e.g., 1992 through 2003). In 2004, when LLNL changed the ingestion assumptions in CAP88-PC to be more realistic (yet still conservative), the dose to an adult predicted by DCART became 110%[30] of the comparable dose predicted using CAP88-PC. This change is reflected in the comparisons shown in Figure 8, where the dose predictions of CAP88-PC, which for

[30] Note that this relationship (which is not obvious in Figure 8) is based on deterministic parameters in DCART. DCART's deterministic doses are always lower than its stochastic doses because of the uncertainty distributions on the dose coefficients (Paterson 2006).

many years were approximately the same as the mean doses predicted using DCART, suddenly drop close to the lower confidence limits of the DCART predictions.

A summary of the known assumptions behind the reported doses may be found in Appendix C. A comparison of dose coefficients used by LLNL throughout the years is provided in Table C4.

Dose predictions based on observed concentrations of HTO in air include the contribution of HT that was converted to HTO in the environment, but, because there is no way to distinguish HTO converted from HT from released HTO, there is no way to determine the fraction of dose contributed by the HTO that arose from converted HT. A model is necessary for this calculation. In DCART, a release of HT has about a 20% greater effect on dose than it does on HTO concentrations in air (Figure 4) because, after a release of HT, soil microorganisms rapidly convert HT to HTO and the soil becomes a much more important source of HTO than it is after a release of HTO. As a result, the concentration of HTO in plant water after a release of HT is higher than the concentration of HTO in air moisture, while, for a release of HTO, the concentrations in the plant water are lower than those in air moisture because the soil is a relatively unimportant source of HTO compared with the air[31]. This effect is most dramatic for root crops, which are assumed to have a concentration approximately equal to the concentration in soil water. After a release of HT, it is estimated that the dietary contribution to ingestion dose from root crops is double that seen after a release of HTO.

The sensitivity analysis demonstrated that the HTO dose coefficient has an uncertainty less than that of the OBT coefficient but that dose is sensitive to it because so much of the dose is derived from ingested HTO (as confirmed by the importance of leafy and fruit vegetables – see Table 6).

That the entire diet could have been contaminated or that a person stayed home 24 hours a day for the entire year, as is assumed in DCART, is highly unlikely. Dose predictions for 1977 (the year of the highest dose impact) were recalculated after the fraction ingested of each food category, except grain, was changed from a deterministic value (1 or 100%) to a triangular distribution of 0 – X – 1, where "X" is the mean fraction of home-grown foodstuff in the Western United States from the U.S. EPA "Exposure Factors Handbook (1999)[32]. The inhalation rate was adjusted to reflect occupancy of the SW-MEI with a rectangular distribution of 0.5 – 0.9 (50% – 90%)[33]. Although distributed, these assumptions are still conservative because they account for a small probability that ingestion (grain excepted) can be 100% contaminated. Consumption of contaminated grain was changed from 100% to 0% for both people and animals, because

[31] See equation 4 in Part 1 of the TDR (Peterson 2006).

[32] "X" equals 0.015 for leafy vegetables, 0.084 for root crops, 0.12 for fruits and no-leafy vegetables, 0.007 for dairy, 0.041 for beef, 0.011 for pork, 0.008 for poultry and 0.021 for eggs.

[33] Lognormal distributions representing the occupancy factor were calculated using Crystal Ball® to be 3400 ± 1250 $m^3 y^{-1}$ for the adult, 3450 ± 1600 $m^3 y^{-1}$ for the child, and 1150 ± 822 $m^3 y^{-1}$ for the infant.

grains were not grown locally. Using the revised assumptions, the total mean doses to an adult, child, and infant were reduced by a factor of three compared with the mean doses obtained using a completely contaminated diet. The 97.5% confidence limit was reduced slightly less by about a factor of 2.5.

The annual dose with the highest upper confidence limit (60.5 μSv or 6.05 mrem) was to an infant in 1977. If this were reduced by a factor of 2.5, the highest dose that could have been received would have been 24.2 μSv (2.42 mrem). As a point of reference, the current EPA annual dose limit from releases to the atmosphere for a member of the public is 100 μSv (10 mrem).

CONCLUSIONS

Tritium in air concentrations at VIS, which drive predicted doses at the Discovery Center, obviously roughly track the magnitude of tritium released by operations to the atmosphere as HTO or as HT (after conversion to HTO in the environment). Doses calculated at the Discovery Center have decreased by as much as a factor of 60 (highest doses in 1977 and 1978 compared with the lowest doses in 2004 and 2005) in thirty years.

CAP88-PC was used to calculate dilution factors at the locations of the SW-MEI for each facility. These dilution factors were then used in DCART to predict tritium in air moisture concentrations at VIS from the estimated release rates and to predict doses to adult, child, and infant at the two locations of the SW-MEI. Predicted air moisture concentrations at VIS were usually within a factor of two of the observed mean annual concentrations, and the confidence intervals on the predictions overlapped the confidence intervals on the observations most of the time. For the few cases when the confidence intervals did not overlap, predicted air moisture concentration exceeded observed air moisture concentrations by more than a factor of two. The cause for this was probably something other than the dispersion model (Appendix B). At no time did the dilution factors derived from CAP88-PC and used as input to DCART cause an underestimation of air moisture concentrations at VIS; even in 1974, when the P/O ratio for air moisture was less than one, the confidence intervals overlapped enough so that the predictions could be considered indistinguishable from the observations.

Even though accidental releases are known to have occurred[34], their impact, if any, cannot be discerned from the mean annual tritium concentrations measured at VIS. This may be because the accidental releases were too small relative to the routine releases to have a noticeable effect on air concentration or because the winds were blowing away from the Discovery Center during the releases.

[34] 1984 – 5,200 Ci HT from LLNL; 1985 – 1,000 Ci from LLNL; 1986 – 200 Ci HTO from SNL/CA; 1987 – 1,100 Ci from SNL/CA.

In DCART, concentrations of HTO from HT releases to the atmosphere are estimated using empirical factors for the conversion of HT to HTO in the environment that were obtained from experiments carried out in a very different climate from that of Livermore. Nevertheless, it appears that overestimations of HTO in air moisture that might have been expected for years of high HT releases had the empirical conversion factor been too large did not occur. Thus the conversion factors can be applied appropriately to conditions at LLNL.

Because monitoring demonstrates what is rather than what might be, a more accurate dose prediction will be obtained if observed air concentrations are used as input to a dose model rather than the amount of tritium released in a year[35]. The decision to estimate probabilistic doses at the Discovery Center was based on the desire to confirm that air concentrations could be predicted with a degree of accuracy that would demonstrate the validity of the assumptions about release rates and dilution factors. This was accomplished, because the confidence intervals of doses calculated from estimated release rates and dilution factors and those calculated from observed air concentrations overlapped for all years except for four years when the predicted air concentrations were distinctly higher[36]. These results are very important, because, for the next phase of the dose reconstruction (1953 – 1972), it is necessary and desirable to have demonstrated that dose predictions using release rates and dispersion modeling are not very different from dose predictions obtained from observed air concentrations, and that, consequently, reasonable confidence can be placed in the doses predicted from release rates, assuming all sources have been assessed appropriately.

All doses to the hypothetical individual living at the Discovery Center have been calculated both from observed air concentrations and from air concentrations derived from dispersion modeling. However, because doses that can be calculated from observed air concentrations are more accurate, they have been treated as the true dose in this TDR. When the SW-MEI was at Location Q, doses had to be calculated from dispersion modeling alone. Without being able to test the dispersion model at Location Q, it is not possible to say whether or not the resulting doses would have been higher (or lower) than they would have been had observed air concentrations been available to use in the dose calculations. Most likely, the magnitude of the uncertainty on the predictions would have overlapped significantly with the uncertainty on potential observations.

The uncertainty about the best-estimated dose is primarily due to uncertainty about the measured air concentrations at VIS or about the uncertainty in the release rate and in the dispersion model. The uncertainty also accounts for uncertainty in the dose coefficients and in the ingestion rates. The uncertainty in the dose predictions averaged 5.6 (upper

[35] As mentioned earlier, the uncertainty on the observed air concentrations was sometimes greater than the uncertainty on the predicted air concentrations. In this context, "more accurate" means that a predicted dose would be more likely to fall within the confidence interval calculated from observed rather than from predicted air concentrations.
[36] This error is conservative and health protective.

confidence limit divided by lower confidence limit) whether or not the doses were predicted from observed or predicted air concentrations.

Doses calculated for this TDR include the contribution from SNL/CA. Releases from SNL/CA were included so that the doses predicted from dispersion modeling could be compared with doses predicted from observed concentrations of tritium in air at VIS (which include the contribution of SNL/CA to the atmospheric tritium sampled). The predicted dose will drop by about 30% from 1992 through 1994 if the SNL/CA contribution is removed; on average, SNL/CA contributed 13% of the dose at the Discovery Center between 1979 and 1995.

