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Improving tritium exposure reconstructions using accelerator mass spectrometry

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Abstract

Direct measurement of tritium atoms by accelerator mass spectrometry (AMS) enables rapid low-activity tritium measurements from milligram-sized samples and permits greater ease of sample collection, faster throughput, and increased spatial and/or temporal resolution. Because existing methodologies for quantifying tritium have some significant limitations, the development of tritium AMS has allowed improvements in reconstructing tritium exposure concentrations from environmental measurements and provides an important additional tool in assessing the temporal and spatial distribution of chronic exposure. Tritium exposure reconstructions using AMS were previously demonstrated for a tree growing on known levels of tritiated water and for trees exposed to atmospheric releases of tritiated water vapor. In these analyses, tritium levels were measured from milligram-sized samples with sample preparation times of a few days. Hundreds of samples were analyzed within a few months of sample collection and resulted in the reconstruction of spatial and temporal exposure from tritium releases. Although the current quantification limit of tritium AMS is not adequate to determine natural environmental variations in tritium concentrations, it is expected to be sufficient for studies assessing possible health effects from chronic environmental tritium exposure.

Keywords

Tree rings; Tritiated; Water vapor; Groundwater

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Introduction

Exposure to radionuclides almost universally causes concern with the public. This concern may be based on reasons such as the fear of cancer or other forms of genetic damage, suspicion of any work performed with radionuclides, or repulsion from images of nuclear weapons testing and humans with severe radiation exposure. Studies have demonstrated that the public's perception of relative risk associated with radiation exposure is disproportionate to the actual risk [1]. As a result, anthropogenic radionuclide exposure remains a unique category of risk that receives greater attention than the risk posed by many other contaminants. Once a radionuclide source is identified as an exposure risk, government regulations act to prevent adverse health effects through the establishment of discharge limits and monitoring guidelines [2]. Because thorough monitoring is the best method of evaluating current exposure to identified contaminants and regulations are generally effective at limiting radionuclide exposure to safe levels, often the most significant exposures could have occurred before the contaminant was identified or adequately monitored. Thus, reconstructed exposure levels are often needed to provide a comprehensive assessment of the potential health impacts for affected populations.

It is not possible to directly measure past exposure. Consequently, surrogate measurements are used to reconstruct contaminant levels. Methods for reconstructing exposure utilize epidemiological data, biomarkers, mathematical models, and environmental measurements [3,4]. The data for each technique are fundamentally different, permitting exposure reconstructions using the information that is available. Thus, reconstructions have been performed using various methodologies for radionuclides at many locations throughout the world [5]. Numerous efforts have led to radionuclide reconstructions that originate from intentional releases, such as atmospheric nuclear weapons detonation at the Nevada test site [6,7,8], the Marshall Islands [9,10], the Semipalatinsk Polygon [11], or Hiroshima [12,13,14,15,16]. In addition, there are studies of releases from operational or mechanical failure, such as accidents at Chernobyl [4,17,18] and Three Mile Island [19,20]. Most other radionuclide reconstructions have been performed at facilities involved with processing nuclear materials, such as such as Hanford Works [21], Fernald Feed Materials Production Center [22], Rocky Flats [23], Oak Ridge [24], Idaho National Engineering Laboratory, Savannah River, Chalk River Nuclear Laboratories in North America, and the Mayak Industrial Association [25,26] in former Union of Soviet Socialist Republics. As early as the 1940s, individuals at these facilities were exposed to plutonium, uranium, cesium-137, strontium-90, iodine-131, and/or tritium both within and outside the plant boundaries. A few other exposure reconstructions have been performed for radon-222 and cobalt-60 from contaminated building materials [27, 28] These reconstructions have been effective in narrowing estimates of the total radionuclide exposure, but few have been able to reconstruct the temporal variation of the radionuclide exposure over the duration of the activities.

Rarely are all of the data needed for each reconstruction available. Despite the need for reliable historical contaminant exposure data, there are relatively few methodologies that utilize environmental measurements in exposure reconstructions. Environmental measurements collected to directly answer the questions of exposure generally have less uncertainty than models that incorporate estimated parameters [29]. Under these situations, development of methodologies using environmental measurements are needed that are fast, accurate, and easily understood by the general public [30].

