



Supplement of

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

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Supplementary Materials

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S1 Leaf volume model for long-term age trends in tree-ring cellulose δ^{18} O and δ^{2} H data for Japanese conifers

By solving the simultaneous Eqs (15)–(18), we can extract the physiological component ($\Delta \delta^{18}O_{cel(physiol)}$) from tree-ring cellulose $\delta^{18}O$ as follows:

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$$\Delta \delta^{18} O_{\text{cel(physiol)}} = \frac{A \times \delta^{18} O_{\text{cel}} - \delta^2 H_{\text{cel}}}{A + B} - \frac{A \times \delta^{18} O_{\text{cel}(0)} - \delta^2 H_{\text{cel}(0)}}{A + B}.$$
 (22)

By applying this equation (assuming A = 5 and B = 5.4; Section 3.9) to the tree in Fig. 4 (No. 65) and the three nearly millennial-length trees cut in the 20th century in Figs S3–S5 (Nos. 43, 49, and 40), we identified distinct long-term positive relationships between $\Delta\delta^{18}O_{cel(physiol)}$ values and tree-ring widths (Fig. S6). This means that increased growth rate results in a lower rate of post-photosynthetic isotopic exchange for oxygen in carbohydrate (f in Eqs (3) and (4)) and in higher cellulose $\delta^{18}O$ values. However, this relationship does not always hold for natural forests, and it is necessary to explain why it holds for the dense Japanese conifer forests where our samples were collected, and in which trees have often been influenced by anthropogenic activities.

- One of the most important characteristics of trees in dense conifer forests is the drastic change in the vertical distribution and volume of leaves during their lives due to both natural and anthropogenic factors. In Arctic or mountainous forests, where trees are sparsely distributed, trees grow with sufficient sunlight, such that the vertical distribution and volume of leaves are
- similar amongst individual mature trees and do not show large temporal variations (Fig. S8a). However, in dense Japanese conifer forests, the average position of leaves increases and the total leaf volume decreases during tree growth due to insufficient sunlight (Fig. S8b). In such a forest, natural or anthropogenic disturbances to the tree or neighboring trees cause the leaf volume and distribution to change markedly. If neighboring trees are removed, then the improved sunlight conditions increase the leaf volume of the tree. If some branches of the tree are cut, then this decreases the leaf volume.

Tree No. 65 in Fig. S6a germinated in the 18th century, grew rapidly in its juvenile stage, and shows a gradual decrease in growth rate during its first 100 yr, probably due to a decrease in leaf volume (Fig. S8b). This is typical of 200–300 yr old trees of Japanese cypress, because the forests in central Japan were intensively logged in the 17–18th centuries at the beginning of the Edo era (17–19th century). Most old trees of Japanese cypress, including No. 65, germinated in open space after forest

- 25 clearing, where abundant light facilitated rapid growth during their juvenile period (Totman, 1989). In addition, this tree experienced a sudden increase in growth rate (i.e., an increase in tree-ring width) at ca. 1950 CE, probably due to the logging of neighboring trees after the end of World War II in 1945 CE. Trees No. 43 and 49 in Fig. S6b–c germinated in the 12th century, and survived the intense logging activity in the 17–18th century, because they were left as seed trees. By the time of logging in the 17–18th centuries, the growth rates of these trees had increased, probably because of gradual cutting of
- 30 neighboring trees, which reached a maximum at ca. 1720 CE. After 1720 CE, the growth rates decreased gradually along with

those of neighboring (younger) trees like No. 65 in Fig. S6a. In contrast, tree No. 40 in Fig. S6d was located in a village and protected by villagers for a long time, and thus its tree-ring width remained high, although there were also long-term variations in growth rate that correlate with the physiological component of cellulose δ^{18} O values ($\Delta\delta^{18}$ O_{cel(physiol)}), probably reflecting intermittent anthropogenic disturbance to tree growth.

- Based on an understanding of the growth histories of these trees, we propose a leaf volume model to explain the long-term opposite age trends in tree-ring cellulose δ^{18} O and δ^{2} H of Japanese conifers, which is shown as the physiological component of cellulose δ^{18} O in Fig. S6. Figure S8c shows the carbohydrate dynamics in a tree. In a xylem cell, cellulose is synthesized using hexose phosphate. If the hexose phosphate is derived directly from the sucrose imported from the phloem, then the isotopic exchange between carbohydrate and xylem water can be minimized. However, if the hexose phosphate is recycled
- 40 from triose phosphate, the rate of isotopic exchange (f in Eqs (3) and (4)) increases and the physiological component of cellulose $\delta^{18}O(\Delta\delta^{18}O_{cel(physiol)})$ decreases. Changes in leaf volume of a tree inevitably influence the sucrose supply from the phloem to xylem. If the decrease in sucrose supply increases the recycling rate of carbohydrate from triose phosphate to hexose phosphate in the xylem cells, and vice versa, then it can explain the co-variations in tree-ring width and $\Delta\delta^{18}O_{cel(physiol)}$ in Fig. S6. Given that the leaf volume of a conifer tree does not change very quickly due to natural factors, this can explain why the
- 45 correlation between tree-ring cellulose δ^{18} O and δ^{2} H is always positive in the short-periodicity variations corresponding to climate signals, and also why it sometimes becomes negative in the long-periodicity variations that reflect physiological mechanisms.

