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Vegetation and Foodstuff Monitoring

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Introduction

Because pollutants originally released to the soil, air, or water can be transported to vegetation, DOE guidance 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). Sampling and analyzing native vegetation can provide information about the presence and movement of radionuclides in the environment. In addition, vegetation monitoring is important because plants can expose humans to radiation through direct ingestion or through ingestion of products from animals that have eaten plants that contain radionuclides.

Since 1972, vegetation and foodstuff sampling in the vicinity of LLNL and Site 300 has been part of a continuing LLNL monitoring program designed to measure any changes in environmental levels of radioactivity, to evaluate any increase in radioactivity that might have resulted from LLNL operations, and to calculate potential human doses resulting from direct and indirect ingestion of these products. During 1997, LLNL collected and analyzed samples of native vegetation and wine. Potential human doses from these foodstuffs are calculated using the monitoring data and dose models presented in Appendix B. Potential human doses from inhalation of water evaporated into the air from non-edible vegetation are determined using the EPA model, CAP88-PC.

Tritium is the nuclide of major interest in the LLNL vegetation and foodstuff monitoring program because LLNL has historically released tritium to the air both accidentally and in the course of routine operations. Tritium is likely to move into the environment as tritiated water and can be assimilated easily into vegetation and foodstuff. It can contribute to human radiation dose burdens if it is inhaled or ingested directly or indirectly. Although other radionuclides are used at LLNL, our assessments show that only tritium could be present in vegetation in detectable concentrations.

Methods

Our methods for monitoring vegetation and wine are presented in the following sections.

Vegetation

LLNL collects vegetation samples, usually annual grasses, quarterly from fixed locations in the Livermore Valley, San Joaquin County, and Site 300, and then analyzes them for tritium.

Location maps are provided in **Figures 10-1** and **10-2**. These locations have been selected so samples would represent vegetation from: (1) locations near LLNL that could be affected by LLNL operations, (2) background locations where vegetation was