Because of its discharge from nuclear fission/fusion research, use in consumer products, and its utilization as a biological or hydrologic tracer, a radionuclide of recent exposure interest is tritium. Public health concerns over the historical releases of tritium near residential or agricultural areas have generated demands for better exposure reconstruction techniques [31].

Nuclear power plants, nuclear weapons laboratories, and industrial operations have discharged tritium into the environment for over 50 years. Tritium continues to be generated and released from energy, industrial, and military operations that are likely to increase over time as our reliance on nuclear fission/fusion increases. Although monitoring is capable of determining contemporary tritium exposure, in many cases historical monitoring and release data are unavailable or suspect. In addition, mathematical models are dependent upon estimates of the release history and if there are large uncertainties, they will propagate the uncertainty into historical exposure estimates. Thus, using environmental measurements could provide the most robust and accurate method for reconstructing historical tritium exposure concentration and discharge.

Reconstructing tritium exposure concentrations using tree rings

Trees have been used in many studies as a natural collection system for dating environmental samples. Transpired water is pumped directly from soil water and groundwater in hours to days [32]. Water vapor in the air can diffuse through stomata and rapidly equilibrate with the leaf water [33,34]. Tree leaves and pine needles have been used as an indirect, short-term monitor of contaminants in the groundwater or air [35,36]. In addition, tree rings can integrate annual exposures to chemicals in their immediate environment and preserve that record in individual tree rings [37,38,39]. Thus, trees act as a passive monitor for environmental exposures and in most cases contain the only long-term exposure record.

Tritium exposure is particularly difficult to reconstruct using environmental measurements because of its low environmental concentrations, mobility, and the low-energy of its radiation. Tree rings are one of the few environmental media that sequesters tritium in a form where the temporal variation in exposure can be reconstructed. Some spatial and temporal tritium signals may be maintained in some unsaturated soil and sediments, but it is more complicated to reconstruct exposure concentrations from measurements of this nature. Tree ring studies have been used to demonstrate the ability of trees to record the tritium levels in the surrounding environment under different conditions. The use of tree rings to study environmental levels of tritium was originally reported in 1961 [40]. Brown [41] demonstrated that organically bound tritium (OBT) from tree rings was the same as tritium levels in precipitation utilized by the tree. Brown also reported the first use of trees as monitors for local anthropogenic tritium exposure. A series of studies by Kozak et al. [42,43,44,45] demonstrated that tree rings had the precision and sensitivity for recording anthropogenic exposure events that occurred during a single year. Other authors [46,47,48] have also used tree rings to reconstruct anthropogenic tritium exposure.

Although studies have demonstrated the ability of trees to act as passive monitors for historical tritium exposure concentrations from local point sources, the time and effort required for these results are prohibitive for its routine use as an assessment tool. Each of the studies reviewed above used decay counting methods for quantifying the tritium activity in the tree ring samples. Because decay counting methods are limited by the inefficiency of measuring the 0.01% of tritium atoms that decay per day, grams to kilograms of wood were used in order to achieve an adequate tritium count rate. Tree ring samples of this size require a tree to be cut down and the separation of the entire circumference of each ring for analysis. As a result, this procedure is too time-consuming and labor-intensive for use at multiple locations near a point source. In addition, if a tree were required to be cut down at each sampling location, the spatial coverage for a geographically limited exposure might remove a significant number of trees in the surrounding community.

Tritium AMS

Accelerator mass spectrometry (AMS) provides an analytical method that eliminates the decay counting inefficiencies for radionuclide measurement. AMS increases measurement efficiency by directly counting the individual ions of the measured isotope. As a result, AMS is typically able to provide more rapid analysis with greater measurement sensitivity using much smaller samples. For OBT measurements, the smaller sample sizes needed for AMS allow collection and sample preparation to be simplified through the use of an increment borer for collecting the annual rings and sample preparation of milligrams instead of kilograms of wood. Improvements in analysis time using AMS increase the number of samples that can be measured each day. In addition, collecting samples using an increment borer is nondestructive. Thus, AMS for measuring tritium from tree rings can facilitate the use of this methodology for exposure reconstructions.

