LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-TR-234644

Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL). Part 4. Routine Releases, 1953 - 1972

S-R. Peterson

September 14, 2007

This work was performed under the auspices of the U.S. Department of Energy by University of California, Lawrence Livermore National Laboratory under Contract W-7405-Eng-48.

ABSTRACT

Lawrence Livermore National Laboratory was founded in September 1952. By 1953, operations involving tritium were underway. Annual doses to an adult, a child (age 10), and an infant (age six months to one year) from tritium released routinely from the Livermore site between 1953 and 1972 were calculated using the tritium dose model, DCART. Uncertainties about sources and release rates are high, particularly for the 1950's, and it was difficult, and sometimes impossible (e.g., when a source was only assumed to have existed) to quantify them accurately. Because of this, every effort was made to assure that the uncertainties applied to the input parameters used in DCART would result in doses that could not have been exceeded. Doses were calculated at the potential locations of the hypothetical site-wide maximally exposed individual (SW-MEI), which were at a residence on Vasco Road inside the present west perimeter of the Laboratory (1953 – 1958), at an automotive garage on East Avenue (1961), and at the Discovery Center (1959, 1960, 1962 – 1972, years which predate the facility). Even with the most conservative, screening model assumptions, the highest dose to the SW-MEI (in 1957) was predicted with 95% probability to have been between 27 and 370 μSv (2.7 and 37 mrem), with the most likely dose being 130 μSv (13 mrem). Using more realistic, but still conservative assumptions about what fraction of the diet could have been contaminated, these predictions were reduced by more than a factor of two. All other annual doses (at the 97.5% confidence limits) to the SW-MEI, calculated with the most conservative and health protective assumptions, were less than 200 μSv (20 mrem), and no dose after 1958 could have exceeded 100 μSv (10 mren). The cumulative dose to the hypothetical individual at the west perimeter location for 1953 through 1972 would have been no greater than 860 μSv (83 mrem), while the dose to the individual born and raised there would not have exceeded 1300 μSv (130 mrem) from routine releases of tritium. Cumulative doses at the location of the Discovery Center were less than half of these values.

This page intentionally left blank.

TABLE OF CONTENTS

TABLE OF CONTENTS CONTINUED

TABLE OF CONTENTS CONTINUED

This page intentionally left blank.

INTRODUCTION

This Tritium Dose Reconstruction (TDR) has been undertaken to account for dose to the public from all sources of tritium released to the atmosphere over the lifetime of the Livermore site of Lawrence Livermore National Laboratory; the probability that the doses predicted in this TDR could not have been exceeded is greater than 97.5%. This report, which is Part 4 of "Historical Doses From Tritiated Water and Tritiated Hydrogen Gas Releases to the Atmosphere from Lawrence Livermore National Laboratory (LLNL)[1]" presents the estimated annual doses to the hypothetical site-wide maximally exposed individual (SW-MEI[2]) from routine releases of tritium between the years 1953 (the first year of operations, LLNL being founded in September 1952) and 1972. The location of the SW-MEI (Figure 1) was determined to have been at a potential residence on Vasco Road (Location T) from 1953 through 1958, at an automotive garage on East Avenue (Location Q) in 1961, and at the LLNL Discovery Center (Location VIS) in 1959, 1960, and 1962 through 1972. The LLNL Discovery Center (opened to the public in 1976) is within about 200 m of the UNCLE[3] Credit Union that has been the location of the LLNL SW-MEI since reporting began 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). The Discovery Center (or, specifically, from 1973 on, the location of the VIS air tritium sampler adjacent) was also the location at which air concentrations and doses were calculated for Part 3 of the TDR. A complete set of annual doses to an adult for 1953 through 1972 was predicted for Location T and for Location VIS.

The years 1953 through 1972 were those before tritium concentrations in ambient air were monitored and before monitoring results and release rates were routinely reported in Site Annual Environmental Reports (SAERs). Although not reported to the public, the releases of tritium from significant sources (e.g., both the original Tritium Facility that opened in 1953 in what is now Building 231[4] and the present Tritium Facility [Building 331] that began operations in 1959) were estimated or measured during these years and, starting at least in 1956, were reported quarterly to the United States Atomic Energy Commission (AEC) (see Table 1, TDR Part 2). The quarterly reports to the AEC included accidental, as well as routine, releases.

---

[1] TDR Part 1. Description of Tritium Dose Model (DCART) for Routine Releases from LLNL (Peterson 2006)
TDR Part 2. LLNL Annual Site-specific Data 1953 – 2005 (Peterson 2007a)
TDR Part 3. Routine Releases 1973 – 2005 (Peterson 2007b)
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] 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.

[3] **UN**iversity of **C**alifornia **L**awrence Livermore Laboratory **E**mployees

[4] In this report, all buildings will be referred to by their current numerical designations. Until about 1966, Building 231 was called Building 102, Building 212 was called Building 153, and Building 331 was called Building 172.

The farther back in history, the more difficult it is to assess releases rates with any degree of certainty. Nevertheless, this TDR is the result of such an attempt. The only known documentation of historical routine tritium releases before 1956 is a letter to the AEC (Olsen 1973) in which release rates for the Tritium Facility (i.e., Building 231) and Building 212 were estimated with ± 50% uncertainty. These release rates were obtained by talking with people who had been involved with operations at that time (Myers 2004). Assumptions about release rates between 1953 and 1972 were described in Part 2 of the TDR. Assumptions about the presence of undocumented area sources were also presented in Part 2 of the TDR. It is very likely that area sources existed historically because, operations then, like today, must have generated waste that had to be stored somewhere before disposal. Because area sources of a very few curies may have a relatively large impact on air concentrations and doses at the perimeter of the LLNL site, the existence of such sources has been assumed for this TDR.

Although tritium emits only low energy β-radiation and can only result in a radiological dose if inhaled, absorbed through the skin, or ingested, tritium has historically been a major contributor to dose to the public from LLNL operations because of the magnitude of the releases. Between 1953 and 1972, an estimated 724,000 Ci (26,800 TBq) of tritium, including two large accidental releases of tritiated hydrogen gas (HT[5]) in 1965 and 1970, were released from the Livermore site of LLNL. This quantity is 91% of the tritium that was estimated to have been released by LLNL between 1953 and 2005 (see Table 7, TDR Part 2). The two large accidents contributed about 83% of the total tritium released between 1953 and 2005. Routine operations contributed about 118,000 Ci (4,370 TBq) (Table 1); the 95% confidence interval for this value ranges from 101,000 (3,740 TBq) to 135,000 Ci (5,000 TBq).

The steady-state tritium dose model, DCART (see Part 1 of the TDR) was used to calculate doses from air concentrations calculated from release rates (Tables A1 – A5) and dilution factors[6] (χ/Q in s m^{-3}) (Tables A6 and A7) obtained using the regulatory model, CAP88-PC (Parks 1992) with a four-year wind file compiled from LLNL site-specific meteorological data from 2000 through 2003.

Although DCART is a probabilistic model[7] and all parameters have been assigned distributed values, the extremely conservative assumption was made that the hypothetical adult, child (age 10) or infant (age 6 m to 1 y) exposed to tritium at a certain location lived there 24 hours a day, 365 days a year. To further assure that the calculated dose consequences were conservative, all food for a complete diet (i.e., all vegetables, animal fodder, and animal products) was assumed grown at that location at the same predicted

[5] Throughout the report, gaseous tritium in any form (e.g., T_2, DT, and HT) will be referred to as HT.
[6] The term "dilution factor" will be used throughout this report to refer to the air concentration for unit source strength (or c/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.
[7] 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).

Table 1. Types, sources of tritium, and curies released routinely from LLNL between 1953 and 1972. CL is the confidence limit on the uncertainty.

Type of release	Best estimate (Ci)	2.5% CL (Ci)	97.5% CL (Ci)
Routine HT	74,100	60,900	88,400
Routine HTO	44,100	34,900	54,700
Sources of chronic releases			
HT from Tritium Facilities[a]	72,600	59,400	86,900
HTO from Tritium Facilities[a]	39,900	30,700	50,500
HT from other stacks	1,500	1,090	1,940
HTO from other stacks	205	148	265
HTO from area sources	4,030	3,180	4,960

Note: Because results from each category above involve sampling from different distributions, the 2.5% and 97.5% limits of the output distributions will be different.
Note: An additional 613,000 Ci of HT were released accidentally from Building 331, and an additional 1,450 Ci of HTO were released accidentally from Building 231 (see Part 5 of the TDR).
[a] The term "Tritium Facilities" refers to operations in both Building 231 and Building 331.

mean annual air concentration for all foodstuffs, even though this is physically impossible. Actual doses to any real individual may have been considerably lower than those at the 2.5% confidence limits of the dose calculations primarily because it is assumed in DCART that all vegetables and animal products are grown or raised at the same air concentration that accounts for dose from inhalation, and this is physically impossible. Furthermore, it is highly unlikely that, even in the early 1950's, a local family would have been able to feed themselves entirely on homegrown products[8]. 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).

Annual routine release rates of HT and tritiated water vapor (HTO[9]) from all sources for 1953 through 1972 are summarized in Tables 2a and 2b. Release rates in Table 2a are based on the assumptions given in Part 2 of the TDR; revised release rates in Table 2b are based on alternative assumptions (see below). Both sets of release rates were used in the dose calculations. A detailed accounting of release rates from each facility, including uncertainty distributions, may be found in Part 2 of the TDR or in the Appendix of this report in Tables A1 – A5.

[8] The most obvious example of this would be homegrown grain. Although grain was once a major crop in the Livermore Valley, it would not have been grown for individual consumption in a home garden.
[9] Throughout the report, aqueous tritium in any form (e.g., T_2O, DTO and HTO) will be referred to as HTO.

SUPPLEMENTAL ASSUMPTIONS

The principal assumptions needed for all dose calculations for the TDR were described in Part 2 of the TDR. Two additional sets of assumptions have been added to this TDR to address uncertainties specific to 1953 through 1972.

Revised Release Rates

Before stack sampling began at Building 331 in mid-1961, releases from the Tritium Facility (whether located in Building 231 or in Building 331) were reported as a certain quantity of HT being released on a single date. The release rates (Tables 2a, A1a and A3a) originally prepared for the TDR (Part 2) were based on the assumption that most tritium released was HT because it was released as puffs. Data from stack sampling of HT and HTO emissions support this assumption because the larger the release of HT in a short period of time, the smaller the release of HTO as a fraction of total tritium released. Upon reflection, however, it seems possible that, although essentially all tritium releases from the Tritium Facility were HT puffs released on single dates, the speciation may nevertheless have been similar to the years when stacks were sampled for both HTO and HT. Souers (2004) stated that only HT has been handled in the Tritium Facility. Yet, during years of high release rates, the speciation from the stacks was about 54% HTO and 46% HT[10] (see Part 2 of the TDR) due to conversion of HT to HTO on surfaces and out-gassing of these surfaces. Thus, the alternative assumption - that the speciation from the Tritium Facilities in the years before the stacks were sampled was the same as that when stack sampling began – appears possible and would guarantee conservative dose predictions. Consequently, two sets of input data for the period 1953 – 1960[11] were prepared based on the original (Tables 2a, A1a and A3a) and alternative assumptions (Tables 2b, A1b and A3b) about the possible speciation of the releases.

A change to the assumption about the fraction of total tritium released as HTO also affects the release rate for any area source if that release rate is derived from the quantity of HTO (rather than total tritium) released from the Tritium Facilities. The alternative assumption[12] only affected estimated release rates from the Building 514 Yard between 1953 and 1960[13].

Doses were calculated using both the original and alternative assumptions about speciation of tritium released from the Tritium Facilities. In this report, the term "original assumptions" will mean that, before 1961, releases from Tritium Facilities were mainly HT, while from 1961 onwards, the releases (with the one exception in 1972[10])

---

[10] When releases from each stack were greater than 630 Ci per year, the speciation of 54% HTO and 46% HT was assumed; in 1972, only 506 Ci were released from Stack 1 of Building 331, so it was assumed that 64% of the release was HTO.

[11] In mid-1961, when stack monitoring began, releases were reported quarterly as "controlled releases" to the AEC; prior to that, specific dates of releases had been mentioned. In practice, of course, releases may still have been puffs on specific dates.

[12] In Part 2 of the TDR, it was explained that HTO release rates from the Building 612 Yard, the Building 514 Yard, and the Evaporation Trays from 1962 through 1964 were estimated as 4% of the HTO released from the Tritium Facility.

[13] The other area source that was assumed to have existed during the early years was the Building 231 WAA. Release rates for this were estimated based on total tritium released from Building 231.

were 54% HTO; “alternative assumptions” will mean that releases from the Tritium Facilities prior to 1961 were 54% HTO. The 2.5% confidence limits and means of the doses presented in this report were calculated based on the original assumptions, and the 97.5% confidence limits were calculated based on the alternative assumptions prior to 1961 (from 1961 onwards, there is no difference between the original and alternative assumptions – all releases from the Tritium Facility were assumed to be 54% HTO).

Table 2a. Estimated radioactivity (Ci HT and HTO), released annually between 1953 and 1972 from routine releases from stack and area sources based on original speciation assumptions, used to calculate means and 2.5% confidence limits through 1960 and all doses thereafter.

	HTO from stacks and/or speciated sources				HTO from area sources					HT from stacks and/or speciated sources			
	B231	B212 Room Air	B212-Stack	B331	B231	B514	B331	B612	Evaporation Trays	B231	B212-Room Air	B212-Stack	B331
1953*	540	0.38			33.8	5.40				4860	2.8		
1954	540	1.5			67.5	10.8				4860	11		
1955	540	9.0			135	21.6				4860	66		
1956	530	9.0			100	21.2				3470	66		
1957	610	9.0			300	24.4				11,400	66		
1958	730	9.0			87.5	29.0	34.0			5530	66		
1959		9.0		377		15.1	39.5				66		2820
1960		9.0		171		6.84	80.0				66		3070
1961		9.0		432		17.3	19.8				66		368
1962		9.0		2250			110		82.2		66		2190
1963		9.0		3250			148		119		66		2770
1964		9.0		6750			309		290		66		5750
1965		1.2		1270			58.1	42.2	4.3		8.8		1080
1966		1.2		1650			75.4	56.6	4.3		8.8		1350
1967		1.2	3.3	3570			163	126	4.3		8.8	24	3040
1968			28.8	3650			167	129	4.3			211	3100
1969			16.8	5260			241	189	4.3			123	4480
1970			7.80	2430			111	85.4	4.3			57.2	2070
1971			31.2	1460			67.0	49.5	4.3			229	1250
1972			6.24	780			33.4	24.3	4.3			45.8	570

* There may have been no releases from B212 room air in 1953

Table 2b. Estimated radioactivity (Ci HT and HTO) released annually between 1953 and 1960 from routine releases from stack and area sources to determine the upper confidence limit for dose predictions. Release rates were based on the very conservative assumption that the tritium released from the Tritium Facilities (Buildings 231 and 331) was 54% HTO and 46% HT.

