11

Vegetation and Foodstuff Monitoring

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Introduction

Lawrence Livermore National Laboratory has a vegetation and foodstuff monitoring program to comply with U.S. Department of Energy (DOE) guidance, which states that periodic sampling and analysis of vegetation should be performed to determine if there is measurable, long-term buildup of radionuclides in the terrestrial environment (U.S. Department of Energy 1991).

Tritium is the nuclide of major interest in the LLNL vegetation and foodstuff monitoring program. LLNL has historically released tritium to the air accidentally and during routine operations and tritium is the only radionuclide released from LLNL activities that occurs in detectable concentrations in vegetation and foodstuff. Tritium moves through the food chain as tritiated water and can be rapidly assimilated into plant water and then incorporated into the organic matter of plants through photosynthesis. It can contribute to human radiation dose if it is inhaled, absorbed through the skin, or ingested via vegetables, or milk and meats from animals that have eaten tritiated vegetation.

LLNL has been monitoring tritium in vegetation to some extent since 1966 and has performed vegetation sampling in the vicinity of the Livermore site and Site 300 as part of a continuing monitoring program since 1972. The monitoring program is designed to measure changes in the environmental levels of radioactivity, to evaluate the environmental effect of LLNL operations, and to calculate potential human doses from radionuclides in the food chain.

In 1977, LLNL added wine to the LLNL monitoring program. Wine is now the most important agricultural product in the Livermore Valley, representing an approximately $80-million annual industry, based on sales. Although the tritium concentrations in all wines are low, the sampling data indicate that Livermore Valley wines contain statistically more tritium than do wines from other California wine-producing regions.

In the past, other foodstuffs (cow milk, goat milk, and honey) leading to potential dose were also monitored for tritium. At present, however, only tritium concentrations in vegetation and wine are used to assess potential ingestion dose from tritium emitted during LLNL operations. During 1999, LLNL collected and analyzed samples of herbaceous vegetation and wine. Potential human doses from these foodstuffs were calculated using the monitoring data and the dose models presented in Appendix A. In addition, as part of a continuing study, LLNL determined the potential tritium dose to the maximally exposed individual from a pine tree at the Livermore site. This tree serves as a diffuse source of tritium because it loses tritium to the air through evapotranspiration of tritium-contaminated water in the root zone. The dose was calculated using the U.S. Environmental Protection Agency (EPA) model, CAP88-PC.

Methods

The methods used for monitoring vegetation and wine are presented briefly in the following sections and in more detail in the Data Supplement. All vegetation and wine sampling was conducted according to written and approved standardized procedures (Tate et al. 1999).

Vegetation

LLNL staff collected vegetation samples, usually annual grasses, quarterly from 22 fixed locations in the Livermore Valley, San Joaquin County, and Site 300. The samples were then analyzed for tritium.

Location maps are provided in **Figures 11-1** and **11-2**. Sample locations were selected to represent vegetation from: (1) locations near LLNL that can be affected by LLNL operations, (2) background locations where vegetation is similar to that growing near LLNL but is unlikely to be affected by LLNL operations, and (3) areas of known or suspected LLNL-induced contamination. Sampling locations for 1999 were the same as those in 1998.

Wine

Three types of wine samples were collected and analyzed for tritium concentrations: wine produced from grapes grown in the Livermore Valley, wine produced from grapes grown in California outside the Livermore Valley, and wine produced from grapes grown in Europe (France, Germany, and Italy). The wines were purchased from local retailers to represent what the general public could buy and drink during 1999.

