UCRL-TR-211722

Historical Doses From Tritated Water and Tritiated Hydrogen Gas Releases to the Atmosphere from Lawrence Livermore National Laboratory (LLNL)

Part 2. LLNL Annual Site-specific Data 1953 — 2003

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May 2005

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Auspices Statement

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

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ABSTRACT

It is planned to use the tritium dose model, DCART (Doses from Chronic Atmospheric Releases of Tritium), to reconstruct dose to the hypothetical maximally exposed individual from annual routine releases of tritiated water (HTO) and tritiated hydrogen gas (HT) from all Lawrence Livermore National Laboratory (LLNL) facilities and from the Sandia National (SNL) Laboratory's Tritium Research Laboratory over the last fifty years. DCART has been described in Part 1 of "Historical Doses From Tritiated Water And Tritiated Hydrogen Gas Released To The Atmosphere from Lawrence Livermore National Laboratory (LLNL)" (UCRL-TR-205083). This report (Part 2) summarizes information about annual routine releases of tritium from LLNL (and SNL) since 1953. Historical records were used to derive facility-specific annual data (e.g., source terms, dilution factors, ambient air concentrations, meteorological data, including absolute humidity and rainfall, etc.) and their associated uncertainty distributions. These data will be used as input to DCART to calculate annual dose for each year of LLNL operations. Sources of information are carefully referenced, and assumptions are documented. Confidence on all data post-1974 is quite high. Prior to that, further adjustment to the estimated uncertainty may have to be made if more information comes to light.

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INTRODUCTION

As of this writing, as much as 3.5 10^{16} Bq (nearly one million Ci) of tritium have been released as tritiated water (HTO[1]) or tritiated hydrogen gas (HT[2]) over the lifetime of the Lawrence Livermore National Laboratory (LLNL) at its Livermore, California site. About two-thirds of this total came from two accidental releases of HT in 1965 and 1970. Tritium from these and smaller routine releases from LLNL and, to a much lesser extent, neighboring Sandia National Laboratory (SNL), has been responsible for nearly all radiological dose received by a hypothetical member of the public over the last fifty years.

A careful reanalysis of annual release rates and resulting doses is needed for several reasons:

1. Doses to the public from routine emissions from LLNL were not calculated prior to 1974.
2. Routine doses to the public during and after 1974 were calculated using three different models with different assumptions.
3. Recently developed tritium dose models include more pathways to dose and account for dose from forms of tritium (e.g., organically bound tritium) not found in models currently used for regulatory compliance.
4. Doses estimated in the past were deterministic and conservative. Dose predictions should be stochastic and define a small probability that a realistic dose should not be exceeded.

Doses from past accidental releases, both large and small, should also be predicted with 95% confidence intervals using the best time-dependent tritium-specific environmental transport and dose model available. Although dose consequences from the two major accidental releases have been assessed and shown to have no impact on public health (Myers et al. 1973; ATSDR 2003), additional analyses would be valuable nonetheless, because all important dose assessments should be made using more than one model and by more than one modeler (Peterson et al. 1996; Thiessen et al. 1997).

In addition to doses predicted by computational models, an empirical confirmation of air concentrations (and, by inference, release rates and dose) can be obtained by analyzing the organically bound tritium (OBT) in annual tree rings.

To accomplish a complete tritium dose reconstruction from LLNL releases, as described above, a six-part series, "Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory" is planned.

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[1] Throughout the report, aqueous tritium in any form (e.g., T_2O, DTO and HTO) will be referred to as HTO. Only an estimated 2.2 10^{15} Bq (about 60,000 Ci) of HTO have been released from LLNL and SNL.

[2] Throughout the report, gaseous tritium in any form (e.g., T2, DT, and HT) will be referred to as HT.

- Part 1 describes DCART, the stochastic model that will be used for the proposed dose reconstruction from routine emissions; it has been published (Peterson 2004a). Corrections to some parameter values and some clarification may be found in Appendix A.
- Part 2 (this report) presents the available information about past releases, both routine and accidental, and the resulting site-specific input to DCART for routine releases.
- Part 3 will present doses with 95% confidence intervals for 1974 – 2003 based on modeling from source terms and on observed air concentrations.
- Part 4 will present doses with 95% confidence intervals for 1953 – 1973 based on modeling.
- Part 5 will present doses with 95% confidence intervals for accidental tritium releases using a tritium-specific time-dependent model.
- Part 6 will present an analysis of concentrations of OBT in tree rings and will relate the concentrations of tritium in the tree rings to concentrations in air. Doses estimated from measured OBT in tree rings will be compared to those predicted from source terms.

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. This document serves as the background and archive of the dose reconstruction.

Dilution factors (χ/Q in s m^{-3}) for each source will be calculated for the closest long-term publicly accessible location (the LLNL Discovery Center, formerly the Visitors Center – see "VIS" in Figure 1) using the regulatory model CAP88-PC (Parks 1992, 1997) and site-specific meteorological input. In DCART, the dilution factors will be multiplied by estimated source terms and added together to predict concentrations of tritium in air that will drive the calculation of annual doses with 95% confidence intervals.

Using the Discovery Center as the location at which to calculate the reconstructed doses was chosen for two reasons. First, it is very close to the UNCLE Credit Union, which has been used for NESHAPs[3] compliance since 1994 as the location of LLNL's site-wide maximally exposed individual (SW-MEI); second, it is the location of an air tritium monitoring station (VIS) that has sampled tritium in air continuously since 1974. For the first phase of the dose reconstruction (Part 3, covering 1974 – 2003), all predicted air concentrations at VIS will be compared with the annual mean observed air concentrations. Thus, coincident with the dose reconstruction, model performance will be analyzed, which may help explain results from 1953 - 1973 (the second phase of the dose reconstruction, to be covered in Part 4) when source terms are not so well known. In Part 3, in addition to calculating doses from predicted air concentrations, doses will be calculated based on the air concentrations observed at VIS. The 95% confidence interval on doses predicted from observed air

[3] 40 CFR Part 61, Subpart H (National Emission Standards for Emissions of Radionuclides Other Than Radon from Department of Energy Facilities) (NESHAPs) (USEPA 1989).

concentrations is expected to be much smaller than that on doses predicted from source terms and dilution factors.

For each year, all principal sources will be included in the model run. Although LLNL is not responsible for doses due to SNL releases, nevertheless releases from SNL must be accounted for because they will have contributed to the air concentrations measured at the Discovery Center. Minor sources are assumed subsumed by the uncertainty on the major sources and will be ignored unless the observed air concentrations at VIS fail to be predicted. The doses predicted by DCART have only a 2.5% chance of being exceeded. Every effort has been made to use realistic assumptions, but if questions cannot be resolved, the assumptions will err on the side of conservatism.

Part 5 of the dose reconstruction will include calculations of the doses from the two large HT releases of 1965 and 1970 and from a few much smaller accidental releases using a state-of-the-art time-dependent tritium model such as UFOTRI (Raskob 1993). Modeling of tritium after an accidental release needs to account for the deposition of HT or HTO and the emission of HTO from the soil over time. In addition, it must account for the time-dependent uptake and loss of HTO from the plant and the production of organically bound tritium (OBT) during exposure to HTO. This detail of modeling requires meteorological data unavailable after so many years, but an attempt would be made to recreate the meteorological conditions to obtain the best possible estimate of dose. Although the ATSDR (2003) dose predictions from the two large releases accounted for deposition of HT and emission of HTO, formation and ingestion of OBT were neglected. At the same time, other assumptions by ATSDR, such as the mean HT deposition velocity, were overly conservative. A re-evaluation of these accidental releases based on different modeling and assumptions will probably result in similar dose estimates. Dose would be estimated for the hypothetical individual at VIS as well as for the individual likely receiving the highest dose.

Rather than wait to estimate doses from accidental releases until such a time as they can be modeled even more accurately, however, DCART can be used in Parts 3 and 4 of the dose reconstruction to predict upper dose limits (i.e., a dose that cannot be exceeded by the accidental release) for the accidental releases. Within certain limitations met by tritium (i.e., only first order reactions and a relatively short biological or environmental half-life), dose from an acute release may be predicted using an equilibrium model, because the dose integral over infinite time per unit of release is numerically equal to the dose at a future steady-state when that release is repeated indefinitely at unit rate. If it is assumed that the accidental release is spread out over the year and that the winds during that year blow only in one direction, then the integrated air concentration to which the individual is exposed is the same as that to which the individual was exposed to during the accidental release. The χ/Q for this can be obtained from a model for short-term dispersion, such as HOTSPOT (Homann 1994). All other assumptions in DCART are conservative, given that the diet is annual; after an accidental release, only a small portion of the diet is likely to be contaminated (albeit at higher levels for shorter periods of time). Significant ingestion exposure after an accidental release is either due to HTO taken up by

vegetables during the accidental release (but rapidly lost) or to OBT in foodstuffs that must have formed either during the accidental release or from HTO re-emitted from the soil for a few weeks after the accidental release. The 95th percentile dose obtained with DCART in this case will likely exceed the dose predicted by a time-dependent model.

The first section of this report (Background Information and Documented Release Data) is comprised of a discussion of what is known in general terms about the history of tritium facilities and releases accompanied by a set of tables that lists reported releases, both routine and accidental, from LLNL and SNL from 1953 to 2003 along with the primary and secondary sources of the information (in footnotes) (Tables 1 – 6). 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 7 – 21. A description of the assumptions and rationale behind the selection of input parameters and associated uncertainties is presented in the text for each table.

The site-specific, annual input for DCART includes:

- Physical descriptions of sources releasing tritium and their locations relative to the air tritium monitor near the Discovery Center at LLNL (location "VIS"). (Table 7)
- All source terms and speciation by year for releases treated as routine from all facilities. These are the release rates and associated uncertainties that will be used as input to DCART. (Tables 8 – 14)
- Dilution factors (χ/Q in s m^{-3}) at VIS for all sources of tritium. Dilution factors were obtained using the Environmental Protection Agency's approved regulatory dispersion and dose model, CAP88-PC (Parks 1992, 1997). Uncertainties are provided for the dilution factors. (Tables 15 and 16)
- Observed mean annual tritium concentrations in air moisture and in air at VIS from 1974 to 2003. (Table 17)
- Absolute and relative humidity at LLNL. (Table 18)
- Observed mean annual concentrations of HTO in the water of the LLNL swimming pool, 1988 – 2001. (Table 19)
- Annual rainfall from 1952 through 2003 and input needed for the DCART precipitation model. (Appendix B)

BACKGROUND INFORMATION AND DOCUMENTED RELEASE DATA

Background information

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. This report addresses releases from:

Building #	Stack; years of operation	Room air or area source; years of operation
231	Tritium Facility Operations (Building 231); 1952 -1958	NA
212	Insulating Core Transformer (ICT); 1966 - 1988	Accelerator; 1953 – 1966 Cyclotron; 1955 - 1971
331	Tritium Facility (TF1, TF2); 1958 - present	Waste Accumulation Area (WAA); 1958 - present
514	NA	Tank Farm (FM); 1960 - 2003
Taxi strip	NA	Evaporation trays; 1962 - 1976
612	Building 624 incinerator; 1977 -1988	Container Storage Area (CSA); 1965 - present
292	Rotating Target Neutron Source (RTNS): 1979 - 1987	NA
SNL	Tritium Research Laboratory (TRL); 1979 - 1995	Probably more than one WAA or storage area

These operations, a mixture of major and minor sources, are all 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 Table 7 for those sources which will be used as model input. Operations, grouped by location and dates, are described below.

Tritium Facility Operations in Building 231

Late in 1952, shortly after the laboratory opened, LLNL began working with tritium when a demand for a source of 14-MeV neutrons arose as a consequence of the infant laboratory's entry into thermonuclear weapons research (Gede and Gildea 1980). Tritium operations began in a wing on the southeast corner of Building 231 (then called Building 102). Work could have occurred in what is now the vault or in a wing adjacent (Room 1128). Both locations have stacks that could have been used for tritium releases between 1953 and 1958. In 1958, operations transferred to Increment 1 of the newly built Tritium Facility (then called Building 172; now called Building 331). 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 be estimated within plus or minus 10% of the likely value (Otsuki, 2004).

With the exception of an informal memo reporting an accidental release in 1954 (see Table 4), the only mention of releases prior to 1956 (see Table 1) is a roughly

estimated 2000 Ci (7.4 10^{13} Bq) per year (see Table 2) for each of 1953, 1954 and 1955. 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 (7.4 10^{13} Bq) each year. Assumptions made in preparing source terms for Building 231 may be found under the discussion of Table 8.

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) accelerator 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 (there was a 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 had to be added to the roof to reduce the neutron dose levels outside the building to reasonable levels. In 1986, molecular sieve samplers were installed to obtain more detail on 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 1966. 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, with just 1% of the beam current of the ICT, the wooden roof provided adequate shielding. Elsewhere in Building 212 (153) 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 were apparently only estimated after the fact with high uncertainty (Table 3); one accidental release of 200 Ci (7.4 10^{12} Bq) from the cyclotron was reported in 1961 (Table 4). Assumptions made in preparing source terms for Building 212 may be found under the discussion of Table 9.

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

Operations began in Increment 1 (south stack) of Building 331 (then Building 172) in 1958. In 1962, Increment 2 (north stack) became operational. Both stacks are still operational and being monitored in 2004, although clean-up in Increment 1 has reduced emissions to extremely low levels.

Before the stacks were monitored, 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.

In the first quarter of 1961, a system for monitoring tritium in the stack effluent of building 172 (i.e., the south stack) was installed. The system extracted a sample of the stack gas at a point above the outlet of the stack blower and into a 1 L ion chamber. The ion current was measured by a Keithley log micro-microammeter (sic) 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; 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 one 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} (3.7 10^{11} Bq m^{-3}); the digital system would alarm when 50 Ci (1.85 10^{12} Bq) was 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 2-liter and a 200 mL ion chamber were reported in 1981 (G. Morris 1981); somewhat later the need to expedite the completion of this new real-time stack monitoring system was noted (R. Morris and Ozaki 1982). The new system (Facility Safety Procedure 1986) consisted of an analog system, primarily for alarming, consisting of a 200 cc ion chamber, an air-flow indicator, and an alarm panel and a digital stack activity integrator, for estimating releases, consisting of a 2 liter ion chamber, a recycling integrator, a scalar, and a printer. 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). This system is still in use in 2004.

By the last weeks of 1971, molecular sieves had been installed to sample stack exhausts in addition to the ion chambers[4]. During 1972, it appears that only HTO was measured (i.e., only the results from one mole 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 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 weekly (historically every 5 to 8 days except in 1992 when samples were collected biweekly), they were processed by heating to 550° C under vacuum and the water and HTO was 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). Starting in 2005, the mole sieves will be replaced by four ethylene glycol bubblers, first in Stack 2 and then in Stack 1. The HTO is trapped primarily by one bottle, but any additional is trapped by a second bottle; HT passes through both HTO-capturing bottles and over a palladium catalyst for conversion to HTO that is then trapped by a second set of bottles. Samples are measured by scintillation counting.