Accelerator mass spectrometry utilizes a high-energy isotope ratio mass spectrometer [49]. Traditional isotope ratio mass spectrometers cannot distinguish ions that have the same charge-to-mass ratio, but by accelerating the ions to MeV energy levels, AMS distinguishes elements with the same charge-to-mass ratio by measuring differences in energy loss inside the detector. The ratio of the radioisotope of interest to a stable isotope from each sample is measured continuously to normalize instrumental and sample heterogeneity. These ratios are then compared to the measured ratios from standards to quantify the results. Results are reported in Tritium Units (TU), where 1 TU is defined as a ${}^3\text{H}:{}^1\text{H} = 10^{-18}$. 1 TU is equal to 0.12 BqL^{-1} . AMS has demonstrated good accuracy and rapid instrument analysis time at very low radionuclide levels [50]. The electrostatic accelerator and negative ion source within the Center for Accelerator Mass Spectrometry (CAMS) at Lawrence Livermore National Laboratory (LLNL) have been used in this study to measure the OBT in tree ring samples.

Sample preparation for tritium AMS analysis requires that OBT be quantitatively converted into a solid form that is compatible with the negative ion source. Robust methods exist in deuterium to hydrogen isotope ratio mass spectrometry (D/H) for converting organic samples into a gaseous form [51,52,53,54]. For AMS analysis, sample preparation is based on these D/H methods with an additional step that incorporated the gaseous sample into titanium, forming titanium hydride. Details of the method are described in Chiarappa-Zucca et al. [55]. Typically, samples contain from 100ng to 1mg of hydrogen if the starting material contained approximately 1mg to 10mg of water or organic material. Current preparation methods limit the sample size to a maximum of 10mg. Below 1mg, it is difficult to produce enough titanium hydride to provide adequate hydrogen current in the AMS system. Samples with as little as 10 μg of hydrogen have been prepared (0.1mg of organic material) with the addition of tributyrin as a hydrogen carrier. However, this decreases the method sensitivity.

The performance of the tritium AMS method determines the range of applications where it is useful and whether it provides analytical capabilities that are better than current techniques. Samples prepared using the method described above typically have H^- currents of 30–60 μA , which is similar to the currents reported by Middleton [56]. Each sample is measured 3–7 times for a 5-min duration each time. The quantification range is from 10^3 to 10^8 TU with variances typically 5–8%, unless the measurement is limited by counting statistics. The sample quantification limit (QL) of 10^3 TU (5 sigma of multiple sample preparation blanks) is a result of laboratory background contamination, as titanium hydride bought from chemical supplier has a QL of ~ 70 TU and likely reflects the current quantification limits of the AMS system, not tritium contamination of the commercial product.

Alternative methods for quantifying tritium activity involve either the detection of the energy released during radioactive decay (liquid scintillation counting/gas proportional counting) or

the subsequent accumulation of decay products (helium-3 in-growth mass spectrometry). The typical instrument QL of 3 TU for decay methods using 25 grams of sample [57] or 0.01 TU for helium-3 in-growth mass spectrometry using 0.5–1 kg of sample [58,59] are much lower than the 70 TU AMS system QL for milligram-sized samples. These methods are only able to achieve these levels with much larger samples than those used in AMS. Because tritium AMS measures the tritium to hydrogen ratio in the sample, it does not benefit from larger samples. Electrolytic tritium enrichment of kilogram-sized samples can decrease the QL of decay counting to 0.1 TU [60]. Similarly, larger samples have the potential to significantly decrease the tritium AMS QL for samples that are tens to hundreds of milligrams in size, although this ability has not been developed. Helium-3 in-growth mass spectrometry can measure samples with as few as 1×10^6 tritium atoms, whereas tritium AMS has measured down to 3×10^5 tritium atoms and lower levels may be possible if sample preparation QL limits can be decreased. To compare measurement efficiency of these methods for milligram-sized samples, a 2-mg sample of water with an activity of 10^4 TU has 0.1 decays per minute for decay counting or helium-3 in-growth mass spectrometry compared to a count rate of 50–100 counts per minute using tritium AMS. There would be a 500–1000 fold decrease in analysis time for tritium AMS compared to the instrument time required for decay counting or the in-growth time required for helium-3 mass spectrometry to count the same number of atoms for quantification.