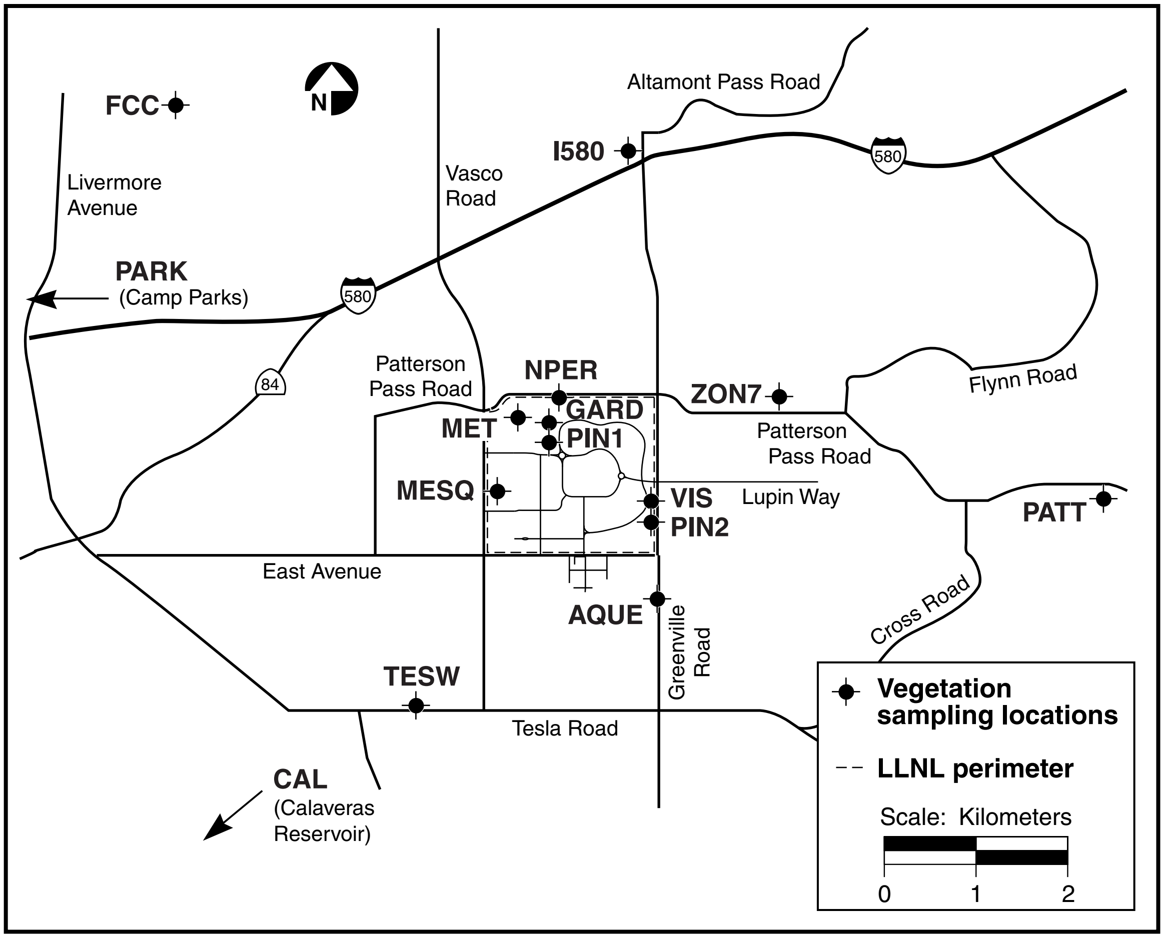

Figure 11-1. Livermore site and Livermore Valley vegetation sampling locations, 1999.

Data from the analysis of tritium in wine can be used to estimate the potential tritium dose received by consumers during the year of purchase. However, because wines purchased in 1999 are from grapes that were harvested in 1996, 1997, and 1998, the 1999 sampling data cannot be used to indicate how LLNL's operations affected concentrations of tritium in wines produced from grapes grown in 1999. To analyze trends and help determine the impact of LLNL operations on tritium in wine for the year when the grapes were harvested, LLNL corrects the wine sample concentrations for the radiological decay that has occurred between the approximate date of the grape harvest and the date when the wine was analyzed in the laboratory. Comparisons can then be made of wine concentrations that represent the year when the grapes were exposed to the tritium.

