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**Environmental Monitoring of Present and Reconstruction of Past Tritium Emissions from the National Tritium Labeling Facility at the Lawrence Berkeley National Laboratory, California**

Committee to Minimize Toxic Waste

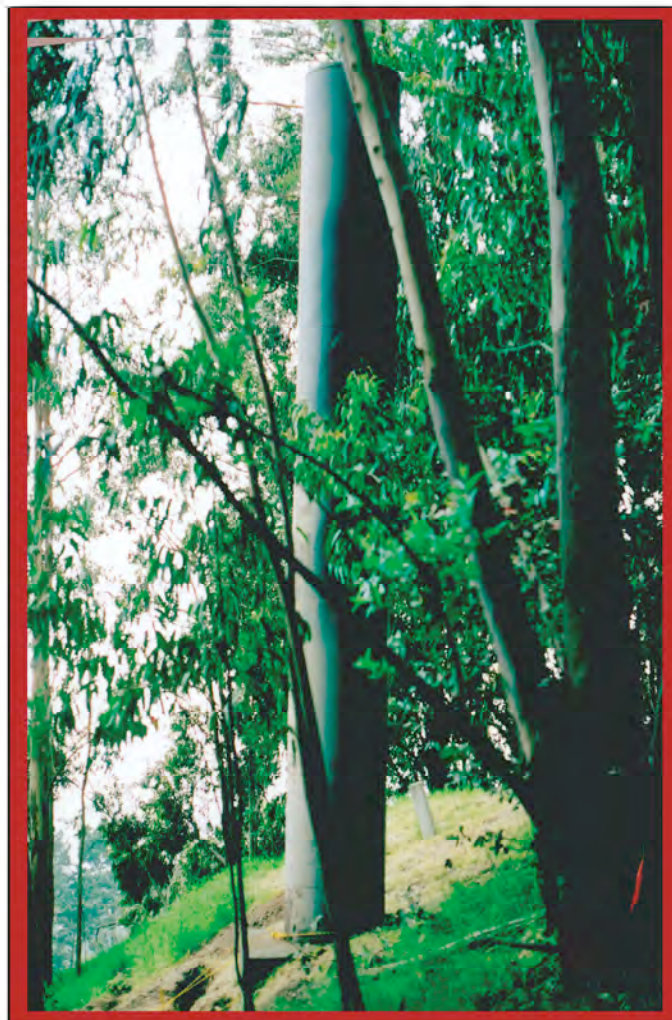
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ENVIRONMENTAL MONITORING OF PRESENT AND  
RECONSTRUCTION OF PAST TRITIUM EMISSIONS FROM  
THE NATIONAL TRITIUM LABELING FACILITY AT THE  
LAWRENCE BERKELEY NATIONAL LABORATORY,  
CALIFORNIA.



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## **Introduction**

The Lawrence Berkeley National Laboratory (LBNL), originally called the University of California Radiation Laboratory, was established on the University of California Berkeley (UCB) central campus in Alameda County in 1932. By 1940, it was relocated to its present site in the steep hills of Strawberry Canyon, east of UCB (Figure 1). The first major facility, the 184-inch synchrocyclotron was built with funds from both private and university sources, and was used in the Manhattan Project in the development of the world's first nuclear bomb. Beginning in 1948 the U.S. Atomic Energy Commission and later its successor agency, the Department of Energy (DOE), funded the lab while it continued to expand its facilities in Strawberry Canyon.

The National Tritium Labeling Facility (NTLF) was located on the eastern edge of the Lawrence Berkeley National Laboratory (LBNL) in Building 75. Just to the north of the NTLF is the Lawrence Hall of Science, a popular children's science museum that is visited by thousands of children every year (Figure 1). The NTLF was established in 1982 as a National User Facility for the labeling of compounds used in pharmaceutical and biological research. The tritiation of these compounds involved an inefficient exchange process that required the use of large quantities of tritium (the radioactive form of hydrogen), i.e., 100+ curies of tritium per tritiation and therefore thousands of curies per year. Commercial tritium facilities can not produce tritiated compounds in this way because Federal Regulations limit them to a total inventory of only 150 curies of tritium. Department of Energy Facilities, in contrast, can have much larger inventories. The NTLF's inventory limit, for example, was 15,000 curies.

The amount of tritium shipped out of the NTLF in tritiated compounds was a very small fraction of the total tritium used. The balance had gone out either as waste or has been emitted via the facility's two stacks. Tritium emissions from the NTLF's main stack were monitored on a weekly basis with a silica gel sampler and in real-time with an Overhoff system. The silica gel data were used

to estimate the annual dose to the public to ensure compliance with National Emissions Standards for Hazardous Air Pollutants (NESHAPs) requirements. Dose estimates were modeled with CAP88, an EPA approved computer program.

The public first became aware that tritium was being released into the environment by the NTLF in the early 1990's. However, at that time very little was known about the nature of the problem. In 1996 community concerns regarding the NTLF began to escalate when Dr. Leticia Menchaca, an LBNL researcher who had been investigating tritium in the area close to the NTLF, reported that the levels were higher than had been previously reported. Community concern was further increased when Susan Monheit, another LBNL employee, produced a master's thesis which showed that the levels of tritium in plants, soils, and rain near the NTLF stack were unexpectedly high. She also reported above background tritium activity in rainfall samples collected 3.8 kilometers from the stack. In the same year, some community members independently collected water samples from Eucalyptus trees between the NTLF stack and the Lawrence Hall of Science and also found that tritium activity levels were unexpectedly high. In addition, Dr. Menchaca analyzed the organically bound tritium activity in Eucalyptus leaves collected in the area around the NTLF stack and again found very high levels, especially in the area between the stack and the Lawrence Hall of Science. Shortly afterwards both Dr. Menchaca and Ms. Monheit were laid off at LBNL.

Another cause for community concern was the publication of a paper by Dr. Tore Straume (1995) in which he pointed out that there is an inverse relationship between the energy level of radiation and its biological effectiveness. He also stated that the biological effects of tritium beta rays were likely to be substantially larger, i.e. 4 to 5 times larger, than previously thought.

In September of 2001 the National Institutes of Health announced that they would discontinue funding the National Tritium Labeling Facility (NTLF). Shortly afterwards Lawrence Berkeley National Laboratory (LBNL) announced

the Facility would be closed by the end of December 2001 and that the process of decommissioning and clean up would begin afterwards. However, the NTLF remained operational through mid-June 2002, while the oxidation of tritiated mixed waste continued. The decision to close the NTLF at LBNL was welcomed by community members who had been concerned about tritium emissions since the early 1990's. In fact in 1998 the US Environmental Protection Agency (US EPA) performed a Superfund reassessment of LBNL concluding that "Based upon a preliminary Hazard Ranking System score, US EPA has determined that LBNL is eligible for the National Superfund Priorities List" for cleanup, due to tritium in air, soil, groundwater, and surface water.

In June of 2005 a National Academy of Sciences panel, formally known as the Committee on Biological Effects of Ionizing Radiation (BEIR), concluded that there is no exposure level found below which dosage of radiation is harmless. The preponderance of scientific evidence shows that even very low doses of radiation pose a risk of cancer or other health problems. The National Academy of Sciences panel is viewed as critical because it addresses radiation amounts commonly used in medical treatment and is likely to also influence the radiation levels that the government will allow at abandoned and other nuclear sites.

The research effort reported on here has three primary objectives: 1) to monitor tritium activity levels in rainfall near the Lawrence Hall of Science, and creeks draining the watersheds close to the NTLF stack; 2) to date wood samples from Eucalyptus trees growing between the NTLF stack and the Lawrence Hall of Science; and, 3) to determine the organically bound tritium content of the dated samples as a means of reconstructing tritium emissions from the NTLF Stack. These three objectives are covered in Part A, Part B, and Part C of this report. The complete report is posted on the following web site:

<http://www.cmtwberkeley.org>.



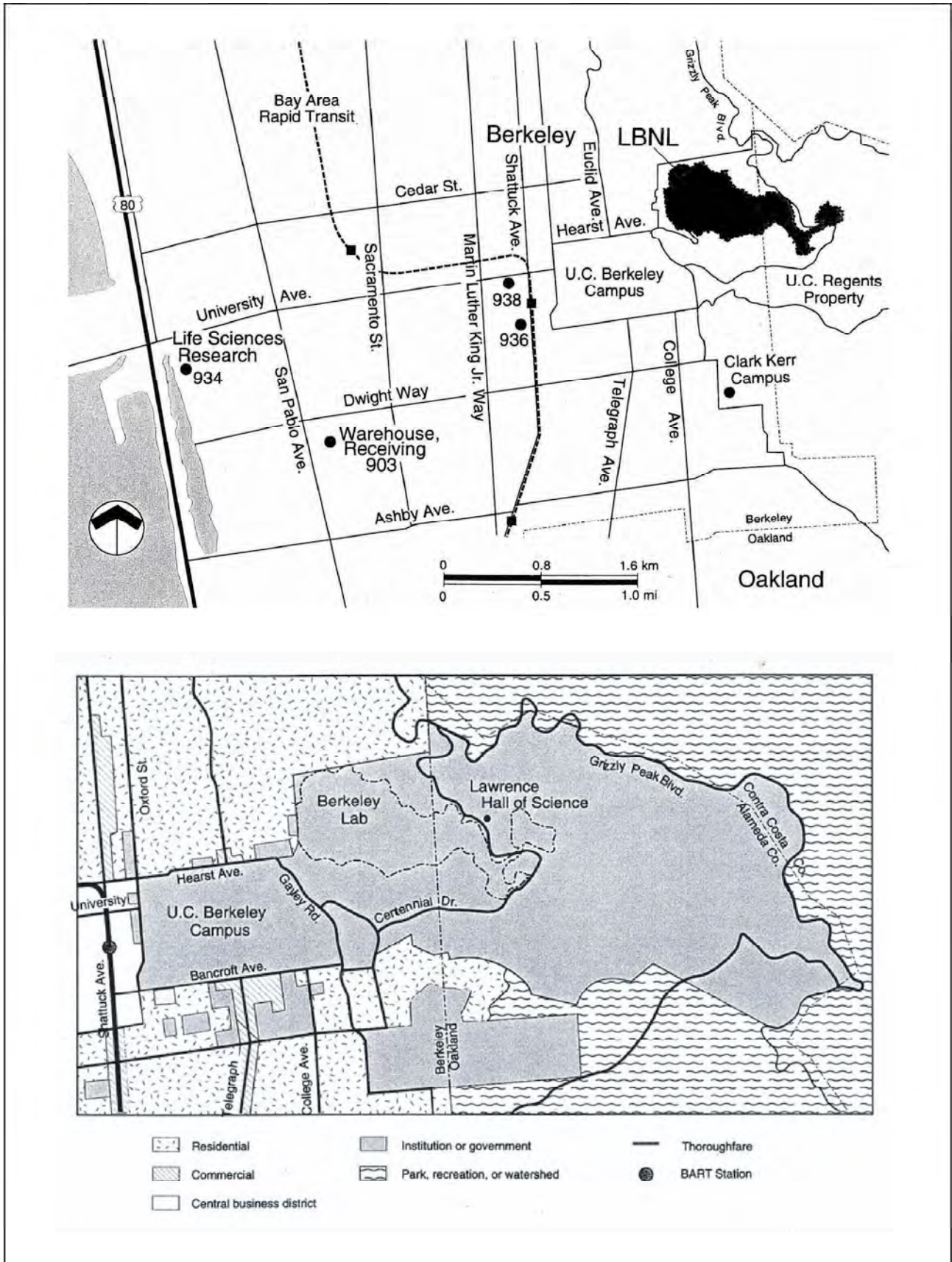


Figure 1. Vicinity and adjacent land use. Source: LBNL RCRA Facility Investigation Report, 2000.

## **Part A: Tritium in Rainfall and Creek Water**

### Tritium in Rainfall

Rainfall in the San Francisco Bay area is primarily associated with mid-latitude storm systems that pass over the area during the winter months. The rainfall year is therefore defined differently from the calendar year; it begins on September 1st and ends on August 31st. Individual storms may produce rainfall amounts that vary from a few millimeters to several centimeters depending upon the size and intensity of the storm system and the time it takes to pass through the area. Also, the number of storms affecting the Bay area varies from year to year.

Because of the inherent variability of rainfall events both in terms of timing and magnitude, tritium activity in rainfall is not used by LBNL to determine compliance with NESHAPs standards. However, LBNL does collect rainfall samples and analyzes them for tritium activity. These activity levels are routinely reported in the Annual Site Environmental Reports. Other DOE Facilities also measure tritium in rainfall as do Nuclear Facilities in other countries. These data are sometimes used to calculate the fallout or "washout" of the radioactive isotope, for example, in picoCuries/per square meter/per unit time. These data obviously do not represent the total amount of tritium emitted from a stack. Rainfall passing through an emissions plume is unlikely to absorb all the tritium within the plume, nor is the tritium activity in the rainfall likely to be the same as the tritium activity in the water vapor in the plume. Tritium activity in rainfall samples can therefore be regarded as minimum estimates of tritium activity in the plume. It should also be emphasized that the washout of tritium by rainfall represents only a small fraction of the tritium emitted from a stack because most of the time it is not raining.

### *Sampling Strategy*

Because of the concern about tritium exposure at the Lawrence Hall of Science we established two rainfall sampling stations (Station 1 and Station 2) approximately half way between the NTLF emissions stack and the Lawrence Hall of Science. The stations were approximately 50 and 60 meters to the north of the stack (Figure A-1). The sampling containers, 1000 ml plastic beakers, were placed on the ground and protected by branches. Initially, we intended to collect samples after every rainfall event but later simplified the process by collecting samples on the weekend of each week in which rainfall occurred. Individual samples were transferred into glass or plastic bottles and labeled as to sample location, time, date, and the approximate amount of rainfall in the sample. The plastic beakers were wiped clean after each sample collection and put back in their original position.

### *Sample Preparation*

Many of the rainfall samples collected were yellow or light brown in color presumably because they contained organic compounds dissolved from Eucalyptus leaves in the canopy. All samples submitted for tritium analysis were therefore distilled. More specifically, each rainfall sample was split into three sub-samples and transferred to plastic bottles: one sub-sample was left untreated; the second sub-sample was filtered; and the third sub-sample was filtered and distilled.

Two distilled samples (5 ml) for each collection from each sampling station were then transferred to glass scintillation vials and sent for tritium analysis to Carbon 14 Centralen, a subdivision of DHI Water and Environment, located in Denmark. All samples sent to Denmark were coded to ensure that replicates would be analyzed as a blind test.



### *Sample Analysis*

The tritium activity in the rainfall samples was determined by liquid scintillation on a Packard Scintillation counter. Prior to counting a scintillation cocktail was added to each 5 ml sample. Counts were calibrated by comparison with the activity of standards and blanks.

### *Results and Discussion*

The results of the tritium analyses of rainfall samples are presented in tabular form in Appendix A-1. Activity levels were "corrected" for radioactive decay by assuming that all samples for a rainfall season were collected on January 1st of that season. Samples plus replicates were analyzed for the three rainfall seasons. Activity levels for each station for the three rainfall seasons are shown separately in Figures A-2, and A-3. Scattergraphs showing the correlation between samples from the two sampling stations and between the replicate samples from each station are shown in Figures A-4 and A-5.

