

Tracing the History of Nuclear Releases: Determination of ^{129}I in Tree Rings

USHA RAO,^{*,||} UDO FEHN,[†]
 YASUYUKI MURAMATSU,[‡]
 HEATHER MCNEIL,^{†,¶}
 PANKAJ SHARMA,[§] AND DAVID ELMORE[§]

Department of Chemistry, Saint Joseph's University, Philadelphia, Pennsylvania 19131, Department of Earth & Environmental Sciences, University of Rochester, Rochester, New York 14627, National Institute of Radiological Sciences, Anagawa 4-9-1, Inage-ku, Chiba, 263-8555, Japan, and PRIME Lab, Purdue University, West Lafayette, Indiana 47907

Concentrations of the long-lived radioisotope ^{129}I were measured in dated tree rings in order to determine whether the distribution of this isotope reflects the history of nuclear deposition. ^{129}I concentrations and $^{129}\text{I}/^{127}\text{I}$ ratios were analyzed in tree rings and bark samples from four trees at West Valley, NY, and from one tree at Rochester, NY. West Valley was the site of short-lived nuclear fuel reprocessing activities (1966–1972), while Rochester, located 115 km to the northeast, provided a regional control site for the study. The selected trees reflect different modes of fluid and nutrient transport in trees, with three species of ring-porous trees (elm, oak, and locust), one semidiffuse (cherry), and one diffuse-porous tree (maple). The results show that ^{129}I levels in ring-porous trees, in which xylem or hydrologic tissue is localized in the outermost growth ring, are generally well correlated with the expected ^{129}I deposition pattern for the region. In contrast, tree rings of the more common semidiffuse to diffuse-porous wood, where xylem is disseminated throughout the trunk, show a less well developed ^{129}I signal, probably due to the transport of iodine ions across annual rings. Iodine concentrations in the tree rings range from 0.04 to 2 mg/kg, $^{129}\text{I}/^{127}\text{I}$ ratios from 6×10^{-10} to 3.8×10^{-6} . Tree bark and the outermost rings show significantly higher ^{129}I concentrations than the wood of the trunk. The $^{129}\text{I}/^{127}\text{I}$ ratios for bark are very similar to values obtained for surface soil and water at the two localities, while inner rings have ratios similar to those in deeper layers of the soil, reflecting different pathways for ^{129}I uptake and the differences in ambient ^{129}I levels between the atmosphere and deep soil. Although ring porous trees preserve the depositional pattern of nuclear releases, rings older than or close to the onset of the nuclear age have $^{129}\text{I}/^{127}\text{I}$ ratios significantly above the preanthropogenic level, suggesting that even in these trees some redistribution of ^{129}I occurs throughout the trunk. Our results indicate that growth rings from ring-

porous wood are useful in time-series analyses of regional ^{129}I deposition, yielding reliable information on relative changes in ^{129}I concentrations but requiring caution in the reconstruction of absolute ambient concentrations during any given time.

1. Introduction

An important consequence of the nuclear age is the presence of anthropogenic radioisotopes in the environment. Initially, this release was limited to the atmospheric testing of nuclear devices carried out mainly between 1945 and 1963, but the development of the nuclear power industry has added another potential source of anthropogenic isotopes to the environment. The addition of these radioisotopes to the environment has caused concern because of potential health risks to humans but has also provided opportunities for new tracer studies in fields such as hydrology, oceanography, and atmospheric studies. In all of these considerations, it is essential to understand the history of release and transport mechanisms and pathways for these radioisotopes in the environment. The goal of the current study was to investigate to what extent the ^{129}I signal in trees can be used to document the deposition of radioisotopes into a local environment. The study is based on the determination of ^{129}I concentrations in several sets of tree rings, collected in the vicinity of West Valley in western New York State, the site of former nuclear fuel reprocessing activities.

