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EXPRESS LETTER

A 180-year-long isotopic record of tree-ring cellulose on Okinawa Island, Japan

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The oxygen isotope ratio ($\delta^{18}O$) of tree-ring cellulose provides a valuable climatic record with an annual timescale. In Japan, the cellulose $\delta^{18}O$ records have been accumulated over the past decade. However, no long-term records have been reported for the southern subtropical island, Okinawa-jima, where many long-lived trees were burned during the Second World War and have been periodically damaged by typhoons. Here, we show a cellulose $\delta^{18}O$ of Ryukyu Pine (*Pinus luchuensis* Mayr.) from the Okinawa-jima island. This $\delta^{18}O$ data, which was obtained from a tree-disk preserved in a University Museum, cover a 95-year period before meteorological observation began on this island in 1891. The cellulose $\delta^{18}O$ variations negatively correlate with the amount of precipitation during the rainy season, suggesting that the cellulose $\delta^{18}O$ values are largely affected by $\delta^{18}O$ of rainwater rather than large scale ENSO variations.

Keywords: tree-ring, oxygen isotope, monsoon, paleo climate, Okinawa

INTRODUCTION

To understand the mechanisms behind natural climate change, it is essential to obtain long-term climate data before instrumental observations started. Tree-ring width has been measured in many regions, revealing climate records in highly populated areas over the past millennia (PAGES2k Consortium, 2017). In addition, the oxygen isotope ratio ($\delta^{18}O$) in the cellulose of tree-rings has proved to be a reliable proxy to estimate regional climate changes, such as precipitation levels and atmosphere-ocean circulation patterns (e.g., Buhay and Edwards, 1995; Burk and Stuiver, 1981).

Based on physical and biochemical mechanisms, the $\delta^{18}O$ of tree-ring is controlled by both the $\delta^{18}O$ of source water and relative humidity through the isotopic fractionations during formation of tree-ring cellulose (Roden *et al*., 2000). Since the $\delta^{18}O$ of source water is influenced by several climate elements, factors controlling the cellulose $\delta^{18}O$ are evaluated using response function analysis. For example, a tree-ring $\delta^{18}O$ chronology from the Nepal Himalaya is primarily controlled by the amount of precipitation and relative humidity during the monsoon season (June–September) (Sano *et al*., 2012a). In northern Japan, a cellulose $\delta^{18}O$ record in a conifer-hardwood mixed forest is governed by the amount of winter precipitation and relative humidity in the summer (Nakatsuka *et al*., 2004). In central Japan, cellulose $\delta^{18}O$ values are governed by rainy season and summer precipitation (Li *et al*., 2015; Sakashita *et al*., 2016). At lower latitudes in Asia, large-scale climate patterns influence the $\delta^{18}O$ of tree-ring. For example, a 300-year-long $\delta^{18}O$ chronology from northern Vietnam shows statistically significant correlation with temperature, precipitation, and the El Niño-Southern Oscillation (ENSO) index (Sano *et al*., 2012b). Recently, an 818-year-long record from Taiwan also revealed the influence of the ENSO index (Liu *et al*., 2017).

Although cellulose $\delta^{18}O$ chronologies in Japan have been collected over the past decade (e.g., Nakatsuka *et al*., 2004; Sano *et al*., under revision), no record has been reported in the southernmost maritime island, Okinawa-jima (hereafter called Okinawa Island; "jima" means island in Japanese). On Okinawa Island, many long-lived trees were burned during the Second World War and have been occasionally damaged by typhoons. In addition, tree-ring analysis in subtropical low-altitude areas involves difficulties in identifying annual boundaries between tree rings. In this study, we analyzed the cellulose $\delta^{18}O$ of Ryukyu pine (*Pinus luchuensis* Mayr.) to reconstruct past

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climate variations on Okinawa Island. We screened out the four $\delta^{18}O$ data from this region by comparing with the $\delta^{18}O$ chronologies from neighboring regions to test the reliability of ambiguous tree-ring identification. We found a large tree-disk preserved in a University museum (Fujukan, University of the Ryukyus) shows significant correlations with the neighboring regions. We then evaluated the climatic factors influencing the $\delta^{18}O$ record by response function analyses.