	HTO from stack sources		HTO from area source	HT from stack sources	
	B231	B331	B514	B231	B331
1953	2920		29.2	2480	
1954	2920		58.3	2480	
1955	2920		117	2480	
1956	2160		86.4	1840	
1957	6480		259	5520	
1958	3380		135	2880	
1959		1730	69.1		1470
1960		1750	69.9		1490

Location Of The Site-Wide Maximally Exposed Individual (SW-MEI)

In Part 3 of the TDR, it was determined that the location of the SW-MEI was at Location Q (Figure 1) for 1974 through 1978 and was at the Discovery Center (Location VIS in Figure 1) for the other years. Similarly, but particularly, for this part of the TDR, potential locations for the SW-MEI had to be investigated because a different group of facilities in different locations released the majority of tritium in the early years of operations.

Aerial photographs from the early and mid-1950s were studied to determine the locations of the potential SW-MEI (Figure 1). Any structure near the Laboratory that looked like a residence (a largish building surrounded by trees) was assumed a potential location for the SW-MEI and is marked in Figure 1 by a triangle[14]. These locations were also marked as structures on the 1953 United States Department of the Interior Geological Survey topographic map of the Altamont Quadrangle. In Figure 1, receptors P, R, S, and T were present in the 1950s, as were a few other potential receptors along East Avenue and Vasco Road that were not modeled because they were slightly farther from the facilities releasing tritium. Receptor Q was an automotive garage that was built about 1957. Receptor VIS is the Discovery Center, which was not opened to the public until 1976, although it served as the SW-MEI for conservatism, continuity, and comparison with later dose calculations.

[14] The exception to this is the Discovery Center area, where no structure existed.

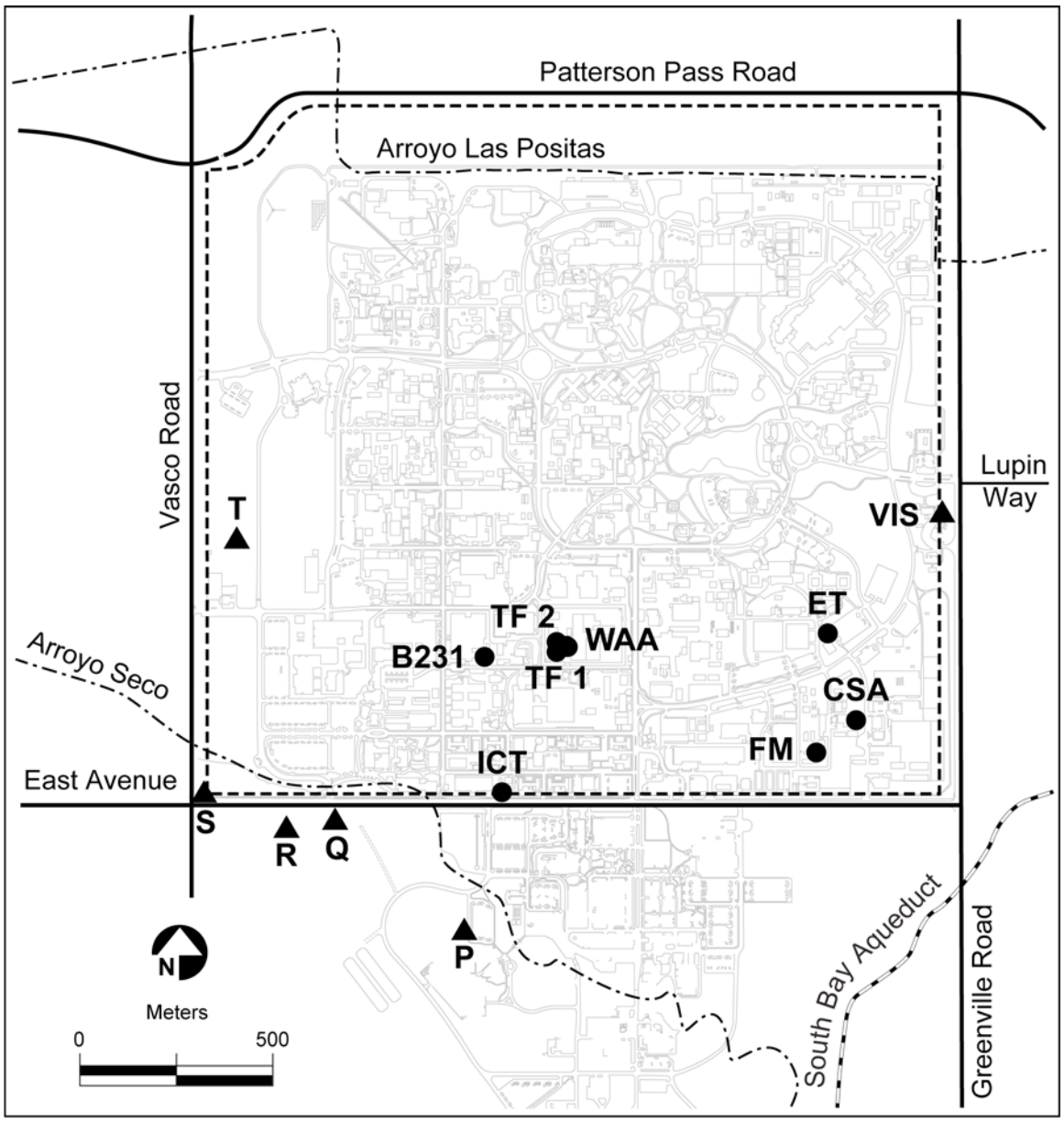

Figure 1. Sources of tritium (●) at LLNL relative to the location of the potential site-wide maximally exposed individual (▲). ▲VIS is location of the Discovery Center; ▲Q was an automotive garage; ▲P, ▲R, ▲S, and ▲T were residences. Modeled sources include TF1 and TF2 (the south and north stacks of the Tritium Facility), WAA (diffuse source at Building 331), B231 (where tritium operations were carried out before 1959, and its presumed WAA), ICT and room air (Building 212), CSA (Building 612 Yard), ET (solar evaporation trays), and FM (the Building 514 Yard).

Distances and directions were estimated to each receptor from each facility, and dilution factors were calculated using the dispersion model in CAP88-PC (Table 3).

Table 3. Directions, distances, and dilution factors from each facility to potential locations of the SW-MEI.

Facility[15]	Location	Direction	Distance in m	χ/Q (s m^{-3})
B231 Stack	Location P	S	683	$2.973 \times 10^{-6} \pm 1.040 \times 10^{-6}$
	Location Q	SW	549	$6.756 \times 10^{-6} \pm 2.552 \times 10^{-6}$
	Location R	SW	646	$6.028 \times 10^{-6} \pm 2.158 \times 10^{-6}$
	Location S	WSW	805	$6.451 \times 10^{-6} \pm 2.357 \times 10^{-6}$
	Location T	WNW	683	$9.399 \times 10^{-6} \pm 4.033 \times 10^{-6}$
B231 WAA	Location P	S	683	$5.363 \times 10^{-6} \pm 2.145 \times 10^{-6}$
	Location Q	SW	549	$1.645 \times 10^{-5} \pm 6.580 \times 10^{-6}$
	Location R	SW	646	$1.213 \times 10^{-5} \pm 4.852 \times 10^{-6}$
	Location S	WSW	805	$1.149 \times 10^{-5} \pm 4.596 \times 10^{-6}$
	Location T	WNW	683	$2.144 \times 10^{-5} \pm 8.576 \times 10^{-6}$
B514 Yard	Location P	WSW	1000	$7.651 \times 10^{-6} \pm 3.060 \times 10^{-6}$
	Location Q	WSW/W	1232	$7.401 \times 10^{-6} \pm 2.960 \times 10^{-6}$
	Location R	WSW/W	1354	$6.345 \times 10^{-6} \pm 2.538 \times 10^{-6}$
	Location S	W	1561	$5.003 \times 10^{-6} \pm 2.001 \times 10^{-6}$
	Location T	WNW	1537	$5.187 \times 10^{-6} \pm 2.075 \times 10^{-6}$
B212 Stack	Location P	S/SSW	366	$4.912 \times 10^{-6} \pm 1.473 \times 10^{-6}$
	Location Q	WSW/W	427	$1.349 \times 10^{-5} \pm 4.046 \times 10^{-6}$
	Location R	WSW/W	561	$1.251 \times 10^{-5} \pm 3.753 \times 10^{-6}$
	Location S	W	768	$1.080 \times 10^{-5} \pm 3.240 \times 10^{-6}$
	Location T	NW	915	$5.868 \times 10^{-6} \pm 1.760 \times 10^{-6}$
B212 Room	Location P	S	366	$1.288\ 10^{-5} \pm 4.508\ 10^{-6}$
	Location Q	WSW/W	366	$4.087\ 10^{-5} \pm 1.430\ 10^{-5}$
	Location R	WSW/W	512	$2.575\ 10^{-5} \pm 9.013\ 10^{-6}$
	Location S	W	707	$1.765\ 10^{-5} \pm 6.178\ 10^{-6}$
	Location T	NW	854	$8.479\ 10^{-6} \pm 2.968\ 10^{-6}$

[15] Dilution factors were also calculated for the "Sunshine Building" (the large structure due south of Location T in Figure 1). Differences between the dilution factors at the Sunshine Building and Location T were slight, and doses were always higher at Location T.

Table 3 continued

Facility	Location	Direction	Distance in m	χ/Q (s m^{-3})
B331 South	Location P	SSW	732	$6.620\ 10^{-7} \pm 1.986\ 10^{-7}$
	Location Q	SW	707	$7.417\ 10^{-7} \pm 2.225\ 10^{-7}$
	Location R	SW/WSW	829	$5.296\ 10^{-7} \pm 1.589\ 10^{-7}$
	Location S	WSW	976	$3.910\ 10^{-7} \pm 1.173\ 10^{-7}$
	Location T	WNW	866	$3.332\ 10^{-7} \pm 9.997\ 10^{-8}$
B331 North	Location P	SSW	780	$5.466\ 10^{-7} \pm 1.647\ 10^{-7}$
	Location Q	SW	732	$5.259\ 10^{-7} \pm 1.887\ 10^{-7}$
	Location R	SW/WSW	854	$4.258\ 10^{-7} \pm 1.259\ 10^{-7}$
	Location S	WSW	1000	$2.850\ 10^{-7} \pm 8.714\ 10^{-8}$
	Location T	WNW	854	$1.795\ 10^{-7} \pm 5.912\ 10^{-8}$
B331 WAA	Location P	SSW	780	$5.460\ 10^{-6} \pm 1.638\ 10^{-6}$
	Location Q	SW	744	$9.314\ 10^{-6} \pm 2.794\ 10^{-6}$
	Location R	SW/WSW	854	$8.742\ 10^{-6} \pm 2.622\ 10^{-6}$
	Location S	WSW	1000	$7.651\ 10^{-6} \pm 2.295\ 10^{-6}$
	Location T	WNW	866	$1.375\ 10^{-5} \pm 4.125\ 10^{-6}$
Evaporation trays	Location P	SW	1183	$4.053\ 10^{-6} \pm 2.026\ 10^{-6}$
	Location Q	WSW	1329	$4.797\ 10^{-6} \pm 2.399\ 10^{-6}$
	Location R	WSW	1463	$4.099\ 10^{-6} \pm 2.050\ 10^{-6}$
	Location S	WSW	1646	$4.596\ 10^{-6} \pm 2.298\ 10^{-6}$
	Location T	W	1512	$5.279\ 10^{-6} \pm 2.640\ 10^{-6}$
B612 Yard	Location P	WSW	1171	$5.888\ 10^{-6} \pm 1.766\ 10^{-6}$
	Location Q	WSW/W	1366	$5.399\ 10^{-6} \pm 1.620\ 10^{-6}$
	Location R	WSW/W	1488	$4.695\ 10^{-6} \pm 1.409\ 10^{-6}$
	Location S	W	1707	$4.324\ 10^{-6} \pm 1.297\ 10^{-6}$
	Location T	W/WNW	1634	$4.669\ 10^{-6} \pm 1.401\ 10^{-6}$

To determine the location with the highest dose each year (the location of the SW-MEI), DCART was used to calculate a deterministic dose using the dilution factors specific to each of the six receptors (the five shown in Table 3 and the Discovery Center [see Table A7]) and the alternative speciation assumptions for the release rates. Doses for all years were calculated. For each year, the receptor locations were ranked from high dose

to low dose. Between 1953 and 1958, the SW-MEI was determined to be at Location T (Figure 1). In 1959 and 1960, Location VIS (the Discovery Center) had the highest deterministic dose using the alternative assumptions, but in 1961, the highest predicted dose occurred at Location Q. From 1962 through 1972, the location for the highest potential dose was again at Location VIS. During the years when the dose at Location T was the highest of all locations, the dose at Location VIS was the lowest. When Location VIS became the SW-MEI, the dose at Location Q ranked second, and the dose at Location T ranked either third or fourth of all locations examined.

Doses for the location with the highest dose each year based on the alternative assumptions prior to 1961, i.e., the location of the SW-MEI for one particular year, were calculated probabilistically. Because both Location T and Location VIS were locations of the SW-MEI for several years, a complete set of doses (1953 through 1972) was calculated for both locations.

Acute Releases with Minimal Dose Impact[16]

There were two acute releases of tritium between 1953 and 1972 that have been modeled as routine emissions. The first, an experimental release of 800 Ci HT in 1969, was probably included in a report of routine emissions (Otsuki 1970). The second was 240 Ci, 135 of which were HTO, released accidentally in 1971.

