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Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL). Part 2. LLNL Annual Site-specific Data, 1953 - 2005

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Historical Doses From Tritiated Water And Tritiated Hydrogen Gas Released To The Atmosphere from Lawrence Livermore National Laboratory (LLNL)

Part 2. LLNL Annual Site-specific Data, 1953 - 2005

S. Ring Peterson

ABSTRACT

Historical information about tritium released routinely and accidentally from all Livermore Site Lawrence Livermore National Laboratory (LLNL) facilities and from the Tritium Research Laboratory of Sandia National Laboratories/California (SNL/CA) between 1953 through 2005 has been compiled and summarized in this report. Facility-specific data (annual release rates and dilution factors) have been derived from the historical information. These facility-specific data are needed to calculate annual doses to a hypothetical site-wide maximally exposed individual from routine releases of tritiated water (HTO) and tritiated hydrogen gas (HT) to the atmosphere. Doses can also be calculated from observed air tritium concentrations, and mean annual values for one air tritium sampling location are presented. Other historical data relevant to a dose reconstruction (e.g., meteorological data, including absolute humidity and rainfall) are also presented. Sources of information are carefully referenced, and assumptions are documented. Uncertainty distributions have been estimated for all parameter values. Confidence in data post-1974 is high.

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INTRODUCTION

Tritium has been released to the atmosphere from facilities at the Livermore site of the Lawrence Livermore National Laboratory (LLNL) as part of routine operations since 1953[1]. LLNL has calculated and reported doses from these emissions to the hypothetical maximally exposed individual (MEI) or the hypothetical site-wide maximally exposed individual (SW-MEI) since 1973. There are several reasons to conduct a comprehensive analysis of operational emissions of tritium and the resulting doses using a probabilistic model and set assumptions.

1. Doses to the public from routine emissions from LLNL were not calculated prior to 1973.
2. Routine doses to the public during and after 1974 were calculated using three different dispersion models with different assumptions.
3. The dose consequences of a release of tritiated hydrogen gas (HT[2]) are much lower than those of an equivalent release of tritiated water (HTO[3]) when modeled appropriately. Based on scientific judgment and regulatory requirements at the time of dose calculations, LLNL either did not calculate dose from HT releases (because inhalation dose consequences of an HT release were 1/25,000th of an equivalent release of HTO) or modeled HT as HTO. The first approach slightly underestimates dose from a release of HT; the second approach greatly overestimates dose from a release of HT. Recently developed tritium dose models, such as DCART (Peterson 2006), estimate dose from releases of HT as well as HTO.
4. Recently developed tritium dose models, again such as DCART, account for dose from organically bound tritium (OBT), while regulatory models only model dose from inhalation and ingestion of HTO. Dose from ingesting 1 Bq of OBT is about 2.4 (or more) times that from ingesting 1 Bq of HTO, so to omit dose from OBT may result in the dose being underestimated.
5. Doses were calculated in the past using a deterministic analysis that produced a single best estimate. In such an analysis, the uncertainty associated with the dose cannot be assessed, which means it is not possible to specify the confidence that can be placed in the prediction. On the other hand, a probabilistic analysis generates a distribution of doses from which meaningful statements can be made regarding the probability that the true dose will fall within a certain specified interval. For a probabilistic set of dose predictions with a 95% confidence interval, the most likely dose will be the mean of the distribution, but the true dose will lie between the 2.5% and 97.5% confidence limits; there will be only a 2.5% probability that the true dose will exceed the upper confidence limit.

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[1] Tritium was also released to the atmosphere from the Livermore site of Sandia National Laboratories/California between 1979 and 1995.

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

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

6. For any assessment, because different assumptions and interpretations of the data are often made, much greater confidence can be placed in the results if two (or more) sets of predictions agree (Peterson et al. 1996; Thiessen et al. 1997).

If doses can be predicted based on annual mean observed air concentrations[4], they will be more accurate (although not necessarily less uncertain) than doses calculated from predicted air concentrations. Another potential way to estimate doses is to estimate the air concentrations to which trees were exposed while growing by analyzing concentrations of OBT in annual tree rings. Analyzing the tritium concentration in a core from a tree will provide a means to estimate the integrated air concentration (and dose) for the years represented by the core. There is considerable uncertainty in this approach, but the predicted doses should show some relationship between those obtained from modeling and from measured air concentrations.

To accomplish a complete tritium dose reconstruction (TDR) from LLNL releases, as described above, a six or seven part series, "Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory" has been undertaken for the Livermore site. All parts of the series will be released as LLNL reports.

- Part 1 describes DCART (**D**oses from **C**hronic **A**tmospheric **R**eleases of **T**ritium), the stochastic model (Peterson 2006) that is being used to calculate dose from routine releases for the TDR.
- Part 2 (this report) presents the data and supporting references about past releases, both routine and accidental. It also presents all values for routine releases, their uncertainties, and the assumptions behind the values that are being used as site-specific input to DCART for routine releases.
- Part 3 will present doses with 95% confidence intervals calculated by DCART from observed air concentrations at the VIS ambient air tritium monitor (adjacent to the LLNL Discovery Center – see Figure 1). These doses will be compared with those calculated by DCART from routine releases for 1973 – 2005 from release rates described in this report and dilution factors (also described in this report) obtained from the Environmental Protection Agency's (EPA) compliance dispersion and dose code, CAP88-PC (Parks 1992, 1997).
- Part 4 will present doses with 95% confidence intervals calculated by DCART at the Discovery Center and at the location of the SW-MEI from routine releases for 1953 – 1972 based on annual release rates and dilution factors described in this report.
- Part 5 will estimate doses to the MEI from the larger accidental tritium releases.
- Part 6 will summarize the results of Parts 3, 4, and 5.
- If time and costs of analysis permit, Part 7 will present doses estimated from the analysis of OBT in cores representing the last 50 years of growth obtained from trees near the perimeter of the Livermore site. These dose estimates will be compared with those calculated in Parts 3 and 4.

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[4] LLNL began measuring air tritium concentrations at some locations in 1973.

This document is as comprehensive as possible and contains all the information about past tritium releases and facilities past and present that has been gathered over a period of several years. This document pulls together tritium information from numerous sources and provides one centralized resource for those interested in all aspects of past tritium releases to the environment. This document also serves as the background and archive of annual release rates and assumptions that are being used to predict doses from tritium released routinely by LLNL and SNL/CA to the hypothetical adult, child (age 10) and infant (age 6 m – 1 y) living (hypothetically) at the Discovery Center (dose predictions are to be released in Parts 3 and 4). For a dose reconstruction, all parameters and their uncertainties and all assumptions must be archived so that dose calculations are transparent, defensible, and can be repeated, if necessary, or changed, if new information comes to light.

As mentioned, doses are being calculated using the annual release rates and dilution factors described in this report as input to DCART. The regulatory model, CAP88-PC (Parks 1992, 1997), was used to calculate the dilution factors. Dilution factors[5] (χ/Q in s m^{-3}) for each source were calculated for the closest long-term publicly accessible location (the LLNL Discovery Center, formerly the Visitors Center – see "VIS" in Figure 1) using site-specific meteorological input also presented in this report; assumptions about the derivation of the dilution factors are described. In DCART, the dilution factors are multiplied by estimated release rates and the resulting air concentrations for each source are added together to predict concentrations of tritium in air at VIS that drive the calculation of annual doses with 95% confidence intervals.

The Discovery Center was chosen as the location at which to calculate the reconstructed doses for two reasons. First, it is very close to the UNCLE[6] Credit Union, which has been used for NESHAPs[7] compliance since 1994 as the location of LLNL's SW-MEI; second, it is the location of an air tritium monitoring station (VIS) that has sampled tritium in ambient air continuously since 1973. For the first phase of the dose reconstruction (Part 3, covering 1973 – 2005), all predicted air concentrations at VIS would be compared with the annual mean observed air concentrations at VIS. Thus, coincident with the dose reconstruction, model performance will be analyzed, which may help explain results from 1953 - 1972 (the second phase of the dose reconstruction, to be covered in Part 4) when release rates are not so well known.

For each year, all sources for which any dose impact is expected are being included in the model run. Although LLNL is not responsible for doses due to SNL/CA releases, nevertheless releases from SNL/CA must be accounted for because they contributed to measured air concentrations and consequent doses for the years the SNL/CA Tritium Research Laboratory (TRL) was operating. Minor sources are assumed included in the uncertainty on the major sources and are ignored. Every effort has been made to use realistic assumptions with the exception of dietary intake. Normal consumption of a

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[5] The term "dilution factor" will be used throughout this report to refer to the air concentration for unit source strength (or χ/Q); units are 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.

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

[7] 40 CFR Part 61, Subpart H (National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities) (NESHAPs) (USEPA 1989).

complete diet is assumed, which should assure conservative predictions because it is impossible that the entire diet could have been contaminated at the same tritium-in-air concentration as measured (or predicted) at the Discovery Center. In any case, when, questions cannot be resolved, requisite assumptions err on the side of conservatism.

The first section of this report (Background Information and Documented Release Data) discusses the history of the facilities at LLNL and SNL/CA that handle(d) tritium. The discussions are accompanied by tables that list reported releases, both routine and accidental, from LLNL and SNL/CA from 1953 to 2005 along with primary and secondary references (in footnotes) (Tables 1 – 6). Table 7a summarizes the estimated quantities of tritium released from LLNL as HT or HTO, either routinely or accidentally, between 1953 and 2005; Table 7b similarly summarizes releases from the TRL at SNL/CA, which operated between 1979 and 1995. Table 8 (also annotated) lists parameter values needed to calculate dilution factors at the Discovery Center from the facilities that released tritium. The second section of the report (Annual Site-Specific Model Input With Uncertainty) summarizes the input data for DCART and the supporting assumptions. Input data are presented in Tables 9 – 22. A description of the assumptions and rationale behind the selection of input parameters and associated uncertainties is presented in the text for each table.

Site-specific, annual input for DCART includes

- All release rates and speciation by year for releases treated as routine from all facilities. These are the release rates and associated uncertainties that are being used as input to DCART. (Tables 9 – 17)
- Dilution factors (χ/Q in s m^{-3}) at VIS for all sources of tritium. Uncertainties are provided for the dilution factors. (Tables 18 and 19)
- Observed mean annual tritium concentrations in air moisture and in air at VIS from 1973 to 2005. (Table 20)
- Absolute and relative humidity at LLNL. (Table 21)
- Observed mean annual concentrations of HTO in the water of the LLNL swimming pool, 1988 – 2000. (Table 22)
- Annual rainfall from 1952 through 2005 and input needed for the DCART precipitation model. (Appendix A)

BACKGROUND INFORMATION AND DOCUMENTED RELEASE DATA

Background information

LLNL began working with tritium late in 1952, very shortly after the laboratory opened, when a demand for a source of 14-MeV neutrons arose as a consequence of the nascent laboratory's entry into thermonuclear weapons research (Gede and Gildea 1980).

Tritium from operations is released either through stacks, from room air, or from area (diffuse) sources. Tritium, being highly mobile, will diffuse from the object of high tritium concentration to the air with a lower concentration. Thus, areas where tritium-contaminated items are stored become sources of tritium.

Major and minor sources of tritium addressed in this report (listed in the table below) are shown in Figure 1, along with the location of VIS. All sources are referred to by their present building numbers rather than those dating from the early years of facility operation. The data needed for the dispersion model (stack height or height of release, stack diameter or area of source, exit velocity, direction towards VIS [both primary and the nearest adjacent sector], distance from VIS, and degrees true north from VIS) are shown in Tables 8a and 8b for those sources used as model input. Operations, grouped by location and dates, are described below.

Building #[8]	**Stack; Years of operation**	**Room air or area source; Years of operation**
231	Tritium Facility Operations; 1953 -1958	Waste Accumulation Area (WAA); [1953 – 1958]
212	Insulating Core Transformer; 1967 - 1988	Accelerator; 1953 – 1967 Cyclotron; 1955 - 1971
514	NA	WAA; 1953 – 1960 [- 1962] Tank Farm; 1960 - 2003
331	Tritium Facility; 1959 - present	WAA; 1959 - present
Taxi strip	NA	Evaporation trays; 1962 - 1976
612	Building 624 incinerator; 1977 -1988	Container Storage Area; 1965 - present
292	Rotating Target Neutron Source; 1979 - 1987	NA
SNL/CA	Tritium Research Laboratory; 1979 - 1995	NA
695	Decontamination and Waste Treatment Facility; 2004 - present	NA

Note: Known years of operation are shown without brackets. Years of operation assumed for the TDR are in brackets.

Tritium Facility Operations in Building 231 and (probable) Building 231 Waste Accumulation Area (WAA)

In the summer of 1952, ideas for two new buildings were being discussed (Fidler 1952). One of these was the first increment of Building 231 (then Building 102)[9]. By October 1953, the building was sufficiently complete to receive shipping[10]. Tritium operations began at some point in 1953 or 1954 in the Chemistry wing (southeast corner) of Increment 1 of Building 231 in what was referred to as the “Tritium Room” (Thaxter 1954). There were apparently three rooms in the Chemistry wing: Rooms A, B and C,

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[8] 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.

[9] Floor plans dating from 1953 are found in Building 231. These were prepared by Deane & Hill, Consulting Engineers, and Albert F. Roller, Architect, all of San Francisco. Only one large room in the Chemistry wing was shown.

[10] Shipment month obtained from a memo in files archived by Lawrence Berkeley National Laboratory (Environmental Health and Safety Chronological Files and Donald Cooksey Administrative Files).

one of which must have been the Tritium Room. Air from these rooms was exhausted to two above roofline ducts. Each exhaust had a flow of 1200 cubic feet per minute (cfm) (Thaxter 1953). Work was carried out in glove boxes (Thaxter 1954) with ventilating stacks. Small diameter external stacks emerging from the windows in the building above ground level may be seen in building plans from May 1978 and in the building today. One or more of these may date to 1953, although none are shown in the building plans for that time. These stacks ran up the outside of the building and exhausted above the roof.

A memo (Thaxter 1954) mentions an additional emergency exhaust blower (5000 cfm) in the Tritium Room that could exchange the air in the room about once per minute. This implies that the room was no larger than 140 m^3. Given that the Chemistry wing was two stories high, the floor plan of the room would have been about 4 m by 5 or 6 m. This memo also discusses desirable changes to better contain and confine the operation and to ventilate the Tritium Room if any HT escaped. One of the changes proposed was to install sampling units in stacks ventilating the glove boxes to make it possible to estimate operational losses and releases to the environment.

Early operations were experiments with known quantities of HT. The amount released was the residual HT remaining in the apparatus after pumping it out and could have been estimated within ±10% of the likely value (Otsuki 2004). With the exception of a memo reporting an accidental release in 1954 (see Table 4), the only mention of releases from Building 231 prior to 1956 (see Table 1) is a roughly estimated 2000 Ci (74 TBq) per year for 1953, 1954 and 1955 (see Table 2). Presumably, the type of operations for 1953, 1954, and 1955 was similar to that of later years (Table 1). Thus there was probably a large puff release or two that exceeded 2000 Ci (74 TBq) each year. Assumptions made in preparing release rates for Building 231 may be found under the discussion of Table 9.

No evidence has been found for a diffuse area source associated with Building 231. However, given that operations with tritium were taking place from 1953 onwards at Buildings 231 and 212, and given the need for an outside location to store wastes or equipment, it has been assumed that there was a WAA, analogous to the one at Building 331, associated with Building 231. Assumptions made in preparing release rates for the Building 231 WAA are listed in Table 10.

It has been assumed that operations in Building 231 were transferred to Increment 1 of the newly built Tritium Facility (then called Building 172; now called Building 331) in early 1959 (LRL 1958).

Insulating Core Transformer, Cockcroft-Walton accelerator, and 90-inch cyclotron (Building 212)

In 1966, the Insulating Core Transformer (ICT, also known as Rotating Target Neutron Source I) was installed in Building 212. The ICT produced neutrons from the bombardment of metal tritide targets. Tritium from the ICT was exhausted through a stack, in which an ion chamber was installed in 1968 to monitor releases. Although the first releases from the ICT were apparently reported in 1968 (observe the factor of about twenty between releases for 1968 and 1967 [Table 3]), Myers (2004c) remembers that the ICT was run as early as 1966, because his measurements towards the end of the year (Myers 1966) indicated that a significant amount of shielding would have to be added to the roof to reduce neutron dose levels outside the building to reasonable levels[11]. In 1986, molecular sieve samplers were installed to obtain more detail about the chemical composition of the tritium and to eliminate the laborious task of hand integrating the plots from the strip chart recorder for the ion chambers. Operations in Building 212 produced only HT, but a small fraction of this was converted to HTO on surfaces and released. The operation of the ICT was discontinued in January 1988.

The ICT had a pit for collecting tritium-contaminated water (Dreicer 1985), but no emissions from it have been estimated.

Building 212 (when it was called Building 153) also housed the Cockcroft-Walton accelerator from 1953 through 1967. The Cockcroft-Walton accelerator was located on the west side of the ICT target pit (Myers 2004b). It also produced neutrons from the bombardment of metal tritide targets, but, because the accelerator had just 1% of the beam current of the ICT, the wooden roof provided adequate shielding. Elsewhere in Building 212 was a 90-inch cyclotron that was used from 1955 through 1971 (Newsline 1971). It too used metal tritide targets. Because no significant radioactive or other toxic effluents were anticipated from the accelerator and cyclotron, the normal building ventilation system, which probably included the accelerator caves as well, would have been adequate. Thus tritium from the accelerator or the cyclotron would have been released to room air and exhausted through small stacks projecting about a meter above the roof (Myers 2004d). Release rates with high uncertainty were apparently only estimated after the fact (Table 3); one accidental release of 200 Ci (7.4 TBq) from the cyclotron was reported in 1961 (Table 4). Assumptions made in preparing release rates for Building 212 may be found under the discussion of Table 11.

LLNL Tritium Facility (Building 331) and Building 331 Waste Accumulation Area (WAA)

Operations probably began in Increment 1 (exhausted by the south stack) of Building 331 (then Building 172) in early 1959[12]. In 1962, Increment 2 (exhausted by the north stack) became operational.

Before the stacks were monitored (see below), releases were estimated from the sample volume of tritiated gas that remained after HT was pumped through the experimental system. It was this residual tritium that went up the stack (Otsuki 2004). Early estimates of tritium releases were all "gas" (Table 1), but HT will convert to HTO if it comes in contact with metal and other surfaces, particularly if the HT remains in contact with the surface for any length of time (Souers 2004). Hence, there will be tritium emissions (mainly HTO but with some HT) not directly attributable to work taking place (Howe and Cate 1985). As a result, releases from the very beginning had to have been partly HT and partly HTO, even though all experiments involved only HT.

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[11] Shielding was added in the form of concrete blocks that were stacked up in pyramid fashion on the heavy wooden timber ceiling. The only time radiation was present outside the building was when the accelerator was in use. The neutron doses were carefully monitored outside the building and particularly along the fence line adjacent to East Ave (Myers 2006).

[12] The projected completion date for Building 172 was February 1959 (construction began in June 1958)(LRL 1958). Certainly construction was completed in 1959 (LRL 1959).

In the first quarter of 1961, a system for monitoring tritium in the stack effluent of the Tritium Facility (i.e., the south stack) was installed. The system extracted a sample of the stack gas into a 1-liter ion chamber at a point above the outlet of the stack blower. The ion current was measured by a Keithley log micromicroammeter (later called a picoammeter) and its output was fed into a recorder. Local and remote alarms set to predetermined levels were incorporated in the system. This system was calibrated using a limited number of calibrated tritium sources obtained from Oak Ridge National Laboratory (Becker 1961). Apparently, the air mover originally installed in Stack 1 was inadequate because of an insufficient sampling rate, high noise level, and excessive variation in sampling rate. In the second quarter of 1961, the system was improved by installing a Gast 0321 pump, which cured the three problems satisfactorily (Becker and Beard 1961). When Increment 2 was built and became operational in 1962, an identical stack-monitoring system was installed to measure total tritium released.

Until 1971, the stacks from Increments 1 and 2 were each monitored continuously by an analog system consisting of 1-liter ion chamber, a Keithley 413A picoammeter, an airflow indicator, and an alarm panel. A two-pen recording chart continuously recorded activity in each stack. This system reliably indicated the total tritium effluent (Silver et al. 1972b). There were two deficiencies, however. One was the need to hand-integrate the total tritium releases during a time period, although this was no problem except after an accidental release because of time constraints. A second deficiency was that, although the alarm was triggered properly by high concentration, a long release of low concentration would not trigger it. Thus in 1971 a new stack integrator was installed (Silver and Chew 1971). In 1976, in each stack there was one analog system consisting of a one-liter ion chamber, a Keithley 413A picoammeter, an airflow indicator, and an alarm panel and one digital stack-activity integrator consisting of a 1-liter ion chamber, a recycling integrator, a scaler, and a printer. The analog system could activate a flashing light and bell if the level of activity exceeded 10 Ci m^{-3} (0.37 TBq m^{-3}); the digital system would alarm when 50 Ci (1.85 TBq) were accumulated in any 15-minute period (Dow 1976). This system was designed primarily for accidental releases. It was cross-calibrated once a month by comparing the analog system’s charts with the integrator printouts (Chew and Colmenares 1973).

Tests of a new system consisting of a 2-liter and a 200-mL ion chamber were reported in 1981 (Morris 1981); somewhat later the need to expedite the completion of this new real-time stack monitoring system was noted (Morris and Ozaki 1982). This system, which is still in use in 2006, was described in the 1986 Facility Safety Procedure, but it was presumably installed closer to 1982 than to 1986. The 200-mL ion chamber, coupled with an air-flow indicator and an alarm panel, was part of an analog system primarily for alarming; the 2-liter ion chamber, along with a recycling integrator, a scaler, and a printer, was part of a digital stack activity integrator to estimate the activity released. The purpose of the small chamber was to reduce recombination effects and accompanying inaccuracy at high tritium concentration levels while the large chamber provided more accuracy and sensitivity at low concentrations (Sherwood 1982).

By the last weeks of 1971, molecular sieves had been installed to sample stack exhausts in addition to the ion chambers[13]. During 1972, it appears that only HTO was measured (i.e., only the results from one molecular sieve in each stack were found in the analytical record files; however Rich et al. (1972a) mention that the molecular sieve stack monitoring system should be installed in the south stack as soon as possible because the system installed in the north stack was performing satisfactorily.) By June the following year (Chew 1973), both HTO and HT were being reported for Stack 2, but there were still problems with Stack 1. The molecular sieve method of Östlund (Östland 1974) was used to measure the relative concentrations of HTO and HT in the stack effluent. In this method, air is drawn in series through two absorbers, each containing a molecular sieve. Stable H_2O and HTO are retained in the first trap. The second trap contains about 3% of palladium catalyst; stable H_2 and HT, which passed through the first trap, are catalytically converted to H_2O and HTO and retained in this absorber. When the traps were replaced[14], the molecular sieve was processed by heating to 550º C under vacuum, and the water and HTO were recovered in cold traps. The HTO activity was determined by liquid scintillation counting. The quantity (Ci) of HTO and HT released during the sampling period was calculated from the counting data and aliquot and total effluent volumes (Facility Safety Procedures 1989).

The processing of molecular sieves to extract the trapped HTO is time-consuming and expensive. Thus, in September 2005, four ethylene glycol bubblers were installed in the north stack of the Tritium Facility, in addition to the molecular sieves. In the bubbler system, HTO is trapped primarily in the first bottle in the series, while any additional HTO is trapped in a second bottle; HT passes through both HTO-capturing bottles and over a palladium catalyst for conversion to HTO that is then trapped by the second set of bottles. Samples are measured directly, without processing, by scintillation counting, making the bubbler system an easier, less expensive alternative to molecular sieves. A nine-week comparative study of the concentrations measured by the molecular sieves and the bubblers demonstrated that the bubblers performed as well as or better than did the molecular sieves. Starting the second quarter of 2006, bubbler data from the north stack were used to estimate the release rate from the stack. By early October 2006, bubblers had replaced the molecular sieves in both stacks (Wilson 2006).

The ionization chambers ran concurrently with the molecular sieves (and now the bubblers). In 1973 (Chew 1973), for a two-month period, the total tritium estimated using molecular sieves was 24% higher than that measured by the ion chamber in Stack 2; a 30% difference was noted in 1975 (Powell 1975), and a need for a controlled release to test the two systems was mentioned. In 1982, a recurring discrepancy in Stack 1 between ion chamber molecular sieve results[15] generated concern that the credibility of the effluent monitoring was poor (Morris and Ozaki 1982). A follow-up study (Sherwood 1982) that assumed the molecular sieve data were correct, or at least more reliable, concluded that the pulse-counter in Increment 1 had so degraded that it could not detect releases but that the ion chambers and molecular sieves in Stack 2 agreed quite

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[13] This is known from the presence of laboratory counting records in the Hazards Control archives.

[14] Replacement has occurred historically every 5 to 8 days except in 1992 when samples were collected biweekly.

[15] Releases of several hundred curies, unexplainable from an operational perspective were being detected by the molecular sieves but not by the ion chambers.

well. On the premise that the new monitoring system would soon be operative, no resources were invested in improving the old system. Presumably when the new ion chambers were installed, which was at least by 1986 (Facility Safety Procedure 1986), if not well before, better agreement was achieved between the ion chambers and the molecular sieves, at least until releases dropped in about 1991. In recent years, releases are far below the sensitivity of the ion chambers[16].

Reported release rates between 1961 and 1974 were estimated based on ion chamber data (Table 1). In 1974, records show that both stacks were monitored for HTO and HT using molecular sieves (Tables 1 and 5). Results of air effluent monitoring of Building 331 have been reported in the LLNL Site Annual Environmental Report[17] (SAER) since 1974, although speciation was not reported until 1986 (Table 5). Assumptions made in preparing release rates for the Tritium Facility may be found under the discussion of Table 12.

Stack flow rates were measured periodically (Table 8) after 1968 when three holes were drilled at 120º intervals about 20 feet above the stack breachings so that traverse velocity measurements could be done (Murrow 1968). There are many memos that describe the efforts made to measure stack flow rates accurately, including at least one monitored release of SF_6 (Wong 1989).

According to Dreicer (1985), no liquid waste from the Tritium Facility was poured down drains. It was collected in carboys[18]. Stone et al. (1982) distinguished between liquid effluent, which drains to the sanitary sewer at levels not considered to be a significant hazard, and liquid waste, which is not released to the sanitary sewer.

