Tree-ring cellulose δ^{18}O variability in pine and oak and its potential to reconstruct precipitation and relative humidity in central Japan

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Oxygen isotope ratios (δ^{18}O) of tree-ring cellulose in pine (*Pinus densiflora*) and oak (*Quercus serrate* and *Quercus variabilis*) growing in central Japan from 1970 to 2011 share a common inter-annual variance among different cores in a tree, different trees in a species, and different species in a forest. The high mean correlation between the two radii ($\bar{r}$ = 0.738) of individual trees and the mean standard deviation of two measurements for the same year from the two radii of eight trees ($\bar{\sigma}$ = 0.361‰) reveals the reliability and representativeness of single cores. The significant correlation (r = 0.667, $p < 0.01$) between pine and oak cellulose δ^{18}O suggests that δ^{18}O in tree-ring cellulose is controlled by common external factors, and that it is possible to establish a unique δ^{18}O chronology using different tree species. The δ^{18}O values of tree-ring cellulose in pine and oak trees are negatively correlated with precipitation from June to August (correlation coefficients are –0.679 for pine and –0.583 for oak), which is attributable to the precipitation amount effect, mainly caused by the stagnant rain front (Baiu) in early summer and typhoons in late summer in this region. The δ^{18}O values are negatively correlated with relative humidity from April to September for pine (r = –0.632, $p < 0.01$), and in June and July for oak (r = –0.437, $p < 0.01$); these correlations probably reflect different lengths of the growing season. Multiple regression analysis reveals that tree-ring δ^{18}O values of pine and oak in this region are largely governed by summer precipitation. Relative humidity does not appear to greatly affect tree-ring δ^{18}O of oak, in contrast to pine, which may be interpreted in terms of differences in leaf morphology and/or cellulose synthesis processes. Our results indicate that δ^{18}O values of tree-ring cellulose can provide a potential crossdating tool, irrespective of the physiological differences between gymnosperms and angiosperms. In central Japan, tree-ring δ^{18}O values of both pine and oak can act as reliable proxies of June–August precipitation, while pine trees tend to exhibit more potential for reconstruction of relative humidity.

Keywords: tree-ring cellulose δ^{18}O, pine, oak, precipitation, relative humidity, central Japan

INTRODUCTION

Dendrochronology is one of the most important of available techniques for reconstructing high-resolution chronologies of past climates (Fritts, 1976; Hughes, 2011). A well-established master tree-ring chronology can be used to estimate the precise ages of wood materials excavated at archaeological sites, and even to reconstruct paleoclimates. Many continuous long-term series have been established using long-lived trees (Cook *et al.*, 1991; Lara and Villalba, 1993; Hantemirov and Shiyatov, 2002) and shorter-lived woody species by extending the chronology beyond their lifetimes using living, archaeological, and sub-fossil samples through crossdating (Grudd *et al.*, 2002; Wilson *et al.*, 2004; Griggs *et al.*, 2007). Despite recent advances in chronology building using Japanese cedar (*Cryptomeria japonica*) (Kojo, 1987;

Ohyama *et al.*, 2013) and Hinoki cypress (*Chamaecyparis obtuse*) (Sweda, 1994; Yonenobu and Eckstein, 2006), long tree-ring chronologies are still sparse in Japan, especially for shorter-lived trees. Many woody gymnosperm and angiosperm species (e.g., Japanese cedar, Japanese umbrella pine, Hinoki cypress, oak, camphor, water elm) excavated at archaeological sites (Itoh, 1990) still require dendrochronological dating. Therefore, the establishment of chronologies for these species would make a significant contribution to the study of dendrochronology in Japan.

Because of developments in stable isotope dendrochronology, the study of oxygen isotope ratios (δ^{18}O) in tree-ring cellulose has developed rapidly in recent decades (e.g., Robertson *et al.*, 2001; Anderson *et al.*, 2002; Waterhouse *et al.*, 2002; Roden *et al.*, 2005; Leavitt *et al.*, 2006; Treydte *et al.*, 2007; Loader *et al.*, 2008). Many gymnosperm and angiosperm tree species have been used to construct chronologies based on δ^{18}O analyses, and to determine past climates; these include fir, beech, oak, and cedar in Europe (Switsur *et al.*, 1994,

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1996; Robertson *et al.*, 2001; Reynolds-Henne *et al.*, 2007); bristlecone pine in the United States (Bale *et al.*, 2011; Berkelhammer and Stott, 2012); larch, spruce, and pine in China (Liu *et al.*, 2008; Li *et al.*, 2011); fir in Nepal (Sano *et al.*, 2012); oak and fir in northern Japan (Nakatsuka *et al.*, 2004; Tsuji *et al.*, 2008); and cedar and cypress in central Japan (Yamaguchi *et al.*, 2010). Because the δ^{18}O value of tree-ring cellulose varies in response to climatic conditions, the values can be a potential source of paleoclimate information (McCarroll and Loader, 2004). Previous studies have reported that δ^{18}O values are strongly related to temperature (Burk and Stuiver, 1981; Ramesh *et al.*, 1986; Anderson *et al.*, 1998), precipitation (Saurer *et al.*, 2002; Liu *et al.*, 2004; Nakatsuka *et al.*, 2004; Treydte *et al.*, 2006), and relative humidity (Craig and Gordon, 1965; Burk and Stuiver, 1981; Ramesh *et al.*, 1986; Flanagan and Ehleringer, 1991; Tsuji *et al.*, 2008). The establishment of long-term chronologies for δ^{18}O can thus provide the basis for the study of paleoclimates.

Most tree-ring cellulose δ^{18}O studies have been conducted using only a single tree species in each study. Only a few studies have compared tree-ring oxygen isotope ratios of different species in the same forest stand, so as to study the responses of cellulose δ^{18}O of different species to climate (Nakatsuka *et al.*, 2004; Marshall and Monserud, 2006; Tsuji *et al.*, 2006; Saurer *et al.*, 2008; Li *et al.*, 2011). Inter-annual variations in δ^{18}O correlate well among species at the same site (Tsuji *et al.*, 2006; Li *et al.*, 2011) and even among different species located at different sites (Yamaguchi *et al.*, 2010; Li *et al.*, 2011). A master chronology of tree-ring δ^{18}O data based on single species can then be applied to other species, and it is therefore possible to build a unique isotope chronology that can be used for crossdating. However, some differences in inter-annual variations of cellulose δ^{18}O time series among different trees within a species and among different species in a forest do exist (Leavitt, 2010), and an estimate of oxygen isotopic consistency among different cores in a tree should provide guidance for sample collection protocols.