It is reassuring to see the activity levels for the two sampling stations are very similar (Figure A-4). Station 1 is ca. 50 meters from the NTLF stack and Station 2 ca. 60 meters from the stack. The close correlation rules out the possibility that samples from one station or another were disturbed prior to collection. Also reassuring is the close correlation between the replicate "blind" samples for each sampling station (Figure A-5). This positive correlation is evidence that the Carbon 14 Centralen scintillator counts are internally reliable.

As far as the reliability of the reported activity levels is concerned, we might point out the annual average values compare reasonably well with tritium in air activities reported by EPA for the Lawrence Hall of Science Sampler located some 50 meters north of our sampling sites. If we assume that the average water vapor content of the air at the Lawrence Hall of Science was  $10\text{ml}/\text{m}^3$  this is equivalent to our rainfall activities of ca. 8,000 pCi/L.

One obvious implication of the 1999-2002 tritium in rainfall data is that the tritiated water (HTO) emissions from the NTLF were significantly lower during this time period than during the early 1990's. During the fall of 1994 Susan Monheit collected 18 rainfall samples from an area within 100 meters of the NTLF stack and reported tritium activity levels ranging from 10,400 to 239,000 pCi/L with an average of 77,300 pCi/L. If we assume this average was typical for that year the implication is that tritium in rainfall activities dropped by roughly 90 percent between 1994 and 2002. In part, this undoubtedly reflects the reduction in tritiations reported by LBNL for the same period. This reduction in NTLF operations was in itself the result of increased public and regulatory scrutiny of the Facility since the mid-1990's.

One important question is whether or not the tritium in rainfall activity reported here is what would be expected given LBNL's annually reported emissions. Even though rainfall samples are obviously only a partial sampling of total emissions, they can be used to make a rough estimate of average air concentrations in the area between the NTLF Stack and the Lawrence Hall of Science. These estimates can then be compared with the CAP88 annual averages calculated on the basis of LBNL's reported emissions and weather data.

#### *Comparative Analysis with LBNL Reported Tritium in Rainfall Data*

LBNL collects rainfall samples for tritium analysis on a monthly basis at three stations. Station ENV-75 is the station closest to the NTLF. In Figure A-6 we plot ENV-75 data for the period November 1999 through December 2001. Also shown are our rainfall data plotted as monthly averages as a basis for comparison. The difference between the two data sets is striking. In part, this may reflect the fact that Station ENV-75 is located upwind of the NTLF stack whereas our sampling stations are downwind. Whatever the reason, it is clear that ENV-75 does not reflect the concentration of tritium in rainfall at the location of primary concern: the location of the maximally exposed individual (MEI), i.e., a person at the Lawrence Hall of Science.

## Tritium in Creek Water

The Lawrence Berkeley National Laboratory is located in the Strawberry Creek Watershed with elevations ranging from 500 to 1,100 feet above sea level. The site is within the jurisdictional boundaries of Berkeley and Oakland. Two major east-west trending creeks, Strawberry Creek and the North Fork of Strawberry Creek (also known as Blackberry Creek), have perennial flow within the site. They drain respectively through Strawberry and Blackberry Canyons through the UC Berkeley Campus and the City of Berkeley into San Francisco Bay. Many of the streams and creeks at and near the site flow at varying intensities throughout the year. The main branch of Strawberry Creek is not within LBNL boundaries, yet seven of its north-south trending tributaries that flow southward, do drain from LBNL. These include Cafeteria Creek, Ravine Creek, Ten-inch Creek, Chicken Creek, No Name Creek, Banana and Pineapple Creeks, the latter two flow into Botanical Garden Creek, which flows into the central reach of mainstream Strawberry Creek, outside the LBNL boundary (Figure A-7).

In September of 1995, the California Department of Health Services (DHS) Environmental Management Branch released an Agreement in Principle (AIP) Annual report. The Report identified LBNL's National Tritium Labeling Facility (NTLF) as a major concern for radioactive contamination in the environment. The Report states:

"This facility (NTLF) handles kilocurie quantities of tritium ( $^3\text{H}$ ) to label a variety of molecules that are subsequently employed in chemical, pharmaceutical, and biomedical research. It is conceded that releases from the tritium-stack as well as fugitive releases from Building 75 are the primary source of tritium at LBNL. Air-fall, rainout, and possibly transport in fog impacts soil, groundwater, and surface water. There is an area of tritium contaminated groundwater in the vicinity of building 75. The Quarterly Progress Report, First Quarter FY 1992, (May 1993) reports sampling ten hydraugers, one immediately down-slope from NTLF, reportedly contained 32,000 pCi/L of tritium."

The AIP Program collected and analyzed surface water samples, which demonstrated that tritium is detectable in surface water around LBNL. The AIP further states:

“One recent investigation, by Leticia Menchaca (LBNL), analyzing for tritium in transpired vapor from plants on LBNL suggest {sic} that there may be significant amounts of tritium in the upper, non-saturated, soil strata. It appears that there may be sufficient evidence to suggest that there may be more tritium in the environment than previously suspected. There are apparently no validated explanations for the appearance of tritium in streams not obviously associated with NTLF.” (See Table A-1).

Location	AIP Results (pCi/L)	AIP Duplicate Results (pCi/L)	LBNL Results (pCi/L)
Blackberry Creek	3335 +/-255		
Claremont Creek	< 328		
Wildcat Creek	1147 +/-218	944 +/- 214	
Lower Strawberry	5902 +/- 294		
Upper Strawberry	< 328	< 328	

Table A-1. Comparison of Tritium Levels from Split LBNL Surface Water Samples. Collection Date June 15, 1995 (Table LBNL -6c, AIP Report, 1995).

During the above referenced investigation, tritium concentration in rainwater was detected as high as 239,000 pCi/L and 197,946 pCi/L in transpired water vapor from trees near the University of California’s Lawrence Hall of Science. See *Summary Tables of Environmental Tritium Measurements, Historical Maximum Result, at Berkeley Lab, April 15, 1997*, which is included here as Appendix A-2. By 1997 the tritium groundwater plume in the Chicken Creek watershed had crossed the LBNL southern boundary. By agreement with the UC Regents the LBNL fence line in that area was moved further south.

US EPA’s Superfund Technical Assessment (July 1998) further stated:

“Residual tritium from labeling activities, conducted at NTLF are {sic} released through the NTLF stack as gaseous tritium and tritiated water vapor, which disperse from the stack and settle to the ground. Ambient air samples collected on and off the LBL site have contained tritium in concentrations that exceed EPA’s cancer risk screening concentration. Tritium also has migrated to groundwater, surface water, soil and soil water both within LBL boundaries and off site.”

### *Sample Collection and Preparation*

In addition to the 1995 California Department of Health Services tritium measurements from Blackberry Creek, in 1993 LBNL had measured tritium concentration of up to 18,100 pCi/L in Chicken Creek. The headwaters of Blackberry Creek are located downwind from the NTLF stack, and the Chicken Creek watershed is directly downstream from the NTLF. We therefore chose these two creeks to be the main sampling sites in our study. The Chicken Creek sampling site (Station CC) is located to the south of the LBNL property at the UC Strawberry Canyon Center. Just below the sampling site the creek enters a culvert under Centennial Drive. The Blackberry Creek sampling site (Station BC) is located in a North Berkeley residential neighborhood at Le Roy Street and Le Conte, where the creek flows day-lighted under a bridge (Figure A-8). Samples were collected at both sites at the end of each month. Additional samples were also taken from both creeks on the LBNL property when LBNL invited community members to participate in their sampling program. On these occasions split samples were taken. Creek water samples were collected in 500 ml bottles from the center of the actively flowing creek and transferred to the lab for distillation to eliminate any organic matter/debris, prior to shipment to Denmark for tritium counting.

### *Results and Discussion*

Fifteen of our creek water samples taken during the period December 1999 through March 2002 had tritium activities above the US EPA's Cancer Risk Screening Concentration (CRSC) of 600 pCi/L. In contrast only 4 of LBNL's Environment, Health and Safety Division's (EH&S) samples did (Appendix A-3). Furthermore, in 2003 LBNL's on-site sampling of two tributaries of Chicken Creek produced tritium levels as high as 1756 pCi/L. This indicated that there was a seep of contaminated groundwater into Chicken Creek with concentrations three times the CRSC level (Figure A-9).

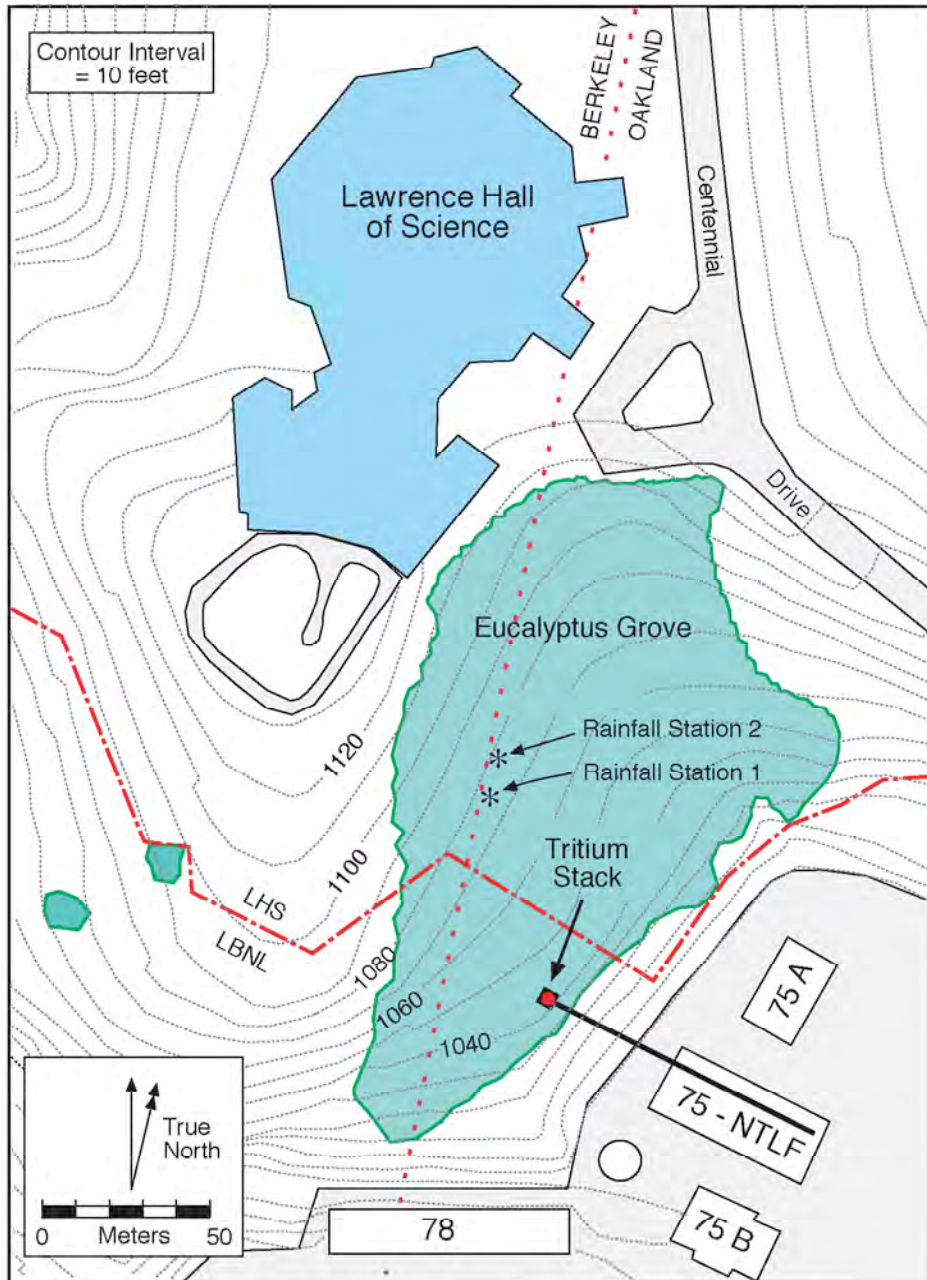


Figure A-1. Topographic map showing the location of the rainfall sampling stations.



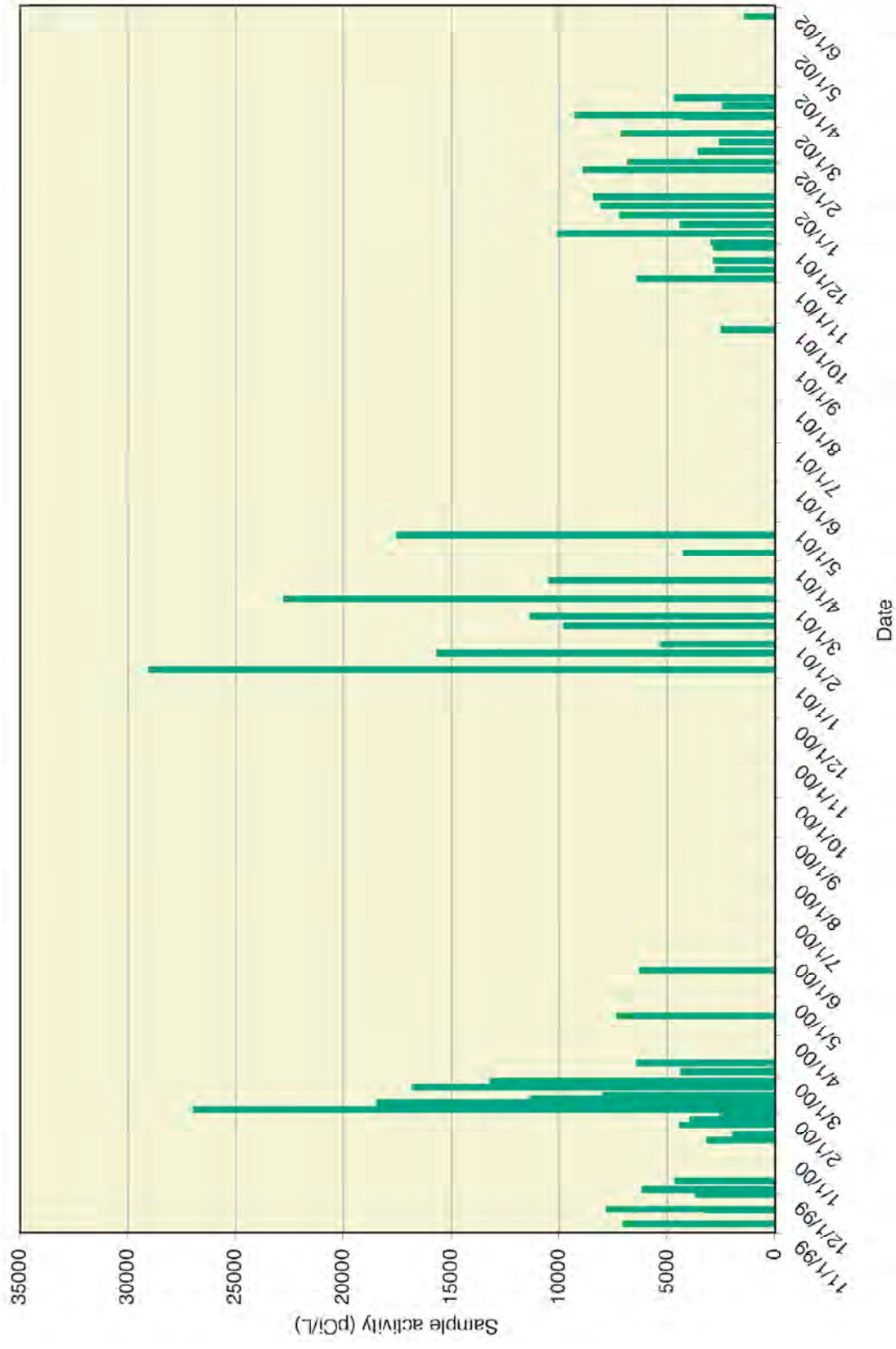


Figure A-2. Tritium activity in rainfall at Station 1 (1999-2002).

