A 2600-year summer climate reconstruction in central Japan by integrating tree-ring stable oxygen and hydrogen isotopes

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Abstract. Oxygen isotope ratios (δ\textsuperscript{18}O) of tree-ring cellulose are a novel proxy for summer hydroclimate in monsoon Asia. In central Japan, we collected 67 conifer wood samples, mainly Chamaecyparis obtusa, with ages encompassing the past 2600 years. The samples were taken from living trees, archeological wood, architectural wood, and buried logs. We analyzed stable isotope ratios of oxygen (δ\textsuperscript{18}O) and hydrogen (δ\textsuperscript{2}H) in tree-ring cellulose in these samples (more than 15 000 rings in total) without using a pooling method and constructed a statistically reliable tree-ring cellulose δ\textsuperscript{18}O time series for the past 2500 years. However, there were distinct age trends and level offsets in the δ\textsuperscript{18}O record, and cellulose δ\textsuperscript{18}O values showed a gradual decrease as an individual tree matures. This suggested it is difficult to establish a cellulose δ\textsuperscript{18}O chronology for low-frequency signals by simple averaging of all the δ\textsuperscript{18}O time series data. In addition, there were opposite age trends in the cellulose δ\textsuperscript{2}H, and δ\textsuperscript{2}H gradually increased with tree age. There were clear positive correlations in the short-periodicity variations between δ\textsuperscript{18}O and δ\textsuperscript{2}H, probably indicating a common climate signal. A comparison of the δ\textsuperscript{18}O and δ\textsuperscript{2}H time series in individual trees with tree-ring width suggested that the opposite age trends of δ\textsuperscript{18}O and δ\textsuperscript{2}H are caused by temporal changes in the degree of post-photosynthetic isotope exchange with xylem water (physiological effect), accompanied by changes in stem growth rate that are influenced by human activity in the forests of central Japan. Based on the assumptions that cellulose δ\textsuperscript{18}O and δ\textsuperscript{2}H vary positively and negatively with constant proportional coefficients due to climatological and physiological effects, respectively, we solved simultaneous equations for the climatological and physiological components of variations in tree-ring cellulose δ\textsuperscript{18}O and δ\textsuperscript{2}H in order to remove the age trend. This enabled us to evaluate the climatic record from cellulose δ\textsuperscript{18}O vari-
This requires three analytical criteria to evaluate the age sets between different individual trees, in a conifer species. Of years after germination, as well as significant level offsets. Moreover, the ecological and artificial disturbance of this is because tree-ring widths measured in trees from northern Japan are responsible for long-term age trends remain unclear.

In general, it is not straightforward to extract low-frequency climate signals from tree-ring time series because there are typically age trends in tree-ring width. To overcome this, the regional curve standardization (RCS) method is used to determine the typical growth curve in a region for a tree species, which is subtracted from the tree-ring width time series of individual trees to explicitly estimate climatological influences (Esper et al., 2003), but it is difficult to fully preserve low-frequency climate signals of tree-ring width and density.

Tree-ring cellulose oxygen isotope ratios (\(\delta^{18}O\)) are a novel proxy for past summer hydroclimate, and it has been reported that such data are not affected by age trends (Young et al., 2011; Kilroy et al., 2016; Xu et al., 2016; Büntgen et al., 2020) or only show short-lived juvenile effects of higher \(\delta^{18}O\) values (Szymczak et al., 2012; Duffy et al., 2017). These juvenile effects are interpreted to be related to the less-developed root system of young trees and the evaporative enrichment of \(^{18}O\) in soil water in the near surface. However, Esper et al. (2010) demonstrated the presence of long-term age trends, whereby \(\delta^{18}O\) gradually decreases for hundreds of years after germination, as well as significant level offsets between different individual trees, in a conifer species. This requires three analytical criteria to evaluate the age trends and level offsets in the tree-ring cellulose \(\delta^{18}O\) data: (1) well-replicated data, (2) non-pooled data, and (3) composite chronologies from multiple species and regions. However, given that subsequent studies of tree-ring cellulose \(\delta^{18}O\) have reported that age trends are negligible, the mechanisms responsible for long-term age trends remain unclear.

In Japan, it has been difficult to establish millennial-scale tree-ring chronologies applicable to reconstructing past changes in summer climate, even though such information is critical for rice paddy cultivation and for historical and archeological studies in Japan. Although there are ancient archeological and architectural woods that have been used for dendrochronological analysis of wooden artifacts with ages of 1000–2000 years (Mitsutani, 2000; NNRIPCP, 1990), these have not been utilized for past climate reconstructions. This is because tree-ring widths measured in trees from normal Japanese forests do not have a high sensitivity to climate. Moreover, the ecological and artificial disturbance of tree growth in dense Japanese forests prevents reconstructions of past climate based on the relatively small number of old sub-fossil wood samples that are available.

However, tree-ring cellulose \(\delta^{18}O\) data are sensitive to summer hydroclimate in Asia (Treydte et al., 2006; Grießinger et al., 2011; Li et al., 2011; Xu et al., 2011; Sano et al., 2012; Liu et al., 2017), including in Japan (Nakatsuka et al., 2004; Tsuji et al., 2008; Yamaguchi et al., 2010; Li et al., 2015). High correlation coefficients for tree-ring cellulose \(\delta^{18}O\) data amongst different individual trees and species make it possible to reconstruct past summer climate using only a small number of sub-fossil trees. Therefore, during the past decade we have analyzed tree-ring cellulose \(\delta^{18}O\) in various types of conifer wood with large tree-ring numbers (mainly Chamaecyparis obtusa), including from living trees, archeological wood, architectural wood, and buried logs, in order to reconstruct past variations in summer hydroclimate in central Japan (Nakatsuka, 2015, 2018).

We do not use the pooling method (Szymczak et al., 2012; Loader et al., 2013) for the tree-ring isotope measurements. Instead, we analyze all the tree rings in the wood samples using a novel method, involving direct cellulose extraction from 1 mm thick wood sections (Kagawa et al., 2015). This allows us to evaluate age trends in individual trees and offsets between different individuals in the tree-ring cellulose \(\delta^{18}O\) data, following the criteria proposed by Esper et al. (2010). This approach has shown that most old trees in Japan (mainly Chamaecyparis obtusa) are significantly affected by long-term age trends and level offsets such that it is not possible to reconstruct long-term summer climate variations without taking these effects into account.

However, we have also found that hydrogen isotope ratios (\(\delta^2H\)) of tree-ring cellulose, which have been measured simultaneously with the \(\delta^{18}O\) data, have age trends opposite to those of \(\delta^{18}O\) and that \(\delta^2H\) is positively and negatively correlated with \(\delta^{18}O\) in terms of short- and long-periodicity variations, respectively. This \(\delta^{18}O\) and \(\delta^2H\) relationship provides insights into the mechanisms responsible for age trends in tree-ring cellulose \(\delta^{18}O\) data and makes it possible to remove the age trends in such data.

