

## Article

# Climate Response of Oxygen Isotopic Compositions in Tree-Ring Cellulose in Java: Evaluation Using a Proxy System Model

Ryo Hisamochi <sup>1</sup>, Yumiko Watanabe <sup>1,\*</sup>, Naoyuki Kurita <sup>2</sup> and Takahiro Tagami <sup>1</sup>

<sup>1</sup> Division of Earth and Planetary Sciences, Graduate School of Science, Kyoto University, Kyoto 606-8501, Japan; ryohisamochi@gmail.com (R.H.); tagami@kueps.kyoto-u.ac.jp (T.T.)

<sup>2</sup> Institute for Space-Earth Environmental Research, Nagoya University, Nagoya 464-8601, Japan; nkurita@nagoya-u.jp

\* Correspondence: watanabe.yumiko.4e@kyoto-u.ac.jp

**Abstract:** Tree-ring cellulose oxygen isotopic composition ( $\delta^{18}\text{O}$ ) is controlled by several hydrological factors such as precipitation, relative humidity, and temperature. A proxy system model can reveal how these factors affect tree-ring cellulose  $\delta^{18}\text{O}$ . In this study, to identify a key control on tree-ring cellulose  $\delta^{18}\text{O}$  variations, we performed model calculation of year-to-year variation of tree-ring cellulose  $\delta^{18}\text{O}$  of Javanese teak in Indonesia from 1960 to 1998. Our model results reasonably reproduce the observed  $\delta^{18}\text{O}$  values and their temporal variations ( $r = 0.6$ ;  $p < 0.001$ ). Moreover, the sensitivity test shows that the cellulose  $\delta^{18}\text{O}$  values are sensitive to the teak growing period. The simulation result with earlier or later shifts of the growing period captured the amplitude of observed  $\delta^{18}\text{O}$  variations over 39 years. These results indicate that the tree-ring cellulose  $\delta^{18}\text{O}$  of Javanese teak might be influenced by a subtle shift of the intra-annual growing period.

**Keywords:** isotope; tree ring; cellulose; proxy system model; Indonesia



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## 1. Introduction

Java Island of Indonesia, a densely populated region, often experiences droughts and floods due to strongly varying intensity of tropical monsoon precipitation. Because instrumental meteorological data are temporally and spatially limited in this region, a long-term and high-resolution hydroclimatic proxy is required to elucidate its mid-term to long-term monsoon variation.

Tree rings are useful to reconstruct terrestrial paleoclimates because of their precise dating and high time resolution. The tropics have been the subject of comparatively few dendroclimatological studies because collecting tree samples with annual rings is difficult. In Indonesia, dendroclimatological studies have been conducted using tree-ring widths of teak (*Tectona grandis*) on Java Island [1–4]. Moreover, a few reports have described oxygen isotopic composition ( $\delta^{18}\text{O}$ ) of tree-ring cellulose in Java [4–6]. According to Schollean et al. [4], cellulose  $\delta^{18}\text{O}$  of teak collected from Java shows positive correlation with precipitation during the dry season before the growing season and shows negative correlation with precipitation during the rainy season. Moreover, Hisamochi et al. [6] measured cellulose  $\delta^{18}\text{O}$  of ten teak disks collected at four sites in Java, and verified the synchronicity of the annual time-series variations. These results of earlier studies suggest that tree-ring cellulose  $\delta^{18}\text{O}$  is a reliable proxy for hydroclimate (i.e., precipitation and relative humidity) over Java Island.