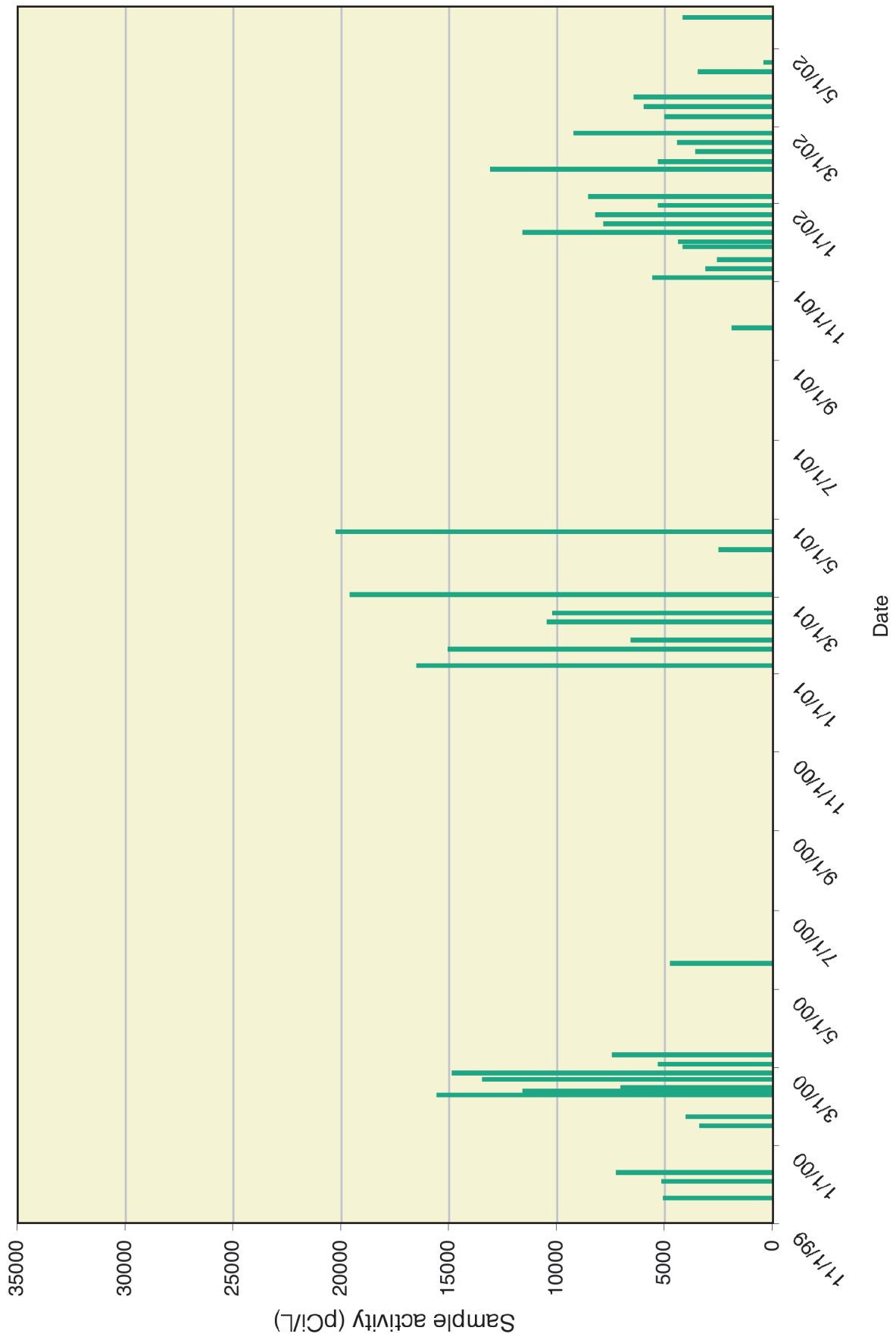


Figure A-3. Tritium activity in rainfall at Station 2 (1999-2002).

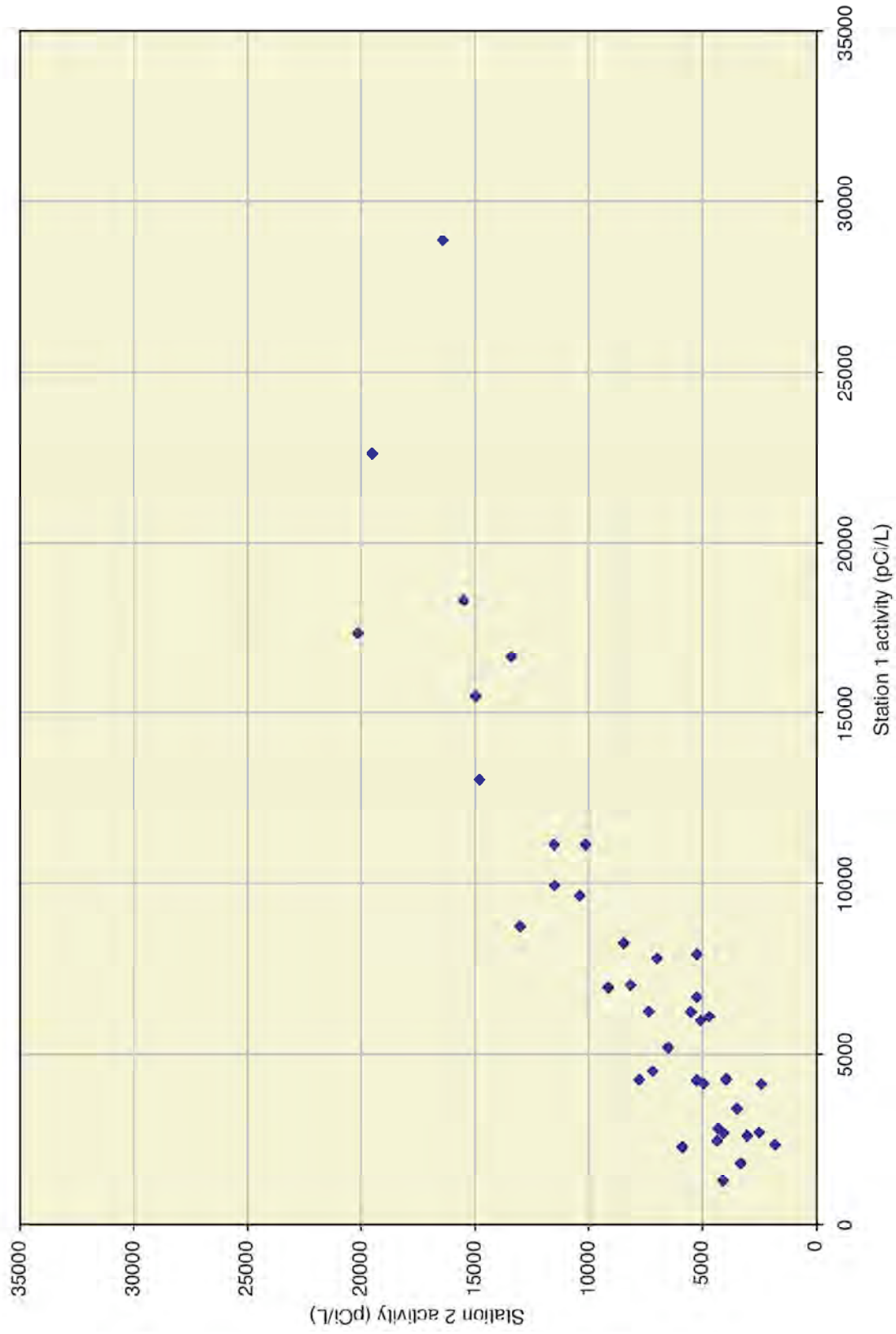


Figure A-4. Correlation between sample means at Station 1 and Station 2.

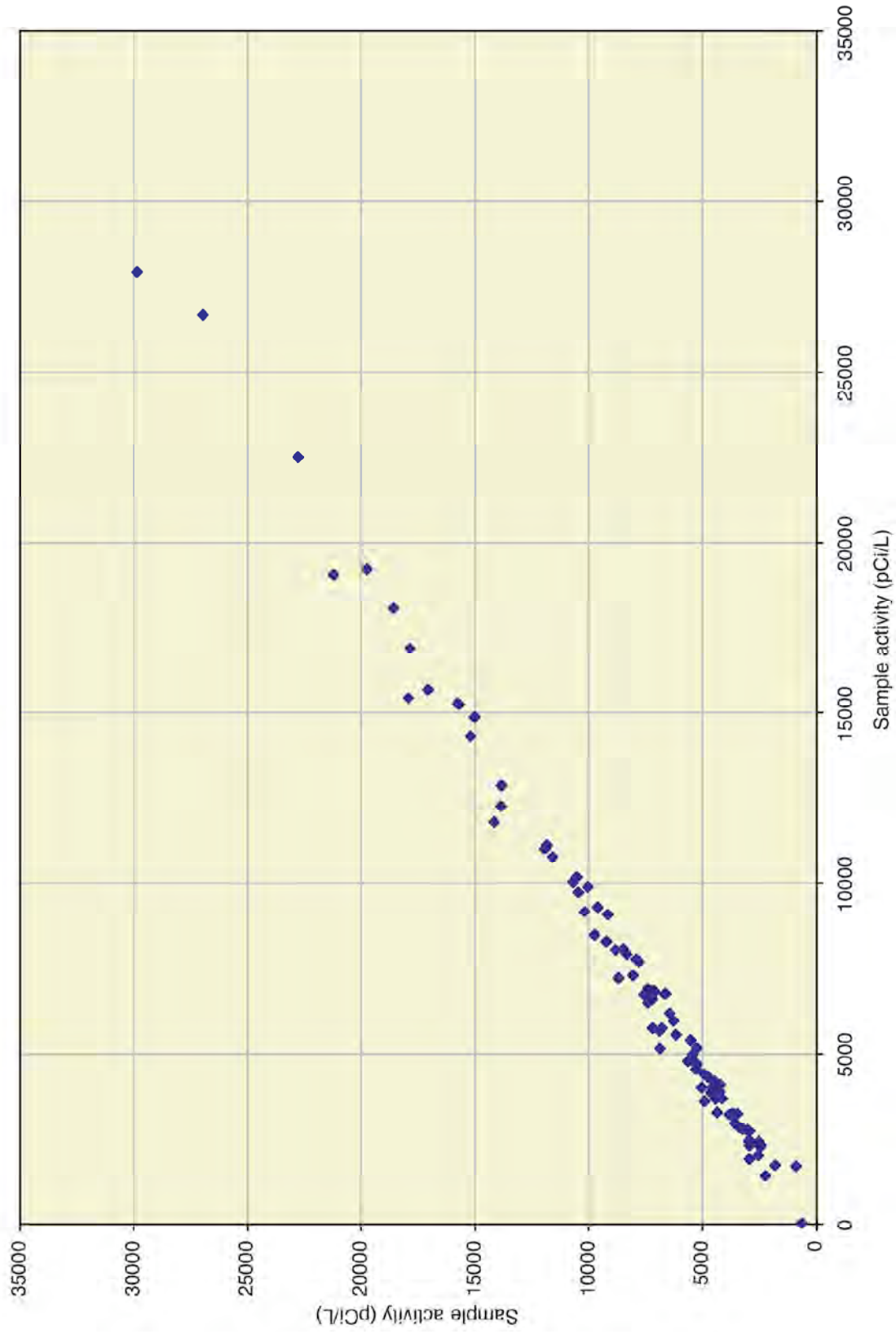


Figure A-5. Correlation between samples and replicates at Station 1 and Station 2.

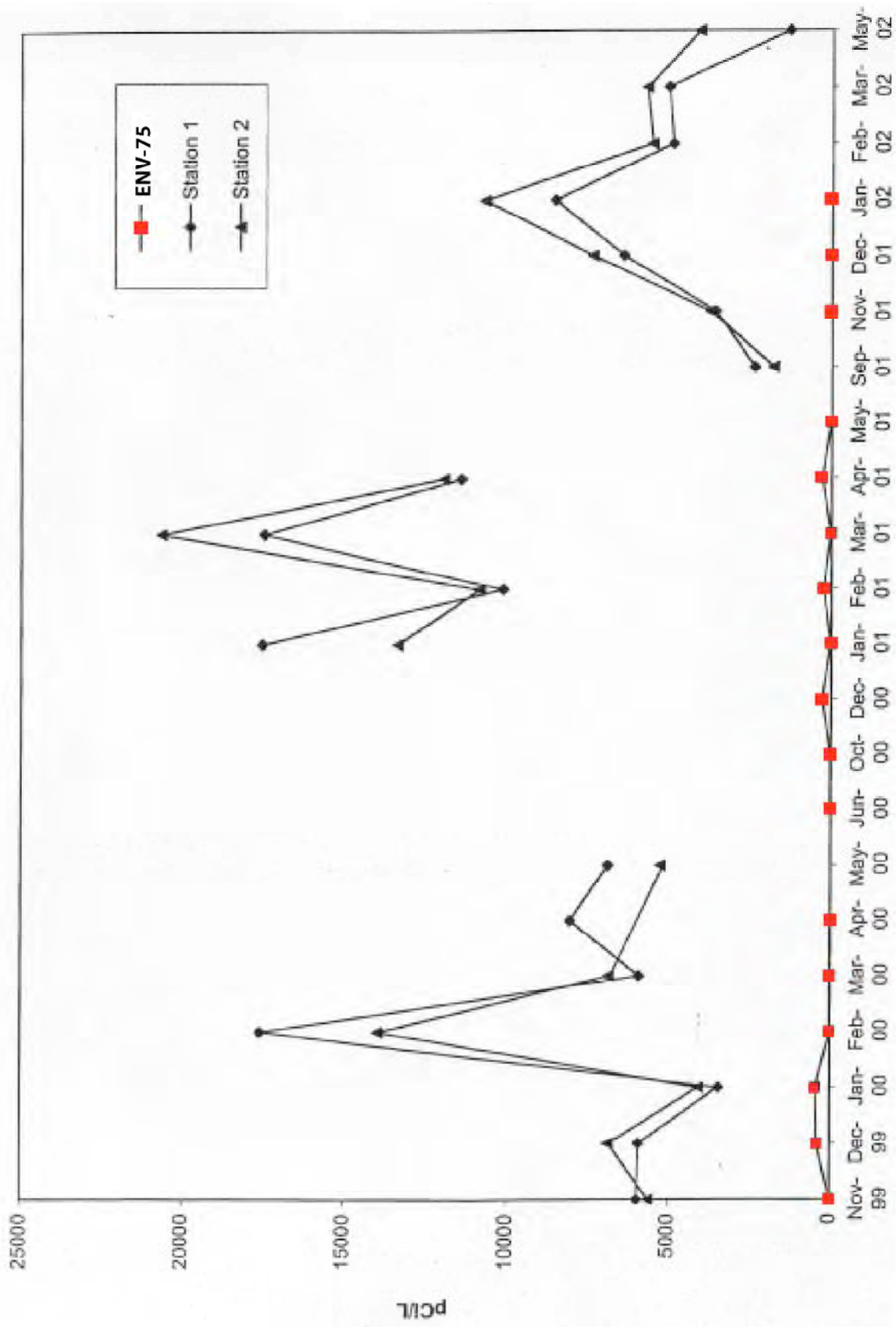


Figure A-6. Tritium activities in rain at LBNL's Station ENV-75 and Stations 1 and 2