Methods

Tree-ring sample

Tree-ring samples were obtained in Okinawa Island, Okinawa prefecture, Japan (~26°N, ~128°E; Fig. 1). Two samples (named FJY and SIN) were collected from the disk-shaped samples of Ryukyu pine tree (*Pinus luchuensis*). The largest tree-disk sample (FJY; major axis of 1.8 m and minor axis of 1.7 m, number of tree-ring of 183) was preserved in a museum at the University of the Ryukyus. It was taken from a tree that had been in Okinawa city, middle of Okinawa Island, Japan (*ca*. 50 m in elevation) and was cut down in 1980 because of insect damage. The other disk sample (SIN; 58 cm in radius, number of tree-ring of 100) had been cut in Kunigami village before the year 2000 and preserved in the Forest Owners' Co-operative Association of Kunigami. Unfortunately, the exact location of SIN samples is unknown (elevation of this area ranged from 20–300 m). From these two disks, triangular prism subsamples (one side *ca*. 2 cm in length) were taken using a circular saw. To improve the reliability of the isotope data, two series were sampled from each disk (hereafter called subsample-A and -B).

Using an increment borer, we also took two samples (GNW1 and GNW2) from living Ryukyu pine trees located in the city of Ginowan in 2014 (*ca*. 100 m in elevation). The numbers of tree rings in the GNW1 and GNW2 samples were 50 and 55, respectively. Tree ring widths were not measured.

Isotopic measurement

Stable isotope composition of tree-ring cellulose was measured by extracting α-cellulose from tree-ring cross-sectional laths (Kagawa *et al*., 2015; Xu *et al*., 2013). Briefly, a 1 mm thin plate was prepared from the tree-ring sample. Then, cellulose was extracted using sodium chlorite, NaOH, and toluene. The boundary between the latewood of the previous year and earlywood of the current year was identified visually, and then the cellulose sample was cut at annual tree-ring intervals. Each cellulose subsample of 150–350 μg in weight was wrapped in silver foil. Subsequently, it was converted to CO and H_2 using a Pyrolysis furnace of thermal conversion elemental analyzer (TCEA, Thermo Fisher Scientific Co. Ltd.). To measure the $\delta^{18}O$ values, the CO gases were sent to an isotope ratio mass spectrometer (Delta V advantage, Thermo Fisher Scientific Co. Ltd.) at the Research Institute for Humanity and Nature (RIHN). The $^{18}O/^{16}O$ ratio was expressed as relative value with respect to the Vienna Standard Mean Ocean Water (VSMOW) using a δ notation. The 1σ reproducibility of $\delta^{18}O$ is ±0.2‰.