Because tree-ring cellulose  $\delta^{18}\text{O}$  is influenced by several hydrological factors such as precipitation, relative humidity, and temperature, it is generally difficult to estimate each element. A forward model of the tree-ring cellulose  $\delta^{18}\text{O}$ , a so-called a proxy system model, has already been established and can predict isotopic compositions precisely by giving

the surrounding meteorological data [7,8]. Proxy system models are able to address the issues how various meteorological conditions affect tree-ring cellulose  $\delta^{18}\text{O}$ , and help to elucidate tree-ring cellulose  $\delta^{18}\text{O}$  as paleoclimate proxy. In this study, we performed model calculations using the proxy system model modified by Kurita et al. [8], in order to identify the key hydroclimate factor control on  $\delta^{18}\text{O}$  variability of tree-ring cellulose of Javanese teak in Indonesia. In this study, we also presented the calibration of a proxy system model and the detail process how teak  $\delta^{18}\text{O}$  records the environmental conditions.

## 2. Experiments

### 2.1. Study Site

Here, we study the 39 y long temporal variability of teak tree-ring cellulose  $\delta^{18}\text{O}$  from four distant sites on Java, Indonesia (1960–1998). Java has a rainy season lasting from November–April and a dry season from May–October because of the seasonal movement of the Inter Tropical Convergence Movement (ITCZ). Relative humidity cycles fluctuate between high levels during the rainy season and low levels during the dry season, whereas air temperature is almost constant throughout the year. Rainfall  $\delta^{18}\text{O}$  has a seasonal cycle, alternating from lower values in the rainy season to higher in the dry season, according to rainout process at regional scale [9].

### 2.2. Tree-Ring Cellulose Oxygen Isotope Data

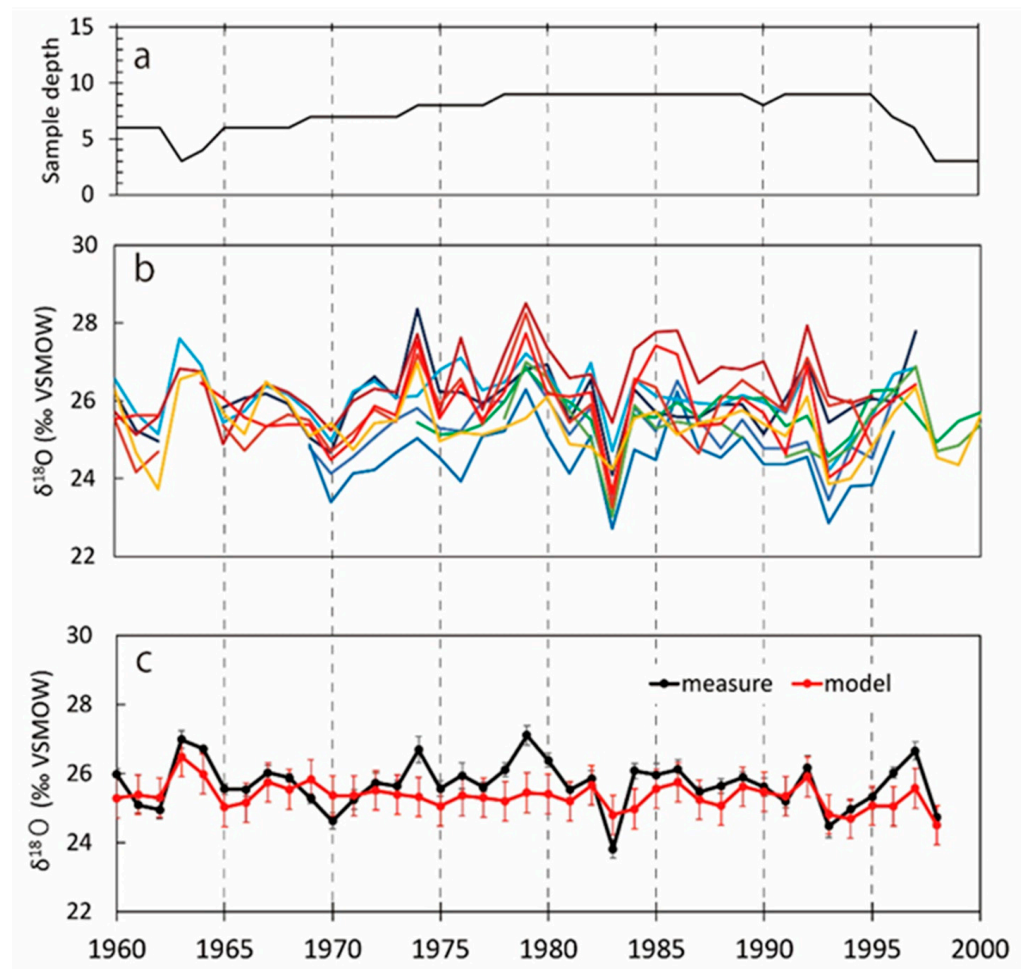
To validate the model performance, we use the observed tree-ring cellulose  $\delta^{18}\text{O}$  in Java [6]. Figure 1b shows the time series of  $\delta^{18}\text{O}$  of ten teak trees collected from four plantations on Java Island. The expressed population signal (EPS) was 0.93 during 1960–1998. The cellulose  $\delta^{18}\text{O}$  time series of teak has high spatial coherence over Java [6]. For this study, we referred to the average tree-ring cellulose  $\delta^{18}\text{O}$  of ten individual teak trees as measured values (Figure 1c). As shown in our previous study [6], tree-ring cellulose  $\delta^{18}\text{O}$  of Javanese teak is negatively correlated with precipitation and relative humidity during the rainy season. In addition, it is positively correlated with precipitation during the dry season preceding the growing season.