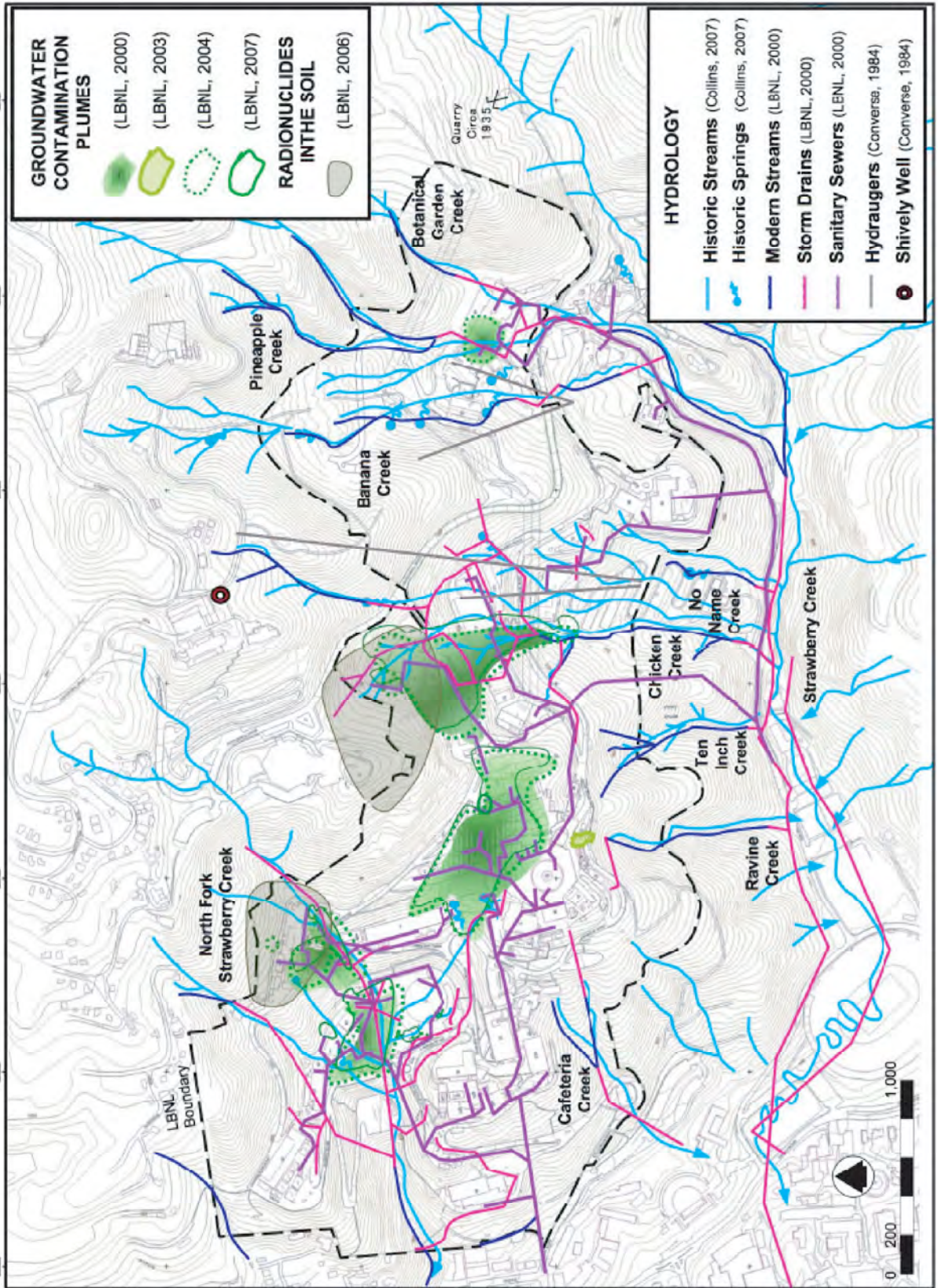


Figure A-7. Modern and historic drainage networks at LBNL in relation to groundwater contamination plumes





Figure A-8. Creek water sampling locations, Stations CC and BC

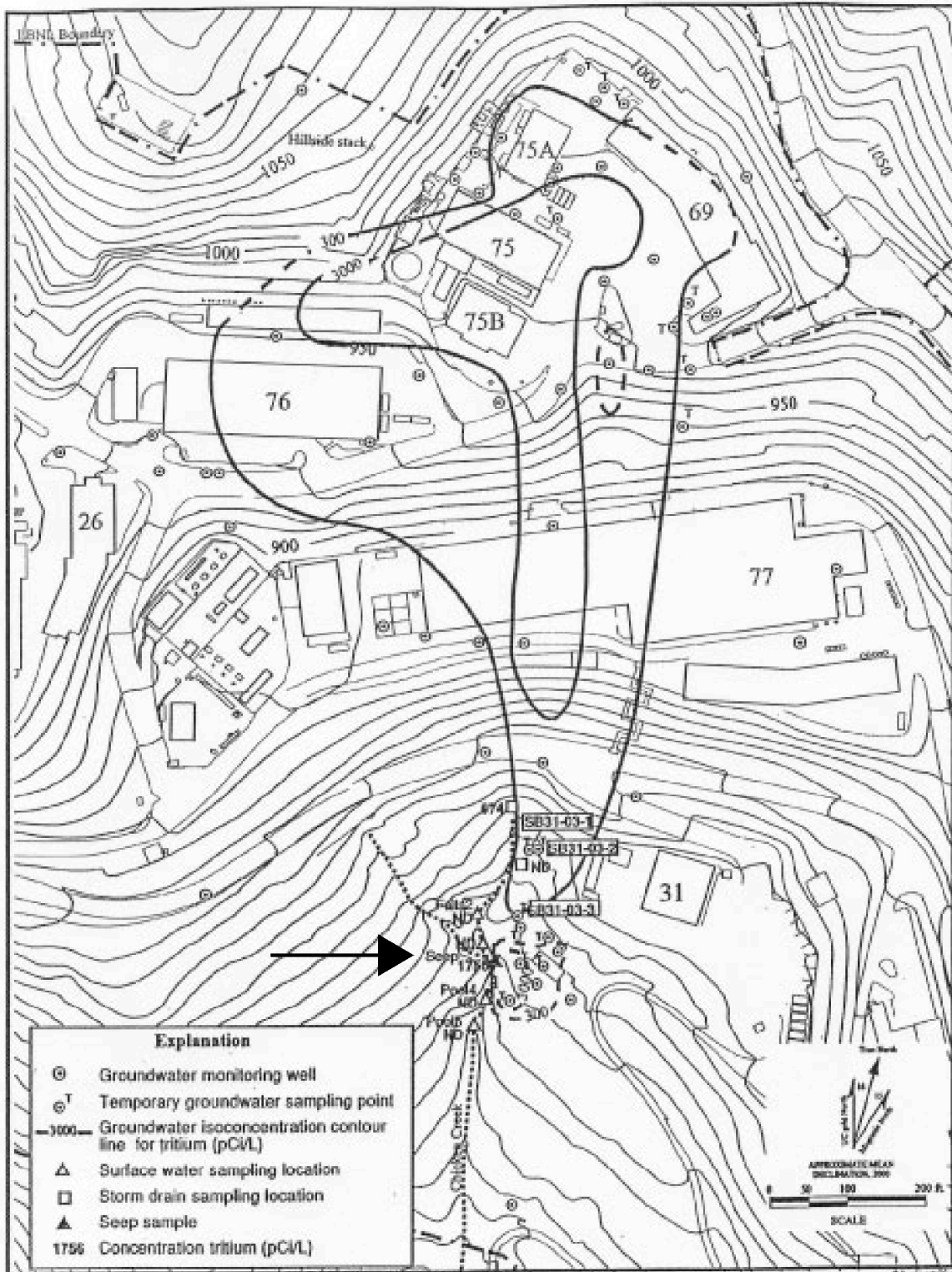


Figure A-9. Arrow indicates source of tritiated ground water seeping into Chicken Creek  
 Source: LBNL ERP Program II QT FY2003

## **Part B: Methods of Dating Bluegum (*Eucalyptus globulus*) Wood**

### *Introduction*

Trees are important archives of environmental contamination. Numerous studies have shown that trees growing in contaminated areas accumulate heavy metals and other toxic substances in their wood, and that if individual wood samples can be accurately dated the history of contamination can be reconstructed. Local concerns about NTLF tritium emissions arose in the mid-1990's when several LBNL scientists reported that tritium activity levels in rainfall and organic samples collected near the Lawrence Hall of Science (LHS) were unexpectedly high. This led community members to propose to LBNL that wood samples from eucalypts growing near the LHS be used to reconstruct exposure levels (McGraw, 2000). LBNL subsequently had a dendrochronologist evaluate the potential of the trees but he concluded that the indistinct nature of the ring structure would make it impossible to date individual samples (Brown, 2002). Not being convinced by this assessment we started to investigate the possibility that wood of Eucalyptus trees growing in the vicinity of the NTLF emissions stack could be accurately dated and thereby provide a temporal record of tritium emissions (Figure B-1). At about the same time, LBNL helped initiate a similar study by Adam Love, then a graduate student in the Department of Environmental Engineering at the University of California in Berkeley. Love took core samples from three Eucalyptus trees growing near the emissions stack and analyzed their organically bound tritium content. He dated his samples by measuring their radiocarbon content with Accelerator Mass Spectroscopy (AMS) at Lawrence Livermore National Laboratory. Love's research effort in many ways paralleled our own although the methods he used were very different from ours.



In the discussion that follows, we compare the two attempts to establish radiocarbon-based chronologies for Eucalyptus trees growing near the tritium stack.

### *Growth Rings in Eucalyptus*

The identification of annual growth rings in eucalypts is not easy for several reasons. One is that eucalyptus wood is diffuse porous and normally does not show well marked seasonal differences in cell structure. A second problem is that eucalypts are known to put on “false rings,” i.e., more than one ring may be laid down within a year because of short term changes in weather. A third problem, often encountered in older trees, is that rings may be extremely narrow or discontinuously present around the tree’s circumference. This often happens when a tree is growing on a slope and compensates for the angle by producing tension wood on the uphill side. Largely because of these problems eucalypts have not attracted much attention from dendrochronologists and dendroclimatologists.

On the positive side, there is some evidence that in certain environments eucalypts can produce countable annual rings. Mucha (1979) in a study carried out near Darwin, Australia showed that one of the local eucalypts (*Eucalyptus tetradonta*) produces annual rings that could be counted with careful effort. He also showed that ring growth was positively correlated with summer rainfall. Darwin at 12 degrees south has a strongly monsoonal climate.

Morrow and LaMarche (1978) studied insect grazing pressures on trees in the Snowy Mountains of southeastern Australia and found that a local eucalypt (*Eucalyptus stellulata*) produced annual rings that could be counted without too much difficulty. These trees grow in a sub-alpine environment (36 degrees south, 1585 m altitude) in which tree growth is again strongly seasonal.

The Tasmanian Blue Gum (*Eucalyptus globulus*), the eucalypt of interest in this project, is not naturally adapted to a seasonal climate (Figure B-2). It’s natural

range covers a small area of southeastern Australia (southern Victoria and Tasmania), an area that has a very equable climate. Summers are cool, winters warm, and rainfall occurs throughout the year. In eastern Tasmania, the core area of the species' distribution, *Eucalyptus globulus* is largely restricted to relatively moist coastal sites in well-drained frost free valleys. It's elevational range is sea level to 450 m.

During the past 200 years *Eucalyptus globulus* has been introduced to many countries around the world. It's rapid growth has made it valuable in some areas as a timber tree; in other areas it is valued as a source of fuel. It is reportedly the world's most important temperate hardwood as far as pulp production is concerned, *Eucalyptus globulus* was first introduced to California in the 1850's and during the late 19<sup>th</sup> century extensive plantings were made in the belief that the tree's rapid growth would lead to a profitable supply of lumber. This never materialized because *Eucalyptus globulus* wood cracks extensively on drying.

The numerous introductions of *Eucalyptus globulus* into so many different areas of the world have been tests of the bioclimatic tolerances of the species. In the coast ranges of central California there are two important climatic hazards facing *Eucalyptus globulus*: frost and drought. As was pointed earlier, freezing temperatures are rarely or never encountered in the coastal lowlands of Tasmania and southern Victoria, and *Eucalyptus globulus* has therefore not evolved the adaptations to withstand sub-freezing temperatures. The tree is also vulnerable to drought. According to Jordon et al. (1993), *Eucalyptus globulus* in its native habitat is adapted to a climate in which the mean precipitation of the driest month ranges from 30 mm to 73 mm. In the coast ranges of central California summers are typically rainless from May to September, and although this drought is to some extent modified by coastal fog the stress on *Eucalyptus globulus* is not insignificant. In the context of this project, summer drought is especially important in that it leads to a slowing of growth and the production of late wood which is denser and darker in color than the early wood produced in the spring and early summer.

## *Eucalyptus globulus* Wood Anatomy

An understanding of the cellular structure of *Eucalyptus globulus* wood is important in the counting of annual growth rings even though it is not necessary to work at the microscopic level. *Eucalyptus globulus* wood (xylem) consists of four kinds of cells.

1. Fibers. These cells make up the bulk of the xylem and provide structural support for the tree. They are small (less than 1 mm in vertical length) and are arranged, in the tree studied here, in a spiral-grained pattern.
2. Vessels. These cells are larger than the fibre cells and are connected to allow the vertical movement of water from the roots to the leaves. A cross section across the trunk exposes the vessels as holes, or pores. They are scattered across individual growth rings and are sometimes arranged radially. The pores can be seen without the aid of a hand lens as small white dots on the surface of a cross section; and on an x-radiograph as small black dots.
3. Parenchyma cells. These narrow walled cells are inconspicuous in *Eucalyptus globulus*. They form sheaths around the vessels.
4. Ray cells. As their name implies, ray cells are aligned along the radius of the trunk. They allow for the horizontal movement of materials from cambium (the layer between the wood and bark where cells actually form) into the xylem. In a tangential section they can be seen to be one, two, or three cells thick. In *Eucalyptus globulus* the rays are very small and can only be seen with the aid of a microscope or good hand lens.

On the outer side of the cambium layer is the bark (phloem). In the samples analyzed here it forms a very thin layer ca., 1 mm thick. The outer surface is smooth and light brown in color. *Eucalyptus globulus* bark is relatively dense and on x-radiographs of cross sectional cuts it can be seen as a light band ringing the outer circumference. On regular color film the most striking contrast in a cross sectional view is between the darker heartwood and lighter sapwood (Figure B-3). The latter represents that part of the wood in which water is actively transported through the vessels; the heartwood is non functional in this respect because the cells have filled with parenchyma. The dark area seen on Figure B-3 at the boundary between the heartwood and sapwood is fungal in origin.



