Reconstructing Tritium Exposure Using Tree Rings at Lawrence Berkeley National Laboratory, California

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Annual tritium exposures were reconstructed using tree cores from *Pinus jeffreyi* and *Eucalyptus globulus* near a tritiated water vapor release stack. Both tritium (^{3}H) and carbon-14 (^{14}C) from the wood were measured from milligram samples using accelerator mass spectrometry. Because the annual nature of the eucalyptus tree rings was in doubt, ^{14}C measurements provided growth rates used to estimate the age for ^{3}H determinations. A 30-yr comparison of organically bound tritium (OBT) levels to reported ^{3}H release data is achieved using OBT measurements from three trees near the stack. The annual average ^{3}H, determined from atmospheric water vapor monitoring stations, is comparable to the OBT in proximal trees. For situations without adequate historical monitoring data, this measurement-based historical assessment provides the only independent means of assessing exposure as compared to fate and transport models that require prior knowledge of environmental conditions and ^{3}H discharge patterns.

Introduction

On numerous occasions, industrial and commercial operations release a variety of contaminants into the environment without adequate monitoring of exposure levels. Still, there are relatively few techniques to reconstruct exposure associated with contaminant releases. The use of tree rings to study historical environmental levels of contaminants is well-documented (*1−3*). Trees are especially useful for determining historical isotopic levels of hydrogen, carbon, and oxygen on an annual basis because these elements are the primary constituents in cellulose. In most studies, tree ring isotopic records are used to determine natural variations in isotopic composition, but in more limited cases, isotopic variations in trees can record anthropogenic discharges of isotopes into the environment.

From 1969 to 2001, Lawrence Berkeley National Laboratory (LBNL) discharged tritium (^{3}H) into the atmosphere through an emission stack from a facility that was primarily

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responsible for synthesizing ^{3}H-labeled organic molecules for use in biomedical research. As a result, ^{3}H activities above those currently encountered in natural precipitation were measured in the air, soil, and vegetation near the stack. The surrounding residential community was concerned about the operation of the facility and the adequacy of both monitoring data and exposure models. This study undertook an historical assessment of ^{3}H exposure near the stack by measuring the ^{3}H content in tree cellulose. Previous efforts at tree ring analysis at this location were hampered by the uncertainty in the annual ring formation of *Eucalyptus globules* (*4*). To address this concern, carbon-14 (^{14}C) measurements were matched to known atmospheric ^{14}C levels to provide an age estimate for the ^{3}H results.

Site Description

LBNL is located on the steep slopes of the west facing hills in Berkeley, CA, approximately 25 km east-northeast of San Francisco (Figure 1). Berkeley has a moderate climate, with temperatures that generally range from 10 to 25 °C and rainfall that averages 60 cm/yr (*5*). Virtually all of the rain occurs during the months of November−March (*5*). Sea breezes from the Pacific Ocean result in the dominant wind direction from west to east. The topography of this area causes distinct meteorological variation on the local scale. Throughout much of the year, fog covers this section of the Berkeley hills from evening until morning.

Building 75 (B75) was the location of the ^{3}H-labeling facility at LBNL. This building is at approximately 300-m elevation, and tritiated water vapor was discharged through a stack ∼10 m high located above it on the adjacent south-facing hillside (slope of ∼30°). The aerial photograph in Figure 1 shows that trees surround the stack on three sides. These trees are all *E. globules* (eucalyptus) and currently extend above the top of the stack. Near B75, the wind direction is primarily in the regional west to east direction, but less often the wind follows the local topography and is from the southeast. The wind records from LBNL monitoring stations show that the eucalyptus grove around the stack is often very calm with the wind below 0.5 m s^{-1} 38% of the time during 2001.

Although B75 was built in 1965, ^{3}H was not used in the building until 1969. In 1982, this facility was formally established as the National Tritium Labeling Facility (NTLF), which brought an increased level of activity. The annual amount of ^{3}H released ranged from 4 Ci (1972) to 575 Ci (1988) (*6*), with both planned and unplanned releases occurring (Figure 2). The ^{3}H handling of the NTLF was described in detail by Morimoto and Williams (*7*). Although most of the ^{3}H that was not used for labeling was recycled, some was released into the atmosphere through a stack that vented laboratory gloveboxes. The emissions were mostly in the form of tritiated water (75−90%), and the rest was released as tritiated hydrogen gas (*6*, *8*). Procedural and hardware improvements in waste recovery during the 1990s dramatically decreased the amount of ^{3}H released during normal operations (Figure 2). The NTLF was closed in December 2001.