Here, we compare \(\delta^{18}O\) and \(\delta^2H\) data for tree-ring cellulose and elucidate the mechanisms responsible for age trends in tree-ring cellulose \(\delta^{18}O\) data. We then propose and apply a new method to the tree-ring cellulose \(\delta^{18}O\) and \(\delta^2H\) time series for reconstructing climate over the past 2600 years in central Japan. This enables us to remove the age trend and extract the climatological component of the tree-ring cellulose \(\delta^{18}O\) data using many ecologically and artificially disturbed trees that are available for the last several thousand years, even in Japan.
2 Samples and chemical analyses

2.1 Collection of tree-ring samples

A total of 67 tree samples covering the past 2600 years were collected in central Japan (Fig. 1). These samples consist of living trees, architectural wood, archeological wood, and buried logs (Table S1 in the Supplement). Most of the trees are Japanese cypress (Chamaecyparis obtusa), but there are some samples of other conifer species (Chamaecyparis pisifera, Sciadopitys verticillata, and Cryptomeria japonica). All samples contain hundreds of rings. Although some samples have already been dated by traditional dendrochronological methods using tree-ring width (Mitsutani, 2000; NNRICP, 1990), the new tree-ring δ18O time series were used in the final dendrochronological determination of their ages (Roden, 2008; Yamada et al., 2018), which provided results consistent with the tree-ring width dating results (Fig. S1).

2.2 Measurement of tree-ring cellulose δ18O and δ2H

Here, we analyzed cellulose δ18O and δ2H for all tree rings in all wood samples using a novel method, involving direct cellulose extraction from 1 mm thick wood laths (Kagawa et al., 2015), without using the pooling method. Firstly, 1 mm thick cross-sectional laths perpendicular to the cellulose fibers were sliced from all wood samples regardless of ring width and age and directly subjected to chemical treatment to remove components other than cellulose. In order to keep the original shape of samples during chemical treatments, 1 mm thick wood laths were put between two Teflon punching sheets and set into test tubes. Given that buried wood sometimes shows extraordinary δ18O values due to alteration of chemical components and/or deposition of iron oxides (Savard et al., 2012; Yapp, 2001), we carefully selected less-degraded wood segments and completely removed all components other than cellulose. A 100–300 µg fragment of cellulose representing the whole annual layer was then cut from the cellulose lath year by year using a fine blade under a microscope (Fig. S2). Each sample was wrapped in silver foil for δ18O and δ2H measurements using a mass spectrometer combined with a pyrolysis elemental analyzer (TCEA/Delta V Advantage; ThermoFisher Scientific, Bremen, Germany). In this study, we analyzed both δ18O and δ2H values for each cellulose sample simultaneously by making the focus of the mass spectrometer automatically jump between outflows of H2 and CO gases from the pyrolysis elemental analyzer. After measurements of every eight samples, a cellulose standard (cellulose reagent; Merck) with known δ18O and δ2H was measured to calibrate analytical drift and calculate the δ18O and δ2H of samples against the international standard of VSMOW, following Eqs. (1) and (2). Analytical reproducibility (1σ) based on repeated measurements of homogeneous cellulose samples is ca. ± 0.15 and ± 1.5‰ for δ18O and δ2H, respectively. Absolute accuracies of δ18O and δ2H may be a little bit worse because we use only one working standard to minimize the memory effect in the δ2H measurement. However, it does not influence the discussion of this study because we do not utilize absolute isotope values but focus only on their temporal variations.

\[
\delta^{18}O_{\text{vsmow}}(\%) = \left( \frac{^{18}O/^{16}O}_{\text{sample}} \right) \left( \frac{^{18}O/^{16}O}_{\text{vsmow}} \right) - 1 \times 1000
\]  
\[
\delta^{2}H_{\text{vsmow}}(\%) = \left( \frac{^{2}H/^{1}H}_{\text{sample}} \right) \left( \frac{^{2}H/^{1}H}_{\text{vsmow}} \right) - 1 \times 1000
\]

Given that cellulose contains 30% OH-group hydrogen that is exchangeable with experimental water during chemical treatment, it is necessary to remove the OH group by nitration of the cellulose (synthesis of nitrocellulose) before the isotope measurements (Epstein et al., 1976; DeNiro, 1981). However, it was nearly impossible to nitrate all the cellulose samples analyzed in this study due to the very small sample amount (Fig. S2) and the extremely large sample number (15 924 rings for isotopic measurements). Moreover, it was desirable to measure both δ18O and δ2H simultaneously for each sample, not only to reduce the sample number to half, but also to treat the δ18O and δ2H data as completely originating from the same sample, as discussed later, but the nitration of cellulose makes it impossible to measure its δ18O together with δ2H due to the contamination of oxygen in the NO2 group. However, we can expect that all OH-group hydrogen in a cellulose lath will have a unique δ2H value, as it would have been replaced by the hydrogen of homogeneous water in the test tube during the process of cellulose extraction such that the measured tree-ring cellulose δ2H time se-
Correlations between hydrogen isotope ratios (δ2H) of tree-ring cellulose and nitrocellulose in two Japanese cypress trees. (a) No. 65 and (b) no. 64 in Table S1. Red lines indicate the slope observed when all OH-group hydrogen in cellulose is exchanged with hydrogen with a unique δ2H value.

3 Results and discussion

3.1 Variations in the tree-ring cellulose δ18O and δ2H data

After constraining the exact dates for all the tree rings by the cross-dating method using δ18O data (Fig. S1; Bunn, 2008), we compared all tree-ring cellulose δ18O and δ2H time series (Fig. 3a and c).

Given that the average correlation coefficient (R bar) amongst the different δ18O time series is ca. 0.6–0.8, the expressed population signal (EPS) (Wigley et al., 1984), defined as $N \times R/[1 + (N - 1) \times R]$, where the $N$ and $R$ values are the number of time series and $R$ bar, respectively, is > 0.85 for almost all periods during the past 2500 years (Fig. 3b), indicating that the δ18O data in Fig. 3a are sufficient to establish accurate δ18O chronologies. However, there are distinct long-term age trends in that the δ18O values gradually decrease as the trees mature, as also noted by Esper et al. (2010). These effects must be detrended in order to reconstruct long-term climate variations using the δ18O data. There are also significant offsets in absolute δ18O values for each year amongst different trees, probably reflecting differences in the sample location and magnitude of the age trend. Given that the original locations of the trees cover a wide region from coastal to inland areas and low to high altitudes (Fig. 1), this would result in different precipitation δ18O values and relative humidity (Waseda and Nakai, 1983; Aragúas-Araguás et al., 2000; Shu et al., 2005). This would account for the offset absolute δ18O values of tree-ring cellulose in different trees.

The R bar for the different δ2H time series is ca. 0.2–0.4, resulting in lower EPS values (Fig. 3d), which must reflect more complicated post-photosynthetic changes in isotopic ratios in hydrogen than in oxygen (Roden et al., 2000), as discussed later, but may be partly owing to the analytical uncertainty from the direct measurement of cellulose hydrogen including the OH group in the δ2H measurements. However, there are also distinct age trends, which are opposite to those exhibited by δ18O in that δ2H values gradually increase as the trees mature.

3.2 Relationship between short- and long-periodicity δ18O and δ2H variations

To investigate the environmental causes of the age trends in tree-ring cellulose δ18O and δ2H data, we decomposed the δ18O and δ2H time series into short- and long-periodicity variations (Figs. 4, S3, S4, and S5). These trees (no. 65 and nos. 43, 49, 40; Table S1) are Japanese cypress (Cha-mae-cyparis obtusa) from Nagano and Gifu Prefectures. In all cases, short-periodicity variations (i.e., <11 years) in the tree-ring cellulose δ18O and δ2H data show a clear positive correlation, indicating that both the δ18O and δ2H variations reflect a common climatological signal (i.e., precipitation isotope ratios and relative humidity) as reported in previous studies (e.g., An et al., 2014). However, the long-periodicity variations (i.e., > 11 years) in the tree-ring cellulose δ18O and δ2H data show negative correlations (Figs. 4b and S3b), suggesting the influence of opposite age trends in Fig. 3. However, the negative correlations are not very clear in Figs. S4b and S5b, probably because the climatological signals also overlap the long-periodicity variations and make the correlation between the tree-ring cellulose δ18O and δ2H positive to a certain degree.