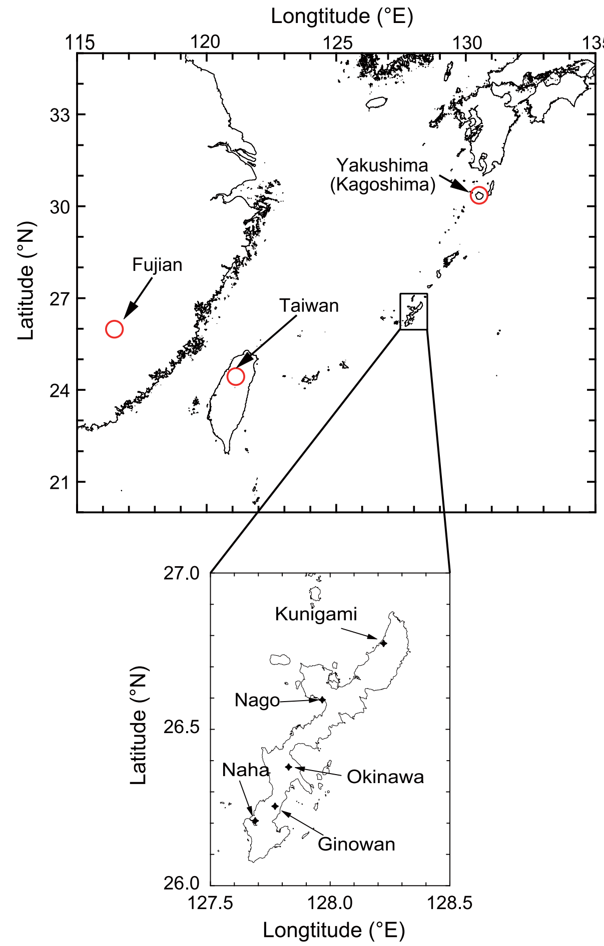

Fig. 1. Map showing tree-ring records and meteorological stations.

Cellulose $\delta^{18}O$ data in neighboring regions

In tropical region, the tree-ring identification is often difficult because of ambiguous tree-ring boundaries. Thus, to check the accuracy of tree-ring boundary identification, our $\delta^{18}O$ records from Okinawa were compared with three $\delta^{18}O$ chronologies from neighboring regions: Japanese cedar (Yakushima, Kagoshima, Japan; hereafter called Kagoshima $\delta^{18}O$) (Sano *et al*., under revision), *Fokienia hodginsii* (Fujian Province, China; hereafter called Fujian $\delta^{18}O$) (Xu *et al*., 2013), and *Chamaecyparis*

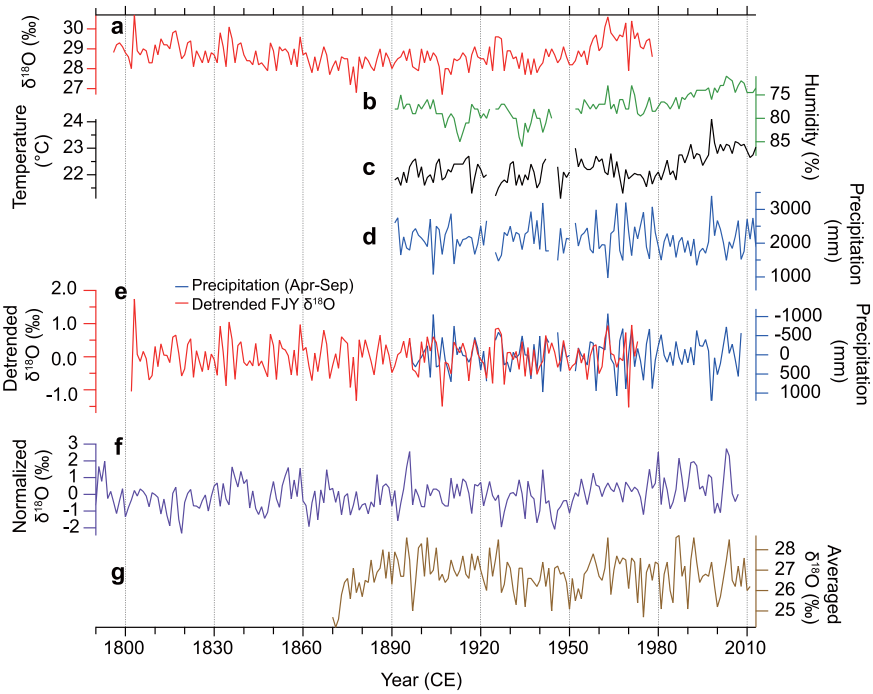

Fig. 2. Meteorological data in Okinawa island and oxygen isotope tree-ring records. (a) Cellulose δ^{18}O record of FJY (red). (b) Annual mean relative humidity in Okinawa. (c) same as (b) but for air temperature. (d) same as (c) but for precipitation amount. (e) detrended δ^{18}O record of the FJY sample (red) and precipitation amount during rainy season (April–September, blue). (f) normalized δ^{18}O record of Taiwan (Liu et al., 2017) (g) δ^{18}O record of Fujian δ^{18}O) (Xu et al., 2013).