Tritium quantities are expressed in a variety of units: 1 ^{3}H atom/10^{18} ^{1}H atoms = 1 tritium unit (TU) = 3.2 pCi L^{-1} of water = 120 Bq m^{-3} of water. Assuming a humidity of 0.015 kg of water m^{-3} and using the reported stack flow rate of ∼280 m^3 min^{-1} (10^4 cfm) (*7*), the average annual ^{3}H activity of the stack water vapor increases ∼10^5 TU for each Ci of ^{3}H released during that year.
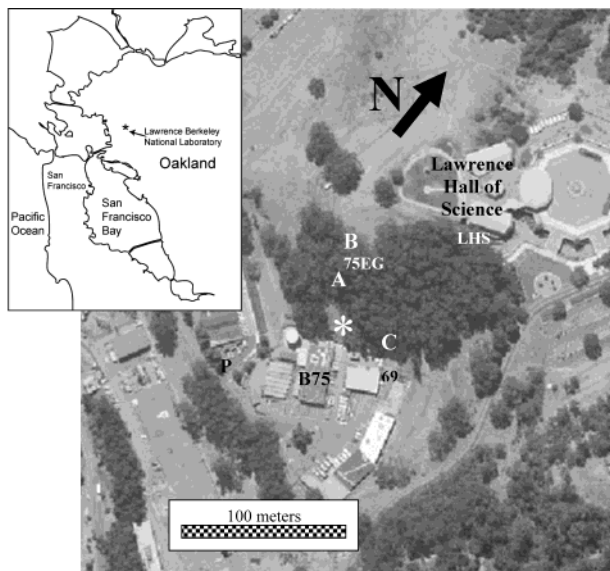
FIGURE 1. Aerial photograph of area around Building 75 (B75). Location of emission stack from B75 is indicated by asterisk. P indicates location of sampled pine, and A–C are locations of sampled eucalyptus. 75EG, 69, and LHS are locations of LBNL water vapor monitors.
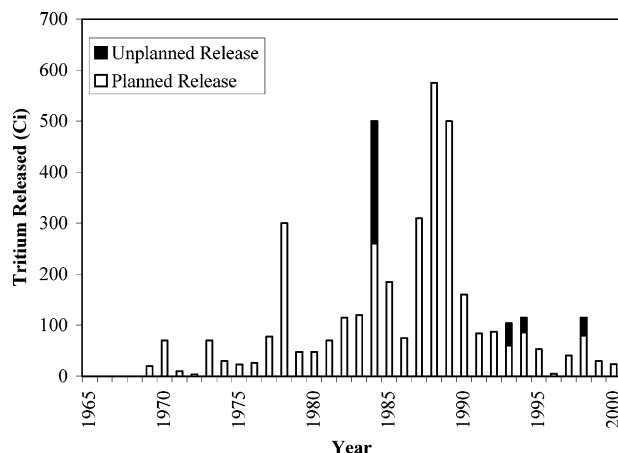

FIGURE 2. Reported ^{3}H released from Building 75 emissions stack. Planned releases are from normal operation. Unplanned releases are from human or mechanical failure.

Methods

Accelerator mass spectrometry (AMS) provides an analytical method that eliminates the decay counting inefficiencies for radionuclide measurement, by directly counting the isotope of interest. For OBT determination within tree rings, the smaller sample sizes permitted by AMS enable sample collection using a narrow diameter increment borer, which is nondestructive, and sample preparation using only milligrams of wood. Relatively rapid analysis using AMS also enables the analysis of 50−100 samples each day. Thus, measuring ^{3}H from tree rings is facilitated by the use of this methodology for exposure reconstructions.

Samples were collected for ^{14}C and ^{3}H analysis on two occasions, September 25 and December 18, 2001. Tree cores 244−400-mm-long were collected using a 500-mm-long increment borer that was 5.15 mm diameter. During the first sampling, two trees were sampled. One tree was a *Pinus jeffreyi* (Jeffrey pine) located upwind and downhill of the ^{3}H stack (Figure 1). The second tree was a *E. globulus* 15 m northwest of the emission stack (eucalyptus A), directly uphill from the stack (Figure 1). Two cores were collected from

each tree, one for ^{3}H analysis and one for ^{14}C analysis. During the December sampling, an additional core was sampled from eucalyptus A to obtain greater resolution in ^{3}H measurements and to verify some of the ^{14}C measurements. Two additional eucalyptus trees were also sampled: one tree located 50 m directly uphill from the stack (eucalyptus B) and a tree located 50 m northeast of the stack at the same elevation (eucalyptus C) (Figure 1). Each tree had one core sampled, as there was enough wood to measure both ^{14}C and ^{3}H.