Figures 4b, S3b, S4b, and S5b show that δ18O and δ2H often changed oppositely in the long-periodicity variations, regardless of the tree age. Therefore, the long-term opposite variations in the tree-ring cellulose δ18O and δ2H data are not a simple age trend. As such, it is impossible to apply the RCS or negative exponential curve proposed by Esper et al. (2010) to remove the apparent age trend of tree-ring cellulose δ18O in central Japan.

To date, the age trend in tree-ring cellulose δ18O data has been explained by the relatively shallow root system of ju-
Figure 3. The tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ time series and their statistical properties. (a) Tree-ring cellulose $\delta^{18}O$ time series of the 67 analyzed trees. (b) $R$ bar (average correlation coefficients between different individual trees), EPS (see text), and sample depth (tree sample numbers) in the corresponding periods for the tree-ring cellulose $\delta^{18}O$ dataset. (c) Tree-ring cellulose $\delta^2H$ time series of the 67 analyzed trees. (d) $R$ bar, EPS, and sample depth for the tree-ring cellulose $\delta^2H$ dataset. All data in panels (b) and (d) were calculated for a total period of 51 years, including 25 years before and after the year shown.

venile trees and the evaporative enrichment of $^{18}O$ in soil water at the near surface. However, this cannot explain the long-term variations in tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ data in this study (Figs. 4, S3, S4, and S5) because the soil water evaporation must increase both $\delta^{18}O$ and $\delta^2H$. As illustrated in Sect. S1 and Fig. S6, the long-term opposite variations in tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ are tightly related to changes in tree-ring width (growth rate of trees), suggesting that they are influenced by some physiological effect corresponding to long-term changes in tree growth rate.

3.3 Mechanical model of cellulose $\delta^{18}O$ and $\delta^2H$

What physiological mechanism can explain the long-term opposite variations in the $\delta^{18}O$ and $\delta^2H$ values? Tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ values can be expressed using the following equations, which are modified and simplified from Roden et al. (2000):

$$\delta^{18}O_{cel} = f \left( \delta^{18}O_{rain} + \varepsilon_{ho} \right) + (1 - f) \left[ \delta^{18}O_{rain} + (\varepsilon_{eo} + \varepsilon_{ko})(1 - h) + \varepsilon_{ao} \right],$$

(3)
δf is constant. Because there are always positive correlations between δf in Eqs. (3) and (4) in the same direction, we can expect any change in δf only by climatological factors (Nabeshima et al., 2018), rather than directly using photosynthetic products for rapid tree growth during the juvenile period. The change in the f value could also occur in the period following an abrupt improvement in the growth environment due to logging of neighboring trees (see Sect. S1 and Fig. S6).

Although the long-periodicity variations in δ18Ocel and δ2Hcel are influenced by predominant physiological effects (Figs. 4b, S3b, S4b, and S5b), this does not mean that the long-periodicity variations in δ18Ocel and δ2Hcel do not con-
tain climatological components. In fact, climate varies at all timescales such that long-term variations in $\delta^{18}O_{\text{cel}}$ and $\delta^2H_{\text{cel}}$ inevitably include climatological components. It is therefore challenging to resolve the climatic signals from the physiological effects. In dendrochronological studies based on tree-ring width, RCS is used to separate and estimate climatological components in tree-ring width time series (Esper et al., 2002; Grudd et al., 2002; Büntgen et al., 2005). However, it is difficult to create a regional standardized $\delta^{18}O_{\text{cel}}$ curve like the RCS for the samples analyzed in this study because the physiological effects on $\delta^{18}O_{\text{cel}}$ are not solely an age trend and also reflect past human disturbances to the trees such as logging activity (see Sect. S1; Totman, 1989).

3.4 Classification of $\delta^{18}O$ and $\delta^2H$ variations into climatological and physiological components

To extract the climatological component of the variations in the tree-ring cellulose $\delta^{18}O$ data, we modified the model of cellulose $\delta^{18}O$ and $\delta^2H$ in Eqs. (3) and (4) into climatological and physiological components. Given there are four variables ($\delta^{18}O_{\text{rain}}, \delta^2H_{\text{rain}}, h,$ and $f$) in Eqs. (3) and (4), we first define their variations as follows:

\[
\delta^{18}O_{\text{rain}} = \delta^{18}O_{\text{rain}(0)} + \Delta \delta^{18}O_{\text{rain}},
\]

\[
\delta^2H_{\text{rain}} = \delta^2H_{\text{rain}(0)} + \Delta \delta^2H_{\text{rain}},
\]

\[
h = h(0) + \Delta h,
\]

\[
f = f(0) + \Delta f,
\]

where $\delta^{18}O_{\text{rain}(0)}, \delta^2H_{\text{rain}(0)}, h(0),$ and $f(0)$ are $\delta^{18}O_{\text{rain}}, \delta^2H_{\text{rain}}, h,$ and $f$ in a fixed year (0), respectively, and $\Delta \delta^{18}O_{\text{rain}}, \Delta \delta^2H_{\text{rain}}, \Delta h,$ and $\Delta f$ are deviations in $\delta^{18}O_{\text{rain}}, \delta^2H_{\text{rain}}, h,$ and $f$ from the fixed year (0) to an arbitrary year, respectively. By substituting Eqs. (5)–(8) into Eqs. (3)–(4) and neglecting the second-order minor terms ($\Delta f \Delta h, \Delta f \Delta \delta^{18}O_{\text{rain}},$ and $\Delta f \Delta \delta^2H_{\text{rain}}$), Eqs. (3) and (4) can be rewritten as follows:

\[
\delta^{18}O_{\text{cel}} = f(0) \left[ \delta^{18}O_{\text{rain}(0)} + \epsilon_{ho} \right] + \left( 1 - f(0) \right) \left[ \delta^{18}O_{\text{rain}(0)} + (\epsilon_{eo} + \epsilon_{ko}) \left( 1 - h(0) \right) \right] + \epsilon_{ao} + \Delta f \left[ \epsilon_{ho} - \epsilon_{ao} - (\epsilon_{eo} + \epsilon_{ko}) \left( 1 - h(0) \right) \right],
\]

\[
\delta^2H_{\text{cel}} = f(0) \left[ \delta^2H_{\text{rain}(0)} + \epsilon_{hh} \right] + \left( 1 - f(0) \right) \left[ \delta^2H_{\text{rain}(0)} + (\epsilon_{eh} + \epsilon_{kh}) \left( 1 - h(0) \right) \right] + \epsilon_{ah} + \Delta f \left[ \epsilon_{hh} - \epsilon_{ah} - (\epsilon_{eh} + \epsilon_{kh}) \left( 1 - h(0) \right) \right].
\]

