

Akira Kagawa · Steven W. Leavitt

Stable carbon isotopes of tree rings as a tool to pinpoint the geographic origin of timber

Received: May 28, 2009 / Accepted: September 14, 2009 / Published online: January 2, 2010

Abstract Illegal logging is a major cause of worldwide deforestation, and demands for scientific methods to identify the geographic origin of timber are increasing. “Dendroprovenancing” is one such method, in which the origin of unknown wood is estimated by calculating correlations of the ring-width series of the unknown wood with reference trees of known geographic origins. We applied the dendroprovenancing method to carbon isotope network data of pinyon pines (*Pinus edulis* and *Pinus monophylla*) from the southwestern United States to test the efficacy of using a carbon isotope time series for provenancing wood. First, we calculated correlations (t values) between test trees temporarily assumed to be of unknown origin and reference trees from 13 surrounding sites. Then, we plotted the t values on a map. When provenancing was successful, the tested trees showed the strongest correlation with reference trees from sites close to the actual origins of the test trees, and the correlations decreased with the distance between the original sites of test and reference trees. This conical distribution of t values enabled provenancing of wood with precision of 114–304 km. Although isotope measurement is more expensive and laborious than ring-width measurement, our tests of provenancing pinyon pines in the southwestern United States showed a higher success rate with carbon isotopes.

Key words Illegal logging · Isotope fingerprint · Traceability · $\delta^{13}\text{C}$ · Forest law enforcement governance and trade (FLEGT)

Introduction

Deforestation accounts for up to one-third of total anthropogenic carbon dioxide emissions, and illegal logging is one of the major causes of worldwide deforestation.¹ However, the lack of scientific methods to estimate the geographic origin of timber has made the identification of illegally logged or traded timber technically difficult. Economical damages through illegal timber trade in the world are currently assessed at 10 billion USD per year.² Illegal logging is severe in nontropical regions such as the Russian Far East and the Baltic States,^{3,4} as well as in tropical regions including Indonesia. For example, 40% of all wood products imported by China are supplied by countries with a high incidence of illegal logging, such as Russia and Indonesia.⁴ To prevent illegal logging, consumer countries need to ensure traceability of the wood supply from the source to the market, thereby preventing “laundering” of illegally logged timber before it enters consumer markets. Geographic origin of wood is the most important aspect of traceability.

Genetic, geochemical, and climatic information preserved in wood all have the potential to serve as tools for provenancing. In general, identification of tree species at the genus level is possible by observing the anatomical structure under a microscope. DNA analysis⁵ allows identification of wood at the species level. The origin of timber can then be constrained to the distribution of the genus or species. Because DNA has natural geographic variation, one can also estimate the geographic origin of unknown wood by comparing the chloroplast DNA with chloroplast DNA from reference trees from different regions.⁶ However, DNA fingerprinting cannot be applied to plantation trees because these DNA fingerprints do not match the natural

A. Kagawa (✉)
Wood Anatomy and Quality Laboratory, Forestry and Forest Products Research Institute, 1 Matsunosato, Tsukuba, Ibaraki 305-8687, Japan
Tel. +81-29-829-8301; Fax +81-29-874-3720
e-mail: akagawa@ffpri.affrc.go.jp

S.W. Leavitt
Laboratory of Tree-Ring Research, University of Arizona, Tucson, AZ 85721, USA

Part of this report was presented at the Fall Meeting of the American Geophysical Union, San Francisco, California, December 2008, and the 59th Annual Meeting of the Japan Wood Research Society, Matsumoto, Japan, April 2009

distribution of DNA. An alternative method to identify the geographic origin of wood is thus required.

Geochemical factors, such as isotope ratios and concentrations of inorganic elements, have also been used to provenance agricultural products.⁷ Isotope ratios of strontium in wood^{8,9} are known to change geographically. However, methods based on stable isotope ratios of carbon, nitrogen, and oxygen have proved more effective than using inorganic elements for provenancing tropical woods.¹⁰

The ring widths of wood reflect the climatic conditions in which the source tree grew, and this knowledge has been used for paleoclimate reconstruction.^{11,12} Local climatic fluctuations are reflected in ring-width fluctuations, resulting in region-specific ring-width patterns. Trees growing close to each other at distances less than a few hundred kilometers show higher similarity than the trees growing farther apart.¹³ Ring-width series can thus be used for provenancing wood.^{14,15} One can estimate the likely origin of unknown wood by calculating correlations of the ring-width series of the unknown wood with reference trees of known origins.¹⁶ By plotting the correlation of a series from an unknown site with each reference chronology on a map, the estimated origin can be identified as the location of maximum correlation.¹⁷ This origin identification method, called “dendroprovenancing,” is mainly used for provenancing historical wooden artifacts.

Stable isotope ratios of tree rings have been used for climate reconstruction in a way similar to use of ring-width series.^{18,19} Isotope ratios reflect the climatic conditions when photosynthesis and ring formation took place.^{20,21} Therefore, all timber produced anywhere in the world, including tropical timber,^{22,23} has a unique isotopic fingerprint that reflects the climatic history of its origin.^{18,19} This uniqueness suggests the potential use of stable isotope fingerprints for provenancing wood.

In fact, isotope fingerprinting is already in use for estimating the origins of various plant-based products.^{24–28} Both oxygen and hydrogen isotope ratios of plants reflect source water and leaf transpiration.²⁹ Isotope ratios of source water and leaf transpiration reflect temperature and vapor pressure deficit signals, respectively. Furthermore, carbon isotope ratios in C₃ plants reflect the balance between stomatal conductance and photosynthetic rate,³⁰ which is dominated by factors such as relative humidity, soil water status, summer irradiance, and temperature. In general, using oxygen and hydrogen isotopes to provenance agricultural products is more successful than using carbon isotopes. Despite recent developments in food and forensic fields, to date very few studies have so far applied these fingerprinting methods to wood provenancing. If the timber comes from nontropical area and has distinct tree rings, then the isotopic fingerprint can be easily expressed as a time series of yearly stable isotope ratios.