In the laboratory, the cores were lightly sanded to remove surface contamination and subsequently separated using a scalpel. Because ^{14}C was used only to estimate the age as a function of distance from the bark, a continuous ^{14}C record was not necessary, and 2-mm discrete samples were taken at regular intervals throughout the cores. As a consequence, there were intervals of each core without ^{14}C measurements. In contrast, all sections of the core were measured for ^{3}H to provide a continuous exposure history. Prior to making ^{3}H measurements, ^{14}C profiles were utilized to determine the growth rate of the tree. These growth rate estimates were then used to equally space 100 samples for ^{3}H analyses between the bark and the point in the core corresponding to the year 1950. This sub-annual sampling interval provided on average 2 samples/yr.

Samples were pretreated to remove most noncellulose components using an acid/base procedure (*9*). To remove any ^{3}H that had equilibrated onto exchangeable sites in the cellulose, the wood was immersed with deionized water at 90 °C for 8 h, the water was removed, and the process was repeated (*10*). The samples were then dried on a heating block overnight at 90 °C to remove water. Dried cellulose was then converted to titanium hydride for ^{3}H-AMS analysis (*11*) or graphite for ^{14}C AMS analysis (*12*), both performed at the Center for Accelerator Mass Spectrometry at Lawrence Livermore National Laboratory.

The measured ^{3}H values are reported as tritium units (TU) for the year 2002. Eucalyptus A and C were measured at 2-mm intervals, whereas eucalyptus B was measured at 3-mm intervals. The measurements of ^{3}H from the three eucalyptus trees are reported after subtracting the average ^{3}H contamination in the sample preparation blanks that result during sample preparation procedures. The innermost section of each core was used as sample preparation blanks because the wood was formed before the 1960s and should contain very low amounts of ^{3}H. The 10 samples closest to the core were used as sample preparation blanks for each eucalyptus core. These blanks had background levels for eucalyptus A of 2000 ± 400 (1 SD) TU, eucalyptus B of 900 ± 80 TU, and eucalyptus C of 700 ± 300 TU. This background level of ^{3}H is greater than the AMS background (<100 TU) and is thought to result from contamination in LLNL laboratory facilities also employed for biological tracing experiments using tritium-labeled compounds. The quantitation limit (QL) for each core was determined using 5σ of the sample preparation blank, except the standard deviation of the blank was unusually low for eucalyptus B, so the QL from eucalyptus C (which has similar levels of background contamination) was also used for eucalyptus B. As a result, 2000 TU is the QL for eucalyptus A, and 1500 TU is the QL for eucalyptus B and C. Because the inner rings of the pine were not older than 1960, 1500 TU was also used as the QL for the pine ^{3}H measurements.

Results and Discussion

^{14}C. Because the eucalyptus tree rings were diffuse and the annual nature of the rings was in doubt, ^{14}C measurements enabled growth rate determination. Although ^{14}C is typically used for dating samples thousands of years old, the spike in $^{14}CO_2$ levels in the atmosphere from above-ground nuclear
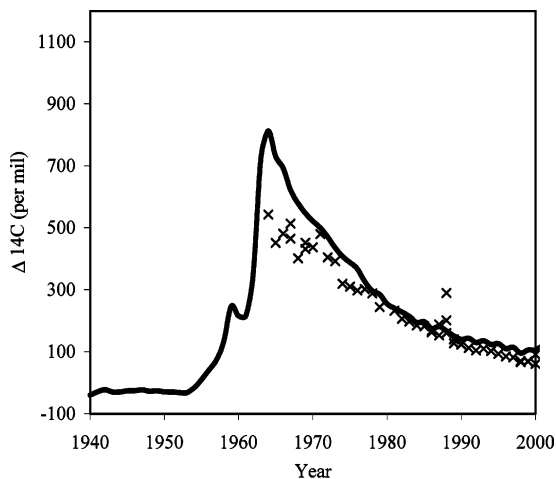
FIGURE 3. Comparison of pine tree ^{14}C upwind of NTLF with Northern Hemisphere clean-air ^{14}C measurements.