Experimental release modeled as routine

On October 27, 1969 at 10:30 am, 800 Ci of HT were released over the course of an hour from the south stack of Building 331 as part of an experiment to determine how well downwind air concentrations could be predicted and to estimate the conversion rate of HT to HTO (Silver 1970). HT samplers and cold fingers to trap HTO were placed along an arc 1000 m from Building 331. The quantity of HT released measured only about 30% above the detection limit at 1000 m, but it was felt that the agreement between predicted and observed HT concentrations in air was reasonable (with a predicted to observed [P/O] ratio of 0.5). Although variations in tritium concentrations along the sampling arc were too great to permit a definite statement about conversion rates from HT to HTO, the rate was reported as less than 10%.

The highest HTO concentration observed was 926 pCi m^{-3} (34.3 Bq m^{-3}). This experimental release, being no different in magnitude than many "routine" puff releases, was modeled as a routine release. Although the release was known to be 100% HT, it was modeled as 432 Ci HTO and 368 Ci HT (based on the assumption applied to all

[16] In addition to these small acute releases, there were four or five accidents that would have had dose consequences to a member of the public. On October 13, 1954, 1,450 Ci (53.7 TBq) of HTO were released from Building 231; on January 20, 1965, 350,000 Ci (13,000 TBq) of HT were released from Building 331; on April 7, 1966, 11,000 Ci (410 TBq) of HT were released from Building 331; and on August 6, 1970, 290,000 Ci (11,000 TBq) of HT were released from Building 331. In addition, there may have been a release of 24,000 Ci (8900 TBq) of HT in 1964. All accidental releases will be described and doses for them will be predicted in Part 5 of the TDR.

releases between 1961 and 1972 that 54% of the tritium released was HTO). The dose predicted for this release was thus higher (and more health protective) than it would have been had it been modeled as HT.

Small accidental release modeled as routine

On April 12, 1971, about 21 Ci of tritium were released at 9:05 am followed by a release of about 200 Ci at 9:40 am. About five hours later, 16 Ci more were released under controlled conditions. Stack sampling indicated that about 135 of the 240 Ci released were HTO. At the time of the release, the atmosphere was characterized by strong incoming solar radiation, winds from the north to northeast at 1.8 to 4.5 m s^{-1} and unstable conditions (Yoder 1971).

Extensive environmental monitoring followed over the next three days. The highest concentration of HTO in air (13,000 dpm m^{-3} or 220 Bq m^{-3}) was measured using a cold finger between 11:00 am and noon on-site near Building 415. A vegetation sample collected closer to the Tritium Facility had a concentration of 926 dpm mL^{-1} (15,400 Bq L^{-1}); collection time was unspecified. Although a couple of high measurements in vegetation were found off-site, based on the pattern of concentrations, the values are questionable. The highest credible off-site concentrations of HTO in vegetation (time unspecified) would have been about 100 - 200 dpm mL^{-1} (1,700 – 3,400 Bq L^{-1}). No tritium was detected in two urine samples taken from the public downwind of the release. All concentrations were well below off-site maximum permissible concentrations (MPC) at the time[17].

An inhalation dose of 0.15 mrem (1.5 μSv) was calculated to a hypothetical individual standing on the centerline of the plume at the nearest downwind off-site location. A dose to a child of 6 mrad was calculated[18] on the conservative assumption that the child drank milk from cows grazing on the most highly contaminated grass.

Because of the high uncertainty associated with modeling acute releases, and because this release, although accidental, was relatively small compared with total routine releases (2,700 Ci) in 1971, it was included as part of the annual estimated routine releases for 1971.

DOSE PREDICTIONS

Doses to the SW-MEI

Location T was the SW-MEI from 1953 to 1958. In Figure 2, the 2.5% confidence limit, the mean, and the 97.5% confidence limit of doses predicted using the original assumptions are shown for Location T for all years modeled for this part of the TDR. In

[17] 6,600 dpm mL^{-1} (1.1 10^5 Bq L^{-1}) for vegetation and 4.4 10^5 dpm m^{-3} (7.3 10^3 Bq m^{-3}) for air. Myers (2006) explains that, because there was no specific MPC for vegetation, the MPC for water was used.

[18] The Ng et al. (1968) model was used for this calculation; it was the same one used by Myers et al. (1973) to calculate dose after the August 6, 1970 HT release.

addition, for 1953 through 1960, the upper confidence limits of the dose predictions that were calculated using the alternative assumptions are shown. The upper confidence limit calculated from the alternative assumptions is always the higher of the two upper confidence limits because of the higher dose consequences of a release of HTO.

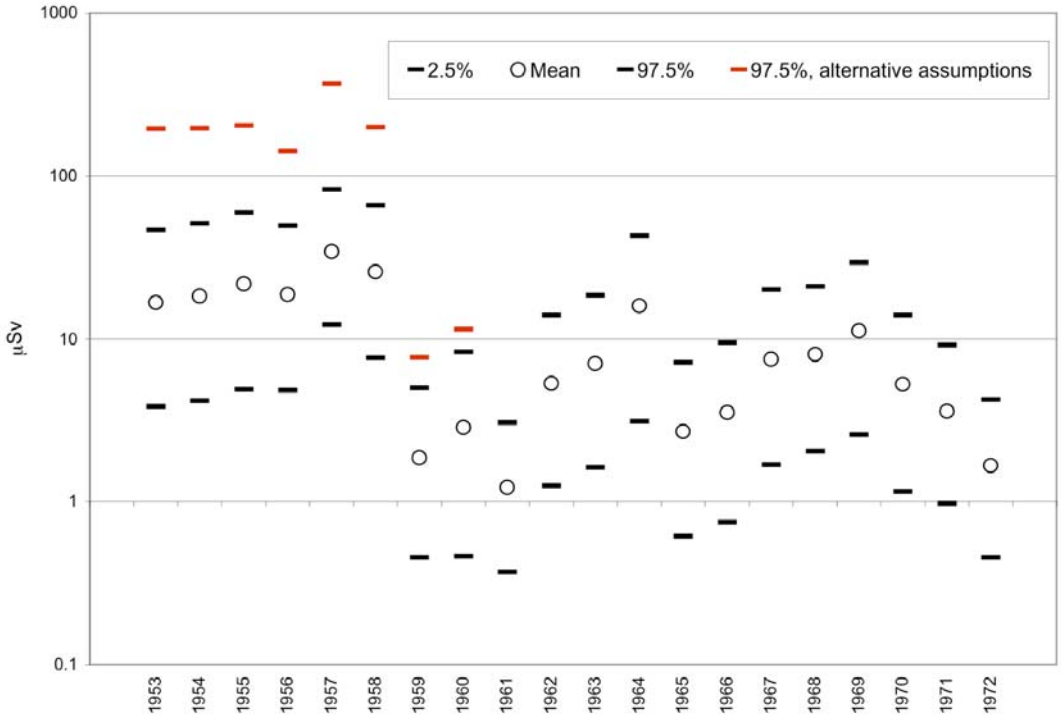

Figure 2. Annual mean doses with 95% confidence intervals predicted by DCART to an adult from routine releases at Location T. Prior to 1961, both the upper confidence limits calculated from original and alternative assumptions about speciation are shown.

Release rates used to calculate doses predicted from the original assumptions are shown in Table 2a; release rates used to calculate the upper confidence limits of doses predicted from the alternative assumptions between 1953 and 1960 are shown in Table 2b. The likely doses that would have been received by the hypothetical SW-MEI are assumed to lie between the 2.5% confidence limit and the higher of the two upper confidence limits. Because of this approach, the uncertainty (higher of the two upper confidence limits divided by the lower confidence limit) on the 1953 through 1960 predictions is much greater (on average, a factor of 33) than on the predictions from 1961 through 1972 (on average, a factor of 11). The greatest uncertainty (a factor of 51) occurred in 1953. The highest dose to an adult (at the 97.5% confidence limit) was 370 µSv (37 mrem) in 1957. All mean annual doses were less than 35 µSv (3.5 mrem). The median dose for the twenty-year period was 7.2 µSv (0.72 mrem).

In Figure 3a, the mean predicted doses at Location T attributed to each facility are plotted on a log scale for each year. The same predictions are plotted in Figure 3b as fractions of the total dose attributable to each facility. Predictions in both figures were calculated based on the original speciation assumptions. Until 1958, the Building 231 stack contributed more than 65% of the dose received at Location T. From 1959 through 1972, the Building 331 Waste Accumulation Area (WAA) was the most important contributor to dose at Location T, providing on average about 60% of the total dose. Releases from Building 212 contributed more than 5% of the total dose for six years.

In 1959, 1960, and 1962 onwards, the SW-MEI was at Location VIS. Dose predictions at Location VIS for routine releases between 1953 and 1972 are compared in Figure 4. Two sets of 97.5% confidence limits are shown for 1953 through 1960. The lower of the two upper limits represents calculations based on the original assumptions about speciation (Table 2a), while the higher of the two upper limits was calculated using the alternative assumptions (Table 2b). The 2.5% confidence limits and means shown were calculated using the original assumptions, as were the 97.5% confidence limits for 1961 through 1972. The true doses that would have been received by the hypothetical individual living at Location VIS are assumed to lie between the 2.5% confidence limit and the higher of the two upper confidence limits. Because of this approach, the uncertainty (higher upper confidence limit divided by lower confidence limit) on the 1953 through 1960 predictions is much greater (on average, a factor of 31) than on the predictions from 1961 through 1972 (on average, a factor of 7.3). The greatest uncertainty (a factor of 41) occurred in 1953. The highest dose (at the 97.5% confidence limit), was 94 μSv (9.4 mrem) in 1957. Mean doses exceeded 10 μSv (1 mrem) for only five years, and the highest mean dose was just 29 μSv in 1964. The median dose for the twenty-year period was 6.0 μSv (0.60 mrem).

In Figure 5a, the mean predicted doses at Location VIS attributed to each facility are plotted on a log scale for each year. The same predictions are plotted in Figure 5b as fractions of the total dose attributable to each facility. Predictions were calculated based on the original speciation assumptions. Clearly, the early Tritium Facility (Building 231) was the most important source of tritium at Location VIS in the early years, contributing more than 70% of the predicted dose. In later years, the present Tritium Facility (Building 331) was the most important contributor to dose at Location VIS, but it only contributed about 60% of the total dose; the Building 612 Yard contributed about 30% of the dose. Releases from Building 212 never contributed more than 4% to the dose at Location VIS.

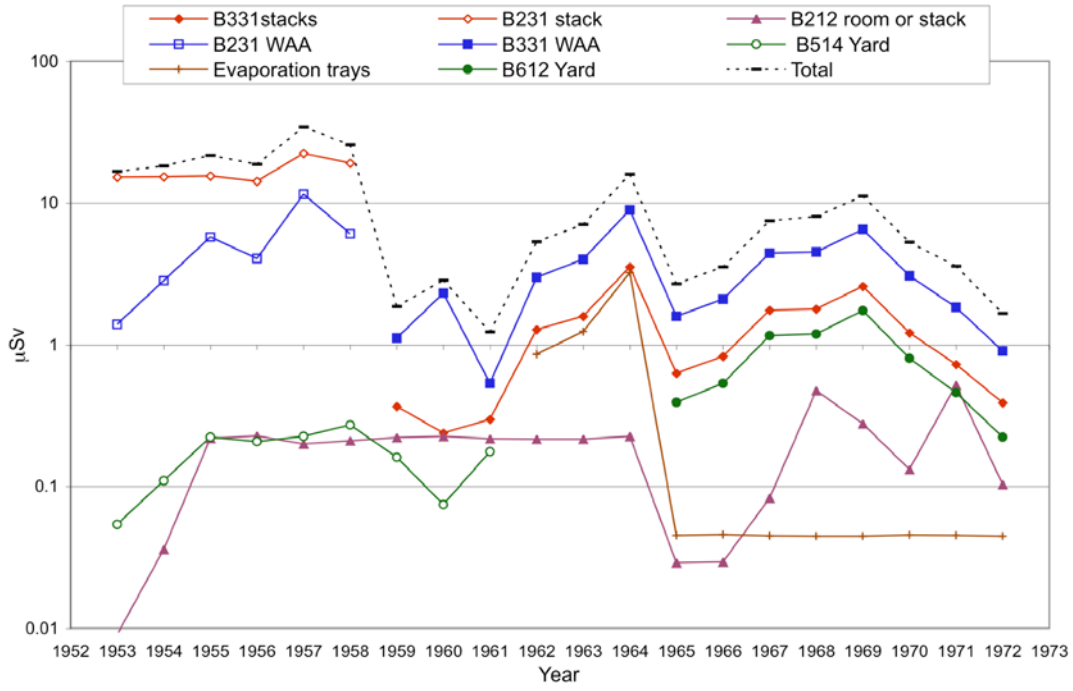

Figure 3a. Annual mean doses from each facility predicted by DCART to an adult at Location T using the original assumptions.

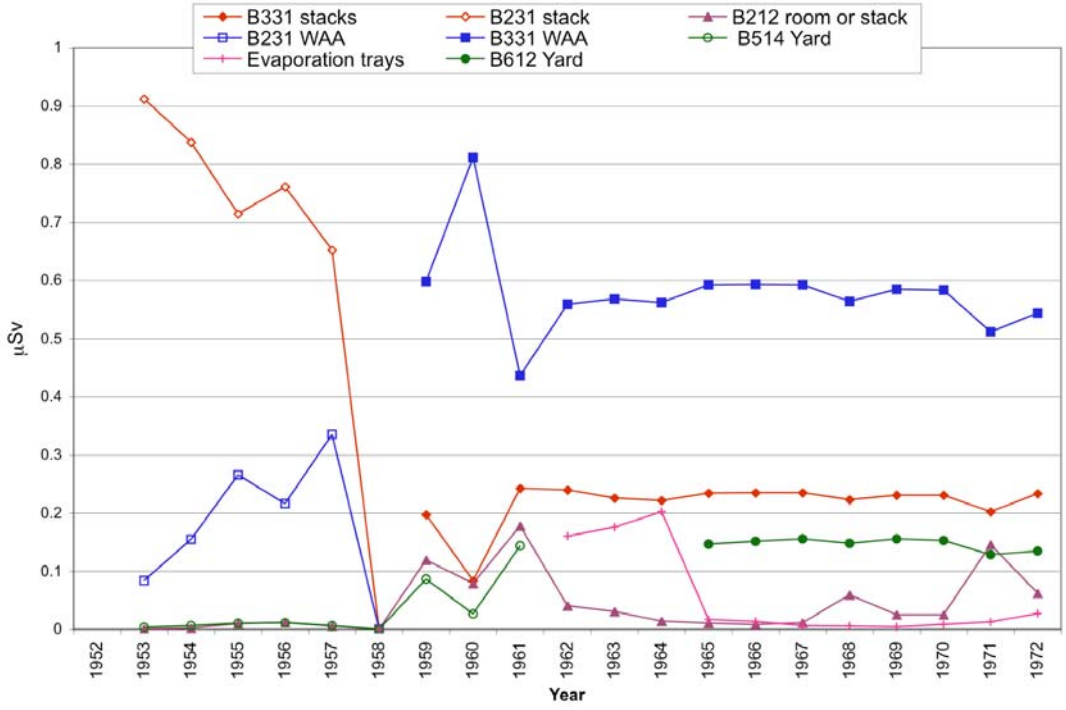

Figure 3b. Fraction of total dose from routine releases contributed by each facility predicted by DCART at Location T using the original assumptions.