Application of the “dendroprovenancing” method to tree-ring isotope series may further improve the accuracy of wood provenancing, because stable isotope ratios of tree rings are generally more coherent (i.e., show less within-site variation among the individuals) than ring widths.^{31–33} High-

frequency fluctuations can be coherent even among sites hundreds of kilometers apart.^{34–36}

The objective of this study was to evaluate the possibility of “isotope dendroprovenancing,” i.e., using stable isotope series of tree rings to identify the geographic origin of wood. As large-scale network data for tree-ring oxygen and hydrogen isotopes were not available, we used carbon isotope network data from the southwestern United States.³⁷

Materials and methods

Carbon isotope network data

We used tree-ring carbon isotope network data both previously published^{35,37–39} and currently available to the public online (<http://www.ncdc.noaa.gov/paleo/treering/isotope/>). The network consists of tree-ring samples from a set of 14 pinyon pine (*Pinus edulis* and *Pinus monophylla*) sites [Kane Spring, Alton, Dry Canyon, Lower Colonias, Aztec, Cerro Colorado, Ozena, Hawthorne, Mimbres, Owl Canyon, north-central Arizona (NC AZ), northeastern Arizona (NE AZ), Gate Canyon, and Lamoille] collected across six states in the American Southwest (Fig. 1). Site elevations ranged from about 1400 m, but the sites were typically in mountainous terrain with intervening topography exceeding 2500 m. In general, the rings of four cores from four of the trees were used in the isotopic analysis. After ring-width measurement for cross-dating, rings were separated into pentads (e.g., rings formed in 1900–1904, 1905–1909, etc.). Pentads from the four cores from

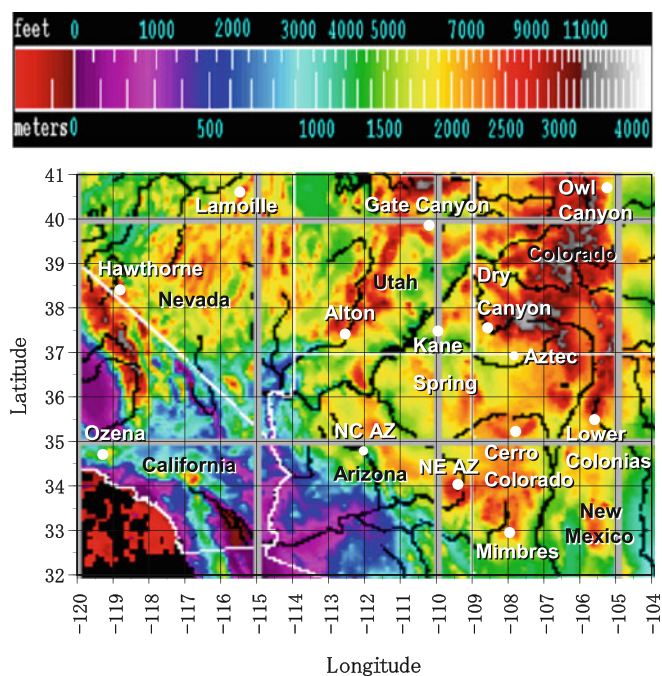


Fig. 1. Locations of the 14 sites and topography in the southwestern United States

four trees were pooled into composite samples for each site except for NC AZ and NE AZ. The pentads of individual trees from NC AZ and NE AZ were analyzed without pooling.³⁸ Stable carbon isotope composition ($\delta^{13}\text{C}$) was expressed as follows:

$$\delta^{13}\text{C} = \left(\frac{^{13}\text{C}/^{12}\text{C}_{\text{sample}}}{^{13}\text{C}/^{12}\text{C}_{\text{standard}}} - 1 \right) \times 1000 \quad (1)$$

Dendroprovenancing

We desired to assess the efficacy of isotope dendroprovenancing, and one necessary condition for successful dendroprovenancing is that the actual origin of the unknown tree must be surrounded by sites from which reference trees were collected. We therefore chose the 6 pooled $\delta^{13}\text{C}$ chronologies in the interior of the 14-site network – 1 pooled chronology each from Kane Spring, Alton, Dry Canyon, Aztec, Cerro Colorado, and Gate Canyon (Fig. 1) to test as though they were from unknown sites (we will hereafter refer to these as test chronologies). For the reference network, we used the remaining 6 marginal chronologies from Lower Colonias, Ozena, Hawthorne, Mimbres, Owl Canyon, and Lamoille as well as the other 7 chronologies from interior sites for a total of 13 chronologies. (We will refer to these 13 chronologies as reference chronologies. Note that there are a total of 14 chronologies. For each test chronology, the remaining 13 chronologies compose a specific reference network.)

By calculating correlations of a test chronology with each of the other 13 reference chronologies, the geographic origin of the test chronology can be estimated by the dendroprovenancing method.^{13,14,16,17} Both ring widths and $\delta^{13}\text{C}$ capture high-frequency (less than 10 years) common forcing (macroclimate) better than low-frequency (on the order of 25 years or more) forcing.^{31,36,39} Therefore, conventional dendroprovenancing techniques preferentially utilize high-frequency variations, and we reason that this should apply to $\delta^{13}\text{C}$ dendroprovenancing as well.

As a first step in isotope dendroprovenancing, we removed the influence of the Suess effect [effects of changing $\delta^{13}\text{C}$ of atmospheric CO_2 ($\delta^{13}\text{C}_a$)] from tree-ring $\delta^{13}\text{C}$ ($\delta^{13}\text{C}_{\text{TR}}$). For this, we used $\delta^{13}\text{C}_a$ data to calculate carbon isotope discrimination (Δ).^{19,40}

$$\Delta = \frac{\delta^{13}\text{C}_a - \delta^{13}\text{C}_{\text{TR}}}{1 + \delta^{13}\text{C}_{\text{TR}}/1000} \quad (2)$$

Then, we extracted high-frequency variations from the carbon isotope discrimination. We investigated several methods to accomplish this. In the first method, the logarithm of the ratio of ring parameter of a given year to the following year (Eq. 3),⁴¹ is used for extracting high-frequency variation as follows:

$$\Delta_i(\text{Holl}) = \log\left(\frac{\Delta_i}{\Delta_{i+1}}\right) \quad (3)$$

where Δ_i is the carbon isotope discrimination of the i th pentad, and $\Delta_i(\text{Holl})$ is the standardized carbon isotope discrimination, after Hollstein's transformation.

