1	Insights into deglacial East Asian Monsoon seasonality and inter-regional teleconnections from
2	Lake Suigetsu, Japan
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26 Abstract

27 The past evolution of the East Asian Monsoon (EAM) was affected by remote climate processes. 28 However, there is uncertainty surrounding these teleconnections and their impact on each seasonal EAM mode during periods of rapid global change. Here we use the unique biannual precipitation 29 patterns of central Japan to reconstruct winter and summer EAM evolution during the last glacial 30 31 termination (22,000 – 10,000 cal BP). Oxygen isotope analysis of diatoms and hydrogen isotope 32 analysis of n-alkanoic acids from the Lake Suigetsu sediment cores show that in Japan the winter 33 EAM weakened and the summer EAM strengthened with deglaciation. Only the summer mode exhibited variations coeval with stade-interstade fluctuations. A relationship between the summer 34 EAM and Antarctic temperature post-16,000 cal BP supports a remote link to the Southern 35 Hemisphere. These trends were different to continental EAM records and highlight the value of 36 37 broadening the geographical range of reconstructions to rationalise system heterogeneities.

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39 Introduction

40 The East Asian Monsoon (EAM) region is located at a point of convergence of multiple, globally interconnected climatic systems, with the palaeo-EAM system demonstrably affected by 41 42 remote surface and atmospheric processes on centennial to millennial timescales. This is most 43 eminently exemplified by the teleconnection between summer EAM strength and North Atlantic 44 temperature during the late Pleistocene; first evidenced by the relationship between the Hulu Cave stalagmite and Greenland ice core oxygen isotope (δ^{18} O) records (Wang *et al.*, 2001; Zhang *et al.*, 45 2019) and subsequently corroborated by studies from across the region (Herzschuh, 2006; Liu et al., 46 2022). This relationship is thought to have been driven by the influence of Atlantic Meridional 47 Overturning Circulation (AMOC) on the position of the Intertropical Convergence Zone (ITCZ) (Zhang 48 49 et al., 2019). However, other reconstructions of EAM strength have noted an inverse relationship to Antarctic temperatures in this interval (Han et al., 2016; Zhang et al., 2016) and, as a result, the view 50

51 of teleconnections acting on the EAM region has expanded to consider links to other climate 52 systems, including Antarctica and the Pacific Ocean.

53 The behaviour of the EAM during the last glacial termination (TI; 22,000 to 10,000 cal BP) is an ideal focus for characterising the changing seasonality of this system and the persistence of these 54 teleconnections. As a global transition, TI exemplifies a substantial, detectable change in many 55 56 palaeoclimate archives with the climate at the geographic poles (representing two possible "end 57 member" teleconnection origins) decoupled, with a Southern Hemisphere lead (Denton et al., 2010; WAIS Divide Project Members, 2013). In the North Atlantic, TI was characterised by colder 58 conditions during Greenland Stade 2.1 (GS-2.1; ~22,900 to 14,700 cal BP) and Greenland Stade 1 59 (GS-1; ~12,800 to 11,700 cal BP), alternating with warmer conditions during Greenland Interstade 1 60 (GI-1; ~14,700 to 12,800 cal BP) and the Holocene (~11,700 cal BP onwards) (Rasmussen et al., 2014). 61 62 GS-2.1, GI-1 and GS-1 were equivalent to the Last Glacial Maximum (LGM), Bølling–Allerød (BA) 63 Interstade and Younger Dryas (YD) Stade of the wider North Atlantic region. In the far Southern Hemisphere, warming was more gradual and interrupted only by the Antarctic Cold Reversal (ACR; 64 65 14,500 to 12,900 cal BP), which bestrode GI-1 and GS-1 (EPICA Community Members, 2006; WAIS Divide Project Members, 2013). 66

Whilst current evidence supports a weakening East Asian Winter Monsoon (EAWM) and 67 68 strengthening East Asian Summer Monsoon (EASM) during TI on an orbital scale (Wang et al., 1999; 69 Gallagher et al., 2018), centennial- to millennial-scale fluctuations in EAM strength and the relative 70 seasonal contributions to annual precipitation are poorly constrained (particularly in the case of the 71 EAWM; Wen et al., 2016) and appear spatially heterogeneous (Zhang et al., 2019). It is unsurprising, therefore, that there is uncertainty surrounding deglacial EAM evolution on submillennial 72 timescales (Yoshida and Takeuti, 2009; Hayashi et al., 2010; Liu et al., 2022), particularly in terms of 73 74 conflicting evidence for teleconnections to other regions. A possible source of this uncertainty is the limited number of available high-resolution EAM reconstructions. Consideration of both EAWM and 75

EASM strength in isolation is vital for deconvolving EAM behaviour during TI because they do not
always show a perfectly inverse relationship (Wang *et al.*, 2012; Wen *et al.*, 2016; Yan *et al.*, 2020)
and hence have the potential to exhibit unique behaviours (and teleconnection signals).

Due to its location, the EAM dominantly controls the climate of Japan; however, the 79 relationship between the EAM and precipitation in Japan is unique. Japan is situated directly 80 beneath the seasonally migrating monsoon front (Fig. 1), a critical geoclimatic boundary which 81 82 moves northwards during summer and southwards during winter and separates the influence of the EAWM and EASM modes (Nakagawa et al., 2006, Yoshida and Takeuti, 2009). The Japanese 83 archipelago lies between two water bodies (the Pacific Ocean and the Sea of Japan), hence both 84 EAM modes bring heavy precipitation to the country; mixed rainfall and snowfall concentrated along 85 the Sea of Japan coast during the EAWM, and rainfall across the entire country during the EASM 86 (Chowdary et al., 2019; Amekawa et al., 2021). This is in contrast to Continental Asia, where the 87 88 EASM brings heavy rainfall from the Pacific Ocean, but the EAWM is predominantly dry because it originates inland (Yancheva et al., 2007; Yan et al., 2020). As such, the annual distribution of rainfall 89 90 in Japan uniquely reflects a balance between both seasonal modes of the EAM. In light of these distinctive climatic characteristics, EAM records from Japan are well suited to deconvolve the 91 complexities of EAM evolution during TI. In this study we present new stable isotope-based proxy 92 93 records of both EAWM and EASM evolution in central Japan from the Lake Suigetsu sediment cores 94 and compare these to a series of global benchmark records of EAM variability and temperature 95 changes (Fig. 1), to better understand the factors affecting both seasonal modes of the EAM in Japan during this interval. 96





98	Fig. 1 – Lake Suigetsu and other key sites. a) the key global benchmark record sites discussed in the main text. b) the
99	sites within the EAM region, alongside the average positions of the modern monsoon front in January and July (adapted
100	from Nakagawa et al., 2006). c) the location of Lake Suigetsu in Fukui Prefecture, central Honshu, relative to key cities
101	and geographical features in Japan. Red boxes show the extent of subsequent panels in the sequence. Basemap is World
102	Imagery (WGS84) from Esri (2023) (scale 1:37,314,123 (a), 1:7,851,046 (b), 1:530,430 (c)).

104	Lake Suigetsu is a tectonic lake located ~1 km from the Sea of Japan in Fukui Prefecture,
105	Honshu Island (35° 35′ N, 135° 53′ E; Fig. 1; Nakagawa <i>et al.</i> , 2012). Suigetsu was a freshwater lake
106	for the last glacial-interglacial cycle, except during global sea level highstands in the Eemian and the
107	mid-Holocene, and post-1664 CE when the lake was artificially connected to the Sea of Japan
108	(Nakagawa et al., 2021). Four previous deep coring campaigns carried out in 1993 (SG93 core), 2006

109 (SG06), 2012 (SG12) and 2014 (SG14) recovered a series of overlapping sediment core sections from 110 the present day to >150,000 yr BP (Nakagawa et al., 2012). The preserved sequence, covering >98 111 m of composite sediment depth, provides a palaeoenvironmental archive that captures two glacial and two full interglacial periods of continuous sedimentation. The Suigetsu cores contain annual 112 laminations (varves) between ~70,000 yr BP and 10,000 cal BP, and between ~50,000 and 10,000 113 cal BP comprise the longest continuously varve-counted record from the Quaternary (Schlolaut et 114 al., 2018). Thin-section microscopic analysis showed that the varves comprise a spring layer of 115 116 Aulacoseira diatoms with some siderite, a detrital layer containing silt-sized quartz and feldspar, a summer layer of light amorphous organic material, an autumn layer of Encyonema diatoms within 117 118 siderite, and a transitional autumn-to-winter clay layer (Schlolaut et al., 2012).