Iodine has one stable isotope, ^{127}I , and one long-lived radioisotope, ^{129}I (half-life 15.7 Ma (1)), the focus of this study. All other isotopes, such as ^{125}I and ^{131}I , are short-lived with half-lives up to 60 days. Natural ^{129}I is produced by the interaction of cosmic rays with Xe atoms in the atmosphere (2) and by the spontaneous fission of ^{238}U in the earth's crust (3). Both of these sources contribute at similar rates to the ^{129}I budget in surface reservoirs (4), with minor additions coming from neutron-induced fission of crustal ^{235}U . As an element that is very active biologically, iodine exchanges rapidly between all surface reservoirs such as the oceans, biosphere and atmosphere, which are assumed to have had identical preanthropogenic $^{129}\text{I}/^{127}\text{I}$ ratios. The preanthropogenic $^{129}\text{I}/^{127}\text{I}$ ratio in surface reservoirs has been determined to be 1.5×10^{-12} (5). With this ratio, the concentration of preanthropogenic ^{129}I in wood can be estimated. For iodine concentrations between 0.2 and 2 mg/kg (as reported, for example, for a black locust tree near Karlsruhe, Germany by Hauschild and Aumann (6)), ^{129}I concentrations in wood were between 1 and 10 atoms/mg before the onset of the nuclear age.

The addition of anthropogenic ^{129}I has raised the ^{129}I concentration in all surface reservoirs by several orders of magnitude. Anthropogenic ^{129}I is the direct product of neutron induced fission, either of ^{235}U , the main reaction in conventional fission reactors, or of ^{239}Pu in nuclear devices. Releases during approximately 500 atmospheric nuclear weapons tests added substantial amounts of ^{129}I in the late 1950s and early 1960s, but nuclear fuel reprocessing at facilities (such as Sellafield in England, Cap La Hague in France, and Hanford, WA) has been the predominant source of anthropogenic ^{129}I in the recent past (7–12). Although operations at nuclear power plants do not routinely release ^{129}I (10), accidental releases occur with the Chernobyl accident being the most widely reported case. This accident resulted in a small but discernible addition of ^{129}I to the global

* Corresponding author phone: (610)660-1786; fax: (610)660-1780; e-mail: urao@sju.edu.

^{||} Saint Joseph's University.

[†] University of Rochester.

[‡] National Institute of Radiological Sciences.

[§] PRIME Lab, Purdue University.

[¶] Current address: Mercer County Soil District, 508 Hughes Drive, Hamilton Square, NJ 08690.

environment (13). As a result of these ^{129}I additions, the natural budget for this isotope has been overwhelmed by anthropogenic ^{129}I , and current $^{129}\text{I}/^{127}\text{I}$ ratios in all global surface reservoirs are several orders of magnitude higher than the natural ratio (10, 14–16), providing a very strong signal in tracer studies. This pronounced signal, combined with the long half-life of ^{129}I and the close affinity of iodine to organic matter, makes the ^{129}I - ^{127}I isotopic system well suited for a large range of potential applications in a number of fields. The development of accelerator mass spectrometry (AMS), the preferred detection method for this system, has made a highly sensitive method available for the detection of ^{129}I (17–19).

The biogeochemical cycling of iodine isotopes is as yet poorly understood. The $^{129}\text{I}/^{127}\text{I}$ ratio in the atmosphere is controlled by many factors. The oceans are the main reservoir for natural iodine, with distance from the oceans controlling the flux of iodine to the continents. Moran et al. (11) have reported that samples of rainwater and epiphytes collected in continental interiors show higher $^{129}\text{I}/^{127}\text{I}$ ratios than those from coastal regions. Although many iodine compounds are volatile, the short residence time of iodine in the atmosphere does not allow for uniform mixing. In addition, iodine isotopes exist in multiple chemical forms depending on production sources and atmospheric interactions. Wet and dry deposition transfer iodine to the soil, where penetration depths appear to be shallow. For example, Rao and Fehn (10) found 85–90% of ^{129}I within a soil core to be present in the top 15 cm of soil, probably related to chemical and biological interactions with the organic-rich soil litter layer, and to high root density in the top few centimeters of surface soil. Natural ^{127}I and anthropogenic ^{129}I may be cycled through the soil-vegetation system at different rates, related to differences in chemical lability (10).