weapons testing allows age dating of samples less than 50 years old (*13*). Because trees obtain almost all of the carbon for photosynthesis from atmospheric CO_2, the ^{14}C of tree wood reflects the ^{14}C in the atmosphere around the tree. The atmospheric ^{14}C record when unaffected by fossil fuel combustion is referred to as clean-air ^{14}C. The age of the wood is determined matching measured ^{14}C levels with the historical record of atmospheric ^{14}C. The ^{14}C data are reported in delta notation (Δ^{14}C) (eq 1), which compares the sample ^{14}C content to the standard reference material that represents the ^{14}C content of the 19th century atmosphere prior to its dilution by large-scale fossil fuel CO_2 releases and referred to as 'modern carbon'. Delta notation is used to correct for operational differences in isotope ratio mass spectrometry by reporting data as the difference between the measured ^{14}C/^{12}C ratio (R_{sample}) of the sample and a known reference material ($R_{standard}$) normalized by the measured ratio of the known reference material. Since these differences are usually very small, results are expressed as part per thousand or per mil (‰) notation by multiplying by a factor of 1000:

$$\Delta^{14}\text{C (‰)} = \frac{R_{sample} - R_{standard}}{R_{standard}} \times 1000 \qquad (1)$$

The radial profile of ^{14}C in the eucalyptus cellulose is matched with known atmospheric ^{14}C levels over the last 60 yr. The ^{14}C levels in the upwind pine tree were used to determine the background ^{14}C levels for this location because the annual tree rings of the pine tree are not in doubt.

The 38 annual rings in the Jeffery pine spanned a radial distance of 244 mm, indicating a growth rate that averaged 6.5 mm/yr. The ^{14}C in these rings closely matches the clean-air atmospheric ^{14}C between 1970 and 2000 (Figure 3). The ^{14}C in the earliest rings (i.e., 1964–1969) is lower than clean-air atmospheric levels of ^{14}C during those years. These lower ^{14}C levels are the result of sources that are depleted in ^{14}C that may have resulted from fossil fuel combustion at the site (*14, 15*), as the tree is less than 1 m from a road. The tree ring that corresponds with the year 1988 has an elevated amount of ^{14}C as compared to clean-air atmospheric levels. Although one of the samples from this ring is similar to clean-air ^{14}C levels, two of the samples are elevated. Because these samples are not continuous over the entire ring width, the different ^{14}C values may represent intra-annual variability and a localized source of ^{14}CO$_2$ as discussed later.

The details of the growth rate determination for the eucalyptus are in the Supporting Information Section A, and the results are discussed below. An initial inspection of the eucalyptus A core yielded 67 rings in 300 mm or an average
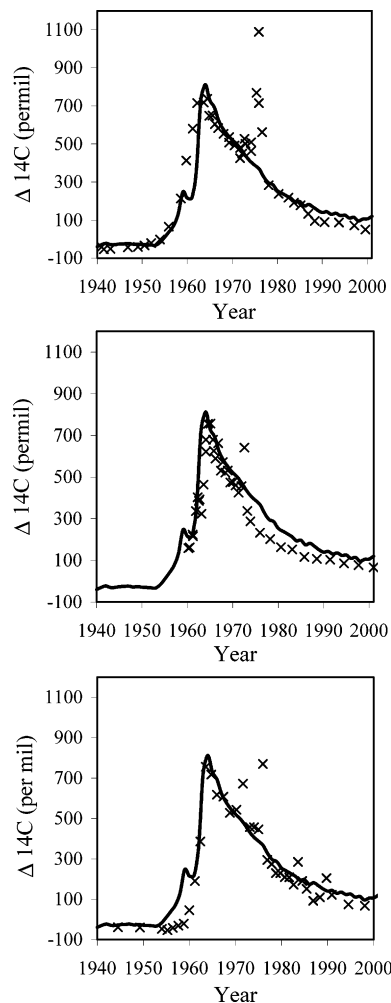

FIGURE 4. Comparison of eucalyptus A (top), B (middle), and C (bottom) ^{14}C with Northern Hemisphere clean-air ^{14}C measurements.

growth rate of 4.5 mm/yr but left significant uncertainty in the ability to discriminate individual rings. Matching the profile of ^{14}C to atmospheric levels (Figure 4) yields an average growth rate of 4.1 mm/yr for eucalyptus A, 4.5 mm/yr for eucalyptus B, and 4.2 mm/yr for eucalyptus C. The uncertainty in the ^{14}C age estimate is approximately 1–2 yr.