As shown by the tree-ring width variations in Fig. S6, long-term variations in leaf volume of trees in a dense forest or a village must be different for individual trees, which reflects individual micro-ecological conditions, although there may be

- 50 some similarities due to coordinated anthropogenic impacts to forests in the past. In fact, the physiological component of treering cellulose δ^{18} O values in three old trees (Fig. S7a) shows a relatively weak correlation, which reduces the correlation between raw tree-ring cellulose δ^{18} O time-series and obscures climatological signals. However, the climatological component of tree-ring cellulose δ^{18} O values (Fig. S7b–c), which can be extracted by Eq. (21), shows a clear correlation for the three trees and corresponds well with existing low-frequency climate reconstructions. The three trees in Fig. S7 were significantly
- 55 influenced by anthropogenic activities in the past, and have not been used for paleoclimatological studies by traditional dendrochronology, including isotope dendrochronology. However, our new method makes it possible to utilize such trees for paleoclimatological studies, which is important given that most trees in highly populated Japan have experienced some anthropogenic effects.

No. of sample	Type of wood *1	Tree species	Start year*2	End year*2	Location of municipalities (Prefecture)	Latitude & Longitude (Local government*3)
1	А	Chamaecyparis obtusa	-612	-354	Nagoya city (Aichi)	35.18N, 136.91E
2	N	Chamaecyparis obtusa	-543	-194	Iida city (Nagano)	35.51N, 137.82E
3	А	Chamaecyparis obtusa	-537	-434	Nagoya city (Aichi)	35.18N, 136.91E
4	А	Chamaecyparis obtusa	-530	-288	Nagoya city (Aichi)	35.18N, 136.91E
5	А	Chamaecyparis obtusa	-465	-340	Nagoya city (Aichi)	35.18N, 136.91E
6	А	Chamaecyparis obtusa	-380	-279	Nagoya city (Aichi)	35.18N, 136.91E
7	А	Cryptomeria japonica	-348	-238	Nagoya city (Aichi)	35.18N, 136.91E
8	А	Chamaecyparis pisifera	-276	-158	Nagoya city (Aichi)	35.18N, 136.91E
9	Ν	Chamaecyparis obtusa	-267	71	Iida city (Nagano)	35.51N, 137.82E
10	А	Chamaecyparis obtusa	-253	-183	Nagoya city (Aichi)	35.18N, 136.91E
11	А	Sciadopitys verticillata	-225	-70	Kasugai city (Aichi)	35.25N, 136.97E
12	А	Sciadopitys verticillata	-216	-66	Kasugai city (Aichi)	35.25N, 136.97E
13	Ν	Chamaecyparis obtusa	-177	207	Koga city (Shiga)	34.97N, 136.17E
14	А	Chamaecyparis obtusa	-167	-56	Nagoya city (Aichi)	35.18N, 136.91E
15	А	Sciadopitys verticillata	-167	-57	Kasugai city (Aichi)	35.25N, 136.97E
16	А	Chamaecyparis obtusa	-83	32	Nagoya city (Aichi)	35.18N, 136.91E
17	Ν	Chamaecyparis obtusa	-65	169	Iida city (Nagano)	35.51N, 137.82E
18	Ν	Chamaecyparis obtusa	-20	442	Koga city (Shiga)	34.97N, 136.17E
19	Ν	Chamaecyparis obtusa	61	552	Iida city (Nagano)	35.51N, 137.82E
20	Ν	Chamaecyparis obtusa	76	600	Iida city (Nagano)	35.51N, 137.82E
21	А	Chamaecyparis obtusa	77	210	Nagoya city (Aichi)	35.18N, 136.91E
22	А	Chamaecyparis obtusa	118	277	Nagoya city (Aichi)	35.18N, 136.91E
23	Ν	Chamaecyparis obtusa	250	511	Iida city (Nagano)	35.51N, 137.82E
24	Ν	Chamaecyparis obtusa	332	640	Miyata village (Nagano)	35.77N, 137.95E
25	Ν	Chamaecyparis obtusa	450	621	Iida city (Nagano)	35.51N, 137.82E
26	Ν	Chamaecyparis obtusa	496	820	Iida city (Nagano)	35.51N, 137.82E
27	Ν	Chamaecyparis obtusa	539	774	Nirasaki city (Yamanashi)	35.71N, 138.45E
28	0	Chamaecyparis obtusa	584	792	Ikaruga town (Nara)	34.61N, 135.73E
29	0	Chamaecyparis obtusa	683	832	Okuwa village (Nagano)	35.68N, 137.66E
30	0	Chamaecyparis obtusa	709	890	Okuwa village (Nagano)	35.68N, 137.66E
31	Ν	Chamaecyparis obtusa	717	919	Iida city (Nagano)	35.51N, 137.82E
32	Ν	Chamaecyparis obtusa	719	1138	Iida city (Nagano)	35.51N, 137.82E
33	0	Chamaecyparis obtusa	802	1159	Okuwa village (Nagano)	35.68N, 137.66E
34	А	Sciadopitys verticillata	896	1116	Inazawa city (Aichi)	35.25N, 136.78E
35	А	Chamaecyparis obtusa	908	1196	Inazawa city (Aichi)	35.25N, 136.78E
36	А	Sciadopitys verticillata	1051	1169	Inazawa city (Aichi)	35.25N, 136.78E
37	А	Sciadopitys verticillata	1068	1207	Inazawa city (Aichi)	35.25N, 136.78E
38	А	Chamaecyparis pisifera	1087	1386	Inazawa city (Aichi)	35.25N, 136.78E
39	А	Sciadopitys verticillata	1100	1331	Inazawa city (Aichi)	35.25N, 136.78E
40	L	Chamaecyparis obtusa	1120	1930	Nakatsugawa city (Gifu)	35.49N, 137.50E
41	А	Chamaecyparis obtusa	1126	1224	Inazawa city (Aichi)	35.25N, 136.78E
42	А	Sciadopitys verticillata	1134	1267	Inazawa city (Aichi)	35.25N, 136.78E
43	L	Chamaecyparis obtusa	1139	1978	Okuwa village (Nagano)	35.68N, 137.66E
44	А	Sciadopitys verticillata	1148	1221	Inazawa city (Aichi)	35.25N, 136.78E