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

The ionization chambers ran concurrently with the molecular sieves. In 1973 (Chew 1973), for a two-month period, the total tritium estimated using mole 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 mole sieve results[5] 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 mole sieves in Stack 2 agreed quite well. On the premise that the new monitoring would system would soon be operation, 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 mole sieves, at least until releases dropped in about 1991. In recent years, releases are far below the sensitivity of the ion chambers. Because of this, ion chamber estimates of releases are more than an order of magnitude higher than those estimated from the mole sieves.[6]

Release rates after 1961 were estimated based on ion chamber data (Table 1). After molecular sieves were introduced in 1974, results were measured as HTO and HT (Tables 1 and 5). Results of air effluent monitoring of Building 331 have been reported in the LLNL Site Annual Environmental Report (SAER) since 1974, although speciation was not reported until 1986 (Table 5). The first accidental release that was acknowledged to be an accident at the time it took place was the 360,000 Ci (1.33 10^{16} Bq) release of HT on January 20, 1965. Assumptions made in preparing source terms for the Tritium Facility may be found under the discussion of Table 10.

Stack flow rates were measured periodically (Table 7) after 1968 when three holes were drilled at 120° 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 several monitored releases of SF_6.

According to Dreicer (1985), no liquid waste from the Tritium Facility was poured down drains. It was collected in carboys[7]. Stone et al. (1982) distinguished between liquid effluent that drains to the sanitary sewer at levels not considered to be a significant hazard to ground water and liquid waste, which is not released to the sanitary sewer.

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

[6] Based on a comparison of Ci/week estimated by ion chambers and mole sieves found in the annual facility-specific air effluent spreadsheets for 1996 through 2003, which are maintained by the Terrestrial Atmospheric Monitoring and Modeling Group with the support of data management.

[7] Carboys contained somewhat higher concentrations of radioactive liquids than did the retention tanks (see Building 514 Tank Farm) (Odell and Toy 1979).

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 apparently 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 need to relinquish the 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 has been monitored with an ambient air tritium silica gel sampler. 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 reported for NESHAPs and for the SAER from the WAA is a value agreed upon between TAMM and the Tritium Facility (Table 3). Assumptions made in preparing source terms for the Building 331 WAA before the air tritium sampler was installed may be found under the discussion of Table 11; assumptions after the sampler was installed may be found under the discussion of Table 12.

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 Tank Farm

Built in 1943, Building 514 (originally Building 127B) served as a storage area for solid wastes until 1960, when it was converted into a treatment facility for large volume, low-level radioactive waste (Dreicer 1985). 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). Both above ground and in-ground tanks were used to store liquid wastes from Building 331 and other facilities. Wastes were released in controlled manner to the sanitary sewer system if they met permitted criteria for tritium (e.g., in 1979, 20 mCi d^{-1} (7.4 10^8 Bq) (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 surge tanks (113,562 L in Dreicer 1985) for later treatment or disposal. In 1979, 189,000 L of tritium-contaminated water was being stored at Building 514. 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 had concentrations of less than 5 pCi g^{-1} soil.

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); by 1989 it was no longer mentioned in the Radiation Safety Plan for Building 514. Dreicer (1985) mentions 8300 gal (4 Ci; 1.48 10^{11})) of tritiated water that were evaporated over the course of one year (probably1984).

Because of the off-gassing of the storage tanks and the need to report diffuse sources of tritium to determine compliance with NESHAPs, an ambient air tritium monitor was placed in the Building 514 Tank Farm. It was operational from August 1991 through December 2003 when sampling terminated because Building 514 was being decommissioned.

Potential releases of tritium from the tank farm have been assessed each year for NESHAPs compliance. Estimates range from a minimum of 1.4 10^{-7} Ci (5180 Bq) for 1993 to a maximum of 5.1 10^{-4} Ci (1.89 10^{7} Bq) for 1992 with an average for the years 1992 – 2003 of 1.2 10^{-4} Ci (4.44 10^{6} Bq). These estimates are based on the EPA factor of 1000 used to reduce the actual aqueous tritium inventory on site to that potentially released. Doses from these releases to the hypothetical member of the public at the Discovery Center will be minute compared with the contributions of other diffuse sources (specifically the Building 331 WAA and the Building 612 yard) and will be ignored for the dose reconstruction, as will the unknown, but undoubtedly small, releases from the thin-film evaporator.

The inventory of tritium at Building 514 that has been estimated annually for NESHAPs compliance has ranged over more than three orders of magnitude. Thus to estimate the inventory for years when air tritium was not sampled is quite difficult, even when records of concentrations in tanks are available, as they are for all of 1975 (in 1976, a partial year's worth of counting data showed mean concentrations in the tanks). 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 samples were not released because they were too high in copper). Thus, although 75 Ci (2.78 10^{12} Bq) of tritium passed through Building 514 in 1975 (tanks with the three highest Ci contents – 14.6 (5.4 10^{11} Bq), 12.3 (4.55 10^{11} Bq), and 5.8 (2.15 10^{11} Bq)- were not released and perhaps went to the surge tanks), if the total tritium recorded for each tank is weighted by month, the total average inventory for Building 514 is 5 Ci (1.85 10^{11} Bq). This number

itself is inflated because the tanks were sometimes emptied without any tritium being present, and in this estimate, it is assumed that the tank contains tritium until it is once again emptied and refilled with tritium-contaminated liquid. A similar exercise cannot 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 (1.52 10^{10} Bq); for 1976, the median was 0.21 Ci (7.77 10^{9} Bq). The 1975 inventory in this estimation is 10 times the highest inventory submitted for NESHAPs (0.51 Ci - 1.89 10^{10} Bq -in 1992). Doses from this to the hypothetical person at the Discovery Center should still be very small compared with other sources. Thus, at least initially for the dose reconstruction, the Building 514 Tank Farm will be considered a minor source and will not be included in the dose calculations. If the air concentrations predicted for 1975 (and other years after air tritium sampling was begun) are lower than those measured at VIS, then the contribution of the Building 514 Tank farm may have to be reconsidered and source terms developed.

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 of liquid waste (1500 by 1959) 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[8] and volumes of water evaporated have not been found. Potential evaporation from each pan could equal the average annual evapotranspiration rates for towns in California with similar meteorological conditions to Livermore. Based on evapotranspiration rates for eleven locations[9] obtained from the California Irrigation Management Information System, Department of Water Resources, Office of Water Use Efficiency on the web, the evaporation rate for Livermore could be about 1.34 m y^{-1} of water. For one evaporation pan of 18 m^2, this means a potential 24 m^3 could be evaporated in a year, assuming the tray was kept filled with water at all times. At present drinking water standards, this would mean just 4.8 10^{-4} Ci (1.78 10^{7} Bq) of tritium

[8] In 1959, the contents of 200 carboys were sampled for gross alpha and gross beta only (Buerer 1983).
[9] Davis, Dixon, Brentwood, Carneros, Morgan Hill, Twitchell Island, Concord, Manteca, Modesto, Patterson, and Tracy.

evaporated from one pan in one year. At the regulatory levels for worker exposure to aqueous tritium for 40 h at the time (National Bureau of Standards 1959) this number could be 10,000 times higher or 4.8 Ci (1.78 10^{11} Bq) evaporated from one pan each year. At the maximum concentration observed at the Building 514 tanks in 1975, the tritiated water evaporated would be close to 100 Ci (3.7 10^{12} Bq).

Although about (sic) five trays may be seen in an aerial photograph from 1970 (Dreicer 1985), 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 per week (20 – 60 L), 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. 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, according to Buerer, the liners of the trays were rolled up with the residual solids for disposal. Given that tritium was so easily disposed of into the sewer and/or had such high concentrations that occupational exposure concentrations would have been exceeded, it might be assumed that some evaporation of tritium might have occurred, in that tritium was a co-contaminant of the true contaminant to be evaporated, but that large quantities were not evaporated to the environment. If this assumption is true, then the potential dose to the hypothetical individual at the Discovery Center will be only a tiny fraction of the dose received from other sources. It is thus inadvisable to attempt to estimate source terms for the evaporation trays except in the case that air concentrations predicted by the model from better-quantified sources fail to meet or exceed the observed air concentrations at VIS. If this occurs, then the assumptions for the evaporation pans must be revisited.

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), 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 pans indicated a peak tritium concentration of 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 pans.

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

Building 612 was constructed in 1965 to package generated 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 Building 514, 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 releases from the Buiding 612 Yard arose when LLNL had to demonstrate compliance with NESHAPs. Consequently, an air tritium monitor was installed in the Building 612 Yard in August 1991. Release rates for NESHAPs have been estimated by back-calculating a source term from either the mean or the median measured concentration of tritium in air. Assumptions made in preparing source terms for the Building 612 Yard before the air tritium sampler was installed may be found under the discussion of Table 11; assumptions after the sampler was installed may be found under the discussion of Table 12.

The incinerator at Building 624 was used between 1977 and 1988. 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). Stack monitoring may have been evaluated (Myers and Gordon, 1987) but was not instigated. The incinerator underwent removal and disposition in 1992 (Winstanley 1992). Stack parameters are known quite well, but release rates are not. If the liquid and solid wastes containing carcinogens had a concentration of less than 1 μCi g^{-1} (3.7 10^4 Bq g^{-1}) tritium (considered trace quantities (Odell and Toy 1979), it 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 (1.85 10^{10} Bq) day, and the number of days that burns could take place was limited to 100 annually (Godwin 1988). Assumptions made in preparing source terms for the Building 624 incinerator may be found under the discussion of Table 13.

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 (1.11 10^{13} Bq) 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 Plant Engineering. 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 20-30 Ci (7.4 10^{11} to 1.11 10^{12} Bq) per hour when both accelerators were operating. The scrubbing system was very efficient, with output concentrations being a factor of 10^6 – 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. Thus 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 source terms for Building 292 may be found under the discussion of Table 13.

In 1984 (Dreicer 1985), waste for Building 292 consisted of tritium-contaminated solvents, spent tritium targets and mole sieve cylinders from the catalytic scrubbers. The tritiated solvents were incinerated at Building 514; 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 migrated through the soil and was evapotranspired by local vegetation. The tank had been drained by November 12, 1991 (Mallon 1991), but soil moisture was still contaminated with tritium (e.g., about 36,000 pCi/L [970 Bq/L] in piezometer UP-292-001 in February 1994 [Hoffman et al. 1994]). An ambient air tritium monitor was positioned not far from Building 292 from August 1991 through July 2003. Between 1991 and 1996, the median annual air moisture concentration measured at the Building 292 ambient air tritium monitor fell about two orders of magnitude from 44,000 (1630 Bq) to 460 (17 Bq) pCi L^{-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.

Similarly to the Building 331 WAA, the Building 514 Tank Farm, and the Building 612 Yard, doses from Building 292 as a diffuse source (the soil, the tree, the building itself) were estimated initially for NESHAPs compliance based on concentrations in air measured at the adjacent air tritium sampler. Even at the two highest mean annual air moisture concentrations (44,000 (1630 Bq) and 19,000 pCi (703 Bq) L^{-1} in 1992 and 1993 respectively), the estimated dose to the

SW-MEI was not greater than 2.5 10^{-5} μSv (2.5 10^{-6} mrem) y^{-1}. 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 x 10^{-5} μSv (1.7 x 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. Sampling could have been discontinued based on this alone, but, as well, the tree was badly infected with red turpentine beetles and very unhealthy.

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

SNL Tritium Research Laboratory

The SNL Tritium Research Laboratory (TRL) was operational from 1979 to 1995. In November 1979, SNL installed ethylene glycol bubblers to sample releases from the TRL stack (Hafner, 2004). As well, there was an Overhoff ion chamber. Presumably releases were characterized as either HT or HTO at this point, although releases were reported as total tritium until 1983. The ethylene glycol 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. Assumptions made in preparing source terms for the TRL may be found under the discussion of Table 14.

Because the TRL was similar to the LLNL Tritium Facility, it is expected that there would have been at least one waste storage area from which tritium would off-gas. In addition, there are known to have been small releases from tritium operations at SNL that took place in B913 from mid-1970s until about 1984 (Hafner 2004). The effect of these potential contributions on air concentration or dose at VIS can only be estimated with much more difficulty than experienced with the diffuse sources on LLNL property; no attempt will be made to estimate source terms for these SNL sources unless air concentrations at VIS are under-predicted during the dose reconstruction.

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).

Estimates of tritium releases documented in Tables 1 - 6

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

Reported estimates (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 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 Hazard Control archives.

Sometimes the reporting makes it difficult to know whether or not the releases listed in this table truly represent 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 with the exception of an accidental release in 1954 (Bldg. Monitor 1954) that predates the memos summarized in this table.

Estimated source terms (HT and HTO summed) for LLNL Tritium Facility, both routine and accidental (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 B172), the present Tritium Facility. Data under the "Memos" column from 1953 to 1971 came from a 1973 memo from LLNL to the US AEC (Olsen 1973) estimating past releases from the Tritium Facility. This memo was generated on fairly short notice by D. Myers (2004f) and others and didn't involve a detailed review of the records. It primarily involved the health physicists talking to the senior people at a given facility and having them make a "guesstimate" of the tritium emissions for the years data weren't 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. The different reported release rates demonstrate one of the sources of uncertainty in the assessment of the source terms to be used in DCART. 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 and potential releases from most LLNL tritium sources other than the Tritium Facility (Table 3)

All the sources shown in this table were reported at some point in the SAERs (after 1974). All release rates shown after 1974 were published in the SAERs unless otherwise noted. Releases for Building 212 up 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 summation of the monthly releases; unfortunately, the summation was incorrect and should have been 34 Ci.

All sources have been discussed in detail in the appropriate sections of "Background Information" and will be discussed under the appropriate table-headings in the "Annual Release Rates" section. The rationale for each annual release rate and its uncertainty distribution based on numbers in this table will be presented below as will the rationale for estimating missing source terms will and an explanation for differences between some of the source terms in this table and those being used for the dose reconstruction (particularly for the Building 331 WAA).

Accidental releases of tritium from LLNL (Table 4)

This list of accidental tritium releases has been included with the summary of annual tritium release rates for LLNL for the sake of completeness and because a rough attempt will be made to estimate the upper limit of dose from accidental releases as part of Parts 3 and 4 of the dose reconstruction. All the incidents shown in Table 4 occurred at the LLNL Tritium Facility, with the exception of 1954 (Building 231) and 1961 (Building 212). As well, Otsuki (2004) remembers the very first accidental release at LLNL in 1953 that involved an accelerator making neutrons in Building 212 (then B153). Until the large release in 1965, the releases shown as accidental in this table, with the exception of the accidental release in 1954, were reported as normal operations because they were planned puff releases (Table 1). As sensitivity to larger releases grew, they were redefined as accidental (Souers 1988); by 1989, any unplanned tritium release greater than 100 Ci had to be reported, at least within LLNL (Facility Safety Procedures 1989)

Although, in an ideal world, all the releases in Table 4 should be modeled as accidental, in practice it is impossible to model them as such, primarily because the meteorological conditions driving the dispersion after each accidental release are often unknown. About the best that can be done to recreate conditions at the time of the accidental release is to develop the most likely weather conditions given the time of year, and, if known, the time of day. An extremely conservative approach would be to assume all accidental releases occurred with the wind blowing towards the Discovery Center with F stability. Neither of these approaches is reasonable. Also, a specialized time-dependent tritium model that accounts for deposition of HT or HTO and emission from soil needs to be used, along with assumptions about what food was growing at the time of the accidental release and how much was ingested at various times after the passage of the plume. Only a few models (e.g., Russell and Ogram 1992; Raskob 1993) can accomplish this.