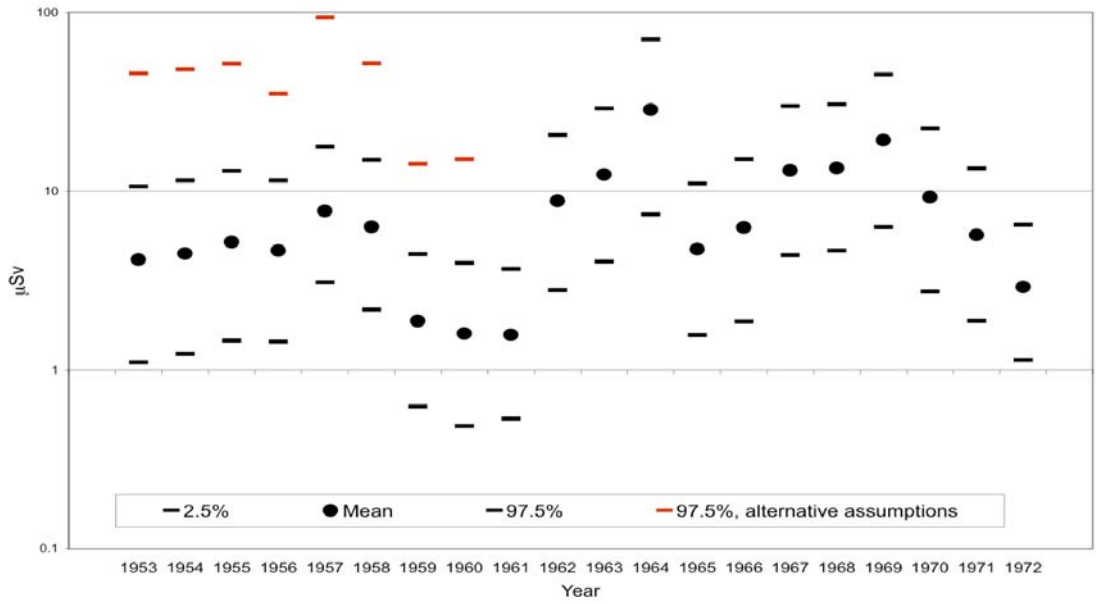

Figure 4. Annual mean doses to an adult from routine releases with 95% confidence intervals predicted by DCART at Location VIS. Prior to 1961, both the upper confidence limits calculated from original and alternative assumptions about speciation are shown.

In 1961, the SW-MEI was determined to be at Location Q (Figure 1). The dose at the 97.5% confidence limit was 5.4 µSv (0.54 mrem); this value is 48% higher than the comparable dose at Location VIS and 78% higher than the dose predicted at Location T.

Doses predicted for 1953 through 1972 for the locations of the SW-MEI (Location T, Location Q, and Location VIS) are combined in Figure 6 so that the dose relationship between the locations can be seen easily. Only the doses for Location T and Location VIS are graphed for the entire time period because they were the locations exposed to the greatest overall doses. The lower confidence limits and the means calculated using the original assumptions, and the 97.5% confidence limits calculated assuming 54% of the tritium released from the Tritium Facility was HTO, are plotted. Between 1953 and 1958, given the large uncertainty in the predictions, the doses predicted at Location T and at Location VIS overlap, but the doses at Location T are about a factor of four higher than those at Location VIS, with the mean dose at Location VIS being essentially the same as the lower limit of the output distributions for Location T. In 1959, and 1960, doses were essentially indistinguishable between Location VIS and Location T. Although upper confidence limits demonstrate that Location VIS was the location of the SW-MEI, the mean doses were the same for Location VIS and Location T in 1959, and in 1960, the mean dose at the Location T was actually higher than at Location VIS. In 1961, Location Q (the garage) edged out the Discovery Center and Location T to become the SW-MEI. Doses in 1961, especially when uncertainty is taken into account, were the lowest for all years. Doses at Location VIS were on average less than a factor of two higher than doses at Location T for the years when Location VIS was the SW-MEI.

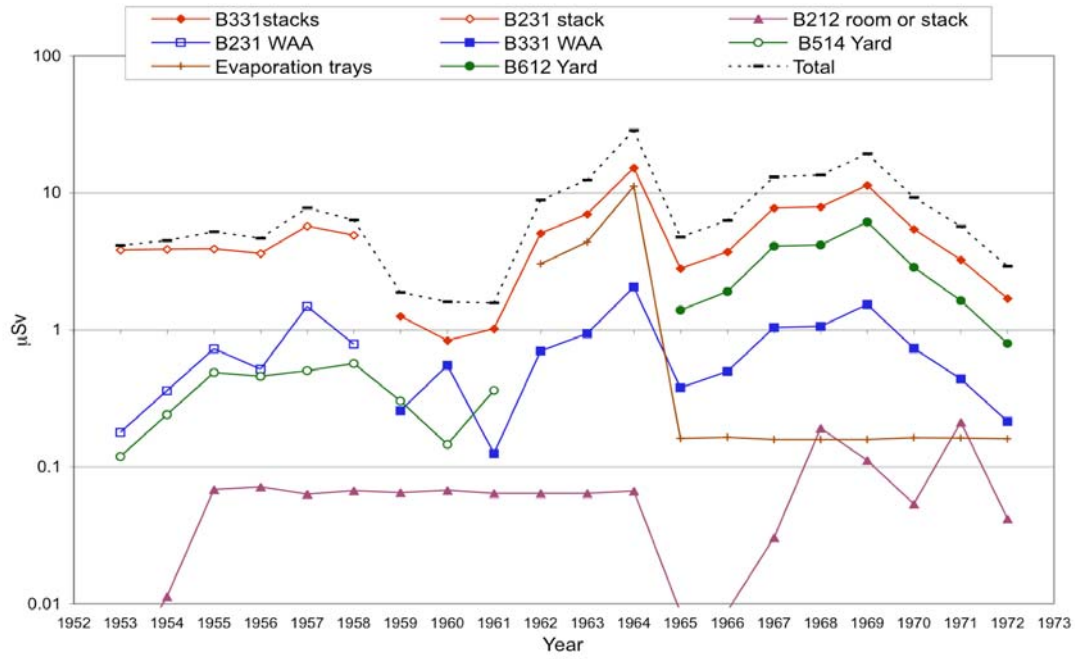

Figure 5a. Annual mean doses to an adult from routine releases by facility predicted by DCART at Location VIS using the original assumptions. Doses have been truncated at 0.01 µSv.

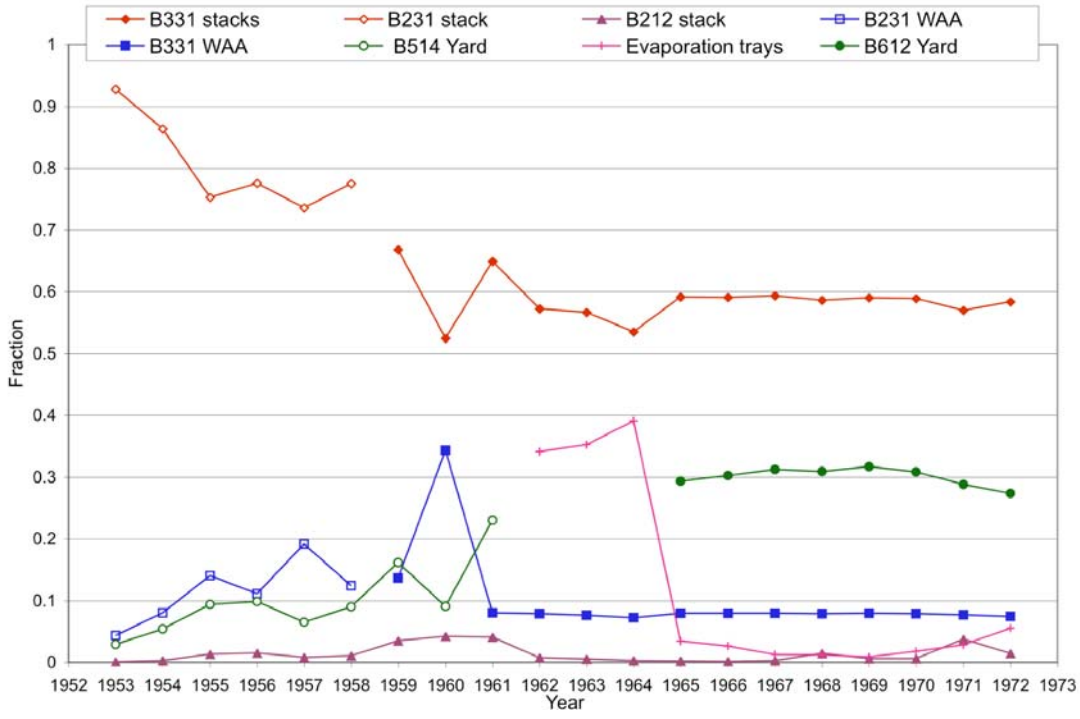

Figure 5b. Fraction of total dose from routine releases contributed by each facility predicted by DCART at Location VIS using the original assumptions.

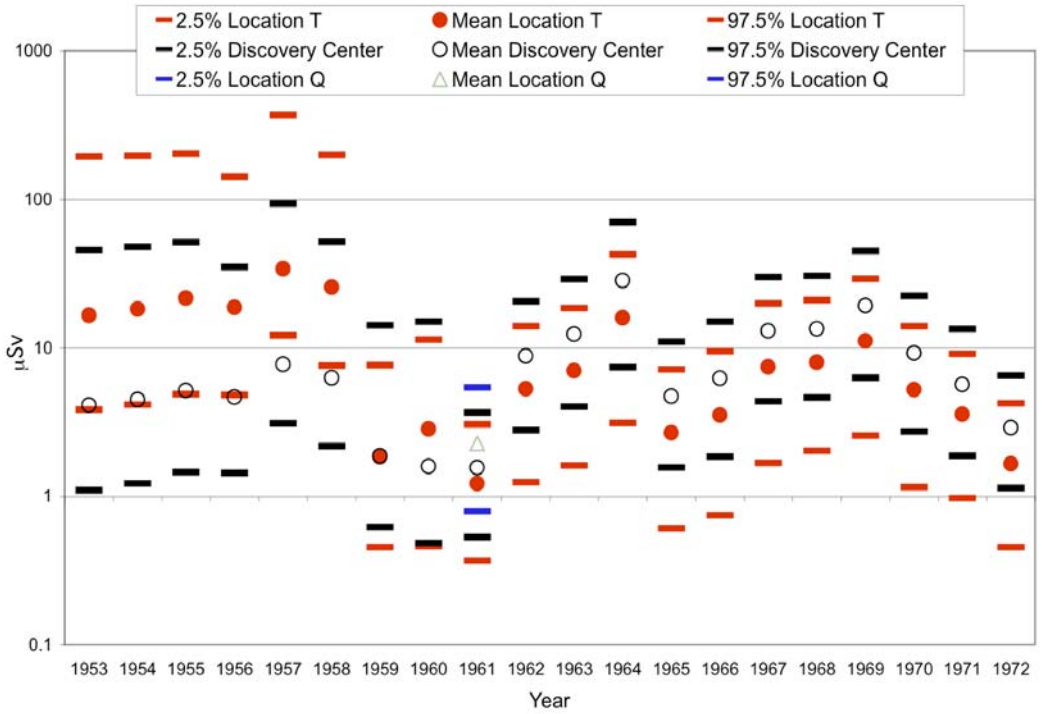

Figure 6. Annual mean doses to an adult from routine releases predicted by DCART at locations of the SW-MEI: 1953-1958, Location T; 1959, 1960, 1962 – 1972, Location VIS; 1961 Location Q. Through 1960, the upper confidence limit was calculated from the alternate speciation assumption.

Relative Importance of Doses

Adult, child, infant

All doses were calculated to an adult, child (age 10) and infant (6 months to 1 year). In DCART, the mean tritium dose to an infant is about 42% higher than that to an adult, and the dose to a child is about 22% higher than that to an adult. The highest possible annual dose (97.5% confidence limit) predicted by DCART to adult, child, or infant was 590 µSv (59 mrem) to an infant at Location T in 1957 assuming the alternative speciation assumptions.

The dose from inhalation calculated by DCART is higher for a child than for either an adult or an infant. The child's inhalation dose is 30% higher than an adult's because the child's inhalation rate is slightly higher than the adult's and the inhalation dose coefficient is also higher; the infant's inhalation dose is 10% less than an adult's because, although the dose coefficient is 2.4 times higher than an adult's, the breathing rate is nearly three times less. As modeled by DCART, the inhalation dose relative to the total tritium dose is about 20%, 19%, and 11% for adult, child, and infant, respectively. These percentages are obtained when more than 50% of the released tritium is assumed to be HTO; when HT releases dominate, these percentages are reduced by about 2%.

In DCART, the child's ingestion dose is about 40% higher than the adult's, and the infant's is about 70% higher than the adult's because of lower ingestion rates and higher dose coefficients for the child and infant. A very small effect due to speciation is seen when the ingestion dose to an infant is compared with an adult's: when it was assumed that 54% of the stack releases were HTO, the infant/adult ingestion dose ratio is about 2.5% higher than when most of the stack releases were assumed to be HT.

That the entire diet could have been contaminated to the same extent as the air concentration at the predicted point (1 m^3) 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 1957 (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 rectangular distribution of 0 – 1 (or 0% - 100%). The inhalation rate was adjusted to reflect occupancy of the SW-MEI with a rectangular distribution of 0.5 – 0.9 (50% - 90%)[19]. Consumption of contaminated grain was changed from 100% to 0% for both people and animals, because grain, although grown in the Livermore Valley, was not grown as close to the Laboratory as the locations of the SW-MEI and was most certainly not grown in the garden of the hypothetical SW-MEI. Using the revised assumptions, the total mean doses to an adult, child, and infant were reduced to about 40% the mean doses obtained using a completely contaminated diet; dose to an infant was reduced to about 45%. The 97.5% confidence limit was reduced slightly less – about a factor of two.