### 2.3. Proxy System Model of Tree-Ring Cellulose Oxygen Isotopic Compositions

The tree-ring cellulose oxygen isotope model used for this study is based on the model reported by Kurita et al. [8], as modified from Roden et al. [7]. It is a mechanistic model used to quantify both physical and biochemical fractionation associated with tree-ring cellulose  $\delta^{18}\text{O}$ , including the uptake of soil water (source water), transpiration and photosynthesis in leaves, and cellulose synthesis. The uptake of soil water at roots occurs with no isotope fractionation and the water reaches leaves through a conduit [10,11]. Thereafter, the following two processes are important in the model: (1) isotopic fractionation accompanying the transpiration of leaf water and (2) isotope fractionation transforming from leaf water to cellulose. Craig and Gordon [12] modeled the fractionation processes during the evaporation of water. The model was applied to the transpiration of leaf water [13]. Leaf water  $\delta^{18}\text{O}$  is given as [8,14]

$$\delta^{18}\text{O}_{leaf} = \delta^{18}\text{O}_{source} + \epsilon_{eq} + \epsilon_k + \left( \delta^{18}\text{O}_{atm} - \delta^{18}\text{O}_{source} - \epsilon_k \right) \left( \frac{e_a}{e_i} \right) \quad (1)$$

where  $\delta^{18}\text{O}_{leaf}$ ,  $\delta^{18}\text{O}_{source}$  and  $\delta^{18}\text{O}_{atm}$  represent the isotopic compositions of leaf water, source water, and atmospheric vapor,  $\epsilon_{eq}$  stands for the equilibrium isotope fractionation factor between water and vapor,  $\epsilon_k$  denotes the kinetic isotope fractionation factor associated with vapor diffusion into unsaturated air, and  $e_a/e_i$  expresses the ratio of the vapor partial pressure in the atmosphere to that in leaf. In addition,  $\epsilon_{eq}$  is calculated as a function of the leaf temperature ( $T_{leaf}$ ) as shown below [15].