It is likely that there has always been some sort of temporary waste storage adjacent to Building 331 during the lifetime of the facility. As early as 1972 (Rich et al. 1972a) there was concern about the quantity of tritium stored in waste barrels at Building 331 and the need to determine if a significant portion of tritium effluent from the facility originated with the barrels. A program was started (Silver et al. 1972a) to assure that the tritium released did not constitute a health hazard. Myers (1977) noted that a considerable amount of tritium-contaminated equipment was being stored outside Building 331. Plans needed to be made for the safe storage of this and other waste on site before the waste was relinquished to storage at the Taxi Strip. Chew (1977) also expressed concern about the build-up of contaminated equipment around the loading dock and apron area of Building 331. Dreicer (1985) mentions a waste pickup area in use in 1984 on the east side of Building 331.

Since September 1991, the WAA adjacent to Building 331 (or a storage room after the WAA was closed in early 2003) has been monitored with an ambient air tritium sampler

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[16] Because ion chambers do not have the sensitivity to accurately measure low release rates, ion chamber estimates of low release rates are more than an order of magnitude higher than those from molecular sieves. This estimate was based on weekly emissions in Ci calculated from ion chamber data compared with those calculated from molecular sieve data. Ion chamber and molecular sieve data were obtained from the annual facility-specific air effluent spreadsheets for 1996 through 2003. These spreadsheets are maintained by the Terrestrial Atmospheric Monitoring and Modeling Group with the support of data management.

[17] SAERs may be found in the LLNL Library "Documents Online" under UCRL-[TR]-50027-yy, where "yy" is the last two digits of the year.

[18] Carboys contained somewhat higher concentrations of radioactive liquids than did the retention tanks (Odell and Toy 1979) (see Building 514 History in this report).

that uses silica gel as the collection medium. Monitoring by the Terrestrial and Atmospheric Monitoring and Modeling Group (TAMM) was begun to estimate releases from the WAA using dispersion modeling. In addition, the Tritium Facility makes independent estimates of releases based on facility knowledge (i.e., what is expected), swipes, and occasional measurements of off-gassing. The estimated annual release rate reported for NESHAPs and for the SAER from the WAA is a value derived from input from these information sources (Table 3). Assumptions made in preparing release rates for the Building 331 WAA before the air tritium sampler was installed may be found under the discussion of Table 13; assumptions after the sampler was installed may be found under the discussion of Table 14.

A 1987 memo (Homann 1987) described the methodology apparently used at the time to estimate the tritium inventory contained in Building 331 waste drums: the tritium emission rate from a small piece of tritium-impregnated stainless steel was measured, and afterwards the tritium inventory was determined by dissolving the steel in hydrochloric acid.

Building 514 History

Building 514 (originally Building 127B) was built in 1943 by the United States Navy[19]. Solid waste was known to be stored outside Building 514 presumably from sometime after 1952 until 1960 (Odell and Toy 1979; Dreicer 1985). In 1960, Building 514 was converted into a treatment facility for large volume, low-level liquid radioactive waste and became known as the Tank Farm. Carboys of similar description to those stored at the Taxi Strip (see below under "Solar Evaporation Trays") accumulated there and were being disposed of, cleaned up, and organized in 1963 (Tyler 1962). In addition, an experimental evaporation pit existed south of Building 514 before solar evaporation was carried out on the Taxi Strip (Dreicer 1985). Estimated release rates and their uncertainty for the Building 514 Yard are listed in Table 10.

At the Building 514 Tank Farm (1960 – 2003), both above ground and in-ground tanks were used to store liquid wastes from Building 331 and other facilities. Wastes were released in a controlled manner to the sanitary sewer system if they met permitted criteria for tritium (e.g., in 1979, 20 mCi [7.4 10^8 Bq] d^{-1} [Odell and Toy 1979]). (Based on retention tank records from Hazards Control archives, there is no evidence that tritium was analyzed in the retention tank water between 1968 and 1972; only alpha and beta were analyzed. Tritium was apparently analyzed in some samples by November 1973. By 1975 it was analyzed routinely.) Wastes that did not meet the criteria for release to sewer were stored in the 123,000 L (113,562 L [Dreicer 1985]) surge tanks for later treatment or disposal. In 1979, 189,000 L of tritium-contaminated water was being stored at Building 514. Over the years, soils at the tank farm were contaminated with leaked tritium; tritium concentrations at two locations were as high as 240 pCi g^{-1} soil at depths between 0.6 and 1.5 m in late 2004 when the site was undergoing decontamination and decommissioning. Most of the area tested in 2004 had concentrations of less than 5 pCi g^{-1} soil (Fish 2006).

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[19] The U.S. Navy purchased the land that is now the Livermore site for a flight training base on March 24, 1942 (Dreicer 1985).

Potential releases of tritium from the Tank Farm to the atmosphere were assessed each year between 1991 and 2003 for NESHAPs compliance. Release estimates based on radionuclide inventory ranged over three orders of magnitude from a minimum of 1.4×10^{-7} Ci (5,180 Bq) for 1993 to a maximum of 5.1×10^{-4} Ci ($1.89\ 10^{7}$ Bq) for 1992; the mean for the years 1992 – 2003 was 1.2×10^{-4} Ci ($4.44\ 10^{6}$ Bq). These estimates are one-thousandth of the actual annual inventory because they have been reduced by the Environmental Protection Agency's (EPA) physical state factor whereby emissions are estimated from radionuclide inventories depending on their physical states for use in dispersion/dose assessment models.

To estimate release rates for years when the inventory was not prepared is quite difficult, even when records of concentrations in tanks are available. A complete set of records (tank number, tank volume, sampling dates, and tritium concentrations on those dates) is available for 1975; a partial set of data is available for 1976. Tanks were apparently filled over fairly short time periods, their contents tested, and the contents released to the sewer if they met permitted criteria (e.g., in 1975 some samples were not released because they were too high in copper). In 1975, 75 Ci (2.78 TBq) of tritium passed through Building 514[20]. The contents of the three tanks with the highest quantities of tritium - 14.6 Ci (0.54 TBq), 12.3 Ci (0.46 TBq), and 5.8 Ci (0.21 TBq) - were not released to the sanitary sewer, and there is no record of how long the contents of the tanks remained at Building 514. When the tritium concentrations recorded on known sampling dates for each tank in 1975 were weighted by month, the mean annual inventory for the Tank Farm in 1975 became 5 Ci (0.19 TBq). This number itself is undoubtedly inflated because the tanks were sometimes emptied without any tritium being present; in this estimate, it was assumed that the tank contained tritium until it was once again emptied and refilled with tritium-contaminated liquid. A similar exercise could not be done for 1976 because of insufficient data. The median total quantity of tritium in the tanks at time of emptying for 1975 was just 0.41 Ci (0.015 TBq); for 1976, the median was 0.21 Ci (0.0078 TBq). The 5 Ci (0.19 TBq) estimated for the 1975 inventory is 10 times the highest inventory submitted for NESHAPs (0.51 Ci [0.019 TBq] in 1992). The annual release rate for modeling this 5 Ci (0.19 TBq) inventory would be 0.005 Ci (1.9×10^{-4} TBq). An estimated dose even this high to the hypothetical MEI or SW-MEI should be very small compared with other sources. Thus, the Building 514 Tank Farm has been considered a minor source and will not be included as a source for the TDR.

In the early 1980's, a thin-film evaporator was installed at Building 514 (it was under construction in 1979 [Odell and Toy 1979]). One of the reasons it was installed was to reduce releases of tritium to the sanitary sewer. This evaporator was operated through the mid 1980's. It was included in the routine swipe program in 1985 (Radiation Safety Program 1985). Dreicer (1985) mentions 8,300 gal (4 Ci; 0.15 TBq)) of tritiated water that were evaporated over the course of one year (probably 1984). This source is assumed to contribute insignificantly to dose and has not been included in the TDR.

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[20] This number was obtained by summing the quantities observed in all tanks on all sampling dates throughout 1975. In some cases, it appears as if an occasional tank might not have been emptied and refilled between sampling dates. Because of this, the 75 Ci estimate may be high.

Solar evaporation trays

Solar evaporation was used as a method to treat and dispose of wastes from 1962 through 1976 (Buerer 1983). Except for an initial experimental pit set up south of Building 514 (Dreicer 1985), solar evaporation occurred on the Taxi Strip in the southeast of the site, where carboys[21] of liquid waste from the Tritium Facility and elsewhere (e.g., ICT and the Biomedical Program) were stored (Buerer 1983). The early evaporation pits were depressions in the ground lined with boards that supported a plastic liner. They were replaced by 10-foot by 20-foot fiberglass trays (Kerns 1998), which were constructed and torn down over their operational life. Later, liquid wastes were evaporated from monolithic concrete units (10 feet x 20 feet x 1 foot deep above ground) coated with 12 mils of polyamide cured epoxy paint and usually lined with polyvinylchloride (Buerer 1983). Two concrete trays were equipped with rolling covers. When the liquids were nearly evaporated to dryness, dry-sorb® was mixed in using rakes and the residue was shoveled into drums for disposal (Kerns 1998).

Records of concentrations of tritium[22] and volumes of water evaporated have not been found. Potential evaporation from each tray could equal the average annual evapotranspiration rates for towns in California with similar meteorological conditions to Livermore. Based on evapotranspiration rates for eleven locations[23] obtained from the California Irrigation Management Information System, Department of Water Resources, Office of Water Use Efficiency on the world-wide web, the evaporation rate for Livermore could be about 1.34 m y^{-1} of water. A potential 24 m^3 could be evaporated in a year from a single evaporation tray of 18 m^2, assuming the tray was kept filled with water at all times.

According to Dreicer (1985), although five trays could be seen in an aerial photograph from 1970, the number of trays being used at any one time is not known. The total volume of liquid evaporated therefore cannot be estimated with any degree of certainty. However, it would seem that, once the backlog of wastes had been evaporated, one 10 x 20 x 1 foot evaporation tray would have been sufficient to evaporate all liquid wastes produced annually at LLNL. If wastes were generated at 1-3 carboys (20 – 60 L) per week, as in 1959 (Buerer 1983), even at the high end, only 3000 L (3 m^3) of this type of waste would have been generated annually.

It seems odd, however, that so many evaporation trays would have been needed to evaporate such a small amount of liquid, so perhaps the amount of liquid being generated (and evaporated) was greater than the quantity generated in 1959. If that were the case, the evaporation trays may be important to dose, particularly as a diffuse source quite close to the perimeter of the site. Releases from the evaporation trays have been estimated (see the discussion under Table 13).

Carboys and contaminated equipment continued to be stored at the Taxi Strip even after solar evaporation had ceased (Chew 1977; Snyder 1978). Asphalt and dirt with only minor contamination were removed from the Taxi Strip area in 1981 (Patterson 1981),

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[21] 1500 carboys had accumulated by 1959.

[22] In 1959, the contents of 200 carboys were sampled for gross alpha and gross beta only (Buerer 1983).

[23] Davis, Dixon, Brentwood, Carneros, Morgan Hill, Twitchell Island, Concord, Manteca, Modesto, Patterson, and Tracy.

although there were still portions of the Taxi Strip that had not been surveyed (Toy 1981). A depth profile in 1982 (Ruggieri 1982) taken near the solar evaporation trays indicated a peak tritium concentration of about 4 µCi (150,000 Bq) L^{-1} at about 3.5 m. Cobalt-60 and ^{241}Am contamination well above background levels were found at about 1 m depth but not deeper. Tritiated water, however, would have percolated downwards with rain. This contamination is probably associated with disposal pits found on the Taxi Strip (Buerer 1983) rather than being related directly to any activities associated with the solar evaporation trays.

Waste disposal: Container Storage Area (Building 612 yard) and Building 624 incinerator

Building 612 was constructed in 1965 as a location for the packaging of solid radioactive waste and toxic waste for shipment. After 1977 and the discontinuation of the solar evaporations ponds, carboys of liquid waste and drums of solid waste were assembled in what became the Building 612 Yard (Figure 1). Liquids with trace amounts of tritium would be incinerated in Building 624; most other liquids would be sent to the Building 514 Tank Farm, with solids being sent to Building 612 (Odell and Toy 1979). Tritiated wastes were stored in transportainers stacked no more than two high (because of earthquake risk) around the yard. The need to monitor tritium releases from the Building 612 Yard arose when LLNL had to demonstrate compliance with NESHAPs. In August 1991, an air tritium monitor was installed in the Building 612 Yard. Since then, release rates for NESHAPs compliance have been estimated by back-calculating a release rate from either the annual mean or median measured concentration of tritium in air. Assumptions made in preparing release rates for the Building 612 Yard before the air tritium sampler was installed may be found under the discussion of Table 13; assumptions after the sampler was installed may be found under the discussion of Table 14.

The incinerator at Building 624 was used between 1977 and 1988 (Radian Corporation 1989) or February 1978 to February 1989 (Ridley et al. 1992). It was a dual chamber unit consisting of an ignition chamber, combustion chamber, and a liquid injection and solid waste feed system. The liquid injection system was installed to inject the contents of the glass carboys directly into the incinerator (Radian Corporation 1989). The maximum capacity of the incinerator was 480 gallons of liquids per day or 1.6 tons of solids per day (Ridley et al. 1992). Stack monitoring may have been evaluated (Myers and Gordon 1987) but was not implemented. The incinerator underwent removal and disposition in 1992 (Winstanley 1992). Stack parameters are known quite well, but release rates are not. Liquid and solid wastes containing carcinogens with concentrations of tritium less than 1 µCi g^{-1} ($3.7\ 10^4$ Bq g^{-1}) (considered trace quantities [Odell and Toy 1979]) may have been incinerated. Waste that was incinerated included non-aqueous tritium-contaminated waste such as liquid scintillation fluid and other contaminated organic fluids. Tritium releases were limited to administrative levels in 1981 (Radian Corporation 1989) of a maximum of 0.5 Ci (0.019 TBq) per day, and the number of days that burns could take place was limited to 100 annually (Godwin 1988). Wastes were only incinerated on Tuesdays, Wednesdays, and Thursdays (Ridley et al. 1992). Assumptions made in preparing release rates for the Building 624 incinerator may be found under the discussion of Table 15.

Rotating Target Neutron Source II (Building 292)

Building 292 was the home of the Rotating Target Neutron Source II (RTNS II) that was operational from 1979 to 1987. As at the ICT, metal tritide targets were bombarded by a beam from an accelerator with consequent release of HT. The goal for limiting radioactive releases at RTNS II was 300 Ci (11 TBq) per year of tritium (Kintner 1981). Although titanium tritide particles were a potential source of contamination, it is unlikely that any would have been released to the atmosphere because they would have stuck to the internal surfaces of the stack (Trent 1998). The exhaust stack was monitored with Overhoff ion chambers, providing real-time data, and a silica gel sampler. The Overhoffs were calibrated every six months by LLNL's Plant Engineering Department. No tritium was normally used in the calibration, but in 1981 a known amount of tritium gas showed a response within 20% of the expected value (Hazards Control 1982). The facility was equipped with an effluent scrubbing system, which converted HT to HTO using a room-temperature catalyst and then trapped the HTO in molecular sieves (Schumacher 1980). The tritium output from the accelerators was estimated to be 20 to 30 Ci (0.74 to 1.1 TBq) per hour when both accelerators were operating. The scrubbing system was very efficient, with output concentrations being a factor of 10^6 to 10^7 lower than the total input tritium concentrations (Schumacher 1980) (99.9995% efficient; Facility Safety Procedures 1985). Although the scrubbing system was very efficient, the increased flow that occurred during rough vacuum pumping, when the scrubbing system was bypassed, was exhausted directly to the stack. Nearly all of the releases from the building occurred during this process; about 98% of the tritium released during the rough pumping operations was in the form of HTO (the conversion to HTO was internal to the accelerator rather than being converted in the effluent recovery system) (Myers 2004a). The primary effluent monitoring system consisted of a silica gel sampler that adsorbed the HTO being exhausted (Logan et al. 1980). Assumptions made in preparing release rates for Building 292 may be found under the discussion of Table 15.

In 1984 (Dreicer 1985), waste for Building 292 consisted of tritium-contaminated solvents, spent tritium targets and cylinders from the catalytic scrubbers. Tritiated solvents were incinerated; other waste was packaged and sent to the Nevada Test Site.

Building 292 was also equipped with an underground retention tank. If the concentration was below 10 mCi (3.7×10^8 Bq) total activity, the contents could be released to the sanitary sewer (Collins 1989). During tank testing in July 1989, a leak was discovered in the tank system (Mallon 1995). Tritiated water had migrated from the tank through the soil and was being evapotranspired by local vegetation. Although the tank was drained by November 12, 1991 (Mallon 1991), soil moisture was still contaminated with tritium (e.g., about 36,000 pCi L^{-1} [1330 Bq L^{-1}] in piezometer UP-292-001 in May 1994 [Hoffman et al. 1994]), as was the local vegetation. Because of this tritium diffusing to the atmosphere, the Building 292 area was treated as a diffuse source for NESHAPs compliance.

For 1992, 1993, and 1994, release rates for NESHAPs compliance were based on measured concentrations of tritium evapotranspired by the trees and soils in the area. In 1995 and 1996, release rates were estimated using measured ambient tritium in air moisture monitored by an air tritium sampler that had been installed in August 1991[24]. Between 1991 and 1996, the median annual air moisture concentration measured at the sampler fell about two orders of magnitude from 49,000 to 500 pCi L^{-1} (1810 to 18.5 Bq L^{-1}). The highest dose to the hypothetical MEI at the perimeter of the site from the Building 292 diffuse source, calculated for 1993, was 2.5×10^{-5} μSv (2.5×10^{-6} mrem) y^{-1}.

From 1997 through 2004, needles from one particular pine tree (PIN1) with the highest concentrations were collected first monthly and then quarterly; the highest concentration (8800 pCi L^{-1} [326 Bq L^{-1}]) was observed in September 1997. After sampling of the pine needles began, the tree was treated as a diffuse source of tritium for NESHAPs purposes, with the maximum dose to a hypothetical individual living at the nearest perimeter location being 1.7×10^{-5} μSv (1.7×10^{-6} mrem) in 1997. In 2003, dose calculations for NESHAPs using PIN1 as a source were discontinued because LLNL obtained permission from the EPA to demonstrate compliance by using monitoring data in place of modeling dose from releases from small sources. The tree succumbed to an infection of red turpentine beetles in 2005. A core representing the body burden of the tree was analyzed for HTO and OBT. Concentrations of 2.18 pCi (0.081 Bq) g^{-1} HTO and 12.3 pCi (0.456 Bq) g^{-1} OBT were found. Because the total tritium concentration of 14.5 pCi (0.537 Bq) g^{-1} was greater than the 5 pCi (0.185 Bq) g^{-1} tritium concentration permitted at local landfills, the tree was taken to the Nevada Test Site for disposal.

Based on dose predictions using these approaches for the Building 292 diffuse source, it was decided not to include this diffuse source in the dose reconstruction. The Building 292 diffuse source contributed at most 0.007% of the dose contributed by the Building 331 stacks alone to the SW-MEI.

Decontamination and Waste Treatment Facility (Building 695 complex)

Livermore's Decontamination and Waste Treatment Facility (DWTF) began operations in September 2003. DWTF is an integrated facility for storing and processing hazardous, low-level radioactive, transuranic, radioactive, or mixed wastes. The facility includes new indoor storage areas, and solid and liquid waste processing facilities. Activities that took place at the Building 514 Tank Farm or the Building 612 complex have been or will be transferred to the DWTF. Essentially all releases of tritium from the DWTF should be through the building stack, although some transportainers containing tritium may be stored outside and serve as a very minor diffuse source. Stacks were monitored for 18 days in November 2004 and have been monitored continuously since mid-February 2005 using ethylene glycol bubblers; samples are collected weekly.

Assumptions about the derivation of estimated release rates for HT and HTO releases for 2004 and 2005 are found in the discussion of Table 16.

Sandia National Laboratories Tritium Research Laboratory

The SNL/CA Tritium Research Laboratory (TRL) was operational from 1979 to 1995. In November 1979, SNL/CA installed ethylene glycol bubblers to sample releases from the TRL stack (Hafner 2004). Releases could have been characterized as either HT or HTO at this point, although they were reported as total tritium until 1983. The ethylene glycol

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[24] The sampler was removed in July 2003.

bubblers were in series. The first set of bubblers trapped HTO. HT passed through the first series of bubblers, came in contact with a palladium catalyst, was converted to HTO and was then trapped by the second series of bubblers. There was also an Overhoff ion chamber. Assumptions made in preparing release rates for the TRL may be found under the discussion of Table 17.

In addition to the TRL, there were small releases from tritium operations at SNL/CA that took place in B913 from mid-1970s until about 1984 (Hafner 2004). Other facilities where tritium was handled are summarized in Garcia and Gorman (1996).

The TRL also had an evaporator that was permitted to evaporate up to 100 Ci per year (Department of Energy 1999) of tritium-contaminated wastewater to the environment. Between 1990 and 1995 this system evaporated 97,500 gallons of extremely low-level tritium-contaminated water with a total tritium content of 31.06 Ci. For at least 1990 and 1991, the contribution from evaporation (22.7 and 1.4 Ci respectively) was included in the total releases for the year from the TRL (Garcia and Gorman 1996). Waste water from 1992 through 1994 was also evaporated (1992: 0.65 Ci; 1993: 1.95 Ci; 1994: 4.39 Ci), but it is not clear (Garcia and Gorman 1996) whether or not these releases were included in the total reported stack releases for those years. The quantity of tritium evaporated, whether included in the stack releases or not, was always very small compared with the quantity of tritium released from the stack (Hafner 2005).

Documented tritium releases (Tables 1 – 7)

All release data are presented in the units in which they were reported. All references for the data reported in the tables have been cited. Although some sources may be more reliable than others, the chance that an informal, hand-written value, for example, may actually be the most accurate cannot be discounted.

Reported quantities (1956-1979) of total tritium (HT and HTO) released from Lawrence Livermore National Laboratory (Table 1).

Data reported from 1956 through 1971 were obtained from internal memos from the Chemistry Division to the Director's Office or from memos, originally marked "secret", "restricted data", or "confidential", to the United States Atomic Energy Commission (US AEC). The classification of all memos was changed to "unclassified" in 1975. After 1971, the memos were not classified. Memos were found in the LLNL Hazards Control Department archives.

Sometimes the memos or reports did not clearly state whether or not the releases listed in Table 1 truly represented releases from all facilities or only releases from the Tritium Facility. Certainly for the early years, it appears that the releases from the Tritium Facility are the only ones reported to the AEC, although they purport to be laboratory-wide releases (compare Table 1 with Table 2). It is likely that the magnitude of early releases from the accelerator and cyclotron in Building 212 (see Table 3) was not considered large enough to report relative to releases from the Tritium Facility; furthermore, the releases from Building 212 at that time were from room air.

No memos to the AEC have been found for the earliest releases before August 1956. Early known releases were tritiated hydrogen gas (HT) and occurred as discrete releases on known dates. In mid-1961, reporting changed from mention of discrete events to quarterly reporting of "controlled releases." As well, mention is made of releases from the mass spectrometer and the 90-inch cyclotron, which were located in Building 212 (then Building 153).

Accidental as well as routine releases are included in the list in Table 1.

Quantities of total tritium (HT + HTO), both routine and accidental, released from the LLNL Tritium Facility (Table 2)

The term "Tritium Facility" used here embraces the operations that were carried out in Building 231 (then Building 102) in 1953-1958 before being moved to Building 331 (then Building 172), the present Tritium Facility. Data under the "Memos" column from 1953 to 1971 came from a 1973 memo from LLNL to the United States Atomic Energy Commission (Olsen 1973) estimating past releases from the Tritium Facility. This memo was generated on fairly short notice by D. Myers (2004e) and others and didn't involve a detailed review of records. It primarily involved the health physicists asking senior people at a given facility to "guesstimate" tritium emission rates for the years when data were not readily available. For the presentation to the Director's Office (Souers 1988), the numbers seem to have been quite well researched and are very similar to those in Table 1. There are also differences between what was reported in the LLNL Site Annual Environmental Report (SAER) and other sources, although mostly these differences are negligible.

Reported releases from most LLNL tritium sources other than the Tritium Facility (Table 3)

All sources and release rates after 1974 shown in this table were published in the SAERs, unless otherwise noted. Releases for Building 212 through 1972 were obtained from the Olsen (1973) memo.

Tritium from the Cockcroft-Walton accelerator and cyclotron would have been released to room air between 1953 and 1971; tritium from the ICT was exhausted through a stack from about 1967 onwards. Releases from Building 212 were largely HT, although some conversion to HTO must have taken place, as demonstrated by the speciation shown for 1986 and 1987. The transition from estimated release rates for room air to measured stack releases was probably not instantaneous, as might be implied by the abrupt shift in reported emissions from an approximate value (10 ± 50% Ci) in 1967 to a more precise value (240 Ci) in 1968. Also, in 1982, a release of 44 Ci was reported in the SAER as well as in a hand-written summary of the monthly releases (Radiological Air Effluent Records 1982); unfortunately, the correct sum obtained from the hand-written list of monthly releases was 34 Ci, not 44 Ci.

Acute releases of tritium from LLNL (Table 4)

This list of acute tritium releases has been included with the summary of routine tritium release rates for LLNL for the sake of completeness and because the upper limits of dose from acute releases will be estimated in Part 5 of the TDR. All the incidents shown in Table 4 occurred at the LLNL Tritium Facility, with the exception of the ones in 1954

(Building 231) and 1961 (Building 212). In addition, Otsuki (2004) remembered the very first accidental release at LLNL in 1953 that involved an accelerator making neutrons in Building 212 (then B153). Before the large accidental release in 1965, the acute releases shown in Table 4, with the exception of the accidental release in 1954, were reported as normal operations because they were planned puff releases (Table 1). As sensitivity grew to the quantity of tritium released at any one time, the larger routine puff releases were redefined as accidental (Souers 1988); by 1989, any unplanned tritium release greater than 100 Ci was required by LLNL internal controls to be reported, at least within LLNL (Facility Safety Procedures 1989).