45	А	Chamaecyparis pisifera	1151	1417	Inazawa city (Aichi)	35.25N, 136.78E
46	Ν	Chamaecyparis obtusa	1169	1656	Nakatsugawa city (Gifu)	35.49N, 137.50E
47	А	Sciadopitys verticillata	1189	1351	Inazawa city (Aichi)	35.25N, 136.78E
48	А	Sciadopitys verticillata	1193	1326	Inazawa city (Aichi)	35.25N, 136.78E
49	L	Chamaecyparis obtusa	1197	1968	Otaki village (Nagano)	35.81N, 137.55E
50	А	Sciadopitys verticillata	1213	1313	Inazawa city (Aichi)	35.25N, 136.78E
51	А	Chamaecyparis obtusa	1218	1294	Kiyosu city (Aichi)	35.20N, 136.85E
52	А	Cryptomeria japonica	1219	1380	Inazawa city (Aichi)	35.25N, 136.78E
53	А	Chamaecyparis obtusa	1231	1374	Inazawa city (Aichi)	35.25N, 136.78E
54	А	Chamaecyparis obtusa	1265	1419	Kiyosu city (Aichi)	35.20N, 136.85E
55	А	Chamaecyparis obtusa	1276	1465	Kiyosu city (Aichi)	35.20N, 136.85E
56	А	Chamaecyparis obtusa	1278	1384	Kiyosu city (Aichi)	35.20N, 136.85E
57	А	Chamaecyparis obtusa	1290	1454	Kiyosu city (Aichi)	35.20N, 136.85E
58	А	Chamaecyparis obtusa	1331	1510	Kiyosu city (Aichi)	35.20N, 136.85E
59	А	Chamaecyparis obtusa	1348	1502	Kiyosu city (Aichi)	35.20N, 136.85E
60	А	Chamaecyparis obtusa	1485	1611	Kiyosu city (Aichi)	35.20N, 136.85E
61	L	Chamaecyparis obtusa	1689	1988	Otaki village (Nagano)	35.78N, 137.69E
62	L	Cryptomeria japonica	1718	1993	Otsu city (Shiga)	35.02N, 135.85E
63	L	Chamaecyparis obtusa	1723	1993	Otsu city (Shiga)	35.02N, 135.85E
64	L	Chamaecyparis obtusa	1730	2005	Agematsu town (Nagano)	35.78N, 137.69E
65	L	Chamaecyparis obtusa	1761	2005	Agematsu town (Nagano)	35.78N, 137.69E
66	L	Chamaecyparis obtusa	1827	2005	Agematsu town (Nagano)	35.78N, 137.69E
67	L	Chamaecyparis obtusa	1839	2005	Agematsu town (Nagano)	35.78N, 137.69E