**Figure 1.** Tree-ring cellulose oxygen isotopic compositions of Javanese teak. (a) The number of studied tree individuals. (b) Measured time series data of ten Javanese teak trees [6]. (c) Tree-ring cellulose  $\delta^{18}\text{O}$  time series calculated by the proxy system model and mean tree-ring isotope chronology. The error of measurement data is presented as standard error. The error of model calculation is described in detail in Section 2.4.

$$\epsilon_{eq} = \left[ \exp\left(1137/T_{leaf}^2 - 0.4156/T_{leaf} - 0.002067\right) - 1 \right] * 1000 \quad (2)$$

Moreover, leaf temperature is also calculated as [16]

$$T_{leaf} = \frac{22}{33} * T_{air} + 10 \text{ (}^\circ\text{C)} \quad (3)$$

where  $T_{air}$  denotes the air temperature. In addition,  $\epsilon_k$  is calculated using the following equation [17].

$$\epsilon_k = \frac{32r_s + 21r_b}{r_s + r_b} \quad (4)$$

Therein,  $r_s$  and  $r_b$ , respectively, represent the resistances of stomata and boundary layer. That is,  $r_s$  is the inverse of stomata conductance ( $S_c$ ); also,  $r_b$  is the inverse of boundary layer conductance ( $B_c$ ). In this study,  $S_c$  and  $B_c$  are based on work by Managave et al. [18], who studied Indian teak using a tree-ring oxygen isotope model:  $S_c = -0.67 * VPD + 1.63$ ,  $B_c = 1$ , where VPD represents the vapor pressure deficit calculated from the temperature and relative humidity [18].

The  $\delta^{18}\text{O}_{leaf}$  modelled using Equation (1) is known to often overestimate the actual value because of the Péclet effect which considers the backward diffusion of enriched water

into the leaf water at the transpiration site [8,19]. The oxygen isotopic ratios after Péclet effect correction ( $\delta^{18}O_{leaf\ corr}$ ) are given as

$$\delta^{18}O_{leaf\ corr} = \delta^{18}O_{leaf} * \alpha + \delta^{18}O_{source} * (1 - \alpha) \tag{5}$$

where  $\alpha$  is the correction term of the Péclet effect, expressed by the following equation.

$$\alpha = \frac{1 - \exp(-\phi)}{\phi} \tag{6}$$

$$\phi = \frac{L * E}{C * D} \tag{7}$$

Therein,  $L$ ,  $E$ ,  $C$ , and  $D$ , respectively, represent the effective path lengths for water transport from the veins to the site of transpiration, the leaf transpiration rate ( $\text{mol}/\text{m}^2/\text{s}$ ;  $E = (Sc * VPD)/P$ , where  $P$  denotes pressure), the molar density of water ( $55.5 \times 10^3 \text{ mol}/\text{m}^3$ ), and the  $\text{H}_2^{18}\text{O}$  diffusion coefficient ( $D = 119 * \exp(-\frac{637}{T}) * 10^{-9}$ ) [20]. The effective path length depends on the tree species; the value for teak is unknown. We therefore assumed the value of a sunflower (0.01061 m) [21] because of the leaf shape similarity, comparing teak with a sunflower. Substituting Equation (1) into Equation (5),  $\delta^{18}O_{leaf\ corr}$  is given as presented below.

$$\delta^{18}O_{leaf\ corr} = \delta^{18}O_{source} + \alpha * \left[ \epsilon_{eq} + \epsilon_k + \left( \delta^{18}O_{atm} - \delta^{18}O_{source} - \epsilon_k \right) \left( \frac{e_a}{e_i} \right) \right] \tag{8}$$

Actually,  $\delta^{18}O_{leaf\ corr}$  is incorporated in sucrose because of photosynthesis in the leaf; then the sucrose is converted into cellulose. During cellulose synthesis, some oxygen atoms of sucrose are replaced by those of xylem water (source water) [17]. Accordingly, the oxygen isotopic ratios of cellulose ( $\delta^{18}O_{cell}$ ) is given as

$$\delta^{18}O_{cell} = f * \left( \delta^{18}O_{source} + \epsilon \right) + (1 - f) * \left( \delta^{18}O_{leaf\ corr} + \epsilon \right) \tag{9}$$

where  $f$  represents the proportion of oxygen atoms exchanged with xylem water,  $\epsilon$  denotes the biochemical fractionation associated with cellulose synthesis. Substituting Equation (8) into Equation (9), tree-ring cellulose isotopic compositions are calculated as shown below.