The first step in reconstructing paleoclimate using δ^{18}O chronologies is to determine the response of cellulose δ^{18}O to recent climate conditions. Responses of tree-ring δ^{18}O to climate can be consistent or variable among different species. Li *et al.* (2011) found that in northern China, δ^{18}O time series of three gymnosperm species were negatively correlated with precipitation and relative humidity during the growing season, not only at the same site but also among different sites. Of all the tested climate variables, relative humidity in June showed the strongest negative correlation with tree-ring δ^{18}O values at different sites in central Japan (Yamaguchi *et al.*, 2010). However, in northern Japan, the response of tree-ring δ^{18}O

to climate varied between gymnosperm and angiosperm species (spruce and oak) at the same site; the difference was attributed to their different root depths, combined with heavy snowfall in the region (Tsuji *et al.*, 2006). Therefore, combining the different responses of tree-ring cellulose δ^{18}O to climate from different species at the same site may provide us with more information on the past climate of that site.

Pine and oak are typical trees in the forests of central Japan, and many wooden materials made from those species have been excavated at archaeological sites. A comparison of tree-ring cellulose δ^{18}O values between these two tree species may reveal the usefulness of their tree-ring δ^{18}O values as a universal tool for dendrochronological dating of excavated woods in Japan, and/or as proxies of past climate.

In this study, we measured variations in individual tree-ring cellulose δ^{18}O values of pine (*Pinus densiflora*) and oak (*Quercus serrate* and *Q. variabilis*) growing in the same forest stand in Nagoya, located on the Pacific coast of central Japan. Our main goals were to: (i) estimate the inter-species consistency of tree-ring cellulose δ^{18}O inter-annual time series; (ii) examine the relationships between tree-ring cellulose δ^{18}O values and meteorological parameters; and (iii) assess the reliability of tree-ring δ^{18}O as a unique basis for chronology development and paleoclimate reconstruction using different tree species. We discuss the potential uses of our results for laying the foundations for further isotope dendroarcheological and dendroclimatological work in this region.

OXYGEN ISOTOPE THEORY

The first source of oxygen isotope variation in tree rings is related to differences in the isotope composition of source water, which is usually precipitation. The isotopic composition of atmospheric precipitation is partly dependent on environmental factors (Dansgaard, 1964; Welker, 2000). At middle and high latitudes, average monthly stable isotope ratios in precipitation are positively correlated with air temperature (i.e., the temperature effect; Dansgaard, 1964; Araguás-Araguás *et al.*, 1998). By contrast, at low latitudes and in maritime regions such as Japan, stable isotope ratios often decrease in months with higher precipitation, even during summer (i.e., the precipitation amount effect; Dansgaard, 1964).

The biological pathways that control how trees fractionate oxygen isotopes and incorporate them into their rings are well understood. Since trees absorb water from the soil rather than directly from precipitation, fractionation and mixing can occur before biological incorporation (McCarroll and Loader, 2004). If a tree takes up significant amounts of deeper soil water, the critical
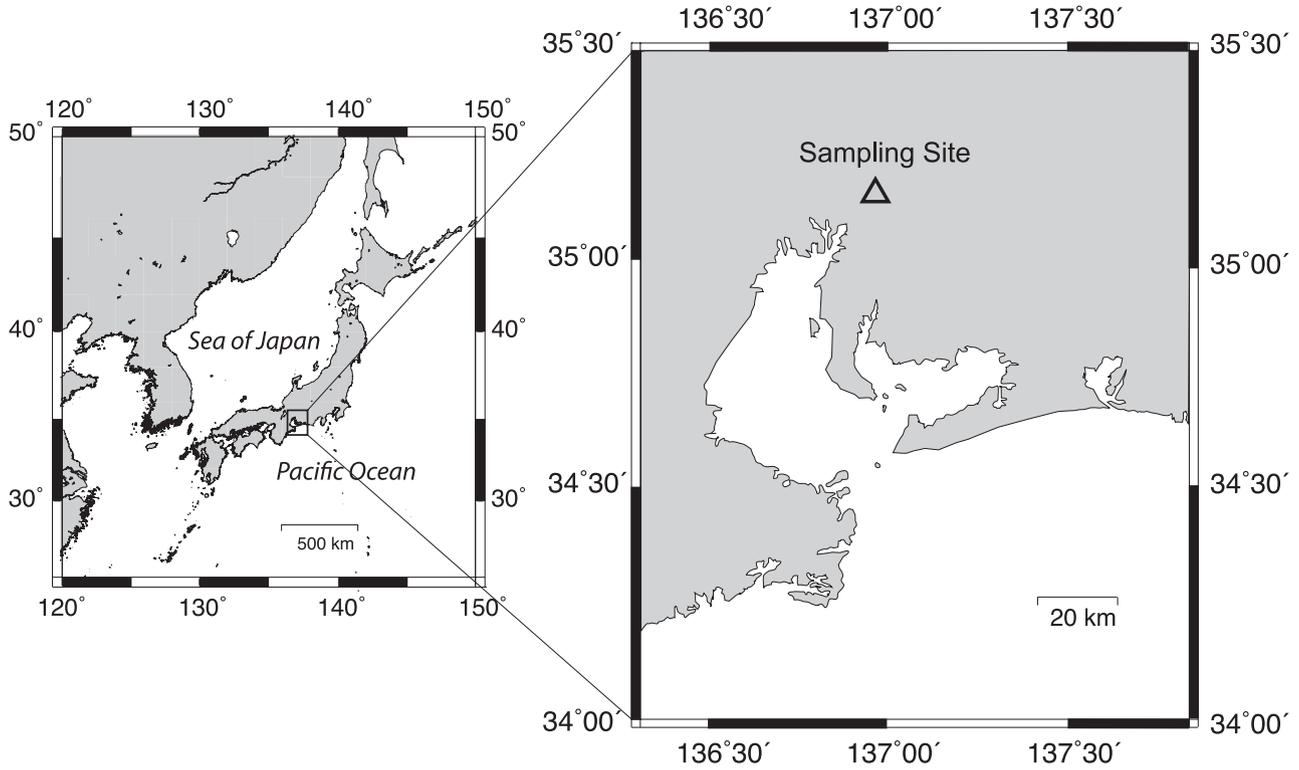
Fig. 1. The study region showing the sampling site (empty triangle) in south-central Japan.

factors controlling isotopic ratios are the total precipitation during the growing season and the rooting depth of the tree (Dawson and Pate, 1996). In the optimum case for climate reconstruction, source water (soil water) taken by trees is predominantly precipitation with the $\delta^{18}O$ signal of atmospheric water vapor (Rozanski *et al.*, 1992). Soil water is then transported to the leaves through the xylem.