The ^{14}C profile of eucalyptus A was far above the clean-air levels between 1973 and 1977 with the maximum measurement at almost +1100‰ in 1976. This value is ~700‰ above clean-air levels during that time. Elevated ^{14}C levels in eucalyptus A are verified in a second core.

Although the overall tree ring ^{14}C pattern for eucalyptus B matches the clean-air curve, the values between 1970 and 1990 are consistently lower than clean-air levels for the corresponding years. The age of the eucalyptus B wood in this core interval would be better correlated with clean-air levels if they were adjusted a few years younger than the age assigned. An adjustment procedure using both ^{3}H and ^{14}C data is described in Supporting Information Section C. As observed in eucalyptus A, a ^{14}C measurement from eucalyptus B that corresponds to 1973 is elevated ~200‰ above clean-air levels.

The ^{14}C profile of eucalyptus C exhibits more scatter than for eucalyptus A but still resembles the clean-air ^{14}C signal. Four measurements corresponding to 1972, 1976, 1984, and 1990 were above clean-air levels. The elevated point in 1976 corresponds with the elevated interval from eucalyptus A, although the ^{14}C level from eucalyptus C is ~400‰ above clean-air levels.
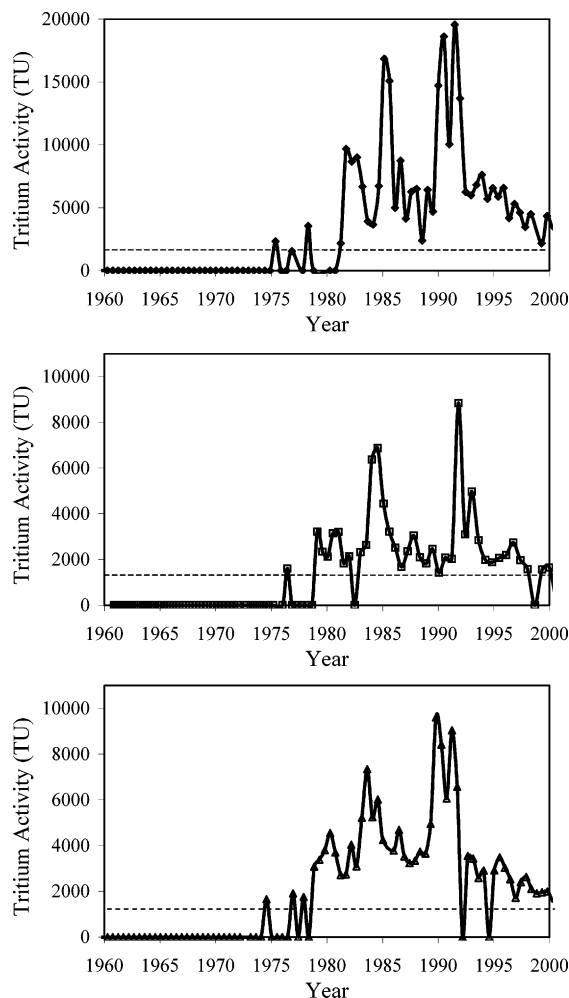
FIGURE 5. Measured ³H activity from wood of eucalyptus A (top), B (middle), and C (bottom) near the ³H stack. The preliminary estimate of the wood age was assigned by determining the average growth rate through comparisons of ¹⁴C measurements in the wood to known atmospheric ¹⁴C values over time. Quantitation limit indicated by dotted line.

³H. Tritium levels in the pine tree are below the QL of 1500 TU for all rings. The results of ³H measurements from the three eucalyptus trees are shown as a function of distance from the bark in Supporting Information Section B. Using the growth rates from the ¹⁴C data, the ³H data are assigned a preliminary growth year. The measured ³H activities from the three eucalyptus trees versus year are shown in Figure 5. OBT levels from each of the three eucalyptus trees show three major peaks, corresponding to the years 1990, 1985, and 1982. The magnitude of the ³H activity from eucalyptus A is consistently twice as large as the activity from eucalyptus B and C, which are similar. Prior to 1970, there was no quantifiable ³H in any of the trees. Tritium values below the QL are indicated on the figures by the symbol placement on the x-axis.