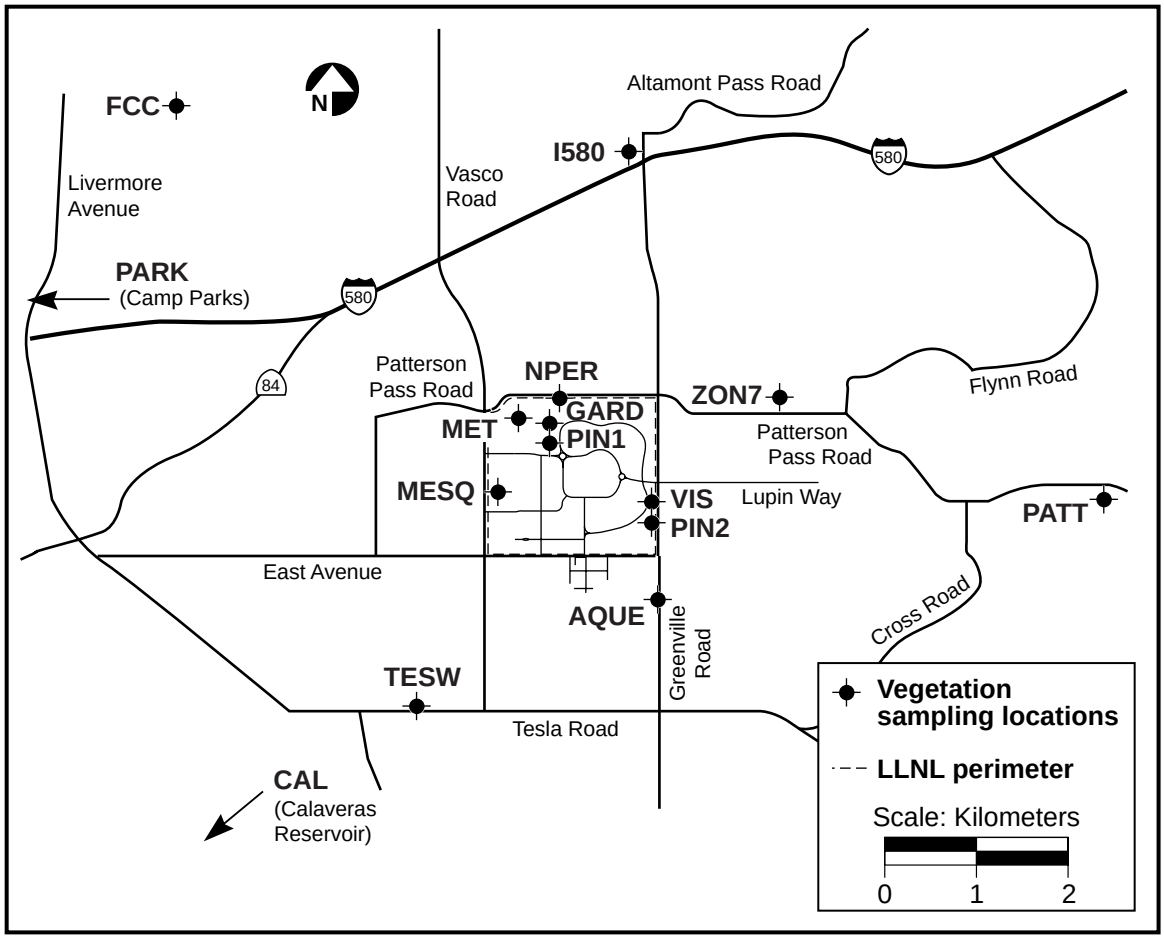

Figure 10-1. Livermore Valley vegetation sampling locations, 1997.

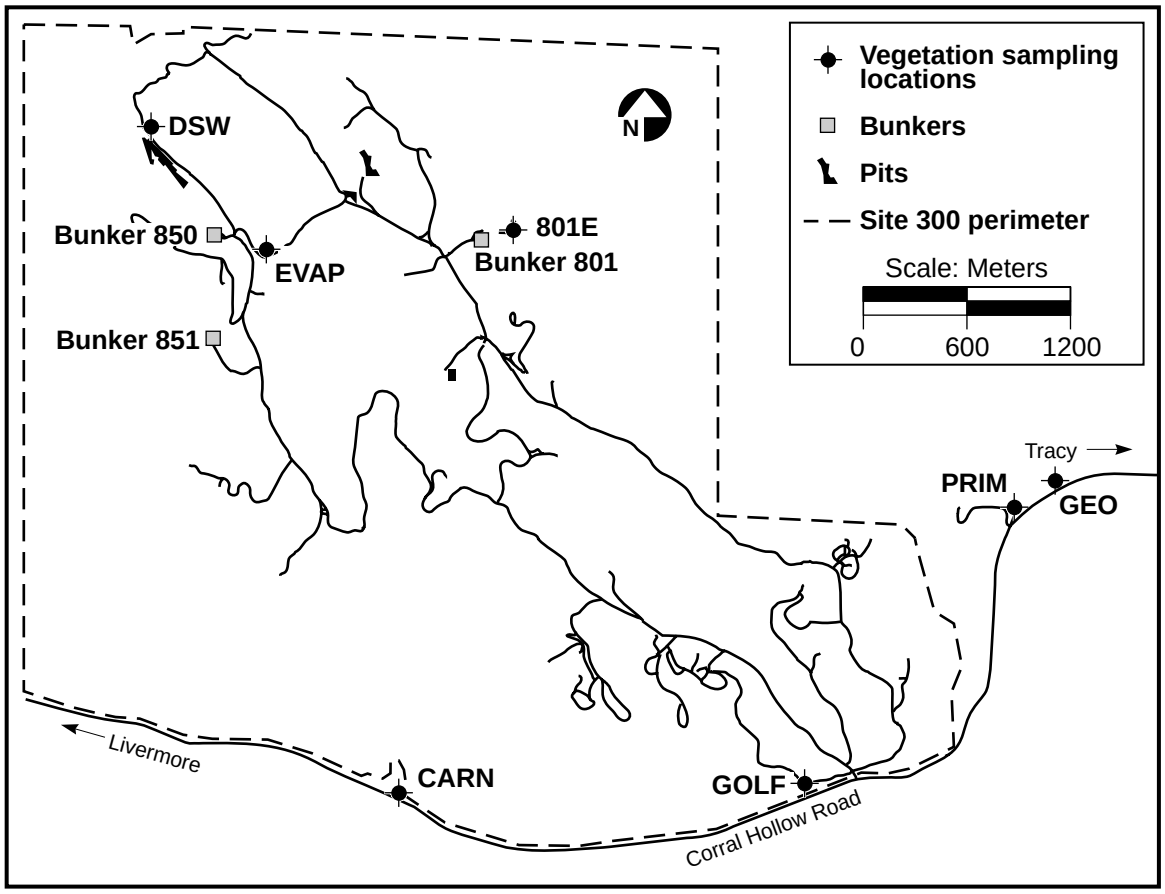

Figure 10-2. Site 300 vegetation sampling locations, 1997.

similar to that growing near LLNL but was unlikely to be affected by LLNL operations, and (3) areas of known or suspected LLNL-induced contamination. Sampling locations PIN1, PIN2, and PRIM were added in the fourth quarter of 1996. PIN1 and PIN2 were added to evaluate the emissions of tritium from a pine tree that is rooted in tritium-contaminated soil (PIN2 is a tree rooted in soil that is not contaminated with tritium). PRIM is located off site and downwind of Site 300.