In Figures 6 through 9, annual doses reported by LLNL (Tables C1 and C2) have been included along with those predicted by DCART to demonstrate that the Laboratory's assessment of dose has generally been reasonable and conservative. The annual doses reported by LLNL never included dose from SNL/CA, and for many years, only inhalation dose was reported. Yet most LLNL dose predictions fell within the confidence intervals predicted using DCART. In general it seems that, no matter what doses are accounted for (inhalation, ingestion of HTO, or ingestion of OBT), essentially all tritium doses from routine releases will fall within a factor of five of each other, even when dispersion models are different.

Diffuse area sources are relatively more important to dose at the perimeter than stack sources because of how the tritium is dispersed by winds close to the ground. It is clear that waste accumulation areas and storage areas are very important sources of dose to the SW-MEI, especially if the area source is near the perimeter. SNL/CA must have had area sources similar to LLNL's although it is likely the annual release rate(s) from the source(s) would have been much smaller. The impact of any SNL/CA area source would have also been minimal at VIS, because of the distance between the source and receptor. Certainly, any contribution to dose at the Discovery Center from an SNL/CA area source has been accounted for by using the measured air tritium concentrations at VIS to estimate dose.

Dose from swimming in the LLNL pool contributed negligibly to total dose and will not be included as a pathway to dose in Part 4 of the TDR.

In general, the goal of dose reconstruction is to estimate, to the extent possible, actual exposures and doses received by real people. This TDR has not done that. Rather, it, like regulatory models, has adopted the conservative assumption that 100% of the diet was contaminated at a concentration equal to the concentration in air at the location of the SW-MEI, that all the food ingested was equally contaminated with tritium, and that all food was home-grown. Additional conservatism was added by selecting the locations of the SW-MEI (the Discovery Center and Location Q) to be where no one had actually lived. Annual air concentrations and resulting doses at the location of the SW-MEI were therefore higher than at any actual residence. Exposure to LLNL (and SNL/CA) tritium

by real people at real residences would have been lower because air concentration is more or less inversely proportional to the square of the distance from the source, and most of the diet would not have been homegrown. Further conservatism at VIS is due to known overestimation of air concentrations at that location.

The end result of all these conservative assumptions is that it can be stated confidently that no individual could have exceeded what was calculated as the 97.5 percentile of the dose distribution. It is even possible that the dose to a real individual fell below the 2.5 percentile of the distribution. The assumptions behind this TDR could be changed to reduce the predicted dose, but this is an unnecessary effort given that these conservative doses are already well below the current[37] compliance limit of 10 mrem from atmospheric releases. The estimated maximum annual dose for the years 1973 through 2005 was to a hypothetical infant living at Location Q in 1977. This dose (60.5 μSv [6.05 mrem]) was calculated from release rates and dilution factors because no ambient air monitor was located there. At VIS, a maximum annual dose (97.5% confidence limit) for the years 1973 through 2005 of 34.4 μSv (3.44 mrem) was calculated from ambient air concentrations to an infant in 1976.

The total maximum dose (97.5% confidence limit) that could have been received at the Discovery Center from exposure to all releases between 1973 and 2005 was estimated from observed ambient HTO concentrations in air to be 150 μSv (15 mrem) and would have been received by an individual born in 1973 who had lived at that location through 2005. Over the same period of time, the external dose from naturally occurring cosmic and terrestrial radiation would have amounted to about 20 mSv (2 rem). Clearly, the Laboratory's tritium releases between 1973 and 2005 added less than 1% to the dose that would have been received by an individual from naturally occurring cosmic and terrestrial radiation had the Laboratory not existed; at 1%, the dose impact from LLNL (and SNL/CA) tritium cannot be distinguished from doses from natural environmental radioactivity.

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[37] Prior to December 15, 1989, the EPA radiation dose standards which apply to air emissions (contained in 40 Code of Federal Regulations Part 61 Subpart H of the Clean Air Act) were 25 mrem/y dose equivalent for whole-body exposures from the air pathway and 75 mrem/y dose equivalent for exposure of any organ from the air pathway.

APPENDIX A

INPUT DATA

Appendix A contains all annual input used in DCART for the LLNL tritium dose reconstruction from 1973 through 2005. Data in these tables are also found in Part 2 of the TDR.

Table A1. HTO (Ci) estimated to have been released annually from the Building 331 WAA, the Building 612 Yard and the evaporation trays before concentrations of tritium in air were measured near area sources. The distributions for both are normal with one standard deviation (σ). A positive correlation (0.5) exists between releases from the Building 331 WAA and releases for HT and HTO from the Tritium Facility; for the Building 612 Yard and the evaporation trays, there is a positive correlation (0.4) with HTO released by the Tritium Facility.

Year	B331 WAA	B612 Yard	Evaporation Trays
1973	63.3 ± 50.7	46.4 ± 21.3	4.30 ± 4.15
1974	43.2 ± 34.1	19.8 ± 9.92	4.30 ± 4.15
1975	58.7 ± 45.8	36.7 ± 14.3	4.30 ± 4.15
1976	75.8 ± 59.4	62.2 ± 22.7	4.30 ± 4.15
1977	83.8 ± 65.8	77.6 ± 27.0	-----
1978	110 ± 85.8	78.3 ± 26.5	-----
1979	101 ± 78.3	59.7 ± 18.6	-----
1980	54.3 ± 42.3	52.0 ± 17.0	-----
1981	63.5 ± 49.4	56.2 ± 18.1	-----
1982	47.3 ± 36.8	47.9 ± 15.1	-----
1983	73.6 ± 57.4	44.4 ± 14.3	-----
1984	47.7 ± 37.6	38.2 ± 12.1	-----
1985	24.5 ± 19.0	23.6 ± 7.29	-----
1986	26.9 ± 20.9	23.1 ± 7.36	-----
1987	65.3 ± 50.8	45.8 ± 14.5	-----
1988	98.3 ± 76.5	61.2 ± 19.5	-----
1989	73.4 ± 57.0	59.4 ± 18.6	-----
1990	31.1 ± 24.1	35.5 ± 14.4	-----
1991	28.4 ± 22.1	25.5 ± 8.09	-----
1992	-----	3.79 ± 1.21	-----

Table A2. Estimated routine annual releases in curies of HT and HTO from the LLNL Tritium Facility (Building 331). Uncertainty is one standard deviation (σ) of a normal distribution. In 1973, before speciation was known, HT and HTO releases are negatively correlated (-0.4 for Stack 1 and -0.5 for Stack 2).

	Stack 1 HT	Stack 1 HTO	Stack 2 HT	Stack 2 HTO
1973	442 ± 209	518 ± 226	736 ± 318	864 ± 338
1974	463 ± 149	226 ± 70.8	629 ± 215	431 ± 144
1975	636 ± 131	344 ± 71.0	622 ± 147	773 ± 183
1976	296 ± 70.2	841 ± 201	958 ± 254	973 ± 260
1977	531 ± 143	1170 ± 319	742 ± 218	953 ± 284
1978	793 ± 178	954 ± 229	1510 ± 382	1180 ± 317
1979	1040 ± 189	734 ± 134	1410 ± 255	894 ± 163
1980	262 ± 49.1	509 ± 93.6	517 ± 116	908 ± 201
1981	516 ± 93.7	1130 ± 204	520 ± 94.4	403 ± 72.7
1982	122 ± 26.9	490 ±108	483 ± 94.5	818 ± 160
1983	255 ± 57.7	318 ± 70.3	1510 ± 306	892 ± 175
1984	285 ± 12.4	487 ± 111	602 ± 254	556 ± 113
1985	140 ± 25.9	359 ± 65.1	198 ± 36.7	293 ± 53.2
1986	155 ± 30.4	228 ± 45.8	302 ± 56.8	402 ± 77.8
1987	242 ± 43.5	606 ± 109	1150 ± 207	644 ± 116
1988	437 ± 79.3	553 ± 101	1870 ± 340	1120 ± 205
1989	354 ± 65.7	896 ± 169	995 ± 185	725 ± 136
1990	143 ± 28.5	338 ± 66.5	419 ± 81.4	356 ± 68.3
1991	40.0 ± 7.76	204 ± 37.0	415 ± 80.3	492 ± 89.5
1992	20.1 ± 4.55	52.5 ± 11.9	59.5 ± 13.3	51.0 ± 11.4
1993	15.2 ± 3.39	41.5 ± 9.25	109 ± 23.9	72.9 ± 16.0
1994	4.11 ± 0.922	22.4 ± 4.99	56.8 ± 12.4	53.7 ± 11.8
1995	1.08 ± 0.283	5.95 ± 1.40	27.9 ± 6.19	56.8 ± 12.6
1996	0.813 ± 0.176	11.3 ± 2.10	32.7 ± 5.94	170 ± 32.8
1997	0.936 ± 0.174	10.1 ± 1.81	31.0 ± 5.57	257 ± 46.1
1998	0.608 ± 0.122	9.76 ± 1.89	24.4 ± 4.71	74.7 ± 14.4
1999	0.263 ± 0.0613	5.45 ± 1.09	66.5 ± 13.3	208 ± 41.5
2000	0.0485 ± 0.0140	4.05 ± 0.834	4.86 ± 1.01	31.1 ± 6.40
2001	0.274 ± 0.0968	2.40 ± 0.582	1.44 ± 0.308	15.9 ± 3.37
2002	0.0120 ± 0.00629	0.0126 ± 0.00558	3.47 ± 0.759	32.8 ± 7.12
2003	0.506 ± 0.271	0.0603 ± 0.0222	5.66 ± 1.41	103 ± 23.9
2004	0.600 ± 0.292	0.030 ± 0.0118	3.82 ± 0.950	12.1 ± 3.33
2005	0.480 ± 0.254	0.030 ± 0.0111	1.09 ± 0.313	30.1 ± 7.61

Table A3. Annual routine releases (Ci) of HT and HTO from the Insulating Core Transformer (Building 212); uncertainty is one standard deviation (σ) of a normal distribution except for 1973, which is triangular.