119 Previous studies of the Suigetsu sediments from TI substantiate that this was a period of 120 significant climatic change at the catchment. Pollen-derived temperature reconstructions of this 121 interval show a structure which resembles that of North Atlantic temperature: abrupt warming at the start of a "Late Glacial Interstade", a more gradual decrease in temperature into a "Late Glacial 122 123 Stade", and an abrupt warming at the start of the Holocene (Nakagawa et al., 2021). However, whilst the onset of the Late Glacial Stade and the Holocene at Lake Suigetsu were synchronous with the 124 125 Younger Dryas and Holocene periods in the North Atlantic, the Late Glacial Interstade at Lake 126 Suigetsu commenced ~200 years prior to its North Atlantic counterpart (the Bølling–Allerød) 127 (Nakagawa et al., 2021). This behaviour was rationalised by a sudden repositioning of the westerly jet to the north of the Himalayas (a topographical barrier which causes bimodality in the positioning 128 of the jet) during boreal summer, which allowed warm air from the Pacific Ocean to propagate 129 130 northwards and caused the step-change in summer temperatures in Japan. Conversely, it was an 131 AMOC switch-on ~200 years later (due to a higher threshold response to insolation forcing), which 132 resulted in a shift to interstadial conditions in the North Atlantic. On a subcentennial scale, multiproxy analysis of the Suigetsu cores showed a bipartite structure to the Late Glacial Stade with 133

a transition that was synchronous, albeit slower and more muted, to those observed at Lakes
Meerfelder Maar (Germany) and Kråkenes (Norway) (Schlolaut *et al.*, 2017). A shift from *Aulacoseira ambigua* to *Aulacoseira subarctica* dominance during deglaciation was attributed to variations in
the EAWM, affecting vegetation coverage and the water nutrient status (Kossler *et al.*, 2011).
Carbon and nitrogen isotope ratios from chlorins also suggest an increase in aquatic primary
productivity from glacial to interglacial conditions in the lake (Tyler *et al.*, 2010).

Here we present EAM precipitation reconstructions from Lake Suigetsu based on two proxy 140 systems: the oxygen isotope composition of aquatic diatom silica frustules ($\delta^{18}O_{diatom}$) and the 141 compound-specific hydrogen isotope composition of n-alkanoic acids ($\delta^2 H_{acid}$) produced by aquatic 142 algae (C₁₆ and C₁₈ homologues) and terrestrial plants (C₂₈ and C₃₀ homologues). Analysis was 143 conducted using 100-year contiguous (continuous adjacent) samples. These proxies were selected 144 145 based on target material abundances in the Lake Suigetsu cores and the ability to reconstruct both 146 autochthonous (aquatic; $\delta^{18}O_{diatom}$, $\delta^{2}H_{C16acid}$ and $\delta^{2}H_{C18acid}$) and allochthonous (terrestrial; $\delta^{2}H_{C28acid}$) and $\delta^2 H_{C30acid}$) changes during TI (Leng and Barker, 2006; Holtvoeth *et al.*, 2019). Both isotope proxy 147 148 systems are versatile tools in palaeolimnological reconstructions of climate and follow the principle 149 that the isotope composition of palaeo-catchment water was incorporated (via biosynthetic processes) into target materials by diatoms, algae, and plants, and preserved in the lake (Leng and 150 151 Barker, 2006; Holtvoeth *et al.*, 2019). $\delta^{18}O_{diatom}$ and $\delta^{2}H_{acid}$ can be affected by fractionation 152 processes during the life cycle of their respective source organisms but are dominantly controlled by the isotope composition of their water source (Leng and Marshall, 2004; Holtvoeth et al., 2019). 153 Hence, broader-scale palaeohydrological inferences can be made, relating to regional-scale shifts in 154 atmospheric circulation and water vapour transport (Leng and Marshall, 2004; García-Alix et al., 155 2021; Tierney et al., 2022). At Lake Suigetsu, the modern isotope composition of water within the 156 157 catchment is closely related to that of precipitation (with minimal effects of evaporation in summer), which is dominantly controlled by both the EAWM and EASM (Rex et al., 2023, preprint), supporting 158

the utilisation of $\delta^{18}O_{diatom}$ and $\delta^{2}H_{acid}$ for reconstructing past EAM behaviour. Other potential influencing factors are water temperature in the aquatic domain, and air temperature and vegetation changes in the terrestrial domain (Leng and Barker, 2006; Castañeda and Schouten, 2011; Sachse *et al.*, 2012).

- 163
- 164 <u>Results</u>

165 **Observations at Lake Suigetsu**

The $\delta^{18}O_{diatom}$ values increased smoothly during TI with no abrupt shifts and the Holocene 166 167 values were ~2.5 ‰ higher than in the LGM (Fig. 2a). Key features of this profile were high frequency centennial-scale fluctuations and higher values between 22,000 and 21,000 cal BP. The $\delta^2 H_{C16acid}$ 168 values followed a smooth trend to lower values through the record, except from 22,000 - 21,000 169 170 cal BP (when low values were observed), 19,000 – 17,000 cal BP (when values fluctuated drastically on a centennial scale), and sections of the Late Glacial Interstade and Late Glacial Stade (when this 171 172 proxy exhibited periods of lower-than-average values; Fig. 2b). The difference between the $\delta^2 H_{C16acid}$ values at 21,000 cal BP and 10,000 cal BP was ~50 ∞ . The $\delta^2 H_{C18acid}$ profile resembled that of 173 $\delta^2 H_{C16acid}$, however long-term trends were harder to identify due to limited data coverage (see 174 Methods) and rapid high-amplitude fluctuations (Fig. 2c). Many of these coincided with equivalent, 175 lower amplitude, variations in the $\delta^2 H_{C16acid}$ values, suggesting that these were closely related. The 176 $\delta^2 H_{C28acid}$ and $\delta^2 H_{C30acid}$ profiles also showed very similar behaviour to one another; decreasing 177 178 through the LGM and into the Late Glacial Interstade before initially increasing smoothly for ~2000 179 years, and then decreasing from the early Holocene onwards (Fig. 2d. Fig. 2e). Like the other $\delta^2 H_{acid}$ proxies, the $\delta^2 H_{C28acid}$ and $\delta^2 H_{C30acid}$ records showed a period of lower values between 22,000 and 180 21,000 cal BP before a marked shift at 21,000 cal BP to higher values. The difference between the 181 182 $\delta^2 H_{C30acid}$ values at 21,000 cal BP and 10,000 cal BP was ~35 ‰. The $\delta^2 H_{C28acid}$ and $\delta^2 H_{C30acid}$ profiles

were only decoupled between 19,000 and 17,000 cal BP; within this section both proxies showed
rapid high amplitude changes which were offset by as little as one sample (100 years).

185 It appears that the shortest (C₁₆) and longest (C₃₀) chain lengths exhibited $\delta^2 H_{acid}$ profiles with the most unique trends and the most complete data coverage, and hence we focus on these in 186 subsequent discussions, whereas the $\delta^2 H_{C18acid}$ values were limited in quantity and the $\delta^2 H_{C28acid}$ 187 values showed some features similar to the $\delta^2 H_{C18acid}$ profile, possibly due to C₂₈ n-alkanoic acids 188 originating from a combination of aquatic and terrestrial sources. Whilst C₃₀ n-alkanoic acids 189 190 originate from strictly terrestrial sources, C₂₈ n-alkanoic acids have been known to originate from 191 aquatic and terrestrial sources (van Bree et al., 2018; Tierney et al., 2022). None of the trends exhibited by the isotope proxies resembled those of the pollen-derived temperature 192 193 reconstructions at Lake Suigetsu (mean temperature of the warmest month, MTWA or mean 194 temperature of the coldest month, MTCO; Fig. 2f, Fig. 2g; Nakagawa et al., 2021). Proxies reflective of local catchment change (e.g., total organic carbon, n-alkanoic acid concentrations and diatom 195 196 frustule concentration) exhibited smooth fluctuations during TI (Supplementary Fig. 1). Of these, only diatom frustule concentration showed any step-changes (at the onset of the Late Glacial 197 Interstade), suggesting that the other proxies were driven by gradual changes, as for the $\delta^{18}O_{diatom}$ 198 and $\delta^2 H_{acid}$ values (Supplementary Discussion 1). 199



Fig. 2 – Proxy changes at Lake Suigetsu during Termination I. a) $\delta^{18}O_{diatom}$ (this study), b) $\delta^{2}H_{acid}$ of C₁₆ n-alkanoic acids (this study), c) $\delta^{2}H_{acid}$ of C₁₈ n-alkanoic acids (this study), d) $\delta^{2}H_{acid}$ of C₂₈ n-alkanoic acids (this study), e) $\delta^{2}H_{acid}$ of C₃₀ nalkanoic acids (this study), f) MTWA; mean temperature of the warmest month (pollen-derived; Nakagawa *et al.*, 2021), g) MTCO; mean temperature of the coldest month (pollen-derived; Nakagawa *et al.*, 2021). Panels A to E are displayed inverted, and show data derived from 100-year integrated contiguous samples. All panels are on the IntCal20 timescale. Vertical dashed lines indicate the phases of temperature change at Suigetsu as designated by the pollen-derived temperature reconstructions (Nakagawa *et al.*, 2021).