We can now introduce new equations for the climatological and physiological components in the tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ time series, as follows:

\[
\Delta \delta^{18}O_{\text{cel}(\text{climate})} = \Delta \delta^{18}O_{\text{rain}} - \Delta h (\epsilon_{eo} + \epsilon_{ko}) \left( 1 - f(0) \right),
\]

\[
\Delta \delta^2H_{\text{cel}(\text{climate})} = \Delta \delta^2H_{\text{rain}} - \Delta h (\epsilon_{eh} + \epsilon_{kh}) \left( 1 - f(0) \right),
\]

\[
\Delta \delta^{18}O_{\text{cel}(\text{physiol})} = \Delta f \left[ \epsilon_{ho} - \epsilon_{ao} - (\epsilon_{eo} + \epsilon_{ko}) \left( 1 - h(0) \right) \right],
\]

\[
\Delta \delta^2H_{\text{cel}(\text{physiol})} = \Delta f \left[ \epsilon_{hh} - \epsilon_{ah} - (\epsilon_{eh} + \epsilon_{kh}) \left( 1 - h(0) \right) \right],
\]

where $\Delta \delta^{18}O_{\text{cel}(\text{climate})}, \Delta \delta^{18}O_{\text{cel}(\text{physiol})}, \Delta \delta^2H_{\text{cel}(\text{climate})},$ and $\Delta \delta^2H_{\text{cel}(\text{physiol})}$ are variations in the tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ from the fixed year (0) with respect to an arbitrary year due to climatological and physiological factors, respectively. We can then reformulate $\delta^{18}O_{\text{cel}}$ and $\delta^2H_{\text{cel}}$ as the sum of climatological and physiological components, as follows:

\[
\delta^{18}O_{\text{cel}} = \delta^{18}O_{\text{cel}(0)} + \Delta \delta^{18}O_{\text{cel}(\text{climate})} + \Delta \delta^{18}O_{\text{cel}(\text{physiol})},
\]

\[
\delta^2H_{\text{cel}} = \delta^2H_{\text{cel}(0)} + \Delta \delta^2H_{\text{cel}(\text{climate})} + \Delta \delta^2H_{\text{cel}(\text{physiol})},
\]

where $\delta^{18}O_{\text{cel}(0)}$ and $\delta^2H_{\text{cel}(0)}$ are $\delta^{18}O_{\text{cel}}$ and $\delta^2H_{\text{cel}}$ at the fixed year (0), respectively.

3.5 A method to extract the climatological component from cellulose $\delta^{18}O$

Here we propose a new method to calculate the climatological component in variations of tree-ring cellulose $\delta^{18}O$ (\(\Delta \delta^{18}O_{\text{cel}(\text{climate})}\)) by solving simultaneous equations consisting of Eqs. (15)–(16), with two additional equations based on the relationship between $\Delta \delta^{18}O_{\text{cel}(\text{climate})}$ and $\Delta \delta^2H_{\text{cel}(\text{climate})}$ and between $\Delta \delta^{18}O_{\text{cel}(\text{physiol})}$ and $\Delta \delta^2H_{\text{cel}(\text{physiol})}$ (i.e., Eqs. 17–18). This is based on the theoretical and observational understanding that tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ data correlate positively and negatively due to climatological and physiological factors, respectively. (Note that we restrict the physiological effects to the change in the rate of post-photosynthesis isotope exchange with xylem water here. If there are other types of ecophysiological effects generating a positive relationship between $\delta^{18}O_{\text{cel}}$ and $\delta^2H_{\text{cel}}$, such as root deepening or growth season shift, they are counted as parts of the climatological effects here.)

\[
\Delta \delta^2H_{\text{cel}(\text{climate})} = A \times \Delta \delta^{18}O_{\text{cel}(\text{climate})},
\]

\[
\Delta \delta^2H_{\text{cel}(\text{physiol})} = -B \times \Delta \delta^{18}O_{\text{cel}(\text{physiol})},
\]

where $A$ and $B$ are positive proportional coefficients, as shown in Eqs. (19)–(20) and defined by Eqs. (11)–(14).

\[
A = \frac{\Delta \delta^2H_{\text{rain}} - \Delta h (\epsilon_{eh} + \epsilon_{kh}) \left( 1 - f(0) \right)}{\Delta \delta^{18}O_{\text{rain}} - \Delta h (\epsilon_{eo} + \epsilon_{ko}) \left( 1 - f(0) \right)},
\]

\[
B = \frac{\epsilon_{hh} - \epsilon_{ah} - (\epsilon_{eh} + \epsilon_{kh}) \left( 1 - h(0) \right)}{\epsilon_{ho} - \epsilon_{ao} - (\epsilon_{eo} + \epsilon_{ko}) \left( 1 - h(0) \right)}.
\]
If \( A \) and \( B \) are constant, we can solve the simultaneous Eqs. (15)–(18) in order to cancel out the physiological component (\( \Delta \delta^{18}O_{\text{cell(physiol)}} \)) and extract the climatological component (\( \Delta \delta^{18}O_{\text{cell(climate)}} \)), as follows:

\[
\Delta \delta^{18}O_{\text{cell(climate)}} = \frac{\delta^2H_{\text{cell}} + B \times \delta^{18}O_{\text{cell}}}{A + B} - \frac{\delta^2H_{\text{cell}(0)} + B \times \delta^{18}O_{\text{cell}(0)}}{A + B}.
\] (21)

In fact, \( A \) might not be constant because there are three variables (\( \Delta \delta^{18}O_{\text{rain}}, \Delta \delta^2H_{\text{rain}}, \) and \( \Delta h \)) in Eq. (19). \( \Delta \delta^{18}O_{\text{rain}} \) and \( \Delta \delta^2H_{\text{rain}} \) may change somewhat independently from each other over a wide spatiotemporal range, and the relationship between \( \Delta \delta^{18}O_{\text{rain}} \) (\( \Delta \delta^2H_{\text{rain}} \)) and \( \Delta h \) may not be simple in Eq. (19). However, the significant positive correlations in the short-periodic variations between \( \delta^{18}O_{\text{cell}} \) and \( \delta^2H_{\text{eq}} \) (Figs. 4c, S3c, S4c, and S5c) suggest there may be an empirical constant value for \( A \) in a restricted study area such as central Japan. In contrast to \( A \), \( B \) may be nearly constant because there are no variables in Eq. (20).

Although the assumption that \( A \) and \( B \) are constant might not be valid, this assumption is needed to explicitly calculate \( \Delta \delta^{18}O_{\text{cell(climate)}} \) using Eq. (21). Hence, we tentatively assumed that \( A \) and \( B \) are constant and calculated \( \Delta \delta^{18}O_{\text{cell(climate)}} \) over the past 2600 years using Eq. (21). We then verify this by comparison with numerous local, regional, and global meteorological, historical, and paleoclimatological records of past summer climate over various timescales in order to judge whether the simplest assumption, that \( A \) and \( B \) in Eq. (21) are constant, is practically useful or not for paleoclimatological studies.

### 3.6 Method to determine the proportional coefficients \( A \) and \( B \)

To utilize Eq. (21), we need to determine the proportional coefficients \( A \) and \( B \) in Eqs. (17)–(20). In principle, there are two ways to determine \( A \) and \( B \): a theoretical approach based on Eqs. (19)–(20) and an empirical approach based on Eqs. (17)–(18).