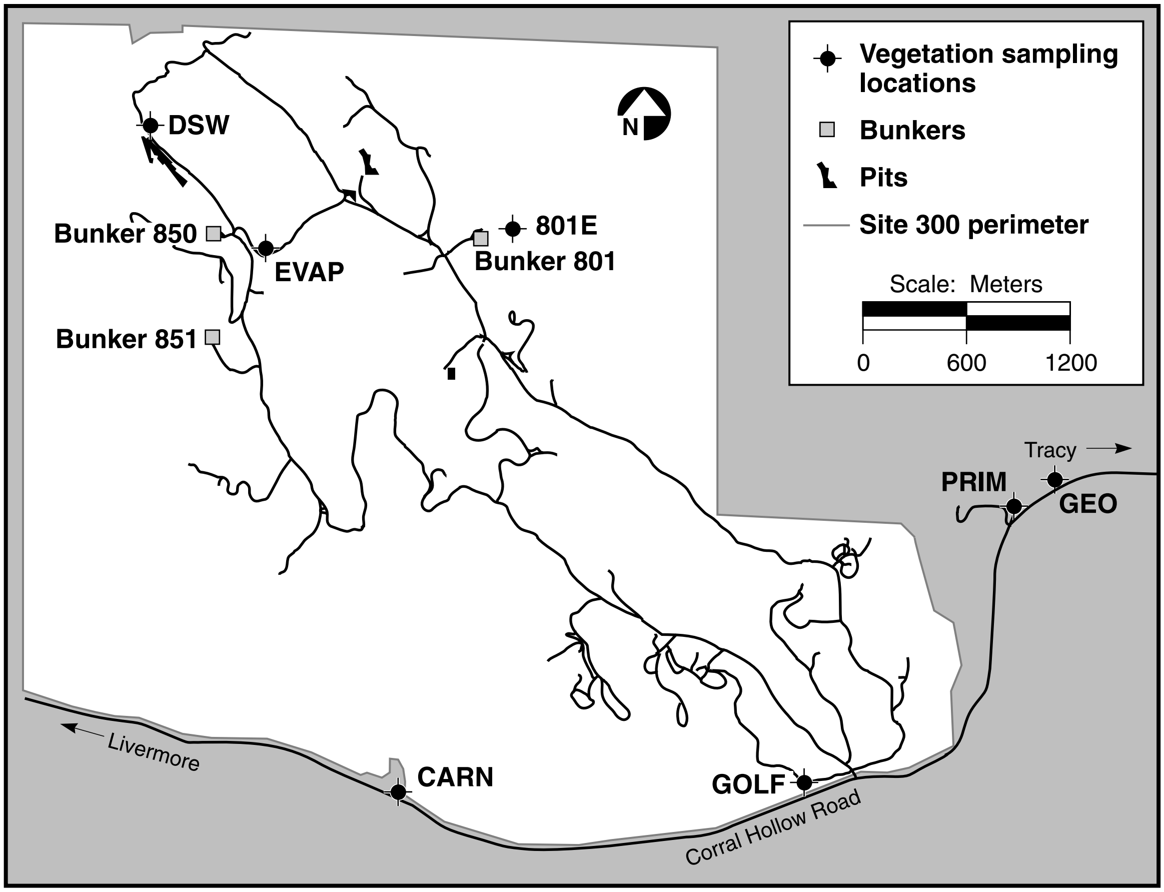

Figure 11-2. Site 300 vegetation sampling locations, 1999.

Results

The results of vegetation monitoring for the Livermore site and Site 300 and of wine monitoring are presented in the following sections.

Livermore Site

Vegetation

The Livermore site and Livermore Valley vegetation locations are put into four groups for statistical evaluation:

- Near—locations within 1 km of the Livermore site perimeter. Near locations include AQUE, GARD, MESQ, NPER, MET, PIN2, and VIS.

- Intermediate—locations in the Livermore Valley 1–5 km from the Livermore site perimeter that are often downwind and, thus, potentially under the influence of tritium releases at the site. The intermediate locations are I580, PATT, TESW, and ZON7.
- Far—locations unlikely to be affected by LLNL operations. One background location (CAL) is more than 25 km away. The other two (FCC and PARK), although in the Livermore Valley, are unlikely to be affected by LLNL operations because they are more than 5 km from the Livermore site and are generally upwind.
- PIN1—location of a pine tree rooted in an area of known tritium contamination on the Livermore site.

Table 11-1 shows summary tritium data for vegetation collected for the LLNL vegetation monitoring program in 1999 (individual sampling values are presented in the Data Supplement of this report). **Figure 11-3** shows the 1999 medians of the tritium concentrations for PIN1, Near, and Intermediate Livermore locations as a continuation of historic median concentrations from 1971 to 1998. The values for 1998 and 1999 Far locations are the lowest positive measured concentrations rather than the medians; medians for 1998 and 1999 are negative for the Far location and, hence, cannot be plotted on a logarithmic scale. Although the concentration in Far vegetation appears to drop by about a factor of 10 in 1998, it is highly unlikely that any difference exists among 1998, 1999, and recent preceding years. The apparent difference is caused by a change in how the analytical laboratory reported concentrations less than the detection limit.

For 1999, the data for tritium in vegetation were compared using Scheffé's and Games/Howell multiple comparisons (Scheffé 1953; Games and Howell 1976). These tests are the most appropriate tests for these distributions of data. Unlike previous years, the Near group was not found to be significantly different from the Intermediate and Far groups. This was caused by unusually high observed values of tritium in vegetation at VIS, PIN2, and I580 for the first quarter of 1999 when tritium releases were unusually high. When the Near group (without first quarter values for VIS and PIN2) is compared with the Intermediate group (without the first quarter value for I580) and the Far group, results are similar to previous years. Both tests show concentrations of the Near group to be significantly higher than concentrations from both the Intermediate and Far groups. The highest median tritium results for individual vegetation sampling locations were found at the Near locations, PIN2 and VIS, which are located near each other downwind of the Livermore site.