Applications

Tritium reconstruction from discharged water

The Nevada test site has been extensively used for studying the subsurface transport of radionuclides resulting from nuclear weapons testing. Since 1962, there have been over 600 subsurface nuclear weapons tests, 30% of which were below the water table [61]. To study radionuclide migration at the site of the Cambrian detonation, the groundwater was pumped nearly continuously for sixteen years, from October 1975 to 1991 [62]. The tritium activity in the pumped water was measured at least monthly during the 16-year experiment. Approximately 50 m from the wellhead, the pumped groundwater was discharged into an unlined ditch that flows along the desert floor. The activity rapidly peaked at 3×10^8 Bq m⁻³ (~2,000,000 TU) and slowly decayed in a manner typical of elution processes in pumped aquifers [62]. Over the course of this experiment, *Tamarix ramosissima* (salt cedar) grew along the length of the ditch. Even after the end of groundwater pumping, salt cedar continued to produce new leaves. The net evapotranspiration rate greatly exceeds the average annual precipitation, so little water is available to these trees from rainfall. As a result, their annual rings are expected to contain the time-varying tritium signal from the pumped groundwater. Love et al. [63] describe sample collection, processing and tritium analysis.

Figure 1 compares the tree ring tritium measurements to the tritium level in the monitored water. Sample preparation blank values averaged 1200 TU and were subtracted from the measured tritium values in the unknown samples. For this comparison, the tritium activity of the water has been decay corrected to the year 2000, the year of OBT analysis in the tree rings. The OBT activity in the tree rings was corrected for the 30% of hydrogen atoms that are exchangeable through equilibration based on the stoichiometry of cellulose. There is good agreement between the organically bound tritium content in the tree rings and the tritium content from the tree's source water. A strong correlation is observed for samples in which both pumped groundwater records and AMS measurements overlap (correlation coefficient of 0.88). This tree, exposed to a relatively high activity tritium source that slowly changed over time, demonstrates how environmental tritium exposure from a single source of tritiated water can be reconstructed using tritium AMS.

Tritium reconstruction from discharged water vapor

The OBT analysis for the tree at the Nevada test site was relatively simple because there were no other sources of water for the tree to utilize. A second example provides a more complicated scenario of a water vapor source of tritium in a more temperate climate. The tritium-labeling facility at Lawrence Berkeley National Laboratory (LBNL) was primarily responsible for synthesizing organic molecules for biomedical research where tritiated molecules can be identified and tracked through complex biological transformations. Although most of the tritium that is not used from this process is recycled, some is released into the atmosphere through an emissions stack as tritiated water vapor during the period of operation from 1969 to 2001. As a result, tritium activities above those currently encountered in natural precipitation have been measured in the air, soil, and vegetation near this location [64]. The tritiated water vapor discharge stack is surrounded by a grove of *Eucalyptus globulus* (Eucalyptus), in which three trees were used for the reconstruction. An assessment of historical tritium releases was performed for comparison with historically reported annual quantities of tritium released to provide a verification of more recent monitoring efforts and evaluate releases during the early history of the facility.

The details of this exposure reconstruction are in Love et al. [65]. Figure 2 compares tree measurements to the total amount of tritium released during that calendar year. To compare the measured data with historical release records, tritium measurements were decay-corrected to the year the sample represents. The relative amplitudes of the decay-corrected tritium measurements from the eucalyptus correlate with the release records throughout the last 40 years. The reconstruction shown is from the Eucalyptus closest to the emission stack, but two other Eucalyptus further away from the stack also had tritium activities that were well-correlated to reported discharge data but with lower tritium levels than the closest tree.