In the second method, high-frequency variation is extracted by calculating first differences^{31,42,43} as follows:

$$\Delta_i(\text{FD}) = \Delta_i - \Delta_{i-1} \quad (4)$$

Finally, in the third method, the difference between the i th pentad and the average of the previous and the following pentad is calculated as

$$\Delta_i(\text{AV}) = \Delta_i - \frac{(\Delta_{i-1} + \Delta_{i+1})}{2} \quad (5)$$

We then calculated the correlation coefficient (r) of high-frequency variations between the test chronology and each reference chronology. We calculated t values for each reference chronology as follows:

$$t = |r| \sqrt{\frac{n-2}{1-r^2}} \quad (6)$$

where n is the number of pentads of the test tree that are common with pentads of the reference trees.

To utilize high-frequency variations in ring-width data [$\text{RW}_i(\text{AV})$], we processed ring-width data in exactly the same way as described above. For both these methods, we used data from the corresponding tree rings formed during 1790–1984 ($n = 37$) for calculating t values of site chronologies from the six interior sites. This step enabled the direct comparison of the dendroprovenancing results obtained from these two different parameters. We estimated the origin of the test chronology to be the location of the reference chronology with which the test chronology showed the highest t value (as in Figs. 3–5 in Wazny¹⁷).

To compare the signal-to-noise ratios (SNR) of tree-ring $\delta^{13}\text{C}$ and width series,⁴⁴ we used a two-way analysis of variance (ANOVA) applied to the high-frequency variations of the five site chronologies [$\Delta_i(\text{AV})$, $\text{RW}_i(\text{AV})$] within 300 km of Kane Spring (Kane Spring, Alton, Dry Canyon, Aztec, and Gate Canyon). Again, we used data from the overlapping tree rings formed during 1790–1985 ($n = 37$). We expected correlations (t values) between two site chronologies to decrease with the distance between the two sites in a similar way as Noda¹³ observed with ring width. To verify this, we calculated and plotted the t values of all possible combinations of two chronologies from the 14 sites (${}_{14}C_2 = 91$) against the distance.

Twelve of the site chronologies were constructed from four pooled trees each, as already described. However, only one tree is expected to be available in most wood provenancing cases. To examine whether using one tree instead of a group of four trees as the test tree can give successful provenancing results, we also used the carbon isotope time series from individual trees to estimate the origin of each tree from NC AZ and NE AZ in the same manner as we provenanced the site chronologies. For calculation of this set of t values, we used data from the tree ring pentads formed during 1845–1984 ($n = 26$).

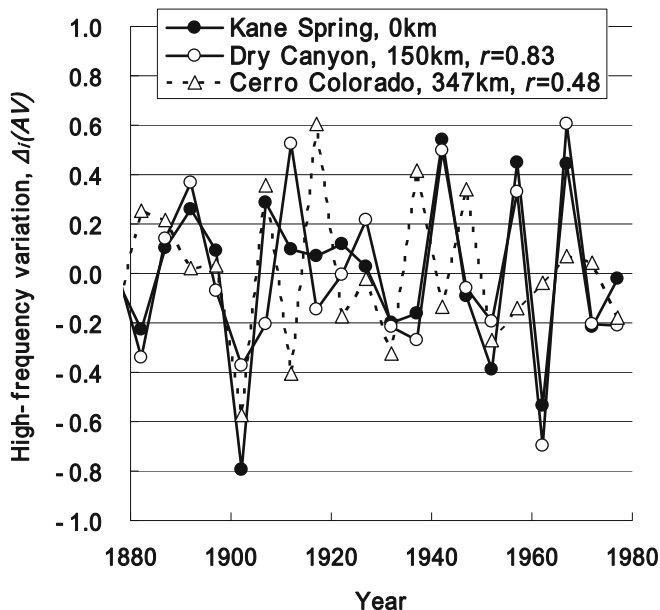


Fig. 2. Change of high-frequency isotope variation with distance. Kane Spring (solid circles) chronology showed more similarity with the chronology from nearby Dry Canyon (open circles) than with the chronology from distant Cerro Colorado (triangles)

Results

Dendroprovenancing of carbon isotope and ring-width site chronologies

Provenancing methods based on Eqs. 3–5 all gave similar results. However, for both isotope and ring-width series, Eq. 5 gave the highest t values, and we therefore adopted this equation for all results and figures reported in this article unless otherwise specified. Carbon isotope chronologies from nearby regions showed very similar high-frequency variation [$\Delta_1(AV)$]. For example, in Fig. 2 we plotted $\Delta_1(AV)$ from Kane Spring and Dry Canyon, which are located 150 km apart (see Fig. 1). However, the similarity decreased at Cerro Colorado, which is located 347 km away from Kane Spring.

We considered provenancing to be successful when the P value of a correlation maximum was less than 2×10^{-7} . Threshold t values to meet this level of confidence were 6.45 in Figs. 3 and 4 ($n = 37$) and 7.18 in Fig. 5 ($n = 26$). Such high t values (>6.45) were observed within 450 km of the actual origin of a test chronology, and t values decreased with distance from the origin (Fig. 4), resulting in a conical t value distribution (Fig. 3a,c,e). For example, the $\delta^{13}C$ (test) chronology from Kane Spring showed the highest t value with the reference chronology from Dry Canyon, which is 150 km away. The t values decreased gradually on the western side over the Colorado River drainage basin and steeply on the eastern side over the Rocky Mountain range (Fig. 3a). The origins of test chronologies were successfully estimated as the locations of t value maxima (Figs. 3a,c,e, 5). This finding strongly indicates the efficacy of carbon isotope dendroprovenancing. Four of the six pooled $\delta^{13}C$

chronologies tested (from Kane Spring, Alton, Dry Canyon, and Gate Canyon) gave successful provenancing results; and the remaining two chronologies (from Aztec and Cerro Colorado) gave unsuccessful provenancing results; these were “complacent” (low interpentadal variation) chronologies, and they showed low t values of less than 5.2 with all the reference chronologies (Figs. 3g, 4). Among the combinations of two individual carbon isotope chronologies that showed successful provenancing ($t > 6.45$), averaged t values were 7.29, 7.26, and 7.74 for Eqs. 3, 4, and 5, respectively.