$$\delta^{18}O_{cell} = \delta^{18}O_{source} + (1 - f) * \alpha \left[ \epsilon_{eq} + \epsilon_k + \left( \delta^{18}O_{atm} - \delta^{18}O_{source} - \epsilon_k \right) \left( \frac{e_a}{e_i} \right) \right] + \epsilon \tag{10}$$

For this study, tree-ring cellulose  $\delta^{18}\text{O}$  of teak was calculated using Equation (10), assuming 0.42 for  $f$  [7] and 27 for  $\epsilon$  [22]. Table 1 presents constants used in this model calculation.

**Table 1.** Constants used in model calculations.

Constant	Description	Value	Reference
$\epsilon$	Biochemical fractionation of cellulose synthesis (‰)	27	[22]
$f$	Oxygen exchange fraction of cellulose synthesis	0.42	[7]
$L$	Effective path length (m)	0.01061	[21]
$C$	Molar density of water ( $\text{mol}/\text{m}^3$ )	$55.5 \times 10^3$	[20]
$B_c$	Boundary layer conductance	1	[18]

#### 2.4. Input Data to the Proxy System Model

Model calculation of cellulose  $\delta^{18}\text{O}$  using Equation (10) requires source water  $\delta^{18}\text{O}$  and atmospheric vapor  $\delta^{18}\text{O}$ , in addition to meteorological data: temperature, precipitation,

air pressure and relative humidity. For meteorological data, we used 6 hourly resolved climate parameters with the resolution of 1.25-degree grid on Japanese 55 y reanalysis (JRA-55), which is the global meteorological product of a state-of-art data assimilation system fed with observations from weather stations around the world [23]. In this study, we picked up the data from the nearest grid point and calculated monthly mean values only using daily time data (00 UTC, 06UTC, 12UTC) with twice the weight of 06 UTC (i.e.,  $00\text{UTC}/2 + 06\text{UTC} + 12\text{UTC}/2$ ) considering photosynthesis during the daytime. Moreover, soil water  $\delta^{18}\text{O}$  as source water was substituted to rainfall  $\delta^{18}\text{O}$ , assuming that they are approximately equal. Data of rainfall  $\delta^{18}\text{O}$  were obtained from Twentieth Century Reanalysis data (V2) by IsoGSM [24,25]. A comparison with observed  $\delta^{18}\text{O}$  in Jakarta (Global Network of Isotopes in Precipitation; GNIP) shows positive  $\delta^{18}\text{O}$  bias for IsoGSM, although temporal variability is well simulated. Thus, rainfall  $\delta^{18}\text{O}$  of IsoGSM was corrected by using the least square method to fit observed value. For atmospheric vapor  $\delta^{18}\text{O}$ , we used  $\delta^{18}\text{O}$  values in equilibrium with rainfall  $\delta^{18}\text{O}$  (i.e.,  $\delta^{18}\text{O}_{\text{atm}} = \delta^{18}\text{O}_{\text{rainfall}} - \epsilon_e$ ).

Input duration of meteorological data was determined based on a linear correlation analysis between cellulose  $\delta^{18}\text{O}$  and the monthly means of climate parameters. Figure 4 of Hisamochi et al. [6] shows the correlation coefficient for temperature, relative humidity, precipitation and PDSI during 1961–1995. The relative humidity is a main factor controlling the enrichment of heavy isotopes in leaf water as with cellulose consequently. The isotopic enrichment is caused by fractionation during the transpiration of leaf water as relative humidity decreases. Accordingly, we expect to find negative correlations between cellulose  $\delta^{18}\text{O}$  and relative humidity. As shown in Figure 4 of Hisamochi et al. [6], cellulose  $\delta^{18}\text{O}$  has the highest negative correlation with relative humidity in February, suggesting that wood cellulose may largely reflect the environmental conditions in February rather than those throughout the entire rainy season. We therefore input the meteorological data (i.e., temperature, air pressure and relative humidity) and atmospheric vapor  $\delta^{18}\text{O}$  in February to the proxy system model. In addition, we input rainfall  $\delta^{18}\text{O}$  during April–January as a substitute of source water  $\delta^{18}\text{O}$  because rain that fell during several months prior to the growing period (February) remains in the soil.