The most important characteristic of *Eucalyptus globulus* wood anatomy, as far as the identification of annual growth rings is concerned, is the contrast between early wood and late wood (see Figures B-3 and B-4). In California's summer dry – winter wet climate early wood is formed in the spring and early summer. It is characterized by a high density of pores that can be seen easily on both x-radiographs and regular film (Figure B-3). As summer progresses drought stress increases and the formation of vessels is reduced. Fiber cells are smaller in size and have thicker cell walls. The resulting latewood is darker in color than the early wood and is also easily identified in x-radiographs by its denser (lighter) appearance. Latewood is also isotopically heavier than early wood because of fractionation during drought stress (MacFarlane and Adams, 1998).

#### *Isotopic Dating of Modern Wood*

At least two radioisotopes can provide independent checks on visually determined ring chronologies for wood that is less than ca., 100 years old: Carbon-14 ( $^{14}\text{C}$ ) and Lead-210 ( $^{210}\text{Pb}$ ). For  $^{14}\text{C}$  this is the reverse of the tree ring calibration for radiocarbon age estimates. Radiocarbon is used in this context because atomic weapons testing of the 1950's and 1960's nearly doubled the  $^{14}\text{C}$  content of the atmosphere. This increase is reflected in organic samples less than 50 years old as a spike in activity that peaked in 1964. Since then radiocarbon activity in the atmosphere has almost dropped back to its pre-testing levels. The age of wood samples less than 50 years old can therefore be determined by fitting their  $^{14}\text{C}$  activities to the bomb spike curve (Figure B-5).

#### *Methods of Analysis*

The primary objective of the present study was to provide a tree ring chronology for wood samples from a *Eucalyptus globulus* tree, hereafter referred to as Eucalyptus-2, growing 70 meters north of the National Tritium Labeling Facility's emissions stack. The upper part of the tree was blown down on

November 24, 2001 during a windstorm that produced 60 mph gusts in the San Francisco Bay area. Prior to the storm the tree was about 6 m tall with a diameter at breast height (dbh) of 27 cm. Ideally a larger diameter tree would be used in a study of this kind because, as we shall indicate later, the mean ring thickness in the fallen tree was less than 0.4 mm.

### *Sample Preparation*

Four consecutive sample logs each representing ca. 35 cm were cut from the fallen trunk with a chain saw. These were then taken to a wood working shop for further preparation. Two disks ca. 2 cm thick were cut from the best preserved part of two of the logs and each log was then cut twice tangentially on either side of the pith to produce ca., 2 cm thick slabs that exposed the growth rings vertically (Figure B-6). Annual growth increment samples are more easily cut in this alignment than in transform (cross) sections. After being cut the disks and slabs were sanded to bring out the grain. The best disk and slab were then selected for detailed measurement.

### *Ring Counting*

In order to facilitate the counting of individual growth rings, the sample disks and slabs were photographed with color film and x-rayed in a cabinet size x-ray machine. Kodak X-Omat 8 inch by 10 inch film was exposed and manually developed, fixed, and washed.

Ring counts were made first directly on the wood samples using the longest pith-circumference radius. Annual growth increments were more easily distinguished on the cross sectional disks than the tangential slabs. The color and pore density differences between early wood and latewood provided the basis for identifying individual rings.

The x-radiographs and color photographs were scanned at 300 dpi resolution to provide a basis for image analysis. The resulting images were then enhanced

for brightness and contrast in Photoshop and imported to NIH Image for analysis (Figures B-7 and B-8). Density plots for both the color photographs (RGB) and x-radiographs (grayscale) were then used to make decisions about individual growth rings. The usually sharp transition between latewood and earlywood was used to define the beginning and end of an individual ring (Figure B-8). Ring thicknesses were then measured as calibrated distances from the pith and also converted to percentages of total radial growth.

#### *Lead 210*

In order to obtain ash for lead 210 analysis we cut 12 samples at consecutive 1 cm increments from bark to pith from a 3.5 by 31.5 cm slab and burned every other sample in a furnace at 600 degrees centigrade for six hours. None of the samples produced the necessary 0.5 g of ash needed for alpha spectrometry and we were therefore unable to analyze for lead 210 activity.

#### *Carbon 14*

As was discussed above, the atomic bomb testing of the 1960's artificially increased the radiocarbon content of the atmosphere and thereby provided a means of dating wood samples that are less than 50 years old. In this project we submitted two sets of samples for radiocarbon dating. The first set was dated by conventional methods, i.e., liquid scintillation, at the Beta Analytic Radiocarbon Laboratory in Coral Gables Florida, as we shall indicate later these dates turned out to be anomalously old and we therefore submitted a second set to be dated at the Center for Accelerator Mass Spectroscopy (CAMS) at the Lawrence Livermore National Laboratory.

The samples for conventional radiocarbon dating were taken as follows. Five samples were cut from a room-dry radial slab (LHS Eucalytus-2 - slab 2) which had previously been x-radiographed (x-radiograph number# 0095) and photographed. The distance from the cambium layer to pith was 14.5 cm. An estimated 0.6 - 1.25 cm of shrinkage had occurred between the time the x-

radiograph was taken and the samples were sent for radiocarbon dating. Each sample was 0.3 cm thick. The samples were labeled by year of sampling and sample number. Each sample weighed approximately 20 g.

1. Sample 2002-1 was taken 13.2 cm from the cambium layer and 0.3 cm from the pith. Estimated date is 1947 - 1948.
2. Sample 2002-2 was taken 7.5 cm from the cambium layer and 6.7 cm from the pith, at 0.95 cm to the cambium side of a dark layer on the x-radiograph. Estimated date is 1957 - 1958.
3. Sample 2002-3 was taken 5.2 cm from the cambium layer and 8.6 cm from the pith, next to a thin dark layer on the x-radiograph. Estimated date is 1966 - 1968.
4. Sample 2002-4 was taken 2.9 cm from the cambium layer and 10.5 cm from the pith, 0.6 cm towards the pith from inner edge of dark band marking the outer edge of the heartwood. Estimated date is 1979 - 1980.
5. Sample 2002-5 was taken 0.2 cm from the cambium layer. Estimated date is 2000 - 2001.

The samples for AMS radiocarbon dating were taken from a thin cross sectional cut that was taken from the same slab sampled for lead 210. Because of the anomalous results from the first set of samples we made no age estimates for these samples.

1. Sample 2007-1 was taken 3 cm from the cambium layer at 76% of the pith - cambium distance.
2. Sample 2007-2 was taken 9.8 cm from the cambium layer at 21.5% of the pith - cambium distance.
3. Sample 2007-3 was taken 12.5 cm from the cambium layer at 0% of the pith - cambium distance.

The results of the radiocarbon dating are shown in Table B-1. With just one exception, sample 2002-5, all the estimated ages are older than expected. Sample 2002-1 was taken from the pith and therefore has to be less than 100 years old because the first plantings of Eucalypts in this area of the Berkeley Hills took place in the first two decades of the last century. The radiocarbon age of 280 +/-

60 years is clearly too old. Especially puzzling is the absence of any “bomb spike” signal. The samples were chosen in the hope that they would straddle the bomb spike of the early 1960’s when radiocarbon concentrations in the atmosphere were almost twice as high as they are today. As can be seen from Table B-1 only sample 2002-5 has a percent modern carbon (pMC) value of more than one hundred percent. Just why the first four samples were too old is not immediately apparent.

Sample number	Distance from cambium (cm)	Lab number	Radiocarbon age BP (before 1950)	Percent modern radiocarbon
2002-1	13.2	Beta - 170839	280 +/- 60 BP	96.53 +/- 0.72
2002-2	7.5	Beta - 170840	170 +/- 60 BP	97.86 +/- 0.73
2002-3	5.2	Beta - 170841	230 +/- 60 BP	97.13 +/- 0.73
2002-4	3	Beta - 170842	70 +/- 50 BP	99.09 +/- 0.62
2002-5	0.1	Beta - 170843	138.06 +/- 0.84 pMC	138.06 +/- 0.84
2007-1	3	CAMS-131372	249 +/- 35 BP	97.05 +/- 0.42
2007-2	9.8	CAMS-131373	235 +/- 35 BP	97.12 +/- 0.37
2007-3	12.5	CAMS-131374	170 +/- 35 BP	97.93 +/- 0.38

Table B-1. Radiocarbon Results for Eucalyptus-2.

### *Adam Love's Chronology*

Adam Love established radiocarbon chronologies for three Eucalyptus trees growing near the NTLF stack. Tree A is located 15 m uphill from the stack; tree B is 50 meters from the stack and 1 meter from the LBNL fence line; and tree C is 50 meters NE of the stack. The circumference and diameter at breast height of A was 203 and 64 cm, B 246 and 78cm, and C 241 and 77 cm. All three trees were therefore more than twice the size of Eucalyptus-2, used in our study, which had a circumference of 84 cm and diameter of 27 cm.

Love dated ~ 200 samples from the three eucalypts and a pine tree growing to the south of the stack. Samples from the latter could be precisely dated by ring counting and therefore provided a useful check on the the  $^{14}\text{C}$  bomb spike curve (Figure B-5).

Love's use of AMS made it possible for him to use very small samples, i.e., milligrams of wood. This in turn meant he could sample his trees with a ~5 mm diameter increment borer of the kind normally used by dendrochronologists and still recover enough material for reliable dates. His results for Eucalyptus A plus our results for Eucalyptus-2 are shown in Figure B-9. Love's Eucalyptus A record shows the bomb spike very clearly and therefore allows for a secure dating of the samples he also took for organically bound tritium analysis. As we indicated earlier, 4 of the 5 radiocarbon results for Eucalyptus-2 are anomalously old. Just why this is the case is not clear. One possibility is that the tree was effected by dead carbon emitted by vehicles in the nearby parking lot or possibly by radiocarbon depleted air from the Lawrence Hall of Science air vents which are located less than 50 meters from Eucalyptus-2. A third possibility is that the low radiocarbon activity on Eucalyptus-2 is a result of the tree being relatively small and more dependent on respired carbon dioxide than the canopy trees that Love sampled.



Fortunately, the problems encountered in dating Eucalyptus-2 did not completely preclude the development of a chronology. As we will indicate in the next section, the organically bound tritium results in themselves provide a means of dating the samples analyzed. They indicate that the tree was at least 30 years old in 2001, which means it is about half the age of the three larger trees sampled by Love. According to Love (2002), tritium was first used in what later became the NTLF in 1969; by coincidence this was about the time that Eucalyptus-2 got established.



Figure B-1. Aerial photograph of the National Tritium Labeling Facility and the Lawrence Hall of Science.



Figure B-2 *Eucalyptus globulus* Labill

Charles Webber © California Academy of Sciences



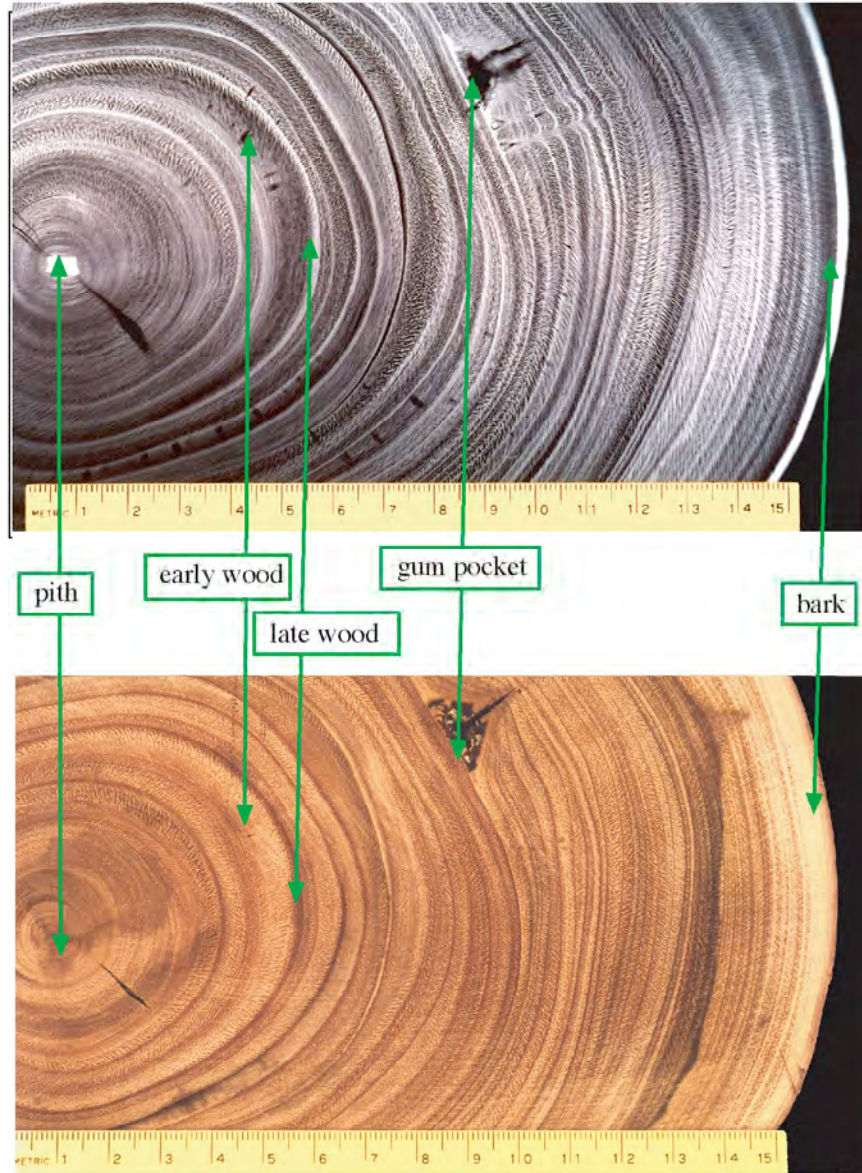


Figure B-3. An x-radiograph (top) and photograph (bottom) of a cross sectional cut of Eucalyptus-2.

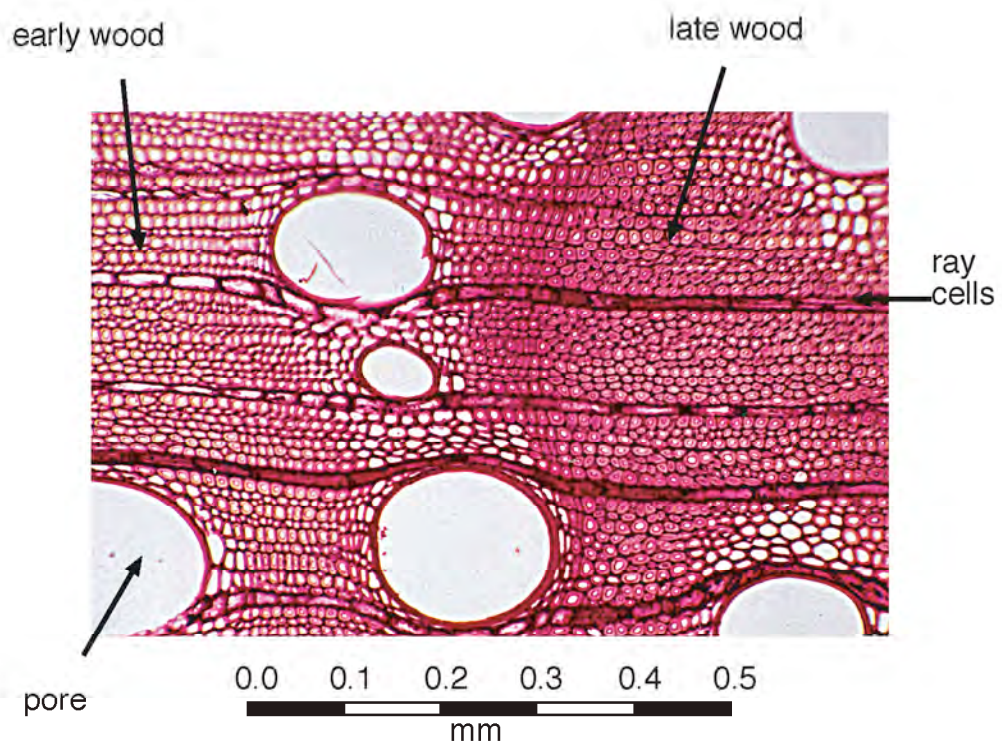


Figure B-4. A transverse thin section of *Eucalyptus globulus*.