209 Proxy Drivers

Determination of the dominant isotope proxy drivers is critical for interpreting the results in 210 211 the context of EAM behaviour. Whilst multiple factors can affect the proxy values presented here, valuable insights can be made by identifying the dominant driver (which can cause the largest 212 amplitude changes) in each case. There was no identifiable relationship between our isotope proxy 213 214 values and the pollen-derived temperature or the primary pollen taxa concentrations, which 215 eliminates air temperature (and thus water temperature) and vegetative change as the main drivers, respectively. There is a temperature dependent fractionation on $\delta^{18}O_{diatom}$ in precipitation and a 216 mineral-water fractionation, which have opposing effects, but the impact is likely to be small. Given 217 the amount of variation, we suggest that changes in water source composition were most likely the 218 dominant control on the $\delta^{18}O_{diatom}$ and $\delta^{2}H_{acid}$ values. The isotope composition of modern catchment 219 220 waters is principally governed by the composition of EAM-borne precipitation, as shown by 221 extended monitoring (Rex et al., 2023, preprint). EAM precipitation composition is determined by a combination of source composition and transport processes; in general, a "stronger" EAM, 222 223 associated with strong winds and large quantities of precipitation integrated across the transport pathway, would act to lower $\delta_{\text{precipitation}}$, and the reverse qualities (for a "weaker" EAM) would 224 provide higher $\delta_{\text{precipitation}}$ values (Yuan *et al.*, 2004; Zhang *et al.*, 2018). EAM strength is ultimately 225 226 driven by an enhanced land-sea pressure gradient, moisture availability and conditions favourable 227 for evaporation at source (Mohtadi et al., 2016). It follows that these were the critical drivers of the 228 isotope proxy systems during TI. On shorter timescales, other influences (e.g., local summer evaporation) could have also affected the isotope proxies, although there is a limited influence of 229 evaporation on catchment isotopes in the modern day (Rex et al., 2023, preprint), so we expect 230 these to have caused small-amplitude effects, if any. The marked seasonality of EAM precipitation 231 232 at Lake Suigetsu, combined with the short residence time of the lake (on the order of a few months; Rex et al., 2023, preprint) means that isotope proxy seasonality is crucial for attribution to one or 233

both seasonal EAM modes. A key element of this designation is related to the modern catchment transit lag of approximately three months (Rex *et al.*, 2023, preprint), which signifies that surface lake water isotope composition (δ_{lake}) reflects that of the precipitation ($\delta_{precipitation}$) delivered in the preceding season, i.e., spring δ_{lake} is equivalent to that of winter $\delta_{precipitation}$ and so on. Consistently, deep water δ_{lake} is homogeneous and reflects annually integrated $\delta_{precipitation}$.

239 In a palaeo context, different diatom taxa were associated with each of the predominant 240 seasonal blooms in the lake (spring and autumn) and consequently, the $\delta^{18}O_{diatom}$ values reflect a 241 combination of seasonal signals. Of the most abundant diatom taxa, mixed planktonic Aulacoseira subarctica and Aulacoseira ambigua diatoms bloomed in the spring and captured surficial spring 242 $\delta^{18}O_{lake}$ (winter $\delta^{18}O_{precipitation}$), and benthic *Encyonema* diatoms bloomed during the autumn and 243 captured annually integrated $\delta^{18}O_{\text{precipitation}}$ from the homogenous deep water (bloom season 244 245 attributions were derived from the varve structure; Schlolaut et al., 2012). The signals captured by 246 other taxa would have been dependent on their habitation depths and bloom seasons, however due to their low abundance relative to Aulacoseira and Encyonema diatoms, their seasonality was 247 not ascribed as part of the varve structure. "Bulk" $\delta^{18}O_{diatom}$ therefore reflects mixed seasonality, 248 249 with the relative weighting of each seasonal signal dependent on the mass contribution of each 250 taxon (Leng and Barker, 2006). In the absence of quantified taxa-specific masses within each sample, 251 the abundance (based on frustule counts) of each taxon can be used to provide an indication of the relative seasonal weighting of the $\delta^{18}O_{diatom}$ values; differences in taxon size can introduce 252 uncertainty to this process, but we believe this to be a reasonable assessment because whilst other 253 taxa are larger in size, the smallest genus of diatom, Aulacoseira, has thick valves and hence 254 contributes significantly to the total diatom mass. Due to its relationship with winter $\delta^{18}O_{\text{precipitation}}$, 255 Aulacoseira $\delta^{18}O_{diatom}$ offers the most robust links to EAWM behaviour. In theory, this connection 256 257 may have been affected by alterations to the transit lag, however we believe that the relationship was maintained throughout TI, despite climatic variability, because the spring diatom bloom in Lake 258

259 Sugetsu is closely related to the delivery of the bulk of winter precipitation to the lake (i.e., both are caused by snow and ice melting). Additionally, winter is the wettest season of the year, 260 minimising the effect of the preceding autumnal precipitation on the composition of surface δ_{lake} 261 (an effect amplified by the short residence time of the lake). In light of this, it is reasonable to utilise 262 the $\delta^{18}O_{diatom}$ values as an indicator of EAWM behaviour (i.e., strongly weighted towards winter 263 $\delta^{18}O_{\text{precipitation}}$), when *Aulacoseira* was the most abundant genus. This was the case for earlier parts 264 265 of the interval (22,000 - 19,800 cal BP) and from 15,000 cal BP onwards, when %Aulacoseira (Aulacoseira frustules as a percentage of total diatom frustules) was consistently greater than 60 % 266 (and often greater than 70 %) on a centennial basis (Supplementary Fig. 2). This was likely to have 267 been even greater in our $\delta^{18}O_{diatom}$ samples, which contained only diatoms which were smaller than 268 80 µm, favouring the (smaller) Aulacoseira over other taxa. However, the presence of other taxa 269 may have contributed to the low amplitude centennial-scale variability in the $\delta^{18}O_{diatom}$ record 270 271 throughout, and had greater influence between 19,800 and 15,000 cal BP. Maturation effects (i.e., post-mortem isotope exchange) remain an unconstrained influence on $\delta^{18}O_{diatom}$ (e.g., van 272 273 Hardenbroek et al., 2018), however (as with other studies) we do not expect this to be a dominant driver because the hydrous (most readily exchangeable) layer of each diatom was removed prior to 274 275 measurement (see Methods) and only the more resistant structurally bound oxygen fixed during 276 early sedimentation was analysed.

The $\delta^2 H_{acid}$ values are associated with less complex seasonality than the $\delta^{18}O_{diatom}$ values, because the growth of terrestrial plants occurs predominantly during spring and summer (Sachse *et al.*, 2012) and lake-based organic matter production in Lake Suigetsu occurs almost exclusively in summer when the lake becomes stratified and aquatic productivity is high (Schlolaut *et al.*, 2012). Indeed, because the terrestrial $\delta^2 H_{acid}$ values would have been unaffected by the catchment transit lag (the source water of terrestrial plants being soil pore water not lake water), we expect the $\delta^2 H_{C30acid}$ values to have been heavily weighted towards summer (EASM) $\delta^2 H_{precipitation}$. However, not 284 only would the aquatic $\delta^2 H_{acid}$ values have been affected by the transit lag (and hence captured the isotope composition of spring $\delta^2 H_{\text{precipitation}}$ = summer $\delta^2 H_{\text{lake}}$), but during periods of cold winters 285 (which TI typifies), we might expect extension of the transit lag time in the first half of the year due 286 to greater longevity of snow on the surrounding hillsides and hence slower delivery of precipitation 287 to the lake system (Rex et al., 2023, preprint). As a result, both winter and summer precipitation 288 would enter the lake in quicker succession. Under these circumstances, summer $\delta^2 H_{lake}$ would have 289 290 become a greater seasonal mix of EAWM $\delta^2 H_{\text{precipitation}}$ (entering the lake in late spring) and EASM $\delta^2 H_{\text{precipitation}}$ (entering the lake in late summer). Consequently, the $\delta^2 H_{\text{C16acid}}$ values reflect the 291 weighted average of annual $\delta^2 H_{\text{precipitation}}$. By calculating the numerical difference between the $\delta^2 H_{\text{recipitation}}$. 292 $_{C16acid}$ and $\delta^2 H_{C30acid}$ values, it is possible to qualitatively infer the relative influence of the EAWM and 293 294 EASM on the lake through time (Supplementary Discussion 2, Supplementary Fig. 3), although there 295 is also an effect of reduced lag time with warming on this signal.