All vegetation sampling is conducted according to written and approved standardized procedures (Tate et al. 1995). Approximately 10% of the sites are sampled in duplicate to comply with quality assurance protocols.

Wine

Wine is the most important agricultural product in the Livermore Valley, representing an approximately $30 million annual industry. Although the tritium concentrations in all wines are low, the data since monitoring began (in 1977) indicate that Livermore Valley wines contain statistically more tritium than do their California counterparts.

Three types of wine samples were collected and analyzed for tritium concentrations: wine produced from grapes grown in the Livermore Valley, wines produced from grapes grown in California outside the Livermore Valley, and wines produced from grapes grown in Europe (France, Germany, and Italy). The latter two groups were divided into 8 and 13 wine-producing regions, respectively, and were used as comparative samples.

The wine samples were purchased from local retailers in a variety of vintages and reflect the body of wines locally available to the general public during 1997. The resulting analytical data can be used to estimate the potential tritium dose received by consumers during the year of purchase. The 1997 sampling data cannot, however, be used to indicate how LLNL's operations affected wines produced in 1996. Some time—in some cases, several years—will have elapsed between the harvest of the grapes and the release of the vintage. However, wine sample data are decay-corrected to original tritium concentrations (given the number of months that have elapsed between wine production and LLNL analysis) to determine trends and to help determine the impact of LLNL operations during a particular vintage year.

The wine samples were submitted for analysis unopened to avoid airborne tritium contamination. Wines were analyzed for tritium using ^{3}He mass spectrometry in the LLNL Isotope Sciences Noble Gas Mass Spectrometry Laboratory (Surano et al. 1991). This highly sensitive method has a detection limit of less than 0.5 Bq/L (13 pCi/L), and is used to determine the small differences in the tritium content of the samples. Conventional scintillation detection systems typically have detection limits between 5 and 10 Bq/L (150–300 pCi/L); therefore, the differences in the samples would not have been detected had conventional detection methods been used.

Approximately 10% of the total complement of wines was sampled in duplicate, 30% of all the samples were analyzed multiple times, and traceable standards were evaluated to comply with quality assurance protocols.

Results

The results of vegetation and foodstuff monitoring for the Livermore site and Site 300 are presented below.

Livermore Site

Vegetation

Table 10-1 shows summary tritium data for vegetation collected in the Livermore site vegetation monitoring program in 1997 (the individual sampling values are presented in the Data Supplement of this report). In general, the 1997 tritium levels in vegetation were not significantly different than the levels measured in 1996.

Table 10-1. Tritium in vegetation (in Bq/L), 1997.

Location[a]	Detection frequency[b]	Median	Interquartile range	Maximum	Dose (μSv/y)[c]	
					Median	Maximum
Livermore site near locations	19/24	5.3	8.1	45.5	0.025	0.22
Livermore site intermediate locations	10/16	2.5	—[d]	9.5	0.012	0.046
Livermore site background locations	3/12	<1.3	—[d]	7.4	<0.006	0.035
Location DSW at Site 300[e]	3/5	2.6	—[d]	1800	0.012	8.7
Location EVAP at Site 300[e]	2/4	<2.9	—[d]	15.8	0.014	0.1
All other locations at LLNL Site 300	0/17	<1.2	—[d]	<1.5	<0.006	<0.007

a See **Figures 10-1** and **10-2** for sampling locations.

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

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

d Insufficient number of detections to calculate IQR.

e Sampling location in known area of contamination.

The Livermore Valley vegetation locations were put into four groups for statistical evaluation:

- Near—locations at or within 1 km of the Livermore site perimeter. Near locations include AQUE, NPER, GARD, MESQ, MET, and VIS.
- Intermediate—locations in the Livermore Valley further from the site (1 to 5 km from the Livermore site perimeter) but close enough and often

downwind so that they are still potentially under the influence of tritium releases at the site. The intermediate locations were I580, TESW, ZON7, and PATT.

- 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) are in the Livermore Valley but are greater than 5 km from the Livermore site and are generally upwind, so they are unlikely to be affected by LLNL operations.
- Special Study—locations taken to represent a tree rooted in an area of known tritium contamination (PIN1) and a similar tree not rooted in a known area of tritium contamination (PIN2). The locations PIN1 and PIN2 were evaluated separately.