The oxygen isotopic composition of leaf water is governed by transpiration through stomata, which leads to consequent enrichment in ^{18}O. A model for evaporative enrichment of $\delta^{18}O$ was developed by Craig and Gordon (1965), as:

$$\delta^{18}O_l = \delta^{18}O_X + \varepsilon^* + \varepsilon_k + e_a/e_i(\delta^{18}O_A - \varepsilon_k - \delta^{18}O_X) \quad (1)$$

where $\delta^{18}O_l$, $\delta^{18}O_X$, and $\delta^{18}O_A$ represent the $\delta^{18}O$ of leaf water, xylem water, and atmospheric water vapor, respectively, ε^* and ε_k are equilibrium and kinetic isotopic enrichment factors, respectively, and e_a/e_i is the ratio of ambient vapor pressure to intercellular vapor pressure, which can be assumed to be equal to the relative humidity (h) (Dongmann *et al.*, 1974). Assuming that atmospheric water vapor is isotopically equilibrated with precipitation, $\delta^{18}O_A$ can be represented as $\delta^{18}O_P - \varepsilon^*$, where $\delta^{18}O_P$ represents the $\delta^{18}O$ of precipitation. Xylem water does not undergo fractionation, and thus $\delta^{18}O$ values are equal to those of soil water and/or precipitation water, since water is taken up by roots without any isotopic fractionation (White *et al.*, 1985). Therefore, $\delta^{18}O_X$ is considered to be equal to $\delta^{18}O_P$. Equation (1) can be simplified as:

$$\delta^{18}O_l = \delta^{18}O_P + (\varepsilon^* + \varepsilon_k) (1 - h). \quad (2)$$

The pathway from leaf water to stem cellulose has been summarized as (Roden *et al.*, 2000):

$$\delta^{18}O_c = f(\delta^{18}O_X + \varepsilon_o) + (1 - f) (\delta^{18}O_l + \varepsilon_o) \quad (3)$$

where $\delta^{18}O_c$ represents the $\delta^{18}O$ value of tree-ring cellulose, f is the proportion of oxygen in carbohydrates that has been exchanged with xylem water, and ε_o is the biological fractionation factor between leaf water and sucrose during photosynthesis, and between xylem water and the exchanged oxygen in carbohydrates during cellulose synthesis (approximately 27‰; DeNiro and Epstein, 1981; Yakir and DeNiro, 1990; Sternberg *et al.*, 2006).

The simple form of Eq. (1) clearly suggests that tree-ring cellulose $\delta^{18}O$ time-series would be consistent among all tree species and individuals that live in the same climate region, which photosynthesize during the same season, and which absorb the same soil water. However,

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growing seasons and/or root depths may differ among different tree species and individuals, even in the same region. Therefore, our goal was to determine if tree-ring $\delta^{18}O$ time series are consistent or different among different species and individuals living in the same forest stand.

MATERIALS AND METHODS

Study site description

The study site is located in a secondary conifer and hardwood mixed forest (35°09′ N, 136°58′ E) in Nagoya, Aichi Prefecture, Japan (Fig. 1). The climate in Nagoya is influenced by its coastal setting on the Pacific Ocean. The climate is humid subtropical. High temperatures and humidity in summer are related to warm moist air masses coming from the Pacific, often accompanied by typhoons. In winter, cold dry air masses from the Asian continent prevail over Japan, where they meet warmer and moister air masses from the Pacific, the latter of which produce less snow but can still be quite cold. Instrumental climate records for the study area were obtained from the Japan Meteorological Agency records of the Nagoya Meteorological Observatory, located 2 km from the sampling site. Mean annual precipitation during 1970–2011 was 1561 mm. Monthly precipitation over the same period was greatest in September (231.5 mm) and least in December and January. Maximum and minimum monthly mean temperatures were approximately 27.5°C in August and 4.3°C in January, respectively. Mean monthly relative humidity reached its peak in July (74.5%) (Fig. 2).

Data collection and analysis

Using an increment borer with an inner diameter of 5 mm, core samples were collected at breast height from several pine (*Pinus densiflora*) and oak trees (*Quercus serrate* and *Q. variabilis*), whose average ages were approximately 50 years old. Two cores were obtained per tree, from opposite directions (north and south). Tree cores of pine (*Pinus densiflora*) were named P01-N and -S, P02-N and -S, P03-N and -S, and P04-N and -S, and tree cores of oak (*Q. serrate*) were named O01-N and -S, O02-N and -S, and O03-N and -S, while tree cores of oak (*Q. variabilis*) were named O04-N and -S. To investigate the inter-annual variations in tree-ring cellulose $\delta^{18}O$ and their relationship with climate, entire annual rings, including early wood and late wood, from the last 42 years (1970–2011) were used for isotopic analyses.

The tree-ring cellulose was extracted using the "plate" method (Kagawa and Nakatsuka, 2013). Each tree core was sliced into 1-mm-thick plates using a diamond wheel saw (FRIES Mecatone T180) along a plane perpendicular to the fibrous direction to extract α-cellulose. After packing the plates with Teflon-punch sheets, lipids were removed using organic solvents (acetone and a 1:1 tolu-
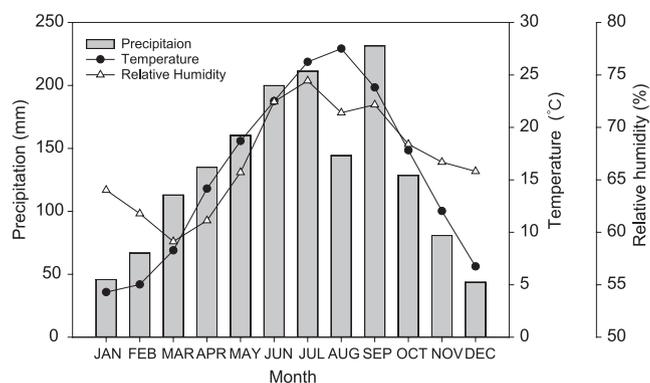

Fig. 2. The monthly mean precipitation, temperature and relative humidity averaged during 1970–2011 in Nagoya.

ene/ethanol mixture). Lignin and hemicellulose were decomposed and removed by acidified $NaClO_2$ solution and NaOH solution, respectively. After neutralization and drying, the cellulose plates were cut under a microscope using a design knife to separate the annual rings.

Approximately 0.12–0.22 mg of α-cellulose from each ring was packed in silver foil (in duplicate for each sample) and then pyrolyzed (Thermo Finnigan TC/EA, Bremen, Germany) into CO molecules at 1375°C. The CO gases were continuously introduced into a mass spectrometer (Delta V Advantage, Finnigan, Bremen, Germany) via an interface (ConFlo III) using He as the carrier gas, and the oxygen isotope ratios of the CO gases were measured.