Table 11-1. Concentrations of tritium in plant water (Bq/L), 1999.

Location[a]	Detection frequency[b]	Median	Interquartile range	Maximum	Dose (µSv/y)[c] Median	Dose (µSv/y)[c] Maximum
Near Livermore site[d]	25/28	7.0	9.9	100	0.034	0.48
Livermore site PIN1[e]	4/4	150	120	280	8.0×10^{-6}[f]	1.5×10^{-6}[f]
Livermore site intermediate locations	7/16	1.4	4.2	100	0.0068	0.48
Livermore site far locations	0/12	–0.72	1.8	0.68	—[g]	0.0033
Location DSW at Site 300[e]	1/4	1.6	1.3	3.1	0.0078	0.015
Location EVAP at Site 300[e]	3/4	170	370	480	0.81	2.3
All other locations at Site 300	5/20	–0.015	1.6	4.0	—[g]	0.019

Note: Radioactivities are reported as the measured concentration and either an uncertainty (±2σ counting error) or as being less than or equal to the detection limit. If the concentration is less than or equal to the uncertainty or the detection limit, the result is considered to be a nondetection. See the main volume, Chapter 14, Quality Assurance.

a See **Figures 11-1** and **11-2** for sampling locations.

b Detection frequency means the fraction of samples taken having a measured value above the detection limit.

c Ingestion dose is calculated based on conservative assumptions that an adult's diet is exclusively vegetables with this tritium concentration, and that meat and milk are derived from livestock fed on grasses with the same concentration of tritium. See Appendix A, Methods of Dose Calculations.

d Includes PIN2; plant water concentrations are similar among plant types.

e Sampling location in known area of contamination.

f For this dose calculation, PIN1 is treated as a diffuse source of tritium (because human beings do not eat pine needles). Dose, calculated using CAP88-PC, is to the maximally exposed individual.

g Dose is not calculated when the median concentration is negative.

In 1997, PIN1, a pine tree growing in a known area of tritium contamination at the Livermore site, was monitored on a monthly basis to estimate emissions for compliance with National Emissions Standards for Hazardous Air Pollutants (NESHAPs). In 1998, the tree sampling was coordinated with the quarterly vegetation sampling. Since 1998, NESHAPs calculations to the sitewide maximally exposed individual (SW-MEI) are based on quarterly observations. To assess the contribution of soil water tritium to PIN1, LLNL also sampled a second tree (PIN2) that is not in tritium-contaminated soil. Concentrations of tritium in PIN2, like in all other vegetation sampled near the Livermore site, are from air and soil water in quasi-equilibrium with air. When samples from PIN1 were compared with samples from each Near location, concentrations of tritium in PIN1 were found to be significantly higher than concentrations at all other locations except the downwind locations, VIS and PIN2.