296

297 EAM Behaviour in Japan During TI

298 By considering the high %Aulacoseira intervals in isolation (Supplementary Fig. 2), variations in the $\delta^{18}O_{diatom}$ values strongly support a reduction in EAWM strength during TI in Japan (Fig. 3d). 299 This aligns with Japanese stalagmite reconstructions of EAWM behaviour from the LGM and 300 301 Holocene (Sone et al., 2013; Amekawa et al., 2021). It is possible that EAWM weakening caused higher $\delta^{18}O_{diatom}$ values both directly (as discussed above) and indirectly (by reducing EAWM surface 302 303 runoff into the Sea of Japan, resulting in a transition from low- to high-salinity conditions in these EAWM source waters (Amekawa et al., 2021)). The $\delta^{18}O_{diatom}$ profile does not exhibit stade-304 interstade-like fluctuations as seen in the North Atlantic (typified by GRIP $\delta^{18}O_{ice}$; Fig. 3a; Rasmussen 305 et al., 2014) or Antarctica (typified by WAIS Divide $\delta^{18}O_{ice}$; Fig. 3h; WAIS Divide Project Members, 306 2013), refuting the dominance of any such teleconnection between these regions and the EAWM 307 during TI. Indeed, the $\delta^{18}O_{diatom}$ record does not show long term trends which parallel any of the 308

benchmark records shown in Fig. 3 suggesting that, instead, EAWM weakening during TI followed a more gradual trend akin to Northern Hemisphere summer insolation (albeit inversely; Fig. 3j; Laskar *et al.*, 2004). The trend in the $\delta^{18}O_{diatom}$ values between 22,000 and 21,000 cal BP inversely aligns with sea surface temperature (SST) records from the Northwest Pacific (Fig. 3i; Oba and Murayama, 2004; Oba *et al.*, 2006), suggesting that EAWM strengthening during this 1000-year interval was due to the intensification of glacial conditions in the region.

315 Variations in the $\delta^2 H_{C30acid}$ values support an overall strengthening of the EASM in Japan during TI (with lower values in the Holocene than the LGM), however this was non-linear and 316 occurred in three phases: EASM strengthening from the LGM into the Late Glacial Interstade, EASM 317 weakening from the Late Glacial Interstade to the early Holocene, and subsequent EASM 318 strengthening (Fig. 3f). An overall increase of EASM strength in Japan is also sustained by grain size 319 320 variations within the South China Sea core GIK17940-2 (<63 μm fraction; Fig. 3g; Wang *et al.*, 1999); 321 although the trend at Lake Suigetsu is not entirely equivalent to this because the South China Sea record shows a gradual, rather than tripartite, increase in EASM strength. Our reconstruction 322 323 indicates that EASM behaviour in Japan was also characteristically different to EASM behaviour in Continental Asia (Chinese cave composite $\delta^{18}O_{\text{speleothem}}$; Fig. 3b; Cheng *et al.*, 2016) and did not show 324 325 a response to North Atlantic-style stade-interstade fluctuations (Fig. 3a; Rasmussen et al., 2014). 326 However, a close inverse relationship is observed between EASM strength in Japan and Antarctic temperatures (illustrated by WAIS Divide $\delta^{18}O_{ice}$, Fig. 3h; WAIS Divide Project Members, 2013) 327 328 between 16,000 and 10,000 cal BP, including coincident timing of inflections at ~14,500 cal BP, ~13,000 cal BP and ~12,000 cal BP (i.e., a response to the ACR). This provides compelling evidence 329 for a teleconnection between the polar southern hemisphere and the EASM during this interval. 330 Prior to this, the $\delta^2 H_{C30acid}$ values were decoupled from WAIS Divide $\delta^{18}O_{ice}$, suggesting that this 331 332 teleconnection was only activated at this time. During the LGM, the $\delta^2 H_{C30acid}$ profile was distinctive and showed large amplitude fluctuations which did not occur synchronously with changes in the 333

global benchmark records, but many of these were also observed in the $\delta^2 H_{C16acid}$ values. Interpretation of this time period is complex and is presented in the Discussion section below.

The $\delta^2 H_{C16acid}$ values display a composite trend (Fig. 3e) which encapsulates characteristics 336 of both the $\delta^{18}O_{diatom}$ (Fig. 3d) and $\delta^{2}H_{C30acid}$ (Fig. 3f) records. The influence of each of these unique 337 behaviours appears to dominate the $\delta^2 H_{C16acid}$ values at different times during the interval: 338 similarities between the $\delta^2 H_{C16acid}$ and $\delta^2 H_{C30acid}$ profiles are observed during the early part of the 339 340 ACR and the Holocene; in the latter stages of the ACR the $\delta^2 H_{C16acid}$ profile resembles that of $\delta^{18}O_{diatom}$; and prior to 17,000 cal BP the $\delta^{2}H_{C16acid}$ profile shows mixed behaviour. These 341 observations are consistent with our assertions that the $\delta^2 H_{C16acid}$ values reflect a combined EAWM 342 and EASM signal, weighted to the season with a greater quantity of precipitation, and these results 343 support a changing balance between EAWM and EASM dominance in Japan during TI. The $\delta^2 H_{C16acid}$ 344 345 values show more EAWM character during the later stages of the ACR, when the EAWM was strong 346 and EASM strength was declining, but shows greater EASM affinity during the early ACR and the Holocene, when there was a strengthened EASM and a weakened EAWM. The numerical difference 347 between the $\delta^2 H_{C16acid}$ and $\delta^2 H_{C30acid}$ values suggests an increasing EASM influence on Lake Suigetsu 348 349 through time (Supplementary Fig. 3) which is consistent with our independent observations of EAWM and EASM strength from the $\delta^{18}O_{diatom}$ and $\delta^{2}H_{C30acid}$ values. 350



Fig. 3 – EAM evolution at Lake Suigetsu during Termination I with contextual global benchmark records. a) GRIP δ^{18} O_{ice} (Johnsen *et al.*, 1997; Rasmussen *et al.*, 2014; remodelled onto U-Th timescale), b) Chinese Cave Composite (Hulu, Dongge, Sanbao) δ^{18} O_{speleothem} (Cheng *et al.*, 2016; U-Th timescale), c) Lake Suigetsu MTWA; mean temperature of the warmest month (pollen-derived; Nakagawa *et al.*, 2021; IntCal20 timescale), d) Lake Suigetsu δ^{18} O_{diatom}, loess smoothed (span = 0.1) with 1σ confidence bands (this study; IntCal20 timescale), e) δ^2 H_{acid} for C₁₆ n-alkanoic acids, loess smoothed (span = 0.1) with 1σ confidence bands (this study; IntCal20 timescale), f) δ^2 H_{acid} for C₃₀ n-alkanoic acids, loess smoothed (span = 0.1) with 1σ confidence bands (this study; IntCal20 timescale), g) South China Sea (GIK17940-2) grain size

359 proportion <63 μ m (Wang *et al.*, 1999; remodelled on to IntCal20 timescale), h) WAIS δ^{18} O_{ice} (WAIS Divide Project 360 Members, 2013; WD2014 timescale), i) Northwest Pacific (MD01-2421) Sea Surface Temperature reconstruction (Oba 361 et al., 2006; remodelled onto Marine20 timescale), j) 30°N June Insolation (Laskar et al., 2004). Arrows indicate dataset 362 interpretation. Dashed lines approximately align climatic transitions. Abbreviations: Hol = Holocene, YD = Younger Dryas, 363 BA = Bølling–Allerød, H1 = Heinrich Event 1, LGM = Last Glacial Maximum, EAWM = East Asian Winter Monsoon, EASM 364 = East Asian Summer Monsoon, ACR = Antarctic Cold Reversal. Alignment of non-IntCal20/Marine20 timescales was 365 performed as follows: the GRIP $\delta^{18}O_{ice}$ dataset was converted from the GICC05 timescale to the U-Th timescale using 366 the transfer functions of Adolphi and Muscheler (2015) and Adolphi et al. (2018). The WD2014 timescale and the U-Th 367 timescale are compatible at the resolution presented here, with a minimal offset at the Holocene onset (Sigl et al., 2016) 368 and an offset of 225 years in the LGM (Sinnl et al., 2023), which has a negligible effect on alignment due to the gradual 369 nature of the changes observed in the WAIS δ^{18} O record. The U-Th and IntCal20 timescales are compatible because 370 IntCal20, beyond the tree ring limit (circa 13,900 cal BP) is dominated by the Hulu Cave U-Th timescale (Reimer et al., 371 2020).

