

1 **Orbital-scale global ocean sea surface temperatures coupling with cryosphere-carbon**  
2 **cycle changes over the past 4 million years**

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19 **Key Points:**

- 20
- Orbital forcing of global SSTs evolution
  - Enhancement of the obliquity signal in the global SSTs at ~2.7 Ma is closely associated  
21 with the NH glaciation
  - Changes in SST may drive processes of ocean-atmosphere carbon exchange that alter  
22 atmospheric CO<sub>2</sub> concentrations
- 23  
24  
25

## 26 **Abstract**

27 Changes in the thermal conditions of the ocean surface, the interface for air-sea exchange, are  
28 critical for understanding global climate and environmental change. Here we explore the  
29 evolution of sea surface temperature (SST) and the meridional sea surface temperature gradient  
30 (STG) at orbital timescales since 4 million years ago (Ma), along with interactions between  
31 SSTs, the cryosphere, and the global carbon cycle. We observe orbital eccentricity and obliquity  
32 influences on SST evolution and infer that SST changes may have played a key role in  
33 atmospheric CO<sub>2</sub> and cryosphere changes through key climate transitions in the past 4 Ma. We  
34 find a major equator-to-pole STG increase in the Northern Hemisphere (NH) close to the  
35 initiation of major NH glaciation (at ~2.7 Ma). In addition, we find substantial increases in the  
36 obliquity sensitivity ( $S_{obl}$ ) of NH STG at ~2.7 Ma and in Southern Hemisphere (SH) STG at ~1  
37 Ma, which may be responses to important expansions of NH and SH ice sheets, respectively.  
38 Phase analysis shows that SST changes typically lead global ice volume changes throughout the  
39 last 4 Ma. SST changes also lead atmospheric CO<sub>2</sub> changes since ~1.5 Ma, which indicates that  
40 SST changes either drove, or directly reflect, processes that changed ocean-atmosphere carbon  
41 exchange and, thus, atmospheric CO<sub>2</sub> concentrations. Overall, our study emphasizes that SST  
42 changes were a critical component of climate change throughout the last 4 Ma.

## 43 **Plain Language Summary**

44 A compilation of global sea surface temperature (SST) records indicates that SST mainly  
45 fluctuated on orbital eccentricity and obliquity timescales during the last 4 million years. The  
46 compilation also reveals an important impact of meridional ocean circulation changes on global  
47 climate since 4 Ma, with particularly notable change in the equator-to-pole SST gradient during  
48 the initiation of major Northern Hemisphere glaciations at ~2.7 Ma. Finally, we find that SST  
49 changes played a key role in the atmospheric CO<sub>2</sub> changes associated with global climate  
50 transformations.

51

## 52 **1 Introduction**

53 The oceans play a fundamental role in the climate system due to their enormous heat-  
54 storage capacity (Deser et al., 2010), and sea surface temperature (SST) changes since the early  
55 Pliocene are thought to have been a potential cause of the onset of NH glacial cycles (Fedorov et  
56 al., 2013). In addition, SST evolution profoundly affects atmospheric circulation patterns  
57 (Brierley et al., 2009; Fedorov et al., 2013), as the ocean can transfer energy to the atmosphere  
58 through turbulence and radiative energy exchange at the sea surface (Deser et al., 2010). As  
59 such, SST assessment is crucial for understanding the evolution of Earth's climate system and the  
60 mechanisms of climate change. The emergence of increasing numbers of SST records based on  
61 geochemical proxies, such as the alkenone unsaturation index ( $U^{K}_{37}$ ) and foraminiferal Mg/Ca  
62 ratios, has greatly improved understanding of global long-term SST evolution since the Pliocene  
63 (Liu & Herbert, 2004; Lawrence et al., 2006; Lawrence et al., 2009; O'Brien et al., 2014; Max et  
64 al., 2020). However, it remains unclear how SST affects other climate processes under changing  
65 geological boundary conditions, such as different atmospheric CO<sub>2</sub> concentrations (Fedorov et  
66 al., 2015).