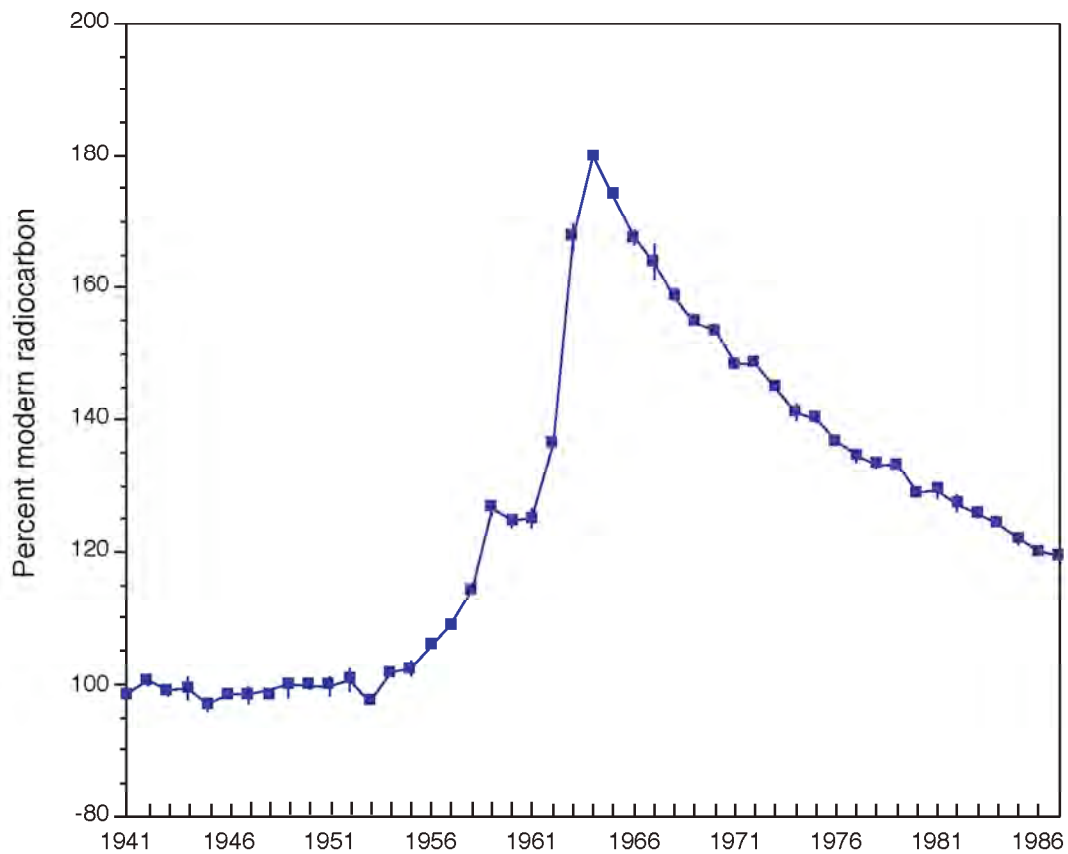


Figure B-5. The radiocarbon "bomb spike" after Yamada et al. (1989).  
 The vertical scale in the original publication is in "Delta 14C per mil" format but is shown here as "Percent modern carbon" to facilitate comparison with the radiocarbon results presented in Table B-1.



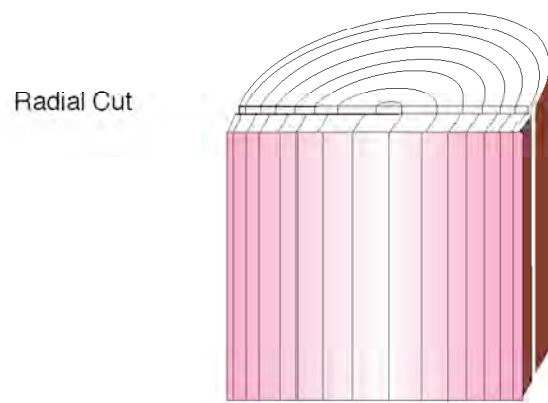


Figure B-6. Block diagrams showing a cross cut and a radial cut.

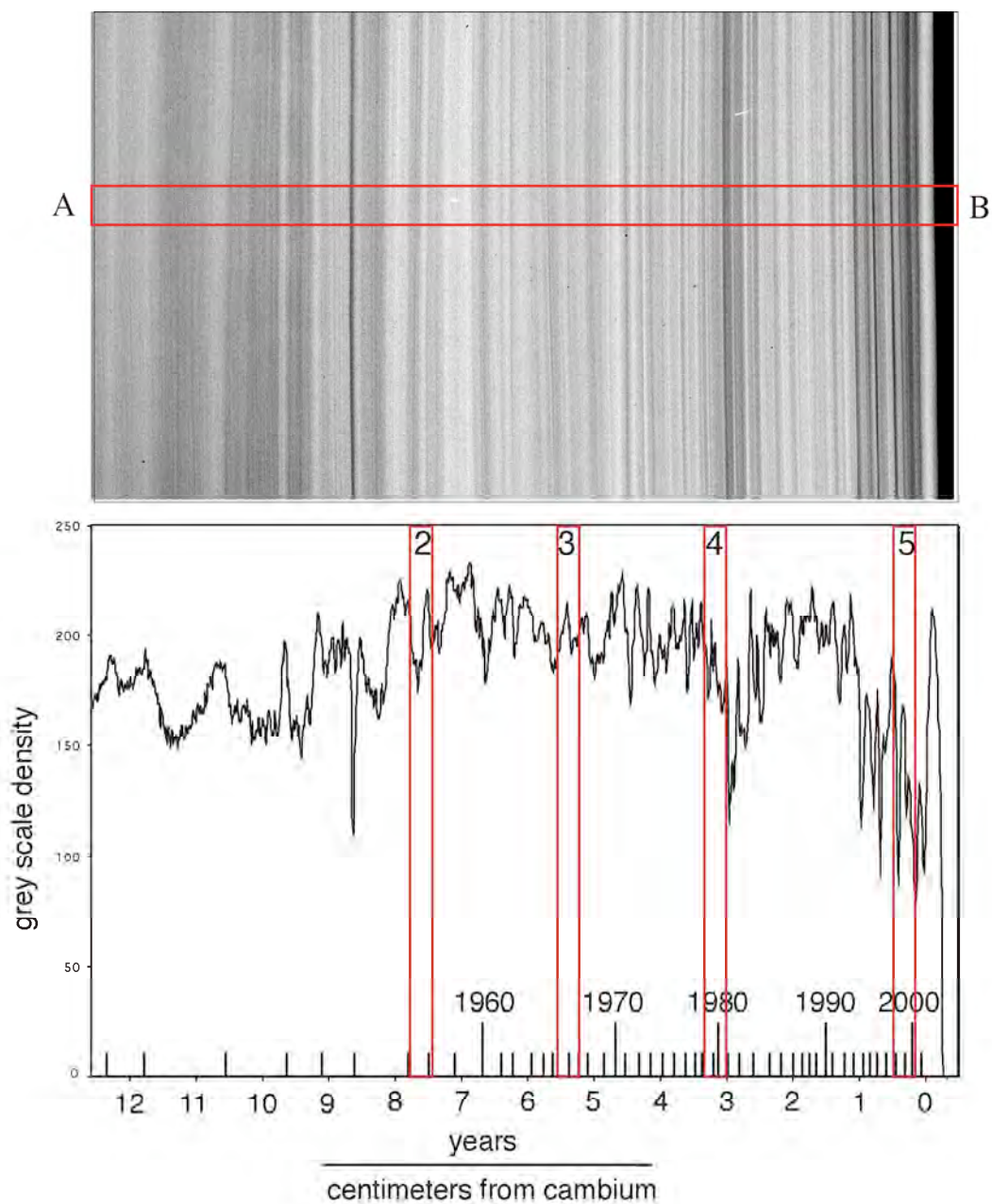


Figure B-7. The upper image shows an x-radiograph of a radial cut of Eucalyptus-2. The pith is on the left and the bark on the right. The lower graphic shows a gray scale density trace from A to B. The red rectangles indicate radiocarbon samples and the years the chronology based on ring counts.

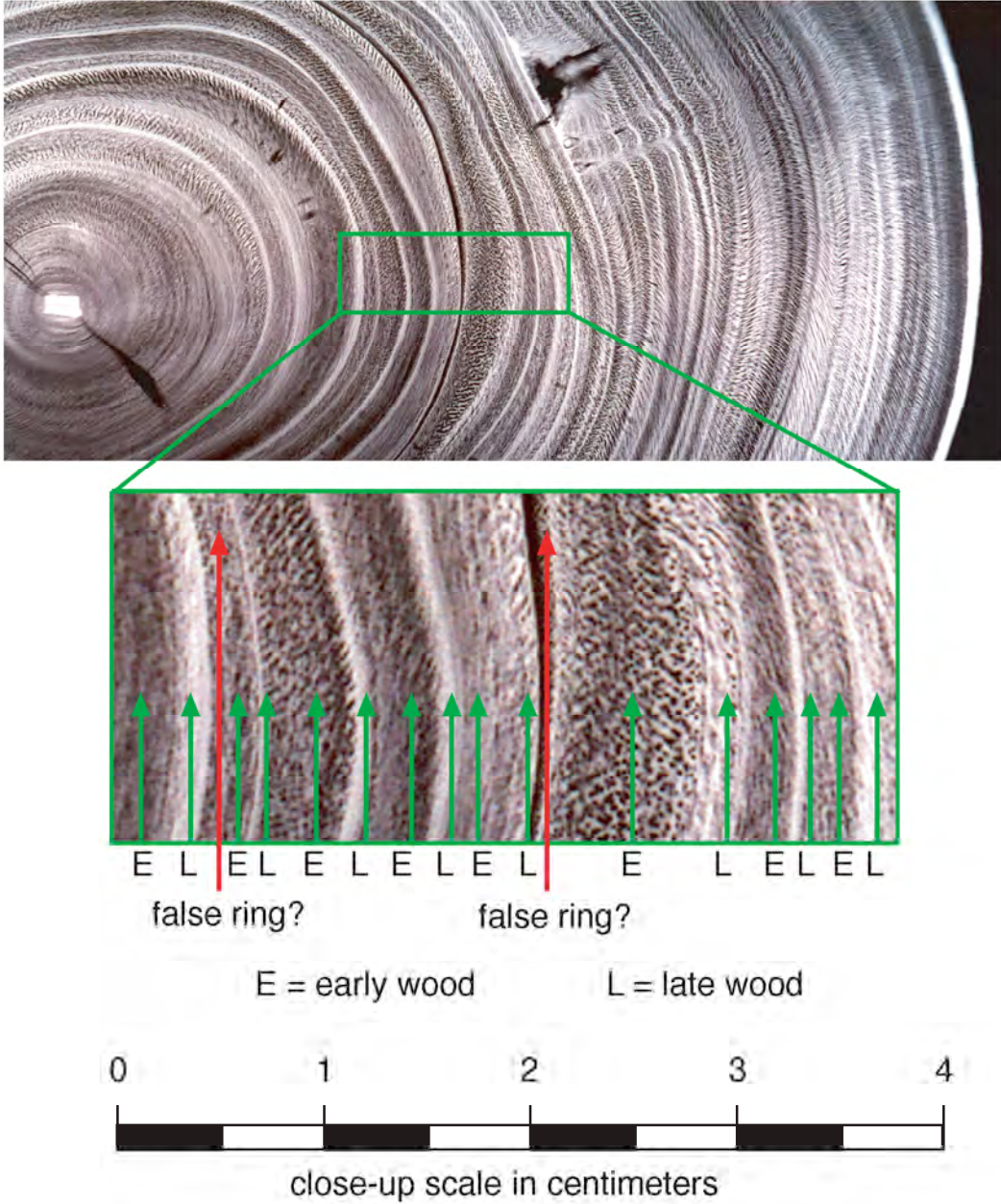


Figure B-8. A close up view of the rings in an x-radiograph of a cross cut from Eucalyptus-2. In the upper image the pith is on the left and the bark on the right.

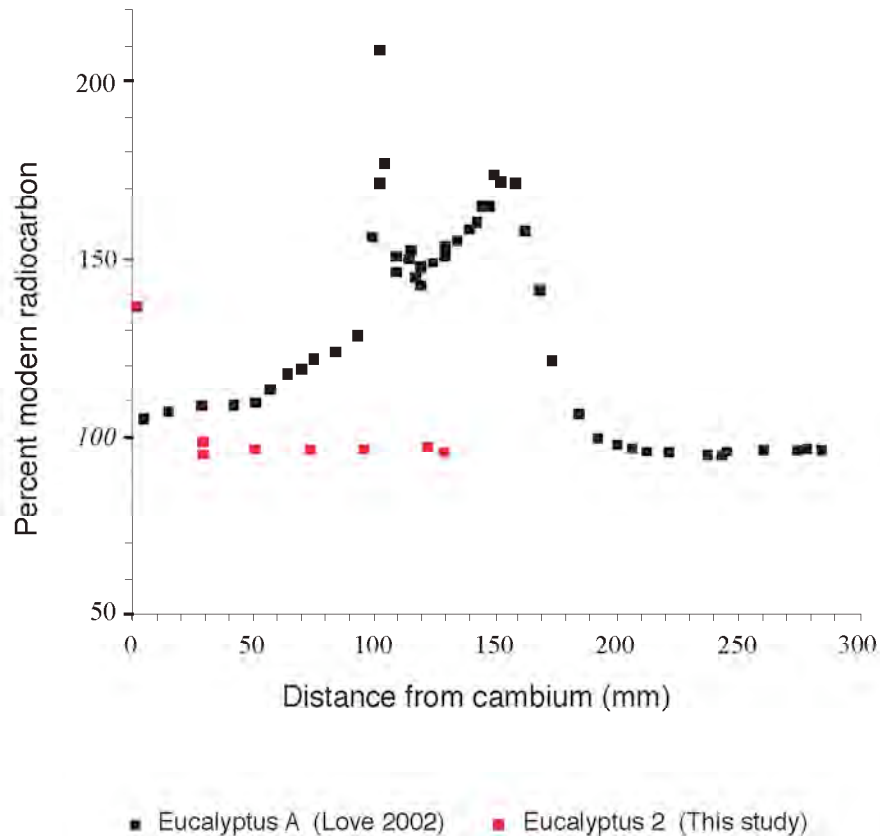


Figure B-9. Radiocarbon dates for Eucalyptus A (Love 2002) and Eucalyptus-2. Love attributes the anomalously high radiocarbon percentages in Eucalyptus A, at ca. 100 mm in from the cambium, to unreported releases from LBNL.