For this dose reconstruction, releases of more than 1000 Ci HT that occurred between 1956 and 1964 will be included as part of the annual routine releases from LLNL. These large puff releases occur randomly in time and are similar in magnitude. Thus, although uncertainty may be quite high for any one particular annual dose, when all doses for this period are added or averaged, the uncertainty may be disregarded. Similarly, dose from smaller accidental releases (less than 1000 Ci) after 1965, which are quite small compared with the annual releases, will average out over time and will be accounted for in the uncertainty on the total activity released routinely from the facility. Thus they will be included as part of the routine releases. This approach leaves just a handful of accidental releases needing to be modeled explicitly (i.e., those that occurred in 1964, 1965, 1966, 1970, 1984, and perhaps 1985).

Curiously, Souers (1988) is the only mention of an accidental release of 24,000 Ci in 1964; no such release is mentioned in any of the memos from the Director's

Office to the AEC (Table 1). In 1964, the release reported to the US AEC for the second quarter was 10,000 Ci (Table 1). It is possible that the 24,000 Ci accidental release and the 10,000 Ci routine quarterly release are the same. Doses from the large releases of 1965 and 1970 have been estimated at the time from measurements (Myers et al. 1973) and calculated (ATSDR 2003). These accidental releases are actually easier to model than some of the smaller ones because wind speed and wind direction were recorded at the time of the accidental release, which tremendously reduces the uncertainty on predicted air concentrations. As mentioned in the introduction, the ATSDR calculations were conservative in many ways but did not include ingestion as a source of dose. Hopefully, as part of this dose reconstruction (Part 5), these two large accidental releases will be re-visited, and modeling the smaller ones will be attempted using an appropriate time-dependent tritium dose model.

Fortunately for any dose reconstruction, the significant releases, with the exception of 1954, have all been HT, so the dose consequences are probably only 5 – 15% those of comparable releases of HTO.

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

Table 5 summarizes the available information on the speciation of releases from the Tritium Facility obtained from spreadsheet or hardcopy, 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 measurements (Table 7). 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 (Mansfield 1987b), the Tritium Effluent Release Spreadsheet used at the time 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 would not have been recognized as such and therefore are 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 from 1978 of release rates, did not account for the change in order of results for those twelve weeks. This suggests that the analyst either didn't notice the change or knew that the laboratory had inadvertently applied the wrong descriptor with the results.
- Sometimes there are two analytical reports for 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. The value of preparing release rates independently from analytical data is to provide some insight into potential uncertainties in reported source terms. The bottom line 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.

<u>Stack effluent emissions from the SNL Tritium Research Laboratory (Table 6)</u>

Data on SNL's TRL obtained for the LLNL dose reconstruction 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 data. Although the SAIC values are likely incorrect, if the release rates for the SAIC and SNL data are summed for 1982 – 1985 and compared, the SAIC total is 86% that of SNL's. The dose for this period will therefore be similar regardless of source term used. Accidental releases from SNL *to the environment* were always included in the total reported release for the year. All will be modeled as part of the routine annual releases except for the 200 Ci HTO release in January 1986 and the 1100 Ci HT release in August 1987, which should be modeled as accidents because each is a large fraction of the annual release.

ANNUAL SITE-SPECIFIC MODEL INPUT WITH UNCERTAINTY

Parameters for dispersion modeling (Table 7)

Parameter values for stack and area sources are known with reasonable certainty, with the exception of Building 231[10]. Stack heights are either those used for compliance with NESHAPS or were measured especially for this dose reconstruction. Exit velocities do vary and cause variability in χ/Q, but the variability in χ/Q due to the exit velocity is less than the variability of the exit velocities. The critical parameter for an area source in CAP88-PC is the source height; although size and exit velocities for the area sources have been estimated, CAP88-PC is completely insensitive to these parameters.

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 Heikkenin (1985), Trent (1986) and Surano (1993); the other exit velocities shown were calculated based on stack flows and an inside stack diameter of 0.71 m (Heikkenin 1985).

All sources 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, but real air concentrations do not exhibit this characteristic. To better assess the potential air concentration at a location, the concentrations are a weighted average of the contributions from both sectors. Bearings shown are the direction from the source to the receptor. Distances were obtained using GPS.

Annual release rates with uncertainty documented in Tables 8 – 14

All annual release rates are shown in units commonly used at LLNL. These units (Ci) will be input into DCART, where they will be converted to becquerels for all calculations. Concentration and dose are in SI units in DCART.

Uncertainty on most source terms has been estimated by assessing the percent standard deviation for each source of uncertainty and taking the square root of

[10] Releases from Building 231 may have come from Room 1128 or the Vault. The stack parameters are different for the two locations according to NESHAPs (1992). The Room 1128 stack is given as 12. 2 m high with a diameter of 0.41 m and an exit velocity of 12 m s^{-1}; the Vault stack is given as 10.6 m high with a diameter of 0.6 m and an exit velocity of 6 m s^{-1}. Figure 2-1 (Larson 2002) shows the Vault Stack as 34 feet (10.36 m) on the above the roof; if the drawing is to scale, the height of the stack will be 18.6 m. From Air Effluent Stack Flow Data Files (1992) maintained by TAMM, the stack diameter is given as 0.5588 m and the exit velocity is given as 12.8 m s^{-1}.

the sum of the squares to give the percent or fraction uncertainty from all sources. Each source term and a numerical standard deviation calculated from the percent standard deviation will be input into the Crystal Ball®[11] software in DCART as a normal distribution. When the uncertainty is large, the lower limit of the distribution will be negative. To correct for this and yet account as much as possible for high uncertainty and potentially very low releases, the lower limit, if negative, will be truncated at 5% of the best-estimated release rate. When the uncertainty on the source is handled differently, the approach is explained in the appropriate section.

For those operations involving HT alone (e.g., the LLNL Tritium Facility), if there are high puff releases of HT, there are fractionally lower releases of HTO. Thus it might be desirable to inversely correlate the parameters for fraction of HTO and fraction of HT. However, counteracting this is the positive correlation between increased releases of HT and HTO in general. There is no satisfactory way to resolve this problem, and thus fractions of HT and HTO will not be correlated. Dose predictions are thus expected to err on the side of conservatism.

Generic uncertainty

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

Uncertainties for which general assumptions can be made include:

- 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 of operation 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 from ion chambers. As yet no information about the performance and reliability of the ion chambers in Building 331 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 efficiency filter. In 1975 (Hazards Control 1976), 2 Ci of HT were introduced over 65 minutes to the system to determine a stack calibration

[11] 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.

constant of 1.73 10^9 Ci/Å-min; it was noted that this calibration constant depends on a set flow rate (0.275 m^3 s^{-1}) and that a variation in flow rate can result in an inaccurate estimate of tritium discharges. Uncertainty on ion chamber release-rate estimates still needs to be addressed because it has been estimated to be as high as ± 80% (Till 2001). For the moment, the uncertainty, based on different ion chambers in use at the Savannah River Site (SRS), is estimated to be ± 14% based on the efficiency of the ion chamber counting system of ± 10% and variations in temperature and pressure that may affect the result (±10%). 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. 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 dose reconstruction because emission data are obtained from the mole 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 source term. 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 has been 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 source term 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 has been assumed to be 5%.

Flow rate through stack tritium samplers

Regardless of the method by which tritium is sampled (mole sieve, silica gel or bubbler) the flow rate through the sampler will vary as conditions change unless a flow controller is used, and the total flow will be uncertain unless a flow totalizer is installed. Based on many years experience with the ambient air silica gel samplers, where the mean flow rate was based on the average of rotameter flow meter readings at startup and finish compared with flow volumes measured with a flow totalizer, the uncertainty on the flow rate is about ± 14% (Peterson 2004b). This uncertainty has been applied to the different stack sampling systems.

Silica gel correction factor
A silica gel correction factor of 1.6 ± 15% (based on nearly four years data; 1600 samples) must be applied to all air tritium samples until 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 exchangeable water bound in the silica gel and is specific to the type of silica gel used by LLNL for the past few years. The LLNL correction factor and uncertainty applies to the two different masses of silica gel (about 1100 g and about 730 g) used since the development of the correction factor, because flow rate was adjusted so that the ratio of water collected to dry mass of gel didn't change. This correction factor and uncertainty has been applied to all years of silica gel sampling because of lack of knowledge about the specific types and mass of silica gel used historically. A few numbers published in the literature[12] 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 average 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 8)

As mentioned, normal operations during the early years of LLNL produced puff releases that strictly speaking should be modeled as accidental. However, because of the fairly random occurrence of these early releases and the difficulty associated with reconstructing meteorological conditions for each puff, all releases for each year will be modeled as if they were chronic.

Uncertainty on the source terms from Building 231 must account for uncertainties on

- reported source term,
- speciation

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 (sic) releases that date to before 1958 when Increment 1 of Building 331 became operational. As noted by Myers (2004f), the estimates provided for the Olsen memo were hurriedly compiled and given a large uncertainty to

[12] 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.

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, it has been assumed that 10% of the tritium released was 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 will be truncated at zero, given the total lack of information about releases.

For the years 1956 through the first half of 1958 (the assumption being that the second half of the year's releases were from the Tritium Facility) assumptions 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 are as follows:

1. The "best estimate" of total tritium released is 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 is estimated based on 1% conversion to HTO for known large releases and 50% conversion to HTO for known smaller releases (≤ 1000 Ci).
3. A lower bound of uncertainty for the HT portion of the release is set using the Crystal Ball software by assuming 10% uncertainty on the total best-estimated release (Otsuki 2004). This lower limit is:
    - 1956: 2800
    - 1957: 8400
    - 1958: 2450
4. To account for missing releases, the standard deviation of the estimated curies of HT released has been increased using the Crystal Ball software to include an upper bound for HT that is derived based on conservative assumptions:
    - 1956: assume a release of 3000 Ci for each of the missing quarters. Thus the standard deviation must set at 2000 so that the upper limit reaches 3470 plus 6000.
    - 1957: assume the missing quarter had a release of 5000 Ci. Thus the standard deviation must be set at 1900 so that the upper limit reaches 17000.
    - 1958: assume the missing quarter had a release of 3000 Ci. Thus the standard deviation must be set at 1100 to get to 6500.
5. For HT, the lower uncertainty bound is then truncated at the one determined based on 10% uncertainty on the total release.
6. 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 is the upper limit of the HT distribution times the estimated fraction of HTO:

- 1956: assume the upper limit is 0.1325 x 9470 = 1255. Thus the standard deviation of 530 must be 240. The lower limit of the distribution is truncated at zero.
- 1957: assume the upper limit is 0.051 x 17000 = 870. Thus the standard deviation of 610 must be 90.
- 1958: assume the upper limit is 0.08 x 6500 = 520. Thus the standard deviation of 280 must be 80.

Only recent NESHAPs data are available for the stack parameters, but exit velocity has no effect on the uncertainty of the source term when releases were estimated based on usage rather than on stack flow measurements and concentrations in stack air.

Derivation of annual release rates of HT and HTO from Building 212 and uncertainty (Table 9).

Uncertainty on the source terms from the Building 212 includes uncertainties on

- reported source term,
- 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

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 stack from the ICT.

The only estimates found for releases from Building 212 from 1953 through 1967 are from the memo to the AEC in 1973 (Olsen 1973; Table 3). Because the releases from the accelerators prior to the startup of the ICT were estimates and not measured values, stack sampling or exit velocities cannot contribute to uncertainty. Releases 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. Releases 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 uncertainty on HTO is truncated at zero to account for the possibility of no conversion to HTO.

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 source term after monitoring was instigated is high (± 5%, based on the long-term average for Building 331). Myers (2004c) expressed surprise that the source term 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 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 rectangular uncertainty for total tritium of 5 – 50. In addition, no record of releases has been found for 1973 (see Table 3), so a rectangular 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 been found, but the same parameter values were used every year between 1974 and 1982 to estimate release rates. This implies 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 ended up as HTO in this operation than 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; the uncertainty on the HTO release is truncated at zero. No uncertainty is assumed for speciation for the two years of data based on mole 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.

The releases from the 90-inch cyclotron in Building 212 are apparently not considered for 1968 – 1971 (Table 3). Quite possibly these were small relative even to the Cockcroft-Walton accelerator, with the exception of the apparently accidental release of 200 Ci in 1961. For the purposes of this dose reconstruction, these releases will be assumed to have a negligible impact on VIS and will be ignored.

Derivation of annual releases of HT and HTO from the LLNL Tritium Facility (Building 331) and uncertainty (Table 10)

In addition to generic uncertainties, the source terms from the Tritium Facility includes uncertainties on

- reported source term,
- stack emitting the release
- 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 of releases from Building 331 depend upon the year of release and how the release was estimated or monitored. Between 1958 and when in-stack tritium monitoring began in 1961, releases were estimated based on the known amounts of HT used for each experiment. For 1958, because of potentially large missing releases, the source term and uncertainty has been prepared using similar assumptions to those used for Building 231. It is assumed HTO is 1% for known large releases and 50% for known smaller releases (≤ 1000 Ci). A lower bound of uncertainty for the HT portion of the release is set at 1925 Ci using the Crystal Ball software by assuming ± 10% uncertainty on the total best-estimated release (Otsuki 2004). To account for a potential missing release of about 3000 Ci, the standard deviation of the estimated curies of HT released has set at 1100 Ci using the Crystal Ball software so that the upper bound is 5500 Ci. It is assumed the fraction of the total release that is HTO does not change even though the potential release of HT may increase. Uncertainty for HTO is selected so that the upper limit of the distribution is the upper limit of the HT distribution times the estimated fraction of HTO: 0.19 x 5600 Ci = 1070 Ci. To obtain this upper limit, the standard deviation of 444 Ci must be 210 Ci. The lower limit of the distribution is truncated at zero. This approach should account for uncertainty on the source term and on the speciation.

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 (based on Fleming 1961 and Foster 1961). For these two years, this means about 200 Ci additional from two quarters 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 source term was ± 10.5% for both years. Speciation was assumed 1% HTO for known large releases and 70% for the negligible and potential very small releases. Uncertainty on this speciation was ± 40%.

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 here to 1959 may actually fall in 1960 and vice versa. This does not affect the dose when summed over several years, but it may make the dose for one year higher than it should be, while the adjacent year's dose is lower. 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. Because of this, uncertainty on the speciation was assumed ± 25%, which is the same as observed in stacks monitored by silica gel (see below). As well, all uncertainty associated with stack sampling has been included in the overall uncertainty on the source term for 1961.

From 1962 onwards, with a complete record of releases reported quarterly, the uncertainty on the reported source terms was determined entirely by averaging the annual totals obtained from different memos or reports (Tables 1 and 5). In 1969, an assumption had to made about the activity released during the missing quarterly report; these assumptions were averaged to get the uncertainty. Averaging reported release rates will account 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 learn what stack flow was used to calculate the release rate and how it relates to the known calibration of the stack flows, but by averaging different values for estimated source terms, the uncertainty will be included. If all reported release rates were the same for a year, the uncertainty on the reported source term is given as 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 releases. 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 releases for the year. The apparent release of 24000 Ci HT in 1964 (Table 4) may eventually be modeled as accidental, but its existence is suspect. 10000 Ci was reported released to the atmosphere between May 1 and July 31, 1964, and there is no way to know if all was released on one day. All that can be done is to model the entire year's release as reported, and perhaps remodel it assuming an accidental release of 10000 or 24000 Ci release as well. Smaller accidental HT releases (e.g., 1986, 1987, 1991) have been treated as part of the routine releases (see discussion under Table 4). Only one HTO release might be considered accidental and 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 will be modeled as part of the routine releases; if the release had been spread over a couple of days, it would resemble the routine as much as the accidental. The largest uncertainty based on reported values for a Building 331 source term is for 1964. This uncertainty (± 105%) has been applied to releases from both stacks because the specific stack from which the release occurred was not identified until 1973. Uncertainty 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). Until 1974, only releases that summed both stack contributions were reported. Although both stacks are essentially collocated

with respect to VIS, knowing the quantity of tritium released from each stack is important because the dilution factor (s m^{-3}) is different for each stack due to different exit velocities (Table 7). Between 1974 and 1993, when the quantities of tritium released from each stack were reasonably proportional year-after-year, 38% (± 17%) was released from Stack 1 and 62% (± 29%) was released from Stack 2. These fractions and uncertainties have been used to assign annual release rates to each stack based on reported total releases for the second half of 1962 through 1973.