67 The temperature of the ocean surface, the interface of air-sea exchange, is controlled by a  
68 combination of atmospheric and oceanic processes. The air-sea temperature contrast and wind

69 velocity dominate air-sea heat exchange, mainly in the form of latent and sensible heat (Cayan et al., 1992). Ocean circulation (Dowsett et al., 2009), vertical mixing (Hasenfratz et al., 2019), and thermocline depth (Wara et al., 2005) also significantly affect SST. SST records from different latitudes (Fig. 1) show large-scale variations with time similar to those in records of the oxygen isotope composition of benthic foraminifera ( $\delta^{18}\text{O}_{\text{benthic}}$ ) and atmospheric  $\text{CO}_2$  concentrations, but substantial differences exist at more detailed levels (Fig. 2).  $\delta^{18}\text{O}_{\text{benthic}}$  is a well-established proxy for the evolution of deep-sea temperatures and seawater  $\delta^{18}\text{O}$  ( $\delta^{18}\text{O}_{\text{sw}}$ ), the latter of which is related to global ice volume (Fedorov et al., 2013; De Vleeschouwer et al., 2020; Rohling et al., 2021). Hence, similarities between large-scale variations in  $\delta^{18}\text{O}_{\text{benthic}}$  and SST reflect the global nature of glacial-interglacial cycles, while differences reflect leads, lags, and amplitude response differences that may hold clues about the nature of the processes by which they are connected.

81 Here we focus on three issues that remain poorly explored within the past 4 million years, namely: (1) global-scale SST evolution at orbital time scales, (2) the evolution of the meridional sea surface temperature gradient (STG) between the tropics, extra-tropics, and polar regions, and (3) the relationships between SST, global ice volume, and atmospheric  $\text{CO}_2$ . We investigate the role of meridional ocean circulation changes in some of the major climate events of the last 4 Myr. The first event concerns the initiation of major NH glaciation (NHG) during the Late Pliocene-Early Pleistocene (LP/EP, ~3-2.7 Ma), when the NH transitioned from warm, virtually ice-free climatic conditions to a cold, persistent ice age climate, marked by an emerging dominance of 41-kyr obliquity cycles in  $\delta^{18}\text{O}_{\text{benthic}}$  records (Lisiecki & Raymo, 2005; Westerhold et al., 2020). The second event concerns the mid-Pleistocene transition (MPT, ~1.2-0.7 Ma), when dominant 41-kyr obliquity cycles gave way to high-amplitude 100-kyr eccentricity cycles in  $\delta^{18}\text{O}_{\text{benthic}}$  records (Lisiecki & Raymo, 2005; Raymo et al., 2006; Westerhold et al., 2020).

## 94 **2 Materials and Methods**

### 95 **2.1 Data sources**

96 The International Ocean Discovery Program (IODP) and its predecessors provided the high-quality sediment sequences used in generation of a valuable set of Cenozoic climate and sea surface temperature (SST). We use alkenone unsaturation index ( $U^{K'}_{37}$ ) based SST records for Site 982 from the North Atlantic (58°N, 16°W; Lawrence et al., 2009), Site 607 from the North Atlantic (41°N, 33°W; Lawrence et al., 2010), Site 1012 from the North Pacific (32°N, 118°W; Brierley et al., 2009), Site 722 from the Arabian Sea (16.6°N, 59.8°W; Herbert et al., 2010), Site 846 (3°S, 91°W; Liu and Herbert, 2004; Lawrence et al., 2006), Site 1239 (1°S, 82°W; Etourneau et al., 2010) from the Equatorial Pacific, Site 662 from the Equatorial Atlantic (1°S, 12°W; Herbert et al., 2010), Site 709C from the Indian Ocean (3°54.9'S, 60°33.1'E; Karas et al., 2011), Site 1237 from the South Pacific (16°S, 76°W; Dekens et al., 2007), Site 1082 (21°S, 12°E; Etourneau et al. 2009) from the Southern Ocean, Site 1090 (43°S, 9°E; Martínez-García et al. 2010) from the Subantarctic, and Site 1094 (53.2°S, 5.1°E; Hasenfratz et al., 2019) from the Antarctic Zone. The age models for the sites used in this study, with exception of Site 1237, are based on orbital tuning of deep-sea oxygen isotope records (LR04 stack; Lisiecki & Raymo, 2005; see Table S1 for details). The  $U^{K'}_{37}$  index and foraminiferal Mg/Ca are widely used to reconstruct ocean temperatures. Use of the  $U^{K'}_{37}$  proxy is limited in warm tropical regions, where the  $U^{K'}_{37}$  index can saturate at temperatures >29°C (Li et al., 2011; Fedorov et al., 2013). We