Cellulose $\delta^{18}O$ was calculated by comparison with a standard material with a predetermined isotope ratio (commercial cellulose; Merck KGaA, Darmstadt, Germany); the standard material was inserted frequently during the measurement process. The isotope ratio is expressed in delta notation $\delta^{18}O$ (‰) relative to the international standard (VSMOW) for oxygen. The oxygen isotope ratio for an annual tree-ring cellulose sample was obtained as the mean of duplicate analyses. The standard deviation (1σ) of repeated analyses of standard material and the mean standard deviation of replicate samples were 0.15‰ and 0.22‰, respectively.

RESULTS AND DISCUSSION

Coherence among $\delta^{18}O$ time series in oak and pine trees

Absolute tree-ring cellulose $\delta^{18}O$ values from the same year but from different trees and from different radii are in good agreement, especially when comparing data from pine (P03) and oak (O02) trees (Supplementary Fig. S1). Inter-annual variations in $\delta^{18}O$ values of different trees are similar, and the correlation between the $\delta^{18}O$ values obtained from the two radii in eight trees is high ($\bar{r}$ = 0.738), although significant differences can be found in

some cases. The mean standard deviation of two measurements from the same year from two radii from the same tree is $\bar{\sigma} = 0.361‰$. Trends in $\delta^{18}O$ values from a single radius have generally been found to parallel those of other radii from the same tree, even though absolute values may differ. Ramesh *et al*. (1985) found variances in $\delta^{18}O$ values between different radii of 0.5–2‰, based on $\delta^{18}O$ analyses of *Abies pindrow*. Robertson *et al*. (1995) found isotopic variances among three radii of *Quercus petraea* of 0.5–1.5‰ for $\delta^{18}O$. Generally, compression wood is isotopically enriched. Specialized reaction wood in conifers is formed in response to tilting of a tree (Luckman and Gray, 1990). However, all the samples were collected from erect trees without reaction wood rings. Therefore, reaction wood cannot explain observations (the circumferential $\delta^{18}O$ differences) in this study. In other words, it is unlikely that reaction wood is important here.

In addition, circumferential $\delta^{18}O$ differences can be related to the influence of different source water compositions in different root directions (Edwards, 1993). In this paper, the mean $\delta^{18}O$ time-series of the two radii from each tree were reported to ensure accuracy; however, the mean deviation between the values of the two radii is small in comparison with values reported in previous studies, as mentioned above.

The absolute values of tree-ring cellulose $\delta^{18}O$ from individual trees were strongly correlated; i.e., absolute values of $\delta^{18}O$ values did not differ significantly among individuals within a given species (Supplementary Fig. S2 and Supplementary Table S1). Correlations between $\delta^{18}O$ variations from individuals of different species were slightly lower than those within the same species. No significant differences were observed between the $\delta^{18}O$ values of *Quercus variabilis* and those of *Q. serrate*.

To evaluate the coherence among tree-ring $\delta^{18}O$ chronologies from different trees, we calculated the Rbar (the mean inter-series correlation) and the expressed population signal (EPS; Wigley *et al*., 1984), which indicates how well the chronology estimates a theoretically infinite population. The EPS values, calculated using four individuals from each species, were 0.927 for pine (Rbar = 0.760, $N = 4$) and 0.910 for oak (Rbar = 0.717, $N = 4$), with a combined value for both species of 0.929 (Rbar = 0.62, $N = 8$). These EPS values are greater than the threshold value of 0.85, which is used to infer that a time-series actually represents a common regional signal (Wigley *et al*., 1984). These results therefore indicate that the four pine trees and four oak trees sufficiently capture the species and regional signals at the study site.

Non-climate-related trends in $\delta^{18}O$ time series should be considered in dendroclimatological studies. The "age-related effect" for $\delta^{18}O$ was first observed by Treydte *et al*. (2006) in young juniper trees from Pakistan. This juvenile effect is characterized by a reduction in values of
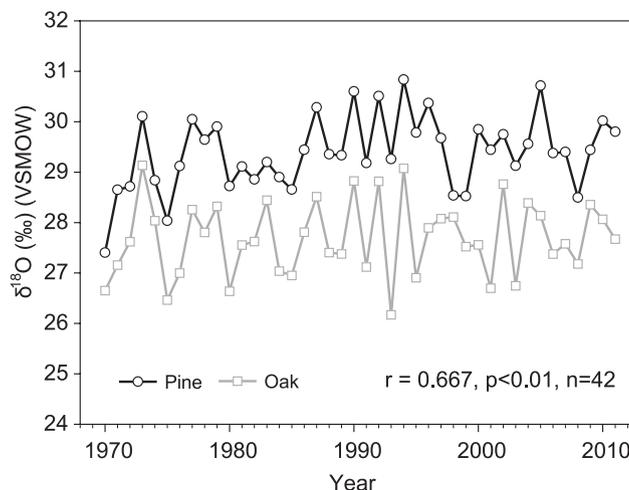

Fig. 3. Inter-annual variation in tree-ring cellulose $\delta^{18}O$ of pine and oak (averaged among four individuals) during 1970–2011. The correlation coefficient and significance are reported.

$\delta^{18}O$ in the years of growth after germination. For purposes of climate construction, this early portion has been typically removed, or the series has been detrended (Esper *et al*., 2010). However, age-related effects have not been observed in tree-ring cellulose $\delta^{18}O$ values of oak (Raffalli-Delerce *et al*., 2004) or Scots pine (Young *et al*., 2011). No evidence of a declining trend can be detected in the tree-ring cellulose $\delta^{18}O$ series in our study (Fig. S2). Thus, the $\delta^{18}O$ data may be considered as free of age-related growth effects, and are thus suitable for climate-related analyses.

Pine and oak show very similar inter-annual variabilities in their tree-ring $\delta^{18}O$ time series data (Fig. 3). Means and standard deviations of tree-ring cellulose $\delta^{18}O$ were $29.4 \pm 0.7‰$ for pine and $27.7 \pm 0.7‰$ for oak. The mean absolute tree-ring cellulose $\delta^{18}O$ of pine trees was 1.7‰ higher than that of oak trees. This difference may be attributable to physiological factors and biochemical effects. Recent studies have reported that enrichment of whole leaf water is significantly less than enrichment at the site of evaporation, because backward diffusion of enriched water is opposed by the convection of isotopically lighter source water to evaporation sites (Péclet effect; Farquhar and Lloyd, 1993; Barbour and Farquhar, 2000; Barbour *et al*., 2004). The Péclet effect is clearly important, especially in arid regions where transpiration rates are high (Barbour *et al*., 2004). However, in the humid climate of Japan, the Péclet effect does not seem to exert a strong influence on leaf water $\delta^{18}O$. In addition, the proportions of isotopic exchange between xylem water and carbohydrate oxygen exhibit species-specific variations (Hill *et al*., 1995; Roden *et al*., 2000), as do net biological fractionation factors (Gessler *et al*.,

2013), which affect the isotopic fractionation of oxygen.