Cumulative dose 1953 – 1972

The total probable dose that could have been received by a hypothetical individual was calculated for Location VIS and for Location T because they were essentially the only two locations of the SE-MEI (Figure 7). The doses were calculated by using Crystal Ball® to sum the distributions for the mean dose for each year. Two sets of sums were calculated: the first set used means and their distributions obtained from the original assumptions; the second set used means and distributions obtained from the alternative assumptions for the years prior to 1961 and means and distributions from original assumptions for the remaining years[20]. The means and 2.5% confidence limits (Figure 7) were obtained from the sum of the first set, and the 97.5% confidence limits were obtained from the second set. Doses were estimated for an individual born in 1953 that grew up at either location and lived there through 1972 and for an adult who lived at either location for the same twenty years. Because DCART only calculates dose to three age groups (infant, child, and adult), it was assumed conservatively[21] that the infant born in 1953 received an annual dose based on an infant dose coefficient and infant intake until age 10, an annual dose

[19] Lognormal distributions representing the occupancy factor were calculated using Crystal Ball® to be 3,400 ± 1,250 m^3/y for the adult, 3,450 ± 1,600 m^3/y for the child, and 1,150 ± 822 m^3/y for the infant.
[20] The input data for the both sets of calculations was identical for the years after 1960.
[21] The magnitude of the dose coefficient decreases with age and size, and infant doses are highest even with low intake.

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 two years.

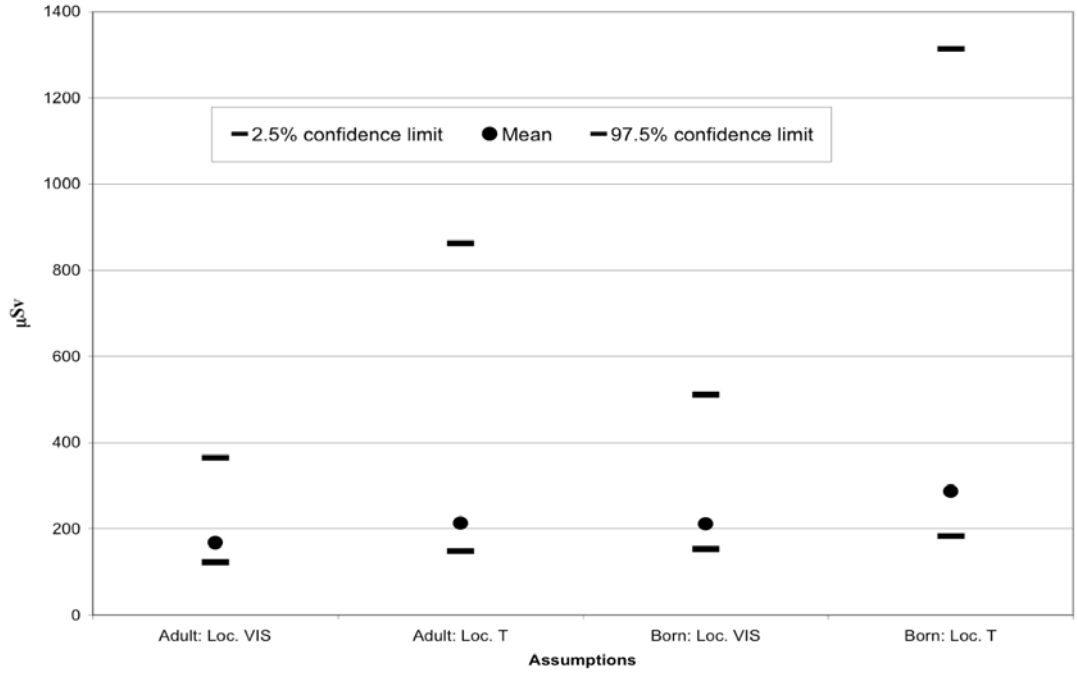

Figure 7. Cumulative doses and 95% confidence intervals (1953 – 1972) predicted by DCART to adults and individuals born and raised at Location VIS and at Location T.

The cumulative dose predicted with 97.5% confidence (1953 – 1972) from routine releases to the hypothetical adult who lived at Location T was 860 µSv (86 mrem); dose to a hypothetical individual who was born and grew up at Location T was 1300 µSv (130 mrem). The upper confidence limits for dose at Location T averaged 4.3 times higher than the mean doses.

Similar cumulative doses were calculated for the hypothetical individual who either lived as an adult at Location VIS (360 µSv [36 mrem]) or was born and grew up there (510 µSv [51 mrem]) (Figure 7). These doses at the 97.5th percentile are a factor of about 2.3 times higher than the mean doses.

Dose contribution of HT

Obviously, the higher the fraction of total tritium released that is HT, the higher the dose impact from HT. Between 1953 and 1960, assuming most of the tritium released was HT, at Location VIS, HT contributed 29% of the mean dose from tritium to an adult, on average; at Location T, HT contributed 29% for 1953 – 1958 (and 24% for 1953 – 1960). The highest fractions of dose contributed by HT occurred in 1957 – 40% at Location VIS and 36% at Location T. For the years when it was assumed that releases were 54% HTO, the contribution of HT to mean predicted dose to an adult averaged 3.3% at Location VIS and 2.3% at Location T. Maximum contributions from HT to dose (assuming that 54%

of the Tritium Facility release was HTO) occurred in 1961: 4.6% at Location VIS and 6.8% at Location T. In 1961 at Location Q, the contribution of HT to mean dose was 16%.

Sensitivity

With the exception of release rates and dilution factors, 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 to adult, child, and infant was examined (all parameter values varying) for the years 1953 (the year with the lowest fraction of total release being contributed by area sources and the Tritium Facility was at Building 231), 1957 (the year with the highest fraction of total release being contributed by area sources and the Tritium Facility was at Building 231), 1961 (the year when stack releases from Building 331 were a typical fraction [48%] of total releases, the releases from Building 212 were their highest, and the contribution of area sources was low), 1964 (the year when area sources, with evaporation trays interim between the Building 514 and 612 Yards, contributed the largest fraction of total release and when the Tritium Facility was at Building 331), and 1970 (the year that both area sources and the Tritium Facility were important). Because sensitivity to dose was expected to be receptor-dependent, sensitivity analyses were carried out for both Location T and Location VIS. The values obtained from the sensitivity analyses were ranked for both locations for both for original and alternative assumptions, if appropriate, by correlation coefficient for each year to obtain the four parameters to which the endpoint, dose, was most sensitive.

Results are shown in Tables 4a and 4b for Location T and in Tables 5a and 5b for Location VIS. The “a” and “b” tables look at the sensitivity of dose to various parameters from two different perspectives. In the “a” tables, the sensitive parameters that are shared across years are shown; in the “b” tables, the sensitive parameters that are common to adult, child, and infant are shown. Between the “a” and “b” tables, a complete set of the parameters to which dose is sensitive is demonstrated.

Unlike in the dose reconstruction for the years 1973 – 2005 (Part 3 of the TDR), in which dose was primarily sensitive to food chain parameters or dose coefficients, for the years addressed in this part of the TDR, the large uncertainty in sources and dilution factors is the driver behind the sensitivity of dose to these parameters. The uncertainty associated with Building 231 dominates the early years, whether the dose is to someone at Location T or Location VIS, regardless of the assumptions about speciation. Only infant milk intake and the HTO dose coefficient in 1957 (original assumptions) are not related to parameters whose values vary from year to year. When the major releases shifted from Building 231 to Building 331, so did the sensitivity of dose shift to the new source. Dose at the Discovery Center was sensitive to releases from the Building 612 Yard, which did

not affect Location T; otherwise the parameters for tritium sources to which dose is sensitive are essentially the same at the two locations.

Table 4a. Parameters to which dose is sensitive at Location T for representative years.

	Dose to adult	Dose to child	Dose to infant
1953 and 1957 – releases mostly HT	B231 dilution factor Release B231 WAA	B231 dilution factor HTO dose coefficient	Milk intake B231 dilution factor
1953 and 1957 – releases 54% HTO	Release of HTO from B231 Stack B231 dilution factor	Release of HTO from B231 Stack B231 dilution factor	Milk intake Release of HTO from B231 Stack B231 dilution factor
All three years tested, 1961, 1964, 1970	Release of HTO from B331 S Release B331 WAA	Release of HTO from B331 S Release B331 WAA	Milk intake Release B331 WAA
Two of the years 1961, 1964, 1970	B331 dilution factor	Release of HTO from B331 N	Release of HTO from B331 S Release of HTO from B331 N

Table 4b. Parameters common to adult, child, and infant to which dose at Location T is sensitive for each year analyzed.

Year	Release mostly HT	Release 54% HTO
1953	Release of HTO from B231 Stack B231 stack dilution factor Release from B231 WAA	Release of HTO from B231 Stack B231 stack dilution factor Release from B231 WAA
1957	B231 stack dilution factor Dose coefficient for HTO	Release of HTO from B231 Stack B231 stack dilution factor Release from B231 WAA
1961		Release from B331 WAA Release of HTO from B331 S
1964		Release from B331 WAA Release of HTO from B331 S Release of HTO from B331 N
1970		Release from B331 WAA Release of HTO from B331 S Release of HTO from B331 N

Table 5a. Parameters to which dose is sensitive at Location VIS for representative years.

	Dose to adult	Dose to child	Dose to infant
1953 and 1957 – releases mostly HT	B231 dilution factor Release B231 WAA	B231 dilution factor	Milk intake B231 dilution factor
1953 and 1957 – releases 54% HTO	Release B514 Yard Release of HTO from B231 Stack B231 dilution factor	Release B514 Yard Release of HTO from B231 Stack B231 dilution factor	Milk intake Release of HTO from B231 Stack B231 dilution factor
1961, 1964, 1970	Release of HTO from B331 S Release B331 WAA	Release of HTO from B331 S Release B331 WAA	Milk intake Release of HTO from B331 S

Table 5b. Parameters common to adult, child, and infant to which dose at Location VIS is sensitive for each year analyzed.

Year	Release mostly HT	Release 54% HTO
1953	Release of HTO from B231 Stack B231 stack dilution factor B231 WAA	Release of HTO from B231 Stack B231 stack dilution factor B231 WAA
1957	B231 stack dilution factor Dose coefficient for HTO	Release of HTO from B231 Stack B231 stack dilution factor
1961		Release of HTO from B331 S
1964		Release evaporation trays Release of HTO from B331 N Release of HTO from B331 S
1970		Release B612 Yard Release of HTO from B331 N Release of HTO from B331

DISCUSSION

The higher of the two upper confidence limits for doses from releases between 1953 and 1960 (Figure 2 and Figure 4) is comparable to the upper confidence limit for doses predicted between 1961 and 1972 because both were calculated using the assumption that

54% of the releases from either Building 231 or Building 331 were HTO. HT converts to HTO when it comes into contact with metal and other surfaces in a facility for any length of time. Nevertheless, for known large puff releases, the fraction of HTO released is much less than 54%. Because the true speciation is unknown, the assumption that 54% of the total tritium released was HTO was used because it is conservative[22]. The magnitude of the confidence intervals on doses between 1953 through 1960 exists because the 2.5% confidence limit and mean (and the lower set of upper confidence limits) were calculated based on the assumption that most of the tritium released was HT.

From 1961 onwards, when stack sampling occurred and releases were reported quarterly rather than as occurring as puffs on a specific date, the assumed speciation of 54% HTO and 46% HT released from the Tritium Facility was used to estimate the mean and the 95% confidence intervals of the annual doses. In reality, the shift from releases of mostly HT to 54% HTO (a ratio based on monitoring data after 1973) undoubtedly was gradual and did not change abruptly, as it did for the TDR modeling in 1961. The real rate-of-change of the speciation is unknown, of course, and, because the upper confidence limit will be conservative when 54% of the release is assumed HTO[23], there was no incentive to attempt to model a gradual, and unknown, increase in the fraction of HTO released over time. The means and 2.5% confidence limits for 1961 through 1972 may in reality be lower than shown in Figures 2 and 4 because of this probable overly conservative assumption about speciation. The mean (or best estimate) of the distributions for all years may be higher or lower than their positions within the confidence intervals depending upon the unknown relationship between actual and assumed speciation of releases and on the presence of area sources.

Mean doses predicted at Location VIS and Location T were compared each year using the original speciation assumptions (Figure 8) and the alternative assumptions (Figure 9). Location Q has been included in the figures for 1961 when it was the SW-MEI. After 1960, Figures 8 and 9 are identical, although they look different because of the different scales (the scale in Figure 9 is 3.5 times greater than that in Figure 8). Doses were clearly higher from 1953 through 1958 at Location T than at Location VIS, regardless of the assumptions. In 1959 and 1960, which location had the higher doses was determined by the assumptions. With the original assumptions, doses at Location T and Location VIS were the same in 1959, while, in 1960, the SW-MEI was clearly at Location T. With the alternative assumptions, Location VIS was the SW-MEI in 1959 and 1960.

[22] If the relatively large HT releases from routine operations could have been modeled as puffs, the recipient of the largest dose impact could easily have been at a location different from that of the SW-MEI for annual releases. The dose, however, being modeled as HT would have been smaller than the dose assuming a routine release that was 54% HTO received by the SW-MEI.

[23] Uncertainty on speciation was included with the uncertainties on the release rate and ranged from ± 18% to ± 40% depending upon whether or not the release was HT or HTO and from which stack the tritium was emitted.