The purpose of the present study was to determine the degree of association of the tree-ring record of ^{129}I with the known nuclear history of a region and to potentially to identify suitable tree species for future studies of ^{129}I in tree rings. This study is a subset of a larger investigation carried out on the sources, distribution, and reservoirs of anthropogenic ^{129}I in North America, with an emphasis on western and central New York State (10, 20, 21). The only previous study of ^{129}I in tree-rings is that of a black locust tree at the small nuclear fuel reprocessing facility in Karlsruhe, Germany (6). The WAK reprocessing plant at Karlsruhe was operated during the years 1971–1990, processing approximately 200 tons of nuclear fuel during that time, although it is a much smaller plant than the West Valley facility.

2. Study Area and Sample Collection

The main study area was outside the West Valley nuclear fuel reprocessing facility in western New York State where 630 tons of spent nuclear fuel were reprocessed during the years 1966–1972. Releases of ^{129}I and other radioisotopes from the West Valley site have been previously documented (20, 22–26). Selected tree rings from the years 1950–1995 and tree bark were collected outside the site from Black Locust (*Robinia pseudoacacia*), Northern Red Oak (*Quercus rubra*), Black Cherry (*Prunus serotina*), and Sugar Maple (*Acer saccharum*) trees and from an American Elm tree (*Ulmus americana*) located in Rochester, NY, approximately 115 km to the northeast of West Valley.

The study area was selected because it provided not only the input from atmospheric fallout from weapons testing and recent reprocessing observed on a global basis but also the specific signal associated with the short period of reprocessing activity at the West Valley facility (1966–1972). During reprocessing activities in the region, $^{129}\text{I}/^{127}\text{I}$ ratios increased by 9 orders of magnitude, averaging approximately 10^{-3} in surface soils around West Valley (23). Since the facility

stopped reprocessing activities in 1972, $^{129}\text{I}/^{127}\text{I}$ ratios have decreased substantially in the region due to the dispersal of ^{129}I away from the site (10). However, additional small releases from the remaining nuclear material and the remobilization of already deposited ^{129}I have resulted in levels there that are still elevated ($^{129}\text{I}/^{127}\text{I} = 1.5 \times 10^{-5}$ in top soil as reported by Rao and Fehn (10)) by more than 3 orders of magnitude above that observed for nonpoint source areas in North America. The signal from West Valley can be traced to about 200 km east from the site, following the predominant wind direction in this region, and is thus diminished but still detectable at Rochester (10).

The trees sampled in this study can be broadly divided into two groups on the basis of the mode of water transport in the tree. Black Locust, Northern Red Oak, and American Elm are ring-porous woods, in which xylem has become specialized so that most of the water is transported via the outermost growth layer. In the more common diffuse porous (Sugar Maple) and semidiffuse porous woods (Black Cherry), xylem is disseminated throughout the sapwood, thus providing a greater potential for the transport of ^{129}I (and ^{127}I) across rings of different years. For all the trees, rings were selected to reflect the height of the atmospheric bomb tests (1960), the activity at West Valley (1970/1971), and the period after reprocessing ended at West Valley (1986). In addition, bark or the outermost rings reflecting the collection age (1995) were sampled and, where available, rings corresponding to the prenuclear age or the onset of the nuclear age were also collected.