113 include  $U^{K}_{37}$  based SSTs for three tropical sites (Sites 662, 846 and 1239; Supplementary Fig.  
114 S1), given that values remained below that 29°C limit (Fig. S1).

115 Atmospheric CO<sub>2</sub> concentrations are obtained from reconstructions based on leaf wax  $\delta^{13}C$   
116 at IODP Site U1446 (0-1.46 Ma; Yamamoto et al., 2022) and foraminiferal  $\delta^{11}B$  (0-4 Ma; Fig.  
117 2o; Hönisch et al., 2009; Bartoli et al., 2011; Martínez-Botí et al., 2015; Stap et al., 2016). The  
118 IODP Site U1446 leaf wax  $\delta^{13}C$  record has a resolution of ~1.7 kyr, which we use for orbital-  
119 scale phase analysis. Its age model is derived from oxygen isotope tuning to the global  $\delta^{18}O_{\text{benthic}}$   
120 (LR04) stack by Clemens et al. (2021). The  $\delta^{18}O_{\text{sw}}$  record in Fig. 2b has been derived from the  
121 CENOGRID  $\delta^{18}O_{\text{benthic}}$  record of Westerhold et al. (2020) by Rohling et al. (2021), and has a 1  
122 kyr resolution. The resolution of the collated datasets makes them suitable for analyzing changes  
123 in Earth's climate system at orbital time scales.

124 Meridional SST gradients (STG) are created by calculating the difference in SSTs between  
125 2 ocean cores from different latitudes in the same ocean. For this study, we take these calculated  
126 STGs to be representative of the basin-wide situation as suggested by previous studies (Wara et  
127 al., 2005; Brierley et al., 2009). Due to differences between the temporal resolutions of the  
128 different SST records, we interpolated SST records from the different sites to 2 kyr spacing and  
129 then calculated the gradients. The North Atlantic STG is determined between ODP sites 982 and  
130 662; the North Pacific STG is that between ODP Sites 846 and 1012; the South Atlantic STG  
131 uses ODP sites 1082 and 1090; and the South Pacific STG is the difference between ODP Sites  
132 1239 and 1237.

133

## 134 2.2 Spectral Analysis

135 SST power spectra and sliding window spectrograms were calculated using Acycle 2.6 (Li  
136 et al., 2019). All records were detrended by fitting and removing LOESS functions (35%). SST  
137 power spectra were analyzed using the  $2\pi$ -MultiTaper Method (MTM) with robust red noise  
138 modeling to ascertain 95% confidence levels. Eccentricity and obliquity cycles in the SST  
139 records were extracted using Gaussian band-pass filtering in Acycle 2.6 (Li et al., 2019).

140

## 141 2.3 Obliquity sensitivity

142 Obliquity sensitivity ( $S_{\text{obl}}$ ) is calculated as  $\sigma^2_{\text{STG}}/\sigma^2_{\text{La04}}$ , where  $\sigma^2_{\text{STG}}$  is the temporal variance  
143 of obliquity in the STG in units of °C<sup>2</sup>, and  $\sigma^2_{\text{La04}}$  is the temporal variance of obliquity in units of  
144 degrees<sup>2</sup>, taken from the Lasker et al. (2004) orbital solution (La04). Obliquity variance was  
145 quantified using multitaper time-frequency power spectra by integrating the variance between  
146 0.023 and 0.027 cycles/kyr (i.e., from ~43.5 kyr to 37 kyr). All time-frequency analyses were  
147 performed using three  $2\pi$  prolate data tapers, with a 400-kyr window and a 10-kyr time step. The  
148 analysis was carried out in R and the R code was modified after Levy et al. (2019).