The changes in tritium levels between 1996 and 1997 for the vegetation from within each of the Near, Intermediate, and Far groups were statistically insignificant.

Because the data for tritium in vegetation were lognormally distributed, the means of the logarithms were compared, using the Tukey-Kramer honestly significant difference (HSD) test. This evaluation of the 1997 data showed a significant difference between the Near group and the other two groups; that is, the Near values are significantly different from the Intermediate and Far values, but the Intermediate values are not significantly different from the Far values. **Figure 10-3** shows the historic averages for the three groups. The highest tritium results for individual vegetation sampling locations were found at AQUE and VIS, which are located downwind of the Livermore site and historically have exhibited higher values than other locations.

In 1997, the tritium content of a pine tree growing in a known area of contamination (PIN1) was studied. Our purpose was to provide monthly data for a year, and to use the resulting data to estimate emissions from the tree for compliance with National Emissions Standards for Hazardous Air Pollutants (NESHAPs). At the completion of the year, the tree sampling was coordinated with the quarterly vegetation sampling, and subsequent NESHAPs calculations will be based on the results of quarterly sampling. To obtain a foundation for understanding the contribution of contaminated soil, a second tree that was not growing in tritium-contaminated soil (PIN2) was also sampled. Any effects of LLNL operations on the second tree would be from air deposition. **Table 10-2** provides the data for the monthly sampling of these pine trees. The results for PIN1 are higher than for any other vegetation sampling location, whereas the results for PIN2 are similar to those for the nearby location VIS, which is a routine vegetation monitoring location where annual grasses are sampled.

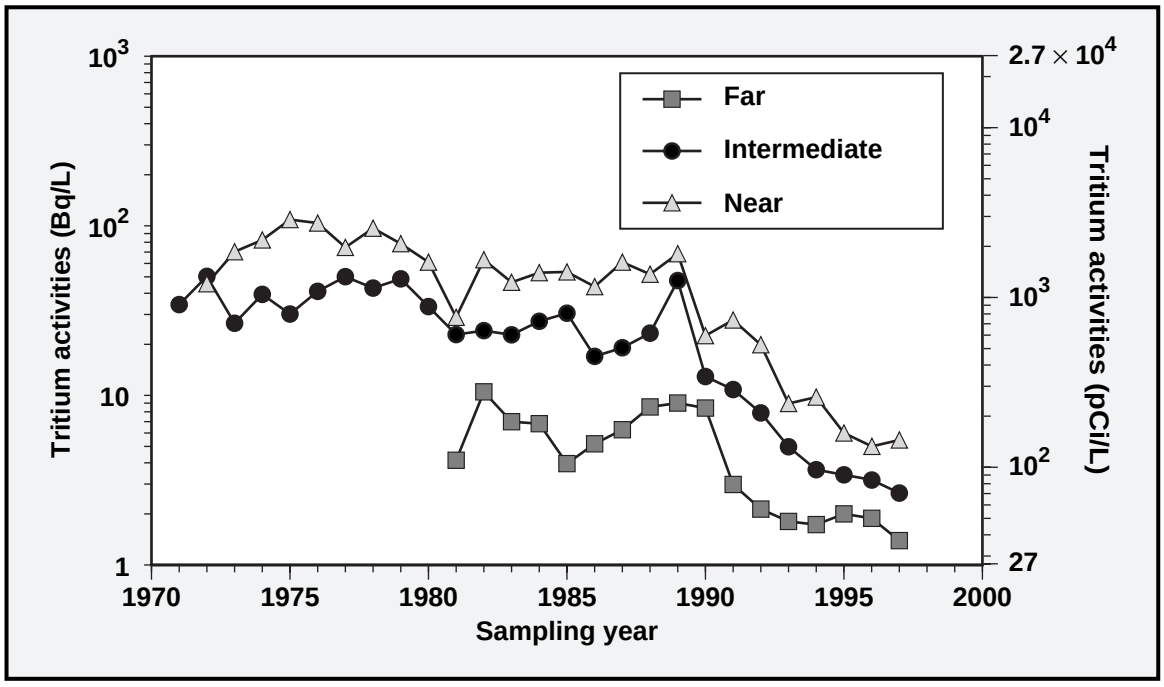

Figure 10-3. Median tritium activities in Livermore Valley vegetation samples, 1971 to 1997.

Table 10-2. Special study of tritium content (Bq/L) in a pine tree growing in a known area of contamination.