When we used ring width instead of $\delta^{13}C$ for dendroprovenancing, the results were less successful. For all test chronologies except for the chronology from Aztec, we failed to observe the conical t value distributions with the maxima greater than 6.45 (Figs. 3b,d,f, 4). For the test chronology from Aztec, the highest t value ($t = 7.2$; Fig. 4) was observed at NC AZ, which is located 522 km away from Aztec (Fig. 3h). There were eight sites closer to Aztec than NC AZ; however, they all showed lower t values than NC AZ. Thus, for our study in the southwestern United States, carbon isotope dendroprovenancing was more successful than ring-width dendroprovenancing.

Decrease of t values with the distance

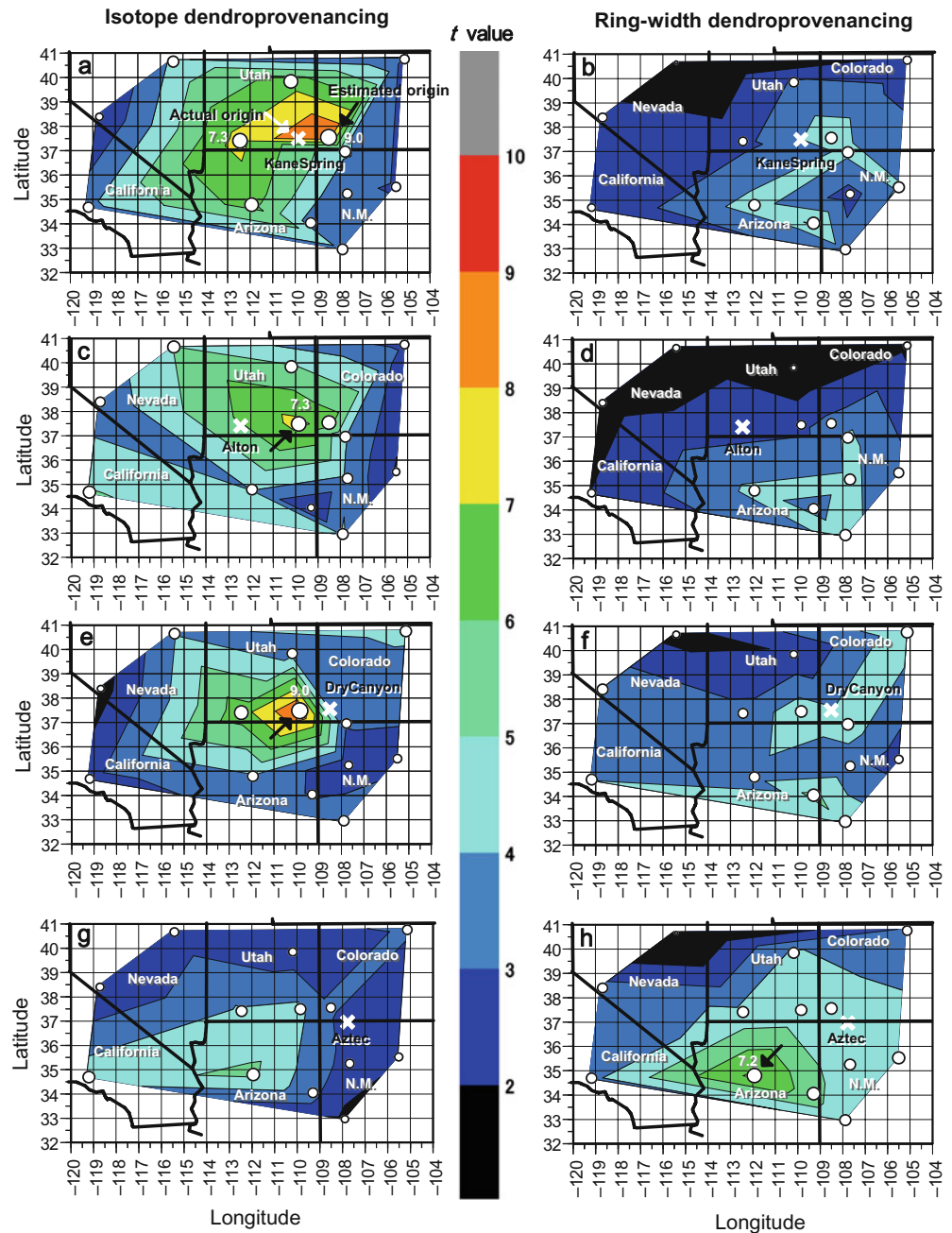
The t values tended to decrease with distance from the origin of the test chronologies (see Fig. 4). This trend was more evident with carbon isotope ratios than with ring widths. We observed high t values exceeding 7 with carbon isotopes at distances of 150–300 km, showing the efficacy of using tree-ring $\delta^{13}C$ for provenancing wood. However, 10 of 14 such combinations of chronologies within 300 km of each other showed t values lower than 5; this tended to happen especially when high mountains lay between the two chronologies. With ring widths, we observed t value maxima exceeding 6.45 at distances 198–522 km away from the actual origin of test chronologies (Fig. 4). In other words, maxima for ring-width chronologies were dispersed over wider areas (~522 km) than maxima for carbon isotope chronologies (~304 km). Widely dispersed t value maxima for ring width are also reported with European oaks,^{16,17} which is problematic because dispersed distribution hinders precise determination of geographic origin.

Signal-to-noise ratios (SNR) of high-frequency variations of carbon isotope discrimination [$\Delta^{13}_i(AV)$] and ring width [$RW_i(AV)$] were 8.1 and 4.6, respectively. The higher SNR of $\Delta^{13}_i(AV)$ that we observed in our study confirms earlier findings that tree-ring $\delta^{13}C$ is more coherent than ring width.^{31–33}

Isotope dendroprovenancing of individual trees

At first, we used 14 pooled + 8 individual = 22 series of $\Delta_1(AV)$ for correlation calculation. Four of eight individual trees from NC AZ and NE AZ showed t value maxima greater than 7.18. However, the other four trees did not show t values greater than 7.18 with any of the reference series. Because high t values exceeding 7.18 ($P < 2 \times 10^{-7}$)

Fig. 3. Results of isotope and ring-width dendroprovenancing of test chronologies from Kane Spring (a, b), Alton (c, d), Dry Canyon (e, f), and Aztec (g, h). The size of the circles of the reference source sites in each panel represents the strength of the relationship (t value) with the site used as the unknown source. The color contour plot in each panel illustrates t value distribution



are required for successful provenancing, the 4 low-sensitivity individual series from NC AZ and NE AZ were excluded from the analysis, leaving 18 reference series (=14 pooled + 4 individual).