A significant positive correlation was observed between pine and oak cellulose $\delta^{18}O$ over the period 1970–2011 ($r = 0.667$, $p < 0.01$, $n = 42$), during which time, for example, obvious low $\delta^{18}O$ values were present during specific years of both time series (e.g., 1975, 1993, and 2003). Both inter-species and intra-species correlations of $\delta^{18}O$ values are significant, revealing that tree-ring cellulose $\delta^{18}O$ values of species in central Japan are controlled by common external factors. Our results indicate that $\delta^{18}O$ values of tree-ring cellulose provide a potential crossdating tool, irrespective of differences between gymnosperm and angiosperm species, and are thus useful in archaeological and geological studies in central Japan; however, some exceptional species, such as evergreen broadleaf trees, which may have very long growing seasons, or trees with very long roots tapping deep ground water, may exhibit departures from the general trend.

Correlation between cellulose $\delta^{18}O$ and climate

A correlation analysis was conducted between tree-ring cellulose $\delta^{18}O$ values of pine trees, oak trees, and climatic factors (mean monthly temperature, monthly precipitation, and monthly relative humidity) during the period 1970–2011. For the correlations of tree-ring cellulose $\delta^{18}O$ with temperature, the values of tree-ring $\delta^{18}O$ from both species, were positively and significantly correlated with the temperature in July (for pine: $r = 0.381$, $p < 0.05$, $n = 42$; for oak: $r = 0.375$, $p < 0.05$, $n = 42$) and August (for pine: $r = 0.409$, $p < 0.01$, $n = 42$; for oak: $r = 0.393$, $p < 0.05$, $n = 42$) of the current year, while in June of the current year, the tree-ring $\delta^{18}O$ values of pine trees were significantly correlated with the temperature ($r = 0.405$, $p < 0.01$, $n = 42$) (Fig. 4 and Supplementary Fig. S3). For the correlations of tree-ring cellulose $\delta^{18}O$ with precipitation, the values of tree-ring cellulose $\delta^{18}O$ were significantly negatively correlated with precipitation, with the most significant highest absolute values observed from June to August. And the maximum correlation occurred in June of current year for pine tree ($r = -0.479$, $p < 0.01$, $n = 42$), while the maximum correlation occurred in August of current year for oak tree ($r = -0.447$, $p < 0.01$, $n = 42$). It is noteworthy that a significant negative correlation was observed between tree-ring $\delta^{18}O$ of pine and precipitation in April ($r = -0.343$, $p < 0.05$, $n = 42$). For the correlations of tree-ring cellulose $\delta^{18}O$ with relative humidity, the values of tree-ring cellulose $\delta^{18}O$ were negatively correlated with precipitation, with the most significant highest absolute values observed in April for pine tree ($r = -0.589$, $p < 0.01$, $n = 42$) and the high correlations persisted until September. And the maximum correlation occurred in July of current year for oak tree ($r = -0.371$, $p < 0.05$, $n = 42$). Although the correlations between the $\delta^{18}O$ values of some individuals with tempera-

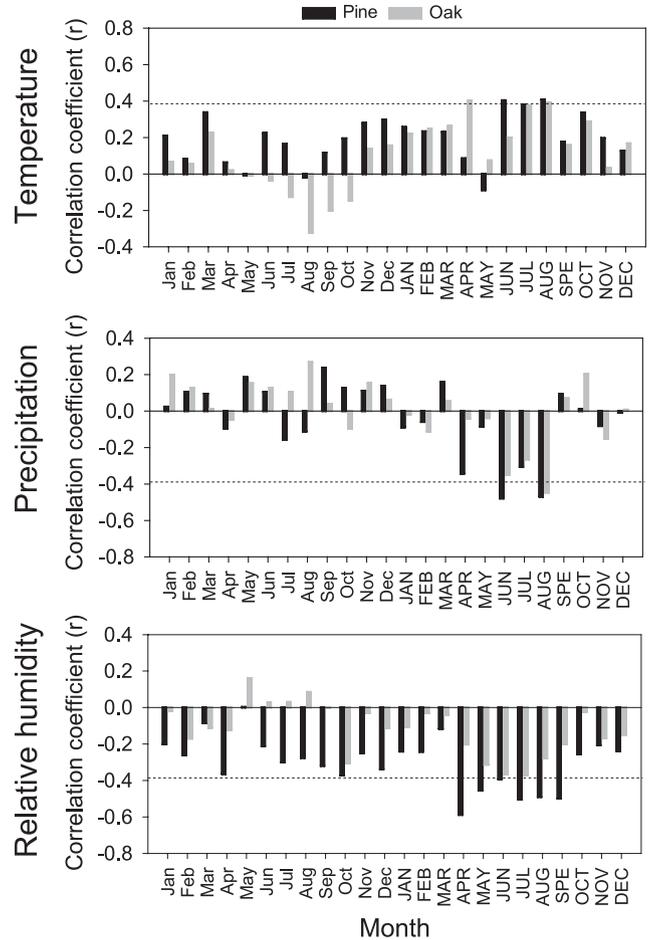*Fig. 4. Correlation coefficients of monthly temperature, monthly precipitation and relative humidity with $\delta^{18}O$ values of pine and oak, respectively. Correlation coefficients were calculated for average in the four individual pine trees as well as oak trees against monthly average of the climate data obtained in the current year (capital letters) and the previous year (small letters). Horizontal dotted line indicates 99% confidence level.*

ture and relative humidity were low (as compared with other trees) (Fig. S3), high correlations were observed for the averages of the four trees of each species (Supplementary Table S2).