149

## 150 2.4 Phase analysis

151 SST- $\delta^{18}O_{\text{sw}}$  and SST-CO<sub>2</sub> phase relationships were calculated using cross-spectral analysis  
152 in the IRISseismic package in R (Callahan et al., 2018). To visualize results, we invert  $\delta^{18}O_{\text{sw}}$   
153 to make it positively correlated with sea level. Our study focuses on phase relationships between  
154 SST,  $\delta^{18}O_{\text{sw}}$ , and CO<sub>2</sub> on eccentricity scales. We calculated cross spectra using a 400-kyr sliding  
155 window and a 10-kyr step size in individual datasets. Phase analysis calculated at the ~100-kyr  
156 eccentricity scale was chosen for a 90-135 kyr period range with the highest period coherence.  
157 The phase analysis was carried out in R and the R code was modified after De Vleeschouwer et  
158 al. (2020).

## 2.5 Recurrence analysis

Non-linear structure within proxy time series can help to reveal regime/system changes that cannot be validated otherwise by visual inspection or linear methods like spectral analysis (Marwan et al., 2007; Han et al., 2020). Recurrence analysis is used to identify transitions between different types of dynamics within a time series. Recurrence plots are matrix plots that visualize a fundamental property of dynamical systems – namely, when a system “repeats” itself, returning to a previous state (See Text S1 for a detailed description). Recurrence plots are a binary matrix where the coordinates of each entry mark the pair of time points with recurring states (Marwan et al., 2007). If climate dynamics have consistent patterns, they will show up as darker areas in the plot; if they have no common dynamics, the plot will remain white (Marwan et al., 2007; Westerhold et al., 2020). We conducted recurrence analyses of non-detrended SST data in Matlab’s CRP Toolbox (Marwan, 2020).

## 3 Results and Discussion

### 3.1 Combined high- and low-latitude forcing of regional SSTs since 4 Ma

There is a strong statistical relationship between  $U^{K'}_{37}$  and mean annual SST in many regions (Rosell-Melé, 1998; Müller et al., 1998); hence its use as an SST proxy. However, many studies have shown that coccolithophorid productivity (i.e., the main source of alkenone) is highest in subpolar ocean regions during the summer to autumn months (Samtleben & Bickert, 1990; Lawrence et al., 2009; Max et al., 2020). A study on coccolithophore fluxes in the Bering Sea and subpolar Pacific (Tsutsui et al., 2016) has further demonstrated that the main blooming season and export of coccolithophores is heavily biased to October-November. Despite the potential seasonal bias in the  $U^{K'}_{37}$  proxy implicit in these studies, other work has suggested that alkenone production may have been much less seasonal in the warm early Pliocene (Lawrence et al., 2009). Overall, there is potential for the SSTs reconstructed from the  $U^{K'}_{37}$  index to be biased. Our analyses provide some perspective on this, as follows.

Our analyses highlight prominent eccentricity (both 400-kyr and 100-kyr) cycles in SST records in both low and mid-latitude, and high-latitude oceans since 4 Ma (Figs. 3, 4, 5; Figs. S2, S3), and this strong eccentricity cycle is also present in SST records reconstructed using foraminiferal Mg/Ca (Site 709C and 1094, Figs. 4d, 5d). The eccentricity cycles in SST records are consistent with theoretical eccentricity cycles (Fig. S3). Yet, annual mean insolation variability at the various sites is almost entirely dominated by obliquity, and the direct impact of eccentricity is very weak (Fig. S4). This agrees with obliquity cycle influences on insolation (Laskar et al., 2004) and relatively negligible direct contributions of eccentricity to insolation changes (<0.1%) (Clemens & Tiedemann, 1997). However, eccentricity still modulates the amplitude of precession impacts, which controls the amount of summer insolation (e.g., equatorial Pacific planktonic foraminiferal Mg/Ca-based SSTs show dominant precession variability since 140 ka; Jian et al., 2020). This, with additional carbon cycle changes and ice-albedo effects (Pälike et al., 2006), can drive major temperature responses at low latitudes (De Boer et al., 2014; Liebrand et al., 2017; Westerhold et al., 2020). Coupling between eccentricity pacing and carbon cycle variations has been inferred throughout the late Neogene based on carbon isotope variations (De Vleeschouwer et al., 2020), and is apparent also from orbital-scale covariation between global SST and atmospheric CO<sub>2</sub> concentrations (Fig. 2) (also Pälike et al., 2006; Liebrand et al., 2017). We infer that whatever is measured by the  $U^{K'}_{37}$  index (either