3. Methods

For the study, wood was drilled from individual tree rings, oven dried, and finely crushed to obtain up to 12 g of wood per sample. Extraction of iodine for the determination of ^{129}I concentrations was carried out using an alkali leach and fusion method originally developed for meteorites (27) and adapted for soil, vegetation, and wood (21). Between 1.5 and 4.2 mg of carrier iodine with a known, low $^{129}\text{I}/^{127}\text{I}$ ratio was added to the samples prior to alkali leach and fusion. Samples were placed in disposable nickel crucibles and were treated with saturated NaOH solution to reduce the temperature of fusion and with Na_2O_2 to oxidize the samples. Dried samples were heated in a high-temperature muffle furnace at 300 °C for 30 min and at 600 °C for 2 h. A solution of H_2SO_4 , NaHSO_3 , and deionized water was used to leach iodine from the resulting fusion cake into an aqueous phase, followed by extraction into carbon tetrachloride and precipitation as silver iodide (18, 21). Extraction efficiency during alkali leach and fusion was between 70 and 85%, monitored using organic material standards obtained from NIST. These relatively large losses during sample preparation can be attributed to the difficulties associated with the separation of iodine from an organic matrix. $^{129}\text{I}/^{127}\text{I}$ ratios in the silver iodide precipitate were measured using the AMS system of PrimeLab, Purdue University (28). ^{129}I concentrations in the samples were calculated by correcting the $^{129}\text{I}/^{127}\text{I}$ ratios measured by AMS for the presence of carrier iodine. Samples of leaves and lichen growing on the bark of the black locust were rinsed thoroughly with deionized water and treated in the same manner as the wood.

Iodine concentrations in three of the trees were determined by ICP-MS after combustion in a tube furnace following established methods (29, 30). For these samples, $^{129}\text{I}/^{127}\text{I}$ ratios were calculated from iodine and ^{129}I concentrations. Errors (all 1σ) were in general below 10% for the iodine concentrations and for the AMS measurements. The combined errors from sample preparation and measurement were estimated to be between 25 and 50% for the $^{129}\text{I}/^{127}\text{I}$ ratios.

TABLE 1: ^{129}I , Iodine Concentrations, and $^{129}\text{I}/^{127}\text{I}$ Ratios in Tree Rings and Associated Samples

tree (porousness)	location	year	^{129}I (atoms/mg)	iodine (mg/kg)	$^{129}\text{I}/^{127}\text{I}$
Black Locust (ring porous)	West Valley	1952	3782.0	0.21	3.8E-6
		1961	332.0	0.57	1.2E-7
		1971	1447.0	0.40	7.7E-7
		1987	300.0	0.25	2.5E-7
		1994	300.0	0.09	7.0E-7
		Bark (1995)	27810.0	0.35	1.7E-5
		Lichen (1995)	3580.0	NA	NA
Red Oak (ring porous)	West Valley	1961	50.0	0.04	3.0E-7
		1971	285.0	0.07	8.0E-7
		1987	41.0	0.09	9.0E-8
		1994	3165.0	0.17	3.9E-6
		Bark (1995)	72.0	NA	NA
Black Cherry (semidiffuse porous)	West Valley	1950-1955	1.0	NA	NA
		1970	6.0	NA	NA
		1987-1995	16.0	NA	NA
Sugar Maple (diffuse porous)	West Valley	1946	39.0	NA	NA
		1959	16.0	NA	NA
		1971	28.0	NA	NA
		1981	4.0	NA	NA
		Bark (1995)	72.0	NA	NA
Elm (ring porous)	Rochester	1935	6.0	0.35	3.6E-9
		1950	4.0	1.21	6.08E-10
		1959	58.0	0.90	1.4E-8
		1970	130.0	1.10	2.49E-8
		1980	28.0	1.25	4.77E-9
		1986	10.0	0.72	2.9E-9
		Bark (1995)	2178.0	2.19	2.10E-7