The small sample size permitted by AMS enables very small rings to be measured or sub-annual measurements to be performed. The Eucalyptus at this location has a radial growth rate of ~4-mm per year and was sampled at 2–3-mm intervals for the entire length of the core. Core sections 2-mm wide from a core with a 5.4-mm radius provided much more sample than was needed for the analyses. In order to examine the sub-annual variability of tritium in the wood, small sections of a core from the Eucalyptus closest to the stack were measured at a 0.5-mm sampling interval. Because two different cores were used for the comparison, the compared distances may not be the exact same interval from the tree because it is difficult to determine corresponding intervals with 0.5-mm precision. The comparison between the 2 mm measurements and 0.5-mm sampling interval is shown in Fig. 3. Although the smaller sampling intervals captures some sub-annual variation that 2-mm sampling interval does not, the 2-mm sampling interval is sufficient to integrate the sub-annual variation and is adequate for reconstructing average annual exposure. These results also demonstrate the ability to precisely measure tritium profiles using 0.5-mm core widths.

A mechanistic model for the transport and fate of the tritium vapor released from the emission stack would require a quantitative understanding of many process that are difficult to quantify at the relevant spatial and temporal scales. As a result, these environmental measurements are likely to be the only reliable assessment of historical anthropogenic exposure at this location.

Future directions

Complex pathways in environmental systems can be better understood through the use of isotopic labels. Although the focus of this study is on reconstructing tritium exposure, the ability of tritium AMS to rapidly measure tritium from small samples with relatively low activity permits greater spatial and temporal resolution for tritium tracer experiments using

lower amounts of radiation in hydrological, chemical, and biological research. Tritium AMS has possible environmental applications in:

1. understanding water transport in the upper soil zone on millimeter-scales without the production of waste that is classified as radioactive,
2. identifying the fate of labeled contaminants in natural or engineered systems,
3. measuring the kinetics of microbial processes, and
4. studying environmental toxicology at environmentally-relevant doses and without significant radiation risk.

Therefore, the development of tritium AMS as a robust and routine analysis technique provides an opportunity for addressing a greater array of environmental tritium applications.

Although tritium AMS is an improvement over decay methods for milligram-sized samples, current limitations of the sample preparation were unable to quantify natural levels of tritium in the environment. Currently, tritium AMS is limited to anthropogenic tritium levels because of the elevated background levels resulting from tritium sample preparation procedures. Tritium background levels could be lowered if samples were prepared in facilities where background tritium levels were lower. Combining improvements to eliminate the background tritium contamination during sample preparation and tritium enrichment of environmental samples would permit quantification of natural levels of tritium from samples tens to hundreds of milligrams in size. Tritium enrichment currently is performed on samples with natural levels of tritium undergoing tritium decay counting and provides approximately 100-fold enrichment. Although these enrichment procedures involve tens to thousands of grams from samples, the techniques already developed provide a model for the enrichment from samples only tens to hundreds of milligrams. Implementation of these improvements to the tritium AMS capability would then permit the use of tritium AMS instead of decay methods in a broader range of applications.

Summary

Accelerator mass spectrometry for tritium analysis allows the practical use of tree rings for tritium exposure reconstructions. The tritium that is organically bound to the cellulose in wood provides an accurate record of the tritium exposed to the tree on an annual basis. Traditional tritium quantification using decay methods often is impractical for use in tritium reconstruction because of the time and labor involved in sample collection and analysis. Instead, the development of a robust method to count the tritium atoms from the sample directly using AMS decreased the sample size required for analysis. As a result of this improvement, the use of tree rings for exposure reconstruction is more practical because 1) a small core from each tree could be sampled instead of felling the entire tree for analysis 2) small samples are easier to prepare for analysis and 3) the rapid analysis time for small samples allows a reasonable commitment of instrument time while still providing data for spatial and time resolution. This has been demonstrated for the tritium reconstruction at Lawrence Berkeley National Laboratory, where cores were collected and ~400 samples were prepared and analyzed by a single investigator within a few months. Applications of tritium as a tracer are expected to also benefit by the ability to rapidly measure tritium from milligram-sized samples.