203 annual mean SSTs, or summer and autumn SSTs) reflects predominantly precession-based  
204 summer-insolation control, where persistence of insolation-driven summer and autumn warming  
205 affects winter SSTs due to the great thermal capacity of ocean water. Our observed prominence  
206 of eccentricity variability then reflects longer-term amplitude modulation of such precession  
207 influences.

208 Sliding window spectrograms of SSTs in the NH and equatorial regions reveal strong  
209 obliquity (41-kyr) variability between ~2.7 Ma and ~0.7 Ma (except at Site 846; Figs. 3, 4; see  
210 also Fig. S5). Although our collection of SST records from three Southern Hemisphere sites do  
211 not show a change in the obliquity signal during the Late Pliocene, a recent Southern  
212 Hemisphere SST stack that integrates additional records reveals substantial significant obliquity  
213 enhancement around 2.7 Ma (Clark et al., 2024). Our spectral analysis of the global SST stack  
214 from the same publication shows that this enhancement of the obliquity signal at ~2.7 Ma is  
215 global in nature (Fig. S6).

216 Obliquity signals at high latitudes are commonly interpreted in terms of annual mean solar  
217 radiation variations (Naish et al., 2009; Bosmans et al., 2015), but the reasons for weaker  
218 obliquity before ~2.7 Ma and after ~0.7 Ma are uncertain. At low latitudes, the proportional  
219 impact of obliquity on annual mean insolation is relatively small (cf. spatio-temporal illustrations  
220 in Loutre et al., 2004; Rohling et al., 2012), but we still observe a marked obliquity signal in  
221 tropical SST records between ~2.7 and ~0.7 Ma (Fig. 4). Obliquity signals have also been found  
222 after ~2.7 Ma in other low latitude records (e.g., Lourens et al., 2001). Some studies have  
223 suggested ocean circulation as a possible mechanism for transfer of high-latitude obliquity  
224 influences to lower latitudes (Jouzel et al., 2007). Other studies have suggested that  
225 interhemispheric temperature gradients may be critical (Raymo & Nisancioglu, 2003; Li et al.,  
226 2017; Bosmans et al., 2015). Meridional temperature gradients drive both atmospheric heat and  
227 moisture transport toward the poles (Raymo & Nisancioglu, 2003; Brierley & Fedorov, 2010;  
228 Fedorov et al., 2013). However, the hemispheric insolation gradient has not changed  
229 significantly since 4 Ma, and no consensus explanation has yet been reached for the abrupt  
230 appearance of the obliquity signal at that time, for its strength across the LP/EP, or for its  
231 weakness after the MPT (especially at low latitudes) (Raymo et al., 2006; Kender et al., 2018;  
232 Yehudai et al., 2021). The explanation must include a mechanism that amplifies the Earth system  
233 response to obliquity variations, including especially its unexpected predominance at low  
234 latitudes.

235 To investigate this issue, we calculate meridional sea surface temperature gradients (STG)  
236 in the North Pacific, North Atlantic, South Pacific and South Atlantic (Fig. 6). We also present a  
237 new assessment of STG sensitivity to obliquity forcing ( $S_{obl}$ ), which represents the ratio of  
238 obliquity-band variance in STG to the variance in obliquity (Fig. 6; see Methods). Given that  
239 previous studies have demonstrated that the influence of obliquity-driven ocean dynamics  
240 changes is amplified when Antarctic or Northern Hemisphere ice sheets expand into the marine  
241 environment (Levy et al., 2019; Cao et al., 2021), variations in  $S_{obl}$  may reflect major changes in  
242 terrestrial versus marine-based ice sheet responses to obliquity forcing (Levy et al., 2019).