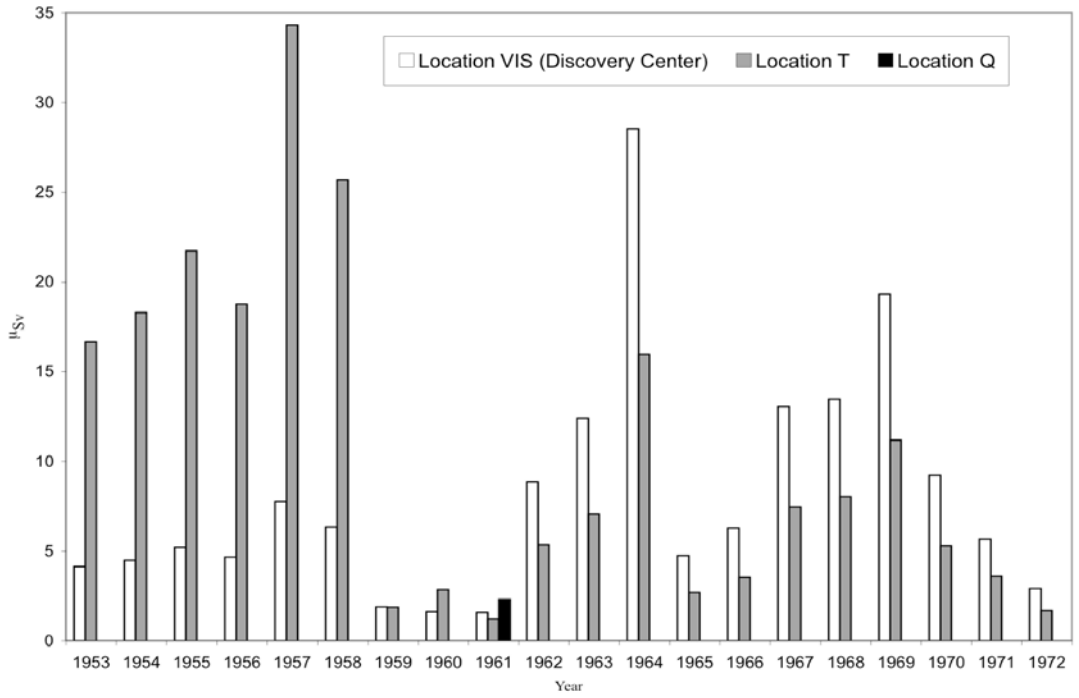

Figure 8. Comparison of mean doses at locations of the SW-MEI (1953 – 1958, Location T; 1961, Location Q; 1959, 1960, 1962 – 1972, Location VIS) assuming releases from the Tritium Facilities were primarily HT prior to 1961.

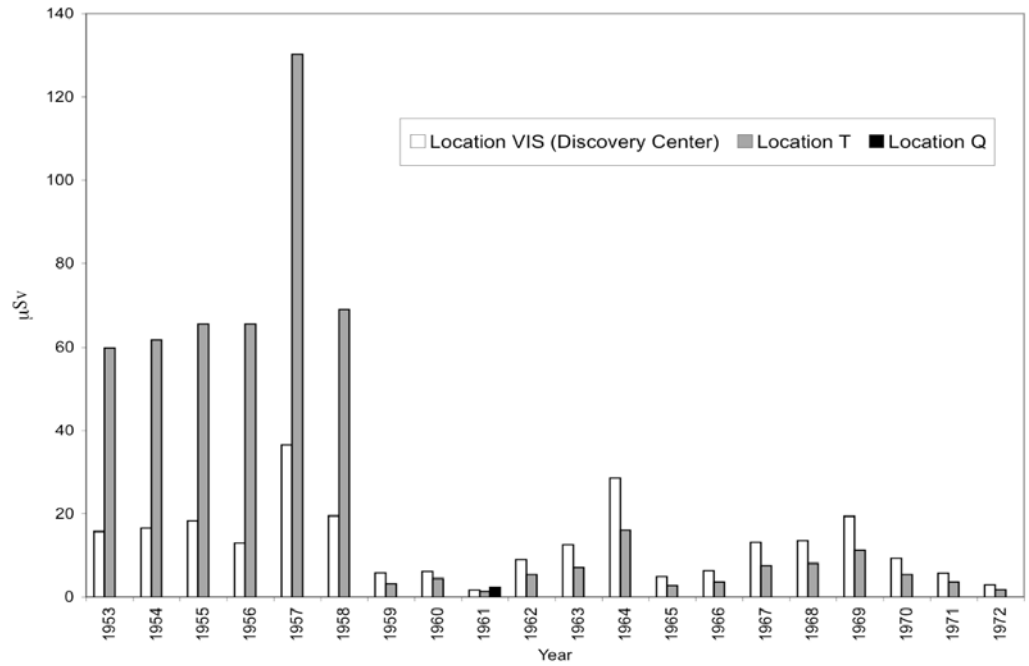

Figure 9. Comparison of mean doses at locations of the SW-MEI (1953 – 1958, Location T; 1961, Location Q; 1959, 1960, 1962 – 1972, Location VIS) assuming releases from the Tritium Facilities were 54% HTO.

Location T was the SW-MEI when releases were primarily from Building 231 and its WAA (Figure 3b). Although the fraction of total dose contributed by releases from Building 231 was similar at Location T (Figure 3b) and Location VIS (Figure 5b), the actual dose was higher at Location T because of its proximity to Building 231 (compare Figure 3a with Figure 5a). Doses in 1957 were higher than other years (Figure 6) because of the more than 11,000 Ci of HT released from Building 231 (the upper confidence limits were calculated on the assumption that 6,480 of the curies released were HTO). As soon as Building 331 started to release tritium, because of the magnitude of releases and the location of Building 331 relative to Location T and Location VIS, the shift began towards Location VIS as the SW-MEI.

The comparisons shown in Figures 3a and 3b and Figures 5a and 5b, prior to 1961, are based on the assumption that most of the tritium released from Buildings 231 and 331 was HT. The dynamics of the comparison will change if the mean doses and fractions of total dose contributed by each facility are based on the alternative speciation assumption. If it is assumed for those years that 54% of the tritium released from the Tritium Facilities was HTO, the dose attributed to the Tritium Facilities will rise because of the higher dose impact of HTO. In addition, because the Building 514 Yard's release rate is derived from the quantity of HTO released from the Tritium Facilities, the dose impact of the Building 514 Yard will rise as well. With the Tritium Facilities and Building 514 Yard contributing more to the dose, the fractional contribution of the Building 231 or 331 WAAs will decrease.

Results in 1959 and 1960 are not consistent with other years. The mean dose predicted for Location T is either the same or higher than at Location VIS, but the upper confidence limits at Location VIS are higher (Figure 6). Overall, however, the doses at the two locations are essentially indistinguishable. These results are due to the assumptions about the quantity of tritium released from the Building 331 WAA. Note in Figures 3a and 3b that, because the release rate from the WAA is directly derived from the total tritium released each year from the stack(s), the Building 331 WAA releases track the Building 331 stack releases perfectly from 1961 onwards because the assumption - that 54% of the released tritium is HTO - is constant; in 1959 and 1960, the contribution of HT to the stack releases was greater. Furthermore, the WAA release rates in 1959 were assumed half the later release rate because the WAA was newly opened. Because Location T is more sensitive to releases from the Building 331 WAA than is Location VIS (compare Figure 3b with Figure 5b), its mean doses were higher (compare Figure 3a with Figure 5a) than those at Location VIS.

Doses at all potential locations of the SW-MEI were lowest in 1961 (Figure 6) because releases of HT and HTO from Building 331 totaled about 800 Ci in 1961, which was the lowest annual release rate from a Tritium Facility during the time period of this TDR[24]. Not only were the mean doses the lowest, but the upper confidence limits were also the

[24] The year with the next lowest release rate was 1972, when 1,350 Ci were released.

lowest because 1961 was the first year that less uncertainty was applied to the assumption that 54% of the tritium released from the Tritium Facilities was HTO.

Cumulative dose was higher at Location T than at Location VIS, because, even though the SW-MEI was there for fewer years than it was at Location VIS (Figure 7), doses at Location T were about a factor of four higher than those at Location VIS for 1953 through 1958. However, because the highest mean cumulative dose, that to the hypothetical individual born at Location T (290 μSv [29 mrem]), is only 70% greater than the lowest mean cumulative dose, that to an adult at the Discovery Center (170 μSv [17 mrem]), the main difference between the cumulative doses at Location T and Location VIS is the magnitude of the uncertainty, which determines the upper confidence limit. The magnitude of the uncertainty on the cumulative dose is a factor of 3 for the adult at Location VIS compared with a factor of 7 for the individual born at Location T.

The location of the SW-MEI is dependent upon the assumptions made about relative release rates, stack parameters, and the presence and location of assumed area sources. The effect on dose at any location from any one source cannot be predicted without dispersion modeling, and dispersion modeling must also be used to predict the location of the SW-MEI when release rates of sources change relative to each other. The location of the SW-MEI when it was at Locations T or Q was very sensitive to the assumptions about stack heights, stack diameters and exit velocities used to determine the dilution factor for Building 231. A relatively small change in these assumptions would have shifted the location of the SW-MEI from Location T to Location S, at least for 1956, and from Location T to Location Q for 1960. This is of no concern, however, because doses to the chosen SW-MEI and the other possible contending locations were within 10 – 40% of each other, well within the uncertainty of the calculations. Changing the assumptions about speciation only changed the ranking of the potential SW-MEI in minor ways.

The relative importance of each source at Location T and at Location VIS for a unit release (i.e., 1 Ci y^{-1}) is shown in Figure 10. This figure demonstrates the dynamic relationship between sources and receptors. The figure was obtained by dividing the dilution factor for each source "x" at location "y" by the highest dilution factor, that for the Building 231 WAA at Location T. The relative effect of these sources would be somewhat different at the other potential locations of the SW-MEI. From this figure, it is clear that the potential effect of a source depends upon two variables: 1) whether the release is an area source or a stack release, and 2) the location of the release relative to the receptor. The actual effect will depend upon these two variables and a third variable – the magnitude of release rate (see Tables A1 – A5). Unit release rates from the Building 331 stacks have very little effect on dose to the SW-MEI; the importance of the tritium releases from the Tritium Facility is due to their magnitude. The effects of releases from the Building 231 stack or Building 212 stack are higher per unit release than those of the Building 331 stacks because the release heights and exit velocities are lower and the facilities are relatively close to the receptor. The effect of an area source

upon the receptor is roughly dependent upon the distance from a source to the receptor, but, given the wind and stability patterns of the Livermore site, there is a tendency for air concentrations to be somewhat higher at any set distance from a source to a receptor on the south and west sides of the site. Thus, for example, although the Building 331 WAA is slightly closer to Location VIS than it is to Location T, the dilution factor at Location T is four times higher than that at Location VIS from this source.

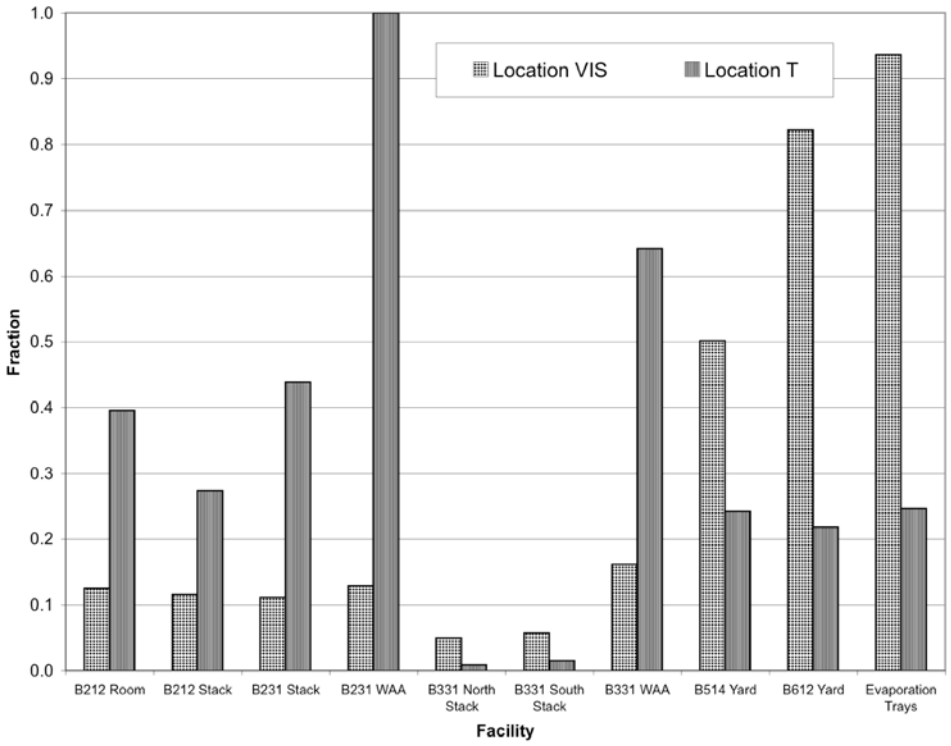

Figure 10. Comparison of the relative importance of unit release from all facilities to two receptors. All values were normalized to the highest dilution factor (that of releases from the Building 231 WAA at Location T).

The impact of area sources on dose was investigated by determining the fraction of total dose contributed by area sources for each year under different assumptions (Figure 11). The deterministic total dose from all sources to an adult was compared with the dose to an adult from area sources alone (the stack release rates in DCART were zeroed). Area sources contributed a greater fraction of the dose when the releases were assumed to be primarily HT because release rates for the area source that was a major contributor to dose (the Building 231 WAA) were based on total tritium, which meant that the air concentration contributed by the WAA did not change when the assumptions about speciation changed. The effect of this becomes clear for both Location T and Location VIS when predicted HTO concentrations in air (HT converted to HTO-equivalent) from each source are compared for 1957 for the two assumptions about the speciation of the tritium released from the Tritium Facility (Table 6).

Table 6. Air concentrations (Bq m^{-3} HTO) contributed by each source under different assumptions.