372

373 Discussion

Our results provide novel insights into the evolution of the EAWM and EASM during TI, 374 alongside their driving mechanisms and teleconnections to other regional climate systems. The 375 gradual decrease in EAWM strength and overall increase in EASM strength in Japan strongly support 376 increasing Northern Hemisphere insolation (driven by Milankovitch cycles) as the primary drivers of 377 378 EAM evolution (Cheng et al., 2016; Mohtadi et al., 2016; Zhang et al., 2019). However, this common orbital driver breaks down as a result of different responses of each seasonal mode to submillennial 379 380 temperature fluctuations in either hemisphere and it becomes necessary to evoke alternative mechanisms to rationalise these decoupled (rather than inversely related) seasonal behaviours. 381

EAWM evolution in Japan appears unaffected by stades in either hemisphere, however other 382 records from Continental Asia (e.g., Lake Huguang Maar (Yancheva et al., 2007)) exhibit significant 383 North Atlantic character (i.e., a BA-YD structure). Despite high quality EAWM records being relatively 384 sparse for this interval, we do not dispute the evidence for a North Atlantic-EAWM teleconnection 385 386 during TI, and suggest that EAWM wind strength increased (decreased) during stades (interstades) in the North Atlantic, driven by Northern Hemisphere cooling (warming) and an intensified 387 (weakened) Siberian High as previously suggested (Nakagawa et al., 2006). Indeed, the pattern of 388 winter temperatures at Lake Suigetsu provides evidence for the impact of North Atlantic 389 temperatures on the Eurasian airmass (Nakagawa et al., 2021). However, as a consequence of the 390

391 interaction of the EAWM winds with the Sea of Japan, the decrease in EAWM strength here was 392 gradual (Fig. 3d). Specifically, we suggest that any response to North Atlantic stade-interstade 393 fluctuations was obscured from our reconstruction by a smoothed, low amplitude response of winter Sea of Japan SSTs to insolation (Wu et al., 2020) and greater inertia in the oceanic domain 394 compared to the terrestrial. Furthermore, we are able to clarify that the previously evidenced bi-395 396 partite Late Glacial Stade at Lake Suigetsu (Schlolaut *et al.*, 2017) was not associated with any shifts 397 in EAWM precipitation; whilst there is a 300-year period of lower $\delta^{18}O_{diatom}$ values within this 398 interval (between 12,284 and 12,093 cal BP; Fig. 3d), this fluctuation is not substantially different from other multi-centennial oscillations seen in the profile. However, it remains possible that there 399 was a bipartite pattern in EAWM wind strength. An Antarctica-EAWM teleconnection was precluded 400 401 by local EAWM moisture transport, the isolation of the Sea of Japan, and the blocking of Pacific 402 Ocean signals by the monsoon front, all of which limited the influence of interhemispheric 403 teleconnections.

Conversely, evidence from Lake Suigetsu supports a response of the EASM to stadial 404 405 conditions in the southern hemisphere (specifically the ACR) post-16,000 cal BP (Fig. 3f). Previous studies which have noted a relationship between EASM strength and Antarctic temperatures have 406 407 evoked the bipolar seesaw and migration of the ITCZ as a mechanism to explain this connection 408 (Han et al., 2016; Zhang et al., 2016). Whilst this would facilitate an inverse correlation between 409 EASM strength in Japan and the ACR, it would also suggest that equivalent shifts in the position of 410 the ITCZ caused by North Atlantic behaviours could affect EASM strength, and we do not see evidence of such a relationship in our records. Instead, we suggest that the Antarctic-EASM 411 teleconnection during TI resulted from Antarctic cooling (warming), which caused coupled 412 atmospheric-ocean circulation changes that resulted in warming (cooling) in the Western Pacific 413 414 Warm Pool, enhanced (decreased) evaporation and a strengthened (weakened) Western Pacific Subtropical High, which caused a stronger (weaker) EASM (Wang and Fan, 2005; Xu et al., 2020). 415

The coexistence of a North Atlantic teleconnection to summer temperature and an Antarctic 416 417 teleconnection to summer (EASM) precipitation at Lake Suigetsu is reconcilable by noting that 418 temperature is much more likely to exhibit patterns common to the same hemisphere, whereas the relationship of the EASM to Pacific Ocean behaviours facilitates the influence of the polar Southern 419 Hemisphere on precipitation. Indeed, our observations of EASM strength at Lake Suigetsu, 420 combined with the pollen-derived summer temperature record, support a "Southern Hemisphere" 421 422 lead" to stade/interstade fluctuations during TI; changes in EASM strength led changes in summer 423 temperature because the former was driven by Antarctic processes and the latter by North Atlantic behaviours. 424

425 Our observations of EASM behaviour in Japan directly contradict those of the Chinese cave 426 composite, which supports an EASM response to North Atlantic stade-interstade fluctuations and 427 not Antarctic ones (Fig. 3b; Cheng et al., 2016). Because all of the caves included in the composite 428 (Hulu, Sanbao and Dongge) and Lake Suigetsu are located south of the monsoon front during the 429 summer (and hence under the influence of the Pacific air mass; Nakagawa et al., 2006), differences 430 cannot be resolved by the blocking of teleconnections by the monsoon front, as previously suggested (Yoshida and Takeuti, 2009), and instead must be determined by a different geoclimatic 431 432 boundary. One possible mechanism involves the overprinting of Antarctic signals in Continental Asia by the partitioning of precipitation between the EASM and post-EASM stages (jet-transition 433 434 hypothesis; Zhang et al., 2018). This phenomenon is linked to the prevailing climatic conditions in the North Atlantic by the timing of the pre-summer northwards shift of the westerly jet and would 435 have caused drier (wetter) conditions in central China during stades (interstades) in the North 436 437 Atlantic For this to not impact on EASM evolution in Japan, the effect of this partitioning must have 438 been spatially restricted to the continent, or limited to latitudes at, or lower than, the southernmost 439 position of the westerly jet (Lake Suigetsu is located to the north of this position and Hulu, Dongge and Sanbao Cave to the south (Nakagawa et al., 2021)). An alternative explanation is that the 440

speleothem records contain an unresolved temperature-controlled component (demonstrated by 441 442 their close relationship to the pollen-reconstructed temperature at Lake Suigetsu; both show a 443 remote North Atlantic influence) which obscured the Antarctic signal in favour of a North Atlantic one. Regardless, we posit that the location of Japan at a higher latitude, adjacent to the Pacific 444 Ocean and geographically separated from the rest of East Asia facilitated the preservation of Pacific 445 446 (and hence Antarctic) signals in our record of EASM behaviour at Lake Suigetsu. Whilst this 447 Antarctica-Japan teleconnection has not been observed in other EASM reconstructions (Wang et al., 448 1999; Ishiwatari et al., 2009; Hayashi et al., 2010), this is likely due to differences in proxy system and a direct relationship between the isotope composition of precipitation and EAM evolution. 449

However, prior to ~16,000 cal BP, EASM strength in Japan and Antarctic temperatures were 450 451 decoupled. We suggest that the establishment of this teleconnection was as a consequence of the 452 northwards repositioning of the westerly jet (relative to the Himalayas), a phenomenon previously 453 used to explain the onset of the Late Glacial Interstade (BA-equivalent; 15,000 cal BP) at Lake Suigetsu (Nakagawa et al., 2021). Not only did this northwards shift of the westerly jet allow warm 454 455 Pacific air to reach Japan, it also moved the EAM front to the north of Lake Suigetsu during boreal summer (because the position of the westerly jet also modulates the seasonal positioning of the 456 457 EAM front), having been situated permanently (year-round) to the south during the LGM (Chiang et al., 2015; Nakagawa et al., 2021). This would allow the EASM to propagate to Japan and connect 458 459 summer precipitation to the aforementioned components of the Pacific atmosphere-oceanic circulation (and hence the southern hemisphere). There were no abrupt shifts in the $\delta^2 H_{C30acid}$ values 460 at the onset of the Late Glacial Interstade (15,000 cal BP) to reflect this expansion of EASM operation 461 to include Japan; however, prior to 16,000 cal BP the $\delta^2 H_{C30acid}$ values were highly variable and 462 structurally distinctive from WAIS Divide $\delta^{18}O_{ice}$. Hence, we suggest that this Antarctic-Japan 463 teleconnection was established at 16,000 cal BP, facilitated in the 16,000-15,000 cal BP period by 464 intermittent northward migrations of the westerly jet and exposure of Japan to the Pacific airmass 465

466 during summer (captured by our centennial sampling resolution), and then continued post-15,000 cal BP when the EAM front was situated permanently to the north of Lake Suigetsu during summer. 467 It follows that prior to the establishment of this teleconnection, precipitation falling during 468 summer in Japan would not technically be equivalent to the EASM (because it would originate and 469 transit north of the EASM front), and instead would reflect local transport of precipitation under the 470 471 influence of the continental airmass (possibly following a similar south-east to north-west trajectory 472 due to pressure gradients, but with a significantly reduced transport distance). The highly variable behaviour of the $\delta^2 H_{C30acid}$ values (and hence the $\delta^2 H_{C16acid}$ values) between 22,000 and 16,000 cal 473 BP may have been caused by the lack of constant, regional driver of summer precipitation (i.e., the 474 EASM) in contrast to the $\delta^{18}O_{diatom}$ values, which were weighted towards the (enduring) EAWM. 475 These fluctuations in the $\delta^2 H_{acid}$ values could be driven by a number of factors, including 476 precipitation amount, changing relative humidity, shifts in precipitation source and variable SSTs 477 478 (particularly between 22,000 and 21,000 cal BP (Oba et al., 2006)). Another possible component of this signal is summer meltwater (which could have caused large negative excursions in the $\delta^2 H_{C30acid}$ 479 480 values, such as those observed between ~19,000 and 18,000 cal BP). The significant expansion of the EASM system at 16,000 cal BP has critical implications for other records from the region, 481 particularly those derived from more northerly latitudes, where the influence of the westerly jet 482 483 migration is more pronounced (Nakagawa et al, 2021). Careful consideration of the location of the 484 monsoon front at this time is vital for robust interpretation of proxy changes and whether they indeed represent EASM evolution or, rather, non-monsoonal summer precipitation. 485

In conclusion, our findings provide centennial-scale reconstructions which highlight the distinctive seasonal behaviours of the EAM during TI, as well as novel insights into the spatially- and temporally- constrained teleconnections acting on the climate of the EAM region during this interval. Our records are unique in their reconstruction of both seasonal EAM modes in Japan from a single

archive. These results may begin to explain the observed heterogeneity between site-specific
 reconstructions of EAM behaviour during deglaciation.