## **Part C: Reconstruction of Tritium Emissions by Analysis of Organically Bound Tritium in Eucalyptus Wood**

### *Introduction*

As stated earlier, reports by LBNL scientists in the mid-1990's, indicating that tritium levels in rainfall and organic samples collected near the Lawrence Hall of Science were unexpectedly high, aroused public concern. In 1996 the Environmental Health and Safety Department at LBNL carried out a study which determined the tritium content of plant material along two 600 meter transects (north-south, and west-east), both centered on the NTLF stack. The organically bound tritium concentrations for several of the samples were extremely high (LBNL-EHS Site Environment Report for 1996). For example, the activity level in the foliage of a tree 100m west of the NTLF stack at the Lawrence Hall of Science was 345 pCi/g, an activity level higher than anything reported in a similar study at Savannah River, one of DOE's major atomic weapons laboratories (Stewart et al. 1972).

Community concern intensified when subsequent LBNL reporting on tritium levels failed to clarify the situation. In 1998 LBNL's EH&S Division began a comprehensive sampling program in which hundreds of plant samples from near the NTLF stack were analyzed for their free water and organically bound tritium activities. The results were typically an order of magnitude lower than those reported in the 1996 Site Environment Report. For example, leaf samples collected ca. 100 m NNW of the stack in 1998 and 2001 (2 sampling dates) produced activity levels of only 50, 24, and 23 pCi/g, respectively (Thomas et al. 2002). Possible reasons for the discrepancy between the 1996 and later results were never discussed.

As we indicated earlier, concern about the reliability of emissions data led community members to propose to LBNL that dated wood samples from eucalypts growing near the LHS be used to reconstruct tritium emissions (McGraw 2000). LBNL subsequently had a dendrochronologist evaluate the

potential of the trees but he concluded that the indistinct nature of the ring structure would make it impossible to date individual samples (Brown, 2000).

In 2001 two independent research efforts were initiated to develop a record of NTLF tritium emissions from the eucalyptus trees growing near the Lawrence Hall of Science. The first was the study reported on here; the second was initiated by Adam Love, as part of his doctoral research in the Department of Environmental Engineering at the University of California, Berkeley (Love, 2002; Love et al. 2003). As we have discussed in Part B of this report, both research efforts attempted to date Eucalyptus wood samples by identifying the radiocarbon bomb spike of the 1960's. For reasons that are still unclear, with one exception our radiocarbon results are anomalously old. In contrast, Love was able to produce reliable chronologies for the three eucalyptus trees he analyzed, which made it possible to correlate the tritium content of his cores to stack emissions data. Furthermore, insofar as our tritium data are generally similar to his, we are also able to use his chronology. The details of the dating efforts have been discussed in Part B. Here we summarize the results of the organically bound tritium analyses. Love's results are discussed first because they provide a check on the reliability of our own data.

#### *Love's Methods and Results*

Love sampled three Eucalyptus trees growing near the NTLF stack (Figure C-1). Tree A is located 15m from the stack, and trees B and C are both 50 meters from the stack. All three are mature trees; tree A had a circumference at breast height of 203 cm, tree B 246 cm, and tree C 241 cm.

Love measured the organically bound tritium in his wood samples by Accelerator Mass Spectroscopy (AMS) at the Center for Accelerator Mass Spectroscopy (CAMS) at the Lawrence Livermore National Laboratory (LLNL). His use of AMS made it possible to use very small samples, i.e., milligrams of wood. This in turn meant he could sample his trees with a ~5 mm diameter increment borer of the kind normally used by dendrologists and still recover



enough material for a high resolution analysis. His sample thickness for tritium was 2 mm for Eucalyptus A and C and 3 mm for Eucalyptus B. A short section of a core from Eucalyptus A was even sampled at 0.5 mm intervals. The bomb spike chronologies indicate average growth rates of 4.1 mm/year for Eucalyptus A, 4.5mm/year for B, and 4.2mm/year for C. These growth rates indicate that Love's sampling intervals provide sub annual resolution. One negative aspect of the AMS technology is the very high cost of analyzing samples. One AMS 14C sample costs \$600 and a tritium sample ca. \$750. Love had ~200 radiocarbon samples analyzed and ~400 tritium samples, so his total sample cost was ~\$420,000. In this study our tritium samples were analyzed by liquid scintillation counting at a cost of less than \$10 per sample.

The decay corrected organically bound tritium activities for Eucalyptus A, B, and C are shown in Figure C-2. The bar graphs show NTLF's reported tritium emissions. In general the organically bound tritium records for the three trees are reassuringly similar with each showing three main peaks with the two higher peaks dating to the 1980's. The curves also match reasonably well with the NTLF emissions data although the match is weaker from 1970 to 1986 suggesting that emissions may have been underreported during the earlier period of the facility's operation. The three records also show that tritium activity decreases with distance from the stack. Tritium levels are more than twice as high in Eucalyptus A which is 15 meters from the stack than in B and C which are both 50 meters from the stack.

Love also compared his results with the reported LBNL EH&S's organically bound tritium results and found general agreement although the very high 1996 values were not supported by his data.

#### *Methods and Results of this Study*

Eucalyptus-2, the tree sampled in this study, is located 70 meters north of the NTLF Emissions Stack (Figure C-1). The upper part of the tree was blown down in the storm of November 24, 2001 and was therefore available for sampling.

Unfortunately, this tree was much smaller than the trees sampled by Love and the circumference of the trunk was only 84 cm. This necessitated the use of a very different sampling geometry than the one he used. As indicated in the previous section, 3 cm thick radial slabs were cut from the trunk of Eucalyptus-2 to facilitate a 2.5 mm interval sampling from bark to pith (Figure C-2). The samples were cut with a fly cutter (a rotating chisel that can be set to cut to a desired depth) and captured in a specially constructed cardboard box. The sampled slab, Eucalyptus-2 slab 1, measured 31.75 cm x 11.5 cm x 3 cm and produced 41 samples. The average sample volume was 25 cm<sup>3</sup> and the mass 22 g.

In order to ensure that the tritium activities in the samples would reflect the tritium activity in the water at the time the wood cells were formed, we used the same acid/alkali/alkali/acid pre-treatment as Adam Love (Love et al. 2003). This removes the more soluble components of the wood, such as resins, that might have been deposited some time after the formation of the cellulose and lignin. The cleaned samples were then allowed to equilibrate in tritium-free distilled water for 24 hours and oven dried for 24 hours at 110 C degrees. It is important to note that the tritium of interest in the present study is the organically bound tritium, i.e., the tritium that was in the water that combined with CO<sub>2</sub> during the process of photosynthesis. The tritium in the tissue free water was not of interest because it postdates the actual formation of the wood cells.

The water of combustion was extracted from the samples by combustion in a 1.8 liter "Parr Oxygen Bomb" specially designed for large (10 g) organic samples. The "Parr Bomb" sits in a bath of ice water and is allowed to cool after firing so the water of combustion condenses and can be poured into a scintillation vial. The average dry weight of the samples was 10 g and the volume of water recovered ca. 5 ml. Some samples were more finely divided than others and this necessitated some variation in the oxygen pressure but in most cases it was 15

combined sample number	batch number	combined samples	top depth cm	bottom depth cm	assigned year	combined H2O volume ml	counting time seconds	cpm	assigned year	uncorrected pCi/L	decay corrected pCi/L	tritium units uncorrected	tritium units decay corrected
2	Q26	1+2+3+4	0	1	2000	5	3600	6.4	2000	575	845	265	265
3.5	Q17	3+4	0.5	1	1998	5.01	3600	7.6	1998	687	1126	353	353
5.5	Q9	5+6	1	1.5	1996	5.02	3600	0.0	1996	0	0	0	0
7.5	Q18	7+8	1.5	2	1994	5.02	3600	2.5	1994	221	451	141	141
9.5	Q1	9+10	2	2.5	1991	5.01	3600	4.1	1991	374	899	282	282
11.5	Q10	11+12	2.5	3	1989	5.03	3600	7.7	1989	697	1870	586	586
13.5	Q2	13+14	3	3.5	1987	5	3600	3.2	1987	287	860	270	270
15.5	Q11	15+16	3.5	4	1986	5.03	3600	0.0	1986	0	0	0	0
17.5	Q3	17+18	4	4.5	1985	5.03	3600	5.4	1985	491	1640	514	514
19.5	Q12	19+20	4.5	5	1983	5.01	3600	2.1	1983	193	720	226	226
21.5	Q4	21+22	5	5.5	1982	5.03	3600	2.8	1982	254	1001	314	314
23.5	Q13	23+24	5.5	6	1980	5.02	3600	3.2	1980	290	1273	399	399
25.5	Q5	25+26	6	6.5	1979	5.02	3600	5.8	1979	520	2415	757	757
27.5	Q14	27+28	6.5	7	1977	5.02	3600	2.2	1977	196	1013	317	317
29.5	Q6	29+30	7	7.5	1976	5.02	3600	1.0	1976	92	505	158	158
31.5	Q15	30+31	7.5	8	1975	5.01	3600	0.0	1975	0	0	0	0
33.5	Q7	33+34	8	8.5	1974	5	3600	0.0	1974	0	0	0	0
35.5	Q16	35+36	8.5	9	1973	5.02	3600	3.0	1973	269	1736	544	544
37	Q8	37	9	9.25	1972	5	3600	2.2	1972	194	1322	414	414
39.5	Q19	39+40	9.5	10	1971	4.97	3600	0.0	1971	0	0	0	0
40	Q25	39+40+41	9.5	10.25	1970	5.02	3600	0.0	1970	0	0	0	0

Table C-1. Eucalyptus-2 organically bound tritium data.

atmospheres. The very finely divided samples tended to blow out of the sample cup, which in turn resulted in incomplete combustion. The amount of water recovered from the incompletely combusted samples was often less than 5 ml and we therefore had to combine adjacent samples to ensure effective counting in the liquid scintillator. The water of combustion was determined to be very acid ( $\text{pH} < 3$ ) and we therefore added a small amount (ca., 0.1 g) of Calcium Oxide ( $\text{CaO}$ ) to each sample to increase the pH. All samples were then distilled in a microstill and 5 ml volume subsamples shipped to the DHI Laboratory in Horsholm, Denmark for tritium counting. The counting time was one hour and the mean counting error  $\pm 12$  percent. The results uncorrected and corrected for decay are shown in Table C-1. The activity levels are expressed both as picoCuries per liter ( $\text{pCi/L}$ ) and as Tritium Units ( $\text{TU's}$ ) to facilitate comparison with Love's results. He presented all his tritium activities in tritium units. One tritium unit is equivalent to  $3.19 \text{ pCi/L}$ .

Our decay corrected results are also shown in Figure C-4 together with the reported NTLF tritium emissions data. The temporal pattern of tritium activity for Eucalyptus-2 is generally similar to the patterns obtained for Eucalyptus A, B, and C. However, the Eucalyptus-2 record differs in that it shows five activity peaks rather than three. Assuming Love's chronology is correct, they date to  $\sim 1973$ ,  $\sim 1978$ ,  $\sim 1984$ ,  $\sim 1988$ , and  $\sim 1999$  with a probable error of  $\pm$  one year in each case. The Eucalyptus A, B, C records do not have a peak in the late 1990's. This discrepancy can't be attributed to differences in the dates the trees were sampled as Eucalyptus A was sampled on September 25, 2001, Eucalyptus B and C on December 18, 2001, and the Eucalyptus-2 trunk was broken during the storm of November 24, 2001. Further analysis of wood from all 4 trees is needed to determine whether or not the late 1990's peak in Eucalyptus-2 is real or not.

Another difference between the four activity curves is that the 1973 and 1978 peaks are higher, relative to the later peaks, in Eucalyptus-2 than in Eucalyptus A, B, and C. The highest tritium activity level in the decay-corrected Eucalyptus-2 curve dates to 1978 whereas in Eucalyptus A it is 1984, and in Eucalyptus B and C it is 1988. In general the similarities between the activity curves for the four

trees is reassuring that the trees are useful monitors of tritium releases from the NTLF. The decay corrected curves do correlate reasonably well with the reported release data, at least for the late 1980's and early 1990's. However for the 1970's and early 1980's the evidence suggests that tritium releases were underreported; and, if the Eucalyptus-2 record is reliable, this was also the case in the late 1990's.

One major difference between the Eucalyptus-2 and the A, B, C activity records is that the former are three orders of magnitude lower than the latter. For example the maximum activity measured in Eucalyptus-2 is 8 pCi/ml as compared with ~9,000 pCi/ml in Eucalyptus B and C. In part this reflects the fact that Eucalyptus-2 is located 70 meters from the stack whereas B and C are only 50 meters away. According to one report on organically bound tritium (OBT) near the NTLF, OBT content in wood is at, or below, the detection limit beyond 40 m of the NTLF stack (Thomas et al. 2002).

There are other possible explanations for the difference. One is that Eucalyptus-2 differs from Eucalyptus A, B, and C in the source of its water. Eucalyptus-2 is a much smaller tree than A, B, and C with a circumference at breast height of only 84 cm as compared with 203 cm, 240 cm, and 241 cm for A, B, and C, respectively. We estimate its average radial growth rate to be ~ 3.7 mm/year which is less than the mean of 4.3 mm/year reported by Love for the trees that he sampled. We were unable to measure accurately the height of Eucalyptus-2 but inspection of the fallen trunk suggests that it was significantly shorter than the older trees growing around it. All of this suggests that Eucalyptus-2 may have been more sheltered from the stack effluent than Eucalyptus A, B, and C. If this was the main reason for its lower tritium concentrations, the implication is that the tall canopy trees absorb more water vapor from the atmosphere than small understory trees such as Eucalyptus-2.

The lower tritium concentrations in Eucalyptus-2 than in Eucalyptus A, B, and C are evidence that trees cannot be used to reconstruct exposure levels in a quantitative sense. The factors that determine a tree's water use are complex,

even within a stand of some 200 trees as in the LHS Eucalyptus grove. On the other hand, the fact that the pattern of temporal variation in tritium activity in Eucalyptus-2 is broadly similar to that found by Love in Eucalyptus A, B, and C is reassuring confirmation of the thesis that trees are useful monitors of environmental emissions.

#### *Wider Implications of the Study*

One of the important implications of this study is that traditional liquid scintillation methods can be used to reconstruct the history of emissions as preserved in the wood of trees. Love et al. (2002) conclude in their paper on reconstruction of tritium exposure at LBNL that liquid scintillation methods would have been impractical in this case because of the time and labor involved in sample collection, processing, and analysis. This is definitely not the case. It is true that larger samples are needed for liquid scintillation than for AMS but sample collection, processing, and analysis are not prohibitively difficult.

A more important question concerns the size of the sample needed to generate meaningful results. The answer to this question depends upon the growth rate of the tree, the sampling size needed, and the accuracy of the counting system used. In this study we used a low cost liquid scintillator to analyze 5 ml water samples. Typically 10 g of dry eucalyptus wood will produce 5 ml of water on combustion. Ten grams of eucalyptus wood has a volume of ~ 12 cc so a sample with a radial thickness of 0.5 cm could have a width of 2 cm and a height of 12 cm. This volume of wood could not be easily obtained by coring the tree as the largest commercially available tree corer has a diameter of only 11 mm. If a more sensitive scintillator is used, and the tree of interest has a faster growth rate than our Eucalyptus, coring would be feasible. For example, if only a 1 ml water sample is needed and the tree has an annual growth rate of 1 cm, just 4 cores would be sufficient. The low cost of liquid scintillation counting, relative to AMS, definitely makes it a more attractive approach for environmentalists interested in reconstructing tritium emissions. The extraction of the water of



combustion from a sample involves an additional cost but it will most likely be less than \$50 per sample.