	B231 HT	**B231 HTO**	**B231 WAA**	**B212 HT**	**B212 HTO**	**B514 Yard**	**Sum**
1957 – primarily HT - Location T	5.88	6.73	7.55	0.0307	0.0895	0.148	20.4
1957 54% HTO – Location T	2.85	71.5	7.55	0.0307	0.0895	1.58	83.6
1957 – primarily HT – Location VIS	1.50	1.71	0.969	0.00970	0.0283	0.329	4.55
1957 – 54% HTO – Location VIS	0.724	18.2	0.969	0.00970	0.0283	3.49	23.4
	B331 HT	**B331 HTO**	**B331 WAA**	**B212 HT**	**B212 HTO**	**B514 Yard**	**Sum**
1960 – primarily HT - Location T	0.0562	0.0688	1.29	0.0307	0.0895	0.0416	1.58
1960 54% HTO - Location T	0.0272	0.684	1.29	0.0307	0.0895	0.425	2.55
1960 – primarily HT – Location VIS	0.207	0.247	0.326	0.00970	0.283	0.086	1.16
1960 – 54% HTO – Location VIS	0.101	2.53	0.326	0.00970	0.283	0.882	4.13
	B331 HT	**B331 HTO**	**B331 WAA**	**B212 HT&HTO**	**Evap Trays**	**B612 Yard**	**Sum**
1970 – Location T	0.0269	0.676	1.79	0.0721	0.0266	0.468	3.06
1970 – Location VIS	0.127	3.20	0.452	0.0306	0.101	1.77	5.68

Once tritium operations switched from Building 231 to Building 331 (south stack) in 1959, a transition began between the relative importance of area sources to doses at Location T and Location VIS between 1953 and 1958 and the pattern seen between 1962 and 1972 (see Figure 11 and the data for 1960 in Table 6). The relative magnitude of the area contributions is quite stable from 1962 through 1972 because releases, which dominated dose, started up from the north stack of Building 331 in 1962 and area sources had the same release rate relationship to Building 331 from 1962 through 1972 (Table 6). During this period, the relative dose impact of area sources was always greater at Location T, i.e., the magnitude of the dose was larger at Location VIS, but the fraction of total dose contributed by the area source was greater at Location T (see 1970 data in Table 6). The large release rate from the Building 331 stacks and the fact that the wind blows primarily toward Location VIS meant that the tritium contributed by the stacks dominated the dose at Location VIS, while the tritium released from the Building 331 WAA dominated dose at Location T.

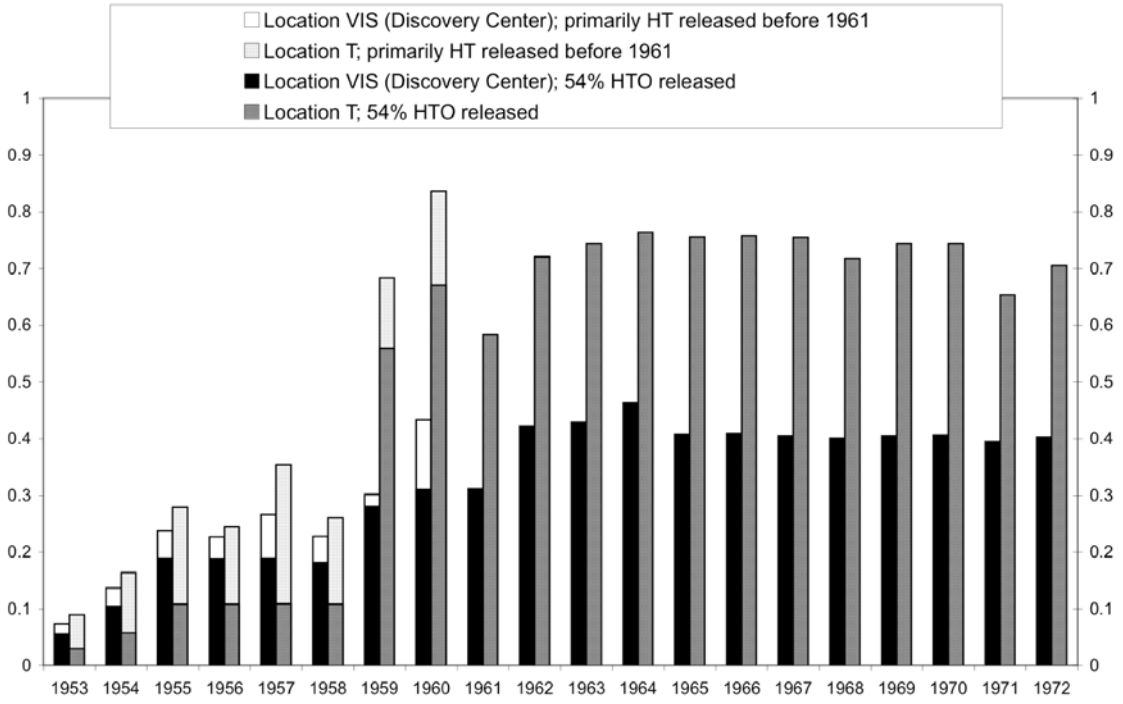

Figure 11. Fraction of total dose contributed by area sources each year at Location VIS and at Location T, as predicted by DCART, assuming either that 54% of the tritium released from the Tritium Facilities was HTO or that the tritium released was mostly HT.

There is considerable uncertainty surrounding the dose predictions for the years 1953 through 1972 (Figure 2 and Figure 4). Even though this TDR attempts to account quantitatively for all sources of uncertainty, the confidence intervals shown in Figures 2, 4, 6 and 7 imply a more accurate dose assessment than is possible. Among other things, it is impossible to quantify the uncertainty for sources that may or may not have existed in the chosen locations (e.g., the Building 231 WAA) or the uncertainty that arises when routine puff releases are modeled as steady-state and speciation is essentially unknown. However, because of deliberately conservative assumptions, reasonable confidence can be placed in the probability that any and all doses received by any member of the public will have been below the 97.5% confidence limit (based on 54% of the releases from the Tritium Facilities being HTO), no matter the location of the SW-MEI.

The Discovery Center was chosen as the location at which to calculate doses from 1973 through 2005 (Part 3 of the TDR) because, besides being the location of an air tritium monitor (VIS) and being close to the location of the SW-MEI for NESHAPs compliance, model testing had demonstrated that CAP88-PC did not underestimate air concentrations at that location. This TDR, using CAP88-PC, has shown Locations T and Q to be the locations of the SW-MEI for the early years. Location T was close to the current location of the MESQ air tritium sampling unit. Observed annual mean tritium concentrations in

air at that unit were used to test air concentrations predicted by CAP88-PC for 1986 - 1999[25] (Peterson 2004). Release rates assumed for the model testing were somewhat different from those assumed for this TDR. Stack releases rates were similar, but, in the model test, the Building 331 WAA was not included as a source prior to 1992, and the estimated release rate from the Building 612 Yard prior to 1992 was assumed to be 2% of the annual HTO releases instead of the 4% assumed in the TDR. With no area release rates included in the test, CAP88-PC underestimated concentrations at MESQ for all years 1986 through 1993 by 2.5 to 4.8 times (median factor of 3.3); when it was assumed that the B612 Yard released 2% of the HTO released from the Tritium Facility, the underestimation was reduced to between a factor of 1.7 and a factor of 3.7 (median 2.2)[26]. For CAP88-PC to correctly predict air concentrations[27] at potential locations of the SW-MEI, e.g., Locations T and Q, the assumption of area sources is necessary (whereas, for the Discovery Center, the overestimation of the dispersion model from stack sources makes the addition of area sources redundant or overly-conservative). Given the reasonable assumptions about missing potential area sources in this TDR, given the large uncertainty associated with the early releases, and given the very conservative assumptions about diet, doses at Locations T or Q should not have been underestimated, although they would not have been overestimated by nearly as much as at the Discovery Center.

The assumptions about the presence of area sources and the quantities of tritium released from them are intended to be conservative. The assumptions do, however, result in some seemingly odd dose impacts. For example, the assumption that all stored tritiated waste was evaporated during the years between the development of Building 514 as a Tank Farm and the opening of the Building 612 Yard results in a dose impact (Figures 3b and 5b) probably much, much larger than any that might have occurred from actual releases from the evaporation trays from 1962 through 1964. The effect at Location VIS is greater than at Location T (Figures 3b and 5b). This assumption was made, however, to account for stored waste that must have existed but about which nothing is known. If the "missing" releases ascribed to the evaporation trays had come from a different location, the dose impact, at least at the Discovery Center, would have been less.

At Location T, the most conservative assumptions resulted in maximum predicted dose to an adult for 1957 of 370 μSv (37 mrem) (see Figure 2) and 590 μSv (59 mrem) to an infant. In 1958 (NCRP 1958), the National Council on Radiation Protection and Measurements (NCRP) recommended a dose limit for continuous exposure of 5 mSv (500

[25] The MESQ sampler in 1986 and 1987 was east of Location T along the then-perimeter of the Laboratory.
[26] Compare this to the results for the VIS air tritium sampler, which, for the same time period, underestimated air concentrations in 1986 by 10%, 1987 by 30% and overestimated by 10 – 80% for the years when no area sources were assumed; when a release rate for the Building 612 Yard was assumed, air concentrations were overestimated by up to a factor 2.3 and only underestimated by 4% for 1987.
[27] At LLNL, light winds blow to the west and northwest mainly at night when the stability classes are E and F. As a result, the highest air concentrations predicted by CAP88-PC miss MESQ and come to ground about 3000 m from a stack source, well beyond MESQ. The plume has passed over MESQ and lack of vertical mixing with E, and especially F, stability class in CAP88-PC causes underestimation close to the source.

mrem) to "persons outside of controlled areas". This limit was reiterated in 1960 in a statement that read, "it is our basic recommendation that the yearly radiation exposure to the whole body of individuals in the general population (exclusive of natural background and deliberate exposure of patients by practitioners of the healing arts) should not exceed 0.5 rem" (FRC 1960). The 500 mrem limit was still in effect in 1971 (NCRP 1971), so all the doses predicted in this part of the TDR fell well below the regulatory limits of the time. In fact, the maximum dose predicted to the hypothetical infant in 1957 using very conservative assumptions is below the current Department of Energy (1993) radiation standard for protection of the public of 1 mSv y^{-1} (100 mrem y^{-1}) effective dose equivalent for prolonged exposure of a maximally exposed individual in an uncontrolled area (DOE 1993) for all types of releases. Dose at Location VIS never exceeded 100 μSv (10 mrem) annually (Figure 4) regardless of the degree of conservatism in the predictions. In other words, the annual dose at Location VIS never exceeded the current regulatory limit for compliance with NESHAPs for releases of radioactivity to air.

Ingestion dose is a larger fraction of the total dose for a release of HT than for a release of HTO. After a unit release of HT, 89% of the adult's and child's dose comes from ingestion, and 93% of the infant's dose is obtained from food; in contrast, for a unit release of HTO, 80% of the total dose to adult and child is from ingestion, and 88% of the infant's dose is from food. This small difference occurs because, for an HT release, the HTO concentrations in air and plants are driven by the HTO concentration in soil. Agricultural crop plants, because they grow close to the soil surface, are exposed to higher HTO concentrations than people are through inhalation. In contrast, for an HTO release, plants and people are exposed to essentially the same concentration of HTO in air, and ingestion plays a less important role.

CONCLUSIONS

Although results are presented as 95% confidence intervals that are calculated from the assessed uncertainty on all parameter values, because of conservative assumptions about the degree to which the diet is contaminated and the fraction of the diet that is contaminated (100%), the upper confidence limit is most certainly a value that could never have been exceeded by any member of the public.

Because Location T lay to the west of the Laboratory, and because of wind patterns and the dispersion model in CAP88-PC, the assumptions about the presence of area sources and the magnitudes of their releases were probably essential so that air concentrations were not underestimated. Dose predictions will be conservative at Location T because of the conservative assumptions about diet. Even though Location T was an actual residence at which vegetables and fruit might have been grown, no actual intake could be as great as predicted by DCART. Neither Location Q nor Location VIS was a residence[28]. At

[28] Location VIS was simply a location until 1976 when the Visitors Center (now Discovery Center) was dedicated. It was used as a location of the SW-MEI for this part of the TDR so that doses could be compared with ones calculated for 1973 through 1975 (see Part 3 of the TDR) and to produce a continuum of doses at a single location.

Location Q, the dose would have been strictly from inhalation for a fraction of the year, and because there was no individual being exposed at Location VIS, dose predictions for these locations must exceed by a large margin any dose that could have been received by a real member of the public. There is additional conservatism built into the dose predictions at Location VIS, because air concentrations are over-predicted by CAP88-PC at that location (Peterson 2004).

The assumptions about area sources, their existence, locations and quantities of tritium released, can have a huge impact on doses (Figure 11). For conservatism, it is important to assume the existence of otherwise unknown area sources, because to neglect to model dose from such an area source could result in an underestimation of dose.

Even with the conservatism built into the calculations, the highest predicted dose to the SW-MEI in any year (Figure 6) was well below the regulatory dose limit to an individual member of the public that applied during the time period of this TDR: 5.0 mSv (500 mrem). All predicted doses can be reduced by a factor of two with more realistic but still conservative assumptions about food ingestion.

The location of the SW-MEI was at Location T for nine years, at Location Q for one year, and at Location VIS for ten years. Predicted doses at Location T were higher than any doses predicted at Location Q or Location VIS. These high doses at Location T dominate the cumulative dose received by the hypothetical adult or the hypothetical individual born and raised there. Dose predicted by DCART from routine releases of tritium from the Livermore site to the adult living at Location T from 1953 through 1972 and to the infant born in 1953 and growing up at Location T through 1972 are not expected to have exceeded 860 μSv (86 mrem) or 1300 μSv (130 mrem) respectively. These doses are about 2.5 times higher than the cumulative doses predicted at the Discovery Center. The cumulative dose received by the adult at the Discovery Center was essentially equal to the highest dose received by the adult at Location T in 1957.

Adverse health effects have been demonstrated conclusively for exposures greater than 100 mSv (10,000 mrem) (ATSDR 1999). No adverse health effects have been documented for doses less than 3.6 mSv (360 mrem per year) (ATSDR 1999); the upper confidence limit of the dose predicted to an infant at Location T in 1957 was a factor of six below this. 360 mrem per year is defined by ATSDR as a No Observed Adverse Effect Level (NOAEL). ATSDR derived a minimal risk level (MRL) from this value by reducing it by a factor of three to account for human variability and further rounded it down to 1 mSv (100 mrem) per year for chronic exposure. 1 mSv (averaged over a five-year period) is also the International Commission on Radiological Protection (ICRP 1991), NCRP (NCRP 1993), and DOE (1993) dose limit for exposure to radiological operations over and above the dose received from natural background radiation by a member of the public. MRLs are intended only to serve as a screening tool to help public health officials decide which release situations require more extensive evaluation. The MRL for chronic exposure to ionizing radiation is considered protective for both cancer

and non-cancer health effects. The very conservative upper limit of annual dose estimated from routine LLNL operations, which was to an infant in 1957, was 59 μSv (59 mrem), which is 59% of the 1 mSv limit.

APPENDIX

Table A1a. Estimated annual releases of HT and HTO (Ci) from Building 231 (then Building 102; the predecessor of the LLNL Tritium Facility) with one standard deviation (σ) uncertainty on a normal distribution when releases were assumed to be mostly HT. The truncated lower limit (LL) indicates that a known minimum of activity was released; it is only mentioned if not zero or irrelevant. The upper limit (UL) was calculated but no truncation occurred because the value falls within the limit determined by the standard deviation.