The highest and most significant correlations were found between tree-ring cellulose $\delta^{18}O$ values of both species and precipitation in summer (June–August), by integrating multiple months from June to August, with correlation coefficients of -0.679 ($p < 0.01$, $n = 42$) for pine and -0.583 for oak ($p < 0.01$, $n = 42$). The relationship between tree-ring cellulose $\delta^{18}O$ and precipitation can be attributed to the negative correlation between $\delta^{18}O$ and the amount of precipitation (i.e., the amount effect) (Dansgaard, 1964) derived from the fractionation of source water before ^{18}O enters into the trees (Anderson
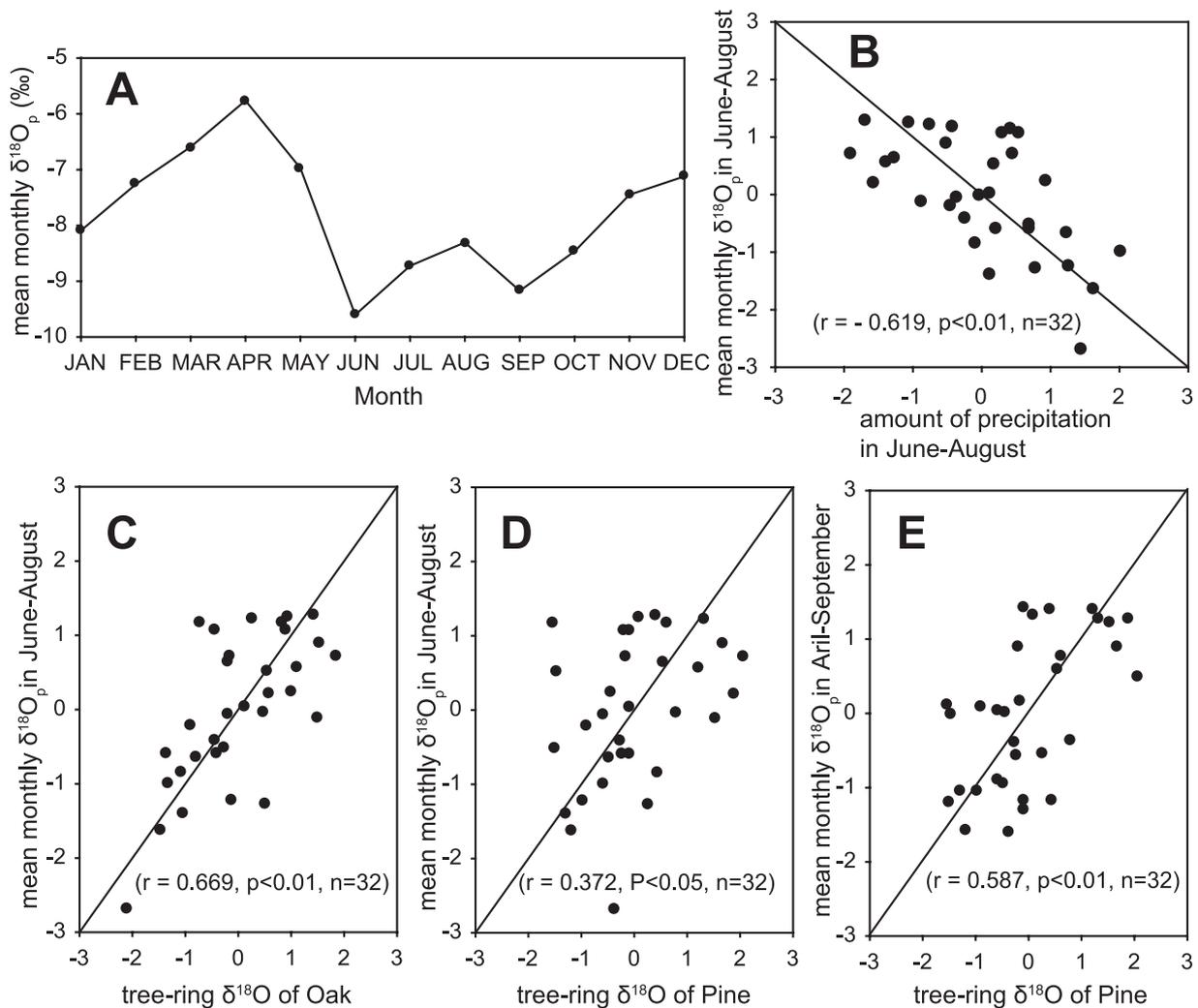
Fig. 5. The monthly modeled isotopic composition of source water $\delta^{18}O_p$ (in precipitation) derived from the nearest grid point of IsoGSM model (Yoshimura et al., 2008) (model dataset from website: http://hydro.iis.u-tokyo.ac.jp/~kei/?IsoGSM1) during 1979–2010 (A). (B) Standard major axis regression of mean monthly modeled $\delta^{18}O$ in precipitation ($\delta^{18}O_p$) in June–August as a function of amount of precipitation in June–August, and the amount of precipitation in June–August, as well as tree-ring cellulose $\delta^{18}O$ of pine and oak trees (C, D). The relationship between amount of precipitation in April–September and tree-ring cellulose $\delta^{18}O$ of pine (E). All of the data had been normalized (converted to z-scores) prior to the Standard major axis regression analysis. The correlation coefficient r and significance of the linear relationship p are reported. The solid lines represent the standard major axis regression namely 1:1 relationship.

et al., 2002). This relationship between tree-ring $\delta^{18}O$ and precipitation has been observed in other tree species and is widely recognized. Rinne et al. (2013) presented a significant correlation between tree-ring $\delta^{18}O$ of *Quercus robur* and precipitation, which has been used to reconstruct May–August precipitation during the last 400 years in England. Tree-ring $\delta^{18}O$ chronologies based on *Juniperus indica*, *Larix griffithii*, and *Picea spinulosa* from the Bhutan Himalaya have been developed, revealing that tree-ring $\delta^{18}O$ values are controlled mainly by summer precipitation (Sano et al., 2013).

The isotopic composition of precipitation is a signifi-

cant determinant of tree-ring cellulose $\delta^{18}O$ values (Saurer et al., 1997; Danis et al., 2006; Li et al., 2011; Brienen et al., 2013). During the summer months (June–August), the amount of precipitation near the study area exerts a strong control on the isotopic composition of the precipitation, as demonstrated by data from the nearest grid point of the IsoGSM model (Yoshimura et al., 2008: website: http://hydro.iis.u-tokyo.ac.jp/~kei/?IsoGSM1) (Fig. 5). Tree-ring cellulose $\delta^{18}O$ of oak is highly correlated with simulated $\delta^{18}O$ of precipitation ($r = 0.669$, $p < 0.01$, $n = 32$), while tree-ring cellulose $\delta^{18}O$ of pine trees shows a lower but still significant correlation with simulated precipita-

Table 1. *Multiple regression of interannual variation in tree-ring cellulose $\delta^{18}O$ with Precipitation (P, mm) and Relative Humidity (RH, %). All of the data had been normalized (converted to z-scores) prior to the analysis, marked as "$_{-N}$") Correlation coefficients and p values of regression and each variable are shown.*

Multiple regression		Correlation coefficients			p		
		Regression	Variables		Regression	Variables	
			P	RH		P	RH
Pine	$\delta^{18}O_{-N} = -0.486 \times P_{(Jun-Aug)-N} - 0.366 \times RH_{(Apr-Sep)-N}$	0.747	−0.486	−0.366	<0.001	<0.001	0.006
Oak	$\delta^{18}O_{-N} = -0.492 \times P_{(Jun-Aug)-N} - 0.163 \times RH_{(Jun-Jul)-N}$	0.598	−0.492	−0.163	<0.001	0.003	0.298

tion $\delta^{18}O$ in June–August ($r = 0.372$, $p < 0.05$, $n = 32$), which may be attributed to the longer growing season of pine as compared to oak. The influence of longer growing season of pine is confirmed by a higher correlation between tree-ring cellulose $\delta^{18}O$ of pine and precipitation $\delta^{18}O$ in April–September ($r = 0.587$, $p < 0.01$, $n = 32$). It appears that tree-ring cellulose $\delta^{18}O$ of pine trees tends to contain more information for months during spring, than for summer months. A significant negative correlation was observed between tree-ring $\delta^{18}O$ of pine and precipitation in April (Fig. 4). In fact, precipitation $\delta^{18}O$ reached a maximum in April as shown in Fig. 5A, in contrast to its values in summer; thus, the signal of precipitation $\delta^{18}O$ in summer is likely neutralized (dampened) in the tree-ring cellulose $\delta^{18}O$ data of pine.