formosensis (Mt. Daxue, Taiwan; hereafter called Taiwan δ^{18}O) (Liu *et al.*, 2017). Generally, the correlations of δ^{18}O in neighboring regions (*ca*. several hundred km in distance) are high (e.g., Sano *et al.*, 2017). Thus, a certain degree of correlation is expected in similar climate regions, including the East Asian summer monsoon. To investigate the correlation between interannual variations of δ^{18}O without the long-term trend, correlation analyses were performed using a detrended δ^{18}O record (i.e., raw data minus the 11-year running average).

Meteorological data and climate indices

Meteorological records were obtained from two stations, Naha and Nago, operated by Japan Metrological Agency (Fig. 1). At the Naha station, located on the southern part of Okinawa Island, climate observations began in 1891 with a seven-year interruption due to Second World War. At the Nago station, located on northern part of Okinawa Island, monitoring began in 1967 for air temperature and precipitation, and has included relative humidity since 1973. The distance between the two stations is *ca*. 50 km, and an averaged record was used for analyses (Fig. 2).

The cellulose δ^{18}O records were also compared with ENSO related indices, an extended version of Multivariate ENSO Index (MEI.ext), and the Niño 3.4 SST Index (Niño 3.4). MEI.ext is defined as the first principal component of the Sea Surface Temperature (SST) and Sea Level Pressure (SLP) field at 30°N–30°S, and 100°E–70°W, excluding the Atlantic Ocean (Wolter and Timlin, 2011). The Niño 3.4 was calculated from the HadISST1 based on the area averaged SST from 5°S–5°N and 170–120°W (Rayner, 2003). The MEI.ext and Niño 3.4 data were obtained from the National Oceanic and Atmospheric Administration (NOAA).

Results and Discussion

Isotopic data and age of FYJ sample

The age of the outermost layer of FJY sample was determined based on its cut year and correlation analysis with tree-ring isotopic chronologies in neighboring regions. The FJY sample was taken from a tree that was cut down in 1980 after it died. A thin early wood layer was observed outside the latest δ^{18}O layer, suggesting that the latest layer corresponds to 1979 or earlier. Then, we conducted correlation analysis by varying the outermost year of the FJY from 1974 to 1980. When we assumed an outermost year of 1977, the FJY δ^{18}O record showed a significant correlation ($p < 0.01$) with the δ^{18}O records of Kagoshima ($r = 0.30$), the Fujian ($r = 0.26$) and the Taiwan ($r = 0.19$). Thus, the outermost year of FJY was des-