Year	HT	HTO
1973	44 – 330 –528 (triangular)	6 – 45 –72 (triangular)
1974	493 ± 281	66.8 ± 38.1
1975	1070 ± 611	145 ± 82.7
1976	1030 ± 561	140 ± 76.0
1977	1810 ± 985	246 ± 133
1978	984 ± 540	133 ± 73.1
1979	384 ± 208	52.0 ± 28.2
1980	61.6 ± 33.5	8.35 ± 4.53
1981	20.6 ± 11.2	2.79 ± 1.52
1982	29.9 ± 16.3	4.06 ± 2.20
1983	123 ± 66.9	16.7 ± 9.07
1984	9.69 ± 5.26	1.31 ± 0.712
1985	4.40 ± 2.51	0.596 ± 0.340
1986	13.0 ± 3.50	2.00 ± 0.548
1987	34.0 ± 9.14	4.00 ± 1.08

Table A4. Annual routine releases (Ci) of HT and HTO from the SNL/CA Tritium Research Laboratory and uncertainty as one standard deviation (σ) of a normal distribution

Year	HT	HTO
1979	1.08 ± 0.377	4.64 ± 1.00
1980	4.77 ± 1.64	20.5 ± 4.36
1981	8.11 ± 2.78	34.8 ± 7.40
1982	38.1 ± 13.0	164 ± 34.7
1983	21.2 ± 3.03	74.2 ± 14.7
1984	18.5 ± 2.64	146 ± 31.7
1985	128 ± 18.1	389 ± 77.0
1986	129 ± 18.2	415 ± 115
1987	159 ± 37.8	573 ± 113
1988	543 ± 75.8	1040 ± 224
1989	180 ± 25.3	659 ± 142
1990	50.8 ± 7.23	244 ± 48.3
1991	113 ± 16.1	352 ± 69.6
1992	130 ± 21.6	134 ± 29.1
1993	55.3 ± 7.87	132 ± 26.4
1994	4.10 ± 0.608	91.2 ± 18.1
1995	1.06 ± 0.170	73.0 ± 14.5

Table A5. Annual HTO releases (Ci) from the Building 331 WAA and the Building 612 Yard estimated from annual mean air tritium concentrations from nearby tritium samplers; uncertainty is one standard deviation (σ) of a normal distribution.

Year	B331 WAA	B612 Yard
1992	4.42 ± 1.62	-----
1993	9.22 ± 3.36	4.24 ± 1.54
1994	3.01 ± 1.10	1.92 ± 0.700
1995	31.4 ± 11.4	4.17 ± 1.52
1996	10.1 ± 3.68	12.4 ± 4.50
1997	8.04 ± 2.92	7.19 ± 2.61
1998	14.4 ± 5.23	8.47 ± 3.08
1999	15.8 ± 5.74	7.61 ± 2.77
2000	2.32 ± 0.850	5.47 ± 1.99
2001	0.679 ± 0.235	2.27 ± 0.689
2002	0.755 ± 0.260	2.33 ± 0.703
2003	8.70 ± 2.89	3.43 ± 1.03
2004	0.695 ± 0.240	3.18 ± 0.956
2005	4.76 ± 1.58	1.48 ± 0.447

Table A6. HTO (Ci) released annually from the Building 624 incinerator and Building 292. A triangular uncertainty distribution on the incinerator results for 1977- 1985 is based on measurements taken in 1988 of all incinerated concentrations. Uncertainty on the Building 292 releases is normal ± one standard deviation (σ).

	Building 624 Incinerator			
Year	Lower limit	Peak	Upper limit	Building 292
1977	0.0001	0.80	17	-----
1978	0.0001	0.80	17	-----
1979	0.0001	0.80	17	18.7 ± 7.31
1980	0.0001	0.80	17	27.7 ± 10.8
1981	0.0001	0.80	17	70.7 ± 27.6
1982	0.0001	0.80	17	89.6 ± 35.0
1983	0.0001	0.80	17	130 ± 50.6
1984	0.0001	0.80	17	229 ± 89.3
1985	0.0001	0.80	17	336 ± 131
1986	3.47	4.43	5.38	178 ± 72.0
1987	0.195	0.216	0.238	126 ± 49.0
1988	0.333	0.343	0.353	8.46 ± 3.30
1989	-----	-----	-----	3.84 ± 1.50

Table A7. Release rates (Ci) and uncertainty (σ) on normal distributions for the Decontamination and Waste Treatment Facility.

Year	DTWF Stack - HT	DTWF Stack - HTO	DWTF Area - HTO
2004	1.0 ± 0.818	19.0 ± 9.08	-----
2005	0.107 ± 0.0532	2.62 ± 0.943	[0.21][1]

[1] This source was not modeled in the TDR because of its minimal relative contribution to dose

Table A8. Dilution factors (χ/Q in s m^{-3}) with uncertainty (σ) on lognormal distributions for modeled sources other than the LLNL Tritium Facility.

Building	Years	χ/Q (s m^{-3})
B331 WAA	1958 - present	$3.474 \times 10^{-6} \pm 1.042 \times 10^{-6}$
Evaporation Trays	1962- 1976	$2.008 \times 10^{-5} \pm 8.032 \times 10^{-6}$
B612 Yard	1965 - present	$1.763 \times 10^{-5} \pm 5.289 \times 10^{-6}$
B212 Stack	1966 - 1987	$2.494 \times 10^{-6} \pm 7.483 \times 10^{-7}$
B624 incinerator	1977 - 1988	$7.925 \times 10^{-6} \pm 2.380 \times 10^{-6}$
B292 Stack	1979 - 1989	$4.425 \times 10^{-7} \pm 1.427 \times 10^{-7}$
Sandia TRL Stack	1979 - 1995	$6.595 \times 10^{-7} \pm 2.008 \times 10^{-7}$
DWTF Stack	2004 - present	$3.472 \times 10^{-7} \pm 1.042 \times 10^{-7}$

Table A9. Dilution factors (χ/Q in s m^{-3}) with uncertainty (σ) on lognormal distributions for Stack 1 and Stack 2 of the LLNL Tritium Facility (Building 331)