492

493 <u>Methods</u>

494 Core Materials

Materials for this study were subsampled from the SG12 core, which was taken from the 495 centre of Lake Suigetsu (35° 35' 08"N, 135° 52' 56"E) and spans a total composite depth of 41.8 m 496 497 (from the present day to 56,100 cal BP). The core consists of a series of overlapping ~90 cm sections from four boreholes. SG12 is a direct stratigraphic copy of the SG06 core (Nakagawa et al., 2012) 498 due to the proximity of the SG12 and SG06 coring locations (the first borehole of each coring 499 campaign was made within 15 m of each other, and both at a water depth of 34 m). As with the 500 501 temporally equivalent sediments from the SG06 core, the SG12 sediment is varved throughout and 502 represents continuous sedimentation (Schlolaut et al., 2018). Chronological control for the SG12 cores is given by correlation to the SG06 cores via distinct visible marker layers. The SG06 chronology 503 504 (Bronk Ramsey et al., 2020; Staff et al., in review) is based on >800 radiocarbon dates for the past ~50 ka BP (Staff et al., 2011; Bronk Ramsey et al., 2020), thin section microscopic varve counts 505 506 (Schlolaut et al., 2018) and tephra tie points (McLean et al., 2018); all ages are presented on the 507 IntCal20 timescale (Reimer et al., 2020).

Subsamples of the SG12 core for this study were taken from 1.2 cm-wide LL-channels (Nakagawa *et al.*, 2012) from the longitudinally cut core across the composite depth (ver. 27 December 2022) range of 1274.8 – 2119.4 cm (9980 ± 30 cal BP to 22,040 ± 38 cal BP). This time period was selected to encompass the maximum range of the existing Lake Suigetsu pollen-derived temperature reconstruction for TI. The LL-channels were cut to produce contiguous (continuous adjacent) subsamples each containing 100 integrated years of sedimentation (based on the SG06 2012 timescale). Sample ages were subsequently converted to the IntCal20 timescale. A contiguous 515 integrated sampling approach was taken to avoid subsampling time periods associated with peaks or troughs of multidecadal cycles, superimposed on the multicentennial and multimillennial scale 516 transitions of interest (Nakagawa et al., 2021). The sampling interval of ~100 years was selected to 517 facilitate the detection of finer resolution variability within the key climatic transitions of the period. 518 The (minimal) core expansion during storage was linearly modelled and accounted for by comparing 519 520 the difference in event layer depth at the time of subsampling to high resolution photographs taken 521 immediately after core extraction. Event layers (floods, turbidites and tephra) were removed during sampling to prevent skewing of the integrated signal by inclusion of a larger quantity of material 522 from a single event. Particular care was taken to avoid contaminating the $\delta^2 H_{acid}$ samples with 523 modern organic compounds. Surfaces and instruments were regularly cleaned with ethanol. 524

Each subsample was split into two, with each half designated for $\delta^{18}O_{diatom}$ (n = 120) and 525 $\delta^2 H_{acid}$ (n = 120) analysis, respectively. The exposed surfaces of one half (for $\delta^2 H_{acid}$ analysis) were 526 527 scraped with a clean instrument prior to splitting to remove the outer sediments containing potential contamination from past handling. Wet weights ranged from 1.8 - 10.3 g (subsamples for 528 δ^{18} O_{diatom} analysis) and 1.1 - 7.1 g (subsamples for δ^{2} H_{acid} analysis). The largest samples were from 529 the younger end of the section, where the sedimentation rate (and thus depth range equating to 530 531 ~100 years of integrated time) was high, and the smallest samples were from the older end of the 532 section, where the sedimentation rate was relatively low. All subsamples were frozen and then 533 freeze dried for 24 hours to obtain dry sediment masses before subsequent preparation steps.

534

535 $\delta^{18}O_{diatom}$ Analysis

536 Subsamples for diatom oxygen isotope analysis were prepared following standard 537 procedures (Swann and Snelling, 2023). Specifically, subsamples were disaggregated overnight in 538 30 % H₂O₂ prior to three successive heavy liquid density separations using sodium polytungstate 539 (SPT) at specific gravities of 2.25 g cm⁻³, 2.20 g cm⁻³ and 2.15 g cm⁻³, removing the denser material 540 each time. The subsamples were subsequently treated with 30 % H₂O₂ at 70 °C for one week to remove any remaining organic material, followed by 5 % HCl for 12 hours to remove carbonates. 541 542 Subsamples were then filtered using an 80 µm nylon mesh to remove large sponge spicules and large diatoms and freeze dried ready for analysis. As with other studies, further taxa-specific 543 separation was not performed beyond this sieve stage due to the similar size of many diatom taxa 544 545 present in the samples, rendering physical separation techniques impossible. Qualitative visual 546 purity checks were made using light microscopy at x800 magnification. The %Al₂O₃ for each subsample was quantified using a Malvern Panalytical Epsilon 3 X-Ray Fluorescence (XRF) 547 spectrometer. Due to elevated %Al₂O₃ values in the majority of samples, a Zeiss Sigma 300 VP-FEG 548 Scanning Electron Microscope (SEM) was used to visually identify the source of this aluminium in 549 five samples of varying %Al₂O₃ (Fig. 4). Samples with %Al₂O₃ exceeding 4.2 % (Al/Si = 0.05) showed 550 551 particulate Al-bearing contamination (most likely a detrital Al-rich silicate, but undetermined due to 552 grain size) which was external to the diatom frustules. These samples (n = 3) were subjected to an additional SPT density separation at 2.15 g cm⁻³ to remove this contamination. Samples with %Al₂O₃ 553 554 below 4.2 % showed only small amounts (on a sub-micron scale) of this Al-bearing contamination adhered to or trapped within the diatom frustules (Fig. 4). Additional density separations would not 555 remove this contamination due to electrostatic charges adhering the contamination to the frustule, 556 557 and air pockets within the cylindrical frustules preventing chemical treatment of internal 558 contamination (Brewer et al., 2008), so a mass balancing approach was taken to correct for the effect of this contamination on the $\delta^{18}O_{diatom}$ values (discussed below). 559



560

561Fig. 4: Diatom SEM Imagery. SEM images for subsample 3D (left) and sample 65D (right) showing Al-bearing562contamination adhered to frustules and inside cylindrical Aulacoseira spp., which are representative of the 120563analysed subsamples for $\delta^{18}O_{diatom}$ analysis. Scale bars are indicated on each image.