*1. "A" = archeologically excavated wood; "N" = naturally buried log; "O" = old architectural wood; "L" = living tree. *2. Minus and plus numbers indicate years BCE and CE, respectively. *3. Sample locations are shown by the latitude and longitude of the governmental offices
65 in the municipalities where samples were collected, because most samples were not living trees for which the actual growth location can be identified.



Figure S1. Statistical reliability of the tree-ring cellulose δ^{18} O dating, which was calculated using "dplR" (Bunn, 2008). Blue sections (50-70 yr length) indicate >99% significance of the correlation between each δ^{18} O time-series and the δ^{18} O master chronology (averages of all individually normalized δ^{18} O variations). Green sections indicate the end of samples with <25-yr overlaps with the master chronology. If the dating fails, a red section emerges. Given that there are no red sections, all samples were successfully dated here. Prior to this calculation, all samples were tentatively dated by comparing their tree-ring cellulose δ^{18} O variations with those of samples previously dated by the traditional tree-ring width method (NNRICP, 1990). Tree-ring cellulose δ^{18} O values have been widely used to date archeological woods in

75 Japan and Korea, because the master δ^{18} O chronology has proven to be effective in dating woods in central–western Japan and southern Korea, irrespective of the tree species (Nakatsuka, 2015, 2018).



Figure S2. Cutting of an individual annual ring of a 1-mm-wide "cellulose" lath from the central part of tree No. 43, viewed under a microscope. Removal of all non-cellulose components from the wood lath (Kagawa et al., 2015) makes it easy to detach annual rings because

- there are cracks in the cellulose fibers at the ring boundaries in conifer trees. Note that the thickness of the blade in this picture is just 0.1 mm (a blade employed for ophthalmological surgery). Using this blade, we can cut annual rings wider than 0.1 mm. Most stem segments in this study have wider rings like tree No. 65, which is a typical case, with maximum, minimum, and average tree-ring widths of 2.10, 0.43, and 1.04 mm, respectively (Fig. S6a). However, two samples (Nos. 43 and 49) have extremely narrow tree-rings with maximum, minimum,
- 90 and average widths of 2.67, 0.04, and 0.42 mm (No. 43), and 2.73, 0.04, and 0.49 mm (No. 49), respectively (Fig. S6b-c). As a result, 38 and 21 annual rings could not be analyzed due to the narrowness of rings in a total of 840 and 772 rings in trees No. 43 and 49, respectively.



Figure S3. Comparison between tree-ring cellulose δ^{18} O (red) and δ^{2} H (blue) time-series in tree sample No. 43. (a) Raw data, (b) long-periodicity components (>11 yr; 11-yr running mean), and (c) short-periodicity components (<11 yr; deviation from the 11-yr running mean).



Figure S4. As for Fig. S3, but for tree sample No. 49'.



110 Figure S5. As for Fig. S3, but for tree sample No. 40'.



Figure S6. Comparison of the physiological component of tree-ring cellulose δ^{18} O values ($\Delta\delta^{18}O_{cel(physiol)}$) (green line) with the tree-ring widths (black line) for trees (a) No. 65, (b) No. 43, (c) No. 49, and (d) No. 40 in Table S1 and Fig. S1. $\Delta\delta^{18}O_{cel(physiol)}$ variations of a tree are shown as the anomaly from its average value over the total growth period of the tree.



Figure S7. (a) Comparison of the physiological component of tree-ring cellulose δ^{18} O values, (b) climatological component of tree-ring cellulose δ^{18} O, and (c) reintegrated climatological component and original δ^{18} O data at the 21-yr threshold for the three old trees No. 40 (black), No. 43 (red), and No. 49 (blue) in Table S1 and Fig. S1. All components in each tree are shown as anomalies from their average values over the total growth period of each tree.



Figure S8. Leaf volume model to explain long-term variations in the physiological component of tree-ring cellulose δ^{18} O values ($\Delta\delta^{18}O_{cel(physiol)}$) in Japanese conifer trees. Typical variations in leaf volume of conifer trees in (a) sparse forests in Arctic or mountainous regions and (b) dense forests in Japan. (c) Biochemical pathway of carbohydrate during formation of tree-ring cellulose.