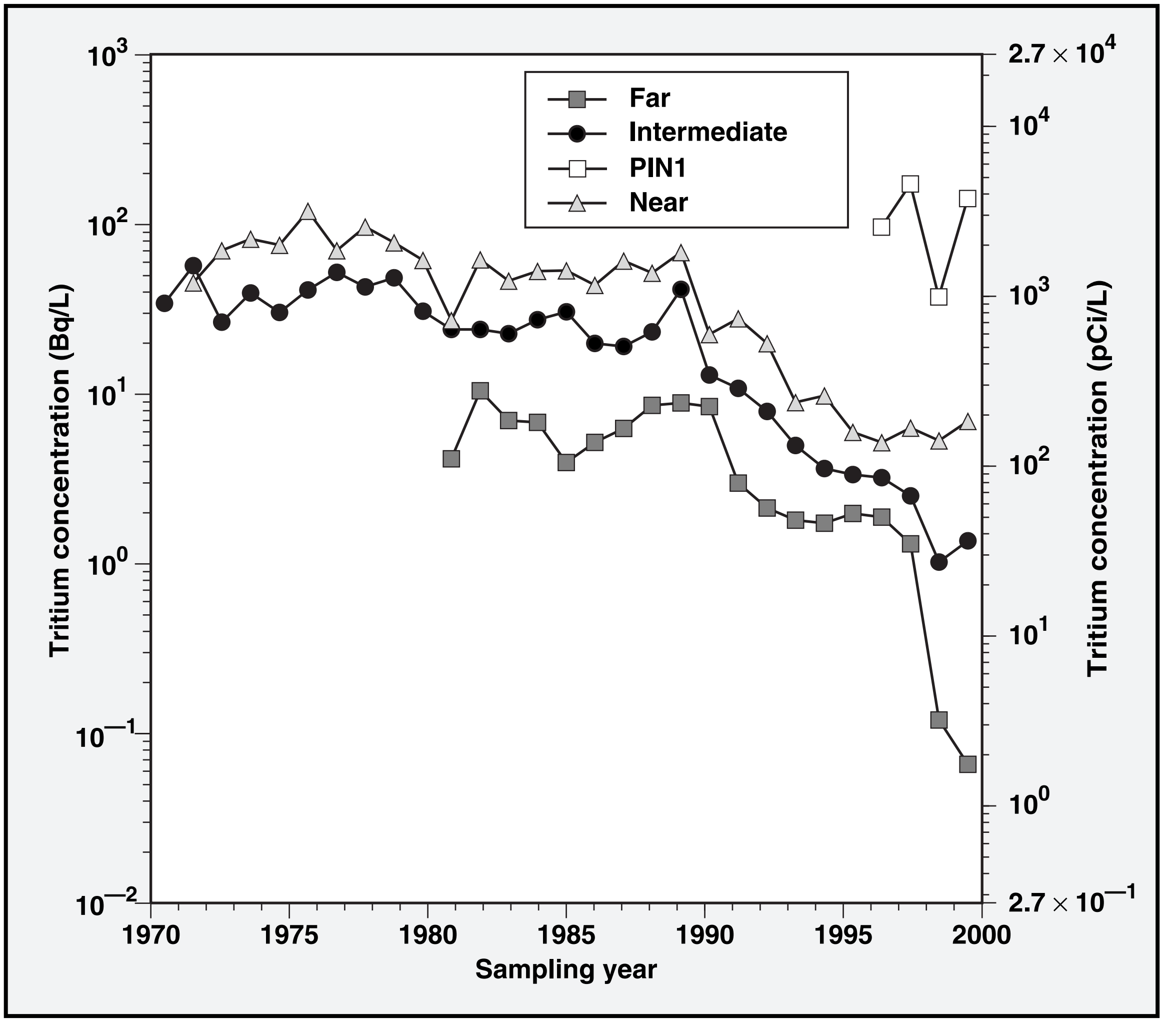

Figure 11-3. Median tritium concentrations in Livermore site and Livermore Valley plant water samples, 1971–1999. (For Far vegetation for 1998 and 1999, the values are the lowest positive.)

Wine

The results from the 1999 wine tritium analyses are shown in **Table 11-2**. Tritium concentrations are within the range of those reported in previous years and remain low in wines from all areas. The data for the 1999 sampling year were analyzed using Scheffé and Games/Howell multiple comparisons. The results of the comparisons are the same as in previous years. Both analyses show that the mean tritium concentration of the 12 Livermore wines is statistically greater than that of the six California wines. When the Livermore, California, and European wines were compared using the Scheffé test, no significant difference was noted among the groups because of the high variability of Livermore Valley wines. The variability in the Livermore Valley wines is due to some grapes being exposed to higher concentrations of tritium in air and in precipitation than others.

Table 11-2. Tritium in retail wine (Bq/L), 1999.[a]

Region	Detection frequency[b]	Median	Interquartile range	Mean	Maximum	Dose[c] (nSv/y)
Livermore Valley	12/12	1.7	0.68	2.4	8.3	2.2
California	6/6	0.43	0.15	0.45	0.57	0.40
Europe	4/4	1.5	0.30	1.3	1.6	1.2

Note: Radioactivity is reported as the measured concentration and an uncertainty (±2σ counting error). If the uncertainty is greater than the concentration, the result is considered to be a nondetection.

a Wines from a variety of vintages were purchased and analyzed during 1999. The concentrations reported are those at the time the bottles were opened.

b Detection frequency means the fraction of samples taken having a measured value above the detection limit.

c This dose is calculated using the assumption of drinking 52 L wine/year and using the mean concentration of sampled wines.

Concentrations corrected to vintage year are plotted in **Figure 11-4**. The downward trend seen in wines from all locations is not statistically significant. Depending upon the test used, 1982 and 1983 are statistically different from several other vintage years. The 1996 Livermore Valley wines were significantly higher in tritium than those from both 1995 and 1997 (Scheffé's test) and were significantly higher than those in just 1997 (Games/Howell test). Data from 1998 could not be included because the sample comprised only two bottles. As mentioned, wines are sampled randomly; and, quite by chance, the 1996 wines unequally represent vineyards close to LLNL.

Site 300

Vegetation

There are seven monitoring locations for vegetation at Site 300. Of these, five (CARN, GOLF, GEO, 801E, and PRIM) detect changes in atmospheric tritium concentrations. Vegetation from locations DSW and EVAP grows in areas of known ground water contamination. Plants with long roots sometimes absorb tritium concentrations from ground water rather than from air.