243 We find substantial  $S_{obl}$  increases in NH STG records at ~2.7 Ma (Fig. 6b,c), without  
244 noticeable  $S_{obl}$  changes in SH STG records (Fig. 6d,e). At the same time, the STG in the NH also  
245 increased significantly at ~2.7 Ma (Fig. 6b,c). The ~2.7 Ma time interval marks the  
246 intensification of Northern Hemisphere glaciation (iNHG), which is recognized in climate proxy  
247 data that include deep-sea oxygen isotopes (Lisiecki & Raymo, 2005; Westerhold et al., 2020),  
248 North Atlantic planktonic foraminiferal assemblages (Dowsett & Poore, 1990) and ice rafted

249 debris records (Smith et al., 2018). This suggests that the strong global expression of obliquity in  
250 SST records at  $\sim 2.7$  Ma likely resulted from  $S_{obl}$  increase associated with establishment of large-  
251 scale NH ice sheets. Conversely, a distinct  $S_{obl}$  increase in SH STG records at  $\sim 1$  Ma may reflect  
252 expansion of the East Antarctica ice sheet (EAIS) margin into the ocean (Raymo et al., 2006),  
253 which would result in an enhanced response to obliquity in the SH (Levy et al., 2019). EAIS  
254 expansion into the ocean at around this time agrees with drill core data that indicate widespread  
255 expansion of a marine-based ice sheet in the Ross Embayment at the MPT (McKay et al., 2012).  
256 A  $S_{obl}$  decrease in the North Atlantic and North Pacific STGs at  $\sim 2.4$  Ma (Fig. 6b, c) corresponds  
257 to a negative  $\delta^{18}O_{sw}$  shift (Fig. 6a), which suggests that a reduction in global ice volume led to a  
258  $S_{obl}$  decrease in the climate records.  
259

### 260 **3.2 Coupling between SST, carbon cycle, and cryospheric changes since 4 Ma**

261 The causes of the iNHG at  $\sim 2.7$  Ma and subsequent frequency and amplitude changes of  
262 glacial cycles remain debated (Chalk et al., 2017; Kender et al., 2018; Willeit et al., 2019;  
263 Yehudai et al., 2021). Explanations for the LP/EP and MPT climate changes often invoke  
264 atmospheric  $CO_2$  and ice volume variations as critical links between orbital forcing and the  
265 climate response (Willeit et al., 2019; Yehudai et al., 2021). Hence, we investigate relationships  
266 between SST, global ice volume ( $\delta^{18}O_{sw}$ ), and atmospheric  $CO_2$  concentrations using time-  
267 evolutive SST- $\delta^{18}O_{sw}$  and SST- $CO_2$  phase analysis on 100-kyr eccentricity timescales (90-135  
268 kyr period range). We use 400-kyr-wide sliding windows and 10-kyr steps to calculate leads and  
269 lags between SST,  $\delta^{18}O_{sw}$ , and  $CO_2$  concentrations (see Methods). We find that SST led  $CO_2$   
270 concentrations for most of the time since  $\sim 1.5$  Ma, with a lead of about 0-17 kyr (0-60°, Fig. 7b)  
271 (ODP Sites 662, 709C and 1237 were not used for phase analysis due to data discontinuities and  
272 their low temporal resolution). SST led global ice volume for most of the time since  $\sim 4$  Ma, with  
273 a lead time of about 0-28 kyr (0-100°). However, some sites show that ice volume led SST  
274 between 2.2 and 1.5 Ma. The phase difference at these times is relatively large (50-180°; 14-50  
275 kyr), which may suggest a decoupling between ice volume and SST between 2.2 and 1.5 Ma  
276 (Fig. 7a).