Results Comparison

The three prominent peaks in ³H activity from all three trees were matched with the three periods of greatest ³H releases by making adjustments in the age estimate within the uncertainty of the ¹⁴C analysis, as described in Supporting Information Section C. Because ³H sampling was sub-annual, ³H values reported for each year were interpolated from the sub-annual data to reflect the OBT pattern in the wood during the intervening years between peaks.
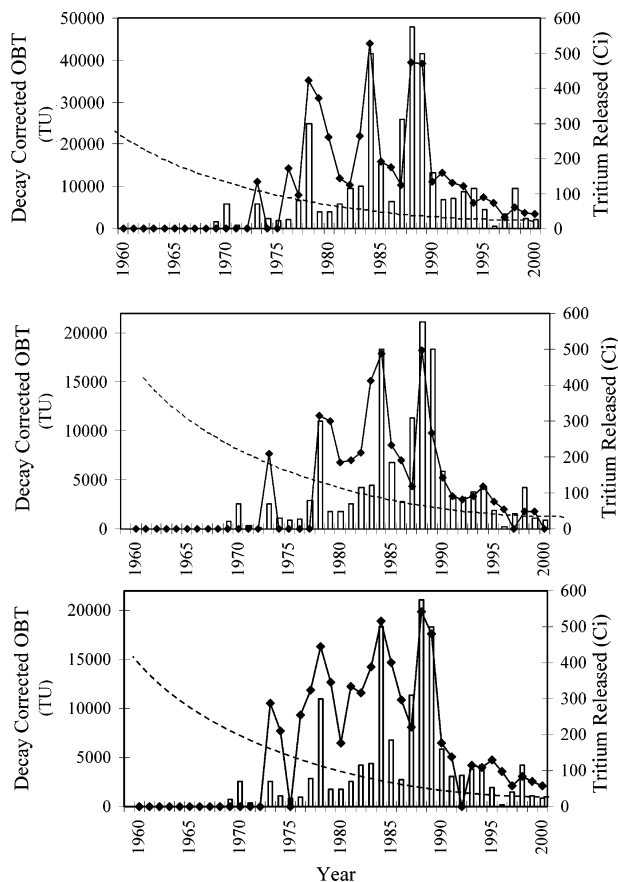

FIGURE 6. Comparison of decay-corrected ³H levels (diamonds) of eucalyptus A (top), B (middle), and C (bottom) to the total annual ³H released from the emission stack (bars). Dotted line represents decay corrected quantitation limit.

To compare the measured data with historical release records, the ³H measurements were decay-corrected to the growth year the sample represents. Figure 6 compares the decay-corrected ³H levels from each of the eucalyptus to the total annual atmospheric ³H releases (planned + unplanned releases) from B75. The relative amplitudes of the decay-corrected ³H measurements from the eucalyptus match the relative amplitudes of the reported releases throughout much of the last 30 yr. All ³H measurements prior to 1975 were below the AMS quantification limit, consistent with the start of ³H operations in 1969.

The ³H age distribution of the eucalyptus trees at LBNL can be compared with monitoring results of atmospheric water vapor using silica gel absorbents. There were three monitoring stations near the NTLF (Figure 1). The monitoring station 75EG in Figure 1 was only active in 2000 (*6*). This monitoring station had an average ³H level of 1300 TU with a maximum of 2900 TU. This amount is between the 3400 TU measured in the wood from eucalyptus A closer to the stack and levels below the quantitation limit (1500 TU) in eucalyptus B and C further away from the stack in the year 2000. Figure 7 compares the eucalyptus C ³H levels with ³H in the water vapor at location 69 in Figure 1 and at the Lawrence Hall of Science (labeled LHS in Figure 1). Although the ³H levels in eucalyptus C exceed the vapor ³H levels from 1997 to 2000, the tree data are virtually always within a factor of 3 of the annual average water vapor levels for 1975–1996. There are additional site monitoring data focused on the soil duff layer and tree leaves.