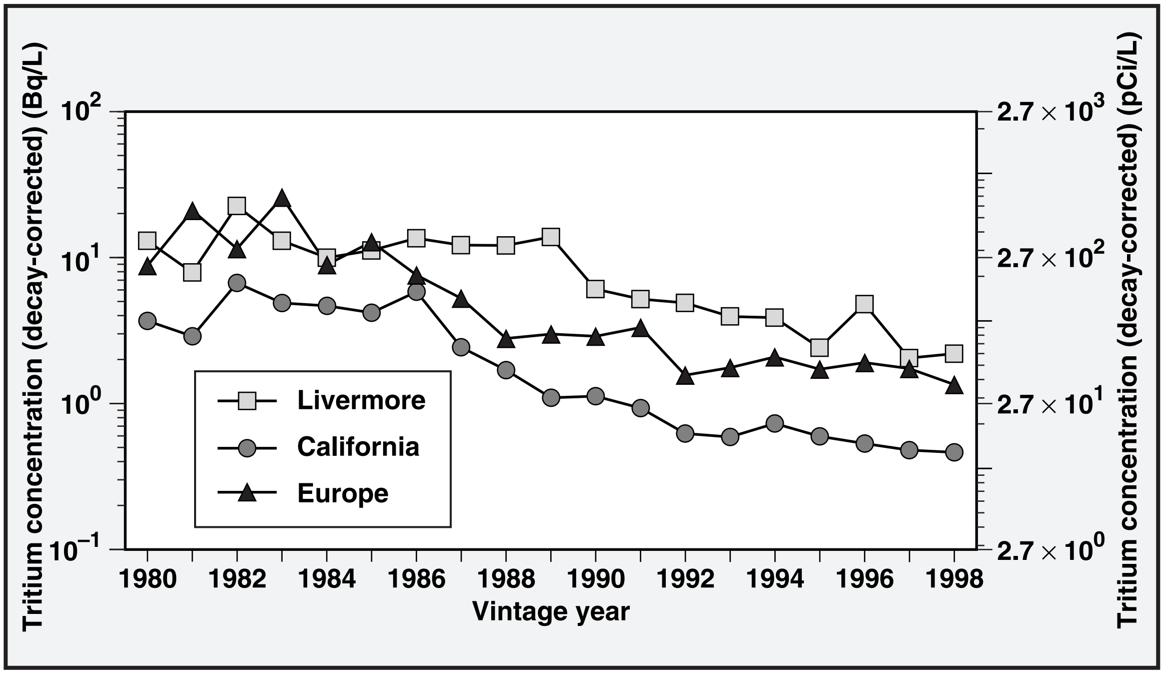

Figure 11-4. Mean tritium concentrations in retail wines decay-corrected from the sampling year to the vintage year.

Table 11-1 shows summary tritium data for vegetation collected at Site 300 during 1999. Historic values for tritium at Site 300 sampling locations are shown in **Figure 11-5**. Of the seven sampling locations at Site 300, six yielded results in 1999 at or near the detection limits. Only EVAP yielded results above detection limits; DSW, normally high (except for 1994), had tritium concentrations at detection limits. The extremely low concentrations for 1998 and 1999 plotted for locations other than DSW and EVAP are caused by having to graph the lowest positive result because the median is negative and the scale of the figure is logarithmic. The apparent difference among 1999, 1998, and preceding years is caused by a change in how the analytical laboratory reported concentrations lower than detection limits.

The highest tritium result for a single vegetation sample occurred at location EVAP (see **Table 11-1**), which is near a spring where ground water flows near the surface and evaporates. The ground water in this area is contaminated with tritium that comes from three sources: Pit 3, Pit 5, and the firing table at Building 850 (see the discussion of wells NC7-61 and NC7-69 in Chapter 9, Ground Water Monitoring). Evaluation of the 1999 data for Site 300 using the Scheffé test yielded no significant differences among the various sampling locations; this is a result of the high variability of the data and the low number of data points.