### *Conclusions*

The important conclusion to be drawn from this study is that the Eucalyptus trees between the NTLF stack and the Lawrence Hall of Science do contain a reliable record of tritium emissions. This conclusion is based on the overall similarities between our study and the parallel investigation of Adam Love. The tree we sampled is located 70 meters from the stack, a distance that, according to one report, should mean that organically bound tritium levels are below the limits of detection (Thomas et al. 2002). This was clearly not the case. Our study also demonstrates that high temporal resolution sampling, at least biannual, can be done without having to process large volumes of wood. It also demonstrates that liquid scintillation counting can provide useful results and that accelerator mass spectroscopy is not essential in a study of this kind. This last point will be encouraging news for environmental groups who might not be able to afford the high cost of AMS sample analysis.

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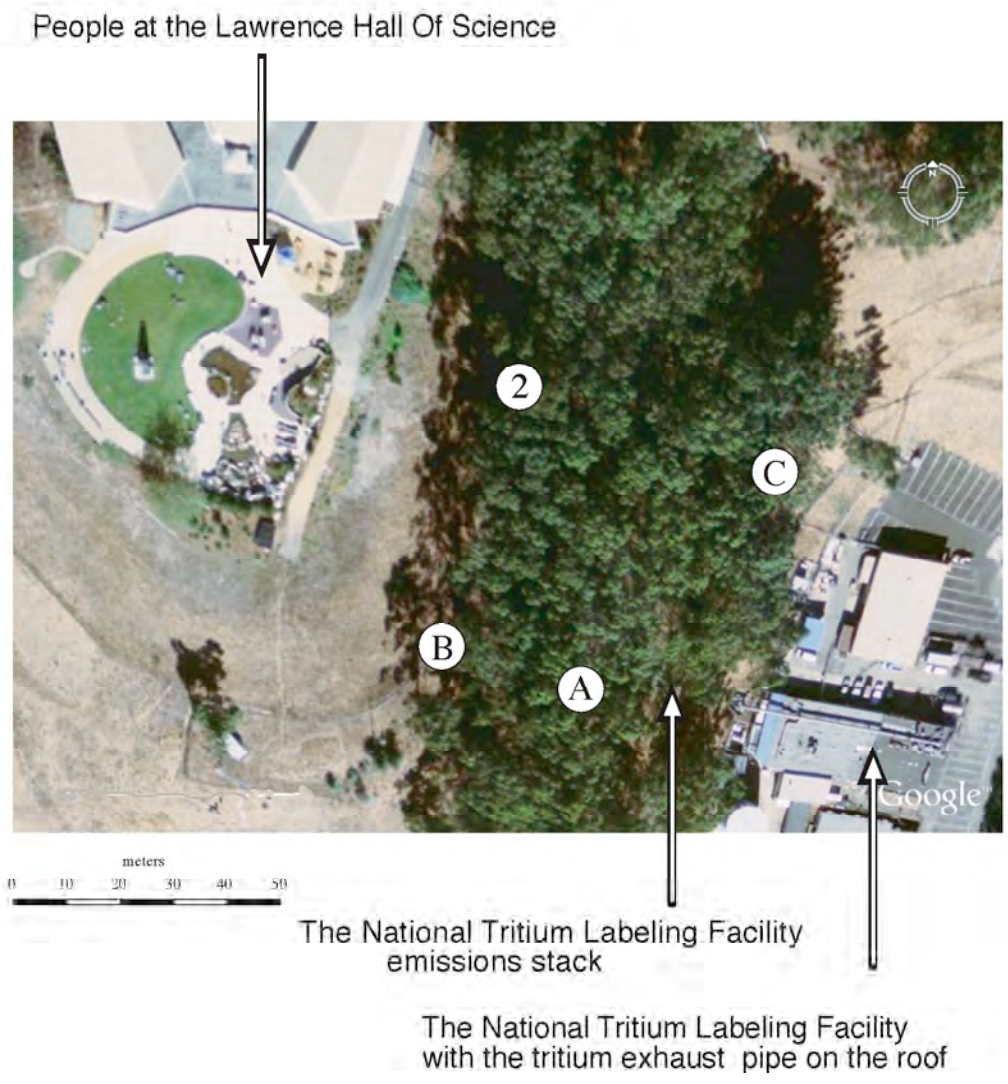


Figure C-1. Google image showing the location of Adam Love's sampled trees and Eucalyptus-2. Note also the shadows of people at the Lawrence Hall of Science.

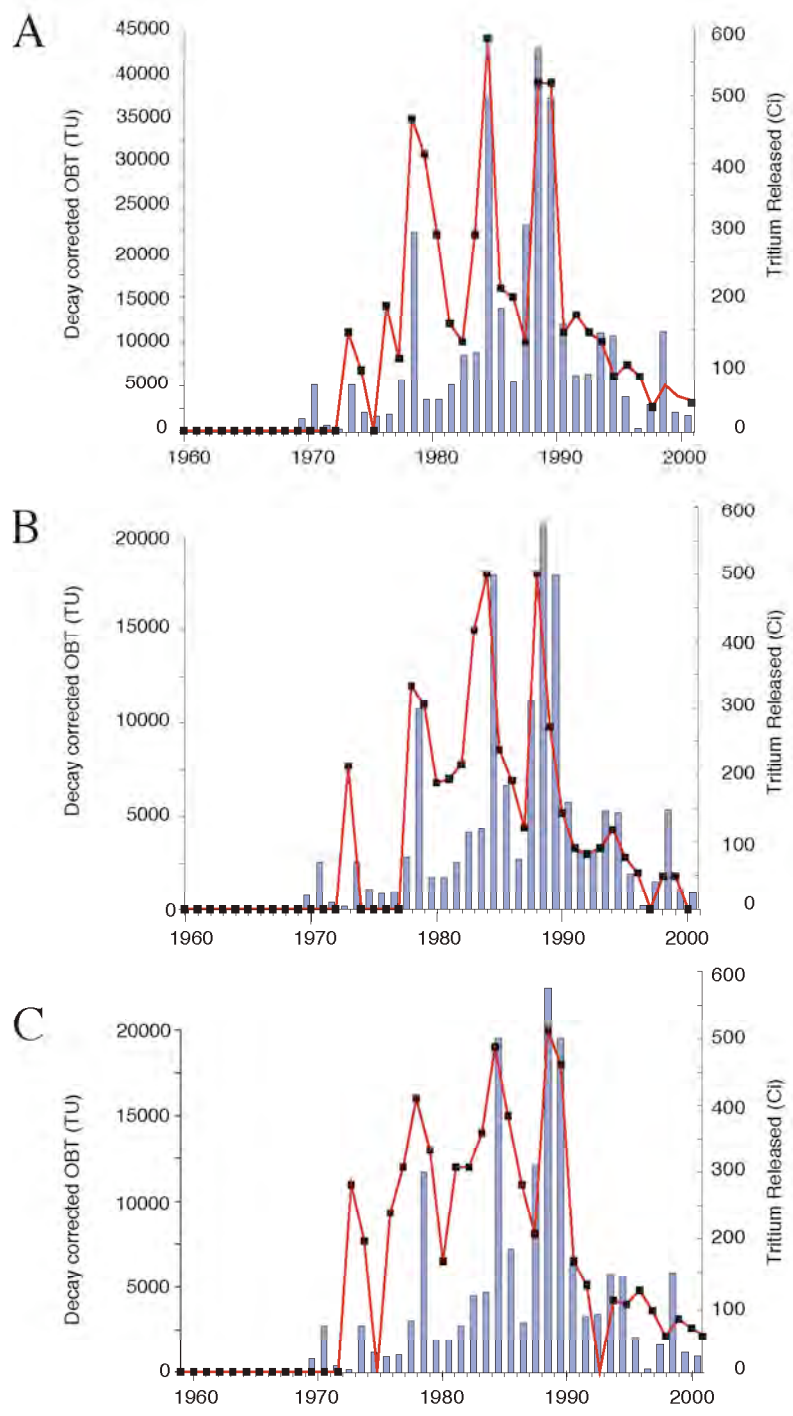


Figure C-2. Decay-corrected organically bound tritium activity in Adam Love's Eucalyptus A, B, and C (red lines) plus NTLF emissions data (blue bars). Redrawn from Love (2002).

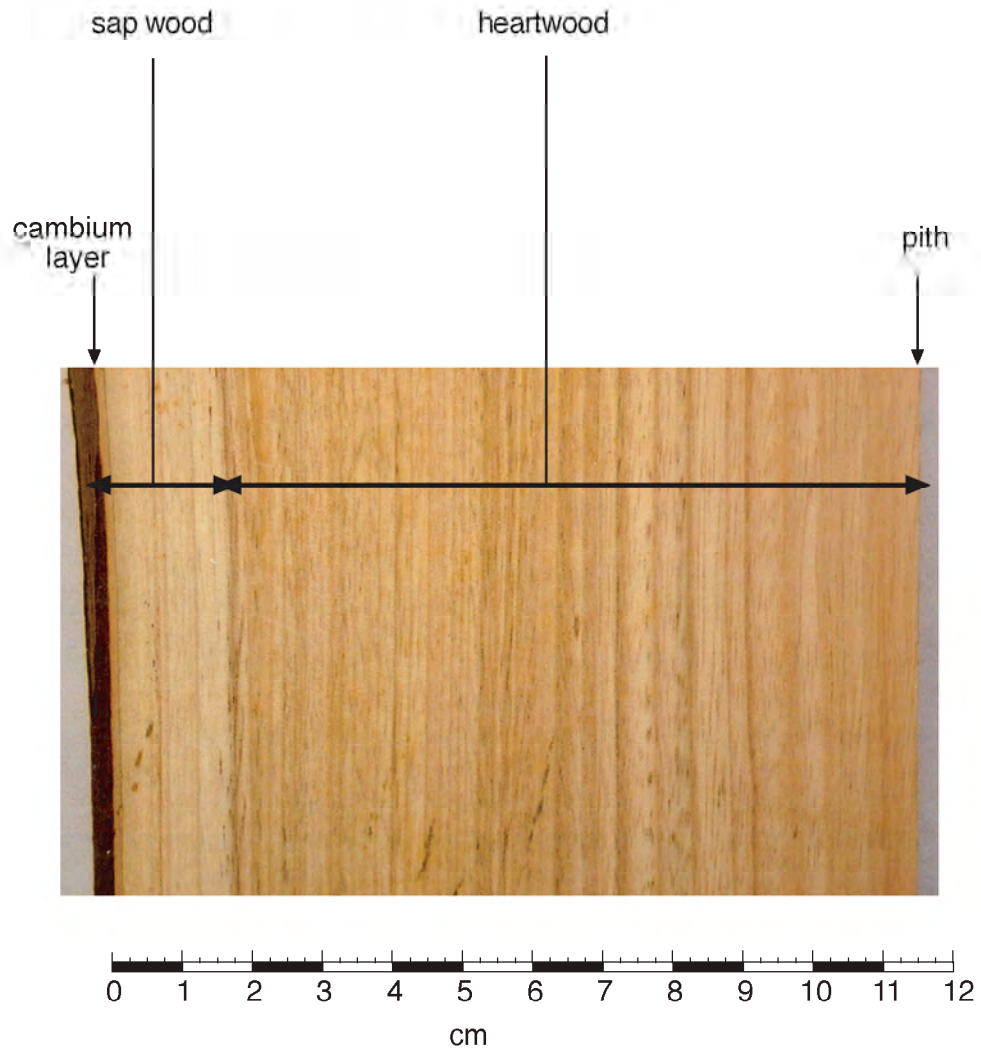


Figure C-3. Photograph of Eucalyptus 2, slab 1. The centimeter scale shows the 2.5 mm sampled intervals.

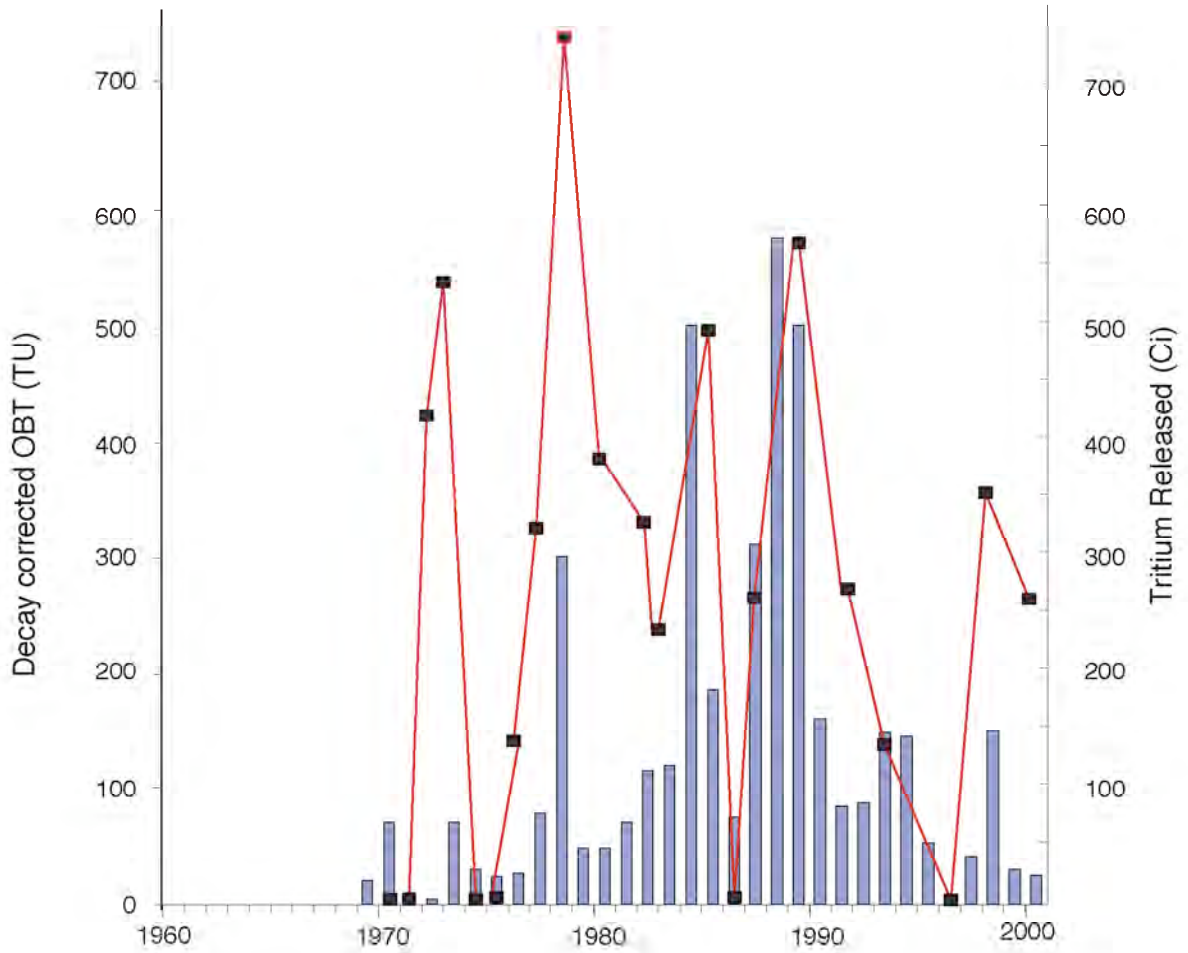


Figure C-4. Decay corrected organically bound tritium activity for Eucalyptus-2 (red line). NTLF reported tritium emissions (blue bars).

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