Speciation of routine releases from the LLNL Tritium Facility (Table 5)

Table 5 summarizes the available information about the speciation of releases from the Tritium Facility obtained from spreadsheet or hardcopy records of stack monitoring data, the SAER, and assorted memos. The "spreadsheet" speciation from 1974 through 1995 was calculated in spreadsheets using laboratory analytical measurements of tritium in the water extracted from the molecular sieves and stack flow rates taken from the last available stack flow measurements (Table 8a). In theory, these spreadsheet calculations should be identical with those in the memos or those reported in the SAER. In practice, they can be quite different for some of the following reasons:

- The assumption (used for the spreadsheet calculations in Table 5) that release rates were calculated using the most recently measured stack flow rates is not necessarily true. For example, in 1988, the estimated radioactivity released was calculated using the 1985 release rate (cubic feet per minute) rather than the most recent measured release rate for 1987 (Biermann 1989). Similarly, the spreadsheet calculation used the flow rates from 1987 to determine the 1987 releases, while the official stack effluent report used the flow rates from 1985 (Mansfield 1987a). Apparently the Tritium Effluent Release Spreadsheet used at the time (Mansfield 1987b) had cells for the flow rates that were not necessarily updated as new flow rates were measured.
- When release rates were prepared by personnel involved with day-to-day activities, suspect data were probably removed; in preparing the spreadsheets, suspect data may not have been recognized as such and therefore may have been included in the releases. One particularly fine example of this is found in a very detailed memo (Mansfield 1990) in which it is explained that apparently high release rates for two periods (2/21 - 2/28/89 and 6/27 -7/03/89) were actually artifacts of stack calibration operations when a measured quantity of HT was injected directly into the stack monitoring system, thus simulating a relatively large release. In this case, the high results from the spreadsheet calculations have been removed for these time periods, but similar corrections may not have been made for other years.
- In 1981, between April 27 and June 1, the molecular sieves in Stack 1 were reversed. Thus no information was collected on the amount of HT released; HTO and HT were both trapped by the same sieve. The assumptions made to account for this will result in different totals of HT and HTO.
- For a twelve-week period in 1978, the order in which HT and HTO results were reported in the analytical data sheets was reversed from the rest of the year. When the spreadsheet was prepared, the analytical results were taken at face value.

However, a hand-written summary of 1978 release rates did not account for the change in order of results for those twelve weeks. This suggests that the analyst either did not notice the change or knew that the laboratory had inadvertently applied the wrong descriptor with the results.

- Sometimes there are two analytical reports for what is apparently the same sample. The preparer of the spreadsheet can only take the result that looks most consistent with all others, but this might not be the right decision.
- Some errors appear to be due to rounding or transcription.

There is no way to resolve the differences pointed out above. This independent preparation of release rates directly from analytical data provides a check on the published release rates and an insight into potential uncertainties in reported release rates. The bottom line, however, is that, even though fairly large discrepancies may exist for individual sampling periods, the totals for the year are not significantly different.

In 1995, flow sensors were installed in the Tritium Facility Stacks so that flow rates would be monitored continuously. From 1996 to the present, air effluent spreadsheets for the Tritium Facility have been prepared by the TAMM analyst, with the support of data management personnel. Stack flows used for these calculations are the real-time flow measurements collected every two hours and averaged for the same duration as the sample. Thus, for these years, there is little more than rounding error between the values reported in the SAER and the values from the spreadsheets.

Stack effluent emissions from the SNL/CA Tritium Research Laboratory (Table 6)

Data on SNL/CA's TRL obtained for the TDR were limited to documents in the public domain. Differences are essentially rounding only except for the large discrepancy between the SAIC data for 1982 – 1985 (Science Applications International Corporation 1993) and the published SNL/CA data. Although the SAIC values are likely incorrect, if the release rates for the SAIC and SNL/CA data are summed for 1982 – 1985 and compared, the SAIC total is 86% that of SNL/CA's. The dose for this period will therefore be similar regardless of release rate used. Accidental releases from SNL/CA to the environment were always included in the total reported release rate for the year.

Estimated total tritium released between 1953 and 2005 (Tables 7a and 7b)

Based on the emission data referenced in Tables 1 – 5, an estimated 792,000 Ci (29,300 TBq) of tritium (either as HTO or HT) have been released to the atmosphere from LLNL from 1953 through 2005 (Table 7a). This estimate has a 2.5% confidence limit of 672,000 Ci (24,900 TBq) and a 97.5% confidence limit of 914,000 Ci (33,900 TBq). About 75% of the total was released accidentally as tritiated hydrogen gas (HT) (Table 7a) in the two large releases of 1965 and 1970. Nearly all of the routinely released tritium came from the Tritium Facilities (Building 231 up through 1958 and Building 331 from 1959 onwards). Tritium was also released from the SNL/CA's TRL between 1979 and 1995 (Table 6). An estimated 7,420 Ci (275 TBq) were released from the TRL, with a 2.5% confidence limit of 6,610 Ci (246 TBq) and a 97.5% confidence limit of 8,240 Ci (305 TBq); about 61% of total tritium released was HTO (Table 7b).

ANNUAL SITE-SPECIFIC MODEL INPUT WITH UNCERTAINTY

Parameters for dispersion modeling (Tables 8a and 8b)

Dispersion for routine releases was modeled using CAP88-PC. The parameter values used in CAP88-PC as input to calculate dilution factors are shown for stacks in Table 8a and for area sources in Table 8b.

Parameter values for stack sources are known with reasonable certainty. Most stack heights are either those used for compliance with NESHAPs or were measured specifically for the TDR. Exit velocities do vary and cause variability in the dilution factor, but the variability in dilution factor due to the exit velocity is less than the variability of the exit velocities.

Stack flow rates are usually measured in cubic feet per minute. Exit velocity is easily calculated from flow rates as long as the interior diameter of the stack is known. All exit velocities for the Tritium Facility were derived in this manner. Exit velocities for Building 292 were mentioned explicitly in Heikkinen (1985), Trent (1986a) and Surano et al. (1993); the other exit velocities shown for Building 292 were calculated based on stack flows and an inside stack diameter of 0.71 m (Heikkinen 1985).

Because work was carried out in glove boxes, air from operations in the Tritium Room of Building 231 was probably exhausted through stacks much like those of 1978 or today. The stacks would have been somewhat higher than the roofline. A stack height of 9 m has been assumed based on the height of the present stacks and the height of the building. Although the diameter of the stacks currently emerging from what used to be the Tritium Room is 0.41 m, to determine the uncertainty on the exit velocity for Building 231 for the TDR, NESHAPs reports were surveyed for representative glove box stack diameters and exit velocities, because the true stack diameter and exit velocity are unknown. Air was also exhausted from the room via ventilation fans in the roof, although this would presumably not have been a source of tritium under normal circumstances.

To model area (diffuse) sources, CAP88-PC requires input for the height of the source, the area of the source, and the estimated exit velocity from the source. The dispersion calculation, however, is independent of values for area and exit velocity. Thus, for LLNL’s area sources (Table 8b), only the estimated release height is given.

All sources, except for the evaporation trays, have been located relative to VIS using the Global Positioning System (GPS). The location of VIS from each GPS-located source is given as the sector in which the sampler lies and the closest adjacent sector (in parentheses). In a sector-averaged model like CAP88-PC, concentrations change, sometimes abruptly, at the sector boundary, while real air concentrations do not exhibit this characteristic. To better assess potential air concentration at a specific location, concentrations have been calculated as a weighted mean of the contributions from both sectors. Bearings shown are the direction from the source to the receptor. The use of GPS to locate area sources implies a certainty about the locations that does not in fact exist.

Derivation of annual release rates for the Tritium Dose Reconstruction (Tables 9 – 17)

All annual release rates are shown in units commonly used at LLNL. These units (Ci) were used as input for DCART and were converted to becquerels for all calculations. Concentrations and doses calculated by DCART are in SI units.

Overall percent uncertainty on most release rates encompasses various different sources of uncertainty, which were combined by taking the square root of the sum of the squared percent standard deviations for each individual source. The resulting distributions were normal. Each normally distributed release rate was used as input to the Crystal Ball®[25] software in DCART. When the uncertainty was large, the lower limit of the distribution, which was negative, was truncated at zero.

Generic uncertainty

Due to the common ways release rates are measured or due to a general inability to reconstruct information about historic technologies, or both, some uncertainties will be the same regardless of the source of the tritium. To avoid unnecessary repetition, these generic uncertainties will first be discussed here independently of the sources to which they apply.

Uncertainties for which general assumptions can be made include those for

- Ion chambers
- Sampled air
- Calibrated exit velocity
- Exit velocity for years not measured
- Flow rate through stack tritium samplers
- Silica gel correction factor

Ion chambers

For the first years that stacks were monitored before molecular sieves or silica gel samplers were used to speciate the releases, release rates for Building 331 and Building 212 were based on measurements obtained from ion chambers.

The ion chambers in the Building 331 stacks were calibrated and cross-calibrated during the early years; after 1973, release rates estimated using the ion chambers were compared to release rates estimated using the molecular sieve data (see the discussion of the Tritium Facility under "Background information"). However, no specific information about the performance and reliability of the ion chambers in Building 331 that can be applied to the uncertainty estimates on the release rates has been found.

Some information was found for the Building 212 stack. In mid-1971 (Hazards Control 1971), a known release of 26.7 mCi of HTO was measured as 24.8 mCi by the ion chamber; the difference was due to tritium absorption in the ductwork, hood, and high

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[25] A risk analysis software package (Decisioneering, Inc. 1515 Arapahoe Street, Suite 1311, Denver Colorado USA 80202) that provides uncertainty and sensitivity analyses for spreadsheet codes, such as DCART.

efficiency filter. In 1975 (Hazards Control 1976), 2 Ci of HT were introduced over 65 minutes to the system to determine a stack calibration constant of 1.73 10^9 Ci/A-min; it was noted that this calibration constant depended on a set flow rate (0.275 m^3 s^{-1}) and that a variation in flow rate could have resulted in an inaccurate estimate of tritium discharges.

Uncertainty on release rates measured by ion chambers has been estimated to be as high as ± 80% (Till 2001). For this TDR, the uncertainty applied to ion chamber measurements was ± 14% based on an efficiency of the ion chamber counting system of ± 10% and variations in temperature and pressure that may affect the result (±10%); these uncertainties are associated with ion chambers in use at the Savannah River Site (SRS) that are different from those at LLNL. Till (2001) concluded no uncertainty due to manual integration or to electronic integrators, given the procedures at SRS. There is no reason to believe that the ion chambers at LLNL would not have been calibrated and maintained in excellent working condition, just as they were normally at SRS. Furthermore, the Building 212 measurements seem to support an uncertainty of ± 14%. Uncertainty on the current ion chambers or others that ran concurrently with molecular sieves or silica gel stack sampling systems is not relevant to the TDR because emission data were obtained from the molecular sieve or silica gel sampling systems.

Sampled air

No matter the type of sampling system, some uncertainty will be associated with whether the sampled air represents the air being released out of the stack; this is important when flow rate is used to calculate the release rate. In 1981 (Industrial Hygiene Group 1981), a test in Building 331 using Freon 12 demonstrated that measured concentrations of sampled air were within 8% of the generated concentrations in both stacks, indicating that the air samples reaching the molecular sieve traps represented gas levels in the building exhaust. In line with this, Biermann (2004) suggested an average uncertainty on sampled air of ± 10%, which was applied to all stack samples.

Calibrated exit velocity

No matter the type of sampling system, the volume of air released per unit time must be known before release rates can be estimated properly; this applies to all stacks where the flow rate is used to calculate the release rate. The exit velocity is directly related to the volume of air exhausted; when freshly calibrated, the uncertainty on exit velocity is about ± 3 - 5% (Biermann 2004); for the dose reconstruction, it was assumed to be ± 5%.

Flow rate through stack tritium samplers

When stack sampling for tritium using molecular sieve or silica gel, the flow rate through the sampler will vary as conditions change unless a mass flow controller is used. If uncertainty in the total flow is to be avoided, the controller should have a flow totalizer. For molecular sieve and silica gel sampling[26], mean flow rate through the sampler was determined by measuring the flow rates at the beginning and end of the sampling period using a rotameter and calculating the total flow from the average of the two readings and the sampling period.

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[26] As mentioned, tritium released through stacks is now sampled using bubblers.

Until 1999, like the stack tritium samplers, estimation of total flow through the ambient air tritium samplers was based on rotameter readings of the flow rate at the start and end of the sampling period. Rotameter readings continued being taken even after the flow totalizers were installed. Based on data for total flows determined by both methods, the uncertainty on the air volume that passed through the sampler was estimated to be about ± 14% (Peterson 2004). It has been assumed that this uncertainty also applies to the tritium samplers in stacks, although it is overly conservative for total flow through bubblers.

Silica gel correction factor

A correction factor of 1.6 ± 15% (based on 1600 samples from the ambient air monitoring network) was applied to all tritium obtained from silica gel sampling prior to 2001, when a correction factor was developed (Guthrie et al. 2002). The correction factor is necessary because silica gel, dried as much as possible without losing structural integrity, nevertheless still contains about 5% exchangeable water by weight. Thus, the concentration of tritium measured in water extracted from the silica gel will be lower than that of the air moisture collected by the silica gel (Rosson et al. 1998; Rosson et al. 2000), because the collected tritium will be diluted by water in the silica gel that contains only background levels of tritium. The magnitude of the correction depends upon the amount of water collected compared with the amount of water bound in the silica gel and is specific to the type of silica gel used by LLNL for ambient air monitoring for the past few years. This correction factor and uncertainty has been applied to silica gel stack sampling due to lack of knowledge about the specific types and mass of silica gel used historically. A few numbers published in the literature[27] indicate that the fraction of latent water in other silica gels is similar to that of LLNL and might even indicate the necessity for a slightly higher correction factor for types of silica gel used in the past. However, without actual measurements, even the mean correction factor for historic silica gel cannot be known with definite accuracy.

Derivation of release rates and uncertainty for Tritium Facility operations carried out in Building 231 (Table 9)

Normal operations during the early years of LLNL produced puff releases (Table 1) that, when greater than 1,000 Ci or so, should strictly speaking be modeled as acute. However, because of the fairly random occurrence of these early releases and the impossibility associated with reconstructing the actual meteorological conditions for each puff, all routine releases for each year will be modeled as if they were chronic. Depending upon meteorological conditions, treating an acute release as chronic may result in either under- or overestimations of dose to the SW-MEI, but, for Building 231, the uncertainty applied to assumptions about release rates (see below) should be large enough that doses are not underestimated.

Uncertainty on the release rates from Building 231 must account for uncertainties on

- reported release rate
- speciation

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[27] Percent water from references cited in Guthrie et al. 2002: LLNL baked silica gel, 5.12; other gels, 5.3, 5.8, 5.9, and 6.3.

Very little is known about releases from Building 231 in the very early years of LLNL. The memo to the US AEC (Olsen 1973) gives uncertainty of ± 50% on Building 331 releases that date to before 1959 when Increment 1 of Building 331 became operational[28]. As noted by Myers (2004e), the estimates provided for the Olsen memo were hurriedly compiled and given a large uncertainty to account for lack of documentation. A comparison of estimates from the Olsen memo with what was reported in LLNL quarterly memos to the US AEC between 1956 and 1963 indicates that annual releases were 2.7 ± 60% times greater than assumed in the Olsen memo. Thus the estimates of tritium released between 1953 and 1955 have been assumed to be 2.7 times the Olsen estimate of 2000 Ci (i.e., 5400 Ci) with an uncertainty of ± 60%. For these years, because the releases are assumed to be large puff releases, it has been assumed that 10% of the total tritium released was HTO and that there is a -0.4 correlation between releases of HT and HTO. This is a comparable percentage to that obtained using the assumptions (below) for 1956 - 1958. Uncertainty on the HT and HTO release rates is ± 60% for both. The lower limit for both HT and HTO was truncated at zero.

For the years 1956 through the end of 1958, the assumptions for Building 231 are somewhat different. A judgment as to the magnitude of possible releases when data were missing (Table 1) was impossible, given the sporadic nature of the early releases. The steps to prepare the input data were as follows:

1. The "best estimate" of total tritium released was assumed to be slightly greater than that reported because some releases, however small, occurred during the quarters for which releases were not reported.
2. Speciation was estimated based on 1% conversion to HTO within the facility for known large releases and 50% conversion to HTO within the facility for known smaller releases (≤ 1000 Ci). A negative correlation of -0.4 was assigned to the distributions for releases of HT and HTO, because the greater the release of HT, the smaller the fraction of HTO released.
3. To account for possibly high release rates for quarters when releases were not reported, the standard deviation of the best-estimated annual release rate of total tritium was increased until the upper limit of the distribution, determined using the Crystal Ball® software, equaled the potential maximum release rate (i.e., the best estimate plus the highest observed quarterly release for the particular year used as the quantity released for each missing quarter). The value for the standard deviation derived this way was then used as the standard deviation for the estimated annual releases of HT.
    - 1956: If it is assumed that 3,000 Ci were released for each of the missing quarters, then, to include this potential maximum release (4,000 + 2 x 3,000 = 10,000 Ci) within a normal distribution, the standard deviation must be set at 2,000.
    - 1957: If it is assumed that 5,000 Ci were released during the missing quarter, then, to include this potential maximum release (12,000 + 5,000 =

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[28] Olsen (1973) apparently used the term "Building 331" in place of "Tritium Facility", which would have referred both to Building 231 prior to 1959 and to Building 331 after that date.

    17,000 Ci) within a normal distribution, the standard deviation must be set at 1,900.

    - 1958: If it is assumed that 3,000 Ci were released during each missing quarter, then, to include this potential maximum release (6,000 Ci) with the best estimate of 6,250 Ci (for an estimated maximum of 12, 250 Ci) within a normal distribution, the standard deviation must be set at 2,000.

4. A lower bound of uncertainty for the HT portion of the release was set using the Crystal Ball® software by assuming 10% uncertainty on the total best-estimated releases of 4,000, 12,000, and 6,250 Ci for 1956, 1957, and 1958 respectively. This lower limit, which was truncated, thus became
    - 1956: 2,800
    - 1957: 8,400
    - 1958: 4,375.
5. It is assumed the fraction of HTO does not change with increased potential release of HT. Uncertainty for HTO is selected so that the upper limit of the distribution equals the upper limit of the HT distribution times the estimated fraction of the total release that was HTO:
    - 1956: The upper confidence limit can be calculated: 0.1325 x 10,000 = 1,325. Thus the standard deviation of the 530 Ci released must be 265 for the upper limit of the normal distribution to equal 1,325. The lower confidence limit of the distribution was truncated at zero.
    - 1957: The upper confidence limit can be calculated: 0.051 x 17,000 = 870. Thus the standard deviation of the 610 Ci released must be 90 for the upper limit of the normal distribution to equal 870.
    - 1958: The upper limit can be calculated: 0.116 x 12,250 = 1,420. Thus the standard deviation of the 725 Ci released must be 240 for the upper limit of the normal distribution to equal 1,420.

For releases from Building 231, exit velocity has no effect on the uncertainty of the release rate because releases were estimated based on usage rather than on stack flow measurements and concentrations in stack air.

One accidental release of HTO occurred at Building 231 in 1954 (Table 4). This release was modeled separately and will be reported in Part 5 of the TDR.

Derivation of release rates and uncertainty for Building 231 WAA and the Building 514 Yard (Table 10)

The existence of the Building 231 WAA is hypothetical, but it is considered analogous to the Building 331 WAA. It would have served the same function for the Building 231 operations as the Building 331 WAA did for the Building 331 Operations. The Building 231 WAA was assumed to be in existence between 1953 and 1958 when Building 231 served as the initial location of Tritium Facility operations at LLNL. It has been assumed that the annual release rate for the WAA when operations were established was 0.025 times the total tritium released from the Building 231 stack; this value is a simplified approximation of the relationship observed between the Building 331 WAA and the Building 331 total tritium releases (see below under the discussion of Table 13). Because the operation was new in 1953, it was assumed that release rate from the WAA was just 25% of the release rate for established operations; in 1954, this was raised to 50%, and in 1955, the percentage was set at 100%. In DCART, release rates from the WAA were assumed correlated with the total tritium released from the Building stack ($r^2 = 0.5$). The uncertainty on the estimated release rates from the WAA was calculated from the considerable uncertainty on the release rates of HT and HTO from Building 231.

For this TDR, it has been assumed that some of the solid waste stored at Building 514 was tritiated and that the Building 514 Yard was analogous to the Building 612 Yard. Although the storage of solid wastes was apparently terminated in 1960 at the Building 514 Yard, there is no record of any other waste treatment/storage area until the evaporation trays were used in 1962 (see below). Therefore, to assume that wastes were stored at Building 514 in the years before the existence of any other known waste storage area is a reasonably conservative assumption. Thus it has been assumed that, although there was a gap of two years between when the Tank Farm was established at Building 514 and when the Evaporation Trays were used, the solid waste remained at Building 514 for these two years. The release rates from equipment and waste assumed stored in the Building 514 Yard were estimated based on the assumption that the annual release rate from the yard was 4% of the HTO released from the both Tritium Facilities. This is the same assumption used for the Building 612 Yard (see below under the discussion of Table 13). Because the operation was new in 1953, it was assumed the release rate for 1953 was 25% of the estimated annual release; in 1954, 50% of the annual release rate was assumed. From 1955 through 1961, the release rate was assumed 4% of the HTO released from the Buildings 231 and 331 stacks. In DCART, release rates were correlated ($r^2 = 0.4$) with HTO releases from the Tritium Facilities. Uncertainty estimates were based on the uncertainty on the release rates and speciation for releases of HTO from Buildings 231 and 331.

Derivation of annual release rates of HT and HTO and uncertainty for Building 212 (Table 11).

Releases from Building 212 fall into two periods. During the first period, through 1967, HT was released to room air from the Cockcroft-Walton Accelerator and the 90-inch cyclotron; during the second period, from about 1968 on, HT was exhausted up a monitored stack from the ICT, although the cyclotron was in use until 1971. Releases from the cyclotron were small relative to those from the ICT, and for dose estimation between 1968 and 1971, it has been assumed that any releases from the cyclotron are accounted for by the uncertainty on the ICT release rates.

Uncertainty on the release rates for the room air releases from Building 212 includes uncertainties on the reported release rate and the speciation. Uncertainty on the release rates for the stack releases from Building 212 includes uncertainties on

- reported release rate
- exit velocity for years not measured
- speciation

- analysis of the tritium content in the water extracted from molecular sieves
- generic uncertainties from above, when applicable.

The only estimates found for releases from Building 212 from 1955 through 1972 are from the memo to the AEC in 1973 (Olsen 1973; Table 3); no estimates are available for the releases from the Cockcroft-Walton Accelerator that certainly was running in 1954 and possibly began running in 1953. Release rates for the Olsen memo would have been estimated based on the initial quantity of tritium on the tritium targets and the initial neutron yield, both of which were known. As the neutron yield dropped with target usage, the tritium on the target was assumed to drop proportionately. Thus, if the neutron yield dropped by a factor of two, one could assume that half the tritium had been released from the target (for release to atmosphere) (Myers 2004c). Emissions for the Olsen memo would have been estimated retrospectively based on the number of targets used, hence the high uncertainty (± 50%) years after the fact. For this TDR, a correspondingly lower release rate in 1954 for the accelerator was estimated because targets used for the accelerator were much smaller than those used for the cyclotron (Myers 2005). An even smaller release rate was estimated for 1953 to account for the small probability that some experiments on the accelerator might have been completed that year.

Releases from the accelerator and cyclotron were entirely HT, but, because nothing is known about the speciation after conversion on surfaces, an uncertainty for speciation of ± 50% for these years is assumed. Because the accelerators and the ICT used similar tritium targets, it has been assumed that the fraction of HTO of the total release should be the same as for the ICT (12%; see below).

The reduction of releases in the period 1965 through 1967 probably was due to reduced use of the Cockcroft-Walton accelerator while the ICT was being brought on-line (Myers 2004c). The date of installation of the ion chamber in the Building 212 stack is not known but is implied from the suddenly high release rate without uncertainty in 1968 (Table 3). Confidence in the reported release rate after monitoring was initiated is high (± 5%, based on the long-term average for Building 331). Myers (2004c) expressed surprise that the release rate estimated for 1967 was as low as 10 ± 50% Ci, because operations of the ICT started in 1967 (Booth 1967). It is likely that estimated emissions for 1967 from the Olsen memo were only those from the Cockcroft-Walton accelerator or cyclotron. Equally likely, the ICT, being operational, would have released some tritium through its unmonitored stack in 1967. Therefore, for 1967, an additional release from the ICT has been assumed, with a uniform uncertainty for total tritium of 5 – 50. In addition, no record of releases has been found for 1973 (see Table 3), so a uniform distribution for total tritium was assigned of 50 – 375 – 600 to account for releases either similar to 1972 or 1974.

No information on the measurement of stack flow rates has been found for the ICT, but the same parameter values were used every year between 1974 and 1982 to estimate release rates. This could imply confidence in the actual stack flow rates, and perhaps the confidence was based on measurement. Even when measured, of course, there is some change over time, so an uncertainty of ± 10% has been applied for each year when the exit velocity is known; for the years when the exit velocity is unknown, the uncertainty applied is ± 20%.

Releases were probably nearly entirely HT, although some conversion to HTO would have taken place as evidenced by measurements in 1986 and 1987, which suggest that much less HT was converted to HTO in this operation than occurred in the Tritium Facility (see below). The fraction of the total release that was HTO in 1986 and 1987 was about 12%, and this percentage has been applied to all releases with an uncertainty of ± 50% for the years the ICT operated and monitoring was done with ion chambers. No uncertainty is assumed for speciation for the two years of data based on molecular sieve sampling.

The uncertainty on the analysis of water collected from the molecular sieves is based on a complete record of counting errors for the Building 212 samples in 1986 and 1987 obtained from Hazards Control archives. The maximum analytical uncertainty (1 σ) was 5.6% for HTO in 1986; the lowest was 0.8% for HT in 1987.

<u>Derivation of annual releases of HT and HTO and uncertainty for the LLNL Tritium Facility (Building 331) (Table 12)</u>

In addition to generic uncertainties, the release rates from the Tritium Facility includes uncertainties on

- reported release rate
- whether the release was emitted from Stack 1 or Stack 2
- speciation
- exit velocities for years the stack flow was not measured
- analysis of the tritium content of the water extracted from molecular sieves.