Month	Location	
	PIN1[(a)]	PIN2
January	52.9 ± 3.5	8.7 ± 2.3
February	96.6 ± 3.4	24.2 ± 2.1
March	69.6 ± 2.9	13.5 ± 1.7
April	102 ± 3	30.4 ± 2.2
May	128 ± 4	30.9 ± 2.1
June	215 ± 5	26.6 ± 2.0
July	243 ± 5	15.6 ± 1.7
August	274 ± 6	12.5 ± 1.9
September	326 ± 6	17.8 ± 1.8
October	221 ± 4	15.2 ± 1.4
November	215 ± 5	13.4 ± 1.6
December	67.3 ± 2.8	5.7 ± 1.3
Median	**172**	**15.4**
Maximum	**326**	**30.9**
Maximum Dose	**1.7×10^{-5} μSv (1.70×10^{-6} mrem)**	**1.6×10^{-6} μSv (1.6×10^{-7} mrem)**

[a] Sampling location in area of known contamination.

Wine

The results from the 1997 wine tritium analyses are shown in **Table 10-3**. Tritium concentrations were within the range of those reported in previous years, and they remained low in wines from all areas.

Table 10-3. Tritium (Bq/L) in retail wine, 1997.[a]

Region	Detection frequency	Median	Interquartile range	Mean	Maximum	Dose[b] μSv/y (mrem/y)
Livermore Valley	12/12	2.45	1.64	2.89	7.96	0.0026 (0.00026)
California	6/6	0.47	0.19	0.51	0.75	0.0005 (0.00005)
Europe	4/4	1.61	0.75	1.91	3.29	0.0017 (0.00017)

a Wines from a variety of vintages were purchased and analyzed during 1997. The concentrations shown are not decay-corrected to vintage year.

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

The data for the 1997 sampling year were analyzed using analysis of variance (ANOVA). The statistical analyses showed that the mean tritium concentration of the Livermore wines sampled was statistically greater than that of the California (other than Livermore) wines. The statistical analyses also indicated that there was no significant difference between the mean tritium values of the European and California wines sampled or between the Livermore and European wines. Multiple comparison tests indicated that the mean levels of the 1997 sampling year data from all areas were not significantly different from those reported for the 1995 and 1996 sampling years. **Figure 10-4**, which shows the results of the wine analyses by sampling year since monitoring began, also shows that 1997 tritium concentrations are among the lowest for all reported Livermore wines.

During the review of historical data in 1995, it was discovered that the data being reported for the 1977 and 1979 sampling years were averages across multiple sampling years. These data have been corrected in **Figure 10-4**, and are the reason for differences observed when comparing this figure to those published before 1995.

Regression analyses and ANOVA of the wine data when decay-corrected and grouped by vintage year (1996 is the last sampled vintage) showed tritium concentrations have statistically decreased for all regions since 1984 (see **Figure 10-5**). Livermore wines, examined by vintage year, show statistically greater tritium concentrations for the period 1986 through 1996 than both European and California wines. Nevertheless, it is important to note the continued downward trend in the tritium concentrations of