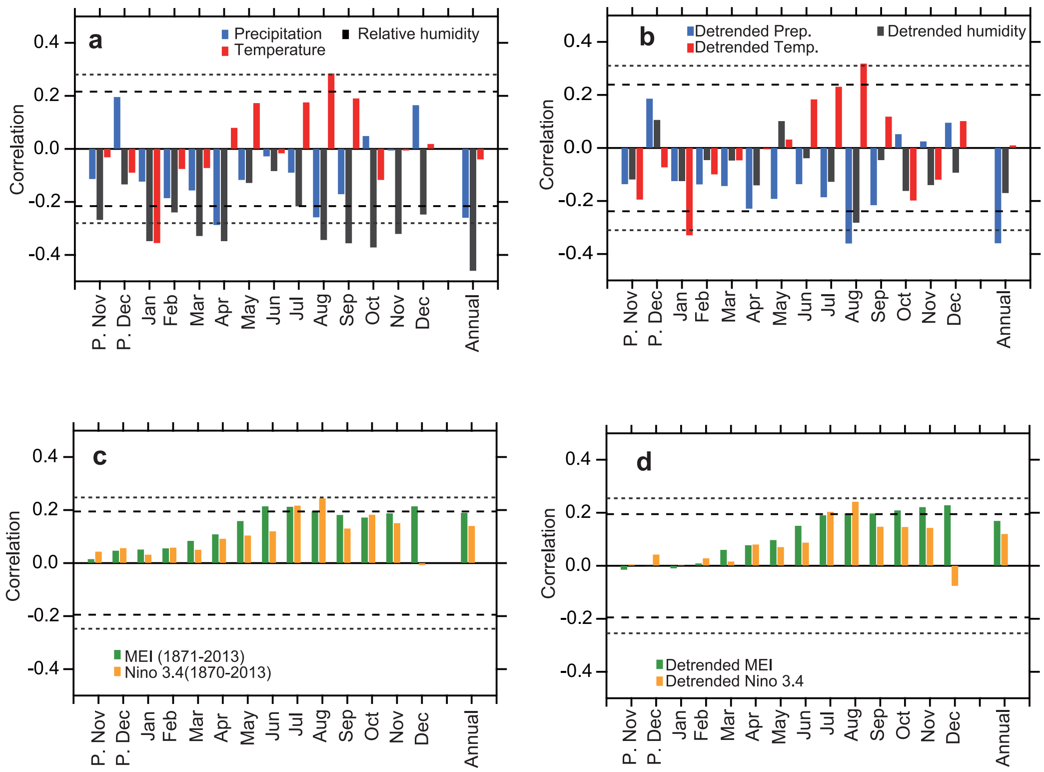

Fig. 3. Correlation between meteorological data and oxygen isotopes of cellulose. (a) Correlation coefficient of FJY $\delta^{18}O$ record with respect to P (blue), H (black) and T (red). (b) Same as in (a) but for the detrended FJY $\delta^{18}O$ record and detrended meteorological factors. (c) Correlation coefficient between FJY $\delta^{18}O$ record and MEI.ext (green) and Nino 3.4 (yellow). (d) Same as in (c) but for the detrended FJY $\delta^{18}O$ record and climate indices. Horizontal lines indicate statistically significant level of the correlation coefficient (long dotted line for $p < 0.05$; short dotted line for $p < 0.01$).

ignated as 1977 (hereafter called "sequence-A").

It is important to note that there is another possible age model for the FJY sample because of two ambiguous annual tree-rings in sequence-A (Supplementary Fig. S1). First, there is a thin layer, which was not identified as an annual tree ring in subsample-A. Thus, this layer, corresponding to year of 1970, may contain two tree rings. Second, a single layer in subsample-B, regarded as a layer of 1957 for sequence-A, involves two thin layers. This layer may be separated into two layers as 1956 and 1957. In summary, we can make another age-model by removing the 1970 layer in subsample-A and separating the 1957 layer into two in subsample-B. This results in a one-year shift of the outermost layer year. Thus, the outermost year of the FJY sample was designated as 1978 for this age model (hereafter called "sequence-B"). At the outermost year of 1978, we found significant correlations ($p < 0.01$) between $\delta^{18}O$ records of the FJY sample and Kagoshima $\delta^{18}O$ (r = 0.36), the Fujian $\delta^{18}O$ (r = 0.31) and the Taiwan $\delta^{18}O$ (r = 0.19) (Fig. 2).

Differences between the two age models were not significant because only recent parts of the 180-year-long time series were affected. In fact, the two FJY $\delta^{18}O$ records (i.e., sequence-A and -B) show high correlation for the entire time series (r = 0.90) (Fig. S2). Thus, the following analysis does not depend on the choice of age model. Here, we adopt sequence-B because (1) correlations with $\delta^{18}O$ chronologies in neighboring regions were slightly higher than age-model A and (2) the year of the outermost layer is consistent with the reported tree removal in 1980.

Isotopic data of the other samples

For the SIN sample, the best correlation with the $\delta^{18}O$ chronology in the neighboring region was found when the outermost year of 1993 was assumed. Although the $\delta^{18}O$ data of the SIN sample showed statistically significant correlation ($p < 0.01$; n = 90) with Fujian $\delta^{18}O$ (r = 0.29), it correlated weakly with the Taiwan $\delta^{18}O$ record (r = 0.20) and did not correlate with the Kagoshima $\delta^{18}O$ record (r = 0.06). Furthermore, at the outermost year of 1997, the SIN data showed significant correlation with Kagoshima $\delta^{18}O$ record ($p < 0.05$; r = 0.25) but no correlations with the chronologies from Fujian and Taiwan. Thus, the outermost age for the SIN data cannot be determined uniquely.