Contributors to uncertainty and the magnitude of uncertainty for releases from Building 331 depend upon the year of release and how the release was estimated or monitored. Between 1959 and 1961 when in-stack tritium monitoring began, releases were estimated based on the known amounts of HT used for each experiment.

Although Building 331 was not completed until about February 1959 (LRL 1958; LRL 1959), it has been assumed for the TDR that all tritium releases for 1959 were from the new Tritium Facility. For both 1959 and 1960, all releases are accounted for, but some are simply termed "negligible". When releases were reported as negligible, it was assumed 23 Ci were released each month[29] (based on Fleming 1961 and Foster 1961). For each of these two years, this meant an additional 115 or 138 Ci from five or six months of negligible releases. The uncertainty on the negligible portions was assumed ± 50%, while the uncertainty on the other puff releases was ± 10%. Thus the uncertainty on the reported release rate was ± 10.5% for both years. Speciation was assumed 1% HTO for known large releases and 70% for the negligible and potentially very small releases. Uncertainty on this speciation was ± 40%. Release rates of HT and HTO were assumed negatively correlated ($r^2 = -0.4$).

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[29] On July 28, 1961, Fleming (1961), in a memo to the Director's Office, reported that approximately 60 to 70 Ci had been released since the end of April. On August 1, 1961, Foster (1961), the LLNL Director, reported to the US AEC that "only negligible amounts" of tritium had been released to the atmosphere in the quarter ending July 31, 1961. To obtain the "negligible release rate" of 23 Ci, 70 Ci for the quarter was divided by 3 (months).

From 1959 through June 1962, quarters were staggered – that is, instead of quarters ending March, June, September, and December, they ended April, July, October, and January. This means that some of the releases allocated to one year may actually fall in another. This does not affect the dose when summed over several years, but it may make the dose for one year higher or lower than it would have been had the actual releases for a calendar year been known. For 1961, the “+” shown in Table 1 indicates only that the year is not exact, not that all releases have not been accounted for. Thus for 1961, just 800 Ci ± 15 % has been assumed released from Stack 1. In the first quarter of 1961, stack monitoring began using ion chambers. Uncertainty on the speciation was assumed ± 25%, which is the same as the speciation observed in stacks monitored by silica gel (see below). The additional uncertainty associated with stack sampling has been included in the overall uncertainty on the release rate for 1961 as well.

Because the records of releases are complete from 1962 onwards (with the exception of 1969 when three months of releases are not accounted for), the uncertainty on the reported release rates was determined entirely by averaging the annual totals obtained from different memos or reports (Tables 1 and 5). In 1969, a release rate for the missing months of data was estimated by assuming that the unknown release equaled the quarterly average of the known three quarters. The standard deviation obtained from averaging reported release rates accounts for uncertainty due to the use of possibly out-of-date flow rates when calculating release rates. As an example, the difference between the flow rates for 1985 and 1987 is about 8% for the South Stack and 1% for the North Stack, 1987 being higher. In most cases, there is no way to determine what stack flow was used to calculate the release rate or how it relates to the known calibration of the stack flows, but the uncertainty is included in the standard deviation when different values for release rates are averaged. If all reported release rates were the same for a year, the uncertainty on the reported release rate was assumed zero.

Acute releases of HT (1965, 1966, 1970, 1984, 1985), over 1000 Ci and recognized at least in a memo, have been subtracted from the reported annual release rates. When reports of accidental releases varied in magnitude, these different values were subtracted from the annual total for the year so that this source of uncertainty was accounted for when averaging reported routine release rates for the year. A release of 24,000 Ci HT in 1964 (Table 4) was modeled as accidental, but its existence is in doubt because it has only been found mentioned by one source (Souers 1988); the routine release rate for 1964 was assumed to equal the quantities reported in the memos to the AEC (Table 1), although the 10,000 Ci release reported between May 1 and July 31, 1964 may have been acute. There was only one HTO release from Building 331 that might be considered accidental and would need to be modeled separately. Although apparently real (450 Ci in 1981; Table 4), so little is known about it (e.g., duration) that it was modeled as part of the routine releases; if the release had been spread over a couple of days, as it may have been, it would have resembled a routine release as much as an accidental one.

The largest uncertainty based on reported values for a Building 331 release rate is for 1970. This uncertainty (± 41%) is high because, when the various values for the accidental release in 1970 are subtracted off estimates of total released, the estimated quantities released routinely are quite different. Uncertainty, notably after 1993, can be as low as ± 0% for variation in reported release rates.

For 1962, it was assumed releases in the first half of the year came from Stack 1 (South), while the releases for the second half of the year were divided between Stack 1 and Stack 2 (North). Both stacks are essentially collocated with respect to VIS, but, because the exit velocity of each stack is different (Table 8), the dilution factors will be different (Table 19). Thus, to best predict tritium concentrations in air and consequent dose at VIS, the release rate of tritium from each stack must be estimated. Based on data from 1974 through 1993, when the quantities of tritium released from each stack were reasonably proportional year-after-year, it has been assumed for the second half of 1962 through 1973 that 38% (± 17%) was released from Stack 1 and 62% (± 29%) was released from Stack 2.

From 1961 and thereafter until stack releases were speciated in 1974, releases were spread fairly uniformly over the year (Table 1). It was assumed, therefore, that the speciation based on the known history of Building 331 (1974 - 1993) would apply to 1961 through 1974. The speciation observed between 1974 and 1993 is summarized below:

Total Ci released from one stack per year	Fraction HTO	Fraction HT
>630	0.54	0.46
400 - 629	0.64	0.36
80 - 399	0.69	0.31
2 - 79	0.88	0.12

Between 1959 and 1973, the annual release rate from each stack was always greater than 630 Ci except for Stack 1 in 1972 (506 Ci). Thus speciation was assumed always to be 54% HTO and 46% HT except for Stack 1 in 1972, when it was assumed that 64% of the release was HTO and 36% was HT. Since 1974, confidence in the quantities of HT and HTO released from each stack of the Tritium Facility is high because releases have been carefully monitored.

To determine the quantities of HT and HTO released from each stack for the years before 1974 when the releases were first speciated, the annual estimated release of total tritium was divided first between the stacks, and then the quantities of HT and HTO released from each stack were estimated. Negative correlations (-0.4 for Stack 1 and -0.5 for Stack 2) were assigned between the distributions of HT and HTO release rates.

Flow rates measured in the Tritium Facility stacks vary over time (Table 8). Given that holes were drilled in the stack in 1968 so that transverse velocity measurements could be taken (Murrow 1968), flow rates prior to 1968 were probably estimated based on rates expected from the air-flow regulation system (Becker and Beard 1961). For years when records of flow rates have not been found, the two values flanking the missing data have been averaged to obtain the exit velocity; the associated uncertainty is the percent standard deviation of the two values. Prior to 1968 and after 1997[30], flow measurements were either not taken or flow rate was not calibrated for several years. Before 1968, the

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[30] Flow velocity sensors were installed in 1995 and calibrated in 1995 and 1997. Flow has been monitored continuously since the flow sensors were installed. Calibration is not required for low-level releases.

exit velocity as well as its uncertainty had to be estimated for both stacks; for 1998 – 2005, mean annual exit velocities are known but uncertainty had to be estimated. For Stack 1 before 1968, an exit velocity of 5.45 m s^{-1} ± 10% has been assumed; for Stack 2 before 1968, an exit velocity of 10.9 m s^{-1} ± 15% has been assumed. For 1998 – 2004, uncertainty is assumed to range from ± 5% in 1998 to ± 16% in 2005. The basis for this assumption is the observation that uncalibrated velocity probes drift about 5% per year (Wilson 2005). Mean uncertainty on the estimated exit velocity (or stack flow) annually over the period when measurements were taken regularly is about ± 5%.

Finally, the analytical uncertainty based on actual counting errors on water collected from molecular sieves since 1974 has been applied. For each year, an average of all errors was taken. This approach may result in a higher mean than would occur if the errors were weighted on the basis of Ci released per sampling period, given that percent errors are smaller the greater the concentration of the sample. Analytical error ranges from insignificant in the earlier years with high release rates to up to 100% for HT or HTO samples from either stack in recent years.

Derivation of tritium release rates and uncertainty for the Building 331 WAA, the evaporation trays, and the Building 612 Yard before monitoring occurred (Table 13)

The Building 331 WAA is assumed to have been in existence for as long as the Tritium Facility. For the years prior to August 1991 and the start of ambient air tritium sampling at the WAA, the empirical relationship between annual total tritium activity released from Building 331 and mean annual concentration (pCi m^{-3}) observed at the nearby ambient air tritium sampler between 1991 and 2003 has been used to estimate the release rate for the Building 331 WAA. Based on this, and disregarding units, the air concentration (pCi m^{3}) at the sampler location 85 m ENE of the WAA can be estimated by multiplying the total Ci released annually from Building 331 by 0.253 ± 71%. The dilution factor from the WAA source at the air tritium sampler, based on meteorological input from 1999 - 2003 used in CAP88-PC, is 3.32×10^{-4} s m^{-3} ± 30%. Using the estimated air concentration at the sampler and the dilution factor, the release rate at the WAA can be back-calculated. The known uncertainties on the Building 331 stack releases of HT and HTO also contribute to the overall uncertainty on the WAA release rates. Overall uncertainty on the WAA release rates for the years 1959 to 1990 approaches or exceeds 80%. It has been assumed that, for the first two years of operation of the facility, the release rate of the WAA was only 50% that calculated above because accumulation of wastes would have occurred gradually over time. Release rates from the Building 331 WAA were correlated ($r^2 = 0.5$) in DCART with both the HT and HTO releases from the Building 331 stacks.

The evaporation trays were used from 1962 to 1976. Part of the uncertainty on the estimated release rates is due to the large uncertainties about the amount of water evaporated annually from the evaporation trays. To account for this, the uncertainty on the volume evaporated was defined as a triangular distribution with a minimum of 3, a midpoint of 20 and a maximum of 120 m^3 (per year) (see the discussion of evaporation under “Solar evaporation trays” earlier).

Some assumptions about what was in the waste can be made. Based on Buerer (1983), it might be assumed that liquids high in the types of solids (like copper) that cannot be released to the sewer would have been evaporated, because the liners of the trays were rolled up with the residual solids for disposal. Given that permitted concentrations of tritium were released into the sewer and high concentrations of tritium would have been treated as waste, it can be assumed that liquid with high concentrations of tritium was not evaporated and that any tritium that was evaporated was probably not the primary reason the waste was a candidate for evaporation.

Because there is no record of a waste storage area on the Livermore Site from 1962 through 1964, it has been assumed, probably extremely conservatively, that the annual release rate from the evaporation trays was equivalent to the release rates from either the Building 514 or 612 Yards (i.e., 4% of the HTO released from the Tritium Facility). This assumption is based on two factors: 1) the need for an interim location for stored waste between the years when waste was stored at the Building 514 and Building 612 Yards, and 2) the back-log of carboys with liquid waste that presumably were disposed of as rapidly as possible. In DCART, release rates between 1962 and 1964 from the evaporation trays were correlated ($r^2 = 0.4$) with HTO releases from the Tritium Facility stacks.

To determine the release rate from the evaporation trays from 1965 onwards, a triangular distribution for the concentration of tritium in the water (minimum of 100, an apex of 2.3×10^4, and a maximum of 1×10^7 Bq L^{-1}) was selected. The minimum is similar to the median on-site tritium concentration in air moisture in 1973; the midpoint of this distribution is about thirty times the current standard for tritium in drinking water (740 Bq L^{-1}); 1×10^7 Bq/L[31] was the mean concentration of tritium in the tanks at Building 514 in 1975. These relatively low concentrations were chosen for the distribution based on the earlier discussion about what kinds of wastes would have been evaporated. When the parameter distribution for concentration in water evaporated is multiplied by the distribution for volume of water evaporated and sampled 25,000 times using the Crystal Ball® software, the derived release rate becomes 4.3 ± 4.15 Ci for a normal distribution left-truncated at 0. This is the value assumed for the release rate from the evaporation trays when both the evaporation trays and the Building 612 Yard coexisted.

The Building 612 Yard historically has held containerized waste. Records of the contents of the containers may exist, but they will not provide estimates of how much tritium was off-gassed annually from containers. It is reasonable to expect that the quantity of wastes stored in the Building 612 Yard was related to the releases of tritium from the Building 331 stacks, because historically the Tritium Facility dominated releases from the LLNL site. To arrive at the estimated release rates shown in Table 13, known release rates from the Tritium Facility between 1993 and 1997 (years when little tritium was handled at Building 612 that did not come from onsite) were correlated with annual mean observed ambient air concentrations at the air tritium sampler (B624) in the Building 612 Yard. The estimated release rate for the Building 612 Yard, obtained by back-calculating from the observed mean annual air tritium concentration at B624 for those years (see under discussion of Table 14), was about 4% of the HTO released from the Tritium Facility

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[31] This seemingly high concentration is nonetheless much lower than the air moisture equivalent concentration of the derived air concentration (DAC) (National Bureau of Standards 1959; Eckerman et al. 1988) for workers. The DAC was $2\ 10^{-5}$ µCi mL^{-1} ($7.41\ 10^5$ Bq m^{-3}) of air; using an annual mean absolute humidity of 8 g water m^{-3} air, the equivalent concentration in air moisture is $9.26\ 10^7$ Bq L^{-1}.

stacks. If this relationship holds over the history of the Tritium Facility, then estimates can be made of releases from the Building 612 Yard before ambient air tritium measurements began. A triangular distribution calculated from 1%, 4% and 6% of the HTO released from the Tritium Facility describes the estimated release rate for the Building 612 Yard. Added to this uncertainty is the uncertainty on the annual Building 331 release rate. This additional uncertainty was taken into account by multiplying the uncertainty on the estimated annual HTO releases from Building 331 times the triangular distribution just described in Crystal Ball® for 10,000 runs with Latin Hypercube Sampling. The results are roughly normal and are shown in Table 13 (and used as input for 1977 through 1992) as normal with ± one standard deviation.

For the years 1965 through 1976, it has been assumed that the sum of release rates from the evaporation trays and release rates from the B612 Yard should only equal the assumed 4% of the HTO released from the Tritium Facility. There is no reason to assume that more "waste" tritium was generated at LLNL just because there was another means by which it was disposed. Through the use of the Crystal Ball® software, distributions of release rates for the Building 612 Yard were calculated by subtracting the tritium estimated to have been released annually from the evaporation trays from the Building 612 Yard release rates estimated at 4% of the annual Tritium Facility releases.

For the DCART calculations, a correlation coefficient (r^2) of 0.4 was applied between the Building 612 Yard release rate and the Building 331 HTO release rates. In addition, release rates for the evaporation trays and the Building 612 Yard were assumed 100% negatively correlated in DCART to maintain the relationship of the release rate from the evaporation trays plus the release rate from the Building 612 Yard equal to 4% of the HTO release rate from the Tritium Facility.

<u>Derivation of annual tritium release rates and uncertainty for the Building 331 WAA and the Building 612 Yard estimated from ambient air monitoring (Table 14)</u>

Sources of uncertainty on releases from the Building 331 WAA and the Building 612 Yard include uncertainty from

- Dispersion model
- Analysis of the tritium content of the water extracted from silica gel.

From 1992 and 1993 for the Building 331 WAA and the Building 612 Yard, respectively, to the present, as part of NESHAPs compliance, data obtained from ambient air tritium samplers placed near these facilities have been used to estimate release rates from their area sources. Obviously, some of the tritium in air at those locations is due to other sources at LLNL, but most of the tritium captured by the samplers does come from the nearby diffuse sources (see tests of CAP88-PC in the NESHAPs reports[32]). Annual estimates of off-gassed tritium can be obtained by back-calculating from the mean[33] observed air concentration to the nearby tritium source using a dilution factor calculated

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[32] NESHAPs reports may be found in the LLNL Library's "Documents Online" as UCRL-ID-113867-yy, where "yy" is the last 2 digits of the year.

[33] As reported in the SAER through 2001, the estimated release rates were based on the median air concentration. However, because CAP88-PC predicts mean air concentrations, the release rates reported here have been recalculated based on means.

by CAP88-PC. Uncertainty on the back-calculation must account for uncertainty on the silica gel correction factor (1.6 ± 15%), the dispersion model (± 30%), the flow through the sampler (± 14%), and the laboratory analysis of tritium concentrations in the extracted water (< ± 10%). Total uncertainty for each year was estimated at about ± 35%.

As mentioned in the section on Background Information, the published estimates of tritium released from the Building 331 WAA are agreed upon based on TAMM calculations and Facility knowledge. However, if the release rates for the Building 331 WAA determined this way for NESHAPs compliance are plotted against measured air concentrations, no correlation is found between release rate and median or mean air concentration. Thus, estimating the release rate by back-calculation may be less uncertain (or at least have more easily quantified uncertainty) than estimates derived as they were for NESHAPs. For the dose reconstruction, therefore, release rates have been back-calculated from the mean air concentration at the air tritium sampler using annual meteorological files for each year and applying the silica gel correction factor.

If the estimated release rates for the Building 612 Yard are plotted against the annual median or mean concentration of tritium in air at the B624 air tritium sampler, there is an excellent correlation between the points, as is expected, because the Building 612 release rate has always been calibrated to observed concentrations at the B624 air tritium sampler. Values prior to 2001 shown in Table 14 are 1.6 times higher than those published in NESHAPs reports and SAERs because of the silica gel correction factor. In addition, the release rates, which were estimated based on median air concentrations between 1993 and 2001, have been multiplied by the mean/median air concentration ratio for each year so that they now are based on mean air concentrations.

Derivation of annual release rates and uncertainty for the Building 624 incinerator and Building 292 (Table 15)

Release rates from the incinerator in Building 624 were only reported for the final three years of operations (Table 3). Hoyt (1989) tabulated total curies per day of all nuclides incinerated in 1988, providing a very accurate assessment of releases for that year. It is not clear how the release rates of 1985 and 1986 were arrived at, and there are discrepancies, particularly for 1985 (see Table 3). Release rates for the remaining years of incinerator operation have been estimated for the TDR based on the daily measured release rates in 1988 (Hoyt 1989). For a triangular distribution, the minimum, mean, and maximum released in one day in 1988 has been multiplied times the 100-day burn limit to give the minimum, peak, and maximum of the distribution. Obviously, the absolute maximum release possible is the 0.5 Ci per day limit times the 100 day per year limit on the use of the incinerator, but this is a highly unlikely maximum.

Even though the releases from the Building 624 incinerator could have had minimal impact on the dose to the hypothetical SW-MEI, because reasonable confidence can be placed in the uncertainty of the release rate, incinerator releases were modeled as part of the TDR. To err on the side of being conservative, releases from the incinerator were modeled for 1977, based on the information provided by the Radian Corporation (1989), even though Ridley et al. (1992) mentioned that start-up was February 1978.

For Building 292, in addition to generic uncertainties, uncertainty on the release rates includes uncertainties on

- stack flow estimates
- reported release rates
- speciation
- analysis of the tritium content in the water extracted from silica gel.

No documentation of stack flow measurement at Building 292 was found, although presumably this was done regularly. Air effluent volumes were given in the air effluent notebooks for 1979 and 1981, but how exit velocity was derived from them was not noted[34]. Heikkenin (1985) reported a nominal exit velocity of 11.1 m s^{-1} obtained from a stack area of 0.4 m^2 (which corresponds to the NESHAPs inside stack diameter of 0.71 m) and a flow rate of 4.4 m^3 s^{-1}. Trent (1987), however, used an exit velocity of 9.1 m s^{-1} and a stack diameter of 0.965 m to obtain a flow rate of 6.65 m^3 s^{-1} for 1986. It would seem that the stack diameter used by Trent was the outside rather than the inside diameter that should have been used to calculate flow rate. The release rate derived for 1986 using the outside stack diameter of 0.965 m was 106 Ci (Table 3); a 1986 release rate derived for the TDR using the inside stack diameter was 58 Ci. Discrepancies such as these can cause release rate estimates to be highly uncertain. In the 1987 calculation of 1986 release rates, the exit velocity appears to be a measured, rather than derived, value and has been taken as the correct exit velocity for 1986 (Table 8). Trent (1986a) reported a stack flow rate for 1985 of 10.5 m^3 s^{-1} used to derive the release rate. If exit velocity had been calculated from this flow rate based on outside diameter, the exit velocity would have been 14.4 m s^{-1}; based on an inside diameter, it would have been an unlikely 26.5 m s^{-1}. Given the unknowns for the Building 292 stack, all exit velocities (except 26.5 m s^{-1}) shown in Table 8 have been averaged to obtain a mean exit velocity that represents all years with an uncertainty of ± 31%. The assumption that release rates for other years were based on the inside stack diameter may be wrong.

An uncertainty (± 5%) has been applied to the reported release rate based on experience with Building 331, because finding agreement between NESHAPs reports and SAERs does not mean there is no uncertainty on the release rate. There is additional uncertainty about the 1986 release because the silica gel sampler was disconnected for about a month (Myers and Silver 1986; Trent 1986b). Although no releases were detected by the ion chambers, releases could have occurred, because the ion chambers were used for alarming purposes and would not have detected less than alarm levels. Based on average release rates and down time, the 1986 release rate for the TDR has been revised upwards by 10% with an additional 10% uncertainty.

It has been assumed that 100% of the tritium released from Building 292 was HTO. No uncertainty has been applied to this assumption. One measurement showed that the fraction of HTO released during rough pumping (Schumacher 1980) was about 98%, thus indicating that only a small amount of HT would have been released. Given that under normal operations about 75-85% of the tritium input to the scrubber was observed to be

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[34] Exit velocities for 1979 and 1981 shown in Table 8 were derived using the stack flow and the inside diameter (0.71 m diameter) of the Building 292 stack obtained from NESHAPs data.

HTO, it is reasonable to assume that 100% of the tritium released from Building 292 was released as HTO. The predicted dose will be on the conservative side if the assumption is incorrect.

Silica gel was used to sample the stack effluent. Thus the release rates are estimated at 1.6 times those reported in the SAERs, with an uncertainty of ± 15%. The silica gel stack sampler at Building 292 was sampled monthly. Mean annual analytical uncertainty for sampled air moisture was obtained by averaging analytical uncertainties from laboratory data sheets. Analytical uncertainty is given as two sigma, but one sigma was used in the uncertainty analysis. Analytical uncertainty was highest in 1986 at just over 1%.

Derivation of annual release rate and uncertainty for the Decontamination and Waste Treatment Facility (Table 16)

In 2004, DWTF preliminary stack monitoring for tritium took place for a few days in November. Because the facility reported that there were no sources of tritium being stored outdoors in 2004, the annual release rate from the DWTF stack was back-calculated from the mean annual tritium concentration observed at the DWTF ambient air tritium monitor. The air tritium sampler is 115 m ENE from the stack, which is far enough from the stack for the tritium to come to ground but not far enough for the dispersion model to work well. Nevertheless, based on stack parameters (Table 8) and CAP88-PC with the wind file for 2004, a release rate of 20 Ci HTO was calculated. Based on the relative amounts of HT and HTO observed during stack monitoring in November 2004 and early 2005, release rates of 1 Ci HT and 19 Ci HTO were used as input to DCART with high uncertainty.

In February 2005, continuous stack monitoring for tritium began. The measured releases plus the estimated releases for the missing weeks were much lower than was estimated for the stack in 2004 by back-calculation from the DWTF air tritium monitor. Presumably because of actual low releases in 2005, the mean annual concentration of tritium in air for 2005 at the DWTF sampler was under-predicted by CAP88-PC when all known sources of tritium were taken into account. The difference between the predicted and observed tritium concentrations at the air tritium sampler was explained by the presence of containers of waste (some tritiated) that were stored outside to the east of Building 693 in 2005. Using back-calculation, an estimated release of 0.21 Ci of tritium was sufficient to approximately match predicted with observed air concentrations. This small diffuse source had a large effect at the DWTF air tritium monitor, but its affect on the air concentrations at either VIS or CRED (a monitor installed in July 2003 at the location of the UNCLE Credit Union, the SW-MEI for NESHAP’s compliance) was minimal; it added less than 1% to the total predicted mean annual tritium concentrations at either air sampler. Thus the outside source at DWTF was not included in the modeling of dose at the Discovery Center in 2005 (and the uncertainty on the release rate did not have to be assessed).

Derivation of annual release rates and uncertainty on HT and HTO from the SNL/CA Tritium Research Laboratory (Table 17)

Uncertainty on the release rates from the SNL/CA TRL includes uncertainties on

- reported release rate

- speciation
- analysis of the tritium content in the water from the bubbler system

Annual release rates from the SNL/CA TRL were published in the SNL/CA Site Environmental Reports. An uncertainty of ± 5% has been applied (similar to Building 331) to the reported routine annual release rate. This 5% accounts for cases when it is unknown whether or not evaporated tritium was included in the stack release rate (see discussion about the SNL/CA TRL under "Background Information").

The history of accidental releases of tritium to the environment is summarized in Garcia and Gorman (1996), Johnson (1997), and Table 6. For the TDR, when accidents were small relative to the annual release rate, the accident was included as part of the routine annual release and the uncertainty on the annual release rate was doubled to account for uncertainty on the magnitude of the accident. The accidents of 1986 and 1987 were large enough that they could not reasonably be considered routine. The uncertainty about the annual release rates for 1986 and 1987 has been set at ± 20%, because the routine releases had to be estimated by subtracting the accidents from the reported annual totals. The doses from these accidents will be assessed in Part 5 of the TDR.

An additional uncertainty arises because only one record of exit velocity from the TRL stack was obtained for the entire operational life of the facility (Garcia 2002), although it is unlikely the exit velocity remained the same throughout the years of operations. It was assumed that the exit velocity varied no more than did the exit velocity of either stack at the LLNL Tritium Facility (about 7% standard deviation over time).

For the first few years, releases were reported only as total tritium, even though the releases were being analyzed as HT and HTO in the bubbler system. For these years, it was assumed that about 19% of the total tritium released was HT, as it was for the first four years that the speciation of the tritium was reported. The uncertainty on this is about ± 7%. If the bubblers were not sampled frequently enough, there was the risk that they would become saturated and that HTO would pass through the first series of bubblers, be caught in the second series and be counted as HT (Hafner, 2002). This source of uncertainty is impossible to quantify. Analytical uncertainty on scintillation counting of the bubbler water was assumed to be similar to that for LLNL samples of comparable magnitude[35]. Based on this approach, an uncertainty of between ± 0.4 and 5% (one standard deviation) was estimated.