We first consider the feasibility of the theoretical approach. It is not easy to determine \( A \) using Eq. (19) because there are three variables (\( \Delta \delta^{18}O_{\text{rain}}, \Delta \delta^2H_{\text{rain}}, \) and \( \Delta h \)). If the rainwater isotope ratios do not change (both \( \Delta \delta^{18}O_{\text{rain}} \) and \( \Delta \delta^2H_{\text{rain}} = 0 \)), then \( A \) is equal to (\( \varepsilon_{\text{eh}} + \varepsilon_{\text{hh}} \))/(\( \varepsilon_{\text{eo}} + \varepsilon_{\text{ao}} \)), which is (80 + 25)/(9 + 29) = 2.76 (Roden and Ehleringer, 2000). However, if relative humidity does not change (\( \Delta h = 0 \)), \( A = \Delta \delta^2H_{\text{rain}}/\Delta \delta^{18}O_{\text{rain}} \) which is equal to 8 if the meteoric waterline is followed (Dansgaard, 1964). Given the wide range of potential \( A \) values from 2.76 to 8, we cannot easily theoretically define \( A \). On the other hand, \( B \) may be easier to determine theoretically because there are no variables in Eq. (20). For example, if the relative humidity \( h(0) \) is 0.5, \( B = 13 \) because \( \varepsilon_{\text{eo}}, \varepsilon_{\text{ko}}, \varepsilon_{\text{ao}}, \varepsilon_{\text{ho}}, \varepsilon_{\text{eh}}, \varepsilon_{\text{kh}}, \varepsilon_{\text{ah}}, \) and \( \varepsilon_{\text{hh}} \) are +9, +29, +27, +27, +80, +25, ca. −150, and +150, respectively (Rodan et al., 2000). However, in fact, it is not easy to fix \( h(0) \) because the relative humidity typically shows large diurnal variations, and the timing of \( \delta^{18}O \) incorporation into leaf carbohydrates is unknown. Moreover, the \( \varepsilon_{\text{ah}} \) and \( \varepsilon_{\text{hh}} \) values of −150 and +150 are just approximations. For example, while Yakir and DeNiro (1990) obtained values of −171 and +158 for \( \varepsilon_{\text{ah}} \) and \( \varepsilon_{\text{hh}} \), respectively, Estep and Hoering (1981) obtained values of −100 to −120 for \( \varepsilon_{\text{ah}} \) and Luo and Sternberg (1992) reported values of +144 to +166 for \( \varepsilon_{\text{hh}} \).

Therefore, we used the empirical approach to estimate \( A \) and \( B \). The \( A \) value in Eq. (17) can be estimated from the relationship between \( \delta^{18}O_{\text{cell}} \) and \( \delta^2H_{\text{cell}} \) in the short-periodicity variations because they are positively correlated due to the climate variations shown in Figs. 4c, S3c, S4c, and S5c. In the case of the \( B \) value in Eq. (18), the situation is more complex. Although there are apparent negative correlations between \( \delta^{18}O_{\text{cell}} \) and \( \delta^2H_{\text{cell}} \) in the long-periodicity variations (Figs. 4b and S3b), these are also affected by long-term climate signals such that it is difficult to resolve the physiological component. Here we propose a practical method to determine the \( B \) value. After fixing \( A \), we first set various \( B \) values in order to calculate the temporal variations in the climatological component of the tree-ring cellulose \( \delta^{18}O \) data (\( \Delta \delta^{18}O_{\text{cell(climate)}} \)) for all 67 tree samples by substituting individual \( \delta^{18}O_{\text{cell}} \) and \( \delta^2H_{\text{cell}} \) time series in Fig. 3a and c into Eq. (21). We then combine the \( \Delta \delta^{18}O_{\text{cell(climate)}} \) time series of all 67 trees calculated from each \( B \) value using the method described below (Sect. 3.7). Finally, we compare the overall trend of the combined time series of \( \Delta \delta^{18}O_{\text{cell(climate)}} \) data with that of a simple average of all 67 raw \( \delta^{18}O_{\text{cell}} \) time series in Fig. 3a in order to find the \( B \) value that produced the best match between the two time series. This procedure assumes that, although individual trees possess different age trends and level offsets, averaging of all data from all trees covering all periods and all locations in this study may cancel out these effects and allow climate variations to be discerned, at least in terms of “the overall trend”.

### 3.7 Method to combine individual tree-ring time series with large level offsets

To reconstruct multi-millennial variations in the climatological component of the tree-ring cellulose \( \delta^{18}O \) data (\( \Delta \delta^{18}O_{\text{cell(climate)}} \)), we must combine individual time series from different trees with variable level offsets due to different sample locations. As such, we cannot simply average all data for individual trees because this produces steps in the composite record at both ends of the time series of individual trees. Numerous procedures have been proposed to combine tree-ring time series (e.g., Hangartner et al., 2012); here we propose a new iterative calculation method (Fig. 5).
that of the combined time series for the corresponding period, without any further offsetting of the individual time series. If you start from the same original dataset on δ¹⁸O variations of stem segments, the pattern of relative variation in the final combined δ¹⁸O time series is mathematically unique although its absolute value has no meaning. That is, there is no room for artifacts to influence resultant time series. This method assumes that the tree-ring absolute isotope ratios of individual trees are not important because they depend on the sample locations but that the temporal variations are well correlated amongst different trees due to a common regional climate signal.

### 3.8 Procedure to calculate the climatological component in tree-ring δ¹⁸O (Δδ¹⁸O<sub>cel(climate)</sub>) data

Figure 6a shows how the combined time series for Δδ¹⁸O<sub>cel(climate)</sub> is constructed. Firstly, we determine A from the short-periodicity variations between δ¹⁸O<sub>cel</sub> and δ²H<sub>cel</sub> in individual trees. Secondly, we use δ¹⁸O<sub>cel</sub> and δ²H<sub>cel</sub> to calculate Δδ¹⁸O<sub>cel(climate)</sub> for individual trees using various B values in Eq. (21). Thirdly, we combine the Δδ¹⁸O<sub>cel(climate)</sub> time series of all trees for each case of different B values. Fourthly, we select the best B value by comparison of the overall trends in the combined Δδ¹⁸O<sub>cel(climate)</sub> and average δ¹⁸O<sub>cel</sub> data. Finally, we reintegrate the long-periodicity variations in Δδ¹⁸O<sub>cel(climate)</sub> calculated using the best B value and short-periodicity variations in δ¹⁸O<sub>cel</sub> for all individual trees at an adequate threshold periodicity and combine all individual data again because the physiological factors do not appear to affect the short-periodicity δ¹⁸O<sub>cel</sub> variation so that we should keep the original δ¹⁸O<sub>cel</sub> variations in the high-frequency domain. In this procedure (Fig. 6a), the iterative calculation used to combine many tree-ring time series in the third step is the most time-consuming because various B values are tested.

Although A can be determined independently for each tree, we can only obtain one B value for all the trees. In a practical sense, it is not meaningful to determine A separately for each tree because all the trees were collected within central Japan (Fig. 1). If we assume that A and B are unique for all trees in this study, we can simplify the procedure as shown in Fig. 6b to reduce the time required for the iterative calculation. Considering that all the calculations used to combine and integrate the time series in Fig. 6 are linear, we can change the order of calculation between combination and integration. In fact, if we use common A and B values, the resultant combined time series for Δδ¹⁸O<sub>cel(climate)</sub> does not change when using the two procedures (Fig. 6a and b). In Fig. 6b, we first combine the δ¹⁸O<sub>cel</sub> and δ²H<sub>cel</sub> time series of all trees in Fig. 3 using the method shown in Fig. 5. Secondly, we determine A by investigating the relationship between short-periodicity variations of δ¹⁸O<sub>cel</sub> and δ²H<sub>cel</sub> in their combined time series. Thirdly, we integrate the combined δ¹⁸O<sub>cel</sub> and δ²H<sub>cel</sub> records to show temporal variations in

**Figure 5.** The iterative calculation method used to smoothly combine many individual tree-ring time series with large level offsets. (a) Pseudo-cross-dated tree-ring cellulose δ¹⁸O time series (Stage 1). (b) Averaging of all time series in Stage 1 (black dotted line) and offsetting individual time series in Stage 1 to the position where the average of the individual time series (colored dashed lines) becomes equal to the average of the black dotted line during the corresponding period (colored dotted lines) by retaining the original temporal patterns. (c) Result of offsetting of individual time series from Stage 1 (Stage 2). (d) Result of nine iterative calculations from Stage 1 (Stage 10). (e) Procedure from Stages 10 to 11. We obtain a unique combined time series when the dotted lines become equal to the dashed lines within a finite number of iterations.