As expected, sensitivity of high-frequency variation [$\Delta_i(\text{AV})$] to environmental forcing was different among the eight individuals. Nevertheless, as few as 28 $\delta^{13}\text{C}$ analysis data points per individual tree were sufficient to pinpoint the geographic origin of the four high-sensitivity series to an accuracy of 114–258 km (see Fig. 5). The rate of success for provenancing individual carbon isotope series was 50% (4 of 8) in our case. The rate of success for provenancing individual ring-width series of European oak is reported to be 8.9%.¹⁶

Discussion

Comparison between isotope and ring-width dendroprovenancing

Tree-ring parameters, such as ring widths or stable isotope ratios, reflect common environmental forcing as well as random factors within a site. Environmental forcing and random factors can be defined as signal and noise, respectively, and we believe our observations of higher SNR of $\Delta_i(\text{AV})$ compared to $\text{RW}_i(\text{AV})$ indicate less influence of random factors on carbon isotope discrimination than on ring width.

Fig. 4. Decrease of correlations (t values) with the distance between two chronologies. There are ${}_{14}C_2 = 91$ possible combinations of 2 sites of 14 sites, and all the t values generated are shown. The origins of test trees could be estimated at 150–304 km accuracy (solid-line box) using carbon isotopes and 198–522 km accuracy (dashed-line box) with ring widths

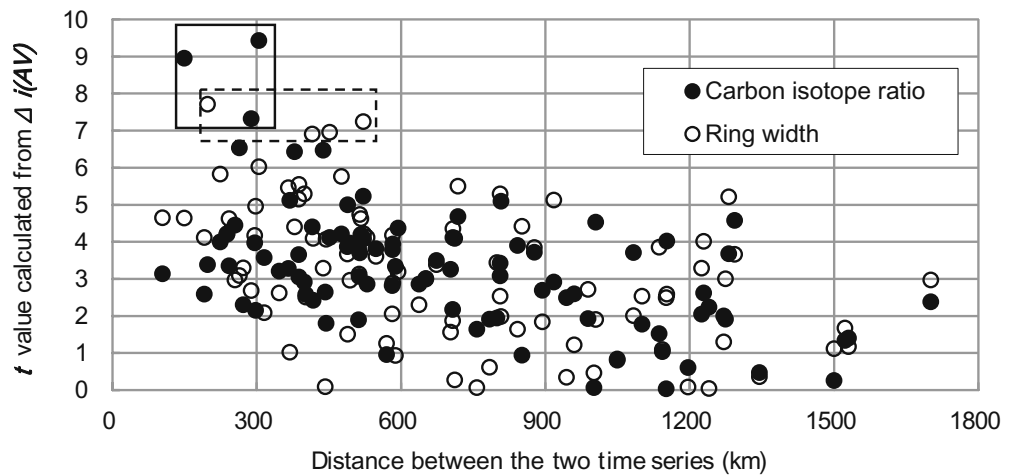
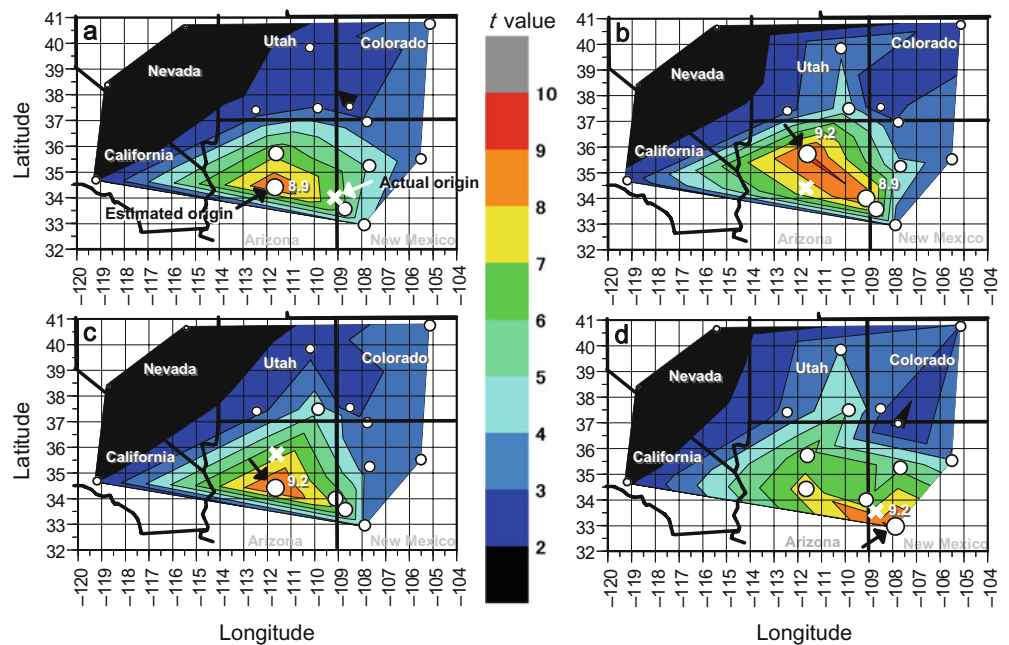


Fig. 5. Results of isotope dendroprovenancing of four single trees from Arizona



Dendroclimatology with ring width is successful when the trees growing in stressful environments, such as high altitudes or arid environments, are used.^{11,12} However, forestry is normally practiced at low altitudes where the environment is optimal for tree growth. Most of the wood we use, whether modern or historical, comes from low altitudes. In such environments, the difference in coherence between tree-ring $\delta^{13}C$ and ring width is expected to become wider. For example, isotope chronologies of the trees from low altitudes in a humid climate, such as oaks from east England and redwood and pine from coastal California and Oregon, are known to show better coherence than ring-width chronologies.^{31,33} Even in semiarid Arizona, where the difference between isotope ratio and ring width is expected to become smaller, we observed a greater success rate for dendroprovenancing based on $\delta^{13}C$ versus ring width. This precision is expected to offer a great advantage for provenancing of timber or wooden artifacts. Further-

more, isotope dendroprovenancing requires fewer trees per site for building reference networks,⁴⁵ and single trees can be provenanced with a higher success rate than methods based on ring-width chronologies. Disadvantages are that isotope measurement is more time consuming (~8 min for one measurement) and costly (approximately \$5–10 US worth of consumables spent for one measurement) than ring width measurement (less than 10 s for one measurement at almost no cost).