In 1961 and thereafter until stack releases were speciated in 1974, releases were spread out enough (i.e., no longer obviously acute) that the speciation based on the known history of Building 331 (1974 - 1993) was used. The fractionation and uncertainty on "normal" speciation is summarized below:

Total Release - Ci	Fraction HTO	% uncertainty	Fraction HT	% uncertainty
>630	0.54	25	0.46	31
400 - 629	0.64	18	0.36	33
80 - 399	0.69	2	0.31	64
2 - 79	.088	10	0.12	76

Between 1958 and 1973, only one annual release rate in each stack was less than 630 Ci (506 Ci from Stack 1 in 1972). Thus speciation was assumed to be 54% HTO and 46% HT from the second half of 1962 through 1973, except for Stack 1 in 1972, when it was assume 64% 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 HT and HTO released from each stack for the years before releases were speciated (1974), first the total tritium was divided between the stacks, and then the tritium released from each stack was speciated.

Flow rates measured in the Tritium Facility stacks vary over time (Table 7). 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 the stacks were measured at more than annual intervals, the average of the two measurements flanking the missing data have been averaged to obtain the exit velocity. For these years, the uncertainty on this derived exit velocity is the percent standard deviation of the mean, which has been applied to the source term. Prior to 1968 and after 1997[13], flow measurements were either not taken or were not calibrated for several years. Before 1968, the exit velocity as well as its uncertainty must be estimated for both stacks; for 1998 – 2003, mean annual exit velocities are known but uncertainty must 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

[13] 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.

been assumed. For 1998 – 2003, uncertainty is assumed to range from ± 5%. in 1998 to ± 13% in 2003. 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 mole 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 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 nearly 50% for 2003 for HT released from Stack 1.

Derivation of tritium release rates and uncertainty from the Building 331 WAA and the Building 612 yard before monitoring occurred (Table 11).

The Building 331 WAA is assumed to have been in existence for as long as the Tritium Facility. For the years prior to ambient air tritium sampling, 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 source term for the Building 331 WAA. Based on this, the total Ci released annually from Building 331 can be multiplied by 0.253 ± 71% to derive the air concentration (pCi m^{-3}) at the air tritium sampler, which is 85 m ENE of the WAA. The dilution factor from the WAA source at the air tritium sampler, based on meteorological input from 1999 - 2003 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 source term at the WAA can be back-calculated. The known uncertainties on the Building 331 stack releases of HT and HTO have also been summed and contribute to the overall uncertainty on the WAA source terms. Overall uncertainty for the years 1953 to 1990 approaches or exceeds 80%. It has been assumed that, for the first two years of operation of the facility, the source term of the WAA was only 50% that calculated above because accumulation of wastes would have taken some time.

Records of what has been stored in the Building 612 yard may exist, but they won't contain estimates of how much tritium was off-gassed from containers annually. It is reasonable to expect that the quantity of wastes stored in the Building 612 yard were and are related to the releases of tritium from the stacks, because historically the Tritium Facility dominated releases from the LLNL site. To arrive at the estimated releases shown in Table 11, known releases from 1993 to 1997 (years when little tritium was handled at Building 612 that did not come from onsite) were correlated with known activities in the Building 612 Yard. The estimated source term for the Building 612 yard (estimate obtained by back-calculating from the mean annual air tritium concentration at the Building 624 air tritium sampler) for those years was about 4% of the HTO released from the Tritium Facility 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 with 1%, 4% and 6% of the HTO released from the Tritium Facility is proposed for the Building 612 Yard. On top of this uncertainty is the uncertainty due to the annual Building 331 source term. This additional uncertainty was accounted for by multiplying the source term uncertainty 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 the table (and used as input) as normal with ± one standard deviation.

Derivation of annual tritium release rates and uncertainty from the Building 331 WAA and the Building 612 Yard estimated from ambient air monitoring (Table 12)

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, respectively, to the present, as part of NESHAPs compliance, air tritium samplers were placed near the Building 331 WAA and in the Building 612 Yard. These samplers provide a biweekly record of the ambient air tritium concentrations at the locations; obviously, some of the tritium in air at those locations is due to other sources at LLNL, but primarily (see tests of CAP88-PC in the NESHAPs reports) these monitors provide a means by which to assess the tritium released from tritium-contaminated wastes stored nearby. Annual estimates of off-gassed tritium[14] can be obtained based on back-calculating from the observed air concentration to the source that would have created it. Uncertainty on the back-calculation must account for uncertainty on the silica gel correction factor (1.6 ± 15%), the dispersion model, the flow through the sampler (± 14%), and the laboratory analysis of tritium concentrations in the extracted water.

Uncertainty on the dispersion model has been assumed to be ± 30% (i.e., a factor of 4 with 95% confidence) for samples close to an area source. Uncertainty on the laboratory analyses was taken from laboratory data sheets for each year. The lowest average uncertainty was for the Building 331 samples in 1998 (0.6%); the highest was for the Building 331 samples in 2001 (9.7%). Total uncertainty for each year came to 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 published source terms for the Building 331 WAA are plotted against measured air concentrations,

[14] As reported in the SAER through 2001, the estimated source terms were based on the median air concentration. However, CAP88-PC predicts mean air concentrations, so a mean should be used. The source terms reported here have been recalculated based on means.

no correlation is found between source term and median or mean air concentration. Thus, estimating the source term by back-calculation may be much less uncertain (or at least have more easily quantified uncertainty) than estimates derived as they were for NESHAPs. For the dose reconstruction, therefore, source terms have been back-calculated from the mean air concentration at the air tritium sampler using the 1999 – 2003 meteorological files and applying the silica gel correction factor. Since ambient air monitoring began, the contribution of the Building 331 WAA to dose to the SW-MEI has averaged 17% of the contribution of the Building 331 stacks.

If the estimated source terms for the Building 612 Yard are plotted against the annual mean concentration of tritium in air at the Building 624 air sampler, there is an excellent correlation between the points, as is expected. Values shown in the table are 1.6 times higher than those published in NESHAPs and SAERs before 2001 because of the silica gel correction factor. Since ambient air monitoring began, the contribution of the Building 612 yard to dose to the SW-MEI has averaged 80% of the contribution of the Building 331 stacks.

Derivation of annual release rates from the Building 624 incinerator and Building 292 and uncertainty (Table 13)

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. The concentrations of tritium being incinerated had to have been quite high[15] so that analytical uncertainty would be undoubtedly very small, and uncertainty on the release for 1988 would have been essentially non-existent. 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 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.

The releases from the Building 624 incinerator will probably be shown to have zero impact on the dose to the hypothetical person at the Discovery Center. However, because reasonable confidence can be placed in the uncertainty of the source term, it will be modeled as part of the initial dose reconstruction.

As well as generic uncertainties, uncertainty on the source terms from Building 292 includes uncertainties on

[15] To achieve the minimum total activity incinerated in one day in 1988 [Hoyt 1989] with concentrations at the detection limit of the time (about 100 pCi L^{-1}), an unlikely amount of liquid (about 250 gallons) would have had to have been incinerated.

- reported source term,
- speciation,
- analysis of the tritium content in the water extracted from silica gel

An uncertainty (± 5%) has been applied to the reported source term based on experience with Building 331, because finding agreement between NESHAPs reports and SAERs does not mean there is no uncertainty on the source term. There is known uncertainty about the release of 106 Ci in 1986 because the silica gel sampler was disconnected for about a month (Myers and Silver 1986; Trent 1986). Although no releases were detected by the ion chambers, releases could have occurred, because the ion chambers were used primarily for alarming purposes and would not have detected less than alarm levels. Based on average release rates and down time, the release rate for 1986 has been revised upwards by 10% with an additional 10% uncertainty.

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[16]. 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 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 stack volume of 6.65 m^3 s^{-1}. It would seem that the stack diameter used by Trent was the outside diameter rather than the inside diameter that should have been used to calculate stack volume. In the 1987 calculation for 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 7). Trent (1986) reports a stack volume of 10.5 m^3 s^{-1} used to derive the curie release rate. If exit velocity had been calculated based on outside diameter, the exit velocity would have been 14.4 m s^{-1}; 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 7 have been averaged to obtain a mean exit velocity that represents all years with an uncertainty of ± 31%. This uncertainty has a large impact on the potential source term. For example, for 1986, if the inside stack diameter had been used in the calculation (as it should have been), the release rate would have been 58 Ci instead of the reported 106. The assumption that release rates for other years were based on the inside stack diameter may be wrong.

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 HTO, it is reasonable to assume that

[16] Exit velocities for 1979 and 1981 shown in Table 7 were derived using the stack flow and the inside diameter of the Building 292 stack from NESHAPs (0.71 m diameter).

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 source terms are 1.6 times higher than 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 is used in the uncertainty analysis. Analytical uncertainty was highest in 1986 at just over 1%.

Derivation of annual release rates of HT and HTO from the SNL Tritium Research Laboratory and uncertainty (Table 14)

Uncertainty on the source terms from the SNL TRL includes uncertainties on

- Reported source term
- speciation,
- analysis of the tritium content in the water from the bubbler system

Annual release rates from the SNL TRL were published in the SNL/CA Site Environmental Reports. No other record has been found, so an uncertainty on the reported routine annual source term of ± 5% has been applied (similar to Building 331). This 5% accounts for cases when it is unknown whether or not the evaporated tritium was included in the stack release rate. In the case of small accidents, the uncertainty on the source term has been doubled to account for uncertainty on the magnitude of the accident. In addition, there were two accidents that were large enough that they cannot reasonably be included as routine releases. One was a 200 Ci release of HTO in January 1986, and the other was an 1100 Ci release of HT in August 1987. These releases have been subtracted from the annual input from SNL, and the uncertainty about the source terms has been set at ± 20%.

An additional uncertainty arises because only one record of exit velocity could be obtained for the entire operational life of the facility, although it is unlikely the exit velocity remained the same throughout the years of operations. Given the importance of releases from the TRL to dose, it was assumed that the exit velocity varied no more than the exit velocity 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 in first four years that were speciated. 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 in the counting of the bubbler water was assumed to be similar to

that for LLNL samples of comparable magnitude. Based on this approach, an uncertainty of 5% (one standard deviation) was estimated for all years.

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[17]), off-gassing from unknown WAAs (both at LLNL and SRL), transpiration of contaminated soil water by vegetation, and releases from contaminated soils undergoing clean-up are being ignored. Some operations from the earliest days of LLNL (pre Building 231) are also being ignored due to lack of any idea of what the releases might have been and their exact time frame. Otsuki (2004) recalls work with liquefying tritium being conducted in B232 (then B147), perhaps concurrent with work in Building 231. His very first tritium research before Building 231 became available was conducted in B119 (then B161), in the hospital wing, in a women's restroom.

Examples of missing sources from recent years can be found be perusing NESHAPs reports looking for "H-3". Some of these sources are known to be so small that any effect at VIS 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 VIS, because it is essentially impossible to characterize them. Sources conspicuous by their absence should be detected at least after 1974 when predicted air concentrations can be compared with observed air concentrations at VIS. If this occurs, the treatment of the missing sources will be reconsidered.

Dilution factors (χ/Q) documented in Tables 15-19

CAP88-PC was chosen as the model with which to calculate dilution factors (and, from them, tritium in air concentrations) at VIS. 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 at the Livermore Site, which is flat, or at VIS, which is not particularly 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

[17] 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.

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 2004a). Although the dose reconstruction will attempt to predict accurate doses with a 95% confidence interval, if there is any bias, it should be slightly conservative.

The wind file used in CAP88-PC to calculate these dilution factors was created from four years (1999 – 2003) worth of hourly data from the LLNL meteorological station. LLNL data from earlier years was rejected; instrumentation has been improving over the years, and these data are the most reliable (Bowen 2004). Quite large differences in dilution factors at any one location are often seen when different annual files or files representing 4- or 5-years worth of data are used as input. These differences are the result of having statistically too few data points in some of the wind speed/stability class categories calculated in CAP88-PC. The large uncertainty on dispersion accounts for these differences.

Based on experience with CAP88-PC, a 25% uncertainty on dispersion has been applied to the Tritium Facility Stacks and a 30% uncertainty has been applied to all other sources. Uncertainty on dispersion is usually lognormal, so these percentages represent factors of about 3 to 4 uncertainty.

Also contributing to the uncertainty of the dilution factor is the effect of the uncertainty of the exit velocity on the dilution factor. For each known exit velocity, χ/Q has been calculated. The assumptions about deriving exit velocity and uncertainty for years when the exit velocity was not calibrated have been described under the Annual Source Term Section; however, to derive the uncertainty on χ/Q, instead of manipulating the exit velocity, the χ/Q calculated by CAP88-PC for a particular exit velocity is manipulated. The effect on χ/Q of uncertainty on the calibrated exit velocity has also been taken into account; a 5% uncertainty on calibrated exit velocity (Biermann 2004) transforms into at most 2.5% uncertainty on χ/Q; the uncertainty is usually much less depending upon the source.

Dilution factors are distributed lognormally. The overall uncertainty on each 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, however, that the difference between the overall distributions being prepared as if all contributing uncertainty distributions were normal compared to being prepared as if they were lognormal (using Crystal Ball sampling each distribution as lognormal) was very small and not worth the considerable extra effort that would be involved. Therefore, the distributions have been prepared as value ± uncertainty that will be entered into Crystal Ball as lognormal.

Dilution factors were calculated for the position of the VIS sampler, located near the Discovery Center, in a sector relative to the closest adjacent sector at known distances and bearings from the sources as determined by GPS. The following equation was used:

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

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 half-width in degrees

Tables 15 and 16 show the estimated dilution factors (χ/Q in s m^{-3}) for all sources with uncertainty for a lognormal distribution. The uncertainty on dispersion dominates the uncertainty on χ/Q. Uncertainty is such that, for all sources except the LLNL Tritium Facility, just one χ/Q with high uncertainty has been derived for all years of operation

<u>Derivation of dilution factors (χ/Q in s m^{-3}) with uncertainty for modeled sources other than the LLNL Tritium Facility (Table 15).</u>

Building 231
Work could have occurred in what is now the vault or in a wing adjacent (Room 1128). The differences in the recent data available for stacks from both locations are summarized in the footnote under Table 7. The uncertainties due to different stack parameters result in 24% uncertainty on the dilution factor. To account for slightly different distances from the stacks to VIS and the fact that the only known exit velocities are forty years removed from the time of operations, an additional 20% uncertainty has been included. Overall uncertainty on the dilution factor is 43%.