Firstly, we simply average all the individual time series to make a preliminary combined time series. Secondly, we offset each individual time series up or down, retaining their original patterns of temporal variations, to the position where the temporal average of the individual time series becomes equal to that of the preliminary combined time series for the corresponding period of the individual tree. Thirdly, we average all the offset individual time series to make a refined combined time series. We iterate this procedure until the temporal average of the individual time series becomes equal to

\[\text{Pseudo-cross-dated tree-ring cellulose } \delta^{18}O \text{ time series} \]

\[\text{(Stage 1)} \]

\[\text{Averaging of all time series in Stage 1 (black dotted line) and offsetting individual time series in Stage 1 to the position where the average of the individual time series (colored dashed lines) becomes equal to the average of the black dotted line during the corresponding period (colored dotted lines) by retaining the original temporal patterns.} \]

\[\text{(Stage 2)} \]

\[\text{Result of offsetting of individual time series from Stage 1 (Stage 2).} \]

\[\text{Result of nine iterative calculations from Stage 1 (Stage 10).} \]

\[\text{Procedure from Stages 10 to 11. We obtain a unique combined time series when the dotted lines become equal to the dashed lines within a finite number of iterations.} \]

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We calculated the temporal variation in the climatological component ($\Delta \delta^{18}O_{cel(climate)}$) of tree-ring cellulose $\delta^{18}O$ (Fig. 10a) by using $A = 5$ and $B = 5.4$, as well as the smoothly combined $\delta^{18}O_{cel}$ and $\delta^{2}H_{cel}$ time series in Fig. 7 and Eq. (21). We used the temporal average of $(\delta^{2}H_{cel} + B\delta^{18}O_{cel})/(A+B)$ during the 30 years from 1961 to 1990 for $(\delta^{2}H_{cel(t)} + B\delta^{18}O_{cel(t)})/(A+B)$ in Eq. (21) so as to make $\Delta \delta^{18}O_{cel(climate)}$ the climatological variation in $\delta^{18}O_{cel}$ from the period between 1961 and 1990. The $\Delta \delta^{18}O_{cel(climate)}$ variation shown in Fig. 10a was influenced by the $\delta^{2}H_{cel}$ time series, not only for the long-periodicity variation, but also for the short-periodicity variation for which it is not necessary to integrate $\delta^{2}H_{cel}$. In fact, when we calculated EPS values for all individual $\Delta \delta^{18}O_{cel(climate)}$ time series from Eq. (21), we found that the EPS values are significantly lower (Fig. 10b), reflecting the low EPS value of the original $\delta^{2}H_{cel}$ time series (Fig. 3d). The purpose of introducing the $\delta^{2}H_{cel}$ signal into the $\delta^{18}O_{cel}$ time series is to remove the physiological effects from the long-periodicity variations in $\delta^{18}O_{cel}$. Therefore, at two threshold periodicities (21 and 51 years), we tentatively separated the long-periodicity component (21- and 51-year running means) from the integrated $\Delta \delta^{18}O_{cel(climate)}$ time series in Fig. 10a and short-periodicity component (deviations from 21- and 51-year running means) from the original combined $\delta^{18}O_{cel}$ in Fig. 7, and we reintegrated these into a new time series for $\Delta \delta^{18}O_{cel(climate)}$ to remove the influence of the $\delta^{2}H_{cel}$ data from the short-periodicity component. We also applied this reintegration procedure between the long-periodicity ($\Delta \delta^{18}O_{cel(climate)}$) and short-periodicity ($\delta^{18}O_{cel}$) components to the data for individual trees and calculated the EPS values for the reintegrated $\Delta \delta^{18}O_{cel(climate)}$ datasets (Fig. 10b). The resultant EPS values were $> 0.85$ and much higher than the original $\Delta \delta^{18}O_{cel(climate)}$ for almost all per-
Figure 7. Smoothly combined individual time series of $\delta^{18}$O$_{cel}$ and $\delta^2$H$_{cel}$ from Fig. 3a and c using the iterative calculation method shown in Fig. 5. The combined time series is after 1000 iterations (Stage 1000).

Figure 8. Relationships between short-periodicity components of $\delta^{18}$O$_{cel}$ and $\delta^2$H$_{cel}$ that are (a) $< 5$ years, (b) $< 11$ years, and (c) $< 21$ years, defined as the anomalies from the 5-, 11-, and 21-year running means of the combined $\delta^{18}$O$_{cel}$ and $\delta^2$H$_{cel}$ time series in Fig. 7, respectively.

ods during last 2500 years when either 21 or 51 years was used as the threshold.

However, this reintegration procedure may recover potential physiological effects with intermediate periodicities of less than 21 and 51 years. Hence, we compared the long-term variations (periodicity $> 11$ years) of the two reintegrated $\Delta \delta^{18}$O$_{cel(climate)}$ time series using 21 and 51 years as the thresholds and the original $\Delta \delta^{18}$O$_{cel(climate)}$ time series (Fig. 10a) in Fig. 10c to investigate whether there are significant differences. Each of the three time series shown in Fig. 10c almost coincide over all timescales, indicating there are no significant physiological effects with a periodicity between 11 and 51 years. However, we used 21 years as the threshold for the reintegration of the long- and short-periodicity components in order to robustly remove the influence of physiological effects. It does not result in a lower-quality reintegrated $\Delta \delta^{18}$O$_{cel(climate)}$ record, given the nearly equal EPS values using the 21- and 51-year thresholds in Fig. 10b. We utilized the reintegrated $\Delta \delta^{18}$O$_{cel(climate)}$ time series between the long-periodicity ($> 21$ years) domain of $\Delta \delta^{18}$O$_{cel(climate)}$ in Fig. 10a and short-periodicity ($< 21$ years) domain of $\delta^{18}$O$_{cel}$ in Fig. 7 as the final time series of the climatological component in the tree-ring cellulose $\delta^{18}$O data (Fig. 11). (You can see the effects of the integration and reintegration procedures more directly in Fig. S7.)