Year	Stack 1 (south)	Stack 2 (north)
1973 - 1978	$1.230 \times 10^{-6} \pm 3.080 \times 10^{-7}$	$1.077 \times 10^{-6} \pm 2.718 \times 10^{-7}$
1979	$1.240 \times 10^{-6} \pm 3.101 \times 10^{-7}$	$1.101 \times 10^{-6} \pm 2.758 \times 10^{-7}$
1980	$1.237 \times 10^{-6} \pm 3.093 \times 10^{-7}$	$1.081 \times 10^{-6} \pm 2.722 \times 10^{-7}$
1981	$1.233 \times 10^{-6} \pm 3.084 \times 10^{-7}$	$1.059 \times 10^{-6} \pm 2.653 \times 10^{-7}$
1982 - 1984	$1.217 \times 10^{-6} \pm 3.055 \times 10^{-7}$	$1.073 \times 10^{-6} \pm 2.694 \times 10^{-7}$
1985	$1.201 \times 10^{-6} \pm 3.004 \times 10^{-7}$	$1.086 \times 10^{-6} \pm 2.719 \times 10^{-7}$
1986	$1.192 \times 10^{-6} \pm 2.985 \times 10^{-7}$	$1.084 \times 10^{-6} \pm 2.715 \times 10^{-7}$
1987	$1.184 \times 10^{-6} \pm 2.965 \times 10^{-7}$	$1.083 \times 10^{-6} \pm 2.711 \times 10^{-7}$
1988	$1.187 \times 10^{-6} \pm 2.969 \times 10^{-7}$	$1.086 \times 10^{-6} \pm 2.720 \times 10^{-7}$
1989	$1.189 \times 10^{-6} \pm 2.974 \times 10^{-7}$	$1.090 \times 10^{-6} \pm 2.729 \times 10^{-7}$
1990	$1.177 \times 10^{-6} \pm 2.951 \times 10^{-7}$	$1.080 \times 10^{-6} \pm 2.709 \times 10^{-7}$
1991	$1.166 \times 10^{-6} \pm 2.919 \times 10^{-7}$	$1.068 \times 10^{-6} \pm 2.675 \times 10^{-7}$
1992 - 1994	$1.184 \times 10^{-6} \pm 2.974 \times 10^{-7}$	$1.088 \times 10^{-6} \pm 2.738 \times 10^{-7}$
1995	$1.203 \times 10^{-6} \pm 3.011 \times 10^{-7}$	$1.108 \times 10^{-6} \pm 2.775 \times 10^{-7}$
1996	$1.209 \times 10^{-6} \pm 3.025 \times 10^{-7}$	$1.095 \times 10^{-6} \pm 2.741 \times 10^{-7}$
1997	$1.216 \times 10^{-6} \pm 3.042 \times 10^{-7}$	$1.102 \times 10^{-6} \pm 2.761 \times 10^{-7}$
1998	$1.225 \times 10^{-6} \pm 3.067 \times 10^{-7}$	$1.098 \times 10^{-6} \pm 2.754 \times 10^{-7}$
1999	$1.240 \times 10^{-6} \pm 3.105 \times 10^{-7}$	$1.088 \times 10^{-6} \pm 2.733 \times 10^{-7}$
2000	$1.240 \times 10^{-6} \pm 3.107 \times 10^{-7}$	$1.115 \times 10^{-6} \pm 2.803 \times 10^{-7}$
2001	$1.245 \times 10^{-6} \pm 3.119 \times 10^{-7}$	$1.146 \times 10^{-6} \pm 2.881 \times 10^{-7}$
2002	$1.253 \times 10^{-6} \pm 3.142 \times 10^{-7}$	$1.125 \times 10^{-6} \pm 2.832 \times 10^{-7}$
2003	$1.254 \times 10^{-6} \pm 3.145 \times 10^{-7}$	$1.125 \times 10^{-6} \pm 2.831 \times 10^{-7}$
2004	$1.253 \times 10^{-6} \pm 3.147 \times 10^{-7}$	$1.125 \times 10^{-6} \pm 2.839 \times 10^{-7}$
2005	$1.231 \times 10^{-6} \pm 3.091\ 10^{-7}$	$1.144\ 10^{-6} \pm 2.890 \times 10^{-7}$

Table A10. Annual mean observed concentrations of tritium in air moisture (Bq L^{-1}) and air (Bq m^{-3}) at the location of the air tritium sampler, VIS, with one standard deviation (σ) uncertainty. Weapons testing fallout and cosmogenic tritium background[38] have been subtracted.

Year	Bq L^{-1}	Bq m^{-3}
1973	385 ± 102	3.91[39] ± 1.17
1974	448 ± 68.8	3.14 ± 0.665
1975	400 ± 66.2	2.97 ± 0.639
1976	572 ± 116	5.94[39] ± 1.46
1977	579 ± 87.4	4.22 ± 0.872
1978	438 ± 66.4	3.07 ± 0.637
1979	349 ± 59.4	2.73 ± 0.604
1980	360 ± 54.6	2.76 ± 0.570
1981	213 ± 36.4	1.51 ± 0.334
1982	290 ± 44.1	2.47 ± 0.512
1983	221 ± 42.2	2.04 ± 0.484
1984	291 ± 45.6	2.28 ± 0.479
1985	203 ± 38.8	1.53 ± 0.363
1986	220 ± 59.2	1.72 ± 0.524
1987	451 ± 70.5	3.20 ± 0.672
1988	347 ± 75.0	2.48 ± 0.639
1989	248 ± 47.3	1.76 ± 0.416
1990	152 ± 24.2	1.02 ± 0.216
1991	104 ± 16.7	0.714 ± 0.153
1992	31.5 ± 5.13	0.254 ± 0.0548
1993	29.5 ± 4.87	0.245 ± 0.0533
1994	18.4 ± 3.29	0.143 ± 0.0327
1995	16.9 ± 3.15	0.144 ± 0.0337
1996	40.9 ± 6.51	0.287 ± 0.0612
1997	41.7 ± 6.72	0.380 ± 0.0813
1998	17.2 ± 3.80	0.133 ± 0.0350
1999	31.5 ± 5.42	0.217 ± 0.0435
2000	11.3 ± 2.65	0.0903 ± 0.0216
2001	6.57 ± 1.37	0.0512 ± 0.0109
2002	8.36 ± 2.20	0.0606 ± 0.0161
2003	26.3 ± 2.47	0.178 ± 0.0180
2004	5.53 ± 1.31	0.0397 ± 0.00959
2005	6.61 ± 2.22	0.0522 ± 0.0176

[38] The highest fallout concentration was 4 Bq L^{-1} in 1974, which was 0.9% of the tritium concentration observed at VIS; the lowest fallout concentration was 0.31 Bq L^{-1} in 2005, which was less than 5% of the tritium concentration observed at VIS.

[39] When Bq m^{-3} is graphed against Bq L^{-1}, these values fall far from the line created by the data from all other years. The reason for this is unknown.

Table A11. Annual absolute humidity (kg m^{-3}), as measured by water absorbed by silica gel and flow rate through the sampler, and relative humidity from the LLNL meteorological tower. Uncertainty on absolute humidity is ± 5%; uncertainty on relative humidity is ± 10%.

Year	Absolute humidity	Relative humidity
1974	0.0071	-----
1975	0.0074	-----
1976	0.0079	-----
1977	0.0071	-----
1978	0.0081	-----
1979	0.0082	-----
1980	0.0080	-----
1981	0.0076	-----
1982	0.0081	-----
1983	0.0088	-----
1984	0.0080	-----
1985	0.0074	-----
1986	0.0077	-----
1987	0.0075	-----
1988	0.0074	-----
1989	0.0073	-----
1990	0.0072	-----
1991	0.0074	-----
1992	0.0078	-----
1993	0.0079	-----
1994	0.0075	-----
1995	0.0082	-----
1996	0.0076	-----
1997	0.0081	-----
1998	0.0081	-----
1999	0.0077	0.760
2000	0.0085	-----
2001	0.0081	0.632
2002	0.0077	0.664
2003	0.0077	0.704
2004	0.0075	0.656
2005	0.0083	0.693
Overall mean	**0.0078**	**0.690**

Table A12. Mean annual HTO concentrations in the LLNL swimming pool in Bq L^{-1}, fraction of 12 months represented by annual sampling, uncertainty (σ), and the annual maximum and minimum concentrations.

	# of monthly samples / 12	**Bq L^{-1}**	**Maximum**	**Minimum**
1988	0.92	67.2 ± 13.9	86.6	48.1
1989	0.58	48.6 ± 18.8	71.4	19.7
1990	0.92	40.3 ± 7.97	49.2	26.6
1991	1.0	39.0 ± 13.2	58.1	22.6
1992	0.92	16.6 ± 5.74	23.0	7.81
1993	0.67	6.76 ± 3.29	10.2	2.08
1994	1.0	4.41 ± 1.33	5.96	3.18
1995	0.83	5.68 ± 2.39	8.92	2.33
1996	1.0	3.34 ± 1.99	5.51	1.62
1997	0.58	6.38 ± 4.36	13.6	1.14
1998	0.33	5.48 ± 2.65	7.81	3.28
1999	0.33	5.76 ± 3.31	10.1	2.98
2000	0.25	2.69 ± 1.36	2.92	2.51

APPENDIX B

AIR MOISTURE PREDICTIONS:

Comparison with Earlier Test of CAP88-PC and Reasons for Over-predictions

The comparison of concentrations of tritium in air moisture predicted by DCART with observed mean annual air moisture concentrations at the air tritium sampler, VIS, located next to the Discovery Center, provides an excellent test of the dilution factors predicted by the dispersion model in CAP88-PC, the assumptions about the magnitude of the release rates, and the uncertainty associated with both dilution factors and release rates. To a lesser extent, assumptions made about mean annual absolute humidity[40] are also tested. By and large, the concentrations of tritium in air moisture obtained by dividing the predicted air concentration by the absolute humidity used in DCART are in excellent agreement with the observations (Figure 2), and the results are well within the factor of two to four uncertainty expected in annual average air concentrations predicted using a Gaussian dispersion model over flat terrain (Miller and Hively 1987). The success rate at VIS, for most predictions within a factor of two, suggests that any P/O ratio greater than two may be the result of incorrect assumptions about the quantity of tritium released during a particular year. This assumption is supported by the fact that, for each year in which the TDR's P/O ratio was greater than 2, the confidence intervals on the predicted and observed air concentrations either failed to overlap or barely overlapped.

An earlier test of CAP88-PC (Peterson 2003, 2004) covered the years from 1986 through 2001 and compared predicted tritium concentrations in air with data from all LLNL air tritium monitors having more than 50% of each year's samples above the detection limit.

There are several differences between the test of CAP88-PC provided by this TDR and the earlier test of CAP88-PC.