The error of the model calculated values was estimated by considering the spatial variations of Java Island on relative humidity, atmospheric vapor  $\delta^{18}\text{O}$ , and rainfall  $\delta^{18}\text{O}$ . Although other parameters (i.e., temperature and air pressure) also have variations, little effect on the calculation results can be observed. Constants such as  $\epsilon$  and  $f$  were excluded from error estimation in this study because they are partly related to systematic errors. Atmospheric vapor  $\delta^{18}\text{O}$  was calculated by substituting rainfall  $\delta^{18}\text{O}$  data, as described above. Accordingly, we verified the spatial variation of rainfall  $\delta^{18}\text{O}$  over Java to estimate the variation of atmospheric vapor  $\delta^{18}\text{O}$ . Based on monthly rainfall  $\delta^{18}\text{O}$  data of ten observation stations in Java and Bali (the data provided from Dr. K. Ichiyanagi, Kumamoto University, personal communication), the standard error among the stations was 0.76‰. For relative humidity, the error was estimated as 0.85%, considering the standard deviation of JRA-55 land surface 4-grid data in Java. The average error of the model calculation value obtained from the error propagation formula was approximately 0.57‰.

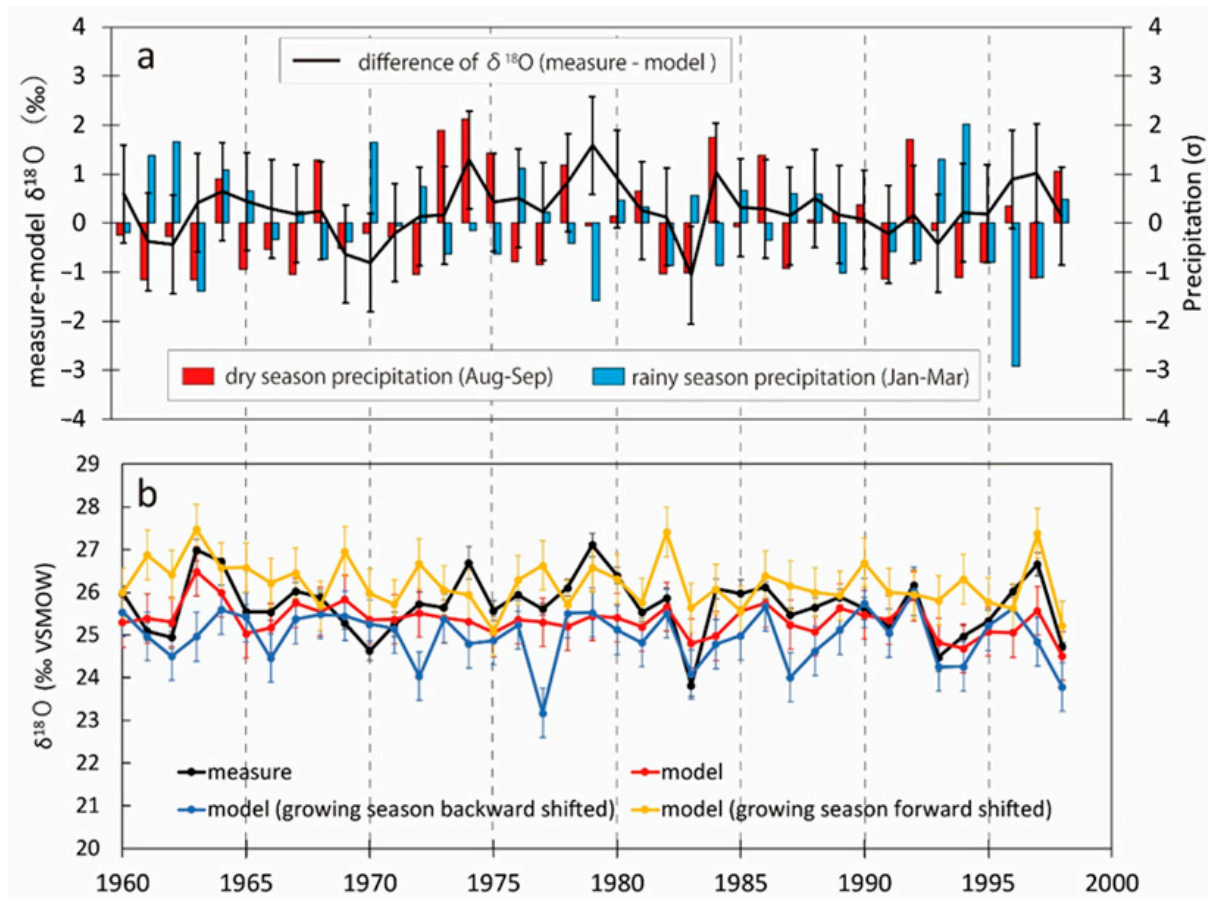
### 3. Results and Discussion

Figure 1c presents the 39 y long temporal variations (1960–1998) of both simulated and observed  $\delta^{18}\text{O}$  of tree-ring cellulose. The absolute values of model calculation almost correspond to the measured values, except for a few characteristic peaks (i.e., 1974, 1979, and 1983). Moreover, interannual variations between model calculation and measurement showed a significant positive correlation, with a correlation coefficient of 0.61 ( $p < 0.001$ ;  $n = 39$ ). These results demonstrate that the model reasonably simulates the observed isotopic values of teak cellulose.

Figure 2a is a time series of the difference between the measured and model calculated values of tree-ring cellulose  $\delta^{18}\text{O}$  and normalized anomalies of precipitation during the dry and rainy seasons, respectively. In years when tree-ring  $\delta^{18}\text{O}$  was underestimated by



the model (i.e., 1974, 1979), more precipitation occurred during the dry season and/or less precipitation occurred during the rainy season. However, in the year when tree-ring  $\delta^{18}\text{O}$  was overestimated by the model (i.e., 1983), less precipitation fell during the dry season and more precipitation occurred during the rainy season.