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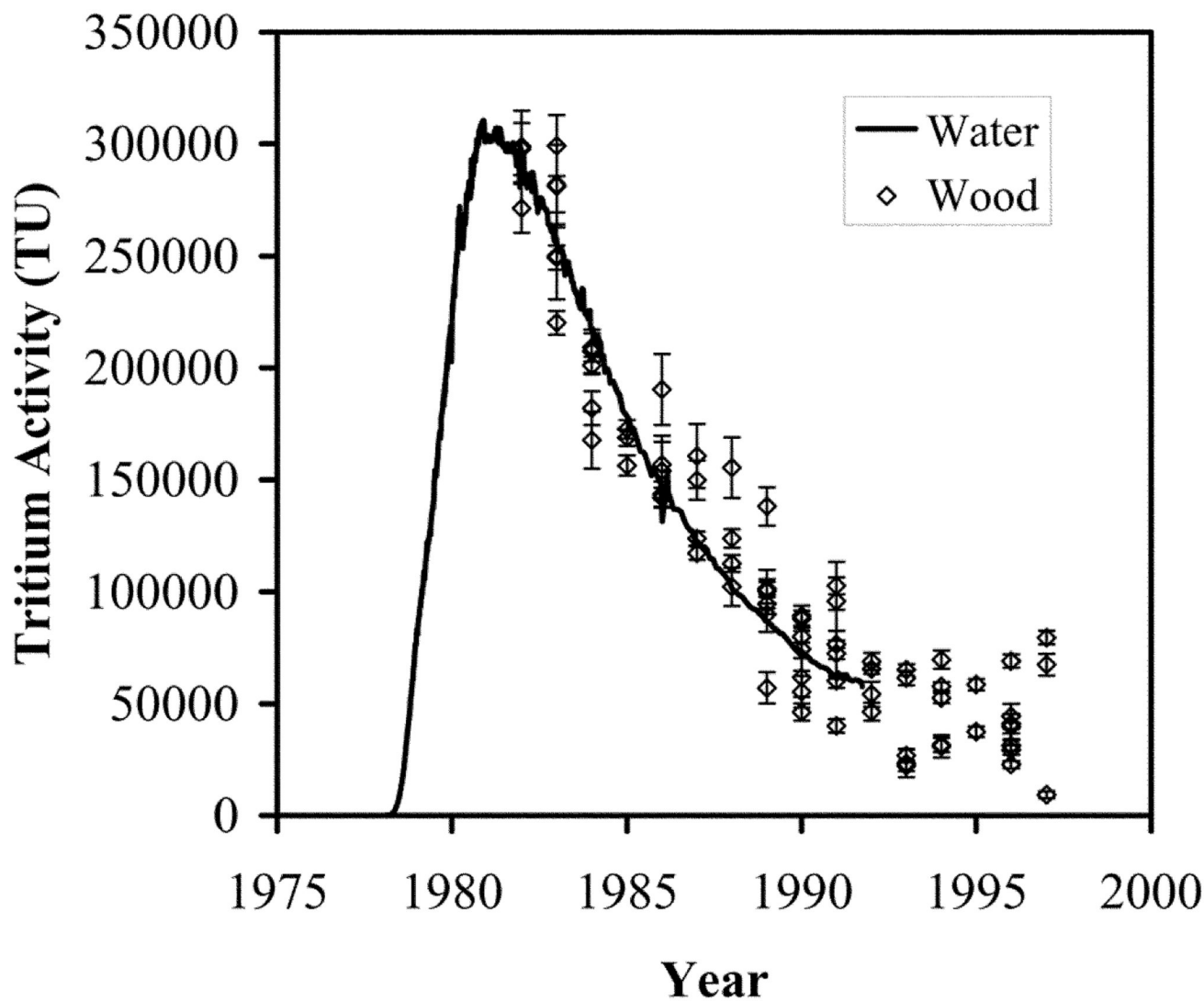


Fig. 1. Tritium levels measured in water from an unlined ditch (*line*) compared with the organically bound tritium (OBT) levels measured in the annual rings of a *Tamarix ramosissima* growing in the ditch. (Reprinted with permission from Ref. [63]. Copyright 2002 American Chemical Society)