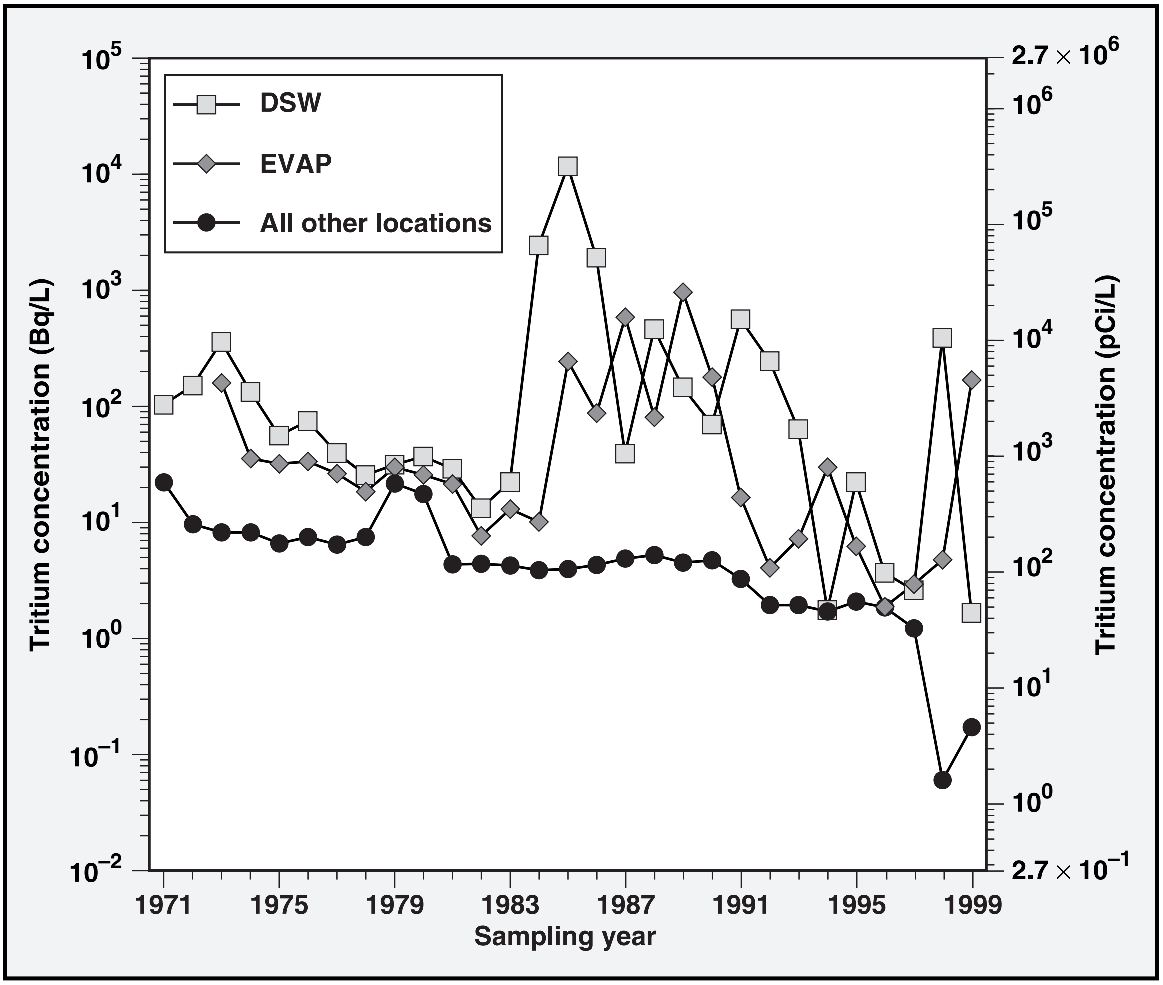

Figure 11-5. Median tritium concentrations in plant water at Site 300 sampling locations, 1971–1999. When the median is negative (e.g., all other locations for 1998 and 1999), the lowest positive concentration has been substituted.

Environmental Impact

In 1999, the environmental impacts of LLNL operations on vegetation and wine were small and are presented below for the Livermore site and Site 300.

Livermore Site

LLNL impacts on vegetation in the Livermore Valley remained minimal in 1999. The effective dose equivalents shown in **Table 11-1** were derived using the dose conversion factors provided by DOE (U.S. Department of Energy 1988) and the dose pathway model from U.S. Nuclear Regulatory Commission (NRC) Regulatory Guide 1.109 (1977). Appendix A provides a detailed discussion of dose calculation methods. The dose from ingested tritium is based on the conservative assumptions that an adult's diet (Table A-2, NRC maximum) consists exclusively of leafy vegetables with the measured tritium concentrations, and meat and milk from livestock fed on grasses with the same concentrations. In actuality, the vegetables consumed by an adult contain tritium at lower levels than those reported because most vegetables are imported from other areas. Similarly, tritium concentrations in food consumed by local livestock are at or below the concentrations in vegetation measured at the Intermediate and the Far locations. Nevertheless, based on these extremely conservative assumptions, the maximum potential dose from ingestion of affected vegetation for 1999 for the Livermore Valley is 0.50 µSv (0.050 mrem).