**Figure 2.** (a) Difference of tree-ring cellulose  $\delta^{18}\text{O}$  between the measured and the model calculated values (black line) and normalized anomalies of precipitation (red bars, August–September of the dry season; blue bars, January–March of the rainy season). (b) Tree-ring cellulose  $\delta^{18}\text{O}$  time series. The black line shows the measured values and the red line represents the modelled values (as shown in Figure 1c). The blue line presents the model calculated values for the growing season shifted backward. The yellow line presents the model calculated values for the growing season shifted forward.

Following the results presented above, we assumed that the years under/overestimated by the model occur because the teak growing period was shifted earlier/later because of the change of seasonal rainfall variation. In fact, the onset of the rainy season in Java is known to show high interannual shifts of a few months. For example, the rainy season onset comes later (earlier) during El Niño (La Niña) years than average [26]. Consequently, it might be true that the growth period of teak in Java is shifted slightly year-by-year. However, Javanese teak  $\delta^{18}\text{O}$  exhibits seasonality, alternating from a peak in the beginning of growth (i.e., rainy season) to nadir level [4,5]. Teak  $\delta^{18}\text{O}$  at the beginning of growth has a maximum, because rainfall  $\delta^{18}\text{O}$  is still heavy, with low relative humidity in the early rainy season [4]. As the rainy season progresses, rainfall  $\delta^{18}\text{O}$  becomes low and relative humidity becomes high, indicating that teak  $\delta^{18}\text{O}$  values become progressively lower [4]. In other words, if there is more precipitation at the end of the dry season or at the beginning of the rainy season, then tree rings are formed earlier and have higher cellulose  $\delta^{18}\text{O}$ . Conversely, if there is less precipitation during the dry season and more precipitation in the rainy season, tree-rings have lower cellulose  $\delta^{18}\text{O}$ . Accordingly, we examined model calculations in two cases, such as the teak growth period shifted forward or backward, as

described in detail in the next paragraph, whereas we originally input rainfall  $\delta^{18}\text{O}$  data during April–January (as source water  $\delta^{18}\text{O}$ ) and meteorological data in February.