	HT		HTO	
Year		**LL**		**UL**
1953	4860 ± 2920	-----	540 ± 324	-----
1954	4860 ± 2920	-----	540 ± 324	-----
1955	4860 ± 2920	-----	540 ± 324	-----
1956	3470 ± 2000	2800	530 ± 240	1250
1957	11400 ± 1900	8400	610 ± 90	870
1958	5530 ± 2000	4380	730 ± 240	1420

Table A1b. Estimated annual releases of HT and HTO (Ci) from Building 231 based on the alternative assumptions that the releases were 54% HTO to determine the conservative upper confidence limits for the dose predictions. Uncertainty is one standard deviation on a normal distribution. HT and HTO are correlated (-0.4).

Year	**HT**	**HTO**
1953	2480 ± 1790	2920 ± 2100
1954	2480 ± 1790	2920 ± 2100
1955	2480 ± 1790	2920 ± 2100
1956	1840 ± 1290	2160 ± 1300
1957	5520 ± 2390	6480 ± 2770
1958	2880 ± 1510	3380 ± 1670

Table A2. Estimated annual releases of HT and HTO (Ci) from Building 212 and uncertainty (σ) as one standard deviation of a normal distribution, unless otherwise noted.

Year	**HT**	**HTO**
	Room Air: Cockcroft-Walton accelerator and 90"cyclotron	
1953	0.13 – 2.8 – 6.9 (triangular)	0.018 – 0.38 –0.94 (triangular)
1954	0.13 –11 - 31 (triangular)	0.018 – 1.5 – 4.2 (triangular)
1955 - 1964	66.0 ± 46.7	9.00 ± 6.36
1965 - 1967	8.80 ± 6.22	1.20 ± 0.849
	Insulating Core Transformer Accelerator	
1967	4.4 – 44 (uniform)	0.6 – 6 (uniform)
1968	211 ± 120	28.8 ± 16.4
1969	123 ± 70.2	16.8 ± 9.57
1970	57.2 ± 32.6	7.80 ± 4.44
1971	229 ± 130	31.2 ± 17.8
1972	45.8 ± 26.1	6.24 ± 3.56

Table A3a. Estimated routine annual releases of HT and HTO (Ci) from the LLNL Tritium Facility (Building 331). Uncertainty is one standard deviation (σ) of a normal distribution. Releases for 1959 and 1960 were assumed to be mostly HT. Releases of HT and HTO are negatively correlated. The correlation is –0.4 for Stack 1 and –0.5 for Stack 2.

	Stack 1 HT	Stack 1 HTO	Stack 2 HT	Stack 2 HTO
1959	2820 ± 1140	377 ± 152	-----	-----
1960	3070 ± 1230	171 ± 68.5	-----	-----
1961	368 ± 142	432 ± 146	-----	-----
1962	1430 ± 668	1360 ± 587	755 ± 322	886 ± 341
1963	1040 ± 482	1220 ± 520	1730 ± 731	2030 ± 773
1964	2160 ± 1190	2530 ± 1320	3590 ± 1870	4220 ± 2050
1965	405 ± 214	476 ± 235	676 ± 332	793 ± 361
1966	526 ± 322	618 ± 361	877 ± 510	1030 ± 568
1967	1140 ± 530	1340 ± 572	1900 ± 804	2230 ± 851
1968	1160 ± 529	1370 ± 569	1940 ± 769	2280 ± 800
1969	1680 ± 822	1970 ± 895	2800 ± 1220	3290 ± 1290
1970	775 ± 473	910 ± 529	1290 ± 734	1520 ± 816
1971	467 ± 264	549 ± 293	779 ± 404	915 ± 443
1972	182 ± 85.4	324 ± 123	388 ± 160	456 ± 139

Table A3b. Estimated routine annual releases of HT and HTO (Ci) from the LLNL Tritium Facility (Building 331) based on the alternative assumptions that the releases were 54% HTO to determine the conservative upper confidence limits for dose predictions. Uncertainty is one standard deviation (σ) on a normal distribution. HT and HTO are correlated (-0.4).

Year	HT	HTO
1959	1470 ± 593	1730 ± 696
1960	1490 ± 597	1750 ± 700

Table A4. HTO (Ci) estimated to have been released annually from the Building 231 WAA and the Building 514 Yard. Distributions are normal with one standard deviation (σ). Release rates from the B231 WAA are correlated (0.5) with annual releases of HTO and HT from the Tritium Facility; release rates from the B514 Yard are correlated (0.4) with HTO released annually from the Tritium Facility Stacks. Values for the "Building 514 Yard" were calculated from HTO values in Tables A1a and A3a; values for the "Building 514 Yard Alternative" were calculated from HTO values in Tables A1b and A3b and were used to determine the conservative upper confidence limits on dose predictions.

Year	Building 231 WAA	Building 514 Yard	514 Yard Alternative
1953	33.8 ± 2$\tilde{8}$7	5.40 ± 3.89	29.2 ± 21.0
1954	67.5 ± 57.3	10.8 ± 7.79	58.3 ± 42.1
1955	135 ± 115	21.6 ± 15.8	117 ± 85.1
1956	100 ± 73.3	21.2 ± 12.8	86.4 ± 52.0
1957	300 ± 66.8	24.4 ± 10.4	259 ± 111
1958	156 ± 76.6	29.0 ± 20.0	135 ± 93.0
1959	-----	15.1 ± 6.08	69.1 ± 27.8
1960	-----	6.84 ± 2.74	69.9 ± 28.0
1961	-----	17.3 ± 5.04	NA

Table A5. HTO (Ci) estimated to have been released annually from the Building 331 WAA, the Building 612 Yard, and the evaporation trays. Distributions are normal with one standard deviation (σ). Release rates from the B331 WAA are correlated (0.5) with annual releases of HTO and HT from the Tritium Facility; release rates from the B612 Yard (and the evaporation pans from 1962 – 1964) are correlated (0.4) with HTO released annually from the Tritium Facility Stacks.

Year	Building 331 WAA	Building 612 Yard	Evaporation Trays
1959	39.5 ± 33.6	-----	-----
1960	80.0 ± 68.8	-----	-----
1961	19.8 ± 16.1	-----	-----
1962	110 ± 87.8	-----	82.2 ± 34.6
1963	148 ± 118	-----	119 ± 48.6
1964	309 ± 298	-----	290 ± 192
1965	58.1 ± 46.9	42.2 ± 20.9	4.3 ± 4.15
1966	75.4 ± 62.1	56.6 ± 30.1	4.3 ± 4.15
1967	163 ± 130	126 ± 52.3	4.3 ± 4.15
1968	167 ± 133	129 ± 53.4	4.3 ± 4.15
1969	241 ± 193	189 ± 80.9	4.3 ± 4.15
1970	111 ± 91.5	85.4 ± 44.4	4.3 ± 4.15
1971	67.0 ± 54.6	49.5 ± 25.8	4.3
1972	33.4 ± 26.5	24.3 ± 11.5	4.3

Table A6. Dilution factors (χ/Q in s m^{-3}) with uncertainty (σ) for lognormal distributions for modeled sources other than the LLNL Tritium Facility.

Facility	Years	χ/Q
B231 Stack	1953 - 1958	$2.389 \times 10^{-6} \pm 7.223 \times 10^{-7}$
B231 WAA	1953 - 1958	$2.758 \times 10^{-6} \pm 1.103 \times 10^{-6}$
B514 Yard	1953 - 1961	$1.076 \times 10^{-5} \pm 4.304 \times 10^{-6}$
B212 Room Air	1953(4) - 1967	$2.678 \times 10^{-6} \pm 8.474 \times 10^{-7}$
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	1967 - 1987	$2.494 \times 10^{-6} \pm 7.483 \times 10^{-7}$

Table A7. Dilution factors (χ/Q in s m^{-3}) at the Discovery Center with uncertainty (σ) for lognormal distributions for Stack 1 and Stack 2 of the LLNL Tritium Facility (Building 331),

Years	Stack 1 (South)	Stack 2 (North)
1958 - 1961	$1.230 \times 10^{-6} \pm 3.076 \times 10^{-7}$	-
1962 - 1967	$1.230 \times 10^{-6} \pm 3.076 \times 10^{-7}$	$1.059 \times 10^{-6} \pm 2.654 \times 10^{-7}$
1968	$1.221 \times 10^{-6} \pm 3.056 \times 10^{-7}$	$1.064 \times 10^{-6} \pm 2.664 \times 10^{-7}$
1969	$1.227 \times 10^{-6} \pm 3.072 \times 10^{-7}$	$1.059 \times 10^{-6} \pm 2.654 \times 10^{-7}$
1970	$1.232 \times 10^{-6} \pm 3.082 \times 10^{-7}$	$1.059 \times 10^{-6} \pm 2.654 \times 10^{-7}$
1971	$1.226 \times 10^{-6} \pm 3.070 \times 10^{-7}$	$1.059 \times 10^{-6} \pm 2.654 \times 10^{-7}$
1972	$1.221 \times 10^{-6} \pm 3.055 \times 10^{-7}$	$1.055 \times 10^{-6} \pm 2.642 \times 10^{-7}$

REFERENCES

ATSDR. Agency for Toxic Substances and Disease Registry. Toxicological profile for ionizing radiation. Atlanta, GA. US department of Health and Human Services.; 1999.

Beller, H.; Eaton, G.; Ekwurzel, B; Esser, B.K.; Hu, Q.; Hudson, G.B.; Leif, R.; McNab, W.W.; Moody-Bartel, C.; Moore, K.; Moran; J.E. California GAMA Program; Sources and transport of nitrate in groundwater in the Livermore Valley Basin, California. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-TR-217189, November 2005; 30 pp.

Department of Energy. DOE Order 5400.5. Radiation Protection of the public and the environment, Change 2. 1/7/93.

Federal Radiation Council (FRC). Background material for development of radiation protection standards. Report No. 1. May 13, 1960.

International Commission on Radiological Protection. ICRP Publication 60. 1990. Recommendations of the International Commission on Radiological Protection. Annals of the ICRP 21 No. 1-3. 1991.

Moran, Jean E. Personal communication, December 2005.

Moran, J.E.; Hudson, G. B.; Eaton, G.F.; Leif, R. A Contamination Vulnerability Assessment for the Livermore-Amador and Niles Cone Groundwater Basins. Report to the California State Water Resources Control Board. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-AR-148831, July 2002; 36 pp.

Myers, D. Memo to Ring Peterson. Subject: Comments on historical doses from tritiated water and tritiated hydrogen gas releases from Lawrence Livermore National Laboratory. August 30, 2004.

Myers, D. Comment on review of draft of TDR Part 4; 2006.

Myers, D.S.; Tinney, J.F.; Gudiksen, P.H. Health physics aspects of a large accidental tritium release. In: Moghissi, A.A.; Carter, M.W., eds. Tritium. Phoenix: Messenger Graphics; 611-622; 1973.

National Committee on Radiation Protection and Measurements (NCRP). Maximum permissible exposure to man (April 15, 1958). Washington, DC: U.S. Department of Commerce' Addendum to U.S. National Bureau of Standards Handbook 59; 1958.

National Committee on Radiation Protection and Measurements (NCRP). Basic Radiation Protection Criteria. NCRP Report No. 39, Washington, D.C. January 15, 1971.

National Committee on Radiation Protection and Measurements (NCRP). Limitation of Exposure to Ionizing Radiation. NCRP Report 116. National Council on Radiation Protection and Measurements, Bethesda, Maryland. March 31, 1993.

Ng, Y.C.; Burton, C.A.; Thompson, S.E.; Tandy, R.K.; Kretner, H.K.; Pratt, M.W. Prediction of the maximum dosage to man from the fallout of nuclear devices, IV Handbook for estimating the maximum internal dose from radionuclides released to the Biosphere. Lawrence Radiation Laboratory, Livermore CA. UCRO-50163, Part IV. 1968.

Olsen, J.L. "Lawrence Livermore Laboratory Radioactivity Data", Subject of letter to D.B. Campbell, U.S. AEC, San Francisco, November 9, 1973.

Otsuki, H.H. Memorandum to Director's Office. Subject: Tritium Release for the Fourth Quarter of 1969. January 19, 1970. Confidential, Restricted Data Unclassified Parks, B.S. User's Guide for CAP88-PC, Version 1.0. U.S. Environmental Protection Agency, Office of Radiation Programs, Las Vegas NV (EPA 402-B-92-001); 1992.

Peterson, S-R. Test of CAP88-PC's predicted concentrations of tritium in air at Lawrence Livermore National Laboratory. Health Physics 87(6):583-595;2004. 2004.

Peterson, S-R. Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory. Part 1. Description of Tritium Dose Model (DCART) for Chronic Releases from LLNL Lawrence Livermore National Laboratory, Livermore, CA. UCRL-TR-205083-REV-1; October 2006.

Peterson, S-R. Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory. Part 2. LLNL Annual Site-specific data 1953 – 2005. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-TR-211722-REV-1; September 17, 2007a.

Peterson, S-R. Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory. Part 3. Routine Releases 1973 - 2005. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-TR-234657; September 17, 2007b.

Silver, W.J. Memo to B331 File. Subject: Summary of Tritium Stack Release Test. January 20, 1970.

Souers, C. Email to Ring Peterson, February 2, 2004.

U.S. Environmental Protection Agency. Code of Federal Regulations. Washington, DC: U.S. Government Printing Office; 40 CFR Part 61: 1989.

Yoder, R.E. Memorandum to J. S. Kane. Subject: LRL Incident/Accident Report Serial No. 003. April 26 (?), 1971.

ABBREVIATIONS, ACRONYMS AND DEFINITIONS

AEC	**A**tomic **E**nergy **C**ommission	
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	
EPA	**E**nvironmental **P**rotection **A**gency	
Evap	Evaporation (trays)	
FRC	**F**ederal **R**adiation **C**ouncil	
HT	Tritiated hydrogen gas	
HTO	Tritiated water	
LLNL	**L**awrence **L**ivermore **N**ational **L**aboratory	

ABBREVIATIONS, ACRONYMS AND DEFINITIONS continued

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	**S**ite **A**nnual **E**nvironmental **R**eport
SW-MEI	**S**ite-**w**ide **M**aximally **E**xposed **I**ndividual
TDR	**T**ritium **D**ose **R**econstruction (LLNL, 1953 – 1972, both routine and accidental releases
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