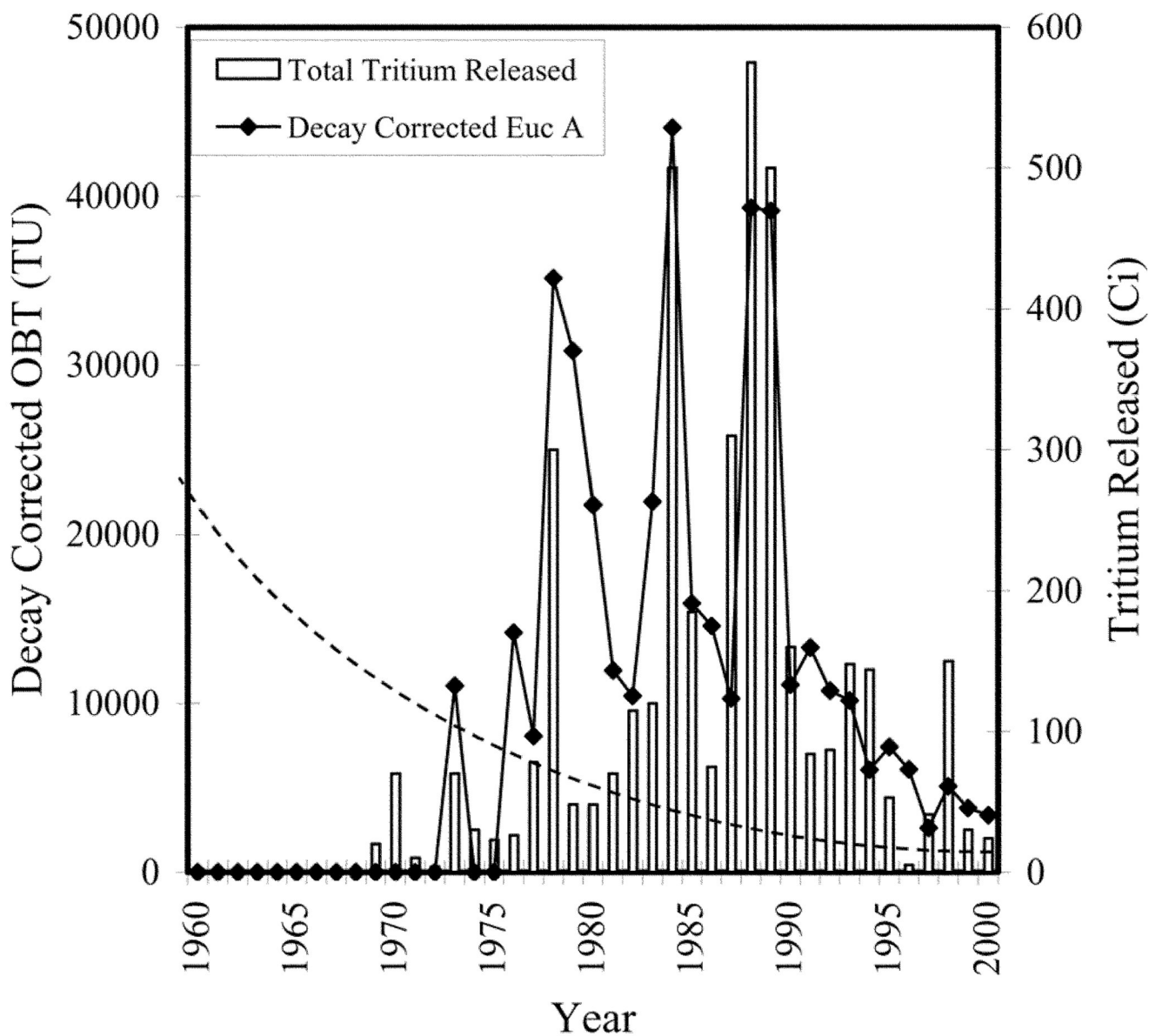


Fig. 2. Comparison of decay-corrected tritium levels in *Eucalyptus globulus* downwind from a tritium emissions stack at Lawrence Berkeley National Laboratory with reported total annual tritium released. The dotted line represents the decay-corrected quantification limit. (Reprinted with permission from Ref. [65]. Copyright 2003 American Chemical Society)