The purified diatom material (6 – 7 mg subsamples) was subjected to stepwise fluorination 565 566 in nickel reaction vessels to liberate structural oxygen following Leng and Sloane (2008) at the British Geological Survey (UK). Briefly, this method involved outgassing samples at 250 °C to remove any 567 water in the sample, "pre-fluorination" at 250 °C using a stoichiometric deficiency of BrF₅ to remove 568 the exchangeable hydrous silica layer from the diatom frustules and minimise exchange effects, and 569 then fluorination with BrF5 overnight at 500 °C. The resulting oxygen gas was cryogenically purified 570 571 and then converted to CO₂ by exposure to hot platinized graphite (Clayton and Mayeda, 1963). The 572 average gas yield of this process was between 68 and 75 %. In each batch of diatom samples, an inhouse laboratory diatomite standard, BFC, with a known value of δ^{18} O (Chapligin *et al.*, 2011) was 573 also reacted and converted to CO₂ in the same manner following the principle of identical treatment. 574 The oxygen isotope ratio of the diatom subsamples ($\delta^{18}O_{diatom}$) and BFC ($\delta^{18}O_{BFC}$) were measured 575

576 relative to a reference CO₂ gas on a Thermo Finnigan MAT 253 dual inlet isotope ratio mass spectrometer. Oxygen isotope ratios were calibrated using a single-point anchoring procedure by 577 analysing CO₂ prepared from an in-house standard (MCS; $\delta^{13}C_{VPDB} = -0.7$ ‰ and $\delta^{18}O_{VPDB} = -9.2$ ‰) 578 via reaction with H₃PO₄ (specific gravity = 1.92) at 25°C. All $\delta^{18}O_{VPDB}$ values were converted to the 579 VSMOW scale using Coplen *et al.* (1983) and normalised such that the $\delta^{18}O_{VSMOW}$ of the within-run 580 581 BFC = +28.9 ‰. The oxygen isotope composition of BFC has been calibrated via inter-laboratory comparison (Chapligin *et al.*, 2011) using the certified international standard NBS 28 ($\delta^{18}O_{VSMOW}$ = 582 +9.57 ‰; Hut, 1987; Gröning et al., 2007). All oxygen isotope data are expressed in standard delta 583 (δ) notation in per mille (∞) deviations relative to VSMOW. External analytical error (1 σ) was ±0.13 ∞ 584 for BFC and ±0.33 ‰ for replicate analysis of the diatom samples. 585

The $\delta^{18}O_{\text{diatom}}$ values were corrected for the aforementioned Al-bearing silicate 586 587 contamination using a geochemical mass balance approach (Mackay et al., 2011; Swann et al., 2018). 588 End member contamination samples for Lake Suigetsu contained 19.45 % Al₂O₃ (from XRF, n = 2), a δ^{18} O composition of 10.36 ± 0.68 ‰ (1 σ range, n = 8) and 48.93 wt% oxygen (from XRF, n = 2). These 589 590 were prepared from raw materials (n = 2) and the residues of SPT separations from the $\delta^{18}O_{diatom}$ subsamples (n = 6), disaggregated overnight in 30 % H_2O_2 and treated with 30 % H_2O_2 at 70 °C for 591 one week to remove any remaining organic material, followed by 5 % HCl for 12 hours to remove 592 593 carbonates and 8 % NaOH at 70 °C for 48 hours to remove biogenic silica. Contamination-corrected $\delta^{18}O_{diatom}$ values were modelled using the %Al₂O₃ for each sample, the end member quantities above, 594 and assuming a %Al₂O₃ of pure diatom material of 1.4 % (Al/Si = 0.016), the minimum value 595 measured across the 120 samples. The uncertainty associated with mass-balancing was calculated 596 assuming a normal distribution for data uncertainty and Monte Carlo simulations (10,000 replicates). 597 Errors presented on Fig. 2 and Fig. 3 show the 1σ range of these simulations (mean = ± 0.36 %). Of 598 599 the 120 samples prepared, 113 datapoints are included in the final dataset. Five samples were excluded due to elevated %Al₂O₃ values (>4.2%; Al/Si = 0.05); these produced anomalously high 600

values during mass balancing, possibly due to atypically large quantities of diatom-bound Al which was unaccounted for by our methods. A further two samples were too small in mass for accurate $\delta^{18}O_{diatom}$ determination via the methods presented above.

604

605 $\delta^2 H_{acid}$ Analysis

Subsamples for $\delta^2 H_{acid}$ analysis were prepared following the Biomarkers for Environmental 606 607 and Climate Science (BECS) group standard protocol at the University of Glasgow (UK). The total lipid extract (TLE) was extracted by a Dionex ASE 350 Accelerated Solvent Extractor using 608 dichloromethane and methanol (9:1, v:v) and the TLE dried and weighed. The TLE was then 609 separated into neutral and acid fractions using solid phase extraction through a LC-NH₂ silica gel 610 column. The neutral fraction was eluted using dichloromethane:propan-2-ol solution (1:1, v:v) and 611 612 the acid fraction using 4 % acetic acid in diethyl ether. The acid fraction was derivatised using 100 613 µL of 12 % boron trifluoride in methanol in sealed glass vials at 70 °C for 60 minutes. This converted n-alkanoic acids into fatty acid methyl esters (FAMEs) in preparation for analysis. These FAMEs were 614 615 then cleaned through a silica gel column (35 – 75 μm particle size). Non-FAME material was eluted using hexane and the FAMEs eluted using dichloromethane. Subsamples were prepared in batches 616 617 of 10 or 11, each with a procedural blank to check for contamination introduced by the stages 618 described above.

Identification of the C₁₆, C₁₈, C₂₈ and C₃₀ n-alkanoic acids (as FAMEs) was performed on subsample 96B using an Agilent 7890B Gas Chromatograph connected to a 5977A mass spectrometer detector (MSD) with an electron impact (EI) ionisation source (GC-MS). A HP1-MS capillary column was used (60 m length, 0.25 mm internal diameter, 0.25 μ m film thickness) with helium carrier gas at a constant flow rate (1.2 mL min⁻¹). The FAMEs were dissolved in DCM (200 μ L) and a volume of 1 μ L used in a splitless injection. The inlet was kept at 315 °C and the following oven programme used: the oven was held for two minutes at 60 °C and raised to 120 °C at 30 °C min⁻¹,

626 then increased to 310 °C at 5 °C min⁻¹ and held for 33 minutes. The auxiliary temperature in the 627 transfer line between the GC and MSD was kept constant at 315 °C. The MS source was set to 230 628 and the MS quad set to 160. The detector analysed all compounds between 12- and 65-minute retention time, except for the period between 34 and 39 minutes where flow was redirected to 629 avoid a large phthalate compound peak from entering the detector. Data acquisition was performed 630 in full scan mode at 2.4 scans s⁻¹. Identification of contaminant peaks was also performed using this 631 method. All large contaminant peaks (retention times: ~24.5, ~33 and ~37 minutes) were shown to 632 633 be phthalate compounds. The largest of these (at ~33 minutes) was present in > 90 % of subsamples, however the others were only present in ~ 10 %. No contaminant peaks obscured the FAME peaks 634 of interest. 635

The concentration of the C₁₆, C₁₈, C₂₈ and C₃₀ n-alkanoic acids (as FAMEs) in each subsample 636 637 were measured using an Agilent 7890B Gas Chromatograph fitted with a flame ionisation detector (GC-FID). A Restek Rtx-1 capillary column was used (60 m length, 0.25 mm internal diameter, 0.25 638 µm film thickness) with helium carrier gas at a constant flow rate (1.2 mL min ⁻¹). FAMEs were 639 640 dissolved in DCM (200 µL - 1250 µL; depending on FAME yield) and a volume of 1 µL used in a splitless injection. The inlet was kept at 320 °C and the following oven programme used: the oven 641 was held for two minutes at 60 °C and raised to 120 °C at 30 °C min⁻¹, then increased to 330 °C at 5 642 °C min⁻¹ and held for 15 minutes. The FID temperature was set to 250 °C. A chromatogram showing 643 644 a representative sample with no phthalate peaks is shown in Supplementary Fig. 4. Peak areas were measured, and concentrations calculated from a set of external calibrations using a standard mix of 645 eleven n-alkanes; ten straight-chained (C16, C18, C19, C23, C25, C26, C28, C30, C32 and C37 n-alkanes) and 646 one branched (squalane). Calibration graphs were made using concentrations of 2.5 µg/mL, 5 µg/mL, 647 648 and 10 µg/mL. A calibration for the 0 - 25 minute, 25 - 35 minute and 35 - 60 minute retention time 649 intervals were made using the C16, C29 and C39 homologues in the standard, respectively. An Rsquared value of >0.99 was achieved for each calibration. The n-alkanoic acid (as FAME) 650

651 concentrations were then normalised to dry sediment weight. Variations in n-alkanoic acid 652 concentrations are shown in Supplementary Fig. 1.

653 The compound specific hydrogen isotopic composition (δ^2 H) of the C₁₆, C₁₈, C₂₈ and C₃₀ nalkanoic acids (as FAMEs) were measured using an Agilent 7890B gas chromatography (GC) system 654 connected to an Elementar GC5 furnace and IRMS. The settings of the GC system were identical to 655 those of the GC-FID outlined above with the difference being that the FAMEs were dissolved in 656 between 30 and 120 µL of hexane and the injection volume set to either 1 or 2 µL depending on 657 658 FAME yield. This ensured that FAMEs were measured as a similar response to the reference H₂ gas. Due to the presence of the largest phthalate compound (retention time ~33 mins) in >90 % of the 659 subsamples, flow was redirected between 1700 s and 1975 s to prevent entry into the IRMS. In 13 660 subsamples, this was extended to the interval 1400 s to 2100 s to remove the second largest 661 662 phthalate peak (retention time ~37 minutes). The smallest phthalate peak (retention time ~24.5 663 minutes) was not larger than the FAME peaks and hence did not require redirection of flow. The 664 furnace temperature was set to 1450 °C with an interface temperature of 350 °C. Subsamples were 665 measured in duplicate and δ^2 H values of each homologue were calculated relative to a calibrated reference H₂ gas. These values were then converted to the VSMOW scale by linear regression to an 666 667 in-house standard (containing C₁₆, C₁₉, C₂₃, C₂₅, C₂₈ and C₃₂ straight chained n-alkanes in addition to squalane) calibrated to the standard Indiana n-alkane mixture B5 containing C₁₆ to C₃₀ n-alkanes. 668 669 The in-house standard was measured four times prior to each batch, after every eight sample measurements, and in duplicate at the end of each batch. All hydrogen isotope data are expressed 670 in standard delta (δ) notation in per mille (∞) deviations relative to VSMOW. Instrument precision 671 was ±2.53 ‰ based on the standard deviation of all standard measurements. The H₃⁺ correction 672 673 factor ranged from 4.24 to 4.57.