Because the TRL was similar to the LLNL Tritium Facility, it is expected that there would have been at least one waste storage area associated with it from which tritium would off-gas. The contribution of such a diffuse source can only be estimated with much greater uncertainty than for the LLNL diffuse sources, and the dose effect at potential locations of the SW-MEI will be small because of the distance between source and receptor and the contributions from larger sources. No attempt was made to estimate release rates for the SNL/CA area sources.

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[35] The uncertainty on the analytical results is based not on the concentrations in the water samples that were counted but on annual release rates that were comparable for the TRL and the Tritium Facility.

Missing Sources

An attempt has been made to document and assess the releases from all major sources of tritium and most of the minor ones. Minor sources that are being overlooked knowingly have been mentioned above if they were associated with an operation described in detail. As well, releases from laser activities (Table 3), off-gassing from the bake-out oven in the decontamination facility (B419[36]), off-gassing from unknown WAAs (both at LLNL and SNL/CA), transpiration of contaminated soil water by vegetation, and releases from contaminated soils undergoing clean-up were ignored. Some operations from the earliest days of LLNL are of necessity not included due to not knowing the magnitude of the releases and their exact time frame. Otsuki (2004) recalled work with liquefying tritium being conducted in B232 (then B147), perhaps concurrent with work in Building 231. Otsuki's very first tritium research before Building 231 became available was conducted in B119 (then B161), in the hospital wing, in a women's restroom. These unknown releases were probably HT. It is hoped that the considerable uncertainty taken into account for doses calculated from known releases will include the impact of these unknown releases.

Examples of missing sources from recent years can be found be perusing NESHAPs reports prior to 2003 looking for "H-3". Essentially all of these sources are known to be so small that any effect at an off-site receptor will be lost in the uncertainty. It is hoped, as well, that the effect of the somewhat larger unaccounted sources will also be subsumed by uncertainty at potential locations of the SW-MEI, because it is essentially impossible to characterize them. No sources conspicuous by their absence were detected after 1974 when predicted air concentrations were compared with observed air concentrations at VIS (see Part 3 of the TDR).

Dilution factors (χ/Q in s m^{-3}) documented in Tables 18 and 19

CAP88-PC was chosen as the model with which to calculate the dilution factors needed as input to DCART, which, in turn, is used to calculate air concentrations from release rates. CAP88-PC is a very simple Gaussian plume model, which does not calculate building wake effects and is suitable only for flat terrain. However, its simplicity is not a drawback for the TDR because the Livermore site is flat and because tritium concentrations measured by the ambient air tritium sampler at VIS are not affected by building wake effects. In general, when a Gaussian plume model has the proper parameters, annual average air concentrations over flat terrain can only be predicted within a factor of two to four, with accuracy decreasing as complexity of meteorological and terrain conditions increases (Miller and Hively 1987). Thus the uncertainty on Gaussian models in general is large, and one model may be no better than another. The high uncertainty exists due to many simplifying assumptions in the model. The choice of CAP88-PC was based on familiarity and its known tendency to slightly overestimate air concentrations at VIS (Peterson 2006). Although the TDR attempts to predict accurate

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[36] There was some concern that the tritium from the oven should be monitored (Rich et al. 1972b). Plans were being made to measure releases (Knezevich et al. 1972; Singh et al. 1973; Graham 1974), but no record of the results of a test has been found.

doses with a 95% confidence interval, the dilution factors calculated by CAP88-PC will provide a conservative bias, at least at the Discovery Center (VIS).

Although annual wind files for CAP88-PC are available from 1989, because it was impossible to model earlier years using annual data, it was decided to model all years of the TDR using a single wind file derived from several years of meteorological data. The wind file used for the TDR was created from four years (2000 – 2003) worth of hourly data from the LLNL meteorological station. LLNL data from earlier years were rejected; instrumentation has been improving over the years, and data for 2000 – 2003 were the most reliable (Bowen 2004) at the time calculations for the TDR were begun. At any one location, quite large differences in dilution factors calculated by CAP88-PC from annual meteorological data for different years can be observed. These differences are the result of having to separate meteorological data for the 8,760 hours in a year into 96 bins (16 wind directions times 6 stability classes); when none or only a few hours of data end up in a bin, the results may not be reliable. This problem is avoided by using the four-year wind file, because, with over 35,000 hours of data, a better statistical sampling can take place.

Based on the normal uncertainty associated with Gaussian plume dispersion models, a ± 25% uncertainty on the dilution factor for the Tritium Stacks has been applied and ± 30% uncertainty has been applied to dilution factors for all other facilities. Uncertainty on dispersion is usually lognormal, so these percentages represent factors of about 3 to 4 uncertainty. This magnitude uncertainty accounts for differences that might be observed between results calculated using annual wind files and those calculated using the four-year wind file of the TDR.

The uncertainty on the exit velocity also contributes to the uncertainty on the dilution factor[37]. For each facility, the standard deviation of a set of dilution factors calculated from a set of exit velocities was used as the percent uncertainty on the dilution factor. The effect on dilution factor of uncertainty on the calibrated exit velocity has also been taken into account; a 5% uncertainty on calibrated exit velocity (Biermann 2004) transformed into at most 2.5% uncertainty on the dilution factor at the Discovery Center; the uncertainty was usually much less depending upon the source.

The overall uncertainty on each lognormally-distributed dilution factor has been calculated as if the contributing distributions were normal (i.e., square root of the sum of the squares). It was determined empirically that there was little difference in the final distribution whether or not it was estimated based on normally or lognormally distributed uncertainty estimates; certainly, the small difference was definitely not worth the considerable extra effort that would have been involved to assign lognormal distributions to each source of uncertainty and calculate the result using Crystal Ball®.

Dilution factors were calculated for the position of the VIS ambient air tritium sampler, located near the Discovery Center, using the weighted average of the sector in which VIS

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[37] For the Discovery Center, the uncertainty on the dilution factors is much smaller than the uncertainty on the exit velocities (see Table 8) that were used for the calculations in CAP88-PC. However, for locations towards which the wind blows less frequently than toward the Discovery Center, the uncertainty on the dilution factor can exceed the uncertainty on the corresponding exit velocities.

is located and the closest adjacent sector at known distances and bearings from the sources as determined by GPS (Table 8). The following equation was used:

$$(\chi/Q)_{\mathrm{eff}} = [(11.25 + b)(\chi/Q)_1 + (11.25 - b)(\chi/Q)_2]/22.5$$

where

$(\chi/Q)_1$	dilution factor for the sector containing the receptor
$(\chi/Q)_2$	dilution factor for the adjacent sector
b	angular distance of the receptor from the boundary of the sector
11.25	half a sector width in degrees (a sector is 22.5 degrees)

Tables 18 and 19 show estimated dilution factors with uncertainty for all facilities; all distributions are lognormal. In Table 18, because one wind file was used for all years and at most one representative exit velocity was known, just one dilution factor with high uncertainty is shown for each facility for all years of operation. Because the data were available, dilution factors were calculated for each year and each stack for the Tritium Facility (Table 19) although the differences between years were negligible.

Derivation of dilution factors (χ/Q in s m^{-3}) with uncertainty for modeled sources other than the LLNL Tritium Facility (Table 18)

Building 231

Very little is known about the Tritium Room in Building 231 except that work was carried out in glove boxes. To estimate a dilution factor, a survey of the range of values for stack diameters and exit velocities of glove boxes was made using several years of NESHAPs reports. Based on thirty-one different sets of glove box stack diameters and exit velocities (Table 8) from NESHAPs reports[38] and what was known about the stacks in 1978[39], thirty-one potential dilution factors were calculated. The standard deviation on the mean of the thirty-one stacks, combined with the 30% uncertainty on dispersion in general, was assumed to be the uncertainty on the dilution factor. The dilution factor at the Discovery Center is quite insensitive to variations in stack parameters at Building 231.

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[38] Specific data for stack height and exit velocity for Room 1228 in the Chemistry wing were found in early NESHAPs reports – forty years after the building was used for tritium, - but the stack was not associated with a glove box. Stack height was 9.1 m, stack diameter was -.41 m, and the exit velocity was 12 m s^{-1}. The oldest-looking stacks on the present Chemistry wing (now the plastics shop) have about a 16 – 20 inch diameter. This agrees well with the NESHAPs inside diameter of 0.41 m.

[39] Building plans (Drawing # PHL-68 231-003 CH found in Building 231) for 1978 show external stacks with indicated flow rates. Four stacks were indicated on the north side of the building (FHE-5 [150 cfm], FHE-32 [1300/2000 cfm], FHG-34 [1300, 2000 cfm], and CD-1 [1800 cfm]) and one (FHE-29 [6000 cfm]) was indicated on the south side. The 1978 plan shows three rooms. Two are large; the dividing wall runs east-west. A small room is found in the southeast corner. Its dimensions are somewhat smaller than the estimated size of the Tritium Room, but the other rooms are too large to have been the Tritium Room if the exchange rate of air for the emergency blower in 1954 was correct (Thaxter 1954). No external stack exited the small room in the 1978 plans.

Building 212 room air

Besides dispersion, the uncertainty for the dilution factor for room air (a point source) includes the height at which the release occurred and the exit velocity. For CAP88-PC, the room air will be modeled as it might have been for NESHAPs compliance; room air at Building 212 would have passively leaked through small stacks on the roof. Fortunately, the dilution factor at VIS from Building 212 is very insensitive to any sort of change in modeling parameter values (compare 2.68 x 10^{-6} s m^{-3} for Building 212 room air with 2.49 x 10^{-6} s m^{-3} for the Building 212 stack), so the details of how the tritium escaped from Building 212 will have very little affect on the dilution factor at the Discovery Center. A generous ± 1% uncertainty has been assumed due to changes in exit velocity, and about ± 10% uncertainty is due to uncertainty about the release height. Overall uncertainty is ± 31.6%.

Area sources (Building 231 WAA, Building 331 WAA, Building 514 Yard, Building 612 Yard, and evaporation trays)

The uncertainty from all area sources except the evaporation trays is primarily from dispersion (± 30%) and potentially from the estimated release height, which is 1 m. However, the effect of estimated release height can be ignored because 1 m is an excellent approximation of the mean release height for all sources, except the evaporation trays. For the evaporation trays, it has been assumed that the release height was at ground level. Uncertainty will be due to dispersion and to not knowing the exact locations of the trays on the Taxi Strip. Three distances and directions to the Discovery Center were estimated from the bottom, center, and top of the Taxi Strip. From these directions and distances (457 m ENE/NE, 419 m ENE (NE), and 400 m ENE) a dilution factor at the Discovery Center was estimated using CAP88-PC with an overall uncertainty of ± 32%.

Building 212 stack

The dilution factor at the Discovery Center from releases at Building 212 is highly insensitive to change in exit velocity. As explained under the discussion about deriving the release rate for Building 212 (Table 11), an uncertainty of ± 10% has been applied for each year when the exit velocity is known; for the years when the exit velocity is unknown, the uncertainty applied is ± 20%. This translates into an uncertainty on the dilution factor of ± 0.57% for years of unknown exit velocity and ± 0.21% for years of known exit velocity. The overall uncertainty on the dilution factor is therefore only that of dispersion (± 30%).

Building 624 incinerator

The parameter values needed as input for CAP88-PC are known quite well for the incinerator (Table 8), so the overall uncertainty is basically the ± 30% from dispersion, even though there are very small contributions from exit velocity unknowns.

Building 292 stack

Reported exit velocities and/or stack volumes for Building 292 are quite variable. Because of this, the average of all known exit velocities was used to calculate the dilution factor at the Discovery Center with an uncertainty of ± 11.9%. The rationale for this is presented in detail in the discussion of Table 15. Using all known exit velocities creates a “global” exit velocity with associated uncertainty and precludes the need to add any uncertainty on the dilution factor contributed by the potential ± 5% uncertainty on the calibrated exit velocity. Overall uncertainty on the dilution factor is ± 32.3% from dispersion and change in exit velocity.

Decontamination and Waste Treatment Facility Stack

Given the confidence that can be placed in the exit velocity (Wilson 2006), essentially the entire uncertainty on the dilution factor at the Discovery Center from the DWTF Stack is the ± 30% uncertainty on the dispersion modeling.

SNL/CA stack

The dilution factor and uncertainty for SNL/CA are the same for all years because SNL/CA was not asked for the kinds of records (stack calibrations, flow rate measurements) that were obtained from LLNL. Assuming similarities in stack maintenance between LLNL’s Tritium Facility and the SNL/CA TRL, the exit velocity might have varied by ± 7%. This results in uncertainty on the dilution factor of ± 4.2%; uncertainty based on problems with calibration adds an additional 3.1% uncertainty. The overall uncertainty is dominated by dispersion and is just over ± 30%.

Derivation of dilution factors with uncertainty for Stack 1 and Stack 2 of the LLNL Tritium Facility (Table 19).

The exit velocities for Stack 1 vary less over time than those of Stack 2, so the effect of the uncertainty on the dilution factor is less. The highest uncertainty on the dilution factor due to unknown exit velocity for the years when measurements were taken regularly is just 2.2 % for Stack 1 and 3.1% for Stack 2; the highest uncertainty due to the 5% uncertainty on the calibrated exit velocity is about 1.5% for both stacks. The uncertainty (± 25%) on the dispersion model dominates the overall uncertainty. As can be seen in Table 19, a single dilution factor could easily have been used for each stack for all years without making a significant difference to the dose calculations.

Other annual input documented in Tables 20 - 22

Annual mean observed concentrations of tritium in air moisture (Bq L^{1}) and air (Bq m^{-3}) at VIS with estimated uncertainty (Table 20)

Observed volumetric air tritium concentrations at VIS can be used in DCART to predict doses that are more accurate and have less uncertainty (at least potentially) than doses predicted based on dispersion modeling. Also, to check that assumptions about release rates and other factors are reasonable and err on the side of slight conservatism, the observed annual mean concentrations of tritium in air moisture at VIS can be compared with air moisture concentrations calculated from CAP88-PC’s predicted air concentration and the absolute humidity used in DCART (Table 21).

To monitor tritium in ambient air, samplers are deployed in the field. Each sampler consists of a flask filled with silica gel, a pump that passes air through the silica gel, and, since 2000, a flow meter that measures the volume of air passed through the sample. In the analytical laboratory, the collected water is extracted from the silica gel by freeze-drying and then analyzed by scintillation counting. This primary concentration is reported in pCi L^{-1}. By relating the air volume passed through the sample to the total activity collected, a derived concentration in air volume is calculated in pCi m^{-3}. Reported concentrations have been converted in this report to Bq L^{-1} and Bq m^{-3} to match DCART's output.

Ambient tritium concentrations in air at the VIS sampling location have been measured since May 6, 1973. Because for most years the mean air concentrations for May through December are very similar to means for the entire year, 1973 has been included with the data used to test CAP88-PC's predictions of air concentrations. 1974 was the first complete year of sampling. Sampling was on a weekly basis in 1973, 1974 and 1975. In 1976, sampling was more or less monthly, but by 1977 sampling had settled into the biweekly pattern that is maintained today. Silica gel was re-used until 1986 or 1987, when a memory effect after exposure to high concentrations of tritium was discovered. Since then, silica gel has been used once and disposed of after analysis. The memory effect is undoubtedly caused by the "bound" water in the silica gel, which necessitated the introduction of the correction factor of 1.6 (see the discussion under the generic uncertainty of release rates that precedes Table 9).

Mean annual concentrations in extracted air moisture and in volumetric air that had passed through the silica gel sampler (Table 20) were calculated by averaging all samples that were not acknowledged as outliers (e.g., Holland et al. 1987), that were above the detection limit, and that were not associated with known accidental releases that obviously affected the concentration of the biweekly sample at VIS but could not be predicted based on routine release rates. For 1976, when samples were taken only roughly on a monthly basis, the results were weighted for the number of days each sample was in the field.

Uncertainty about the mean is based on the number of missing samples, the number of samples below the detection limit, and samples rejected because known accidental releases were reported during the time period of the sample. For numbers (x) of missing or removed samples, the percent uncertainty is assumed to be ± x/# samples in a year. For weeks of known accidental releases that were not detected at VIS, the uncertainty on the observed values has been increased. Uncertainty about the mean also accounts for the silica gel correction, laboratory analysis, and, for the volumetric results, the flow rate through the sampler.

All concentrations prior to 2001 have been multiplied by 1.6 with an uncertainty of ± 15% to account for the silica gel correction factor and associated uncertainty. Prior to 1986 or 1987, the correction factor is likely to be lower but more variable, given that, with a memory effect, the bound water in the silica gel would be expected to have a higher concentration than background because most samples would have been exposed to a tritium signal from LLNL operations. This phenomenon has been demonstrated using Crystal Ball® with assorted assumptions. Correcting by factor of 1.6 ± 15% should either estimate the true concentrations adequately or over-estimate them, which will assure that doses are not under-predicted.

Analytical error was estimated by selecting analytical results close to the calculated annual mean and selecting a representative error. Analytical error (1 standard deviation) ranged from 0.011 in 1977 to 0.175 in 2005. Before flow meters were used, the estimated uncertainty on the sample volume of air was ± 14%; after 2000, the uncertainty on the flow dropped to ± 3.5%, based on manufacturer specifications.

Because air concentrations predicted from the quantities of tritium released do not include bomb test fallout and cosmogenic levels of background tritium that are sampled during air tritium monitoring, background concentrations have been subtracted from observed air tritium concentrations, and associated uncertainty has been taken into account (Peterson 2004). The uncertainty associated with background tritium concentrations is due to assumptions about the ratio (0.8 ± 5%) between concentrations of tritium in rain[40] and those in air moisture and about the long-term absolute humidity at LLNL (0.0078 kg m^{-3} ± 5%) that must be known to convert concentrations in air moisture (Bq L^{-1}) to concentrations in air volume (Bq m^{-3}). Tritium background concentrations in most cases were very small compared with observed annual mean concentrations; at the most extreme, in recent years when LLNL's contributions to ambient air tritium concentrations were low, the estimated background concentrations were about 6-7% of the observed air concentrations.

When observed concentrations were not expected at an LLNL sampling location, an effort, not always successful, has always been made to explain them. For example, Holland and Brekke (1988) reported that between September 1986 and May 1987, air tritium concentrations at several locations were elevated over anything that could be attributed to monitored stack releases at LLNL or SNL/CA. Measurements of tritium in air were corroborated by measurements of tritium in vegetation. No other local source of tritium was identified, even after an exhaustive search (Holland and Carlsen 1987). CAP88-PC under-predicted all air concentrations at all sampler locations except VIS and one other (Peterson 2004) for 1986 and 1987, so perhaps under-predictions due to anomalies are less likely to occur at VIS than at other sampling locations.

When annual mean concentrations in air moisture are plotted against annual mean concentrations in air volume for each year (see figure below), a nearly linear response is expected. Perfect linearity is not expected, of course, because of the role played by absolute humidity (i.e., the quantity of water collected per unit volume of air passed through the sampler): only if water collected per unit volume is the same for all samples will the relationship be truly linear. This will not occur, because absolute humidity varies throughout the year and because silica gel does not necessarily either collect or retain all the moisture that is passed through it[41]. By plotting the observed data, apparent errors in the volumetric concentrations for 1976 and, to a lesser extent, 1973, can be seen. The value that best fits the expected linear relationship between the concentrations is 4.27 Bq m^{-3} for 1976 and 2.87 Bq m^{-3} for 1973.

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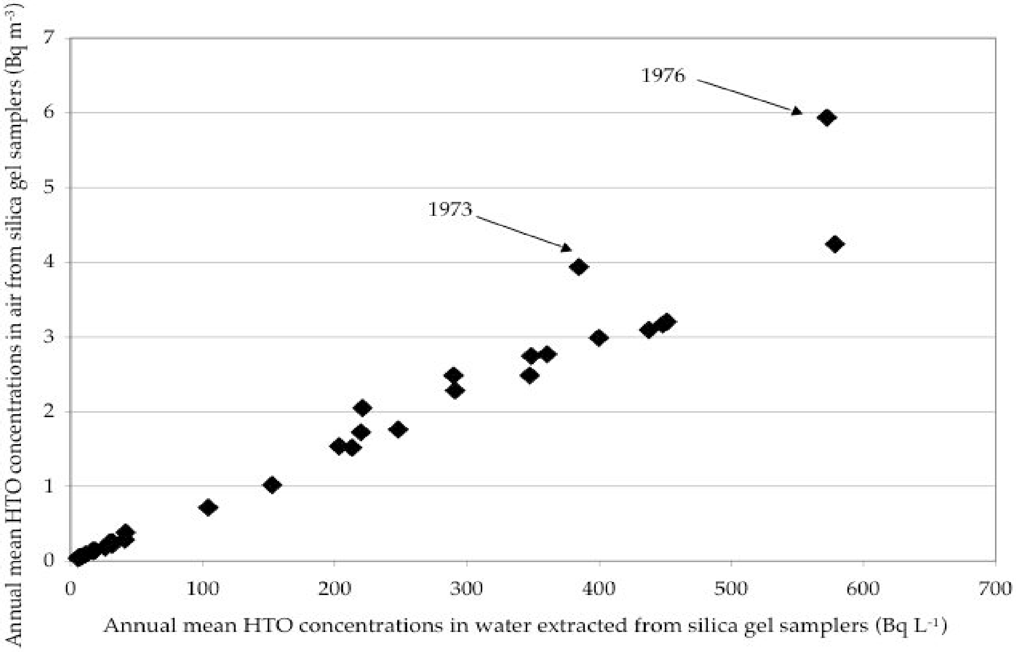
[40] Essentially all data on background concentrations of tritium are obtained from concentrations in precipitation. Concentrations in rain for various locations are available from the International Atomic Energy Agency/World Health Organization. IAEA/WHO (2004) Global Network of Isotopes in Precipitation. The GNIP Database is accessible at http://isohis.iaea.org.

[41] Absorption efficiency increases under higher humidity, higher pressure and lower temperature (Absorption efficiency is above 25% when the relative humidity is 50% and the temperature is 20° C: absorption efficiency is above 65% when the relative humidity is under 100% and the temperature is 20° C) (http://www.silicagel.net/products.asp)



Annual absolute humidity (kg m^{-3}) and relative humidity with uncertainty (Table 21)

Data needed to calculate absolute humidity (i.e., temperature and relative humidity) have only been gathered at the LLNL meteorological station since 1997, whereas air moisture has been collected using silica gel air tritium samplers since 1973. The mass of the water absorbed by the silica gel divided by the volume of air that has passed through the sample gives an estimate of the absolute humidity. The values for absolute humidity shown in Table 21 were obtained from averaging estimates of absolute humidity for all Livermore site air tritium samples for each year. Eberhart (1999) and Paulus et al. (2003) observed that absorption of water by silica gel may not necessarily be a good method by which to estimate absolute humidity under some sampling conditions, because the atmospheric moisture-collection efficiency for silica gel decreases as the ambient temperature increases. Annual average absolute humidity at LLNL for the years of reliable measured relative humidity and temperature data are, on average, 4% higher than the absolute humidity from corresponding silica gel data, but a consistent underestimation of absolute humidity by silica gel is not seen. Any underestimation of absolute humidity will result in higher doses in DCART, so predictions will err on the side of conservatism. The uncertainty on the mean of absolute humidity for a normal distribution for all years is ± 5%. This has been applied to the uncertainty of each annual mean as well.

The values for relative humidity shown are ones that are reasonably trusted from data gathered at the LLNL meteorological station (Bowen 2003). Uncertainty on relative humidity is ± 10%. Uncertainty on the mean of the relative humidity values for the years shown is only 7%, but there are so few data that the uncertainty was increased. Uncertainty on relative humidity is normally distributed.

Analysis of concentrations in the LLNL swimming pool (Table 22)

In DCART (Peterson 2006), tritium concentrations in the LLNL swimming pool can be estimated for the years when the pool water was not sampled from the empirical relationship between air moisture tritium concentrations at VIS, air moisture tritium concentrations at the ambient air tritium sampler (POOL) next to the pool, and pool water concentrations when measurements were taken. For years with measured concentrations, the mean annual pool water concentrations can be directly input to DCART. In Table 22, the means were calculated using the lower limits of detection (LLD) as the values for samples below the LLD; data for 2001 through 2004 have been excluded because all samples were below the LLD. In most cases, removing the samples below the LLD increased the concentrations only slightly. In the early years of sampling the pool, samples were taken monthly for all or most months of the year. More recently, samples have been taken quarterly. For a year in which there were no large fluctuations of atmospheric sources, the maximum concentration divided by the minimum concentration in the pool was only about a factor of 2. In years with large fluctuations in emissions, the annual variation in the pool water concentrations was about a factor of 4. The uncertainty shown here (normal distribution) is based only on analytical uncertainty and the calculated standard deviation of the available concentrations. Because the pool is a good integrator of the atmospheric tritium signal, quarterly samples appear to adequately represent pool water concentrations.

In July 2004, the pool was declared structurally unsafe and drained. It was removed about a year later.

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In a few cases in this report, I point out errors or omissions in the past that affect the values used in this dose reconstruction. No criticism of any individual is intended. This revision seeks to correct omissions and mistakes made in the original report, to update the report to include 2005, and to clarify issues based on experience gained subsequent to the publication of the report in February 2005; every effort has been made not to introduce new errors.