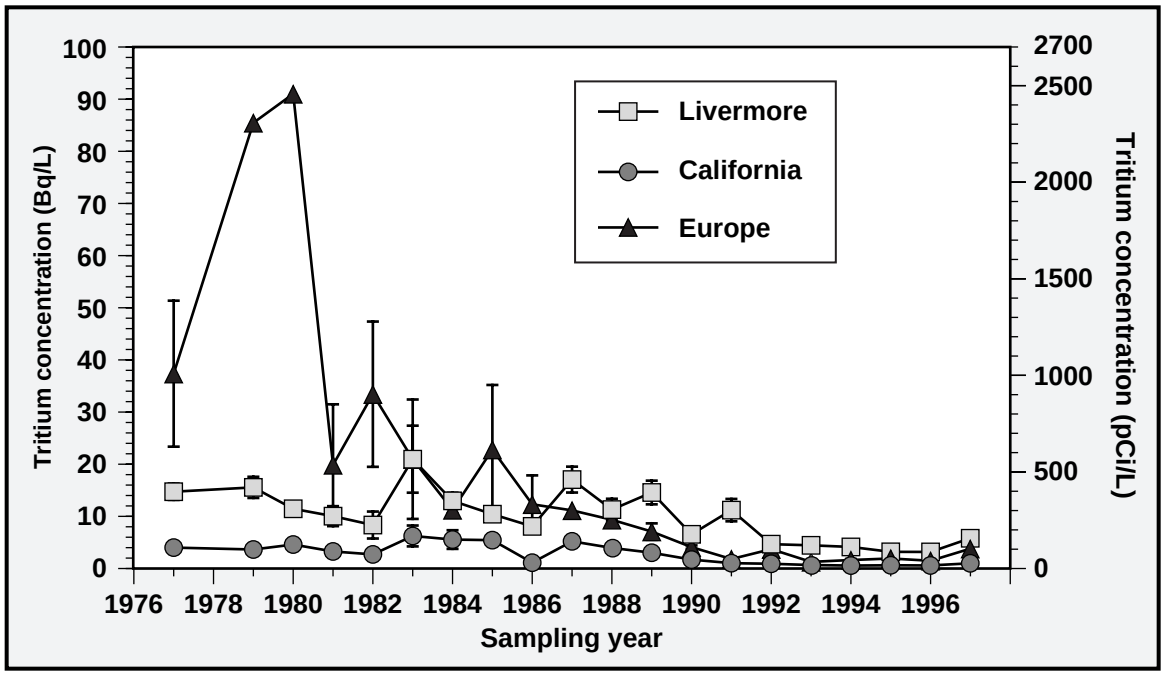

Figure 10-4. Mean tritium in retail wines, 1977 to 1997, plotted by sampling year (error bars are ±1 standard error).

Livermore wines (when decay-corrected and grouped by vintage year) that has been observed since 1984 (when tritium operations at LLNL were scaled down and total amounts of tritium released were reduced).

Site 300

Vegetation

Table 10-1 shows summary tritium data for vegetation collected at Site 300 during 1997. Historic values for tritium at Site 300 sampling locations are shown in **Figure 10-6**. Of the six sampling locations at Site 300, four yielded results at or near the detection limits. Two locations, EVAP and DSW, yielded results above background.

The highest tritium result for a single vegetation sample occurred at the location DSW (see **Table 10-1**). The sample was not the usual native annual grass, but a thorn apple bush (*Datura wrightii*), which, like the stinging nettle (*Urtica dioica)* that in 1996 yielded a high tritium measurement, has a relatively long tap root. The thorn apple bush was

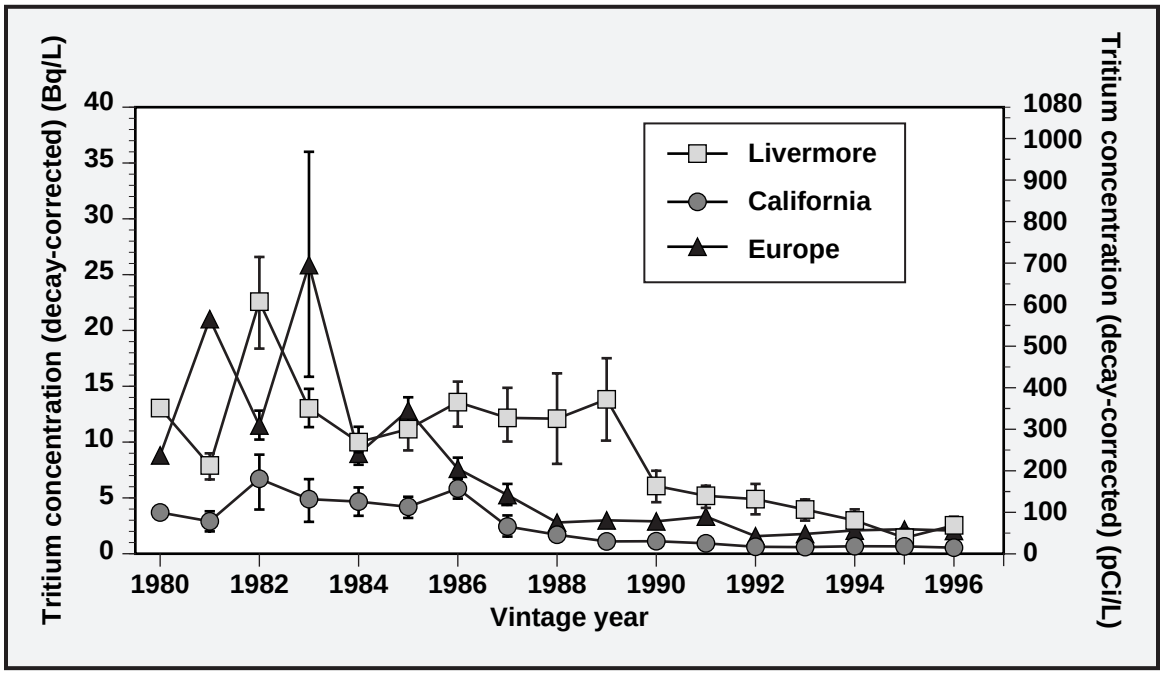

Figure 10-5. Mean tritium in retail wines, vintages 1980 to 1996 values are decay-corrected and plotted by vintage year (error bars are ±1 standard error).

chosen for sampling because it was the only living (i.e., green-colored) vegetation in the area of the sampling location during the summer; its long tap root was evidenced by the plant's greenery, which contrasted dramatically with the brown of the annual grasses.