1. The earlier test used only HTO release rates as input to the model, while the TDR modeled releases of both HT and HTO. The effect of modeling releases of HT varied with each year (Figure 4), but the predicted concentration of HTO in air moisture would have increased by at most 7% when HT releases were included.
2. Release rates in the earlier test were those primarily reported in the LLNL or SNL/CA SAERs, while release rates for the TDR were distributed values based

[40] Although air dispersion models predict tritium concentration per volume of air and air tritium monitoring also provides comparable numbers based on the volume of air that has passed through the sampler in a given time, less error is associated with the concentration in the water from air moisture collected on silica gel. However, to compare the air concentration predicted from release rates and dilution factors with the observed air moisture concentration, the predicted air concentration must be divided by the annual estimated value for absolute humidity (kg water per m^{-3} air).

on additional information. The deterministic release rates used in the earlier test are compared in Table B1 with the means of the distributed release rates from the TDR.

3. The earlier test used the most recent meteorological data for the preceding five years to prepare the wind file used in CAP88-PC. The TDR used four years (2000 through 2003) of meteorological data for the wind file. Improvements are continually being made to instrumentation at the LLNL meteorological tower, and, at the time the wind file for the TDR was prepared, the highest quality data were represented by these years.
4. For the earlier test, the annual release rate from the Building 612 Yard was assumed to be 2% of the HTO released from the Tritium Facility for the years before the release rate was estimated from concentrations of tritium in air measured nearby. For the TDR, this assumption was doubled to 4%. In addition, new assumptions were made in the TDR about quantities of tritium released from the Building 331 WAA before the release rate was estimated from measured air concentrations (see Table B1.)
5. The earlier test was a direct comparison of tritium concentrations in air volume (Bq m^{-3}) predicted by CAP88-PC and observed mean annual tritium concentrations in air volume (Bq m^{-3}). In this TDR, predicted and observed tritium concentrations in air moisture (Bq L^{-1}) were compared.

P/O ratios of the concentration of tritium in air moisture and in air volume at VIS are compared in Table B2 for the test of CAP88-PC and the TDR. The results of the two tests were similar for most years; perhaps they were more similar than might have been expected given the different assumptions. Differences in the predictions of the two approaches will have been caused by a combination of any of the above differences. Both studies over-estimated tritium concentrations in air or air moisture by more than a factor of two in 1991, 1998, and 1999; in addition, the TDR overestimated HTO concentrations in air moisture by more than a factor of two in 1989 and 1995.

The TDR covers more years than did the test of CAP88-PC, and the observed tritium concentrations in air moisture for 1981 and 2004, (not shown in Table B2), were also overestimated by more than a factor of 2. An attempt has been made below to explain why the over-predictions occurred. Fortunately for a TDR, over-predictions of air concentration (and therefore dose) are health-protective and therefore are only of scientific interest. Over-predictions at VIS, of course, do not affect the conclusions about dose for the period 1973-2005 because dose at the Discovery Center was calculated based on the observed air concentrations.

Table B1. Comparison of release rates (Ci) used for the test of CAP88-PC and the TDR. Only HTO release rates are compared.

	CAP88-PC Test	TDR	
	HTO	**HTO**	**HT**
1989			
Tritium Facility	1555	1620	1350
Sandia TRL	659	659	180
B292	2.40	3.84	
B612 Yard	35.0	59.4	
B331 WAA	----	73.4	
1991			
Tritium Facility	711	696	455
Sandia TRL	351	352	113
B612 Yard	14.2	25.5	
B331 WAA	----	28.4	
1995			
Tritium Facility	62.8	62.8	29.0
Sandia TRL	73.0	73.0	1.06
B612 Yard	2.1	4.17	
B331 WAA	4.0	31.4	
1998			
Tritium Facility	85.1	84.5	25.0
B612 Yard	4.6	8.47	
B331 WAA	6.0	14.4	
1999			
Tritium Facility	213	213	66.8
B612 Yard	4.4	7.61	
B331 WAA	7.3	15.8	

Table B2. Predicted-to-observed ratios of tritium concentrations in air volume (CAP88-PC Test) and in air moisture (TDR) for location VIS. Overestimates of a factor of 2 or more are shown as bold.

	CAP88-PC Test	TDR
1986	1.2	1.1
1987	1.0	1.0
1988	1.8	1.9
1989	1.7	**2.4**
1990	1.7	1.9
1991	**2.4**	**2.4**
1992	1.4	1.4
1993	1.6	1.7
1994	1.8	1.6
1995	1.7	**2.6**
1996	1.2	1.7
1997	1.4	1.6
1998	**2.0**	**2.5**
1999	**2.1**	**2.1**
2000	1.8	1.8
2001	1.6	1.4

1981

The over-prediction in 1981 (P/O = 2.3) may have been due to the inclusion in the annual release rate of two small acute releases of 130 Ci[41] HTO (November) and 450[42] Ci HTO in December from the Tritium Facility's South Stack (#1) (Table A2 shows the assumption that more HTO was released from Stack 1 than from Stack 2 in 1981 while in most years the reverse is true). Therefore, a revised release rate for Stack 1, one that did not include the 580 Ci of HTO that might have been accidentally released, was used in DCART on the assumption that the wind was not blowing towards VIS for the duration of the releases[43]. Because the 580 Ci were not routine releases, the release rates for the

[41] The 130 Ci release is documented (Morris 1981) as having been detected by the Ostlund Monitor but not by the ion chambers. Although the reported data indicate that the release occurred from the South Stack (#1), Morris decided that a handling error had occurred, and the release, if it had occurred, would have come from the North Stack (#2). The fact that the release was modeled here from the South Stack, based on other sources, has very little effect on the dose prediction. Furthermore, in another memo (Morris 1982), it is not clear that the 130 Ci release was from the North Stack.

[42] This release was detected by the Ostlund Monitor and not by the ion chambers (Morris 1982). However, the results of environmental monitoring at the time indicated that the release was most probably real.

[43] This assumption can neither be confirmed nor denied from the ambient air tritium data. The 450 Ci release was presumably detected by the monitors, because all samples for the two-week period in which the release occurred were

Building 331 WAA and the Building 612 Yard were scaled down accordingly. When this was done, the P/O ratio dropped to 1.7. This reanalysis does not solve the mystery, however, because the mean air concentration (Table A10) used for the ratio includes the very high value obtained from the sampling period in which the 450 Ci release occurred. If the mean air moisture concentration for 1981 is calculated without this high value, the revised P/O ratio becomes 2.7. The reason for the over-prediction remains unclear.

1989

It is not clear why the air moisture concentration predicted by the TDR was significantly more over-predicted than was the air volume concentration of the earlier test. The assumptions for the TDR or the earlier test about the fractions contributed by the area sources to total tritium released in 1989 were no different than other years, and the absolute humidity used in the TDR (Table A10) is not out of line with other years. Even in the test of CAP88-PC, however, when predicted concentrations were compared with observed concentrations at twelve perimeter and offsite locations, it was clear that predicted air concentrations were unusually high in 1989. That year the P/O ratios for all locations were higher than other years (the mean P/O ratio for the twelve locations of 1.3 was the highest of the years of the test up until 1997, when the area sources began to have an increased effect on predicted concentrations). Given how well air concentrations were predicted at VIS for most of the years, it appears likely that the reported quantity of tritium released from the Tritium Facility and/or the SNL TRL was higher for some reason than the actual quantity of tritium released. There is no way to resolve the issue.

1991

There were three releases during 1991 that may have occurred when the wind was blowing away from the Discovery Center: 140 Ci of HT were released from Stack 1 as a puff in April, and, during two weekly sampling periods of the year, 55 Ci of HTO were released from each stack of the Tritium Facility. The HT release was a small accident, and the 55 Ci HTO releases were routine emissions, but both the TDR and the earlier test included them as part of the annual release rate. However, even with these releases removed from the total quantity of tritium released from the Tritium Facility, the predicted concentration of tritium in air moisture dropped by no more than 10%. The assumptions for the TDR or the earlier test about the fractions contributed by the area sources to total tritium released in 1991 were no different than other years, and the absolute humidity used in the TDR (Table A10) was not out of line with other years. No explanation has been found for these over-predictions.

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ten or more times higher than the mean concentration for the year without that sampling period. However, all concentrations for the six sampled locations were within a factor of three of each other.

1995

In 1995, the predictions for the TDR were more than two times higher than the observations (2.6) while the test of CAP88-PC only overestimated the observations by a factor of 1.7. The large overestimation by the TDR was caused by assuming a release rate from the Building 331 WAA that was about eight times higher than the release rate used in the earlier test[44]. As a result, the Building 331 WAA became the major contributor to tritium concentrations in air at VIS (Table 5). When 4 Ci instead of 31.4 Ci are used for the Building 331 WAA release rate, the P/O ratio for the TDR predictions becomes 1.7, just like the results of the test of CAP88-PC. The release rate for the Building 331 WAA used in the TDR appears to be overly conservative.