Influence of distance and topography between the two sites on t values

As clearly seen in our results, t values are not a simple function of the distance between the two given sites. We saw a strong influence of intersite topography (Fig. 1) on t values as evidenced by the gradual decrease in t value moving

away from Kane Spring over the Colorado River drainage basin compared to the steep decrease over the Rocky Mountain range (Fig. 3a). Among the 91 possible combinations of 2 of the total 14 sites, there were 14 combinations in which the distance between the 2 sites was less than 300 km (Fig. 4). Four of these 14 combinations showed successful provenancing results ($t > 6.45$: Alton–Kane Spring, Dry Canyon–Kane Spring, Gate Canyon–Kane Spring, NC AZ–NE AZ), but the other 10 did not show t values significantly high enough to enable provenancing.

In eight of the ten unsuccessful combinations, high mountains exceeding 2500 m lie between the sites, potentially lowering the t values (Aztec–Dry Canyon, Aztec–Lower Colonias, Cerro Colorado–Mimbres, Cerro Colorado–NE AZ, NE AZ–Mimbres, Alton–NC AZ, Cerro Colorado–Dry Canyon, Cerro Colorado–Lower Colonias). The rugged topography of Colorado causes large variations in climate within short distances, and we must keep in mind that few climatic generalizations apply to the whole area.⁴⁶

Another reason for unsuccessful provenancing (Aztec–Cerro Colorado, Aztec–Dry Canyon, Aztec–Kane Spring, Aztec–Lower Colonias) could be the difference in climate sensitivity among trees.³⁹ For example, Aztec $\Delta_i(\text{AV})$ chronology did not show significantly high t values ($t > 6.45$) with any of the 13 reference chronologies (Fig. 3g). The four low-sensitivity series from NC AZ and NE AZ also did not show a significantly high t value with any of the reference chronologies. We believe that selecting climate-sensitive trees as references and then correcting for intertree difference in climate sensitivity may further improve the provenancing accuracy.

Error was smaller for provenancing results based on $\delta^{13}\text{C}$ (150–304 km) than results based on ring width (198–522 km; Fig. 4). When $\delta^{13}\text{C}$ provenancing was successful, the test chronologies had a maximum t value higher than the threshold t value at one of the neighboring sites close to the actual origin. As our experiment did not include $\delta^{13}\text{C}$ chronologies from sites less than 104 km apart (Fig. 4), we cannot say whether t values increase further at distances less than 104 km. The highest achievable spatial resolution of isotope provenancing methods depends on how much t values increase as the distance between test and reference sites approaches zero. In our study, we found that provenancing based on ring width often gave the highest t values at distant sites instead of sites neighboring the actual origin of test chronologies. We can therefore say that spatial accuracy of provenancing based on ring width was lower than provenancing based on $\delta^{13}\text{C}$. One potential reason for this difference is that $\delta^{13}\text{C}$ and ring widths are affected by partly different climatic parameters.^{11,30} For example, precipitation events are more localized phenomena than temperature-related events. Therefore, temperature observation data from a given station are correlated with those of surrounding stations in wider areas than when precipitation data are used. Carbon isotope chronology from eastern Siberia is also correlated with observed July temperature data from wider areas than July precipitation data (fig. 5D,E in Kirydanov et al.⁴⁷). Carbon isotope ratio is known as a good indicator of plant water-use efficiency,^{20,30} and it is strongly

influenced by precipitation. In fact, carbon isotope chronologies from the southwestern United States were used for reconstructing drought history.^{37,48}

Future prospects of isotope dendroprovenancing

The carbon isotope chronologies we used were pentads, and therefore we could not use cycles shorter than 5 years. Higher coherence of high-frequency versus low-frequency variations has been previously reported for both ring-width and carbon isotope chronologies.^{31,36,39} However, suppressed trees can occasionally experience sudden decreases in neighbor effects as a consequence of tree falls, resulting in individual-specific variations, especially in the low-frequency domain. Such conditions may explain why dendroprovenancing works best with high-frequency variations.¹⁶ Annual^{23,33,45} and subannual^{23,46,49} isotopic variations are reported to be coherent between trees at the same site and even between distant sites. Therefore, we believe methods based on both annual and subannual isotope variations have potential in dendroprovenancing and may even have a higher success rate than the methods based on pentads reported in this article. In theory, the time-resolution of tree-ring isotope archives can reach the monthly level at best.⁵⁰

Both SNR and t value maxima of $\Delta_i(\text{AV})$ were higher than those of $\text{RW}_i(\text{AV})$. The higher is the SNR of the tree-ring parameter (or the higher is the correlation coefficient), the higher the t value becomes. Because the signal amplitude (macroclimate) is common among the trees and constant within a site, we believe that reduction of noise is the key to improve accuracy of isotope dendroprovenancing. The noise of $\Delta_i(\text{AV})$ is primarily caused by within-site variability of tree-ring isotope ratios.^{23,33,49,51} For example, SNR calculated from annual $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ series from five *Quercus crispula* individuals from the same site⁵¹ are 7.8 and 14.6, respectively. The noise is also affected by the analytical error in isotope measurement.

We suggest the following measures to further improve the accuracy of isotope dendroprovenancing.

1. Usage of tree-ring oxygen isotopes³⁶ and/or hydrogen isotopes in combination with carbon isotopes to form network data.
2. Selection of reference trees with high sensitivity to common forcing. Correction for the difference in sensitivity among the reference trees may further improve provenancing results. However, this necessitates isotopic analysis of individual trees and excludes the option of pooling.
3. Usage of high-frequency isotope variation shorter than 5 years. Annual or even subannual fluctuations are coherent among trees within a site,^{23,33,48,49} and therefore dendroprovenancing may work with shorter frequencies than the pentads used in our study. This point is especially important when the number of available tree rings for provenancing is limited.
4. Development of reference isotope chronologies with higher spatial resolution. If future investigations reveal

that t values continue to increase at distances less than 104 km (see Fig. 4), we may be able to provenance trees with higher spatial accuracy.