Doses are calculated based on measured tritium in plant water without the contribution of organically bound tritium (OBT). Dose conversion factors of 1.8×10^{-11} Sv/Bq for tritium in the plant or animal water (HTO) and 4.2×10^{-11} Sv/Bq for OBT have been published by the International Commission on Radiological Protection (1996). These show the relative importance of ingested HTO and OBT to dose. In vegetables, the dose from HTO is greater because the fraction of the plant that is organic matter is quite small (10–25%). For example, about 10% of the ingestion dose from leafy vegetables (about 10% dry matter) is from OBT. OBT becomes increasingly important when the fraction of dry matter increases. Pork, for example, has a dry-matter content of about 30–50% (Ciba-Geigy Ltd. 1981), and the resulting ingestion dose from pork is about half from OBT and half from HTO. The OBT in grain, which is 88% dry matter, contributes nearly 90% of the total grain ingestion dose. Given the differences in OBT dose contribution from different foods, the importance of OBT to ingestion dose depends on what quantities of what kinds of foods are consumed. An extremely unlikely diet very high in OBT would, at most, give an OBT contribution to dose equal to that of HTO. Thus, conservatively, the maximum total tritium dose from ingestion of vegetables and foodstuffs from the Livermore Valley for 1999 could be 1.0 µSv (0.10 mrem), well below any level of concern.

The dose values for PIN1 shown in **Table 11-1** were calculated in a different manner from those for edible vegetation because it is unreasonable to assume that any person is directly ingesting pine needles. The pine tree is treated as a diffuse source of tritium to the atmosphere via the contaminated transpirational stream. LLNL used an estimated tritium transpiration rate from the tree as input data to the EPA regulatory model CAP88-PC. LLNL modeled air dispersion of the transpired tritium and calculated a resulting dose from inhalation, skin absorption, and potential ingestion from air concentrations at the location of the maximally exposed individual. This total dose is based on the conservative assumptions that 100% of the individual's time is spent at this location and that his/her diet consists exclusively of vegetables with the measured tritium concentration, and meat from livestock fed on grasses with the same concentration. The resulting maximum dose for PIN1 of 1.5×10^{-5} µSv (1.5×10^{-6} mrem) is considerably lower than ingestion doses calculated directly from measured concentrations in vegetation because the tree is only an indirect source of air/vegetation contamination.

No health standards exist for radionuclides in wine. However, all the wine tritium levels were far below drinking water standards. In fact, even the highest detected Livermore Valley value (8.3 Bq/L or 224 pCi/L) represents only 1.1% of the California drinking water standard (740 Bq/L or 20,000 pCi/L). Doses from wine consumption can be calculated according to methods for water ingestion, as described in Appendix A.

Based on the conservative assumption that wine is consumed at the same rate as water (370 L/year or 1 L/day), the annual dose that corresponds to the highest detected 1999 Livermore Valley tritium value in wine is 53 nSv (5.3 µrem). Assuming a more realistic average wine consumption (52 L/year or 1 L/week of wine) and the mean tritium values from the three sampling areas, the annual doses from Livermore, European, and California wines would be 2.2 nSv (0.22 µrem), 1.2 nSv (0.12 µrem), and 0.40 nSv (0.040 µrem), respectively.

The potential ingestion dose from all foodstuffs grown near the Livermore site was realistically well below 1.0 µSv (0.10 mrem) for 1999. This is a factor of 3000 lower than an annual background dose (~3000 µSv or 300 mrem) and a factor of 100 lower than the dose from a typical chest x-ray (100 µSv or 10 mrem) (Shleien and Terpilak 1984). Therefore, although tritium levels are elevated slightly near the Livermore site, doses from tritium ingestion are negligible.

Site 300

In general, LLNL impacts on tritium concentrations in vegetation at Site 300 for 1999 were insignificant. With the exception of vegetation from previously identified sites of contamination, the tritium levels at Site 300 were at or near the limits of detection and comparable to those observed in previous years. The areas where tritium is known to be present in the subsurface soil are well delineated and localized.

The calculated maximum potential annual ingestion dose from vegetation at sampling location EVAP, based on the maximum value of 480 Bq/L (13,000 pCi/L), is 2.3 µSv (0.23 mrem). This dose, based on the conservative modeling assumptions described above, is theoretical, because vegetation at Site 300 is not ingested either by people or by livestock. In comparison, the potential annual dose (also theoretical) from vegetation at all other locations at Site 300 could not be calculated because the median concentration is below the limit of detectability.