1998

There is no obvious explanation for the over-predictions, which are too great to be accounted for by differences between the long-term wind files and the probable annual meteorological conditions in 1998. In fact, a comparison of air concentrations predicted using the release rates and the dilution factors at VIS derived from using the five-year wind file of the earlier test, the four-year wind file of the TDR, and the NESHAPs wind file for 1998 in CAP88-PC resulted in only about a 2% difference in tritium concentrations in air moisture between the three sets of input.

1999

In 1999, most of the HTO predicted at VIS came from the Tritium Facility's north stack, and 87% of those releases occurred in the first three months of the year. A release pattern like this automatically puts CAP88-PC and its annual mean wind file at a disadvantage because annual wind patterns may easily not represent a seasonal event. In winter (December, January, February), winds towards the ENE (towards VIS) are much reduced compared with the rest of the year (Gouveia and Chapman 1989). Thus the high releases of the first quarter of the year are over-represented when air concentrations are calculated based on annual wind patterns. Both the test and the TDR had P/O ratios just over 2, so the effect is not large.

2004

The over-prediction in 2004 (P/O = 2.2) by the TDR is likely due to having overestimated the release rate from the Decontamination and Waste Treatment Facility (DWTF) stack. Because the DWTF stack was only monitored for released tritium for

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[44] The 31.4 Ci release rate for the Building 331 WAA was estimated based on the mean air concentration observed at VIS for 1995 adjusted for the dilution of the tritium concentration in the sampled air by bound water in the silica gel (Guthrie 2002). The high mean was due to a few weeks of very high concentrations observed at the Building 331 WAA. The mean air tritium concentration in 1995 was 224 pCi m^{-3} (or 358 pCi m^{-3} when corrected) compared with 52.5 pCi m^{-3} for the median (uncorrected) value used to estimate the 1995 release rate of 4 Ci from the Building 331 WAA. The difference between the corrected mean and the uncorrected median results in a factor of 6.8 that nearly accounts for the difference seen in the release rates (7.8); the remainder of the difference should be due to dispersion modeling.

two weeks in November, an annual release rate was estimated based on the mean concentration of HTO measured at the DWTF air tritium sampler, which had been placed 115 m from the stack to intercept some of the plume. A release rate this large from the stack might have occurred, but, based on the two weeks of monitoring in 2004 and monitoring in early 2005, it was not likely. The most likely source of tritium measured at the DWTF air tritium sampler would have been drums of contaminated waste stored outside (as occurred in 2005[45]).

[45] In 2005, an estimated 0.21 Ci was released from waste stored outdoors (Table A7). This small release affected the DWTF air tritium monitor but had no impact on dose to the SW-MEI.

APPENDIX C

REVIEW OF HISTORICAL DOSES MODELED AND REPORTED BY LLNL

All LLNL SAERs were reviewed to obtain the information in this appendix. The descriptions below of the assumptions and models applied each year to the calculation of dose may raise questions because the background detail provided in each SAER, upon which the descriptions are based, was not always sufficiently complete to understand the calculation of the reported doses. Although it was assumed that what was done one year was probably repeated each year thereafter until a change was mentioned in the report, this may not have been the case.

Only dose to the hypothetical individual at the perimeter is reviewed here. LLNL also reported population doses and doses to the nearest resident.

The contributions of each facility to the reported dose from tritium at the site boundary are shown in Tables C1 and C2. In Table C1, the doses for 1973 and 1974, which were calculated without using a named dispersion model, are included with tritium doses predicted by the Continuous Point Source Code. In Table C2, the doses predicted by the Clean Air Act Code are listed. Dose from each facility was to the facility-specific maximally exposed individual (MEI) until 1992,when the concept of site-wide maximally exposed individual (SW-MEI) came into use for NESHAPs compliance. Starting in 1985, doses to an (the?) MEI were added together, but it is not clear whether or not the sum represented the dose to a SW-MEI or was a very conservative dose based on summing doses to the MEIs for all facilities. The doses shown in Tables C1 and C2 are those that were summed to compare with doses predicted by DCART at the Discovery Center (Figures 6 – 9).

One important assumption that affected the dose prediction each year was whether doses were predicted for releases of HTO only (i.e., no HT was assumed released) or whether they were predicted for all tritium released (i.e., releases of HT were assumed to have been HTO). For years when doses were calculated only for releases of HTO, the dose would have been underestimated by a small amount; on the other hand, for the years when all HT was modeled as HTO, the dose consequences would have been over-predicted. The degree by which dose would have been under- or over-predicted is not as important as the fact that doses between years may not be compared easily. Annual assumptions about modeling all releases as HTO or only HTO releases are summarized in Table C3.

Table C1. Inhalation doses (in μSv) at the site boundary for 1973, 1974, and the years when the Continuous Point Source Code was used to calculate doses.

1973	3.0 (calculated from maximum perimeter measured air concentration)				
Source	**B331**	**B212**	**B292**	**Incinerator**	**Lasers**
1974	1.2	7.5	-	-	-
1975	2.0	16.3	-	-	-
1976	2.6	17	-	-	-
1977	2.9	30	-	-	-
1978	2.0	4.1	-	-	-
1979	7.2	3.7	0[a]	-	-
1980	3.9	0.60	0.090	-	-
1981	3.3	0.20	0.20	-	-
1982	0.9	0.40	0.10	-	-
1983	2.7	2.5	1.0	-	-
1984	0.40	0.20	0.10	-	-
1985	0.50	0.00[b]	1.1	-	-
1986	0.40	0.00[c]	0.30	0.0[d]	-
1987	1.2	0.020	0.20	0.0[e]	-
1988	1.0	-	0.0090	0.0090	-
1989	2.6	-	0.0060	0.0	0.00070 [f]
1990	0.99	-	-	-	0.0010
1991	0.62	-	-	-	0.00087

[a] Dose was not calculated for the first year of operations for the Rotating Target Neutron Source

[b] Based on modeling 5 Ci HTO released

[c] Based on modeling 15 Ci HTO from the Insulating Core Transformer

[d] Based on modeling 5.1 Ci HTO from the incinerator

[e] Based on modeling 0.22 Ci HTO from the incinerator

[f] Based on modeling 0.25 Ci HTO from laser complex

Table C2. Inhalation plus ingestion doses (μSv) predicted by the Clean Air Act Code at the site boundary. Doses from 1992 through 2005 were to the SW-MEI.

	B331	B212	B292	Incin-erator	Lasers	B282	B612 Yard	B331 WAA	DWTF
1986[a]	0.30	0[b]	0.070	0.02	-	-	-	-	-
1987[a]	0.70	0.03	0.040	0[c]	-	-	-	-	-
1988	5.5	-	0.040	0.30	-	-	-	-	-
1989	5.2	-	0.019	-	0.002	-	-	-	-
1990	2.7	-	-	-	0.010	-	-	-	-
1991	2.0	-	-	-	0.0050	-	-	-	-
1992	0.37	-	-	-	-	0.016	-	0.0089	-
1993	0.35	-	-	-	-	0.016	0.20	0.038	-
1994	0.27	-	-	-	-	-	0.13	0.041	-
1995	0.17	-	-	-	-	-	0.14	0.059	-
1996	0.45	-	-	-	-	-	0.25	0.031	-
1997	0.75	-	-	-	-	-	0.16	0.017	-
1998	0.23	-	-	-	-	-	0.19	0.039	-
1999	0.67	-	-	-	-	-	0.18	0.061	-
2000	0.084	-	-	-	-	-	0.15	0.044	-
2001	0.043	-	-	-	-	-	0.082	0.0080	-
2002	0.081	-	-	-	-	-	0.11	0.0087	-
2003	0.22	-	-	-	-	-	0.130	0.059	-
2004	0.014	-	-	-	-	-	0.053	0.0017	0.0069
2005	0.026	-	-	-	-	-	0.02	0.012	0.00084

[a] Only inhalation dose was calculated in 1986 and 1987

[b] Based on modeling 15 Ci HTO from the Insulating Core Transformer

[c] Based on modeling 0.22 Ci HTO from the incinerator

Table C3. Species of tritium used to calculate reported doses[a].

	HTO only	Total T
1974 - 1983		X
1984 - 1985	X (B331)	X (other facilities)
1986 - 1988	X	
1989 - 1991		X
1992 - 1998	X	
1999 - 2005		X

[a] For total T, HT is included in the dose calculations as if it were HTO

The choice of dose coefficients also affects dose to a lesser extent. The dose coefficients used by LLNL when reporting doses are summarized in Table C4. In comparison, the dose coefficient used by DCART for inhalation and ingestion of HTO was 1.8×10^{-11} Sv Bq^{-1} (ICRP 1995). Inhalation dose was multiplied by 1.5 to account for skin absorption. DCART also calculated dose from inhalation of HT and ingestion of OBT.

Table C4. Comparison of dose coefficients for HTO (Sv Bq^{-1}) used over the years.