Murphy et al. (*16*) demonstrated that when ³H in the leaf water was greater than atmospheric water vapor levels, the source of ³H taken up by the plants was from the degradation
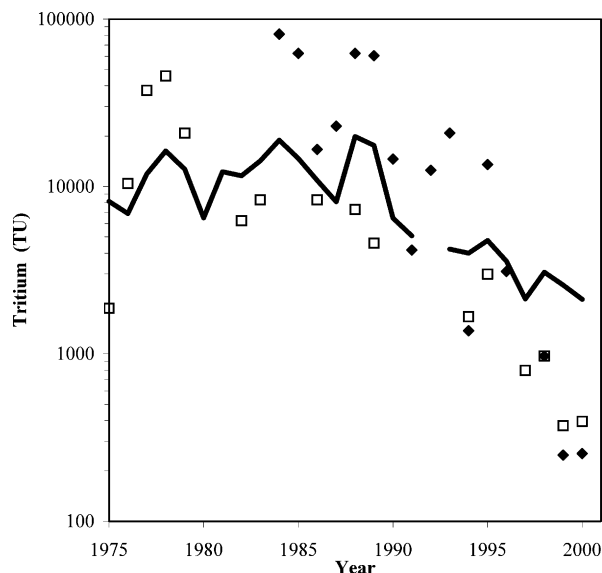
FIGURE 7. Water vapor monitoring data from locations 69 (diamonds) and LHS (squares) as compared to decay-corrected ^3H levels in eucalyptus C. Monitoring data are the annual average reported by LBNL using water vapor monitors. Years without data points and the break in the eucalyptus line represent values below the detection limit (1500 TU for eucalyptus, ~2000 TU from monitoring data between 1975 and 1991, and ~200 TU from 1991 to 2000).

of soil organic matter. Because the vegetation is the principal source of organic matter in soil, the surrounding vegetation can reutilize ^3H incorporated into organic matter from previous tritiated water vapor exposures as it is oxidized in the soil by microbes (16). Near Building 69, unbound water in the duff on the ground ranged from ~0.003 to 0.05 Bq/g of soil over the years 1996–2000. While the water content of the duff is uncertain, these values are equivalent to thousands of TU in the soil water (6). OBT levels in the leaves and duff were approximately an order of magnitude greater in ^3H activity than the unbound water (6). The soil water ^3H levels are similar to the levels measured from the eucalyptus trees (Figure 5) during the period 1997–2000 and may indicate that the litter is a relatively low-level ^3H source to the surrounding trees. Tritiated soil duff and leaves would cause the OBT levels to be higher than water vapor levels during periods where ^3H activity in the air is low but provide little contribution to exposures when atmospheric water vapor is at higher levels.

The ^3H levels from eucalyptus A are consistently two times greater than the levels measured in eucalyptus B and C further away from the emission stack. Monitoring data of OBT and free water in foliage also indicate that the ^3H levels decrease by approximately 50% between the stack and 50 m and again between 50 and 100 m (6). Reported ^3H levels from samples adjacent to the stack collected during 1996 (19 Bq g^{-1} or ~100 000 TU for OBT and ~30 000 TU for unbound water) are greater than decay-corrected measurements from eucalyptus A for that year (6000 TU) (6). Reported ^3H levels from OBT and free water samples collected near eucalyptus B were 2.4 Bq g^{-1} (or ~12 000 TU) for OBT and 3000 TU for unbound water in the wood (6) as compared with the decay-corrected measurement of 3000 TU from this study. Thus, the OBT levels in trees measured using AMS are consistent with results from other monitoring efforts using conventional quantification methods.

The age-dated ^3H measurements from the eucalyptus provide annual estimates of ^3H exposure around the stack. Modeling historical exposures at the hillside location based on stack emissions or water vapor measurements is complicated by two factors; first, the time-integrated information

of ^3H discharges from silica gel measurements does not permit mechanistic tracking of stack emission but instead relies on time-averaged parameters; second, meteorological and eco-hydrological parameters are needed at the same time scale as the discharge data and with a spatial resolution of tens of meters. Because much of this information is poorly known, use of a predictive model based on time-averaged tritiated water vapor measurements is unlikely to provide detailed and reliable historical exposure estimates.

The eucalyptus trees surrounding the stack are successful as passive monitors because they integrate ^3H exposure over the complex temporal variation in releases, wind direction, wind speed, humidity, temperature, and precipitation. Although comparisons between measured OBT data and monitoring data indicated consistent results, there is no way to determine if the tree ring contains a complete annual exposure record or if ^3H is not incorporated into the wood during periods when the tree is dormant for part of the year. Also, the ^3H recorded by the trees may come from either soil water or the air. Although the most important ^3H exposure pathway for humans at this location is via water vapor, the OBT record does not distinguish between water vapor and soil water and may at times reflect soil water ^3H levels. As with models, there are some important uncertainties in using environmental measurements for ^3H exposure reconstructions. At this location, the uncertainties in quantifying the important mechanisms at temporal and spatial scales for a predictive model are likely to be much greater than the uncertainties from the exposure reconstruction using environmental samples.