Tree-ring cellulose $\delta^{18}O$ of pine was also negatively correlated with relative humidity from April to September ($r = -0.632$, $p < 0.01$, $n = 42$), while that of oak was correlated with relative humidity only in June and July ($r = -0.437$, $p < 0.01$, $n = 42$). This relationship between tree-ring cellulose $\delta^{18}O$ and relative humidity can be attributed to lower ^{18}O enrichment in leaf water due to higher relative humidity, based on the leaf-water enrichment model (Craig and Gordon, 1965; Roden *et al*., 2000). The influence of relative humidity on pine trees $\delta^{18}O$ persists longer than for oak trees, suggesting that lengths of the growing seasons of pine and oak are different; e.g., the growing season of the evergreen Japanese red pine (*Pinus densiflora*) is longer than that of the deciduous oak "konara" (*Quercus serrate*) (Kushida, 2000). Many researchers have found similar relationships between tree-ring $\delta^{18}O$ and relative humidity in other species. Ramesh *et al*. (1986) confirmed that $\delta^{18}O$ values of *Abies pindrow* are significantly controlled by relative humidity in India. And, a study of tree-ring cellulose $\delta^{18}O$ in *Pseudotsuga menziesii* in Washington, USA, revealed that relative humidity can be recorded in tree-ring cellulose (Shu *et al*., 2005).

Results of a multiple regression analysis, performed using precipitation and relative humidity as predictors, are shown in Table 1. To make the regression expression more intuitive, all of the data were normalized (converted to z-scores) prior to the analysis. The multiple regres-

sions for pine and oak were both significant, although the significance level for pine was higher than that for oak (multiple $r = 0.747$, $p < 0.001$ for pine, and multiple $r = 0.598$, $p < 0.001$ for oak). These results revealed that tree-ring $\delta^{18}O$ is largely governed by these two climatic factors. The dominant environmental signals in tree-ring $\delta^{18}O$ are precipitation and relative humidity during the growing season, although the relative strengths of these two signals vary according to the tree species.

First, the precipitation amount is a dominant influence on the isotopic ratio of source water for tree growth, while relative humidity influences leaf water enrichment. For pine trees, both precipitation ($r = -0.486$, $p < 0.001$) and relative humidity ($r = -0.366$, $p < 0.01$) appear to contribute to the results of tree-ring $\delta^{18}O$ values. In oak trees, not all independent variables appear to influence $\delta^{18}O$ values greatly, and relative humidity does not seem to affect tree-ring $\delta^{18}O$ values as much as precipitation does. These differences between pine and oak may be interpreted in terms of differences in leaf morphology (Barbour *et al*., 2002). In broad-leaved trees, because of the greater potential for evaporative cooling, leaf physiology is less strongly coupled to environmental parameters. Leaf temperatures vary with stomatal conductance, which affects e_i and ε^* in Eq. (1). Thus, the difference between e_a/e_i and relative humidity is greater in oak than in pine. In pine trees, needle temperature remains close to that of air, and the response of tree-ring $\delta^{18}O$ to changes in stomatal conductance is small. Thus, e_a/e_i is much more likely to be equal to the actual relative humidity.

In addition, biochemical effects play an important role in isotopic fractionation. Broadleaf species, such as oak, show high isotopic exchange rates between carbohydrates and xylem water prior to cellulose synthesis (Hill *et al*., 1995); namely, values of f in Eq. (3) are high, and thus, the oxygen isotope signal derived from leaf water enrichment will be dampened. For the reasons mentioned above, it seems understandable that tree-ring cellulose $\delta^{18}O$ in oak shows less sensitivity than pine to relative humidity.

We found patterns of coherence between inter-annual variations in tree-ring cellulose $\delta^{18}O$ in pine and oak trees and spring and/or summer climate (Fig. 6). For example, remarkably large amounts of precipitation in June–
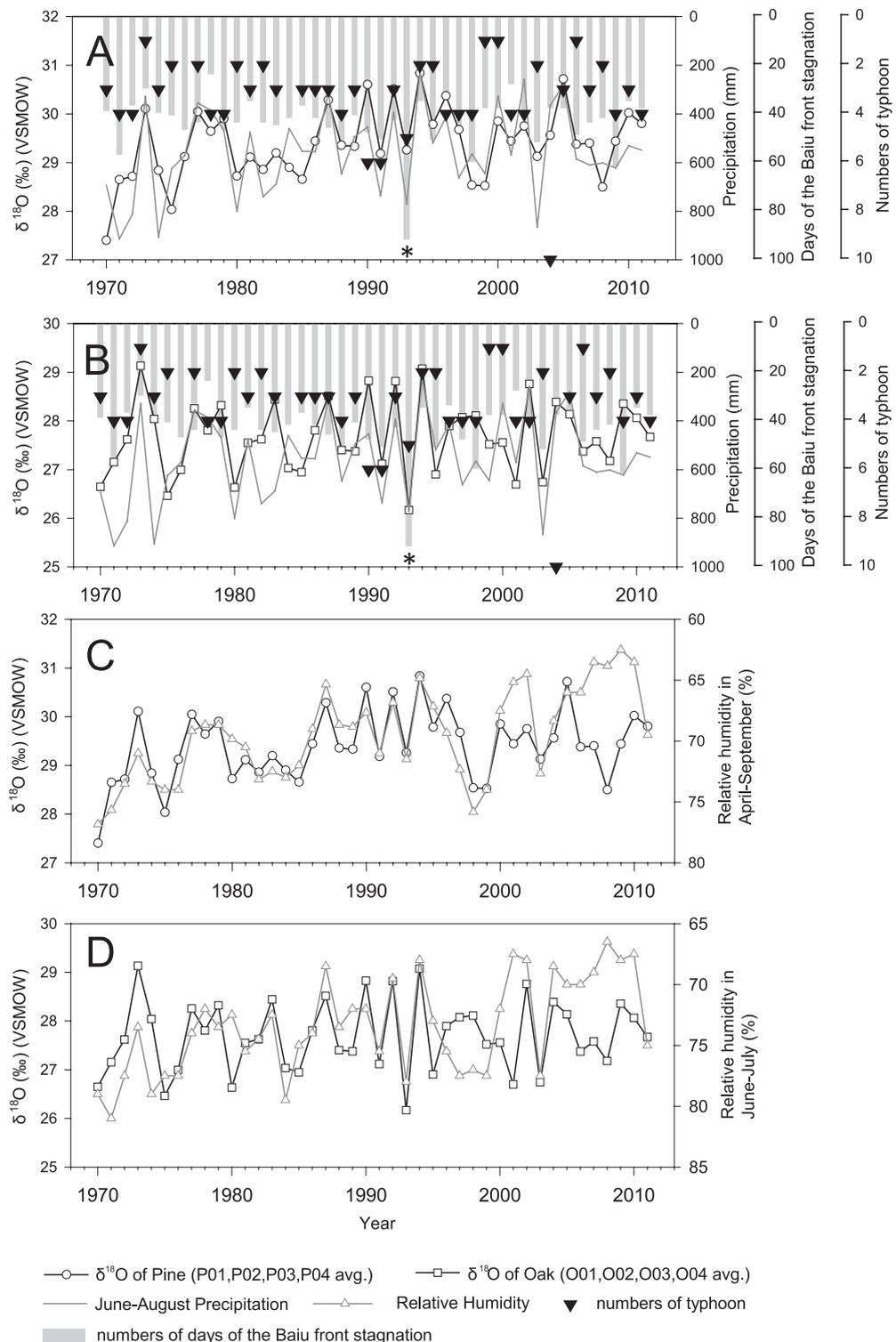
Fig. 6. Inter-annual variations in tree-ring cellulose δ¹⁸O of pine and oak trees and June–August precipitation as well as relative humidity in April–September and June–July respectively. (A) δ¹⁸O of pine and amount of precipitation in June–August; (B) δ¹⁸O of oak and amount of precipitation in June–August; (C) δ¹⁸O of pine and mean relative humidity in April–September; (D) δ¹⁸O of Oak and mean relative humidity in June–July. The inter-annual variations of days of Baiu front stagnation and numbers of typhoon events that passed within 300 km of study area had been added. (*, the end of Baiu in 1993 could not be identified).