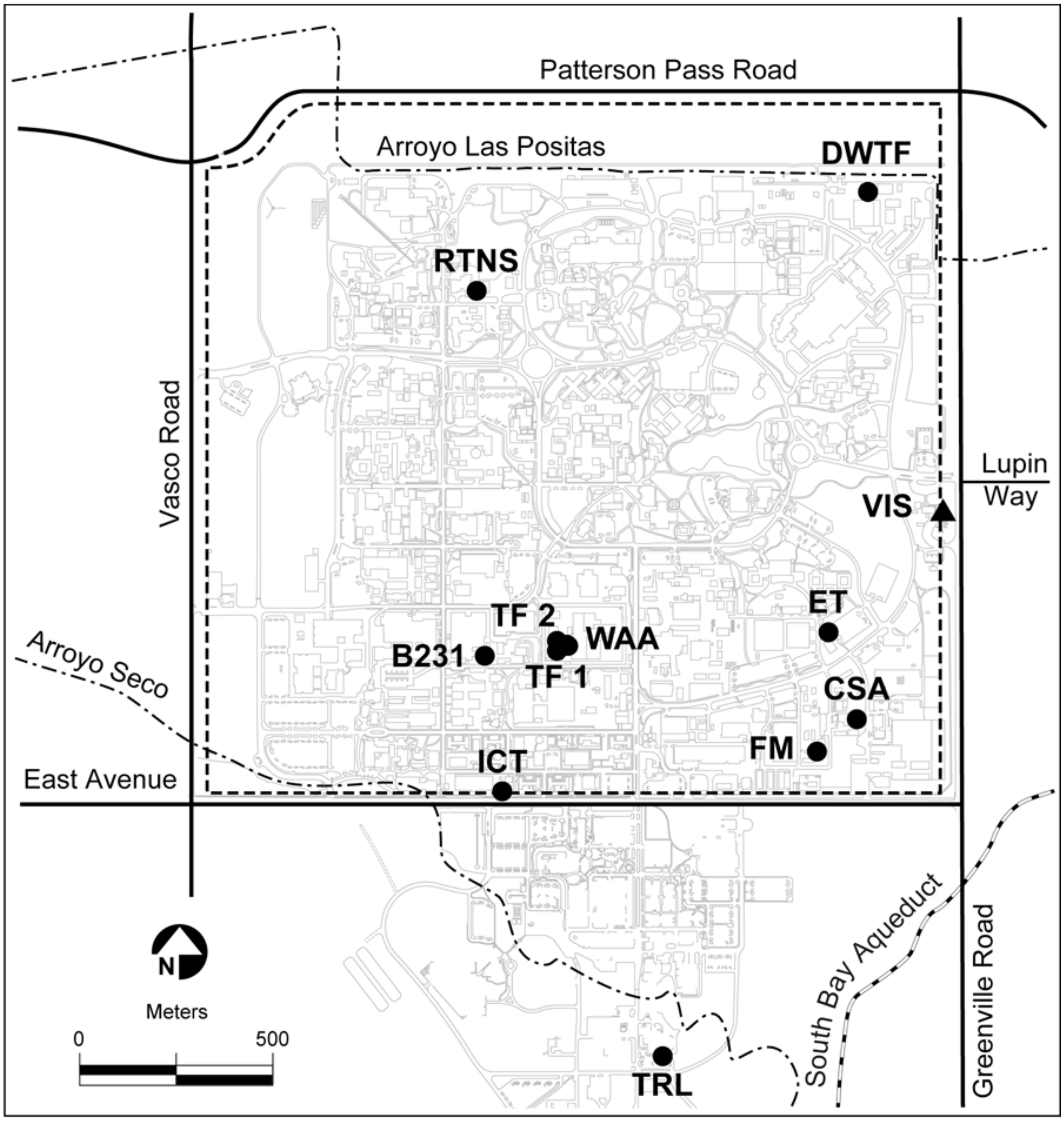

Figure 1. Sources of tritium (●) at LLNL and SNL/CA relative to the location of the potential site-wide maximally exposed individual (▲). ▲VIS is the Discovery Center. Modeled sources include TF1 and TF2 (the south and north stacks of the Tritium Facility), WAA (diffuse source at Building 331), B231 (where tritium work took place before 1958, and its presumed WAA), ICT and room air (Building 212), RTNS (Building 292). CSA (Building 612 Yard and the Building 624 incinerator), ET (solar evaporation trays), and TRL (Tritium Research Laboratory at SNL/CA), FM (the Building 514 Yard), and DWTF (Decontamination and Waste Treatment Facility).

Table 1. Quantities (1956 – 1979) of total tritium (HT and HTO) released from the main site of Lawrence Livermore National Laboratory reported to the Department of Energy and predecessors. A “+” indicates that more emissions are expected because some data for that year may be missing. A “-” indicates the probable number is lower. Building172 was the Tritium Facility before it was renamed Building 331.

Year	**Ci y^{-1}**	**Comments (numbers in parentheses are in Ci)**
1956	3519+	“gas” released 8/2[1] (3000), 8/28[1] (192), 10/5[2] (135), 10/25[2] (192); there were no memos for the first two quarters.
1957	11356+	“gas” released 4/1[3] (32), 5/9[3] (130), 5/16[3] (194), 5/22[3] (6000), 6/12[3] (5000); there was no memo for the third quarter.
1958	5940+	“gas” released 2/19[4,5] (200), 3/31 (3000)[5,6], 1st quarter[7] (240), 10/31[8] (600), 11/5[9] (1900); there were no memos for the second and third quarters.
1959	3060+	“gas” released 2/16[10] (1600), 5/12[11] (1100), 5/25[11] (360); additional negligible releases were reported for the third quarter[12] and fourth quarter.[13]
1960	3100+	“gas” released 2/23[14] (2000), 11/19[15] (1100); additional negligible releases[16,17,18] were reported for quarters ending in January, July, and October.
1961	1005+	“gas” released April 5[19] (6), 6[19] (6), 7[19] (19), 10[19] (25), 11[19] (19), 12[19] (31), 13[19] (19); a negligible release of ~70 Ci[20],[21] was reported for the quarter ending in July. Reporting changed in mid-year, and “gas” is not mentioned: 10 Ci came from the mass spectrometer in Building 172 and 600 from “controlled releases” from Building 172; 200 Ci came from the 90-inch cyclotron.[22]
1962	4430-	All “controlled releases” from Building 172. 300 Ci of first quarter (end 1/31/62) may belong to 1961[23]. February through June, 300 Ci each month[24]. July (500), August (1200), September (800)[25]. October (65), November (65), December (0)[26].
1963	6000	All “controlled releases” from Building 172. First quarter[27] (1100); second quarter[28] (2000); third quarter[29] (2200); fourth quarter[30] (November – 700)
1964	12500	Reporting changed again. Building 172 was no longer mentioned; only “total tritium released from LLNL” was mentioned. February 5[31] (~300); March 17-19[31] (~1200); second quarter[32] (10,000); August through December[33] (1000)
1965	362350-	First quarter[34] (360,000 Ci were released accidentally on 1/20/65; 1000 routine); second quarter[35] (900); third quarter[36] (250-300); fourth quarter[37] (150)
1966	14050	First quarter[38] (850); second quarter[39] (11,000 Ci were released accidentally on 4/7/66; 200 routine); last half[40] (2000)

Table 1 continued.

Year	Ci y^{-1}	Comments (numbers in parentheses are in Ci)
1967	6600	First quarter[41] (1400); second quarter[42] (2000); third quarter[43] (2000); fourth quarter[44] (1200)
1968	6750	First quarter[45] (2000); Second quarter[46] (400); third quarter[47] (2350); fourth[48] quarter (2000)
1969	7300+	January[49] (300); February[49] (1200); March[49] (800); April[50] (1250); May (250); June (700); memo missing for third quarter; October[51] (1900); November[51] (800); December[51] (100)
1970	291463 or 291541[52]	January[53] (440); February[53] (1680); March[53] (260); April[54] (555); May[54] (825); June[54] (360); July[55] (160); August[55] (24); September[55] (30); October[55] (62); November[55] (37); December[55] (30); 287,000 Ci[56] were accidentally released August 6; third quarter[56] (230); fourth quarter[57] (191)
1971	2710	First quarter[58] (560); second quarter[59] (990); third quarter[60] (790); fourth quarter[61] (370)
1972	1350	First quarter[62] (330); second quarter[63] (240); third quarter[64] (510); fourth quarter[65] (270)
1973	2560	First quarter[66] (480); second quarter[67] (1140); third quarter[67] (470); fourth quarter [68](470)
1974	1690	First quarter[69] (460); second quarter[70] (610); third quarter[71] (310); fourth quarter[72] (310)
1975	2440	First quarter[73] (510); second quarter[74] (740); third quarter[75] (410); fourth quarter[76] (880)
1976	3700	First quarter[77] (570 T); second quarter[78] (~365 HTO, ~445 HT); third quarter[79] (~742 HTO, ~685 HT); fourth quarter[80] (~366 HTO; 527 HT)
1977	4700	First quarter[81] (~135 HTO; ~429 HT); second quarter[82] (~650 HTO; ~452 HT); third quarter[83] (~571 HTO; ~697 HT); fourth quarter[84] (~600 HTO, 1166 HT)
1978	5361	First quarter[85] (~473 HTO; ~879 HT); second quarter[86] (~539 HTO, ~776 HT); third quarter[87] (~390 HTO, ~867 HT); fourth quarter[88] (~560 HTO, ~877HT)
1979	3893+	First quarter[89] (~488 HTO, ~1387 HT); second[90] quarter (~614 HTO, ~1046 HT); third quarter[91] (~211 HTO, ~147 HT); no memo for fourth quarter

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[48] May, M.M. Memo “Release of Tritium to the Atmosphere” for period October through December 1968 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. January 7, 1969.

[49] Otsuki, H.H. Memo “Tritium releases for first quarter 1969” to Director’s Office. Confidential memo declassified at some point. April 4, 1969.

[50] Otsuki, H.H. Memo “Tritium release” to Director’s Office. Confidential memo declassified at some point. July 8, 1969.

[51] Otsuki, H.H. Memo “Tritium release for the fourth quarter 1969” to Director’s Office. Confidential memo later declassified. January 19, 1970.

[52] 291463 was obtained by adding all reported monthly release rates and the accident; 29154 was obtained by adding the reported monthly results for January through June and the two quarterly release rates, plus the accident.

[53] Otsuki, H.H. Memo “Tritium release” to Director’s Office. Confidential, restricted data memo later declassified. April 7, 1970.

[54] Otsuki, H.H. Memo “Tritium release – 2nd quarter” to Director’s Office. Confidential, restricted data memo later declassified. July 13, 1970.

[55] Snider, G.D. Memo “Tritium releases third and fourth quarter 1970” to Director’s Office. Confidential, restricted data memo later declassified. February 11, 1971.

[56] May, M.M. Memo “Release of Tritium to the Atmosphere” for period July through September 1970 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. October 19, 1970.

[57] May, M.M. Memo “Release of Tritium to the Atmosphere” for period October through December 1970 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. February 18, 1971.

[58] May, M.M. Memo “Release of Tritium to the Atmosphere” for period January through March 1971 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. April 23, 1971.

[59] May, M.M. Memo “Release of Tritium to the Atmosphere” for period April through June 1971 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. July 22, 1971.

[60] Haussman, C. Letter for period July through September 1971 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. October 13, 1971.

[61] Batzel, R.E. Letter for period October through December 1971 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. January 19, 1972.

REFERENCES (Table 1 continued)

[62] Batzel, R.E. Letter for period January through March 1972 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. April 19, 1972.

[63] Batzel, R.E. Letter for period April through June 1972 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. July 13, 1972.

[64] Batzel, R.E. Letter for period July through September 1972 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. October 17, 1972

[65] Batzel, R.E. Letter for period October through December 1972 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. January 17, 1972

[66] Batzel, R.E. Letter for period January through March 1973 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. April 13, 1973.

[67] Batzel, R.E. Letter for period April through June 1973 to E.S. Pierce, U.S. Atomic Energy Commission, Washington D.C. October 14, 1973.

[68] Olsen, J.L. Letter for period October through December 1973 to E.S. Pierce, U.S. Atomic Energy Commission, Washington D.C. January 14, 1974.

[69] Olsen, J.L. Letter for period January through March 1974 to E.S. Pierce, U.S. Atomic Energy Commission, Washington D.C. April 15, 1974.

[70] Olsen, J.L. Letter for period April through June 1974 to E.S. Pierce, U.S. Atomic Energy Commission, Washington D.C. July 11, 1974.

[71] Olsen, J.L. Letter for period July through September 1974 to E.S. Pierce, U.S. Atomic Energy Commission, Washington D.C. October 14, 1974.

[72] Olsen, J.L. Letter for period October through December 1974 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. January 16, 1975.

[73] Olsen, J.L. Letter for period January through March 1975 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. April 15, 1975.

[74] Olsen, J.L. Letter for period April through June 1975 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. July 17, 1975.

[75] Olsen, J.L. Letter for period July through September 1975 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. October 23, 1975.

[76] Olsen, J.L. Letter for period October through December 1975 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. January 20, 1976.

[77] Olsen, J.L. Letter for period January through March 1976 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. April 7, 1976.

[78] Olsen, J.L. Letter for period April through June 1976 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. September 7, 1976.

[79] Olsen, J.L. Letter for period July through September 1976 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. November 9, 1976.

[80] Olsen, J.L. Letter for period September through December 1976 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. January 27, 1977.

[81] Olsen, J.L. Letter for period January through March 1977 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration, Washington D.C. May 6, 1977.

[82] Olsen, J.L. Letter for period April through June 1977 to E.S. Pierce, Division of Physical Research, U.S. Energy Research and Development Administration. Washington D.C. August 25, 1977.

[83] Olsen, J.L. Letter for period July through September 1977 to E.S. Pierce, Division of Physical Research, U.S. Department of Energy, Washington D.C. November 29, 1977.

[84] Olsen, J.L. Letter for period October through December 1977 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. February 6, 1978.

REFERENCES (Table 1 continued)

[85] Olsen, J.L. Letter for period January through March 1978 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. May 8, 1978.

[86] Olsen, J.L. Letter for period April through June 1978 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. August 1, 1978.

[87] Olsen, J.L. Letter for period July through September 1978 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. November 22, 1978.

[88] Olsen, J.L. Letter for period October through December 1978 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. March 12, 1979.

[89] Olsen, J.L. Letter for period January through April (sic) 1979 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. May 29, 1979.

[90] Olsen, J.L. Letter for period April through June 1979 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. August 29, 1979.

[91] Olsen, J.L. Letter for period July through September 1979 to W.K. Benson, Division of International Security Affairs, U.S. Department of Energy, Washington D.C. January 2, 1979.

Table 2. Quantities of total tritium (HT + HTO), both routine and accidental, released annually from the LLNL Tritium Facility

Year	Ci (from memos other than to Department of Energy or predecessors)	Ci (from Talk to Director's Office[92])	Ci (from LLNL Site Annual Environmental Reports)
1953	2000 ± 50%[93]	2000	
1954	2000 ± 50%[93]	2000	
1955	2000 ± 50%[93]	2000	
1956	3000 ± 50%[93]	3519	
1957	3000 ± 50%[93]	11356	
1958	1000 ± 50%[93]	5940	
1959	1000 ± 50%[93]	3380	
1960	1000 ± 50%[93]	3180	
1961	1000 ± 50%[93]	1100	
1962	3000 ± 50%[93]	4130	
1963	3000 ± 50%[93]	6000	
1964	26000[93]	26500	
1965	360000[93]	363350	
1966	14000[93]	17050	
1967	6600[93]	6600	
1968	6800[93]	6750	
1969	7300[93]	7300	
1970	290000[93]	291480	
1971	1400[93]	1400	
1972		1350	
1973		2960	2510
1974	1350[94]	1690	1300
1975	1353[95]	2540	2167
1976	2819[96]; 2822[97]	3675	2828
1977	2956[97]; 3145[98]	3150	3150
1978	4232[97]; 4243[99]; 4245[100]	5250	4245
1979	4069[101]; 4070[99]	4100	4069

Table 2 continued

Year	Ci (from memos other than to Department of Energy or predecessors)	Ci from (Talk to Director's Office[92])	Ci (from LLNL Site Annual Environmental Reports)
1980	2217[102]; 2219[103]	2179	2218
1981	2552[104,105]	2550	2552
1982	1899[106]; 1914[107]	1914	1914
1983	3024[108]	3024	3024
1984	7200[109]	7200	7200
1985	1989[110]	1989	1989
1986	1128[111,112]	1128	1128
1987	2633.8[113]	2634	2633.8
1988	3977.5[114]		3997.5
1989	2949.5[115]		2949.4
1990			1281.3
1991			1111.6
1992			177
1993			237
1994			137
1995			91.8
1996			214.7
1997			299.3
1998			108.9
1999			280.5
2000			40.0
2001			20.0
2002			36.3
2003			116
2004			16.0
2005			32.0

REFERENCES (Table 2)

[92] Souers, C. Overheads from a talk "Tritium Releases from B331" prepared for the Director's Office, June 1988.

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

[94] Powell, T.J. Building 331 – Health Physics Audit for 1975.

[95] From a hand-written summary (Build Stack Effluent Report) of monthly gaseous releases from B194, B281, B212, and B313 for 1975; particulate releases also.

[96] From a hand-written summary (1976 Effluent Data of monthly gaseous releases from B194, B281, B212, and B313 as well as the 1976 SAER. Sheet found in the 1976 Stack Reports in LLNL Radiological Air Effluent Records.

[97] Extracted from hand-drawn graph by S. Homann, "Building 331 Monthly Tritium Release for 1976"

[98] From a hand-written summary (1977 Effluent Data) of monthly gaseous releases from B194, B281, B212, and B313. Sheet found in the 1977 Stack Reports in LLNL Radiological Air Effluent Records.

[99] Homann, S. Cumulative graphs of total emissions.

[100] From a hand-written summary (1978 Effluent Data) of monthly gaseous releases from B194, B281, B212, and B313. Sheet found in the 1978 Stack Reports in LLNL Radiological Air Effluent Records.

[101] From a hand-written summary (1979 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313 and B292. Sheet found in the 1979 Stack Reports in LLNL Radiological Air Effluent Records.

[102] From a hand-written summary (1980 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313 and B292. Sheet found in the 1980 Stack Reports in LLNL Radiological Air Effluent Records.

[103] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1980, memo to distribution, January 15, 1981

[104] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1981, memo to distribution February 18, 1982.

[105] From a hand-written summary (1981 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313 and B292. Sheet found in the 1981 Stack Reports in LLNL Radiological Air Effluent Records.

[106]From a hand-written summary (1982 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313 and B292. Sheet found in the 1982 Stack Reports in LLNL Radiological Air Effluent Records.

[107] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1982, memo to distribution, February 2, 1983.

[108] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1983, memo to distribution, January 17, 1984.

[109] King. W.C., Radiation and Environmental Safety Division, Hazards Control Department, memo to distribution January 22, 1985.

[110] Printed worksheet showing weekly sample values and figure.

[111] Gordon, L.E.. Memo to D. Brekke, January 30, 1987

[112] Gordon, L.E. Memo to A.L. Buerer, March 16, 1987.

[113] Mansfield, W.G. Memo to G. Morris, January 19, 1988

[114] Biermann, A. Memo to D. Brekke, February 21, 1989

[115] Mansfield, G. Memo to M. Singleton, April 13, 1990

Table 3. Reported releases from Building 212 (Accelerators and Insulating Core Transformer), Building 292 (Rotating Target Neutron Source), Building 624 incinerator, laser complex (Building 391, Building 381, Building 298), Building 612 Yard, Building 331 Waste Accumulation Area and the Decontamination and Waste Treatment Facility. All unmarked numbers were obtained from SAERs or NESHAPs reports; other sources are referenced in superscripts. Species of tritium are noted.

Year	B212[116,SAERs (1974 on),117] Ci HT, unless indicated	B292[118,119] Ci HTO	B624 incinerator Ci HTO	Laser complex Ci HT	B612 yard Ci HTO	B331 WAA Ci HTO	DWTF stack Ci HTO	DWTF stack Ci HT	DWTF area
1955	75 ± 50%								
1956	75 ± 50%								
1957	75 ± 50%								
1958	75 ± 50%								
1959	75 ± 50%								
1960	75 ± 50%								
1961	75 ± 50%								
1962	75 ± 50%								
1963	75 ± 50%								
1964	75 ± 50%								
1965	10 ± 50%								
1966	10 ± 50%								
1967	10 ± 50%								
1968	240								
1969	140								
1970	65								
1971	260								
1972	52								
1973	missing								
1974	560								
1975	1217[120]								
1976	1173[121]								
1977	2057[122]; 2060								
1978	1117[123]								
1979	436[124]	11.7[124]; 12							

Table 3 continued

Year	B212[116,SAERs,117] Ci HT, unless indicated	B292[118,119] Ci HTO	B624 incinerator Ci HTO	Laser complex Ci HT	B612 yard Ci HTO	B331 WAA Ci HTO	DWTF stack Ci HTO	DWTF stack Ci HT	DWTF area
1980	70[125]	17; 17.3[125]							
1981	23; 23.4[126]	44; 44.2							
1982	34[127]; 44	56; 56.2[127]							
1983	140	81							
1984	11	143							
1985	5	210[128]							
1986	13; 2 (HTO)[129]	106[130,131,132]	5.1; 3.75[133]						
1987	34 ; 4 (HTO)[134]	78.5	0.22; 0.213[135]						
1988		5.29	0.343[136]						
1989		2.4[137]		0.25					
1990				0.34					
1991				0.081					
1992						0.8			
1993					2.2	3.0			
1994					1.2	3.0			
1995					2.1	4.0			
1996					3.0	3.0			
1997					4.2	2.5			
1998					4.6	6.0			
1999					4.4	7.3			
2000					3.6	5.2			
2001					2.0	1.0			
2002					2.3	1.0			
2003					3.4	8.7			
2004					3.2	0.7	20		
2005							2.6	0.1	0.2[138]

REFERENCES AND ANNOTATIONS (Table 3)

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

[117] Radioactive effluent release from the LLL ICT (Insulating Core Transformer) consists primarily of tritium gas (Johnston, J.E. and Singh, M.S. Memo to distribution. Subject: Calibration of Building 212 Stack Monitor. September 25, 1975).

[118] Releases were monitored using silica gel; releases were estimated to be about 98% HTO and occurred during rough pumping.

[119] All published release rates, except the last year when molecular sieves were used, must be revised upwards by a factor of 1.6 before modeling to account for underestimation by the silica gel method of collecting HTO.

[120] From a hand-written summary (Build Stack Effluent Report) of monthly gaseous releases from B194, B281, B212, and B313 for 1975 as well as the 1975 SAER. Sheet found in the 1974/75 Stack Reports in LLNL Radiological Air Effluent Records.

[121] From a hand-written summary (1976 Effluent Data) of monthly gaseous releases from B194, B281, B212, and B313 as well as the 1976 SAER. Sheet found in the 1976 Stack Reports in LLNL Radiological Air Effluent Records.

[122] From a hand-written summary (1977 Effluent Data) of monthly gaseous releases from B194, B281, B212, and B313. Sheet found in the 1977 Stack Reports in LLNL Radiological Air Effluent Records.

[123] From a hand-written summary (1978 Effluent Data) of monthly gaseous releases from B194, B281, B212, and B313 as well as from the 1978 SAER. Sheet found in the 1978 Stack Reports in LLNL Radiological Air Effluent Records.

[124] From a hand-written summary (1979 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313, and B292 as well as from the 1979 SAER. Sheet found in the 1979 Stack Reports in LLNL Radiological Air Effluent Records.

[125] From a hand-written summary (1980 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313, and B292 as well as from the 1980 SAER. Sheet found in the 1980 Stack Reports in LLNL Radiological Air Effluent Records.

[126] From a hand-written summary (1981 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313, and B292 as well as from the 1981 SAER. Sheet found in the 1981 Stack Reports in LLNL Radiological Air Effluent Records.

[127] From a hand-written summary (1982 Effluent Data) of monthly gaseous releases from B194, B281, B212, B313, and B292 as well as from the 1982 SAER. Sheet found in the 1982 Stack Reports in LLNL Radiological Air Effluent Records. B212 showed sum of monthly releases to be 44, but numbers actually only summed to 34.

[128] Trent, M. Memo to K. Griggs January 21, 1986

[129] Homann, S. Memo to D. Brekke February 5, 1987. Subject: Annual Stack release from Buildings 194 and 212

[130] Trent, M. Memo to D. Brekke February 5, 1987. Subject: Stack Releases from Building 292 (RTNS-II) and Building 865 (ATA).

[131] Trent. M. Memo to Dave Myers and Bill Silver. Subject: Response to the Health Physics Review of Building 292. September 24, 1986.

[132] Stack sampler disconnected for at least a month, but there was thought to be no significant releases during the down-time based on output from the ion chambers. See references 123 and 124.

[133] Radiation Safety Program, Hazardous Waste Management, 1986 Annual Facility Report

[134] Homann, S.G. Memo to A. Biermann January 7, 1988. Subject: Annual Stack release from Buildings 194 and 212

[135] Radiation Safety Program, Hazardous Waste Management, 1987 Annual Facility Report

[136] Hoyt, Dan. Memo to Susi Jackson. Subject: Radionuclides incinerated at the HWM incinerator for calendar year 1988. HWM 89-161; February 3, 1989.

[137] Myers, D. Memo to K. Lamson February 5, 1990

[138] This source was modeled for NESHAPs in 2005 because its affect was seen at certain air tritium monitors when CAP88-PC’s predicted air concentrations were compared with observations. However, at location VIS, the effect of this source on modeled air concentrations was less than 1% of the total.

Table 4. Record of acute releases of tritium from LLNL.