We found that there are ambiguous tree-ring boundaries in GNW1 and GNW2. The $\delta^{18}O$ records of the samples taken from two living trees (GNW1 and GNW2) correlated very weakly or even negatively with the $\delta^{18}O$ chronologies in the neighboring region. We then compared correlations among the four Okinawa $\delta^{18}O$ time series (Supplementary Fig. S2). We calculated the outermost year of SIN sample either 1993 or 1997 for this analysis. There was no significant correlation among them, except for that for GNW1 versus FJY. However, this correlation between FJY and GNW1 is based on just five data points.

These results suggest that the dating of these samples (SIN, GNW1 and GNW2) were incorrectly identified because of ambiguous annual boundaries between tree rings. Thus, in this study, we rejected these data. For FJY sample, horizontal section of the disk sample allows us to select subsamples where clear annual rings were observed. In contrast, the core samples of living tree samples (GNW1 and GNW2) using the increment borer contains unclear tree ring boundaries. In the future, robust age may be determined even for these samples using seasonally resolved cellulose $\delta^{18}O$ measurements (Xu *et al*., 2014).

Oxygen isotope record and correlations with climate elements

Figure 2 presents the FJY $\delta^{18}O$ record, which covers a 95-year period before meteorological observation began on this island in 1891. Meteorological data, precipitation amount (hereafter, P), relative humidity (H), and air temperature (T) are also shown in Fig. 2. The cellulose $\delta^{18}O$ values varies from 26.7‰ to 30.7‰. The $\delta^{18}O$ record shows *ca*. ±1‰ variations at a 2- to 10-year periodicity. On the decadal time scale, the $\delta^{18}O$ record shows steadily increasing trend since 1950.

Correlation coefficients between the FJY $\delta^{18}O$ record and climate elements are shown in Fig. 3a. For annual mean climate, the FJY $\delta^{18}O$ record indicates statistically significant correlation ($p < 0.01$) with P ($r = -0.26$) and H ($r = -0.46$), but no correlation between the $\delta^{18}O$ and T ($r = -0.03$). The high correlation between H and $\delta^{18}O$ appears to be influenced by the decadal scale variations of H, which includes a broad dry period between 1919 and 1935 and a decreasing trend from 1950 to 2013 (Fig. 2). For monthly mean climate data, the $\delta^{18}O$ data also shows significant correlations with P (April and August) and H (January–April; August–December) (Fig. 3a).

To investigate the interannual variations in FJY $\delta^{18}O$ without the decadal trend, correlation analysis was performed with the detrended records. Correlation coefficients between the detrended FJY $\delta^{18}O$and detrended climate factors are presented in Fig. 3b. For interannual variation, the detrended $\delta^{18}O$ shows a significant correlation ($p < 0.01$) only with P ($r = -0.34$). The detrended $\delta^{18}O$ correlates with P, H and T in several months (April and August for P; August for H, January and August for T). Notably, the FJY $\delta^{18}O$ record shows broad correlation with P using the rainy season (April–September). In fact, we find a high correlation ($r = -0.46$; $p < 0.01$) between the $\delta^{18}O$ and precipitation amount during the rainy season (Fig. 2e).