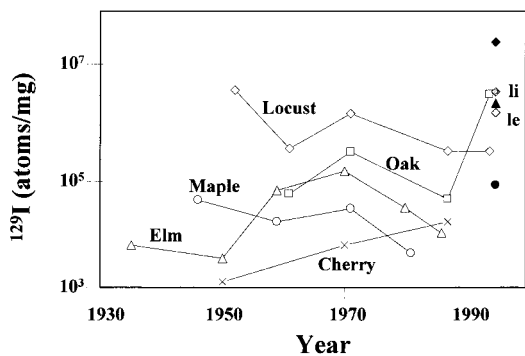


FIGURE 1. ^{129}I concentrations in tree rings, as a function of tree ring age, and associated samples. Results for tree rings are shown as open symbols, bark as solid symbols, leaves (le) and lichen (li) as gray symbols, with the symbols matching the associated tree. ^{129}I concentrations are shown on a logarithmic scale.

4. Results and Observations

Iodine and ^{129}I concentrations as well as $^{129}\text{I}/^{127}\text{I}$ ratios are listed in Table 1. ^{129}I concentrations varied over more than 4 orders of magnitude, from 1000 to 28×10^6 atoms/mg. Even the lowest concentration is substantially above pre-anthropogenic levels in wood (<10 atoms/mg). If ^{129}I concentrations are plotted as a function of tree ring age (Figure 1), the trees (except for the cherry) show a similar general pattern: an increase from 1960 to 1970 and a subsequent decrease to 1986. The highest values are found in the bark or in the outermost ring (oak), following the ranking of concentrations between the trees. In the one case where we also sampled leaves and lichen, the concentrations there fall between those of outermost ring and bark. In elm, maple, and locust, we sampled the center of the tree and found a distinct increase in ^{129}I concentrations, as compared to the next oldest sample. This overall pattern is less well preserved in the maple tree and is absent in the cherry. Material had been combined over several rings in two of the cherry samples to obtain enough sample material for iodine extraction. This might explain the absence of a more

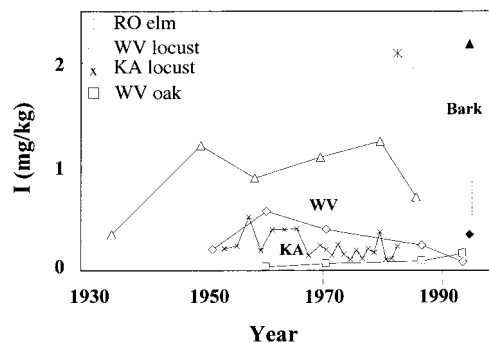


FIGURE 2. Iodine concentrations in elm (triangles), locust (diamonds, WV) and oak (squares). Results for bark samples are shown as matching solid symbols. Results for the locust from West Valley (WV) compare well with those from the locust in Karlsruhe, Germany, (KA), tree rings and bark represented by x and asterisk symbols, respectively; data from ref 6).

distinct pattern in this tree. Since the general level of ^{129}I concentrations in maple and cherry is considerably lower than in oak, locust, and elm, and the pattern is most clearly visible in the latter three trees, we focused our further efforts there.

Iodine concentrations are shown in Figure 2 for oak, locust, and elm, including the bark samples from the latter two trees. Also included are the data from the locust tree near Karlsruhe, Germany (6). There are systematic differences in the concentrations between the three trees, with the highest values in the elm, followed by the locust and the oak. Fluctuations in the concentrations are also visible within the trees, especially in the elm tree. The two bark samples have considerably higher concentrations than the other samples from elm and locust, respectively. Concentrations for the black locust from West Valley are in excellent agreement both in values and range with those found for the black locust in Germany, except for the bark value, which is substantially lower in our study.