Tritium has been observed in the vegetation of the DSW sampling location since 1971; it is in an area presently being investigated under CERCLA for tritium contamination of ground water. This sampling location is adjacent to a landfill that contains debris contaminated with tritium from past experiments. The landfill area is under continued investigation for tritium in soil and ground water, as described in reports published as part of LLNL's Environmental Restoration Program (Lamarre 1989a, b, and c; Taffet et al. 1989a and b; Taffet et al. 1991; Carlsen 1991a and b; and Webster-Scholten 1994). The tritium results in vegetation samples that were above background values also occurred at the location EVAP. The location EVAP is near a spring where ground water flows near the surface and evaporates. The ground water in this area is contaminated with tritium which comes from three sources, Pit 3, Pit 5, and the firing table at Building 850 (see discussion of Wells NC7-61 and NC7-69 in Chapter 8, Ground Water). Evaluation of the 1997 data for Site 300 using the Tukey-Kramer HSD test on the logarithms of the data 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. However, if the 1995 and the 1996 data are combined with the 1997 data, a

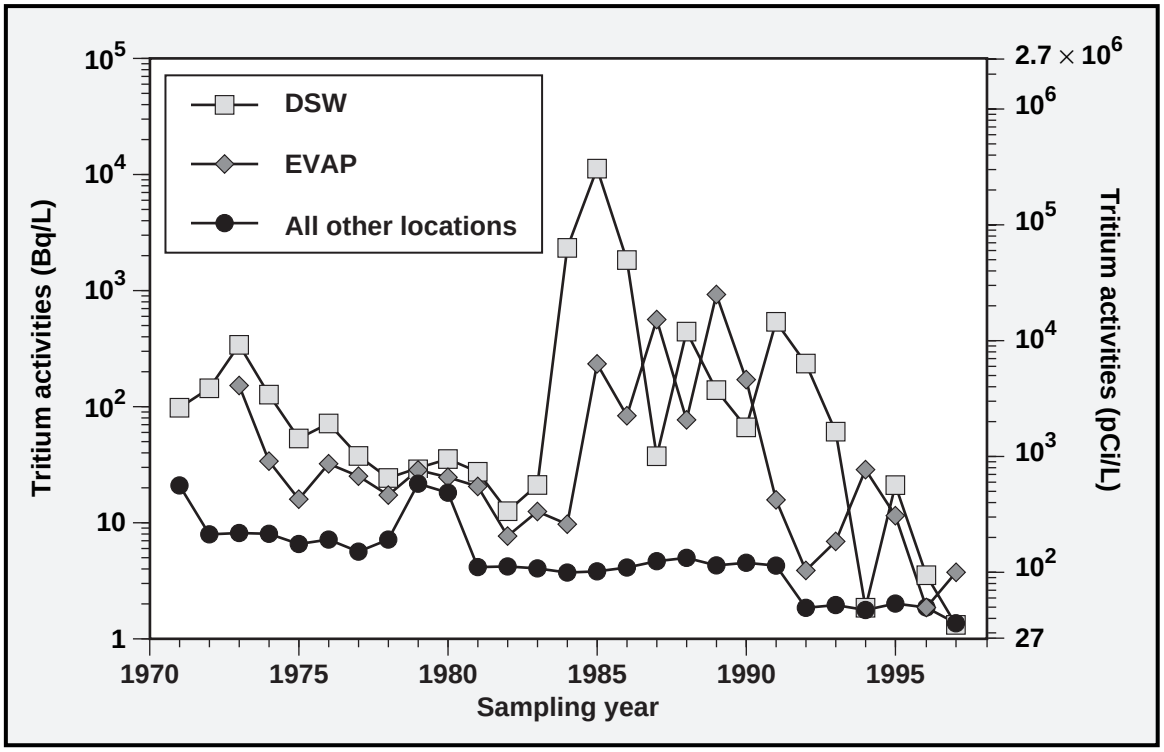

Figure 10-6. Median tritium activities in vegetation at Site 300 sampling locations, 1971 to 1997.

significant difference is found between the set of locations comprising GEO, CARN, GOLF, and 801E, and locations DSW and EVAP. This is a result of the fact that DSW and EVAP are located in areas of known tritium contamination.