Year	Ci	HT or HTO	Facility	Type[a]	Comments
1954	1450	HTO	231	A	1 cc of HTO lost from cold trap and HT October 13, 1954[139]
1956	3000	HT	231	R	Released August 28, 1956[140,141]
1957	11000	HT	231	R	6000 Ci May 22, 1957 and 5000 Ci June 12, 1957[142,141]
1958	4900	HT	331; Stack 1	R	3000 Ci March 31, [143,144,141] and 1900 Ci November 5, 1958[145,141]
1959	2700	HT	331; Stack 1	R	1600 Ci February 16, 1959[146,141] and 1100 Ci May 12, 1959,[147,141]
1960	3100	HT	331; Stack 1	R	2000 Ci May 2, 1960[148,141] and 1100 Ci November 19, 1960,[149,141]
1961	200	HT	212	A	From the 90 inch cyclotron[150] for the period 10/25 to 10/28/61
1964	24000	HT	331	A?	No record except Souers (1988)[141]
1965	360000	HT	331; Stack 1	A	2 minute release at 3:27 pm January 20, 1965; reported to AEC as 360,000 Ci[151]. Estimates based on integrations of the chart trace range from 207,000 to 300,000 Ci, but 350,000 was published[152]
1966	11000	HT	331; Stack 1	A	April 7, 1966[153]; 14,000[141]
1969	800	HT	331	NA	Experimental release October 27, 1969, 10:30 – 11:30 am[154]
1970	287000[155]	HT	331; Stack 1	A	30 minute release at 6:14 August 6, 1970, 289000 Ci[156]
1971	105 + 135	HT + HTO	331; Stack 1	A	April 12, 1971 at 9:05; environmental monitoring followed[157]
1975	340	HTO ?	331; Stack 2	R	Reported by Souers (1988)[141]; 460.7 Ci HTO released 5-12 November[158]
1976	175	HT ?	331; Stack 2	R	Reported by Souers (1988)[141]; 146.8 Ci HT released 3-10 March[158]
1981	130	HTO	331; Stack 1	A?	Possible 130 Ci release in November[159]; possible 142.8 Ci HTO release 2-9 November or possible 133.1 Ci HTO release 16-23 November[158]
	450	HTO	331 Stack 1	A?	December 12 or 13, 450 Ci measured by Ostlund Monitor but undetected by ion chambers[159]; environmental study carried out[160]; 477.8 Ci HTO released 7-14 December[158]

Table 4 continued

Year	Ci	HT or HTO	Facility	Type[a]	Comments
1983	220	HT ?	331; Stack 2	R	No record except Souers (1988)[141]; possibly 317 Ci HT released 7-14 March or 379 Ci HT released 28 November – 5 December.
1984	5200	HT	331; Stack 2	A	June 8, 1984 at 8 am; 6,000 Ci estimated from pressure drop; 5,200 best estimate based on electronic integrator (1,400 ± 300 and 4,300 ± 800[161]; 5,000 Ci reported officially[162]; 5,600 ± 500 Ci also reported[163]; reported in 1984 SAER as 5,000 Ci. HT; 5,155 Ci[141]
1985	1034	HT	331; Stack 2	A	January 24, 1985[164]; reported in 1985 SAER as 1,000 Ci HT; 1,000 Ci[141]; 920 Ci from ion chamber[164]; 935 Ci HT released 22-28 January[158]
1986	125	HT	331 Stack 1	R	December 15, 1986[165]; 135 Ci[141]; 149.8 Ci HT released 15-22 December[158]
1987	198	HT	331; Stack 2	R	April 14, 1987[165]; 185 Ci[141]; 198.9 Ci HT released 14-20 April[158]
	575	HT	331	R	No record except Failor (1999)[165]
1988	145	HT	331; Stack 2	R	January 19, 1988[165]; 121.7 Ci released 11-19 January[158]
	138	HTO	331; Stack 2	R	January 25, 1988[165]; 138.6 Ci HTO released 25 January – 2 February[158]
	653	HT	331; Stack 2	R	May 15, 1988[165]; 651.6 Ci HT released 16-18 May[158]
	120	HTO	331; Stack 2	R	August 1, 1988[165]; 120.4 Ci HTO released 2-10 August[158]
1989	112	HT	331; Stack 2	R	February 28, 1989[165]
	290 + 7	HT + HTO	331; Stack 2	A	August 25, 1989[166] between 10 and 11 am reported as 329 Ci HT in 1989 SAER; reported to National Response Center[167]; 329 Ci HT and 6.73 Ci HTO released 22-28 August[158]
	112	HTO	331; Stack 1	R	October 31, 1989[165]; 112 Ci HTO released 31 October – November 6[158]
1991	~112	HT	331; Stack 1	A	April 2, 1991 at 3:50 pm[168]. Reported in SAER as 144 Ci.[169]; 139 Ci HT released 1-3 April[158]

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[a] “A” is a reported accident; “R” refers to routine releases that were reclassified as acute; “?” is used when the amount released is not known or if the accident itself is questionable.

REFERENCES (Table 4):

[139] Stanhope, Chester M. (Bldg. Monitor, Bldg. 102). Memo to Dusty Meadors. Subject: T_2 accident and personnel involved, October 13, 1954. October 26, 1954.

[140] York, H.F. Memo 11, 127, “Release of Tritium to the Atmosphere” for quarter ending September 30, 1956 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret, restricted, later unclassified. March 1, 1957.

[141] Souers, C. Overheads from a talk “Tritium Releases from B331” prepared for the Director’s Office, June 1988.

[142] Southwick, E.W Memo 57-21, “Release of Tritium to the Atmosphere” for quarter ending June 30, 1957 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, later unclassified. July 3, 1957.

[143] Street, K. Memo 58-17, “Release of Tritium to the Atmosphere” for quarter ending March 31, 1958 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, later unclassified. April 2, 1958.

[144] Fleming, E.H. Memo “MINT” to Director’s Office. April 1, 1958.

[145] Batzel, R.E. Memo “MINT” to Directors Office. November 5, 1958.

[146] Street, K. Memo “Release of Tritium to the Atmosphere” for quarter ending March 31, 1959 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, later unclassified. August 3, 1959.

[147] Brown, H. Memo “Release of Tritium to the Atmosphere” for quarter ending July 31, 1959 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, later unclassified. April 7, 1959.

[148] Brown, H. Memo “Release of Tritium to the Atmosphere” for quarter ending April 30, 1960 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, later unclassified. May 2, 1960.

[149] Brown, H. Memo “Release of Tritium to the Atmosphere” for quarter ending January 31, 1961 to S.G. English, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. February 1, 1961.

[150] Foster, J.S. Jr. Memo ”Release of Tritium to the Atmosphere” for quarter ending October 31, 1961 to D.R. Miller, U.S. Atomic Energy Commission, Washington, D.C. Secret, restricted; unclassified in 1975. November 4, 1961.

[151] Foster, J.S. Jr. Memo “Release of Tritium to the Atmosphere” for period January 1 to March 31, 1965 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. April 8, 1965.

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[155] May, M.M. Memo “Release of Tritium to the Atmosphere” for period July through September 1970 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. October 19, 1970.

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[159] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1981, memo to distribution February 18, 1982.

[160] Handwritten notes from Hazards Control Records

REFERENCES (Table 4 continued)

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[162] King. W.C., Radiation and Environmental Safety Division, Hazards Control Department, memo to distribution January 22, 1985.

[163] Howe, H. Memo to C. Souers January 22, 1985; Subject: Dose Equivalent Estimates – Tritium Release, June 8, 1984

[164] Howe, H. Memo to C. Souers February 26, 1985; Subject: Tritium Release from Building 331 – January 24, 1985

[165] Obtained in 1999 from papers in the possession of Rebecca Failor.

[166] Brown, M. Memo to J. Steenhoven, September 15, 1989. Subject: Reassessment of the 8/25/89 tritium release with actual source term data from Building 331 molecular sieve (reference EQVG memo 2159-89)

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Table 5. Speciation of routine releases from the LLNL Tritium Facility. “B331 Spreadsheets” were calculated for the TDR based on tritium concentrations reported by the analytical laboratory and assumptions about stack flow until 1996, when the TAMM air effluent spreadsheets provide the release rates.

	B331 spreadsheets		**SAER**		**Other sources (rt) = routine; (ac) = accidental**	
Year	**Ci HT**	**Ci HTO**	**Ci HT**	**Ci HTO**	**Ci HT**	**Ci HTO**
1974	1336	812			1422[170]	831[170]
1975	1248	1108				
1976	1150	1672			1163[171]; 1161[172,173]	1659[171]; 1657[172,173]
1977	1215	2034			1127[174]; 1182[172,175]	1829[174]; 1965[172,175]
1978	1981	1923			2183[176]; 2271[177]; 2285[172]; 2284[178]	1853[176]; 1961[177]; 1960[172,178]
1979	2361	1600			2543[172,179]	1526[172,179]
1980	750	1459			800[176]; 802[180,172]; 837[181]	1377[182,180,172,176]; 1380[181]
1981	1097	1609			1007[183]; 1013[176]; 1034[184,172]	1518[184,176]; 1510[172]; 1545[183]
1982	608	1316			593[185]; 608[186,172]	1306[186,186,172,185]
1983	1798	1220			1831[187]; 1611[172]	1193[187,172]
1984[188]	1188	1136			1146[189]; 911[172]	1045[189,172]
1985[190]	1260	649			365[172]	624[172]
1986	487.7[191]	654.4	467.4	660.6	332 (rt) & 135 (ac)[172]; 467.7[192,193]	661[172]; 654.4[192]
1987	1423.0	1283.3	1387.6	1246.2	813 (rt) & 575 (ac)[172]; 1386.7[194]	1246[172]; 1227[194]
1988	2341.6	1636.1	2341.9	1635.6	2341.9[195]; 1542 (rt) & 800 (ac)[172]	1635[172]
1989	1300[196]	1749	1395	1554.4	1395[197,172]	1554[197]; 1555[172]
1990	538	684	581.8	699.5	572[172]	689[172]
1991	478	711	431.4	680.2		
1992	82.1	107	77	100		
1993	125	115	123.5	113.5		
1994	60.8	76.2	61	76		
1995	29.0	62.8	29	62.8	29.01[198]	62.79[198]
1996	33.6	181	33.5	181.2		
1997	31.9	267.4	31.9	267.4		
1998	24.8	85.2	25.1	83.8		
1999	66.8	213.6	66.8	213.7		
2000	4.81	35.4	5	35		
2001	1.72	18.3	1.7	18.3		
2002	3.47	32.9	3.5	32.8		
2003	6.33	103	6.0	104		
2004	4.41	12.1	4	12		
2005	1.57	30.2	2	30		

REFERENCES AND ANNOTATIONS (Table 5)

[170] Derived from the same analytical data as the spreadsheet results but with different assumptions about sample size

[171] Extracted from hand-drawn graph by S. Homann, "Building 331 Monthly Tritium Release for 1976".

[172] Obtained from papers in the possession of Rebecca Failor.

[173] From a hand-written summary (1976 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1976 Stack Reports in LLNL Radiological Air Effluent Records.

[174] Extracted from hand-drawn graph by S. Homann, "Building 331 Monthly Tritium Release for 1977".

[175] From a hand-written summary (1977 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1977 Stack Reports in LLNL Radiological Air Effluent Records.

[176] Hand-written data found reporting weekly Ci released of HTO and HT from both stacks in stack reports of the LLNL Radiological Air Effluent Records.

[177] Extracted from hand-drawn graph by S. Homann, "Building 331 Monthly Tritium Release for 1978".

[178] From a hand-written summary (1978 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1978 Stack Reports in LLNL Radiological Air Effluent Records.

[179] From a hand-written summary (1979 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1979 Stack Reports in LLNL Radiological Air Effluent Records.

[180] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1980, memo to distribution, January 15, 1981

[181] From a hand-written summary (1980 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1980 Stack Reports in LLNL Radiological Air Effluent Records

[182] An additional 40 Ci of tritium (undifferentiated species) was reported.

[183] From a hand-written summary (1981 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1981 Stack Reports in LLNL Radiological Air Effluent Records

[184] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1981, memo to distribution February 18, 1982.

[185] From a hand-written summary (1982 Effluent Data) of monthly gaseous releases from B194, B281, B212, B331 and B292. Sheet found in the 1982 Stack Reports in LLNL Radiological Air Effluent Records

[186] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1982, memo to distribution, February 2, 1983.

[187] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1983, memo to distribution, January 17, 1984

[188] The accidental HT release of about 5000 Ci of June 8, 1984 has been subtracted from the total.

[189] King. W.C., Radiation and Environmental Safety Division, Hazards Control Department, memo to distribution January 22, 1985.

[190] The accidental release of about 1000 Ci of January 24, 1985 has been subtracted from the total.

[191] 150 Ci were released in the week of December 15 – 22 1986 from Stack 1. This would correspond to the 135 Ci accident referred to in Reference 172.

[192] Gordon, Lisa E. Memo to D. Brekke. Subject: 1986 Effluent Monitoring Report for Building 331. January 30, 1987.

[193] 125 Ci were released in the week of December 15 – 22 1986 from Stack 1. This would correspond to the 135 Ci accident referred to in Reference 172.

[194] Mansfield, W. Gary. Memo to George Morris. Subject: Summary of 1987 Tritium Stack Effluents. January 19, 1988. These values assume different treatment of year-end based on attached weekly data than do the summary numbers in the memo.

[195] Biermann, A. Memo to D. Brekke, February 21, 1989.

[196] 1244 Ci from two sampling periods have been subtracted off the total for the year because the apparently high releases were actually artifacts of stack calibration (see Mansfield, memo to M. Singleton, April 13, 1990, below).

[197] Mansfield, G. Memo to M. Singleton, April 13, 1990

[198] Tate, P. Memo to M. Mintz February 9, 1996. Subject: B-331 Routine Tritium Report

Table 6. Stack effluent emissions from the SNL/CA Tritium Research Laboratory, Livermore, California

Year	Bubbler Data[199]	Sandia Data[200]		SAIC Data[201]		Accidents[202]
		HT	HTO	HT	HT0	
1979	5.72 (Nov. on)	5.72				
1980	25.27	25.27				
1981	42.93	42.93				
1982	201.63	201.63		88.4		
1983	104.66	21.2	74.2	200		
1984	164.01	18.5	145.6	110		2.5 HTO July
1985	512.24	128.1	389.3	450		
1986	760.21	128.6	614.7	760		200 HTO January
1987		1258.5	573	1257	570	1100 HT August
1988		542.6	1035.4	543	1047	124 HTO October
1989		180.3	658.8	178	656	11.5 HTO March
1990		50.8	243.9[203]	51	244	
1991		113.4	352.1[204]			
1992[205]		130	133.7			5/28/92; 4 Ci HT[206] 6/1/92; 36 Ci HT[206]
1993[205]		55.3	132.4			2.0 HTO October
1994[205]		4.1	91.2			
1995[207]		1.06	72.9			
1996		0.078[206]				

REFERENCES AND ANNOTATIONS (Table 6):

[199] Hafner, R. Personal communication. 2002.

[200] Garcia, T.B. and T.P. Gorman. Radiological Characterization and Final Facility Status Report Tritium Research Laboratory. SAND96-8004 UC-407; August 1966.

[201] Science Applications International Corporation. Review of Tritium Operations, Emissions, and Measurements in the Livermore Valley. Prepared for Lawrence Livermore National Laboratory, January 1993.

[202] Garcia, T.B.; Gorman, T.P. Radiological characterization and final facility status report Tritium Research Laboratory. Sandia Report. SAND96-8004UC-407. August 1996.

[203] Includes "22.7 Ci Evap"; TRL uses a low-level tritium evaporator to evaporate liquids with low tritium activity. Emissions are limited to 100 Ci per year (see endnote [201])

[204] Includes 1.368 Ci "Evap"

[205] Confirmed by the SNL/CA Site Environmental Reports

[206] Johnson, Alice J. Tritium Research Laboratory Cleanup And Transition Project Final Report. Sandia National Laboratory, Livermore, CA. SAND97-8009. February 1997.

[207] Chavarria, J.J. Memo to Becky Failor, January 22, 1996. Subject: Calendar year 1995 radiological effluent data for SNL/CA.

Table 7a. HT and HTO released from LLNL facilities since 1953 as determined for the Tritium Dose Reconstruction. CL is the confidence limit on the uncertainty.

Type of release	**Best estimate**	**2.5% CL**	**97.5% CL**
Routine HT	104,000	90,300	119,000
Routine HTO	74,700	65,200	85,500
Accidental HT[a]	612,000	490,000	730,000
Accidental HTO[a]	1,450	597	2,300
Sources of routine releases			
HT from Tritium Facilities	95,900	82,500	110,000
HTO from Tritium Facilities	65,800	56,500	76,500
HT from other stacks	8,130	5,570	10,900
HTO from other stacks	2,410	1,900	2,940
HTO from area sources	6,470	5,500	7,490

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.

[a] Accidental releases of HT were from Building 331.

[b] Accidental release of HTO was from Building 231

Table 7b. HT or HTO released from the Tritium Research Laboratory of Sandia National Laboratories/CA between 1979 and 1995. CL is the confidence limit on the uncertainty.

Type of release	**Best estimate**	**2.5% CL**	**97.5% CL**
HT routine	1,590	1,400	1,780
HTO routine	4,540	3,880	5,200
HT accidental	1100	670	1500
HTO accidental	200	80.0	320

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.

Table 8a. Stack, stack diameter, exit velocity, and distance, direction, and bearing towards the air tritium monitor (VIS) near the Discovery Center for LLNL and SNL/CA tritium sources.

Source	Stack height (m)[208]	Stack diameter (m)	Year measured or reported; exit velocity ($m\ s^{-1}$)	Direction towards VIS and closest adjacent sector (in parentheses); Bearing in degrees True N; Distance (m)
B212 ICT Stack	8.2[209]	0.2[210]	1974 through 1982[211]: 4.43	ENE (NE); 64.48°; 1332 m
B231 Stack[212]	9	0.1 – 0.46	0.54 - 23.6	ENE (E); 73°; 1226 m
B292 Stack	20.2[213]	0.71	1979[214]: 6.1 1981[215]: 12.6 1985: 11.1[216]; 14.4[217]; 26.5[218] 1986[219]: 9.1 1989[220]: 17.2 1992[221]: 15.4	ESE (E) 109.83° 1308 m
B331 South Stack (#1) North Stack (#2)	30	1.22	1968[222]: 5.7 (#1); 10.7 (#2) 1970[223]: 5.4 (#1) 1972[223]: 5.7 (#1); 11.1 (#2) 1976[223]: 5.65 (#1); 10.1 (#2) 1979[224]: 5.1 (#1); 9.1 (#2) 1981[225]: 5.3 (#1); 10.9 (#2) 1985[226]: 6.4 (#1); 9.7 (#2) 1987[227]: 6.9 (#1); 9.9 (#2) 1989[228]: 6.8 (#1); 9.6 (#2) 1991[229]: 7.6 (#1); 10.5 (#2) 1995[230]: 6.3 (#1); 8.8 (#2) 1997[231]: 5.9 (#1); 9.1 (#2)	ENE (NE) 75.06° (#1); 76.22° (#2) 1036 m (#1); 1046 m (#2)
B624 Incinerator	11.7[232]	0.46[232]	13.5[232]	NNE (NE); 23°; 542 m
DWTF (B695)	20	1.98	Dec. 2004[233]: 10.5	SSE(S); 167°; 812 m
SNL/CA TRL Stack	30[234]	1.0[234]	27.6[235]	NNE (NE); 32.9°; 1568 m

Table 8b. Area (diffuse) source, height of release, and distance, direction, and bearing towards the air tritium monitor (VIS) near the Discovery Center for LLNL facilities.

	Height of area source (m)[236]	**Direction towards VIS and closest adjacent sector (in parentheses); Bearing in degrees True N; Distance (m)**
B212 Room exhaust	4.0	ENE (NE); 64.48°; 1332 m
B231 WAA	1.0	ENE (E); 73°; 1226 m
B331 WAA	1.0	ENE (E); 75.64°; 1041 m
B514 Yard	1.25	NE (NNE); 35°; 674
B612 Yard	1.0	NNE (NE); 23°; 542 m
Evaporation Trays	0.0	ENE or NE; 400 - 457 m

REFERENCES AND ANNOTATIONS (Tables 8a and 8b)

[208] Taken from NESHAPs reports unless otherwise noted.

[209] The stack was measured in 2004 after being identified by D.S. Myers.

[210] Estimated inside diameter

[211] Calculated from flow rate of 0.275 m^3 s^{-1} mentioned in the air effluent monitoring notebook for 1975.

[212] These parameters were estimated from height of building and existing stacks, stack diameters and exit velocities for LLNL glove boxes reported for NESHAPs, and the assumption that glove boxes in the early 1950s had dimensions and exhaust rates similar to more recent glove boxes.

[213] The stack was measured in 2003 because of discrepancies in stack heights mentioned in various NESHAPs reports.

[214] Based on a flow rate of 2.4 m^3 /s reported in the stack effluent monitoring notebook and inside stack diameter of 0.71 m.

[215] Based on a flow rate of 5.0 m^3 /s reported in the stack effluent monitoring notebook and inside stack diameter of 0.71 m.

[216] Heikkinen, Dale W. RTNS-II Operations Guidebook. Lawrence Livermore National Laboratory, Livermore CA. UCID-20299. April 1, 1985.

[217] Trent, M. Memo to Kyle Griggs. Subject: B-292 Tritium Stack Releases: 1985, January 21, 1986.

[218] Calculated from the air flow rate of 10.5 m^3/s given in [8] and the stack diameter from [7] and NESHAPs reports.

[219] Trent, M. Memo to David Brekke. Subject: Stack releases from Building 292 (RTNS-II) and Building 865 (ATA). February 5, 1987.

[220] Biermann, A. Memo to D. Brekke. Subject: 1988 Air effluent monitoring results for radionuclides. February 21, 1989.

[221] Surano, K.; R.A. Failor; A.H. Biermann; R.L. Berger; R.J. Harrach. LLNL NESHAPs Project 1992 Annual Report. Environmental Protection Department, Lawrence Livermore National Laboratory, Livermore, CA. UCRL-ID-113867-93. 1993.

[222] Industrial Hygiene & Toxicology Section. Memo to Radiation Safety Section. Subject: Bldg. 331. October 7, 1968.

[223] Summary of Building 331 (Tritium Facility) Stack Flow Measurements, 1968-1987.

[224] Industrial Hygiene & Toxicology Group, Hazards Control Department. Memo to Chuck Folkers. Subject: Air flow measurements. November 8, 1979.

[225] Industrial Hygiene & Toxicology Group, Hazards Control Department. Memo to George Morris. Subject: Evaluation of Stack Samplers, Bldg. 331. September 22, 1981.

[226] Eneidi, Walt. Memo to George Morris. Subject: Airflow measurements, Bldg. 331. April 22, 1985.

[227] Eneidi, Walt. Memo to George Morris. Subject: Exhaust airflow measurements, Building 331 (April 3, 1987). April 8, 1987.

[228] Wong, M. Memo to G. Morris. Subject: Building 331 tritium sampler verification test results. May 2, 1989.

[229] LLNL NESHAPs Project Quarterly Progress Report. Environmental Protection Department, Lawrence Livermore National Laboratory, Livermore CA. UCRL-AR-108419-92-2. June 31, 1992.

[230] From calibration dated May 5, 1995 found in LLNL Radiological Air Effluent Emission Data for 1995 Volume 5 (of 5); Laboratory Records Center.

[231] Calibration October 29, 1997; data from LLNL Mac Network, EPD T5475, TAMM Server, Air Effluent for 1997 folder; also hard copy in air effluent archives.

[232] Radian Corporation. RCRA Part B Health Risk Assessment Phase 1; Existing Hazardous waste management incinerator. Lawrence Livermore National Laboratory, Livermore, CA. UCRL-21220; December 1989.

[233] Kent Wilson, personal communication, 2005.

[234] Garcia, Toff. Email to Ring Peterson April 22, 2002.

[235] See [233]. Based on flow rate of 1300 m^3 per minute.

[236] B212 Room Exhaust, B331 WAA and B612 Yard taken from NESHAPs reports; others estimated.

Table 9. Annual release rates of HT and HTO (Ci) from Building 231 (then Building 102; the predecessor of the LLNL Tritium Facility), one standard deviation (σ) uncertainty on a normal distribution, and truncated lower limit, when not zero. The truncated lower limit (LL) indicates that a known minimum of activity was released and is only mentioned if not zero or irrelevant. The upper limit (UL) was calculated but is included in the distribution. Releases of HT and HTO are negatively correlated (-0.4).

	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 ± 265	1250
1957	11400 ± 1900	8400	610 ± 90.0	870
1958	5530 ± 2000	4380	730 ± 240	1420

Table 10. Estimated Ci of HTO released annually from the (hypothetical) Building 231 WAA and the Building 514 Yard. The distributions are normal with one standard deviation (σ), left-truncated at zero if necessary. Release rates from the Building 231 WAA are positively correlated (0.5) with releases of HT and HTO from Building 231; release rates for the Building 514 Yard are positively correlated (0.4) with HTO released from the Tritium Facilities.

Year	Building 231 WAA	Building 514 Yard
1953	33.8 ± 28.7	5.40 ± 3.89
1954	67.5 ± 57.3	10.8 ± 7.79
1955	135 ± 115	21.6 ± 15.8
1956	100 ± 73.3	21.2 ± 12.8
1957	300 ± 66.8	24.4 ± 10.4
1958	156 ± 76.6	29.0 ± 20.0
1959	-	15.1 ± 6.08
1960	-	6.84 ± 2.74
1961	-	17.3 ± 5.04

Table 11. Annual releases of HT and HTO (Ci) from Building 212 and uncertainty as one standard deviation (σ) of a normal distribution.

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

***** A triangular distribution describes the minimum, most likely, and maximum values.

****** A uniform distribution describes an equal probability of sampling all values in the range.

Table 12. 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 of HT and HTO are negatively correlated (-0.4 for Stack 1 and -0.5 for Stack 2) through 1973.

Year	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

Table 12 continued

Year	Stack 1 HT	Stack 1 HTO	Stack 2 HT	Stack 2 HTO
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
1973	442 ± 209	518 ± 226	736 ± 318	864 ± 338
1974	463 ± 149	226 ± 70.8	629 ± 215	431 ± 144
1975	636 ± 131	344 ± 71.0	622 ± 147	773 ± 183
1976	296 ± 70.2	841 ± 201	958 ± 254	973 ± 260
1977	531 ± 143	1170 ± 319	742 ± 218	953 ± 284
1978	793 ± 178	954 ± 229	1510 ± 382	1180 ± 317
1979	1040 ± 189	734 ± 134	1410 ± 255	894 ± 163
1980	262 ± 49.1	509 ± 93.6	517 ± 116	908 ± 201
1981	516 ± 93.7	1130 ± 204	520 ± 94.4	403 ± 72.7
1982	122 ± 26.9	490 ± 108	483 ± 94.5	818 ± 160
1983	255 ± 57.7	318 ± 70.3	1510 ± 306	892 ± 175
1984	285 ± 124	487 ± 111	602 ± 254	556 ± 113
1985	140 ± 25.9	359 ± 65.1	198 ± 36.7	293 ± 53.2
1986	155 ± 30.4	228 ± 45.8	302 ± 56.8	402 ± 77.8
1987	242 ± 43.5	606 ± 109	1150 ± 207	644 ± 116
1988	437 ± 79.3	553 ± 101	1870 ± 340	1120 ± 205
1989	354 ± 65.7	896 ± 169	995 ± 185	725 ± 136
1990	143 ± 28.5	338 ± 66.5	419 ± 81.4	356 ± 68.3
1991	40.0 ± 7.76	204 ± 37.0	415 ± 80.3	492 ± 89.5
1992	20.1 ± 4.55	52.5 ± 11.9	59.5 ± 13.3	51.0 ± 11.4
1993	15.2 ± 3.39	41.5 ± 9.25	109 ± 23.9	72.9 ± 16.0
1994	4.11 ± 0.922	22.4 ± 4.99	56.8 ± 12.4	53.7 ± 11.8
1995	1.08 ± 0.283	5.95 ± 1.40	27.9 ± 6.19	56.8 ± 12.6
1996	0.813 ± 0.176	11.3 ± 2.10	32.7 ± 5.94	170 ± 32.8
1997	0.936 ± 0.174	10.1 ± 1.81	31.0 ± 5.57	257 ± 46.1
1998	0.608 ± 0.122	9.76 ± 1.89	24.4 ± 4.71	74.7 ± 14.4
1999	0.263 ± 0.0613	5.45 ± 1.09	66.5 ± 13.3	208 ± 41.5
2000	0.0485 ± 0.0140	4.05 ± 0.834	4.86 ± 1.01	31.1 ± 6.40
2001	0.274 ± 0.0968	2.40 ± 0.582	1.44 ± 0.308	15.9 ± 3.37
2002	0.0120 ± 0.00629	0.0126 ± 0.00558	3.47 ± 0.759	32.8 ± 7.12
2003	0.506 ± 0.271	0.0603 ± 0.0222	5.66 ± 1.41	103 ± 23.9
2004	0.600 ± 0.292	0.0300 ± 0.0118	3.82 ± 0.950	12.1 ± 3.33
2005	0.480 ± 0.254	0.0300 ± 0.0111	1.09 ± 0.313	30.1 ± 7.61

Table 13. HTO (Ci) estimated to have been released annually from the Building 331 WAA, the evaporation trays, and the Building 612 Yard before measurements were taken. The distributions are normal with one standard deviation (σ). Releases from the Building 331 WAA are positively correlated (0.5) with HT and HTO releases from the Tritium Facility; releases from the evaporation trays and the Building 612 Yard are positively correlated (0.4) with releases of only HTO from the Tritium Facility. After 1965, the correlation between the evaporation trays and the Building 612 Yard is -1.0.