Combining ^{129}I and iodine concentrations allows the calculation of $^{129}\text{I}/^{127}\text{I}$ ratios in these samples (Figure 3). Ratios

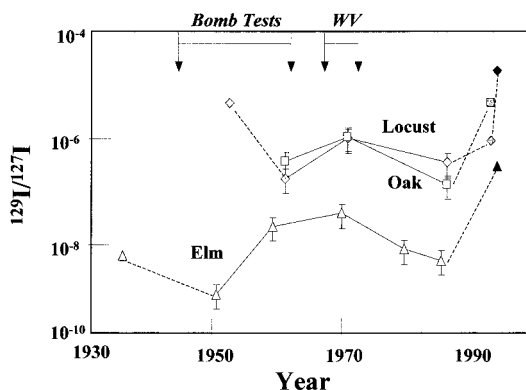


FIGURE 3. $^{129}\text{I}/^{127}\text{I}$ ratios in tree rings and bark from oak and locust, West Valley, and elm, Rochester. The results from the inner parts of the trees are connected by solid lines, bark (solid symbols) and outermost rings (dark gray symbols) as well as innermost rings (light gray symbols) are connected by dotted lines. The periods of atmospheric testing and reprocessing activity at West Valley are indicated at the top.

are essentially identical during the period between 1960 and 1986 for locust and oak, the two trees from West Valley. A very similar pattern is visible in the elm tree from Rochester but shifted to considerably lower levels. The two bark samples and the outermost rings again show substantially higher values as do the two innermost samples, but they still follow the established ranking between the trees. The inclusion of error margins shows that the overall pattern is well preserved despite the rather large errors (up to 50%) of these ratios.

5. Discussion

The initial comparison of ^{129}I concentrations strongly suggests that the depositional pattern is better preserved in ring porous trees (oak, locust, and elm) than in diffuse (maple) or semidiffuse porous (cherry) trees. This finding is not surprising, given the different modes of fluid transport prevalent in the different trees (31). The ring-porous trees also had considerably higher ^{129}I concentrations than the other two types of trees. Because neither center nor bark in the maple and cherry trees had excessively high ^{129}I concentrations, the lower levels probably reflect lower uptake of iodine in these trees rather than a strong redistribution toward the bark or the center. The comparison of the distribution of ^{129}I in different tree types suggests that ring-porous trees maintain the depositional pattern considerably better than other types of trees.

Although the overall range in iodine concentrations is rather small, they apparently are distinctly different for each tree species. The excellent agreement between the two locust trees, both in range and variability, supports this observation and suggests that iodine levels in trees are more strongly influenced by the type of tree than by the availability of iodine in the soil. A substantial variability of iodine concentrations was observed in all trees. The specific role of iodine in wood is not known, but it is assumed that trees derive their iodine from several sources: soil, deposition to leaves from rain or dryfall, and recycling of leaf litter (32, 33). This variability could reflect changes in the availability of soil or atmospheric iodine or, perhaps more likely, changes in the uptake of iodine dependent on the growth pattern in a specific year. Because differences in isotopic composition are much greater than possible from isotope fractionation, the variation in iodine concentrations does not influence the interpretation of the isotopic record in the trees but demonstrates the importance of using $^{129}\text{I}/^{127}\text{I}$ isotope ratios for the final interpretation of the depositional record.

The isotopic patterns in the intermediate portions of the three trees are well correlated with the depositional history

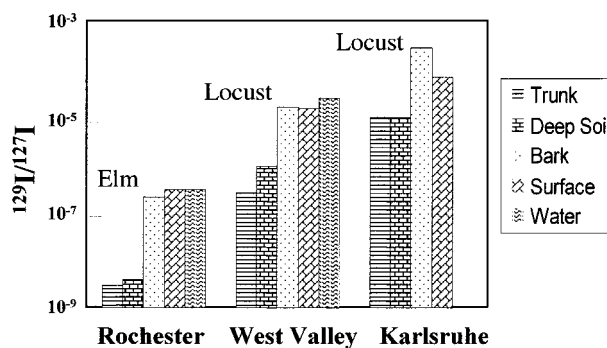


FIGURE 4. Comparison of $^{129}\text{I}/^{127}\text{I}$ ratios in the elm at Rochester, the locust at West Valley and the locust at Karlsruhe with environmental samples. In each case, ratios in an inner ring are compared to ratios in deep soil, and the ratios in bark to top soil and surface river water (not available for Karlsruhe). Data from refs 6, 10, and 34.