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 is for NESHAPs compliance (Table 7); 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 10^{-6} s m^{-3} for Building 212 room air with 2.49 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 χ/Q. 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 % on a lognormal distribution.

Building 331 WAA and Building 612 yard
The uncertainty from both these sources is primarily from dispersion 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 both sources.

Building 212 stack
The dilution factor at VIS from releases at Building 212 is highly insensitive to change in exit velocity. As explained under Table 9, 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 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 χ/Q 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 7), so the overall uncertainty is basically the 30% from dispersion, even though there are 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 has been used to calculate χ/Q, at VIS with an uncertainty of ± 11.9%. The rationale for this is discussed in detail under Table 12. Using all known exit velocities creates a "global" exit velocity with associated uncertainty and precludes the need for any χ/Q uncertainty based on the 5% uncertainty on the calibrated exit velocity. Overall uncertainty on χ/Q is 32.3%.

SNL stack
The dilution factor and uncertainty for SNL are the same for all years because of lack of knowledge. Assuming similarities in stack maintenance between LLNL's Tritium Facility and the SNL TRL, the exit velocity might have varied by ± 5%. This results in uncertainty on the dilution factor of ± 4.2%; uncertainty based on problems with calibration adds an additional 3.1% uncertainty. These uncertainties add essentially nothing to the overall dispersion uncertainty of 30%.

<u>Derivation of dilution factors (χ/Q in s m^{-3}) with uncertainty (for Stack 1 and Stack 2 of the LLNL Tritium Facility (Table 16).</u>

The exit velocities for Stack 1 vary less over time than those of Stack 2, so the effect on the uncertainty in χ/Q is less. The highest χ/Q uncertainty 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 exit velocity is about 1.5% for both stacks. The uncertainty (±25%) on the dispersion model dominates the overall uncertainty.

Other annual input documented in Tables 17 - 19

Annual mean observed concentrations of tritium in air moisture (Bq L^{-1}) and air (Bq m^{-3}) at VIS with estimated uncertainty (Table 17)

Observed volumetric air tritium concentrations at VIS can be used in DCART as an alternative, more accurate estimation of dose, with less uncertainty, than can be obtained through modeling (with any dispersion model). As well, the observed annual mean concentrations of tritium in air moisture at VIS can be compared with air moisture concentrations predicted by DCART to perhaps prompt reexamination and reduction of the uncertainty on input parameters, particularly source terms.

To monitor tritium in 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 in pCi m^{-3} is calculated. Reported concentrations have been converted in this report to Bq L^{-1} and Bq m^{-3} to match DCART's output.

Ambient air sampling for tritium has taken place since 1973; 1974 was the first complete year of sampling. Sampling was on a weekly basis in 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 preceding Table 8).

Mean annual concentrations in extracted air moisture and in volumetric air that had passed through the silica gel sampler (see table) 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 includes that from missing data (uncollected samples or those removed, such as outliers, results of accidental releases, and those below the detection limit), silica gel correction, laboratory analysis, and, for the volumetric results, the flow through the sampler.

Uncertainty on 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 each missing or removed sample, the uncertainty is assumed to be ± 1/# samples in a year in percent. For weeks of known accidental releases that were not detected at VIS, the uncertainty on the observed values has been increased.

All concentrations 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. If possible overestimation of air concentrations at VIS results in under-predictions by dispersion modeling, the use of such a conservative correction factor will have to be reexamined.

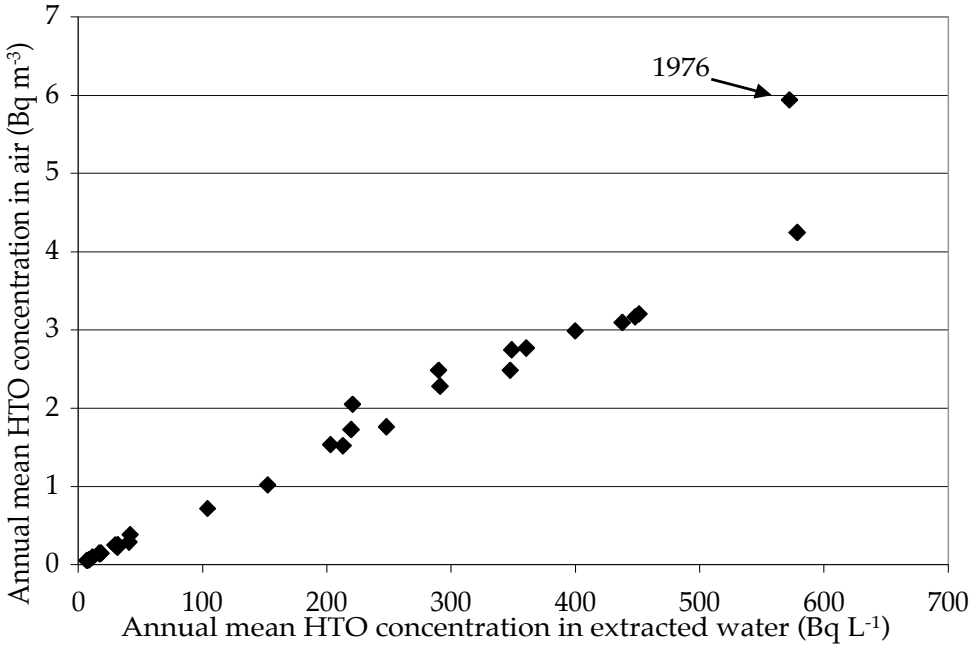
Analytical error was estimated by selecting analytical results close to the calculated annual mean and selecting a representative error. Analytical error (1 standard deviation) ranged from 0.011 in 1977 to 0.175 in 2001.
For concentrations in air volume, once flow meters were used, starting in 2000, the uncertainty on the flow drops to ± 3.5%, based on manufacturer specifications.

The contribution from bomb fallout and cosmogenic tritium has been subtracted from observed air tritium concentrations, and associated uncertainty has been taken into account (Peterson 2004b), because the model uses source terms to predict air concentrations that obviously cannot include background tritium. The uncertainty on background tritium concentrations is due to assumptions about the ratio (0.8 ± 5%) between concentrations of tritium in rain and those in air moisture and about the long-term absolute humidity at LLNL (0.00786 kg m^{-3} ± 6.5%) 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 were low, the estimated background concentrations were about 6-7% of the observed air concentrations.

When observed concentrations are not expected at an LLNL sampling location, an effort has always been made to explain them, although not always successfully. 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. Measurements of tritium in air were corroborated by measurements of tritium in vegetation. No other local source of tritium was identified. CAP88-PC under-predicted all air concentrations at all sampler

locations except VIS and one other (Peterson 2004b) for 1986 and 1987, so perhaps under-predictions due to anomalies are less likely to occur at VIS than at other sampling locations.

When concentrations in air moisture are plotted against concentrations in air volume for each year (see figure below), a nearly linear response is expected. Perfect linearity is not expected because of variations in mean absolute humidity each year. However, by plotting the observed data, an apparent error in the volumetric concentration for 1976 can be seen. The value that best fits the expected linear relationship between the concentrations is 4.25 Bq m^{-3}.



<u>Annual absolute (kg m^{-3}) and relative humidity with uncertainty (Table 18)</u>

Data on relative and absolute humidity have only been gathered at the LLNL meteorological station since 1997, whereas tritium has been sampled using silica gel since 1974. The weight of the water gathered 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 this table were obtained from averaging estimates of absolute humidity for all Livermore site air tritium samples for each year. The values for relative humidity shown are ones that are reasonably trusted from data gathered at the LLNL meteorological station (Bowen 2003). Eberhart (1999) observed that silica gel may not necessarily be a good method by which to estimate absolute humidity under some sampling conditions. 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. The uncertainty on the mean of all years of absolute humidity is ± 5%. This has been applied to the uncertainty of each annual mean as well. Uncertainty on relative humidity is greater (± 10%). Uncertainty on the mean of the years shown is only 7%, but there are so few data that the uncertainty has been increased. Uncertainty on both absolute and relative humidity is normally distributed.

Analysis of concentrations in the LLNL swimming pool (Table 19)

In DCART (Peterson 2004a), tritium concentrations in the LLNL swimming pool can be estimated when unavailable from the empirical relationship between air moisture tritium concentrations at VIS, air moisture tritium concentrations at the air tritium sampler (POOL) next to the pool, and pool water concentrations. For years with measured concentrations, the mean annual pool water concentrations can be directly input to DCART. In Table 19, the means and uncertainties include those samples less than the lower limit of detection (LLD – at the LLD concentration), although data for 2001 through 2003 have been excluded because all samples were below the LLD. In most cases, removing the samples below the LLD increases 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. In July 2004, the pool was declared structurally unsafe and drained. 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 changes in emissions, the fluctuation 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.

ACKNOWLEDGEMENTS

Much of this work was a team effort. I wish to thank Joe Shinn, Nick Bertoldo, Paula Tate, and Gretchen Gallegos of the original team that started this dose reconstruction. I thank Kris Surano for having the vision that made this work possible. Special thanks go to Gretchen Gallegos for continual help, support and inspiration, as well as reviews of this report. Special thanks also go to Dave Myers for ferreting out missing information, for support of this project, and for review of this report. I thank Kent Wilson for explaining stack sampling. I thank other reviewers: Bert Heffner and Art Biermann. Thanks go to Paris Althouse for providing the GPS data. I also wish to thank my colleague Phil Davis of Chalk River Laboratories, Canada, for providing answers to innumerable questions about the preparation of the uncertainty analysis.

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 by pointing these out. Quite probably, this summary contains its own mistakes, although every effort has been made to ensure that this is not the case.

APPENDIX A

Errata

Corrections and changes to "Historical Doses from Tritiated Water and Tritiated Hydrogen Gas Released to the Atmosphere from Lawrence Livermore National Laboratory (LLNL). Part 1. Description of Tritium Dose Model (DCART) for Chronic Releases from LLNL.

- Page 8. Units for wet deposition (Ω) are Bq m^{-3} y. Units for duration of rainfall when plume is present (ΩT) are s y^{-1}.

- General clarification: All calculated concentrations and doses are mean annual concentrations and doses.

- Page 39, Table A1. The generic value for the fraction of year it rains should be 0.018 ± 0.0060. Note, however, that this value has been replaced by the equation in Appendix A of this report.

- Page 47, Table A9. The recommended value for humidity at skin/water temperature is 0.02724; the range of values for the triangular distribution is 0.02240 – 0.02724 – 0.02878

APPENDIX B

Parameters for precipitation pathways

As explained in Part 1 (Peterson 2004a), soil concentrations are calibrated from air concentrations, and the precipitation pathway has been disabled in DCART because it is not needed to calculate concentrations in food or ingestion dose. Nevertheless, the precipitation equations can be used to determine concentrations in rainfall. The following tables describe site-specific LLNL values for the various parameters and a data set of tritium in rain collected at the Discovery Center monitoring location, VIS.

Annual rainfall at LLNL and uncertainty (Table B1)

The uncertainty on the rainfall measured is ± 10% (normal distribution) to apply the value measured at the western edge of the site 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[18]. The uncertainty distribution on these numbers is assumed triangular, with the estimate 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:

Fraction of time it rains in a year = 0.0569 x annual rainfall in m - 0.000195

Note that this equation replaces the generic fraction (0.018 ± 0.0060) from Table A1, Part 1 of this LLNL Dose Reconstruction (Peterson 2004a). Because this equation was derived from fractions that represent the maximum time it could have rained, but 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.

[18] 1997 - 0.0131; 1998 – 0.0291; 1999 – 0.0147; 2000 – 0.0187; 2001 – 0.0202; 2002 – 0.0135; 2003 – 0.0132.

Other input for the precipitation model in DCART (Tables B2 and B3)

Data were prepared from the LLNL 15-minute meteorological data for the years 1997 through 2003:

- 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
- Wind directions from 281.25 to 303.75 degrees (towards ESE)
    - Building 292

The fractions of time that the wind blew from a particular source to the Discovery Center (sampling location VIS) are shown in Table B2 for each year measurements are available. Frequency was obtained by counting the 15-minute segments in which rain occurred 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.

Other input needed by DCART includes the washout coefficient and the wind speed towards the receptor when raining (Table B3). Wind speeds were calculated for each year of data (1997 – 2003). Because, for some sources, the wind blows very infrequently towards VIS, data for each source for all years were averaged to obtain a more representative sample size. The uncertainty on the seven-year mean is the standard deviation of the annual mean wind speeds. This uncertainty includes 5% uncertainty on wind speed measurements plus the uncertainties due to 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 different heights than the release. Washout coefficients, with lognormal distributions are based on Belot (1998), who calculates washout coefficients for 1 mm h^{-1} rains at various release heights and various distances from the source. Washout coefficients from Belot for 1 mm h^{-1} rains were 7.25×10^{-5} for the Building 331 stacks, 8.0 10^{-5} for the Building 331 WAA and Building 212, 9.0 10^{-5} for the Building 612 yard, 7.6 10^{-5} for Building 292, and 6.5 10^{-5} for SNL. Uncertainty for a lognormal distribution on these numbers is about ± 20%. The higher the effective release height, the lower the washout coefficient. In general terms, beyond close in, washout coefficient decreases with distance. A further adjustment is made for higher rainfall rates. The washout coefficient can be considered as 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 precipitation intensity.

The washout coefficients (Table B3) representing the geometric mean of seven-years data for each source were calculated using a mean rainfall rate for the

LLNL site. Rainfall rates were estimated by dividing the total year's rainfall by the 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 global rainfall rate with a geometric mean (GM) of 2.39 and a geometric standard deviation (GSD) of 1.34 has been estimated. 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.

Observed concentrations of tritium in rainfall at the Discovery Center (Table B3)

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 has been collected. Nevertheless, the data can be used to test models in a limited manner.

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 move quite rapidly from a volume with high concentration (the water in the bucket) to a volume of low concentration (air moisture) and vice versa, so 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 samples collected, the more representative 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. Nevertheless, model predictions can be compared with the individual observations.

Mean tritium measurements in rainfall with fallout background subtracted for the sampling location VIS are summarized in Table B4.

Table B1. Rainfall in m at LLNL and one standard deviation (σ) uncertainty

Year	Rainfall (m)	± 1 σ	Year	Rainfall (m)	± 1 σ
1952*	0.540	0.0763	1978	0.403	0.040
1953*	0.198	0.0280	1979	0.377	0.0377
1954*	0.331	0.0468	1980	0.298	0.0298
1955*	0.434	0.0613	1981	0.404	0.0404
1956*	0.272	0.0385	1982	0.531	0.0530
1957*	0.317	0.0448	1983	0.853	0.0853
1958*	0.467	0.0661	1984	0.274	0.0274
1959	0.235	0.0235	1985	0.256	0.0256
1960	0.308	0.0308	1986*	0.334	0.0473
1961	0.246	0.0246	1987*	0.315	0.0446
1962	0.345	0.0345	1988*	0.208	0.0294
1963	0.391	0.0391	1989*	0.243	0.0344
1964	0.298	0.0298	1990	0.230	0.0230
1965	0.375	0.0375	1991	0.277	0.0277
1966	0.228	0.0228	1992	0.305	0.0305
1967	0.433	0.0133	1993	0.402	0.0402
1968	0.345	0.0345	1994	0.303	0.0303
1969	0.401	0.0401	1995	0.541	0.0541
1970	0.480	0.0480	1996	0.501	0.0501
1971	0.237	0.0237	1997	0.250	0.0250
1972	0.303	0.0303	1998	0.523	0.0523
1973	0.531	0.0531	1999	0.245	0.0245
1974	0.245	0.0245	2000	0.295	0.0295
1975	0.330	0.0330	2001	0.339	0.0339
1976	0.167	0.0167	2002	0.271	0.0271
1977	0.200	0.0200	2003	0.244	0.0244

* Uncertainty for these years is ± 14.1%, instead of 10%, because no rainfall measurements were made on site.