674 Reliable δ^2 H values for all chain lengths across all subsamples was not achieved due to some 675 subsamples exhibiting raised baselines resulting in poorly defined peak areas and inaccurate

676 determination of δ^2 H for at least one homologue (affecting 26 subsamples). Raised baselines in some samples predominantly precluded accurate measurement of the aquatic chain lengths, 677 though it was possible to accurately measure the C₂₈ and C₃₀ homologues in many of the impacted 678 subsamples. Low concentrations affected the repeatability of the measurements of $\delta^2 H_{acid}$ of at least 679 one homologue in a further 9 subsamples (most commonly C₁₆ or C₁₈), hence these values were 680 681 excluded from the final dataset. The mean precision of the $\delta^2 H_{acid}$ measurements was ± 2.0 ‰ (1 σ range). The resulting dataset has 95 datapoints for the C₁₆, 86 for the C₁₈, 112 for the C₂₈ and 108 682 for the C₃₀ n-alkanoic acids. Terrestrial and aquatic composites were not generated due to 683 insufficiently strong statistical relationships between the $\delta^2 H_{C28acid}$ and $\delta^2 H_{C30acid}$ values, and the 684 $\delta^2 H_{C16acid}$ and $\delta^2 H_{C18acid}$ values, although there was som visual correlation between the two pairs. 685 Values were corrected for the methylation process (which removes the exchangeable carboxylic 686 acid hydrogen from each molecule and replaces it with a methyl group with three hydrogens) 687 688 (Chivall *et al.*, 2012). GC-IRMS measurements of (Z)-hexadec-9-enoic acid (δ^2 H = -154.02 ‰) and methyl (Z)-hexadec-9-enoate (δ^2 H = -143.13 ‰) were made to calculate the δ^2 H value of a single 689 methanol-derived methyl hydrogen ($\delta^2 H = -37.86 \%$), and a mass balance scheme (Chivall *et al.*, 690 2012) applied to correct the measured FAME δ^2 H values to n-alkanoic acid values (excluding the 691 692 exchangeable hydrogen on the carboxylic acid group).

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701 Data Availability

Accompanying data can be found at <u>http://doi.org/10.5525/gla.researchdata.1439</u>.

703

704 Supplementary Information

705 Supplementary Discussion 1

706 The total lipid extract (TLE) exhibited low values in the early part of the section ($\sim 2 \text{ mg/g}$) 707 before increasing gradually from ~15,500 cal BP (Supplementary Fig. 1). Values were then high (~6 708 mg/g) and fluctuating. This follows the pattern of sediment total organic carbon (TOC) during this period (Tyler et al., 2010). Aquatic n-alkanoic acid concentrations did not show this long-term 709 increasing trend, and the concentration profiles for the C₁₆ and C₁₈ n-alkanoic acids showed 710 711 centennial-scale fluctuations throughout (Supplementary Fig. 1). Conversely, the terrestrial n-712 alkanoic acid concentrations were higher in the latter part of the section; the concentration of the 713 C₂₈ n-alkanoic acid showed a greater increase than the C₃₀, but also exhibited larger fluctuations (Supplementary Fig. 1). Throughout, the terrestrial n-alkanoic acid concentrations were generally 714 715 higher than those of the aquatic n-alkanoic acids, aside from in a small number of samples. The close 716 relationship between the terrestrial n-alkanoic acid concentrations and the TLE concentrations 717 suggests that the terrestrial component was driving the observed TLE increase (allowing for the un-718 resolved non-acid fractions which also contribute but were not measured). The gradual increase in 719 TLE, TOC and terrestrial n-alkanoic acid concentration was similar in character to the changes in 720 EAWM and EASM strength and very different from the temperature reconstruction for the site (Main Text), suggesting that they were driven by precipitation changes rather than temperature 721 changes. None of these variables show ACR-type inversions, however the seasonality of these 722 variables suggests a relationship to the EASM is more likely than a link to the EAWM, despite the 723 724 EAWM showing more gradual changes during this interval. Diatom frustule concentration also increased through the section (Supplementary Fig. 1), but the shift to higher values initiated ~500 725

cal BP later (~15,000 cal BP) and more abruptly than the TLE concentration. Hence, whilst an overall resemblance between frustule concentration and TOC can be argued, there was some decoupling of these variables between ~16,000 and ~15,000 cal BP. The sudden increase in diatom frustule concentration at ~15,000 cal BP was likely driven by temperature changes, in light of the abruptness of this shift and the coincident timing with the onset of the Late Glacial Interstade (Main Text). Aulacoseira dominated the diatom assemblage counts, most notably at 21,500 cal BP, 20,000 cal BP and between 15,000 and 12,000 cal BP (Supplementary Fig. 1). Spikes of Naviculoid and benthic taxa were observed at 21,000 cal BP. Between 20,000 and 15,000 cal BP there were greater proportions of Stephanodiscus, Naviculoid and benthic taxa relative to Aulacoseira, and similarly a period of increased Asterionella between 12,000 and 10,000 cal BP.



Proxy weights, abundances and concentrations. Variations in total organic carbon (TOC; Tyler *et al.*, 2010),
 mass-normalised total lipid extract (TLE), mass normalised n-alkanoic acid concentrations (aquatic (C₁₆ and C₁₈) and terrestrial (C₂₈ and C₃₀)), diatom frustule concentration (Saito-Kato *et al.*, unpub) and diatom taxa
 abundance (Saito-Kato *et al.*, unpub) during TI in the Lake Suigetsu sediment cores.



760Comparison of Lake Suigetsu $\delta^{18}O_{diatom}$ and Lake Suigetsu diatom taxonomic counts. Diatom taxonomic761counts show varying abundances of seasonally assigned genera (*Aulacoseira* and *Encyonema*, a benthic762species) versus other taxa. Intervals associated with high *%Aulacoseira* are interpreted as a proxy of EAWM763strength. The data presented here supports EAWM weakening during Termination I.

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765 Supplementary Discussion 2

766 By considering the numerical difference between the $\delta^2 H_{C30acid}$ and $\delta^2 H_{C16acid}$ values (Supplementary Fig. 3), it is possible to semi-quantify the relative influence of the EAWM and EASM 767 on the $\delta^2 H_{C16acid}$ values (which reflects a combination of the $\delta^2 H_{precipitation}$ of both seasons; Main Text). 768 Whilst traditionally used to consider the effect of evaporation on the aquatic $\delta^2 H$ signal (because 769 770 the terrestrial δ^2 H is not affected by lake evaporation, unlike aquatic δ^2 H), under the circumstances presented here (and expanded upon in the main text), we interpret this measure to represent the 771 balance between EAWM and EASM influence, based on the inference that the $\delta^2 H_{\text{C16acid}}$ values 772 represent a seasonal mix, and the $\delta^2 H_{C30acid}$ values as seasonally constrained to summer. Following 773 this logic, the numerical difference between these measures indicates increasing EASM influence on 774

aquatic δ^2 H during TI (Supplementary Fig. 3); larger values indicate greater EASM influence, and smaller values indicate greater EAWM influence (based on the assumption that EAWM δ^2 H_{precipitation} is higher than EASM δ^2 H_{precipitation}). However, it is inappropriate to apply this interpretation to the ~19,000 – 18,000 cal BP interval, where there was a significant amount of scatter in the δ^2 H_{C30acid} values (discussed in the main text). A reduction in lag time with warming temperatures remains a component of this signal.

- 781
- 782 Supplementary Figure 3



Changing EASM influence on $\delta^2 H_{acid}$ **at Lake Suigetsu.** Comparison of Lake Suigetsu $\delta^2 H_{C16acid}$, $\delta^2 H_{C30acid}$, and the numerical difference between these quantities during TI in the Lake Suigetsu sediment cores. $\delta^2 H_{C30acid} - \delta^2 H_{C16acid}$ is interpreted as an indicator of EASM versus EAWM influence, with a component of a reduced lag time with warming temperatures.

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