The unplanned releases appear to be incorporated into the trees as well as planned releases, except that the relative amplitudes of the 1998 ^3H peaks in the three eucalyptus trees are relatively lower than the reported release data as compared to the other years. The 1998 35-Ci unplanned released occurred while the wind was blowing away from the eucalyptus trees, according to wind direction monitors (17), which is rare at this location. This dry inland air would transport the ^3H away from the eucalyptus trees. In addition, the dry air would provide little opportunity for released water vapor to diffuse through the stomata or to condense on the surrounding leaves. Thus, measured ^3H from the eucalyptus provides a much closer match to the planned ^3H releases during 1998 than the total released ^3H. This demonstrates that the trees represent the local exposure and not just the annually averaged stack release quantity.

In addition to measuring ^3H in the trees around the ^3H stack, the ^{14}C measured in the eucalyptus trees also reflects the atmosphere in the local environment. The ^{14}C levels were expected to closely match clean-air ^{14}C measurements, but instead a few ^{14}C spikes were measured. Because the intent of measuring ^{14}C was to estimate a growth rate, there were sections in the tree core where no ^{14}C was measured.

Episodes of elevated ^{14}C are attributable to a local anthropogenic release, but neither LBNL nor the adjacent University of California at Berkeley report any significant ^{14}C releases over 1965–2000 (6, 18). Eucalyptus A had a peak ^{14}C activity in 1977 that was 700‰ above clean-air $^{14}CO_2$ levels that year. Eucalyptus B and C had large spikes in ^{14}C activity in 1976 that was 200–400‰ above clean-air values. This is a similar pattern observed with ^3H where eucalyptus A had ^3H levels a factor of 2 greater than eucalyptus B and C. The small ^{14}C spike of 100‰ in the pine tree for 1988 may correspond to the elevated level observed in eucalyptus C in 1990. Because the elevated ^{14}C detected in the trees appears to be localized in a way that is similar to local ^3H, it is likely that the source of the ^{14}C was from the stack.

The presence of anthropogenic ^{14}C around the emission stack might be explained by a hazardous waste packaging facility that vented room exhaust through the same emission

stack at B75. Thus, any volatile ^{14}C released in the facility would have been vented into the local environment. Because ^{14}C is used extensively in biomedical and environmental tracer research, it would not be surprising for it to be in hazardous waste at a scientific laboratory. The release of anthropogenic ^{14}C from hazardous waste was recently documented at Oak Ridge Reserve through the analysis of tree cellulose (19). Although the absence of coincident spikes in each of the trees leaves some questions over the historical magnitude and distribution of ^{14}C in the local environment, the existence of the large 1976 spike in eucalyptus A is fairly certain because it was measured multiple times from two different cores of the same tree. The rapid decline in elevated ^{14}C at the other trees also supports the stack as the likely source.

Traditional quantification using decay methods would have been impractical for this reconstruction because of the time and labor involved in sample collection, processing, and analysis. Instead, accelerator mass spectrometry permits a reasonable turnaround time for a relatively large number of samples. In this study, hundreds of samples ($\sim$150 for ^{14}C and $\sim$400 for ^3H) were prepared and analyzed by a single investigator within 6 months.

Exposure reconstructions are an important tool for addressing concerns of historical exposure. The ^3H that is organically bound to the cellulose in wood provides an accurate record of the ^3H exposed to the tree on an annual basis. This study determined the temporal and spatial distribution of ^3H near an emission stack and provided an independent verification of historical release data. Because of difficulties with mathematically modeling the episodic ^3H releases with complex topographical and meteorological variability, these environmental measurements are likely to be the only reliable and independent assessment of historical exposure. The reconstruction was able to address many of the uncertainties and concerns associated with historical releases and demonstrates the consistency of passive monitors to record ^3H exposures in the environment around this emission stack.

Supporting Information Available

The methodology used to determine growth rates from the ^{14}C measurements, the OBT data from each eucalyptus tree as a function of radial distance in from the bark, and the age adjustments of tritium measurement based on both ^3H and ^{14}C results. This material is available free of charge via the Internet at http://pubs.acs.org.

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