We believe our study demonstrates the potential of identifying timber with falsely labeled origins at a spatial accuracy of less than 300 km through development of a tree-ring isotope network in areas affected by illegal logging, such as the Russian Far East and Baltic States. By analyzing disk or core samples with exact information about geographic origin (longitude and latitude), an isotope lab with sufficient facilities, staff, and funding should be able to develop such a network for one illegally logged region within a few years. However, for this method to be applied practically to market timbers, improvement of isotope analysis techniques, such as laser-ablation isotope ratio mass spectrometry,⁵² a method for rapid tree-ring isotope analysis at subannual resolution, is necessary. In the tropics, isotope dendroprovenancing may be first applied to tree species that form annual rings, such as teak.^{22,23} Teaks are one of the most illegally logged species in Indonesia and Burma.⁵³ Thus, there is a strong need for provenancing teak timber. In the future, laser-ablation IRMS may also hold the key for provenancing tropical timber that lacks tree rings, because even visually ringless tropical timber is reported to have annual isotope rings.²³

Acknowledgments This study was funded by research grants #200804 of the Forestry and Forest Products Research Institute, JSPS KAKENHI 21688014 and research fellowship of the Alexander von Humboldt Foundation. The authors thank Jennifer Lue for reviewing early drafts of this paper.

References

- Intergovernmental Panel on Climate Change (2007) Coupling between changes in the climate system and biogeochemistry. In: IPCC Fourth Assessment Report. Working group I report: the physical science basis. Cambridge University Press, Cambridge
- World Bank (2006) Formulation and implementation of national action plans to combat illegal logging and other forest crime. Results of ENA-FLEG. In: World Bank Technical Paper. World Bank, Washington, DC
- Sheingauz A (2004) Overview of the forest sector in the Russian Far East: production, industry and illegal logging. In: Working paper no. 2: forest trends. Asia Pacific Partners, Washington, DC
- Contreras-Hermosilla A, Doornbosch A, Lodge M (2007) The economics of illegal logging and associated trade. In: Report no. SG/SD/RT(2007)1. Organization for Economic Co-operation and Development, Paris
- Ohyama M, Baba K, Itoh T (2000) Wood identification of Japanese *Cyclobalanopsis* species (Fagaceae) based on DNA polymorphism of the intergenic spacer between *trn* T and *trn* L 5' exon. *J Wood Sci* 47:81–86
- Deguilloux MF, Pemonge MH, Bertel L, Kremer A, Petit RJ (2003) Checking the geographical origin of oak wood: molecular and statistical tools. *Mol Ecol* 12:1629–1636
- Kelly S, Heaton K, Hoogewerff J (2005) Tracing the geographic origin of food: The application of multi-element and multi-isotope analysis. *Trends Food Sci Technol* 16:555–567
- English NB, Betancourt JL, Dean JS, Quade J (2001) Strontium isotopes reveal distant sources of architectural timber in Chaco Canyon, New Mexico. *Proc Natl Acad Sci U S A* 98:11891–11896
- Kagawa A, Aoki T, Okada N, Katayama Y (2002) Tree-ring strontium-90 and cesium-137 as potential indicators of radioactive pollution. *J Environ Qual* 31:2001–2007
- Kagawa A, Kuroda K, Abe H, Fujii T, Itoh Y (2007) Stable isotopes and inorganic elements as potential indicators of geographic origin of Southeast Asian timber. In: Fujii T (ed) Proceedings of the international symposium on development of improved methods to identify *Shorea* species wood and its origin. Forestry and Forest Products Research Institute, Tsukuba, pp 39–44
- Fritts HC (1976) Tree rings and climate. Academic Press, London
- Schweingruber FH (1988) Tree rings. Basics and applications of dendrochronology. Kluwer, Dordrecht
- Noda M (1996) Analysis of time-series variation of tree-ring width (in Japanese). *Res Bull Hokkaido Univ For* 53:97–146
- Eckstein D, Brongers JA, Bauch J (1975) Tree-ring research in the Netherlands. *Tree-Ring Bull* 35:1–13
- Eckstein D, Wazny T, Bauch J, Klein P (1986) New evidence for the dendrochronological dating of Netherlandish paintings. *Nature (Lond)* 320:465–466
- Haneca K, Wazny T, Acker JV, Beeckman H (2005) Provenancing Baltic timber from art historical objects: success and limitations. *J Archaeol Sci* 32:261–271
- Wazny T (2002) Baltic timber in Western Europe: an exciting dendrochronological question. *Dendrochronologia* 20:313–320
- Leavitt SW (1993) Environmental information from $^{13}\text{C}/^{12}\text{C}$ ratios in wood. In: Swart PK (ed) Climate change in continental isotopic records. Geophysical Monograph 78. American Geophysical Union, Washington, DC, pp 325–331
- McCarroll D, Loader NJ (2004) Stable isotopes in tree rings. *Quat Sci Rev* 23:771–801
- Farquhar GD, O'Leary MH, Berry JA (1982) On the relationship between carbon isotope discrimination and intercellular carbon dioxide concentration in leaves. *Aust J Plant Physiol* 9:121–137
- Roden JS, Lin G, Ehleringer JR (2000) A mechanistic model for interpretation of hydrogen and oxygen isotope ratios in tree-ring cellulose. *Geochem Cosmochim Acta* 64:21–35
- Evans MN, Schrag DP (2004) A stable isotope-based approach to tropical dendroclimatology. *Geochem Cosmochim Acta* 68:3295–3305
- Poussart PF, Evans MN, Schrag DP (2004) Resolving seasonality in tropical trees: multi-decade, high-resolution oxygen and carbon isotope records from Indonesia and Thailand. *Earth Planet Sci Lett* 218:301–316
- Förstel H (2007) The natural fingerprint of stable isotopes: use of IRMS to test food authenticity. *Anal Bioanal Chem* 388:541–544
- Martin GJ, Guillou C, Martin ML, Cabanis MT, Tep Y, Aerny J (1988) Natural factors of isotope fractionation and the characterization of wines. *J Agric Food Chem* 36:316–322
- Robards K, Antolovich M (1995) Methods for assessing the authenticity of orange juice. A review. *Analyst* 120:1–28
- Suzuki Y, Chikaraishi Y, Ogawa NO, Okoshi N, Korenaga T (2008) Geographical origin of polished rice based on multiple element and stable isotope analyses. *Food Chem* 109:470–475
- Ehleringer JR, Casale JF, Lott MJ, Ford VL (2000) Tracing the geographic origin of cocaine. *Nature (Lond)* 408:311–312
- Barbour MM (2008) Stable oxygen isotope composition of plant tissue: a review. *Funct Plant Biol* 34:83–94
- Farquhar GD, Ehleringer JR, Hubick KT (1989) Carbon isotope discrimination and photosynthesis. *Annu Rev Plant Physiol Plant Mol Biol* 40:503–537
- Robertson I, Switsur VR, Carter AHC, Barker AC, Waterhouse JS, Briffa KR, Jones PD (1997) Signal strength and climate relationships in $^{13}\text{C}/^{12}\text{C}$ ratios of tree ring cellulose from oak in east England. *J Geophys Res* 102:19507–19516
- Kagawa A, Naito D, Sugimoto A, Maximov TC (2003) Effects of spatial and temporal variability in soil moisture on widths and $\delta^{13}\text{C}$ values of eastern Siberian tree rings. *J Geophys Res* 108:1–8
- Roden JS (2008) Cross-dating of tree ring $\delta^{18}\text{O}$ and $\delta^{13}\text{C}$ time series. *Chem Geol* 252:72–79
- Leavitt SW, Long A, Dean JS (1985) Tree-ring dating through pattern-matching of stable-carbon isotope time series. *Tree-Ring Bull* 45:1–10