	US NRC 1.109	ICRP 30	US EPA[a]	CAP88-PC[b]
Inhalation/skin	4.27×10^{-11}	1.7×10^{-11} (× 1.5)	1.73×10^{-11} (× 1.5)	3.41×10^{-11}
Ingestion	2.84×10^{-11}	1.7×10^{-11}	1.73×10^{-11}	2.43×10^{-11}

[a] Eckermann et al. 1988

[b] Parks 1992

1973: Inhalation dose from maximum observed air concentration

Doses were first calculated in the 1973 SAER (Silver et al. 1974)[46]. A dilution factor obtained from the Preliminary Safety Analysis Report for Building 332 (PSAR 1974) was used to calculate a mean annual concentration of 1.6×10^{-10} μCi/mL HTO in air volume at the site boundary, assuming all tritium released from the Tritium Facility (the only source modeled) was HTO, but the location was not named. However, even though an air concentration was predicted, the reported dose of 0.3 mrem (3 μSv) was calculated using the highest observed air tritium measurement (1.8×10^{-10} μCi/mL [180 pCi m^{-3} or 6.7 Bq m^{-3}]), which occurred at the air tritium sampling location CAFÉ. CAFÉ was, and is, on the south boundary of the site adjacent to East Avenue. No breathing rate, dose coefficient or other assumptions were mentioned, but in all probability, only inhalation dose was calculated.

1974: Inhalation dose from air concentrations predicted by a meteorological dispersion model

In 1974, a meteorological diffusion model based on Hillsmeir and Gifford (1962) was used to calculate maximum fence-post dose, dose to nearest residence, and collective dose (to 80 km). The location of the fence-post dose (apparently equivalent to LLNL's MEI) was not described and would have been different for each source. Dose was calculated based on the assumption that all 1,900 Ci emitted from the Tritium Facility and the Insulating Core Transformer (ICT) in Building 212 were HTO. The fence line locations for the reported doses were probably to the south of the laboratory. Inhalation dose was probably the only dose calculated.

[46] In 1971 and 1972, the first years of the SAER for the public, environmental concentrations were compared with environmental activity guide levels.

1975 – 1978: Inhalation dose from air concentrations predicted by the Continuous Point Source Code

The method to estimate reported doses was the same between 1975 and 1978. Doses were once again the maximum fence-post dose, but the dispersion model used was the Continuous Point Source (CPS) code (Peterson et al. 1976). The CPS code provided estimates of concentration through sixteen 22.5°compass sectors and at distances between 0.1 and 100 km from the point of release. The code was based upon meteorologically typical data derived from wind direction, wind speed, and variability measured at half-hour intervals. All tritium released was assumed to have been HTO. No assumptions about breathing rate or the dose coefficient used have been found.

1979 – 1983: Inhalation dose from air concentrations predicted by the Continuous Point Source Code

Between 1979 and 1983, dispersion calculations were done using the CPS code, but it had been modified to adjust for topographic differences not accounted for by the earlier version. Another change to the dispersion calculations resulted because meteorological data (wind direction, wind speed, and atmospheric stability class) used to prepare the wind file for the code were collected quarterly each hour from the LLNL meteorological tower, which had monitoring equipment at 10 m and at 40 m. Variance in the horizontal wind direction was used to estimate Pasquill-Gifford stability categories based on the method described by Slade (1968). Lateral and vertical standard deviations, σ_y and σ_z, were entered into the computer code as functions of these stability categories and respective differences. The inhalation dose coefficient was that from the U.S. Nuclear Regulatory Commission's (NRC) Regulatory Guide 1.109 (USNRC 1977) (Table C4)

1979 (Silver et al. 1980) was the first year that Regulatory Guide 1.109 equations and dose coefficients were used to calculate doses from ingestion of vegetation, milk, beef, and drinking water using tritium concentrations in drinking water and vegetation measured by LLNL[47]. Transfer factors were derived that could be multiplied by an observed environmental concentration to directly predict a dose based on ingestion rates. The derivation of these transfer factors appeared in an appendix to each year's SAER from 1979 through 2002. In 2003, the appendix was replaced by a table of transfer factors in the chapter on radiological dose. Except for 1986 – 1991 when doses based on Regulatory Guide 1.109 were published in dose tables for direct comparison with tables of doses predicted using the CPS and Clean Air Act Codes (CAAC) (see below), doses calculated based on Regulatory Guide 1.109 have always been reported with the measured medium, e.g., ingestion doses are calculated and included with the tables of observed concentrations in vegetation.

[47] From 1972 through 1978, ingestion doses from measured concentrations in vegetation had been calculated using the method of Anspaugh et al (1972); doses from inhalation of measured air concentrations had also been calculated from 1974 through 1978, but the method was not stated in the SAERs.

Population dose from inhalation was also calculated in 1979 using air concentrations predicted by the CPS code and the Regulatory Guide's dose coefficient of 1.58×10^{-7} mrem pCi^{-1} (4.27×10^{-11} Sv Bq^{-1}); because this dose coefficient is 1.5 times higher than the dose coefficient used for ingestion, it may be assumed that the value of the inhalation dose coefficient accounts for additional tritium absorbed through the skin. The Regulatory Guide's inhalation rate is 8000 m^3 per year.

All tritium released was modeled as if it were HTO. Dose from each facility was the maximum fence-post dose. The Tritium Research Laboratory at Sandia National Laboratories (SNL/CA) began operations in 1979. SNL/CA tritium releases were never combined with LLNL releases when calculating dose.

1984 and 1985: Inhalation dose from air concentrations predicted by the Continuous Point Source Code

For 1984 and 1985, the CPS code was used with the ICRP 30 (ICRP 1979) dose coefficient for inhalation (Table C4). Meteorological data for 1984 came from the meteorological tower at SNL because the LLNL 40-m sensors were inoperative awaiting relocation to a new site. In 1985, the LLNL 40-m sensors operated for 10 months, and the LLNL data from 10 m was used for the other two months. From records of these data, wind speed, wind direction and stability classes were tabulated at one-quarter hour intervals over the calendar year. At least for the Tritium Facility, doses from releases of HT were not calculated. In 1984, the 5,200 Ci accidental release of HT was not included in dose estimates[48] published in the SAER; similarly, in 1985, doses from routine releases of HT were not calculated for the SAER, nor was the accidental release of 1,000 Ci HT[49]. Ignoring HT releases when the only dose calculated is that from inhalation results in only a tiny underestimation of dose, because the inhalation dose from unit concentration of HT is about a factor of 15,000 less than the inhalation/skin absorption dose from unit concentration of HTO. No releases from SNL were included in LLNL dose calculations. In 1984, the highest fence-line dose was along the south perimeter in an area not routinely occupied. It was subject only to transient traffic; a parking lot was across the street. Doses in 1985 were summed and reported as a total for the first time, implying that dose was being calculated to a SW-MEI rather than to an MEI.

Doses using the Regulatory Guide 1.109 were calculated with dose coefficients very close to those of ICRP 30 (6.23×10^{-8} mrem pCi^{-1} or 1.68×10^{-11} Sv Bq^{-1}) for ingestion[50]. In the appendix describing the derivation of the transfer factors from environmental measurements to dose using Regulatory Guide 1.109, inhalation to the individual was calculated for the first time using an inhalation rate of 8,400 m^3 per year.

[48] No tritium was detected in environmental samples at the perimeter. Urine analyses of employees were all below the lower limit of detection of the analytical method (< 0.02 mrem or 0.2 μSv) (Hill 1984).
[49] Doses of 22 mrem (0.00022 μSv) to the whole body and 185 mrem (0.00185 μSv) to the lung were calculated to a hypothetical receptor at the perimeter (Howe 1985), but they were not reported in the SAER.
[50] This value might have been used in the CPS Code as well, but the documentation does not provide the value. There is only slightly more than a 1% difference between the two values.

1986 through 1991: Inhalation dose from air concentrations predicted by CPS and inhalation/(ingestion) dose from CAAC

In 1986, the Department of Energy required all contractors to calculate air pathway doses using the Environmental Protection Agency's Clean Air Act Code (CAAC) containing the AIRDOS-EPA (Moore et al. 1979) and RADRISK models. The CAAC calculates air concentrations and dose on a sector-averaged basis for 16 sectors. The CAAC not only complied with newly adopted DOE standards, but also with the National Emissions Standards for Hazardous Air Pollutants, Title 40 Part 61 Subpart H (NESHAPs (Office of the Federal Register 1986). The CPS code was also used as in the past to calculate air concentrations from which inhalation doses to the fence post and nearest residence could be calculated; it was also used to calculate population (inhalation) doses.