Figure 2b presents calculation results with modified input data according to the teak growing season duration. Assuming that the growth period was shifted forward, we input rainfall  $\delta^{18}\text{O}$  data of April–October and meteorological data of November–December. In addition, assuming that the growth period was shifted backward, we input rainfall  $\delta^{18}\text{O}$  data during December–March and meteorological data in March. Specifically examining the years underestimated by the model (i.e., 1974, 1979), the calculated values with growing duration shifted forward (yellow curve in Figure 2b) correspond to measured values within the error bands. Moreover, for the year overestimated by the model (i.e., 1983), the calculated value with growing duration shifted backward (blue curve in Figure 2b) matches with the measured value, within the expected range of error. Therefore, the model can be modified to reproduce actual values of tree-ring cellulose  $\delta^{18}\text{O}$ , strictly considering the shift of the growing period attributable to the change of seasonal rainfall variation. Although we simply examined two cases of earlier/later growing periods in this study, more exact estimation of the growing period requires further investigations based on the phenology of teak, surrounding hydrological monitoring and cellulose intra-annually resolved  $\delta^{18}\text{O}$  measurements of subdivided tree rings.

According to previous studies on Javanese teak [4,6], tree-ring cellulose  $\delta^{18}\text{O}$  has a positive correlation with precipitation during the dry season. Schollaen et al. [4] explained that the positive correlation is attributable to the uptake of  $^{18}\text{O}$ -enriched soil water, in conjunction with enhanced  $^{18}\text{O}$ -enrichment of leaf water (driven by transpiration) at the beginning of the growing season. Thus, we conducted model calculations using rainfall  $\delta^{18}\text{O}$  data during April–January as source water  $\delta^{18}\text{O}$ . The calculated results of tree-ring cellulose  $\delta^{18}\text{O}$  coincided with the measured values (Figure 1c), supporting the speculation of Schollaen et al. [4]. That is, more precipitation during the dry season promotes teak growth earlier and engenders higher tree-ring cellulose  $\delta^{18}\text{O}$ , because rainfall  $\delta^{18}\text{O}$  is higher under lower relative humidity at the end of the dry season. Future work must analyze intra-annual variations of tree-ring cellulose  $\delta^{18}\text{O}$  of teak, as in previous studies in other regions of Asia [27,28], in order to provide new insights into the construction of a more reliable proxy system model for assessing teak tree-ring cellulose  $\delta^{18}\text{O}$ .

#### 4. Conclusions

In this study, we evaluated the tree-ring cellulose  $\delta^{18}\text{O}$  of Javanese teak as a hydro-climate proxy by using a proxy system model. Based on the results presented above, the following conclusions can be drawn:

1. Simulated values of cellulose  $\delta^{18}\text{O}$  corresponded to the measured values (except for a few peaks) with a correlation coefficient of  $r = 0.61$ , demonstrating that the model of this study can reproduce the actual isotopic compositions of teak cellulose.
2. Characteristic seasonal rainfall patterns were recognized in years under/overestimated by the model, and thus the growth duration could slightly shift according to the seasonal rainfall pattern.
3. Model calculations assuming that the growth duration is shifted earlier or later than usual matched with the measured peaks within the expected range of error, indicating that tree-ring cellulose  $\delta^{18}\text{O}$  is also influenced by subtle shifts of growth duration because of the change of seasonal rainfall pattern.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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