35. Leavitt SW, Long A (1989) The atmospheric $\delta^{13}\text{C}$ records as derived from 56 pinyon trees at 14 sites in the southwestern United States. *Radiocarbon* 31:469–474
36. Treydte K, Frank D, Esper J, Andreu L, Bednarz Z, Berninger F, Boettger T, D'Alessandro CM, Etien N, Filot M, Grabner M, Guillemain MT, Gutierrez E, Haupt M, Helle G, Hiltunen E, Jungner H, Kalela-Brundin M, Krapiec M, Leuenberger M, Loader NJ, Masson-Delmotte V, Pazdur A, Pawelczyk S, Pierre M, Planells O, Pukienė R, Reynolds-Henne CE, Rinne KT, Saracino A, Saurer M, Sonninen E, Stievenard M, Switsur VR, Szczepanek M, Szychowska-Krapiec E, Todaro L, Waterhouse JS, Weigl M, Schleser GH (2007) Signal strength and climate calibration of a European tree-ring isotope network. *Geophys Res Lett* 34:L24302
37. Leavitt SW, Chase TN, Rajagopalan B, Lee E, Lawrence PJ, Woodhouse CA (2007) Southwestern U.S. drought maps from pinyon tree-ring carbon isotopes. *EOS Trans Am Geophys Union* 88:39–40
38. Leavitt SW, Long A (1985) The global biosphere as net CO_2 source or sink: evidence from carbon isotopes in tree rings. In: Caldwell DE, Brierly JA, Brierly CL (ed) *Planetary ecology*. Van Nostrand Reinhold, New York, pp 89–99
39. Leavitt SW, Long A (1988) Stable carbon isotope chronologies from trees in the southwestern United States. *Global Biogeochem Cycles* 2:189–198
40. Francey RJ, Allison CE, Etheridge DM, Trudinger CM, Enting IG, Leuenberger M, Langenfelds RL, Michel E, Steele LP (1999) A 1000-year high precision record of $\delta^{13}\text{C}$ in atmospheric CO_2 . *Tellus B* 51:170–193
41. Hollstein E (1980) Mitteleuropäische Eichenchronologie, Trierer dendrochronologische Forschungen zur Archäologie und Kunstgeschichte. In: *Trierer Grabungen und Forschungen*, Rheinisches Landesmuseum Trier, 11. Verlag Philipp von Zabern, Mainz am Rhein, p 273
42. Saurer M, Siegenthaler U, Schweingruber F (1995) The climate carbon isotope relationship in tree rings and the significance of site conditions. *Tellus B* 47:320–330
43. Loader NJ, Switsur VR, Field EM (1995) High resolution stable isotope analysis of tree rings: implications of “microdendrochronology” for palaeoenvironmental research. *Holocene* 5:457–460
44. Wigley TML, Briffa KR, Jones PD (1984) On the average value of correlated time series, with applications in dendroclimatology and hydrometeorology. *J Appl Meteorol Clim* 23:201–213
45. Leavitt SW, Long A (1984) Sampling strategy for stable carbon isotope analysis of tree rings in pine. *Nature (Lond)* 311:145–147
46. Doesken NJ, Pielke RA, Bliss OAP (2003) *Climate of Colorado. Climatology of the United States*, no. 60. National Climatic Data Center, Asheville, NC
47. Kirilyanov AV, Treydte KS, Nikolaev A, Helle G, Schleser GH (2008) Climate signals in tree-ring width, density and $\delta^{13}\text{C}$ from larches in Eastern Siberia (Russia). *Chem Geol* 252:31–41
48. Leavitt SW (1993) Seasonal $^{13}\text{C}/^{12}\text{C}$ changes in tree rings: species and site coherence, and a possible drought influence. *Can J For Res* 23:210–218
49. Roden JS, Johnstone JA, Dawson TE (2009) Intra-annual variation in the stable oxygen and carbon isotope ratios of cellulose in tree rings of coast redwood (*Sequoia sempervirens*). *Holocene* 19:189–197
50. Kagawa A, Sugimoto A, Yamashita K, Abe H (2005) Temporal photosynthetic carbon isotope signatures revealed in a tree ring through $^{13}\text{CO}_2$ pulse-labelling. *Plant Cell Environ* 28:906–915
51. Nakatsuka T, Ohnishi K, Hara T, Sumida A, Mitsuishi D, Kurita N, Uemura S (2004) Oxygen and carbon isotopic ratios of tree-ring cellulose in a conifer-hardwood mixed forest in northern Japan. *Geochem J* 38:77–88
52. Schulze B, Wirth C, Linke P, Brand WA, Kuhlmann I, Horna V, Schulze ED (2004) Laser ablation-combustion-GC-IRMS: a new method for online analysis of intra-annual variation of $\delta^{13}\text{C}$ in tree rings. *Tree Physiol* 24:1193–1201
53. Araya A (2003) Present state of decrease of forest area, illegal logging and illegal timber export in Indonesia. *Annual Report of Japan Federation of Wood-industry Associations*, Tokyo