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August 1991, 1993, and 2003 correspond to low $\delta^{18}O$ values in pine and oak trees. This correspondence can be attributed to the amount effect in the oxygen isotope ratio of precipitation, which results in lower $\delta^{18}O$ values of heavy rainfall events in summer (Dansgaard, 1964). In central Japan, the main factor influencing summer rainfall $\delta^{18}O$ is the arrival of the East Asian summer monsoon and its characteristic stationary front, the Baiu front. In early summer, during the rainy season, the Baiu front develops over most areas of Japan (except for on Hokkaido Island), along the boundary of the maritime subtropical high in the western Pacific and the maritime subarctic high in the Okhotsk Sea; the front is characterized by heavy rains generated by interactions between the air masses (Yoshimura, 1967). The Baiu front is stationary for approximately 40 days each a year (the stagnation period), during which rainfall amounts constitute as much as one-third of the total annual precipitation in Japan (Hirahara *et al.*, 2012).

In central Japan, the Baiu season usually starts in June and ends in July, according to the positions of the subtropical and subarctic air masses, respectively (Akiyama, 1973), but it sometimes stagnates for a prolonged interval due to the sustained presence of subarctic high pressure until August, resulting in prolonged cloudy and rainy summers in Japan. We found that the low tree-ring cellulose $\delta^{18}O$ values of pine and oak in 1991 and 1993 corresponded with years of long-term stagnant Baiu front conditions (based on meteorological records from the Japan Meteorological Agency, http://www.data.jma.go.jp/fcd/yoho/baiu/kako_baiu08.html). Particularly, in 1993, the end of the Baiu could not be identified because wet and cloudy weather continued until late August. This anomaly caused significant socio-economic damage, especially to agricultural sectors. In fact, the summer of 1993 was recorded as the coldest summer on record since 1954 (JMA, 1994). This prolonged unsettled weather caused by the Baiu brings not only large amounts of precipitation but also high relative humidity, which is a likely reason for lower tree-ring cellulose $\delta^{18}O$ values, as per Eq. (2).

Because of its geographical location, the study area also tends to be affected by typhoons. The precipitation in heavy rainfall events related to typhoons is relatively isotopically depleted (Lawrence and Gedzelman, 1996; Lawrence *et al.*, 2004), and such events may be recorded in tree-ring $\delta^{18}O$ chronologies. The number of typhoons occurring annually in the study area was counted as the number of typhoon events that have passed within 300 km of the study area (Fig. 6). For instance, the low tree-ring $\delta^{18}O$ values of pine and oak in 1991 and 1993 correspond to years when devastating typhoons influenced central Japan. These observations indicate that the high negative correlation between precipitation in summer and the $\delta^{18}O$ values of pine and oak are the result of substantial rainfall events that occur during the growing seasons.

In 1993, the extraordinarily long and cold rainy period in summer was the result of a combination of intense typhoon events and prolonged Baiu; these conditions also caused low rice production, resulting in importation of large amounts of rice (Davidson *et al.*, 1998; Hosoe, 2004; Krishnan and Sugi, 2001). Conversely, high $\delta^{18}O$ values of tree rings coincide with years with few typhoons (e.g., in 1973); in fact, most parts of Japan suffered from drought conditions in the summer of 1973.

CONCLUSIONS

We found evidence for circumferential, intra-species, and inter-species coherences in inter-annual tree-ring cellulose $\delta^{18}O$ time-series data from pine and oak trees. We demonstrated that the inter-annual variations between species are similar, suggesting that $\delta^{18}O$ values of the tree-ring cellulose of each species, each tree, and each radius is controlled by common external factors, despite significant differences in absolute $\delta^{18}O$ values among different species. High negative correlations are present between summer precipitation and $\delta^{18}O$ values of tree-ring cellulose in pine and oak trees, indicating that the tree-ring $\delta^{18}O$ values of these species can serve as proxies for the amount of summer precipitation in central Japan, which, in turn, is mainly influenced by the stationary rain front (Baiu front) in early summer and typhoon events in late summer. Tree-ring cellulose $\delta^{18}O$ values of pine showed strongly negative correlations with a long period of relative humidity during April–September, while those of oak showed negative correlations with relative humidity mainly during June and July. This difference between pine and oak might be attributable to different growing seasons. Multiple regression analysis revealed that tree-ring $\delta^{18}O$ is largely governed by summer precipitation in the region, for both pine and oak. Relative humidity does not seem to greatly affect tree-ring $\delta^{18}O$ values of oak, in contrast with pine, which may be interpreted as a difference in leaf morphology or biosynthesis processes of cellulose in the cambium.

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Supplementary Materials

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Figures S1 to S3
Tables S1 and S2