Table B2. Annual and long-term (1997 – 2003) mean values ± uncertainty (1) for the fraction of time the wind blows towards the Discovery Center from tritium sources when raining, Uncertainty on the annual frequencies is ± 10%.

	LLNL TF Stacks	B331 WAA; B212; B231	B612 Yard; B624 Incinerator	B292 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 B3. Mean wind speed during rain and washout coefficients with 1 uncertainty to apply to all years for each tritium source at LLNL. Values are specific to a receptor at the Discovery Center. Wind speed has a normal distribution; washout coefficient has a lognormal distribution.

	Mean wind speed (m s^{-1}) during rain	Washout coefficient (s^{-1}) GM	GSD
LLNL TF Stack	4.23 ± 0.446	1.19 10^{-4}	1.52
B331 WAA; B212; B231	3.18 ± 0.301	1.31 10^{-4}	1.52
B612; B624 incinerator	3.76 ± 0.302	1.47 10^{-4}	1.52
B292 Stack	2.13 ± 0.279	1.25 10^{-4}	1.52
SNL TRL	4.86 ± 0.439	1.07 10^{-4}	1.52

Table B4. Number of rainfall samples collected each month for all months except July and mean annual tritium concentration (fallout background subtracted) of rain sampled at the Discovery Center, LLNL. Non-detects are shown as shaded or as (n). Uncertainty on the means for 1991 and 1992 is ± 30%.

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

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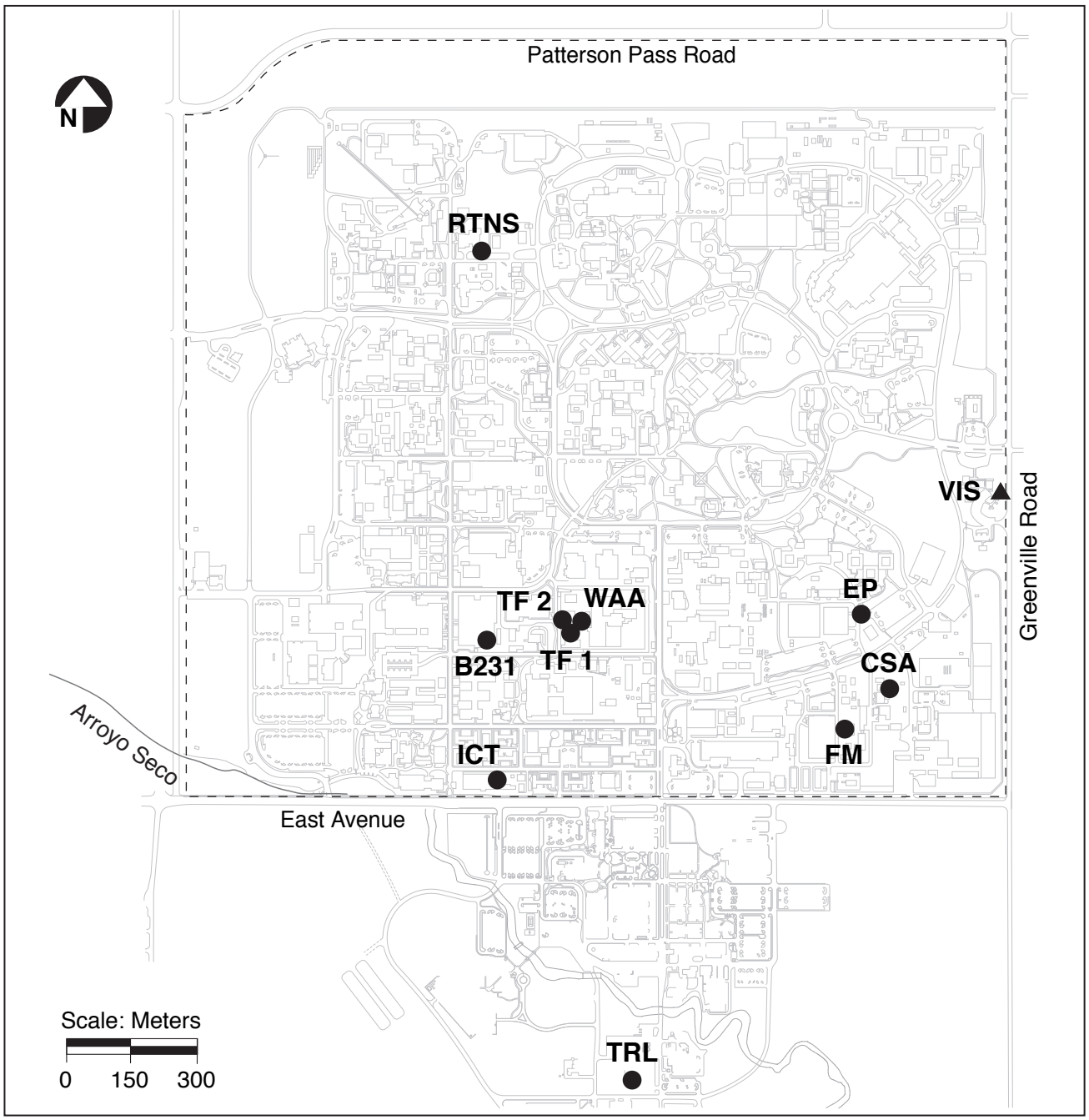

Figure 1. Sources of tritium (●) at LLNL and SNL relative to the location of the maximally exposed individual (VIS; ▲). Modeled sources include RTNS (Building 292), TF 1 and TF 2 (the south and north stacks of the Tritium Facility, Building 331), WAA (the diffuse source at Building 331), B231 (previously numbered Building 102, where tritium work took place before 1958), ICT and room air (Building 212), TRL (the Tritium Research Laboratory, SNL), and CSA (Building 612 yard diffuse source and Building 624 incinerator). Sources not included in the dose modeling include the solar evaporation pans (EP) and the Building 514 Tank Farm (FM).

Table 1. Source estimates from memos (1956 – 1979) of total tritium (HT and HTO from all facilities) released from Lawrence Livermore National Laboratory. A "+" indicates that more emissions are expected because some data for that year may be missing. A "-" indicates the probable number is lower. B172 was the Tritium Facility before it was renamed B331.

Year	Ci/y	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); no memos for 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); no memo for 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); no memos for second and third quarters
1959	3060+	"gas" released 2/16[10] (1600), 5/12[11] (1100), 5/25[11] (360); additional negligible releases third quarter[12] and fourth quarter[13]
1960	3100+	"gas" released 2/23[14] (2000), 11/19[15] (1100); negligible releases[16,17,18]
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); negligible release of ~70 Ci[20],[21] reporting changes in mid-year – no mention of gas. 10 Ci came from the mass spectrometer in B172 and 600 from "controlled releases" from B172; 200 Ci came from the 90-inch cyclotron.[22]
1962	4430-	All "controlled releases" from B172. 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 B172. First quarter[27] (1100); second quarter[28] (2000); third quarter[29] (2200); fourth quarter[30] (November – 700)
1964	12500	Reporting changes again – B172 not mentioned - only total tritium released from LLNL. 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)
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)

Table 1 continued.

Year	Ci/y	Comments (numbers in parentheses are in Ci)
1970	291541?	January[52] (440); February[52] (1680); March[52] (260); April[53] (555); May[53] (825); June[53] (360); July[54] (160); August[54] (24); September[54] (30); October[54] (62); November[54] (37); December[54] (30); 287,000 Ci[55] accidentally released August 6; third quarter[55] (230); fourth quarter[56] (191)
1971	2710	First quarter[57] (560); second quarter[58] (990); third quarter[59] (790); fourth quarter[60] (370)
1972	1350	First quarter[61] (330); second quarter[62] (240); third quarter[63] (510); fourth quarter[64] (270)
1973	2560	First quarter[65] (480); second quarter[66] (1140); third quarter[66] (470); fourth quarter [67](470)
1974	1690	First quarter[68] (460); second quarter[69] (610); third quarter[70] (310); fourth quarter[71] (310)
1975	2440	First quarter[72] (510); second quarter[73] (740); third quarter[74] (410); fourth quarter[75] (880)
1976	3700	First quarter[76] (570 T); second quarter[77] (~365 HTO, ~445 HT); third quarter[78] (~742 HTO, ~685 HT); fourth quarter[79] (~366 HTO; 527 HT)
1977	4700	First quarter[80] (~135 HTO; ~429 HT); second quarter[81] (~650 HTO; ~452 HT); third quarter[82] (~571 HTO; ~697 HT); fourth quarter[83] (~600 HTO, 1166 HT)
1978	5361	First quarter[84] (~473 HTO; ~879 HT); second quarter[85] (~539 HTO, ~776 HT); third quarter[86] (~390 HTO, ~867 HT); fourth quarter[87] (~560 HTO, ~877HT)
1979	3893+	First quarter[88] (~488 HTO, ~1387 HT); second[89] quarter (~614 HTO, ~1046 HT); third quarter[90] (~211 HTO, ~147 HT); no memo for fourth quarter

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Table 2. Source terms for LLNL Tritium Facility releases (total tritium {HT + HTO}, both routine and accidental).

Year	Ci (from memos)	Ci from (Talk to Director's Office[1])	Ci (from LLNL Site Annual Environmental Reports)
1953	2000 ± 50%[2]	2000	
1954	2000 ± 50%[2]	2000	
1955	2000 ± 50%[2]	2000	
1956	3000 ± 50%[2]	3519	
1957	3000 ± 50%[2]	11356	
1958	1000 ± 50%[2]	5940	
1959	1000 ± 50%[2]	3380	
1960	1000 ± 50%[2]	3180	
1961	1000 ± 50%[2]	1100	
1962	3000 ± 50%[2]	4130	
1963	3000 ± 50%[2]	6000	
1964	26000[2]	26500	
1965	360000[2]	363350	
1966	14000[2]	17050	
1967	6600[2]	6600	
1968	6800[2]	6750	
1969	7300[2]	7300	
1970	290000[2]	291480	
1971	1400[2]	1400	
1972		1350	
1973		2960	2510
1974	1350[3]	1690	1300
1975	1353[4]	2540	2167
1976	2819[5]; 2822[6]	3675	2828
1977	2956[6]; 3145[7]	3150	3150
1978	4232[6]; 4243[8]; 4245[9]	5250	4245

Table 2 continued.

Year	Ci (from memos)	Ci from (Talk to Director's Office[1])	Ci (from LLNL Site Annual Environmental Reports)
1979	4069[10]; 4070[8]	4100	4069
1980	2217[11]; 2219[12]	2179	2218
1981	2552[13,14]	2550	2552
1982	1899[15]; 1914[16]	1914	1914
1983	3024[17]	3024	3024
1984	7200[18]	7200	7200
1985	1989[19]	1989	1989
1986	1128[20,21]	1128	1128
1987	2633.8[22]	2634	2633.8
1988	3977.5[23]		3997.5
1989	2949.5[24]		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

REFERENCES (Table 2)

[1] Souers, C. Overheads from a talk "Tritium Releases from B331" prepared for the Director's Office, June 1988.
[2] Olsen, J.L. "Lawrence Livermore Laboratory Radioactivity Inventory Data", Subject of letter to D.B. Campbell, U.S.AEC, San Francisco, November 9, 1973
[3] Powell, T.J. Building 331 – Health Physics Audit for 1975.
[4] From a hand-written summary (Build Stack Effluent Report) of monthly gaseous releases from B194, B281, B212, and B313 for 1975; particulate releases also.
[5] 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.
[6] Extracted from hand-drawn graph by S. Homann, "Building 331 Monthly Tritium Release for 1976"
[7] 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.
[8] Homman, S. Cumulative graphs of total emissions.
[9] 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.
[10] 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.
[11] 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.
[12] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1980, memo to distribution, January 15, 1981
[13] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1981, memo to distribution February 18, 1982.
[14] 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.
[15]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.
[16] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1982, memo to distribution, February 2, 1983.
[17] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1983, memo to distribution, January 17, 1984.
[18] King. W.C., Radiation and Environmental Safety Division, Hazards Control Department, memo to distribution January 22, 1985.
[19] Printed worksheet showing weekly sample values and figure.
[20] Gordon, L.E.. Memo to D. Brekke, January 30, 1987
[21] Gordon, L.E. Memo to A.L. Buerer, March 16, 1987.
[22] Mansfield, W.G. Memo to G. Morris, January 19, 1988
[23] Biermann, A. Memo to D. Brekke, February 21, 1989
[24] Mansfield, G. Memo to M. Singleton, April 13, 1990

Table 3. Reported and potential 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, and Building 331 waste accumulation area. All unmarked numbers were obtained from SAERs or NESHAPs reports; other sources are referenced in superscripts. Species of tritium are noted.

Year	B212[1,SAERs (1974 on),2] Ci HT, unless indicated	B292[3,4] Ci HTO	B624 incinerator Ci HTO	Laser complex Ci HT	B612 yard Ci HTO	B331 WAA Ci HTO
1954	75 ± 50%					
1955	75 ± 50%					
1956	75 ± 50%					
1957	75 ± 50%					
1958	75 ± 50%					See Table 11
1959	75 ± 50%					"
1960	75 ± 50%					"
1961	75 ± 50%					"
1962	75 ± 50%					"
1963	75 ± 50%					"
1964	75 ± 50%					"
1965	10 ± 50%				See Table 11	"
1966	10 ± 50%				"	"
1967	10 ± 50%				"	"
1968	240				"	"
1969	140				"	"
1970	65				"	"
1971	260				"	"
1972	52				"	"
1973	missing				"	"
1974	560				"	"
1975	1217[5]				"	"
1976	1173[6]				"	"
1977	2057[7]; 2060		See Table 13		"	"
1978	1117[8]		"		"	"
1979	436[9]	11.7[9]; 12	"		"	"
1980	70[10]	17; 17.3[10]	"		"	"
1981	23; 23.4[11]	44; 44.2	"		"	"

Table 3 continued.