in this region: initial increase in $^{129}\text{I}/^{127}\text{I}$ isotope ratios due to the atmospheric bomb tests from the late 1940s until 1963 (with a possible addition from reprocessing releases during the late 1950s and early 1960s from Hanford, WA), a further increase due to the reprocessing activity at West Valley from 1966 to 1972, followed by a gradual decrease after the closing of this facility. The high levels in bark and outermost ring probably reflect two depositional paths: uptake of iodine through the root system and direct atmospheric deposition. The increase toward the center, observed in both trees where the center was analyzed, is not easily explained. Although the increase in the elm from 1950 to 1935 in itself is not very pronounced, the 1935 value is higher by a factor of 2000 than the value of 1.5×10^{-12} (5) expected for this preanthropogenic period. Also contrary to expectations is the difference in 1960 ratios between Rochester and West Valley, i.e., before the start of reprocessing activities at West Valley. Both of these observations suggest that some degree of reequilibration of iodine occurs throughout the trunk even in ring-porous trees. A similar observation can be made for the locust tree from Karlsruhe, Germany, where the innermost ring also shows values considerably higher than those measured in the environment at that time (6). Although the overall depositional history is well preserved in ring porous trees, some transport of iodine occurs even in these types of trees. The transport could take place either across tree rings or, perhaps more likely, through uptake of fluids from the root system into the inner part of the trunk. Although the inner rings do not show a corresponding increase in iodine concentrations along with ^{129}I concentrations, this may result from preferential uptake of recently released (potentially more biolabile) ^{129}I over stable iodine from an older geochemical pool. The presence of anthropogenic ^{129}I in older heartwood warrants further study.

The different pathways of ^{129}I in trees are also visible if trees from the two locations in western New York are compared to the tree in Germany (Figure 4; data compiled from refs 6, 10, 34). For the three locations (using data from the locust for West Valley), the following five ratios are compared: main trunk (i.e., intermediate rings, 1987 for West Valley, 1986 for Rochester, 1980 for Karlsruhe); deep soil (30 cm for Rochester and West Valley; 20–30 cm for Karlsruhe); bark; top soil (0–2 cm); surface water (not available for Karlsruhe). Although the general levels of the ratios strongly reflect the specific location, a striking similarity exists in all three cases between values of the main portion of the trunk and deep soil, on one hand, and between bark values and those in surface soil and water, on the other hand. These comparisons indicate that the sapwood receives iodine from the soil but that the values in the bark reflect uptake mainly

from the atmosphere. The ratio found for the locust at Karlsruhe in 1960 is 5×10^{-7} , which is considerably higher than those found in West Valley (1.2×10^{-7}) or Rochester (1.4×10^{-8}) for the same time. Since at 1960, reprocessing activity had not yet started at either Karlsruhe or West Valley, these differences are again an indication that some degree of reequilibration occurs even in ring-porous trees.

We conclude that ring-porous trees such as oak, locust, and elm appear to be well suited for an investigation of the local depositional history of ^{129}I , while transport across the tree rings in diffuse porous or semidiffuse porous trees leads to a much less well preserved signal. The records in ring-porous trees clearly show the history of ^{129}I releases in western New York, in particular the global increase associated with atmospheric bomb tests and the local increase due to the reprocessing activity at West Valley. Although the overall nuclear history is well preserved, there is also clear evidence for reequilibration between rings, even in ring-porous trees. The results of the present study of trees at West Valley and Rochester are in excellent agreement with those from an investigation carried out near a reprocessing plant in Karlsruhe, Germany (6). $^{129}\text{I}/^{127}\text{I}$ records in ring-porous trees give a good indication of the depositional pattern in a specific location, although some level of transport across tree rings causes a degree of equilibration. This transport would have to be taken into account if a quantitative assessment of the depositional history in a given location is desired.

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