In contrast to the combined $\delta^2$H$_{cel}$ record (Fig. 7), the multi-centennial variation in the combined $\delta^{18}$O$_{cel}$ record (Fig. 7) does not appear to be very similar to that of the climatological component $\Delta \delta^{18}$O$_{cel(climate)}$ (Fig. 11). This is partly because there is an apparent age trend in the combined $\delta^{18}$O$_{cel}$ record, which overlaps the multi-millennial climatological decrease in Fig. 9. This means the multi-centennial variations are ambiguous in the combined $\delta^{18}$O$_{cel}$ record (Fig. 7). However, there may be an anthropogenic explanation for this, whereby in wetter and cooler periods, the number of trees in Japanese forests might have decreased due to logging for fuel. Rapid tree growth in the resultent more open and lighter forest would have increased $\delta^{18}$O$_{cel}$ and decreased $\delta^2$H$_{cel}$ values due to physiological effects (see Sect. S1). The wetter and cooler climate may have also lowered $\delta^{18}$O and $\delta^2$H in leaf water. The combined effects of climate variations and anthropogenic factors on Japanese forests might have reduced and enhanced the multi-centennial variations in $\delta^{18}$O$_{cel}$ and $\delta^2$H$_{cel}$, respectively (Fig. 7). However, we can robustly extract the climatological component $\Delta \delta^{18}$O$_{cel(climate)}$ independently of the local forest history by integrating $\delta^{18}$O$_{cel}$ and $\delta^2$H$_{cel}$ data. This is the most important paleoclimatological innovation of our study.
in Japan, corresponding to the baiu–meiyu front, which is an early summer stagnant rain belt characteristic of the East Asian summer monsoon (Fig. 13a). $\delta^{18}O_{\text{cel(climate)}}$ has a significant positive correlation with June–July mean temperature across a wide region of Japan, Korea, and China, suggesting that $\Delta \delta^{18}O_{\text{cel(climate)}}$ may be a good proxy for the East Asian summer monsoon (Fig. 13c).

The negative correlations of $\Delta \delta^{18}O_{\text{cel(climate)}}$ with relative humidity and precipitation can be explained by the direct negative relationship between $\Delta \delta^{18}O_{\text{cel(climate)}}$ and relative humidity in Eq. (11) as well as the amount effect, whereby $\delta^{18}O_{\text{rain}}$ becomes lower when precipitation increases (Dansgaard, 1964; Araguás-Araguás et al., 1998). The highest correlation ($< 0.6$) area of June–July precipitation is located just to the south of the sample sites (Fig. 13b). This is because, in the summer season, water vapor usually comes from the south (Pacific Ocean) to the sample sites and $\delta^{18}O_{\text{rain}}$ becomes lower when heavy rainfall occurs just before the arrival of the air mass carrying the water vapor. The positive correlation between $\Delta \delta^{18}O_{\text{cel(climate)}}$ and temperature must be caused by the meteorologically reverse relationship between summer precipitation and temperature in humid monsoonal Asia, including Japan. In fact, the center of the highest correlation area of June–July mean temperature is located slightly to the west of the sample site (Fig. 13c). As such, when the temperature in western Japan is high and characterized by high pressure, the wind blows from the north to the sample site, resulting in dry conditions and low rainfall.

Given the relationship between $\Delta \delta^{18}O_{\text{cel(climate)}}$ and modern meteorological observations evident in Figs. 12 and 13, we compared $\Delta \delta^{18}O_{\text{cel(climate)}}$ in central Japan with long-term historical and paleoclimatological records of summer climate before the 19th century. During the Edo era (1603–1868 CE), people wrote numerous diaries throughout Japan in which daily weather conditions were routinely described. Mizukoshi (1993) compiled many diary weather descriptions for central Japan and reconstructed interannual varia-

Figure 9. Linear regressions of $\Delta \delta^{18}O_{\text{cel(climate)}}$ calculated using 3 (orange line), 5 (red line), 7 (blue line), and 9 (green line) as $B$ values, with $A = 5$ in Eq. (21), and the simple averages of all tree-ring cellulose $\delta^{18}O$ data in Fig. 3a (gray line) with a linear regression (black dotted line).
disaster records during the summer season (June–August) to construct an index of the “flood disaster ratio”, which is the proportion of flood-related documents to total flood- and drought-related documents. Given that documents are scarcer in the older period, we calculated an 11-year running mean of the flood disaster ratio and compared it with $\Delta \delta^{18}O_{\text{cell(climate)}}$ (Fig. 15). In the 10th and 11th centuries during the Medieval Climate Anomaly, there were numerous droughts in Japan, corresponding to the highest values of $\Delta \delta^{18}O_{\text{cell(climate)}}$. After the 11th century, both the documentary records and $\Delta \delta^{18}O_{\text{cell(climate)}}$ values demonstrate that climate became progressively wetter towards the Edo era in the 17th century.

Although we could not find historical records of extreme climate events prior to the 10th century CE in Japan, Sakaguchi (1983, 1989) reconstructed long-term summer temperature variations using the pollen percentage of a cold-region pine ($Pinus pumila$) in the Ozegahara peatland of east Japan (Fig. 16). Although the pollen data after the 3rd century CE are not very reliable due to the lower sedimentation rate and human disturbance, the high sedimentation rate before the 2nd century CE enabled us to compare it with $\Delta \delta^{18}O_{\text{cell(climate)}}$. Both the pollen and $\Delta \delta^{18}O_{\text{cell(climate)}}$ records show similar variations from the 6th century BCE to 2nd century CE, indicating a warmer and drier climate from the 5th to 2nd century BCE and a cooler and wetter climate after the 1st century BCE (Fig. 16). After the 3rd century CE, both datasets show similar millennial variations, although the temporal resolution of the pollen data is not high.

The climatological component of the variations in tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{\text{cell(climate)}}$) correlates well with meteorological, historical, and vegetation data over various timescales in central Japan, and it also shows similar long-term patterns as paleoclimatological global temperature and East Asian precipitation data. Figure 17a shows that $\Delta \delta^{18}O_{\text{cell(climate)}}$ exhibits variations almost identical to air temperature reconstructions for land areas in the Northern Hemisphere (Mann et al., 2008), except for the period of the Medieval Climate Anomaly. Variations in summer precipitation reconstructed from diatom assemblages in lake sediments in Taiwan (Wang et al., 2013) are also similar to the variations in $\Delta \delta^{18}O_{\text{cell(climate)}}$ in central Japan (Fig. 17b). Two time series of carbonate $\delta^{18}O$ values in speleothems from northwest China (Zhang et al., 2008; Tan et al., 2010) also match the variations in $\Delta \delta^{18}O_{\text{cell(climate)}}$ (Fig. 17c). Note that the directions of the y axis are reversed between the speleothem and $\Delta \delta^{18}O_{\text{cell(climate)}}$ data, reflecting the meridional disparity of precipitation patterns in East Asia (Fig. 13a), as demonstrated by Liu et al. (2014) and Chen et al. (2015).

The climatological component of the variations in tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{\text{cell(climate)}}$) in central Japan (Fig. 11) corresponds well over various timescales with summer precipitation and temperature records in central Japan, which have been derived from various meteorological, his-

Figure 10. Calculation of the climatological component of the tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{\text{cell(climate)}}$). (a) Time series of $\Delta \delta^{18}O_{\text{cell(climate)}}$ calculated using Eq. (21), with $A = 5.0$ and $B = 5.4$, and the smoothly combined time series of $\delta^{18}O_{\text{cel}}$ and $\delta^{18}O_{\text{cell(climate)}}$ in Fig. 7. (b) EPS values for the individually calculated time series of $\Delta \delta^{18}O_{\text{cell(climate)}}$ (gray line) and the individually calculated and reintegrated time series between the long-periodicity component of $\Delta \delta^{18}O_{\text{cell(climate)}}$ and short-periodicity component of $\delta^{18}O_{\text{cel}}$, with 21 years (red line) and 51 years (blue line) as the thresholds between long and short periodicity. (c) 11-year running mean of the time series of $\Delta \delta^{18}O_{\text{cell(climate)}}$ in Fig. 10a (gray line) and those of the reintegrated time series between the long-periodicity component in $\Delta \delta^{18}O_{\text{cell(climate)}}$ in Fig. 10a and short-periodicity component in the combined $\delta^{18}O_{\text{cel}}$ in Fig. 7, with 21 years (red line) and 51 years (blue line) as the threshold between long and short periodicity. EPS values in panel (b) were calculated for a total period of 51 years, including 25 years before and after the year shown.
Figure 11. The final combined time series of the climatological component in variations of tree-ring cellulose $\delta^{18}O$ data ($\Delta\delta^{18}O_{\text{cell(climate)}}$). This is the reintegrated time series between the long-periodicity component in the original $\Delta\delta^{18}O_{\text{cell(climate)}}$ data in Fig. 10a and the short-periodicity component in the combined $\delta^{18}O_{\text{cell}}$ data in Fig. 7, with 21 years as the threshold between long and short periodicity.