The CAAC was obtained only shortly before the 1986 SAER was written, so detailed exposure data for the LLNL site were not ready to be used as input to the CAAC; a variety of assumptions and approximations were made to complete the calculations. In 1986 and 1987, doses were calculated using site-specific[51] LLNL data on wind speed and wind direction; data for atmospheric stability was obtained from Oakland Airport records from 1960 – 1964. In 1987, because LLNL had not yet tailored CAAC to site-specific exposure and meteorological parameters, the model was run by Oak Ridge National Laboratory with assumptions and approximations necessary to complete calculations. For both years, the CAAC only calculated inhalation dose. Starting in 1988, the CAAC was used for the first time by LLNL as it was designed to have been used – to calculate both inhalation and ingestion doses. The CAAC's complete radiological dose assessment included each of the environmental transport and exposure pathways for tritium: inhalation/submersion from the air pathway, ingestion of locally grown foods, and ingestion of drinking water[52].

Meteorological data used in the CPS code were obtained from sensors on the LLNL meteorological tower from a height of 40 m. In 1986 and 1987, wind speed, wind direction, and atmospheric stability estimates were tabulated at one-quarter hour intervals over the calendar year from these records[53]. In 1986, the nomenclature changed: "fence-post" doses became known as "site boundary" doses; in 1987, the name changed again to "perimeter" dose. The highest predicted perimeter air concentrations were used to estimate dose.

In April 1988, LLNL began efforts to relocate the Livermore site meteorological tower. This was necessary because of building construction that compromised the horizontal clearance requirements for tower siting. As a result, the two-level 40-m tower was non-

[51] The CAAC requires the use of 10-m data, but the height at which the LLNL data were taken for use in the CAAC was not stated.
[52] In the CAAC it is assumed that the concentration of tritium in drinking water is 1% that of air moisture.
[53] Note that these stability class data used in the CPS code are apparently different from those used in the CAAC for 1986 and 1987.

operational between April through October. A temporary single-level 10-m tower was erected during the downtime. Consequently, only meteorological data from 10-m were collected during this period. By the end of October 1988, the normal 40-m tower was at its new location in the newly acquired land in the northwest portion of the site. From 1989 on, the meteorological data used in the models was site-specific data, but the height above ground (10 or 40 m) at which it was obtained was not specified.

The CPS dose from tritium is the committed effective dose equivalent for 50 years from the tritium inhaled in one year. The CAAC dose from tritium is the committed effective dose equivalent for 50 years from tritium inhaled and ingested in one year.

For these years, two tables in the SAERs presented the doses predicted by the CPS code and the CAAC. In addition, in a third table, inhalation doses predicted using the CPS code and ingestion doses calculated using the Nuclear Regulatory Guide 1.109 were presented and summed. For these calculations, dose to the MEI was defined as the dose that represents credible dose to any "hypothetical" member of the general public. The dose included contributions from all potential exposure pathways and was derived using the most conservative but realistic exposure data and assumptions. The hypothetical person was assumed to reside at the point of highest ground-level radionuclide air concentration continuously for 24 h per day, 365 days per year. During this occupancy period, a portion of this person's diet consisted of locally produced foodstuffs and drinking water containing the highest measured radionuclide concentration. Thus, this was not an actual dose that could have been received by any individual, and it was viewed as a conservative estimate of the highest possible dose to any member of the public. Inhalation and ingestion doses in this table were calculated using dose coefficients from ICRP 30 (Table C4).

1992 – 1998: Inhalation and ingestion dose calculated by CAP88-PC

CAP88-PC (which stands for Clear Air Act Assessment Package-1988) is composed of modified versions of AIRDOS-EPA, for dispersion and transfer through the environment (Moore 1979) and DARTAB, to calculate dose (ORNL 1981)[54]. CAP88-PC differs from the dose assessment software AIRDOS-PC in that it estimates risk as well as dose, it offers a wider selection of radionuclide and meteorological data, it provides the capability for collective population assessments, and it allows the user greater freedom to alter values of environmental transport values (Parks 1992). For compliance with NESHAPs, doses both to the MEI and to the SW-MEI were calculated; dose to the SW-MEI was reported in the SAER. The location of the SW-MEI was determined to be the UNCLE Credit Union just to the east of the laboratory (very close to location VIS where the air tritium sampler was and is). LLNL assumed that 100% of the vegetables and meat consumed by the SW-MEI were contaminated with tritium from the Laboratory but that

[54] CAP-88 also includes the RADRISK module found in the CAAC.

all milk was imported. This assumption was based on the fact that there were no dairy cows in the Livermore Valley during this period.

The dose coefficient used for the Regulatory Guide 1.109 calculations was 6.3×10^{-8} mrem pCi^{-1} or 1.7×10^{-11} Sv Bq^{-1} up through 1997. In 1998, the value was increased slightly to 1.73×10^{-11} Sv Bq^{-1} (Eckerman et al. 1988).

1999 – 2005 – Dose calculated by CAP88-PC

Starting in 1999, the Environmental Protection Agency Region IX required that LLNL model all tritium releases as if they were entirely HTO. Assumptions about the location and diet of the SW-MEI remained unchanged until 2004, when more realistic assumptions were developed for the fraction of diet that is assumed contaminated with tritium. LLNL assumed that 25% of vegetables and 25% of beef were homegrown and that 75% of each was imported; as before, all milk was assumed imported. The effect of this change was to reduce the dose to the individual to 40% of the dose calculated based on the previous, more conservative assumptions.

The inhalation rate used for the Regulatory Guide 1.109 calculations was also changed in 2004. It was reduced from 8400 to 8000 m^3 per year, the original breathing rate given in the Guide, which is more in line with the breathing rate in CAP88-PC (8038 m^3 y^{-1}).

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ABBREVIATIONS, ACRONYMS AND DEFINITIONS

AIRDOS-EPA	The first of the EPA's CAACs (Moore et al. 1979)
CAAC	The EPA's **C**lean **A**ir **A**ct **C**ode
CAP88-PC	The CAAC code used by LLNL for compliance with NESHAPs (Parks 1992)
CPS	Continuous Point Source (code)
CRED	Location of an air tritium sampler near the UNCLE Credit Union
DCART	**D**oses from **C**hronic **A**tmospheric **R**eleases of **T**ritium; a steady-state, stochastic dose model (Peterson 2006)
Dilution Factor	A term that refers to the air concentration for unit source strength (or χ/Q); units are Bq m^{-3} / Bq s^{-1} (or Ci m^{-3} / Ci s^{-1}. The term, although standard for χ/Q, can be misleading, because the higher the dilution factor, the higher the air concentration.
Distribution	A function of a discrete random variable yielding the probability that the variable will have a given value. Types of distributions include
	Lognormal — The probability distribution of any random variable whose logarithm is normally distributed. It can be expressed as a geometric mean and geometric standard deviation. It is commonly used for dilution factors.
	Normal — A theoretical frequency distribution for a set of variable data, usually represented by a bell-shaped curve symmetrical about the mean and is expressed as a mean and standard deviation. Also called *Gaussian distribution*. Source terms are commonly distributed normally.
	Triangular — A distribution with three terms: minimum, likely, and maximum.
	Uniform — A distribution in which all values in the range have an equal probability of being sampled
DT	**d**euterium **h**ydrogen gas
DTO	a form of water in which the hydrogen is replaced by deuterium and tritium
DWTF	Decontamination and Waste Treatment Facility; also an air tritium sampler

ABBREVIATIONS, ACRONYMS AND DEFINITIONS continued

EPA	**E**nvironmental **P**rotection **A**gency
Evap	Evaporation (trays)
HT	Tritiated hydrogen gas
HTO	Tritiated water
Incin	Incinerator
LLNL	**L**awrence **L**ivermore **N**ational **L**aboratory
MEI	**M**aximally **E**xposed **I**ndividual (to releases from one facility)
NESHAPs	**N**ational **E**mission **S**tandards for **H**azardous **A**ir **P**ollutants (40 CFR 61 Subpart H. (National Emission Standards for Emissions of Radionuclides other than Radon from Department of Energy Facilities).
P/O	**P**redicted-to-**o**bserved (ratio)
SAER[i]	**S**ite **A**nnual **E**nvironmental **R**eport
SNL-CA	**S**andia **N**ational **L**aboratory, Livermore, **CA**
SW-MEI	Site-wide Maximally Exposed Individual
TDR	**T**ritium **D**ose **R**econstruction (LLNL, 1953 – 1972, both routine and accidental releases
TRL	Tritium Research Laboratory (SNL/CA)
UNCLE	**UN**iversity of **C**alifornia **L**awrence Livermore Laboratory **E**mployees Credit Union
VIS	Location of an air tritium sampler near the Discovery Center
WAA	**W**aste **A**ccumulation **A**rea

[i] LLNL's annual environmental report has had three different names since 1973. In 1973, it was called "Environmental Levels of Radioactivity in the Vicinity of the Lawrence Livermore Laboratory"; between 1974 and 1987, it was called "Environmental Monitoring at the Lawrence Livermore Laboratory; from 1988 onwards, it has been called simply "Environmental Report". SAER is the term used at LLNL for the "Environmental Report". "ASER" (Annual Site Environmental Report) is the term used at most other Department of Energy facilities. "SAER" will be used in this appendix for any of the LLNL annual environmental reports.