Precipitation and humidity effects

The observed negative correlations of the FJY $\delta^{18}O$ with H and P are consistent with the expected controlling factors for cellulose $\delta^{18}O$ values. Based on physical and biogeochemical theory (Roden *et al*., 2000), the cellulose $\delta^{18}O$ value reflects H and the $\delta^{18}O$ of source rainwater. First, under higher H conditions, the isotope fractionation during leaf-water evaporation results in lower $\delta^{18}O$ values of cellulose because of suppressed evapotranspiration of leaf water. Thus, the negative correlation between H and $\delta^{18}O$ is consistent with the leaf-water evaporation model. Second, rainwater observations in Okinawa Island between 2008 and 2012 showed that the monthly $\delta^{18}O$ of rainwater correlated negatively with P because of the rain-out process (i.e., amount effect) (Uemura *et al*., 2012, 2016). Thus, the negative correlation of cellulose $\delta^{18}O$ with P can be attributable to the $\delta^{18}O$ values of rainwater.

Similar negative correlation between summer precipitation (and H) and cellulose $\delta^{18}O$ values of pine trees was observed in central Japan (Li *et al*., 2015). A stronger influence of humidity on cellulose $\delta^{18}O$ of pine trees was also reported compared to its influence on oak trees (Li *et al*., 2015). These results were consistent with our observations. It is notable that it is difficult to separate these two factors because the detrended P correlates well with the detrended H ($r = 0.40$, $p < 0.01$) on the Okinawa Island.

Correlation with ENSO

The correlation coefficient between the FJY $\delta^{18}O$ record and ENSO-related indices (Niño 3.4 and MEI.ext) are shown in Fig. 3c. Since both indices are related to ENSO, there is a positive correlation between MEI and Niño 3.4. FJY $\delta^{18}O$ records correlates weakly ($p < 0.05$) with MEI.ext record for June, July, August and December and with Niño3.4 index for July and August. Figure 3d presents the correlation analyses using the detrended FJY $\delta^{18}O$ data. The MEI.ext record shows weak correlations ($p < 0.05$) with the detrended FJY $\delta^{18}O$ record from August to December. The Niño3.4 index also correlates weakly ($p < 0.05$) with the FJY $\delta^{18}O$ record for July and August. These results imply that ENSO weakly influences the $\delta^{18}O$ value of tree-ring cellulose at Okinawa from late summer to early winter.

We should note, however, that the degree of correlation observed in Okinawa is much weaker than that found at lower latitudes in the Southeast Asian region. For example, the $\delta^{18}O$ chronology from northern Vietnam correlated strongly with the Niño 3.4.index from 1871 to 2004 ($r = 0.67$) (Sano *et al.*, 2012b). By contrast, for inter-annual variations, the FJY $\delta^{18}O$ record correlates weakly with the MEI.ext ($r = 0.19$, $p < 0.05$) and did not correlate significantly with the Niño3.4 index. These results suggest that the cellulose $\delta^{18}O$ values in Okinawa was partly influenced by the ENSO, but was primarily controlled by the local $\delta^{18}O$ values of rainwater during the rainy season. A 13-year observation of $\delta^{18}O$ values of rainwater in the Galapagos Islands suggests that substantially low $\delta^{18}O$ values are observed only when precipitation is very strong during the warm months associated with El Niño, because the SST threshold of 28°C for deep convection is exceeded during this time (Martin *et al.*, 2018). This analysis suggests that the influence of the ENSO on the $\delta^{18}O$ of rainwater in Okinawa would be limited to strong ENSO periods.

Conclusions

We presented a long $\delta^{18}O$ record of tree-ring cellulose from the maritime island, Okinawa, in the East Asian monsoon region. The data cover climate variations on this island during the nineteenth century, which extend the instrumental observation record by 95 years. Since long-term tree-ring samples from this region are very scarce, the data are valuable for the evaluation of local climate variability and for comparison with historical documents on this island. Response function analysis revealed that the cellulose $\delta^{18}O$ record is controlled by relative humidity and the amount of precipitation. Particularly, the cellulose $\delta^{18}O$ record shows a strong correlation with the amount of precipitation during the rainy season. This result suggests that the cellulose $\delta^{18}O$ record in Okinawa Island is controlled by the local amount effect rather than ENSO variations.

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Supplementary Materials

URL (http://www.terrapub.co.jp/journals/GJ/archives/data/52/MS543.pdf)
Figures S1 and S2
Tables S1 and S2