Figure 12. (a–c) Average values of the monthly mean temperature (red), precipitation (blue), and mean relative humidity (green) from 1901 to 2005 CE. (d–f) Correlation coefficients of variations between $\Delta\delta^{18}O_{\text{cell(climate)}}$ in Fig. 11 and the monthly mean temperature (red), precipitation (blue), and mean relative humidity (green) from 1901 to 2005 CE. The meteorological data were observed at Iida (a, d), Nagoya (b, e), and Kyoto (c, f) (see Figs. 1 and 13b) and recorded in the database of the Japan Meteorological Agency. Asterisks in panels (d), (e), and (f) indicate statistical significance at the $>99\%$ level ($p < 0.01$).
**Figure 13.** Maps of the spatial correlation ($p < 10\%$) of $\Delta \delta^{18}O_{\text{cel(climate)}}$ in Fig. 11 with (a–b) June–July precipitation and (c) June–July mean temperature for 1901–2005 from CRU TS4.03, with sites of reference studies shown as an open square (Huangye Cave), diamond (Wanxiang Cave), triangle (Tsui-fong Lake), and circle (Ozegahara peatland). For this analysis, we used Climate Explorer (http://www.knmi.nl, last access: 21 January 2020) of the Royal Netherlands Meteorological Institute (KNMI) (van Oldenborgh and Burgers, 2005). Sample locations and numbers (Table S1) are shown as white circles (large 10–15; medium 4–9; small 1–3) in panel (b).

**Figure 14.** Comparisons of the climatological component of the tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{\text{cel(climate)}}$) record in Fig. 11 (black line) with diary-based annual reconstructions (orange line) and meteorological observations (blue line) of early summer precipitation at Osaka (Mizukoshi, 1993). Note that the $y$ axis of $\Delta \delta^{18}O_{\text{cel(climate)}}$ is inverted to indicate higher precipitation toward the top of the figure.
Figure 15. Comparisons of the climatological component of the tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{cel(climate)}$) record in Fig. 11 (gray line: annual; black line: 11-year running mean) with the proportion of flood-related documents (11-year running mean) of total flood- and drought-related documents (11-year running mean) during the summer season (JJA) in central Japan (mostly in and around Kyoto) (Fujiki, 2007). Note that the $y$ axis of $\Delta \delta^{18}O_{cel(climate)}$ is inverted to indicate higher precipitation toward the top of the figure.

Figure 16. Comparisons of the climatological component of the tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{cel(climate)}$) record in Fig. 11 (gray line: annual; black line: 11-year running mean) with the pollen percentage of a cold-region pine ($P. pumila$) in the Ozegahara peatland (Sakaguchi, 1983, 1989). Location of the pollen record is shown in Fig. 13. Given that $P. pumila$ becomes dominant during cold periods, the $y$ axis for the pollen percentage is inverted to indicate warmer conditions toward the top of the figure.

torical, and paleo-vegetation archives (Figs. 12–16). This indicates that $\Delta \delta^{18}O_{cel(climate)}$ is a reliable proxy for summer climate, such as the activity of the East Asian summer monsoon. Multi-centennial and millennial $\Delta \delta^{18}O_{cel(climate)}$ variations are similar to those of paleoclimatological reconstructions of global temperatures and East Asian precipitation (Fig. 17), indicating a drier climate during the Medieval Climate Anomaly and wetter climate during the Little Ice Age in central Japan. As such, $\Delta \delta^{18}O_{cel(climate)}$ can be utilized in climatological, historical, and archeological studies.

4 Conclusions

We constructed a statistically reliable multi-millennial tree-ring dataset of cellulose $\delta^{18}O$ in central Japan by analyzing tree rings of 67 trees without using a pooling method. We found that there are distinct age trends in the $\delta^{18}O$ time series. By comparison with the $\delta^2H$ time series, we showed that the age trend in $\delta^{18}O$ is caused by an increase in the degree of post-photosynthesis isotopic exchange with xylem water before cellulose synthesis as the trees mature. Because the physiological conditions of the post-photosynthesis isotopic exchange are not simply controlled by tree age, but also related to the tree growth environment randomly influenced by human activity, it was not possible to remove the age trend by application of the negative exponential curve or RCS.

Given that tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ are correlated positively and negatively due to climatological and physiological factors, respectively, we formulated simultaneous equations for the climatological and physiological components of the tree-ring cellulose $\delta^{18}O$ and $\delta^2H$ data. We solved these equations to cancel out the physiological effects and established a multi-millennial record of the climatological component of tree-ring cellulose $\delta^{18}O$ ($\Delta \delta^{18}O_{cel(climate)}$). The $\Delta \delta^{18}O_{cel(climate)}$ time series is well correlated with local, regional, and global variations in summer climate reconstructed by instrumental, historical, and paleoclimatological methods. This suggests that $\Delta \delta^{18}O_{cel(climate)}$ has recorded summer climate variations in central Japan during the past 2600 years on annual to millennial timescales.

However, further research is needed to make $\Delta \delta^{18}O_{cel(climate)}$ a more reliable index of summer climate. In this study we utilized $\delta^2H_{cel}$ as supplementary data just to remove the long-term physiological effects...
Figure 17. Comparison of centennial- to millennial-scale variations of the climatological component of tree-ring cellulose $\delta^{18}$O ($\Delta\delta^{18}$O$_{\text{cell(climate)}}$) in central Japan with global and East Asian climate reconstructions. The time series of 11-year running means for Fig. 11 (black line) with (a) air temperature over the Northern Hemisphere defined as NH EIV-land (Mann et al., 2008) (red line), (b) precipitation inferred from a principal component analysis (PC1) of diatom assemblages from Tsuifong Lake in southeastern China (Taiwan) (Wang et al., 2013) (orange line), and (c) precipitation inferred from carbonate $\delta^{18}$O values of speleothems from Wanxiang (pink line) and Huangye (blue line) caves in northwestern China (Zhang et al., 2008; Tan et al., 2010). Note that the y axes are reversed between the speleothem data and $\Delta\delta^{18}$O$_{\text{cell(climate)}}$ in panel (c). Locations of the records in panels (b) and (c) are shown in Fig. 13.
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Author contributions. TN designed the research and wrote the paper; TN, MaS, KS, TM, MiS, HO, NH, NN, and MY collected tree-ring samples; TN, MaS, KS, ZL, CX, and KO extracted cellulose from tree rings; TN, MaS, ZL, CX, KO, and AT measured isotope ratios of tree-ring cellulose; TN, MaS, and YS analyzed isotopic data mathematically; all authors discussed the results and provided input to the paper.

Competing interests. The authors declare that they have no conflict of interest.

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