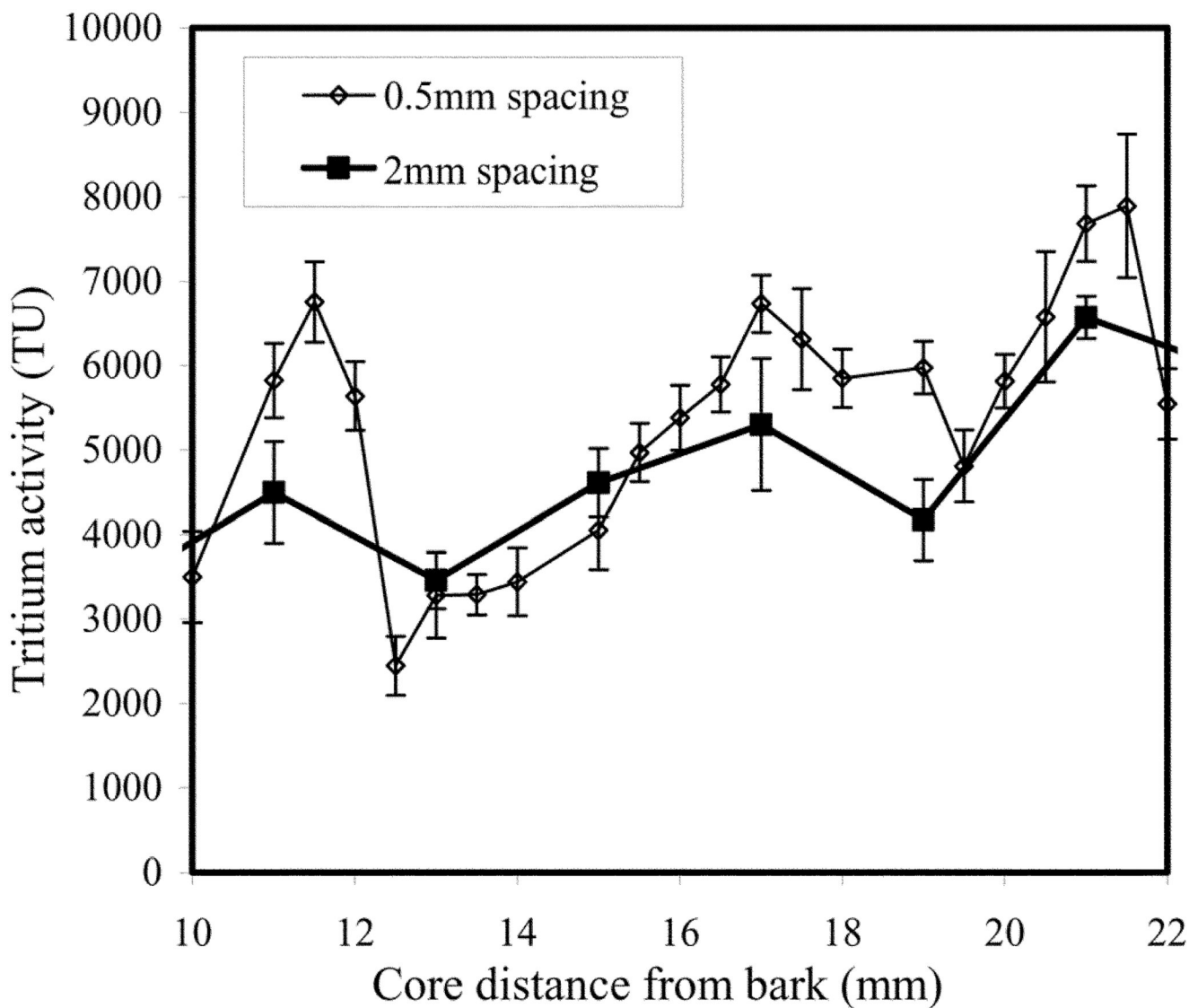


Fig. 3. Tritium measurements using 0.5-mm and 2-mm sampling intervals from two different cores of *Eucalyptus globules* downwind of a tritium emission stack from Lawrence Berkeley National Laboratory. Radial growth rate is ~4 mm per year. From Love [66]