Environmental Impact

The environmental impacts of LLNL operations on vegetation and foodstuff monitoring are 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 1997. The effective dose equivalents shown in **Table 10-1** were derived using the dose conversion factors provided by DOE (U.S. Department of Energy 1988) and the dose pathway model from NRC Regulatory Guide 1.109 (U.S. Nuclear Regulatory Commission 1977).

Appendix B provides a detailed discussion of dose calculation methods. The dose from tritium in vegetation is based on the conservative assumptions that an adult's diet consists exclusively of vegetables with the measured tritium concentration, and meat and milk derived from livestock fed on grasses with the same concentration. These assumptions are conservative because most vegetables consumed directly by an adult will not contain tritium at the levels reported (the tritium levels will actually be much lower), nor will the livestock actually consume vegetation with the reported levels of tritium. Based on these conservative assumptions, the maximum potential dose (from ingestion of affected vegetation) for 1997 for the Livermore site is 0.46 μSv (0.046 mrem). The contribution of any organically bound tritium (OBT) is not included in these calculations; they are based only on the tritium in the water fraction of the plant. A conservative estimate of such a contribution would be to assume that the entire plant is organic matter (the actual fraction of plants that is organic matter varies from plant to plant and also varies among the tissues of the plant), that is—all the calculated dose is from OBT—and use that assumption to calculate a upper-bound estimate of the dose from vegetation. Using the ratio of the dose conversion factors of OBT (4.2×10^{-11} Sv / Bq) and water fraction tritium (1.8×10^{-11} Sv / Bq) from International Commission on Radiological Protection Publication 67 (ICRP 1994) of 2.33 to make such an estimate, the maximum potential dose (from ingestion of affected vegetation) for 1997 for the Livermore site would be 1.07 μSv (0.107 mrem), a dose well below any level of concern.

The dose values shown in **Table 10-2** are calculated in a different manner than those for annual vegetation because it is unreasonable to assume that any person or animal is directly ingesting a pine tree. The dose estimates for the pine trees are based on estimates of transpiration of tritium from the trees into the atmosphere; these estimates are used as input data to the U.S. EPA regulatory model CAP88-PC, which models the air dispersion of the transpired tritium and calculates a resulting dose. These doses are also based on the conservative assumptions that an adult's diet consists exclusively of vegetables with the measured tritium concentration, and meat derived from livestock fed on grasses with the same concentration. The resulting dose for PIN1 of 1.7×10^{-5} μSv (1.7×10^{-6} mrem) is considerably lower than the other calculated vegetation doses because the trees are not directly ingested, rather the dose is calculated based on the subsequent deposition of tritium evapotranspirated from the tree.

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 (7.96 Bq/L or 215 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, which are detailed in Appendix B.

The annual dose that corresponds to the highest detected 1997 Livermore Valley tritium value in wine (7.96 Bq/L [215 pCi/L]) is 0.099 μSv (0.0099 mrem), based on the extremely conservative assumption that wine is consumed in the same quantities as water (730 L/year or 2 L/day). Using a more realistic wine consumption factor (52 L/year or 1 L/week of wine from a single area) and the mean tritium values detected in wines from the three sampling areas, the annual dose from Livermore wine would be 0.0026 μSv (0.00026 mrem), from European wine would be 0.0017 μSv (0.00017 mrem), and from California wine would be 0.0005 μSv (0.00006 mrem). Compared with an annual background dose of approximately 3000 μSv (300 mrem), which includes radon, and a 100-μSv (10-mrem) dose from a typical chest x-ray (Shleien and Terpilak 1984), the potential dose from consuming wine from any area is minute. Therefore, although Livermore wines contained statistically more tritium than wines produced in other areas of California, the effects of the tritium are negligible.

Site 300

In general, LLNL impacts on vegetation at Site 300 for 1997 were insignificant. Tritium levels found in the Site 300 vegetation were comparable to those observed in previous years. With the exception of vegetation from previously identified sites of contamination, the levels were low, near the limits of detection. The areas where tritium is known to be present in the subsurface soil are well delineated and localized.

The calculated maximum potential annual dose from vegetation at sampling location DSW, based on the maximum value of 1800 Bq/L (48700 pCi/L), is 8.7 μSv (0.87 mrem). This dose, which would never actually be received by anyone, is about 11.5 times less than a chest x-ray (Shleien and Terpilak 1984). This calculation uses the same conservative pathway modeling assumptions, as described above. In actuality, this dose never would be received because vegetation at Site 300 is not consumed by people or by grazing livestock. In comparison, the calculated potential annual dose from vegetation at all other locations at Site 300 had a median value of <0.006 μSv (<0.0006 mrem; the value is a "less than" value because all measured tritium levels were less than the detection limit). Tritium levels in vegetation at Site 300 will continue to be monitored.