Year	B212[1,SAER,2] Ci HT, unless indicated	B292[3,4] Ci HTO	B624 incinerator Ci HTO	Laser complex Ci HT	B612 yard Ci HTO	B331 WAA Ci HTO
1982	34[12]; 44	56; 56.2[12]	See Table 13		See Table 11	See Table 11
1983	140	81	See Table 13		"	"
1984	11	143	See Table 13		"	"
1985	5	210[13]	See Table 13		"	"
1986	13; 2 (HTO)[14]	106[15,16,17]	5.1; 3.75[18]		"	"
1987	34 ; 4 (HTO)[19]	78.5	0.22; 0.213[20]		"	"
1988		5.29	0.343[21]		"	"
1989		2.4[22]		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

REFERENCES AND ANNOTATIONS (Table 3)

[1] Olsen, J.L. "Lawrence Livermore Laboratory Radioactivity Data", Subject of letter to D.B. Campbell, U.S. AEC, San Francisco, November 9, 1973.
[2] 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).
[3] Releases were monitored using silica gel; releases were estimated to be about 98% HTO and occurred during rough pumping.
[4] 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.
[5] 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.
[6] 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.
[7] 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.
[8] 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.
[9]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 1978 Stack Reports in LLNL Radiological Air Effluent Records.
[10]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 1978 Stack Reports in LLNL Radiological Air Effluent Records.
[11] 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 1978 Stack Reports in LLNL Radiological Air Effluent Records.
[12] 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 1978 Stack Reports in LLNL Radiological Air Effluent Records. B212 showed sum to be 44, but numbers only sum to 34.
[13] Trent, M. Memo to K. Griggs January 21, 1986
[14] Homann, S. Memo to D. Brekke February 5, 1987. Subject: Annual Stack release from Buildings 194 and 212
[15] Trent, M. Memo to D. Brekke February 5, 1987. Subject: Stack Releases from Building 292 (RTNS-II) and Building 865 (ATA).
[16] Trent. M. Memo to Dave Myers and Bill Silver. Subject: Response to the Health Physics Review of Building 292. September 24, 1986.
[17] 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 8 and 9.
[18] Radiation Safety Program, Hazardous Waste Management, 1986 Annual Facility Report
[19] Homann, S.G. Memo to A. Biermann January 7, 1988. Subject: Annual Stack release from Buildings 194 and 212
[20]Radiation Safety Program, Hazardous Waste Management, 1987 Annual Facility Report
[21] Hoyt, Dan. Memo to Susi Jackson. Subject: Radionuclides incinerated at the HWM incinerator for calendar year 1988. HWM 89-161; February 3, 1989.
[22] Myers, D. Memo to K. Lamson February 5, 1990

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

Year	Ci	HT or HTO	Facility	Comments
1954	? + 3000	HT + HTO	231	1 cc of T_2O lost from cold trap and T_2 October 13, 1954[1]
1956	3000	HT	231	Released August 28, 1956[2,3]
1957	11000	HT	231	6000 Ci May 22, 1957 and 5000 Ci June 12, 1957[4,3]
1958	4900	HT	231?; 331	3000 Ci March 31, [5,6,3] and 1900 Ci November 5, 1958[7,3]
1959	2700	HT	331; Stack 1	1600 Ci February 16, 1959[8,3] and 1100 Ci May 12, 1959,[9,3]
1960	3100	HT	331; Stack 1	2000 Ci May 2, 1960[10,3] and 1100 Ci November 19, 1960,[11,3]
1961	200	HT	212	From the 90 inch cyclotron[12] for the period 10/25 to 10/28/61
1964	24000	HT	331	No record except Souers (1998)[3]
1965	360000	HT	331; Stack 1	2 minute release at 3:27 pm January 20, 1965; reported to AEC as 360000 Ci[13]estimate based on integrations of the chart trace range from 207,000[14] to 300,000 Ci, but 350000 published[15]
1966	11000	HT	331; Stack 1	April 7, 1966[16]; 14,000[3]
1969	800	HT	331	Experimental release October 27, 1969, 10:30 – 11:30 am[17]
1970	287000[18]	HT	331; Stack 1	30 minute release at 6:14 August 6, 1970, 289000 Ci[19]
1971	105 + 135	HT + HTO	331; Stack 1	April 12, 1971 at 9:05; environmental monitoring followed[20]
1975	340	HTO ?	331; Stack 2	Reported by Souers (1998)[3]; 460.7 Ci HTO released 5-12 November[21]
1976	175	HT ?	331; Stack 2	Reported by Souers (1998)[3]; 146.8 Ci HT released 3-10 March[21]
1981	130	HTO	331; Stack 1	Possible 130 Ci release in November[22]; possibly 142.8 Ci HTO released 2-9 November or possibly 133.1 Ci HTO released 16-23 November[21]
	450	HTO	331 Stack 1	December 12 or 13, 450 Ci measured by Ostlund Monitor but undetected by ion chambers[22]; environmental study carried out[23]; 477.8 Ci HTO released 7-14 December[21]
1983	220	HT ?	331; Stack 2	No record except Souers (1998)[3]; possibly 317 Ci HT released 7-14 March or 379 Ci HT released 28 November – 5 December.
1984	5200	HT	331; Stack 2	June 8, 1984 at 8 am; 6000 Ci estimated from pressure drop; 5200 best estimate based on electronic integrator (1400 ± 300 and 4300 ± 800[24]; 5000 Ci reported officially[25]; 5600 ± 500 Ci also reported[26]; reported in 1984 SAER as 5000 Ci. HT; 5155 Ci[3]

Table 4 continued

Year	Ci	HT or HTO	Year	Ci
1985	1034	HT	331; Stack 2	January 24, 1985[27]; reported in 1985 SAER as 1000 Ci HT; 1000 Ci[3]; 920 Ci from ion chamber[27]; 935 Ci HT released 22-28 January[21]
1986	125	HT	331 Stack 1	December 15, 1986[28]; 135 Ci[3]; 149.8 Ci HT released 15-22 December[21]
1987	198	HT	331; Stack 2	April 14, 1987[28]; 185 Ci[3]; 198.9 Ci HT released 14-20 April[21]
	575	HT	331	No record except Failor (1999)[28]
1988	145	HT	331; Stack 2	January 19, 1988[28]; 121.7 Ci released 11-19 January[21]
	138	HTO	331; Stack 2	January 25, 1988[28]; 138.6 Ci HTO released 25 January – 2 February[21]
	653	HT	331; Stack 2	May 15, 1988[28]; 651.6 Ci HT released 16-18 May[21]
	120	HTO	331; Stack 2	August 1, 1988[28]; 120.4 Ci HTO released 2-10 August[21]
1989	112	HT	331; Stack 2	February 28, 1989[28]
	290 + 7	HT + HTO	331; Stack 2	August 25, 1989[29] between 10 and 11 am reported as 329 Ci HT in 1989 SAER; reported to National Response Center[30]; 329 Ci HT and 6.73 Ci HTO released 22-28 August[21]
	112	HTO	331; Stack 1	October 31, 1989[28]; 112 Ci HTO released 31 October – November 6[21]
1991	~112	HT	331; Stack 1	April 2, 1991 at 3:50 pm[31]. Reported in SAER as 144 Ci.[32]; 139 Ci HT released 1-3 April[21]

REFERENCES (Table 4):

[1] Bldg. Monitor, Bldg. 102. Memo to Dusty Meadows. Subject: T_2 accident and personnel involved, October 13, 1954. October 26, 1954.
[2] 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.
[3] Souers, C. Overheads from a talk "Tritium Releases from B331" prepared for the Director's Office, June 1988.
[4] 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.
[5] 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.
[6] Fleming, E.H. Memo "MINT" to Director's Office. April 1, 1958.
[7] Batzel, R.E. Memo "MINT" to Directors Office. November 5, 1958.
[8] 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.
[9] 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.

[10] 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.
[11] 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.
[12] 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.
[13] 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.
[14] Peterson, S-R., G.M. Gallegos, R.J. Harrach. A review of the January 20, 1965 tritium release from Lawrence Radiation Laboratory. Lawrence Livermore National Laboratory, Livermore CA, UCRL-AR-148811, July 2002.
[15] Lawrence Radiation Laboratory. Investigating Board Report for the Lawrence Radiation Laboratory Tritium Release of January 20, 1965 (unclassified title for classified report). UCRL-12402, March 1965.
[16] May, M.M. Memo "Release of Tritium to the Atmosphere" for period April 1 through June 30 1966 to A.R. Van Dyken, U.S. Atomic Energy Commission, Washington D.C. Secret restricted, unclassified in 1975. July 11, 1966.
[17] Silver, W.J. Memo to Building 331 File, January 20, 1970. Subject: Summary of Tritium Stack Release Test.
[18] 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.
[19] Myers, D.S.; Tinney, J.F.; Gudiksen, P.H. Health physics aspects of a large accidental tritium release. In: Moghissi, A.A.; Carter, M.W., eds. Tritium. Phoenix: Messenger Graphics; 1973: 611-622.
[20] Yoder, R.E. Memo to J.S. Kane, April 26, 1971 (?); Subject LRL Incident/Accident Report Serial No. 003
[21] Value from the spreadsheets prepared from analytical data (see discussion under Table 5 in the text).
[22] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1981, memo to distribution February 18, 1982.
[23] Handwritten notes from Hazards Control Records
[24] Hill, R.W. Memo to R.M. Alire June 29, 1984. Subject: LLNL Incident Analysis Report, Serial No. 0317 Tritium Releases B331
[25] King. W.C., Radiation and Environmental Safety Division, Hazards Control Department, memo to distribution January 22, 1985.
[26] Howe, H. Memo to C. Souers January 22, 1985; Subject: Dose Equivalent Estimates – Tritium Release, June 8, 1984
[27] Howe, H. Memo to C. Souers February 26, 1985; Subject: Tritium Release from Building 331 – January 24, 1985
[28] Obtained in 1999 from papers in the possession of Rebecca Failor.
[29] 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)
[30] Ragaini, R. Memo to S. Rosenblum, EPA, San Francisco dated August 30, 1989. Subject Report on Release of Tritium August 30, 1989
[31] Galles. H. Draft letter to Ed Howell, Alameda County Health Care, Oakland, CA. April 5, 1991.
[32] Lee, J.D.; Gary Mansfield. Memorandum to R.A. Failor. Subject: Mole sieve results for 4/1/91 – 4/3/01. April 8, 1991.

Table 5. Speciation of routine releases from the LLNL Tritium Facility. "B331 Spreadsheets" are calculated based on tritium concentrations reported by the analytical laboratory and assumptions about stack flow until 1996, when the TAMM air effluent spreadsheets are used.

	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[1]	831[1]
1975	1248	1108				
1976	1150	1672			1163[2]; 1161[3,4]	1659[2]; 1657[3,4]
1977	1215	2034			1127[2]; 1182[3,5]	1829[2]; 1965[3,5]
1978	1981	1923			2183[6]; 2271[2]; 2285[3]; 2284[7]	1853[6]; 1961[2]; 1960[3,7]
1979	2361	1600			2543[3,8]	1526[3,8]
1980	750	1459			800[6]; 802[9,3]; 837[10]	1377[11,9,3,6]; 1380[10]
1981	1097	1609			1007[12]; 1013[6]; 1034[13,3]	1518[13,6]; 1510[3]; 1545[12]
1982	608	1316			593[14]; 608[15,3]	1306[15,15,3,14]
1983	1798	1220			1831[16]; 1611[3]	1193[16,3]
1984[17]	1188	1136			1146[18]; 911[3]	1045[18,3]
1985[19]	1260	649			365[3]	624[3]
1986	487.7[20]	654.4	467.4	660.6	332 (rt) & 135 (ac)[3]; 467.7[21,22]	661[3]; 654.4[21]
1987	1423.0	1283.3	1387.6	1246.2	813 (rt) & 575 (ac)[3]; 1386.7[23]	1246[3]; 1227[23]
1988	2341.6	1636.1	2341.9	1635.6	2341.9[24]; 1542 (rt) & 800 (ac)[3]	1635[3]
1989	1300[25]	1749	1395	1554.4	1395[26,3]	1554[26]; 1555[3]
1990	538	684	581.8	699.5	572[3]	689[3]
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[27]	62.79[27]
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	104	6.0		

REFERENCES AND ANNOTATIONS (Table 5)

[1] Derived from the same analytical data as the spreadsheet results but with different assumptions about sample size
[2] Extracted from hand-drawn graph by S. Homann, "Building 331 Monthly Tritium Release for 1976";
[3] Obtained from papers in the possession of Rebecca Failor.
[4] 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.
[5] 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.
[6] 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.
[7] 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.
[8] 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.
[9] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1980, memo to distribution, January 15, 1981
[10] 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
[11] An additional 40 Ci of tritium (undifferentiated species) was reported.
[12] 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
[13] Morris, R.L., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 During 1981, memo to distribution February 18, 1982.
[14] 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
[15] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1982, memo to distribution, February 2, 1983.
[16] Velen, S., Health Physics Group, Hazards Control Department; Tritium Releases from Building 331 during 1983, memo to distribution, January 17, 1984
[17] The accidental HT release of about 5000 Ci of June 8, 1984 has been subtracted from the total.
[18] King. W.C., Radiation and Environmental Safety Division, Hazards Control Department, memo to distribution January 22, 1985.
[19] The accidental release of about 1000 Ci of January 24, 1985 has been subtracted from the total.
[20] 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 2.
[21] Gordon, Lisa E. Memo to D. Brekke. Subject: 1986 Effluent Monitoring Report for Building 331. January 30, 1987.
[22] 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 2.
[23] 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.
[24] Biermann, A. Memo to D. Brekke, February 21, 1989.
[25] 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).
[26] Mansfield, G. Memo to M. Singleton, April 13, 1990
[27] Tate, P. Memo to M. Mintz February 9, 1996. Subject: B-331 Routine Tritium Report

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

Year	Bubbler Data[1]	Sandia Data[2]		SAIC Data[3]		Accidents[4]
		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[5]	51	244	
1991		113.4	352.1[6]			
1992[7]		130	133.7			5/28/92; 4 Ci HT[8] 6/1/92; 36 Ci HT[8]
1993[7]		55.3	132.4			2.0 HTO October
1994[7]		4.1	91.2			
1995[9]		1.06	72.9			
1996			0.078[8]			

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[1] Hafner, R. Personal communication. 2002.
[2] Garcia, T.B. and T.P. Gorman. Radiological Characterization and Final Facility Status Report Tritium Research Laboratory. SAND96-8004 UC-407; August 1966.
[3] Science Applications International Corporation. Review of Tritium Operations, Emissions, and Measurements in the Livermore Valley. Prepared for Lawrence Livermore National Laboratory, January 1993.
[4] Garcia, T.B.; Gorman, T.P. Radiological characterization and final facility status report Tritium Research Laboratory. Sandia Report. SAND96-8004UC-407. August 1996.
[5] 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 [3])
[6] Includes 1.368 Ci "Evap"
[7] Confirmed by the SNL/CA Site Environmental Reports
[8] Johnson, Alice J. Tritium Research Laboratory Cleanup And Transition Project Final Report. Sandia National Laboratory, Livermore, CA. SAND97-8009. February 1997.
[9] Chavarria, J.J. Memo to Becky Failor, January 22, 1996. Subject: Calendar year 1995 radiological effluent data for SNL/CA

Table 7. Stack or source height, stack diameter or area of source, exit velocity, and distance, direction, and bearing towards the air tritium monitor (VIS) near the Discovery Center for LLNL and SNL tritium sources.