Year	Building 331 WAA	Evaporation Trays	Building 612 Yard
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	4.30 ± 4.15	42.2 ± 20.9
1966	75.4 ± 62.1	4.30 ± 4.15	56.6 ± 30.1
1967	163 ± 130	4.30 ± 4.15	126 ± 52.3
1968	167 ± 133	4.30 ± 4.15	129 ± 53.4
1969	241 ± 193	4.30 ± 4.15	189 ± 80.9
1970	111 ± 91.5	4.30 ± 4.15	85.4 ± 44.4
1971	67.0 ± 54.6	4.30 ± 4.15	49.5 ± 25.3
1972	33.4 ± 26.5	4.30 ± 4.15	24.3 ± 11.5
1973	63.3 ± 50.7	4.30 ± 4.15	46.4 ± 21.3
1974	43.2 ± 34.1	4.30 ± 4.15	19.8 ± 9.92
1975	58.7 ± 45.8	4.30 ± 4.15	36.7 ± 14.3
1976	75.8 ± 59.4	4.30 ± 4.15	62.2 ± 22.7
1977	83.8 ± 65.8	-	77.6 ± 27.0
1978	110 ± 85.8	-	78.3 ± 26.5
1979	101 ± 78.3	-	59.7 ± 18.6
1980	54.3 ± 42.3	-	52.0 ± 17.0
1981	63.5 ± 49.4	-	56.2 ± 18.1
1982	47.3 ± 36.8	-	47.9 ± 15.1
1983	73.6 ± 57.4	-	44.4 ± 14.3
1984	47.7 ± 37.6	-	38.2 ± 12.1
1985	24.5 ± 19.0	-	23.6 ± 7.29
1986	26.9 ± 20.9	-	23.1 ± 7.36
1987	65.3 ± 50.8	-	45.8 ± 14.5
1988	98.3 ± 76.5	-	61.2 ± 19.5
1989	73.4 ± 57.0	-	59.4 ± 18.6
1990	31.1 ± 24.1	-	35.5 ± 14.4
1991	28.4 ± 22.1	-	25.5 ± 8.09
1992	-	-	3.79 ± 1.21

* These values are unreasonably high based on what is known about tritium concentrations in liquid evaporated from the trays. However, as explained in the text, the assumption was made that there must have been a waste accumulation area somewhere on site for these years. Because the location of such a site is unknown, it was assumed that the site was at the location of the evaporation trays.

Table 14. Annual HTO releases (Ci) ± one standard deviation (σ) uncertainty from the Building 331 WAA and the Building 612 Yard estimated from annual mean ambient air tritium concentrations from nearby tritium samplers.

Year	B331 WAA	B612 Yard
1992	4.42 ± 1.62	-
1993	9.22 ± 3.36	4.24 ± 1.54
1994	3.01 ± 1.10	1.92 ± 0.700
1995	31.4 ± 11.4	4.17 ± 1.52
1996	10.1 ± 3.68	12.4 ± 4.50
1997	8.04 ± 2.92	7.19 ± 2.61
1998	14.4 ± 5.23	8.47 ± 3.08
1999	15.8 ± 5.74	7.61 ± 2.77
2000	2.32 ± 0.850	5.47 ± 1.99
2001	0.679 ± 0.235	2.27 ± 0.689
2002	0.755 ± 0.260	2.33 ± 0.703
2003	8.70 ± 2.89	3.43 ± 1.03
2004	0.695 ± 0.240	3.18 ± 0.959
2005	4.76 ± 1.58	1.48 ± 0.447

Table 15. HTO (Ci) released annually from the Building 624 incinerator and Building 292. A triangular uncertainty distribution on the incinerator results for 1977 – 1985 is based on measurements taken in 1988 of all incinerated concentrations. Uncertainty on the Building 292 releases is normal ± one standard deviation (σ).

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

Table 16. Annual release rates (Ci) and uncertainty (σ) on normal distributions for the Decontamination and Waste Treatment Facility stack and area source.

Year	Stack HT	Stack; HTO	Area; HTO
2004	1.0 ± 0.818	19.0 ± 9.08	-
2005	0.107 ± 0.0532	2.62 ± 0.943	[0.21] (not modeled)

Table 17. Annual routine releases of HT and HTO (Ci) and uncertainty as one standard deviation (σ) from the SNL/CA Tritium Research Laboratory

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

Table 18. Dilution factors (χ/Q in s m^{-3}) with uncertainty (σ) on lognormal distributions at the Discovery Center for modeled sources other than the LLNL Tritium Facility.

Building	Years	χ/Q
B231 Stack	1953 - 1958	$2.389 \times 10^{-6} \pm 7.223 \times 10^{-7}$
B231 WAA	1953 - 1958	$2.758 \times 10^{-6} \pm 1.103 \times 10^{-7}$
B514 Yard	1953 - 1961	$1.076 \times 10^{-5} \pm 4.304 \times 10^{-6}$
B212 Room Air	1953 - 1967	$2.678 \times 10^{-6} \pm 8.474 \times 10^{-7}$
B331 WAA	1959 - 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	$2.494 \times 10^{-6} \pm 7.483 \times 10^{-7}$
B624 incinerator	1977 - 1988	$7.925 \times 10^{-6} \pm 2.380 \times 10^{-6}$
B292 Stack	1979 - 1989	$4.425 \times 10^{-7} \pm 1.427 \times 10^{-7}$
Sandia TRL Stack	1979 - 1995	$6.595 \times 10^{-7} \pm 2.008 \times 10^{-7}$
DWTF Stack	2004 - present	$3.472 \times 10^{-7} \pm 1.042 \times 10^{-7}$

Table 19. Dilution factors (χ/Q in s m^{-3}) with uncertainty (σ) on lognormal distributions for Stack 1 and Stack 2 of the LLNL Tritium Facility (Building 331)

Year	Stack 1 (south)	Stack 2 (north)
1959 - 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	$1.221 \times 10^{-6} \pm 3.055 \times 10^{-7}$	$1.055 \times 10^{-6} \pm 2.642 \times 10^{-7}$
1973 - 1978	$1.230 \times 10^{-6} \pm 3.080 \times 10^{-7}$	$1.077 \times 10^{-6} \pm 2.718 \times 10^{-7}$
1979	$1.240 \times 10^{-6} \pm 3.101 \times 10^{-7}$	$1.101 \times 10^{-6} \pm 2.758 \times 10^{-7}$
1980	$1.237 \times 10^{-6} \pm 3.093 \times 10^{-7}$	$1.081 \times 10^{-6} \pm 2.722 \times 10^{-7}$
1981	$1.233 \times 10^{-6} \pm 3.084 \times 10^{-7}$	$1.059 \times 10^{-6} \pm 2.653 \times 10^{-7}$
1982 - 1984	$1.217 \times 10^{-6} \pm 3.055 \times 10^{-7}$	$1.073 \times 10^{-6} \pm 2.694 \times 10^{-7}$
1985	$1.201 \times 10^{-6} \pm 3.004 \times 10^{-7}$	$1.086 \times 10^{-6} \pm 2.719 \times 10^{-7}$
1986	$1.192 \times 10^{-6} \pm 2.985 \times 10^{-7}$	$1.084 \times 10^{-6} \pm 2.715 \times 10^{-7}$
1987	$1.184 \times 10^{-6} \pm 2.965 \times 10^{-7}$	$1.083 \times 10^{-6} \pm 2.711 \times 10^{-7}$
1988	$1.187 \times 10^{-6} \pm 2.969 \times 10^{-7}$	$1.086 \times 10^{-6} \pm 2.720 \times 10^{-7}$
1989	$1.189 \times 10^{-6} \pm 2.974 \times 10^{-7}$	$1.090 \times 10^{-6} \pm 2.729 \times 10^{-7}$
1990	$1.177 \times 10^{-6} \pm 2.951 \times 10^{-7}$	$1.080 \times 10^{-6} \pm 2.709 \times 10^{-7}$
1991	$1.166 \times 10^{-6} \pm 2.919 \times 10^{-7}$	$1.068 \times 10^{-6} \pm 2.675 \times 10^{-7}$
1992 - 1994	$1.184 \times 10^{-6} \pm 2.974 \times 10^{-7}$	$1.088 \times 10^{-6} \pm 2.738 \times 10^{-7}$
1995	$1.203 \times 10^{-6} \pm 3.011 \times 10^{-7}$	$1.108 \times 10^{-6} \pm 2.775 \times 10^{-7}$
1996	$1.209 \times 10^{-6} \pm 3.025 \times 10^{-7}$	$1.095 \times 10^{-6} \pm 2.741 \times 10^{-7}$
1997	$1.216 \times 10^{-6} \pm 3.042 \times 10^{-7}$	$1.102 \times 10^{-6} \pm 2.761 \times 10^{-7}$
1998	$1.225 \times 10^{-6} \pm 3.067 \times 10^{-7}$	$1.098 \times 10^{-6} \pm 2.754 \times 10^{-7}$
1999	$1.240 \times 10^{-6} \pm 3.105 \times 10^{-7}$	$1.088 \times 10^{-6} \pm 2.733 \times 10^{-7}$
2000	$1.240 \times 10^{-6} \pm 3.107 \times 10^{-7}$	$1.115 \times 10^{-6} \pm 2.803 \times 10^{-7}$
2001	$1.245 \times 10^{-6} \pm 3.119 \times 10^{-7}$	$1.146 \times 10^{-6} \pm 2.881 \times 10^{-7}$
2002	$1.253 \times 10^{-6} \pm 3.142 \times 10^{-7}$	$1.125 \times 10^{-6} \pm 2.832 \times 10^{-7}$
2003	$1.254 \times 10^{-6} \pm 3.145 \times 10^{-7}$	$1.125 \times 10^{-6} \pm 2.831 \times 10^{-7}$
2004	$1.253 \times 10^{-6} \pm 3.147 \times 10^{-7}$	$1.125 \times 10^{-6} \pm 2.839 \times 10^{-7}$
2005	$1.231 \times 10^{-6} \pm 3.091 \times 10^{-7}$	$1.144 \times 10^{-6} \pm 2.890 \times 10^{-7}$

Table 20. Annual mean observed concentrations of tritium in air moisture (Bq L^{1}) and air (Bq m^{-3}) at VIS with one standard deviation (σ) uncertainty. Fallout and cosmogenic tritium background* has been subtracted.

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

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

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

Table 21. Annual absolute humidity (kg m^{-3}), as measured by water absorbed by silica gel and air flow through the sampler, and relative humidity from the LLNL meteorological tower. Uncertainty on absolute humidity is ± 5%; uncertainty on relative humidity is ± 10%.

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

Note: Absolute humidity was not calculated for 1973 because only a partial year of silica gel data were obtained.

Table 22. Mean annual HTO concentrations in the LLNL swimming pool in Bq L^{-1}, fraction of 12 months represented by annual sampling, uncertainty, and the annual maximum and minimum concentrations. Uncertainty is one standard deviation (σ) of a normal distribution.

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

APPENDIX

PARAMETERS FOR PRECIPITATION PATHWAYS

Concentrations of HTO in soil water can be calculated two ways in DCART. The first way uses dry and wet deposition velocities to determine the initial concentration of tritium in the soil water. Then, because dry deposited HTO is re-emitted from the soil, the soil concentrations must be adjusted manually to agree as closely as possible with empirical values. The second, much simpler and no less accurate approach, is to use the empirical values themselves to calibrate the concentrations in the soil based on air moisture concentrations (Peterson2006). This latter, simpler approach is used for the TDR. However, precipitation equations in DCART can be used to determine concentrations in rainfall should the need arise.

Parameters specific to the equations used to calculate concentrations of tritium in rainfall (see Equations 1 and 1a in Peterson 2006) include mean wind speed when raining, the washout coefficient, the fraction of time the wind blows towards the receptor when raining, and the annual rainfall rate. Tables A1, A2, and A3 describe site-specific LLNL values for these parameters when the Discovery Center is the receptor for several of the tritium sources addressed in the TDR. Additionally, the equation that estimates the fraction of time it rains in a year is described under the discussion of Table A3. For model testing, a data set of tritium in rain collected at the VIS monitoring location has been provided (Table A4)

Input (wind speeds, washout coefficients, and fraction of time the wind blows towards the receptor) for the precipitation model in DCART (Tables A1 and A2)

Wind speeds and the fractions of time that the wind blew towards the receptor while raining were calculated for 1997 – 2003[1] using 15-minute data from instruments at 10 m and 40 m on the LLNL meteorological tower. Each set of data (i.e., from 10 m or 40 m) was sorted by wind direction from the source to the receptor at the air tritium and rain sampling location, VIS. The listing below shows how the 15-minute wind directions were used to separate the meteorological data into sets of data to be applied to each source based on source release height. Ideally, meteorological data for modeling is obtained from the same height as the release. When this is not possible (as in these cases), a rough approximation can be made by applying, for example, 10 m data to area sources and sources with low stacks, and 40 m data to sources with high stacks.

- Wind directions from 236.25 to 258.75 degrees (towards ENE)
  - 40 m data for Building 331 stacks.
  - 10 m data for Building 331 WAA, Building 212, and Building 231
- Wind directions from 191.25 to 213.75 degrees (towards NNE)
  - 40 m data for SNL stacks.
  - 10 m data for Building 612 Yard and Building 624 incinerator

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[1] These parameter values were prepared in 2004 for the TDR. When the decision was made not to explicitly model concentrations of tritium in soil moisture for the TDR, there was no incentive to update the values..

- Wind directions from 281.25 to 303.75 degrees (towards ESE)
    - 10 m data for Building 292

Because the wind blows very infrequently towards VIS from some sources, data for each source for all years were averaged to obtain a more representative sample size and, consequently, a representative mean wind speed (Table A1). The uncertainty on the seven-year mean is the standard deviation of the mean of the annual wind speeds, which is large enough to include uncertainties from wind speed measurements (± 5%), sample size, whether or not the wind blew towards the receptor for the entire 15-minute period, and differences due to wind speed being measured at heights other than the release heights.

Washout coefficients, with lognormal distributions were based on Belot (1998) and were estimated from a figure illustrating washout coefficients for a rainfall rate of 1 mm h^{-1} at various release heights and various distances from a source. Estimated washout coefficients were 7.25×10^{-5} s^{-1} for the Building 331 stacks, 8.0×10^{-5} s^{-1} for the Building 331 WAA and Building 212, 9.0×10^{-5} $s^{-1}$for the Building 612 yard, 7.6×10^{-5} $s^{-1}$for Building 292, and 6.5×10^{-5} s^{-1} for SNL. Uncertainty on these numbers for a lognormal distribution is about ± 20%. The higher the effective release height, the lower the washout coefficient. In general terms, beyond close in, washout coefficients decrease with distance. Belot provides an adjustment for higher rainfall rates: the washout coefficient is proportional to $J^{0.7}$, where "J" is the rainfall rate. The high intensities of rain correspond to an increase in the spread of the raindrop size distribution with a shift towards larger drops that are less effective in gas scavenging. This explains why washout coefficient increases more slowly than does precipitation intensity.

Rainfall rates for the Livermore Site for each year were estimated by dividing the total year's rainfall (see Table A3) by the total minutes it rained, based on the assumption that, if any rain was measured during a 15-minute sampling period, it had rained for the entire time. This assumption is poor, however, and the uncertainty on the rainfall rate must account for this as well as for fairly large variability seen between years. On the assumption that rainfall rate is probably on average about the same regardless of the other meteorological conditions, a Livermore site-specific rainfall rate with a geometric mean (GM) of 2.39 mm h^{-1} and a geometric standard deviation (GSD) of 1.34 has been estimated and used to adjust the washout coefficients derived from Belot's figure (Table A2). The GSD of 1.52 on each washout coefficient accounts for the uncertainty on the washout coefficient estimated from Belot and the uncertainty on the rainfall rate.

The fractions of time that the wind blew from a particular source to the Discovery Center (sampling location VIS) are shown in Table A2. Frequency was obtained by counting the 15-minute segments in which rain occurred in a particular direction and dividing by the number of 15-minute segments in a year. An uncertainty of ± 10% on a normal distribution has been applied to account for instrument variability and whether or not it was actually raining. For years annual data were not available, means and standard deviations have been calculated from the 1997 – 2003 data. The standard deviation on the mean values accounts for the uncertainties on each individual value as well as for the variability in the values between years.

Annual rainfall at LLNL and uncertainty (Table A3)

The uncertainty on the annual rainfall rate obtained from past records from LLNL and the Livermore Valley is ± 10% (normal distribution) to apply the measured value to VIS. As well, for the few years when no measurements were taken on site, an additional 10% uncertainty has been applied.

The fraction of time it rains in a year is also needed as input by DCART. It was assumed that if rain was recorded during a fifteen-minute period with the tipping bucket at the LLNL meteorological station, it had rained for fifteen minutes. Fractions of each year that it rained were calculated for 1997 to 2003[2]. The uncertainty distribution on these numbers is assumed triangular, with the fraction of time estimated being the maximum, because it is unlikely that it actually rained throughout all 15-minute periods; the best estimate and minimum are assumed 85% and 50% of the maximum, respectively.

Based on the 1997 to 2003 15-minute LLNL meteorological data, an equation that relates rainfall rate to fraction of time it rains was derived for all other years:

$$\text{Fraction of time it rains in a year} = 0.0569 \text{ x annual rainfall in m} \quad 0.000195$$

Note that this equation replaces the generic fraction (0.018 ± 0.0060) from Table A1, Part 1 of the TDR (Peterson 2006). Because this equation was derived from fractions that represent the maximum time it could have rained, and because the real fraction may be either higher or lower than the best fit to the linear regression, a triangular distribution with maximum, best estimate, and minimum at 1.1, 0.9, and 0.6 times the calculated value, respectively, has been assigned.

Observed concentrations of tritium in rainfall at the Discovery Center (Table A4)

Tritium has been analyzed in rainfall collected at the Discovery Center since 1990 as part of a larger rain-monitoring network at LLNL. The rainy season at LLNL lasts approximately from October through May and rain sampling occurs only during daylight hours during the week, so tritium concentrations in rainwater will never be as representative of annual releases as is ambient air monitoring, even if a representative number of samples were collected. Nevertheless, the data can be used to test models in a limited manner. Mean (all annual data were averaged, including non-detects) tritium measurements in rainfall with fallout background subtracted for the sampling location VIS are summarized in Table A4.

The uncertainty about tritium concentrations in rain is due to collection methods as well as to numbers of samples collected and randomness of sampling. Sample buckets are collected soon after the rain ceases. Because tritium is highly mobile, a molecule of HTO can diffuse quite rapidly from a volume with high concentration (the water in the bucket) to a volume of low concentration (air moisture) and vice versa; the concentration of tritium in the rain may have changed between the end of the rain and the emptying of the bucket. Note also that concentrations of rainfall near a source are much higher towards the beginning of a rain, so rainfall concentrations may well depend on the duration of the rain, whether or not the wind has shifted during the rain, etc. Obviously, the more

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[2] 1997 - 0.0131; 1998 – 0.0291; 1999 – 0.0147; 2000 – 0.0188; 2001 – 0.0202; 2002 – 0.0135; 2003 – 0.0132.

samples are collected, the more representative is the annual mean concentration.

Fairly arbitrarily, uncertainty of ± 30% has been applied to the mean of those years with more than twelve rain samples; for the other years, assessment of uncertainty is too difficult to attempt meaningfully, although model predictions could be plotted against the scatter of the observations.

Table A1. Mean wind speed during rain and washout coefficients with 1 σ uncertainty to apply to all years for each facility that releases tritium at LLNL. Values are specific to a receptor at the Discovery Center. Wind speed has a normal distribution; washout coefficient has a lognormal distribution with a geometric standard deviation of 1.52.

	Mean wind speed (m s^{-1}) during rain	**Washout coefficient (s^{-1})**
Tritium Facility Stack	4.23 ± 0.446	$1.19\ 10^{-4}$
Building 331 WAA; Buildings 212 and 231	3.18 ± 0.301	$1.31\ 10^{-4}$
Building 612 Yard; Building 624 incinerator	3.77 ± 0.282	$1.47\ 10^{-4}$
Building 292 Stack	2.13 ± 0.279	$1.25\ 10^{-4}$
SNL TRL	4.86 ± 0.439	$1.07\ 10^{-4}$

Table A2. Annual and long-term (1997 – 2003) mean values ± uncertainty (1 σ) for the fraction of time the wind blows when raining towards the Discovery Center from facilities releasing tritium. Uncertainty on the annual frequencies is ± 10%.

	Tritium Facility Stacks	**Building 331 WAA; Buildings 212 and B231**	**Building 612 Yard; Building 624 Incinerator**	**Building 292 Stack**	**SNL TRL**
Mean ±	0.124 ± 0.0394	0.119 ± 0.0312	0.147 ± 0.0266	0.0372 ± 0.0119	0.154 ± 0.0234
1997	0.0766	0.0870	0.168	0.0350	0.171
1998	0.113	0.113	0.164	0.0314	0.169
1999	0.190	0.171	0.153	0.0330	0.183
2000	0.108	0.116	0.114	0.0624	0.129
2001	0.0876	0.0833	0.109	0.0410	0.123
2002	0.142	0.117	0.178	0.0297	0.165
2003	0.149	0.147	0.145	0.0281	0.137

Table A3. Annual rainfall in m at LLNL and one standard deviation (σ) uncertainty

Year	Rainfall (m)	Year	Rainfall (m)
1952*	0.540 ± 0.0763	1979	0.377 ± 0.0377
1953*	0.198 ± 0.0280	1980	0.298 ± 0.0298
1954*	0.331 ± 0.0468	1981	0.404 ± 0.0404
1955*	0.434 ± 0.0613	1982	0.531 ± 0.0531
1956*	0.272 ± 0.0385	1983	0.853 ± 0.0853
1957*	0.317 ± 0.0448	1984	0.274 ± 0.0274
1958*	0.467 ± 0.0661	1985	0.256 ± 0.0256
1959	0.235 ± 0.0235	1986*	0.334 ± 0.0473
1960	0.308 ± 0.0308	1987*	0.315 ± 0.0446
1961	0.246 ± 0.0246	1988*	0.208 ± 0.0294
1962	0.345 ± 0.0345	1989*	0.243 ± 0.0344
1963	0.391 ± 0.0391	1990	0.230 ± 0.0230
1964	0.298 ± 0.0298	1991	0.277 ± 0.0277
1965	0.375 ± 0.0375	1992	0.305 ± 0.0305
1966	0.228 ± 0.0228	1993	0.402 ± 0.0402
1967	0.433 ± 0.0433	1994	0.303 ± 0.0303
1968	0.345 ± 0.0345	1995	0.541 ± 0.0541
1969	0.401 ± 0.0401	1996	0.501 ± 0.0501
1970	0.480 ± 0.0480	1997	0.250 ± 0.0250
1971	0.237 ± 0.0237	1998	0.523 ± 0.0523
1972	0.303 ± 0.0303	1999	0.245 ± 0.0245
1973	0.531 ± 0.0531	2000	0.295 ± 0.0295
1974	0.245 ± 0.0245	2001	0.339 ± 0.0339
1975	0.330 ± 0.0330	2002	0.271 ± 0.0271
1976	0.167 ± 0.0167	2003	0.244 ± 0.0244
1977	0.200 ± 0.0200	2004	0.278 ± 0.0278
1978	0.403 ± 0.040	2005	0.455 ± 0.0455

* Uncertainty for these years is ± 14.1%, instead of 10%, because no rainfall measurements have been found for the Livermore site of LLNL.

Table A4. Number of rainfall samples collected each month for all months except July and mean annual tritium concentration (fallout background subtracted) of rain sampled at VIS (Discovery Center), LLNL. Non-detects are shown as shaded. Uncertainty on the means for 1991 and 1992 is ± 30%.

					Month of sample							**Annual mean concentration**
Year	J	F	M	A	M	J	A	S	O	N	D	Bq L^{-1}
1990										2	3	117
1991	1	3	6	1	2	1	2		1	1	3	47.6
1992	2	7	4	1		1			1	1	3	12.7
1993	1	1	1						1	1		23.3
1994	1		1	2	1					1		6.45
1995		1	1	1	1	1					1	5.73
1996	1			2	1				1			5.44
1997	1									5 (1)	2 (1)	6.21
1998		1									1	4.91
1999	1	1	1	1								36.2
2000	1	1	1	1				1	1			3.05
2001	1	1	1	1						1		0.558
2002			1		1					1	1	1.98
2003				1							2 (1)	8.57
2004		2(1)							1			2.41
2005	1	1										NA

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