	Stack (or source) height (m)[1]	Stack diameter (m) or area of source (m^2)[1]	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)
B331 South Stack (#1) North Stack (#2)	30	1.22	1968[2]: 5.7 (#1); 10.7 (#2) 1970[3]: 5.4 (#1) 1972[3]: 5.7 (#1); 11.1 (#2) 1976[3]: 5.65 (#1); 10.1 (#2) 1979[4]: 5.1 (#1); 9.1 (#2) 1981[5]: 5.3 (#1); 10.9 (#2) 1985[6]: 6.4 (#1); 9.7 (#2) 1987[7]: 6.9 (#1); 9.9 (#2) 1989[8]: 6.8 (#1); 9.6 (#2) 1991[9]: 7.6 (#1); 10.5 (#2) 1995[10]: 6.3 (#1); 8.8 (#2) 1997[11]: 5.9 (#1); 9.1 (#2)	ENE (NE) 75.06° (#1); 76.22° (#2) 1036 m (#1); 1046 m (#2)
B231 (Rm 1128 or Vault)	12.2; 10.6 or 18.6-	0.41; 0.6 or 0.56	1992: 12; 6 or 12.8	ENE (E); 73°; 1226 m
B292 Stack	20.2[12]	0.71	1979[13]: 6.1 1981[14]: 12.6 1985: 11.1[15]; 14.4[16]; 26.5[17] 1986[18]: 9.1 1989[19]: 17.2 1992[20]: 15.4	ESE (E) 109.83° 1308 m
B212 Room exhaust	4.0	1.0	0.4	Same as B212 ICT Stack
B212 ICT Stack	8.2[21]	0.2[22]	1974 through 1982[23]: 4.43	ENE (NE); 64.48° 1332 m
SNL TRL Stack	30[24]	1.0[24]	27.6[25]	NNE (NE); 32.9°; 1568 m
B612 diffuse	1.0	600	0.4	NNE (NE); 23°; 542 m
B624 Incinerator	11.7[26]	0.46[26]	13.5[26]	Same as B612 diffuse
B331 diffuse	1.0	1.0	0.5	ENE (E); 75.64°; 1041 m

REFERENCES AND ANNOTATIONS (Table 7)

[1] Taken from NESHAPs reports unless otherwise noted.
[2] Industrial Hygiene & Toxicology Section. Memo to Radiation Safety Section. Subject: Bldg. 331. October 7, 1968.
[3] Summary of Building 331 (Tritium Facility) Stack Flow Measurements, 1968-1987.
[4] Industrial Hygiene & Toxicology Group, Hazards Control Department. Memo to Chuck Folkers. Subject: Air flow measurements. November 8, 1979.
[5] Industrial Hygiene & Toxicology Group, Hazards Control Department. Memo to George Morris. Subject: Evaluation of Stack Samplers, Bldg. 331. September 22, 1981.
[6] Eneidi, Walt. Memo to George Morris. Subject: Airflow measurements, Bldg. 331. April 22, 1985.
[7] Eneidi, Walt. Memo to George Morris. Subject: Exhaust airflow measurements, Building 331 (April 3, 1987). April 8, 1987.
[8] Wong, M. Memo to G. Morris. Subject: Building 331 tritium sampler verification test results. May 2, 1989.
[9] LLNL NESHAPs Project Quarterly Progress Report. Environmental Protection Department, Lawrence Livermore National Laboratory, Livermore CA. UCRL-AR-108419-92-2. June 31, 1992.
[10] From calibration dated May 5, 1995 found in LLNL Radiological Air Effluent Emission Data for 1995 Volume 5 (of 5); Laboratory Records Center.
[11] 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.
[12] The stack was measured in 2003 because of discrepancies in stack heights mentioned in various NESHAPs reports.
[13] 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.
[14] 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.
[15] Heikkinen, Dale W. RTNS-II Operations Guidebook. Lawrence Livermore National Laboratory, Livermore CA. UCID-20299. April 1, 1985.
[16] Trent, M. Memo to Kyle Griggs. Subject: B-292 Tritium Stack Releases: 1985, January 21, 1986.
[17] Calculated from the air flow rate of 10.5 m^3/s given in [15] and the stack diameter from [14] and NESHAPs reports.
[18] Trent, M. Memo to David Brekke. Subject: Stack releases from Building 292 (RTNS-II) and Building 865 (ATA). February 5, 1987.
[19] Biermann, A. Memo to D. Brekke. Subject: 1988 Air effluent monitoring results for radionuclides. February 21, 1989.
[20] 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.
[21] The stack was measured in 2004 after being identified by D.S. Myers.
[22] Estimated inside diameter
[23] Calculated from flow rate of 0.275 m^3 s^{-1} mentioned in the air effluent monitoring notebook for 1975.
[24] Garcia, Toff. Email to Ring Peterson April 22, 2002.
[25] See [24]. Based on flow rate of 1300 m^3 per minute.
[26] 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.

Table 8. Annual releases of HT and HTO from Building 231 (then Building 102; the predecessor of the LLNL Tritium Facility), one standard deviation uncertainty on a normal distribution, and truncated lower limit.

	Building 231 HT			Building 231 HTO		
Year	Ci/y	± sd	LL	Ci/y	± sd	LL
1953	4860	2920	0	540	324	0
1954	4860	2920	0	540	324	0
1955	4860	2920	0	540	324	0
1956	3470	2000	2800	530	240	0
1957	11390	1900	8400	610	90	NA
1958	3220	1100	2450	280	80	NA

Table 9. Annual releases of HT and HTO from Building 212 and uncertainty as one standard deviation of a normal distribution.

	Building 212 HT		Building 212 HTO	
Year	Ci/y	± sd	Ci/y	± sd
Room Air: accelerator and cyclotron				
1954 - 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 (rectangular)[a]		0.6 – 6 (rectangular)	
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)[b]		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	9.10
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] A rectangular distribution describes an equal probability of sampling all values in the range.
[b] A triangular distribution describes the minimum, maximum, and most likely values.

Table 10. Estimated routine annual releases of HT and HTO from the LLNL Tritium Facility (Building 331). Uncertainty is one standard deviation of a normal distribution.

	Stack 1 HT		Stack 1 HTO		Stack 2 HT		Stack 2 HTO	
	Ci	± 1 sd	Ci	± 1 sd	Ci	± 1 sd	Ci	± 1 sd
1958	2306	1100[i]	444	213[ii]				
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	2470	2530	2860	3590	4050	4220	4690
1965	405	214	476	235	676	332	793	361
1966	526	322	618	361	877	510	1030	568
1967	1140	530	1340	572	1900	804	2230	851
1968	1160	529	1370	569	1940	769	2280	800
1969	1680	822	1970	895	2800	1220	3290	1290
1970	775	473	910	529	1290	734	1520	816
1971	467	264	549	293	779	404	915	443
1972	182	85.4	324	123	388	160	456	139
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	144	1170	319	742	218	953	284
1978	799	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

Table 10 continued

	Stack 1 HT		Stack 1 HTO		Stack 2 HT		Stack 2 HTO	
	Ci	± 1 sd	Ci	± 1 sd	Ci	± 1 sd	Ci	± 1 sd
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	58.5	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	0.738	103	23.9

[i] This distribution is left-truncated at 1925 Ci.
[ii] This distribution is left-truncated at 0 Ci.

Table 11. HTO estimated to have been released from the Building 331 WAA and the Building 612 yard before measurements were taken. The distributions for both are normal with one standard deviation (σ).

	Building 331 WAA		Building 612 Yard	
Year	Ci y^{-1}	± σ	Ci y^{-1}	± σ
1958	34.0	29.7	-	-
1959	39.5	33.6	-	-
1960	80.0	68.8	-	-
1961	19.8	16.1	-	-
1962	110	87.8	-	-
1963	148	118	-	-
1964	309	298	-	-
1965	58.1	46.9	46.5	20.6
1966	75.4	62.1	60.8	29.8
1967	163	130	131	52.3
1968	167	133	134	53.1
1969	241	193	193	81.6
1970	111	91.5	89.6	43.9
1971	67.0	54.6	53.9	25.2
1972	33.4	26.5	28.6	10.7
1973	63.3	50.7	50.7	20.9
1974	43.2	34.1	24.1	9.19
1975	58.7	45.8	41.0	13.6
1976	75.8	59.4	66.5	22.3
1977	83.8	65.8	77.6	27.0
1978	110	85.8	78.3	26.5
1979	101	78.3	59.7	18.6
1980	54.3	42.3	52.0	17.0
1981	63.5	49.4	56.2	18.1
1982	47.3	36.8	47.9	15.1
1983	73.6	57.4	44.4	14.3
1984	47.7	37.6	38.2	12.1
1985	24.5	19.0	23.6	7.29
1986	26.9	20.9	23.1	7.36
1987	65.3	50.8	45.8	14.5
1988	98.3	76.5	61.2	19.5
1989	73.4	57.0	59.4	18.6
1990	31.1	24.1	35.5	14.4
1991	28.4	22.1	25.5	8.09
1992	-	-	3.79	1.21

Table 12. Annual HTO releases ± one standard deviation (σ) from the Building 331 WAA and the Building 612 yard estimated from annual mean air tritium concentrations from nearby tritium samplers.

	B331 WAA		B612 Yard	
Year	Ci y^{-1}	± σ	Ci y^{-1}	± σ
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	95.7	34.8	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	3.64	1.10
2002	0.750	0.259	3.68	1.11
2003	8.70	2.89	3.40	1.02

Table 13. HTO 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 (Ci y^{-1})			Building 292	
Year	Lower limit	Peak	Upper limit	Ci y^{-1}	± σ
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 14. Annual routine releases of HT and HTO from the SNL Tritium Research Laboratory and uncertainty as one standard deviation (σ)

	SNL TRL HT		SNL TRL HTO	
Year	Ci y^{-1}	± σ	Ci y^{-1}	± σ
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	1030	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 15. Dilution factors (χ/Q in s m^{-3}) with uncertainty (lognormal distribution) for modeled sources other than the LLNL Tritium Facility.

Building	Years	χ/Q in s m^{-3}	± σ
B231 Stack	1953 - 1958	$1.865\ 10^{-6}$	$8.016\ 10^{-7}$
B212 Room Air	1953 - 1966	$2.678\ 10^{-6}$	$8.474\ 10^{-7}$
B331 WAA	1958 - present	$3.474\ 10^{-6}$	$1.042\ 10^{-6}$
B612 Yard	1965 - present	$1.763\ 10^{-5}$	$5.289\ 10^{-6}$
B212 Stack	1966 - 1987	$2.494\ 10^{-6}$	$7.483\ 10^{-7}$
B624 incinerator	1977 - 1988	$7.925\ 10^{-6}$	$2.380\ 10^{-6}$
B292 Stack	1979 - 1989	$4.425\ 10^{-7}$	$1.427\ 10^{-7}$
Sandia TRL Stack	1979 - 1989	$6.595\ 10^{-7}$	$1.878\ 10^{-7}$

Table 16. Dilution factors (χ/Q in s m^{-3}) with uncertainty (lognormal distribution) for Stack 1 and Stack 2 of the LLNL Tritium Facility (Building 331)

	Stack 1 (south)		Stack 2 (north)	
Year	s m^{-3}	± σ	s m^{-3}	± σ
1958 - 1961	$1.230\ 10^{-6}$	$3.076\ 10^{-7}$	-	-
1962 - 1967	$1.230\ 10^{-6}$	$3.076\ 10^{-7}$	$1.059\ 10^{-6}$	$2.654\ 10^{-7}$
1968	$1.22e\ 10^{-6}$	$3.056\ 10^{-7}$	$1.064\ 10^{-6}$	$2.664\ 10^{-7}$
1969	$1.227\ 10^{-6}$	$3.072\ 10^{-7}$	$1.059\ 10^{-6}$	$2.654\ 10^{-7}$
1970	$1.232\ 10^{-6}$	$3.082\ 10^{-7}$	$1.059\ 10^{-6}$	$2.654\ 10^{-7}$
1971	$1.226\ 10^{-6}$	$3.070\ 10^{-7}$	$1.059\ 10^{-6}$	$2.654\ 10^{-7}$
1972	$1.221\ 10^{-6}$	$3.055\ 10^{-7}$	$1.055\ 10^{-6}$	$2.642\ 10^{-7}$
1973 - 1978	$1.230\ 10^{-6}$	$3.080\ 10^{-7}$	$1.077\ 10^{-6}$	$2.718\ 10^{-7}$
1979	$1.240\ 10^{-6}$	$3.101\ 10^{-7}$	$1.101\ 10^{-6}$	$2.758\ 10^{-7}$
1980	$1.237\ 10^{-6}$	$3.093\ 10^{-7}$	$1.081\ 10^{-6}$	$2.722\ 10^{-7}$
1981	$1.233\ 10^{-6}$	$3.084\ 10^{-7}$	$1.059\ 10^{-6}$	$2.653\ 10^{-7}$
1982 - 1984	$1.217\ 10^{-6}$	$3.055\ 10^{-7}$	$1.073\ 10^{-6}$	$2.694\ 10^{-7}$
1985	$1.201\ 10^{-6}$	$3.004\ 10^{-7}$	$1.086\ 10^{-6}$	$2.719\ 10^{-7}$
1986	$1.192\ 10^{-6}$	$2.985\ 10^{-7}$	$1.084\ 10^{-6}$	$2.715\ 10^{-7}$
1987	$1.184\ 10^{-6}$	$2.965\ 10^{-7}$	$1.083\ 10^{-6}$	$2.711\ 10^{-7}$
1988	$1.187\ 10^{-6}$	$2.969\ 10^{-7}$	$1.086\ 10^{-6}$	$2.720\ 10^{-7}$
1989	$1.189\ 10^{-6}$	$2.974\ 10^{-7}$	$1.090\ 10^{-6}$	$2.729\ 10^{-7}$
1990	$1.177\ 10^{-6}$	$2.951\ 10^{-7}$	$1.080\ 10^{-6}$	$2.709\ 10^{-7}$
1991	$1.166\ 10^{-6}$	$2.919\ 10^{-7}$	$1.068\ 10^{-6}$	$2.675\ 10^{-7}$
1992 - 1994	$1.184\ 10^{-6}$	$2.974\ 10^{-7}$	$1.088\ 10^{-6}$	$2.738\ 10^{-7}$
1995	$1.203\ 10^{-6}$	$3.011\ 10^{-7}$	$1.108\ 10^{-6}$	$2.775\ 10^{-7}$
1996	$1.239\ 10^{-6}$	$3.105\ 10^{-7}$	$1.095\ 10^{-6}$	$2.751\ 10^{-7}$
1997	$1.236\ 10^{-6}$	$3.090\ 10^{-7}$	$1.101\ 10^{-6}$	$2.758\ 10^{-7}$
1998	$1.225\ 10^{-6}$	$3.062\ 10^{-7}$	$1.098\ 10^{-6}$	$2.754\ 10^{-7}$
1999	$1.240\ 10^{-6}$	$3.105\ 10^{-7}$	$1.088\ 10^{-6}$	$2.733\ 10^{-7}$
2000	$1.240\ 10^{-6}$	$3.107\ 10^{-7}$	$1.115\ 10^{-6}$	$2.803\ 10^{-7}$
2001	$1.245\ 10^{-6}$	$3.119\ 10^{-7}$	$1.146\ 10^{-6}$	$2.881\ 10^{-7}$
2002	$1.253\ 10^{-6}$	$3.142\ 10^{-7}$	$1.125\ 10^{-6}$	$2.832\ 10^{-7}$
2003	$1.254\ 10^{-6}$	$3.145\ 10^{-7}$	$1.125\ 10^{-6}$	$2.831\ 10^{-7}$

Table 17. 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}	± σ	Bq m^{-3}	± σ
1974	448	68.8	3.14	0.655
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

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

Table 18. Annual absolute (kg m^{-3}) as measured by silica gel and relative humidity from the LLNL meteorological tower. Uncertainty on all absolute humidity estimates 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
Overall	0.0078	0.690

Table 19. 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.

	# of monthly samples / 12	Bq L^{-1}	± 1 σ	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

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