277 The persistent lead of SSTs over global  $CO_2$  concentrations (since 1.5 Ma) and ice volume  
278 (since 4 Ma, except for 2.2-1.5 Ma) suggests that SST changes played an important role in global  
279 climate cycles since 4 Ma. We find that the phase difference between  $CO_2$  and ice volume is  
280 smaller and more stable (0-25°; 0-7 kyr) than the phase difference between SST and ice volume  
281 (0-100°; 0-28 kyr), which suggests that  $CO_2$  changes are more likely to be directly responsible  
282 for changes in global ice volume. This result could be informative also in the context of future  
283 climate change caused by anthropogenically forced increase in atmospheric  $CO_2$ . Reconstructed  
284  $CO_2$  data suggest a significant decrease from  $\sim 2.7$  Ma (Fig. 2o), associated with the iNHG. Due  
285 to a lack of high-resolution atmospheric  $CO_2$  reconstructions before 1.5 Ma, we have no way to  
286 determine phase relationships between  $CO_2$ , SST, and ice volume at  $\sim 2.7$  Ma. However, we find  
287 that SST has continued to lead atmospheric  $CO_2$  since  $\sim 1.5$  Ma (Fig. 7b; Fig. S7). There is about  
288 39,000 PgC of dissolved carbon storage in the oceans; more than 13 times the amount in the  
289 atmosphere and terrestrial biosphere combined (IPCC, 2007). Therefore, the mechanisms  
290 responsible for long-term changes in atmospheric  $CO_2$  concentrations likely are ocean-related  
291 (Kohfeld and Ridgwell, 2009). Previous work has elucidated the importance of carbon  
292 redistribution between the atmosphere and ocean carbon reservoirs in driving atmospheric  $CO_2$   
293 changes, notably through  $CO_2$  sequestration/release from the deep ocean (e.g., Kohfeld &

294 Ridgwell, 2009; Sigman et al., 2010; De Boer & Hogg, 2014; Kohfeld & Chase, 2017; Yu et al.,  
295 2014a, b, 2016, 2019, 2020; Hasenfratz et al., 2019).

296 Explaining the large (>100 ppm) atmospheric CO<sub>2</sub> drops associated with glacials requires a  
297 complex set of interconnected oceanic processes, including: 1) direct dissolution of atmospheric  
298 CO<sub>2</sub> into seawater (Martin et al., 2005; Kohfeld & Ridgwell, 2009; Yu et al., 2014a); 2)  
299 enhanced ocean biological pump activity (Martínez-García, 2011; Hain et al., 2014); 3)  
300 weakened deep-ocean circulation (Pena & Goldstein 2014; Farmer et al. 2019); and 4) reduced  
301 CO<sub>2</sub> exchange between the deep sea and the surface ocean ("stratification"; Martin et al., 2005;  
302 Hasenfratz et al., 2019). Sequestered CO<sub>2</sub> becomes "trapped" in the deep sea, as confirmed by  
303 deep-sea [CO<sub>3</sub><sup>2-</sup>] reconstructions (Ridgwell & Zeebe, 2005; Anderson et al., 2008; Yu et al.,  
304 2016, 2019, 2020). Air-sea disequilibrium can also enhance ocean carbon storage (Ito &  
305 Follows, 2013; Eggleston & Galbraith, 2018; Odalen et al., 2018; Khatiwala et al., 2019). In fact,  
306 previous studies in the Southern Ocean and the subarctic North Pacific based on biogenic opal  
307 accumulation and nitrogen isotope records indicate that a transition toward permanent polar  
308 ocean water-column stratification at ~2.7 Ma may have been a key driver of the atmospheric CO<sub>2</sub>  
309 decline (Haug et al., 1999; Sigman et al., 2010). This polar ocean stratification is thought to have  
310 been controlled by both temperature and salinity changes (Sigman et al., 2004). Our compiled  
311 dataset indicates that North Atlantic and North Pacific SSTs decreased markedly from 4 to ~2.7  
312 Ma (ODP Sites 982 and 607; Fig. 2c, d), while the corresponding STG increased (Fig. 6b, c). NH  
313 high latitude cooling (ODP Sites 982 and 1012, Fig. 2c,e) would increase the equatorial-to-polar  
314 meridional temperature gradient (Fig. 6b,c), strengthening water vapor transport from the tropics  
315 (tropical Pacific and tropical Atlantic) to the Arctic (Loutre et al., 2004; Brierley & Fedorov,  
316 2010) through the temperature-moisture feedback (Källén et al., 1979; Raymo & Nisancioglu,  
317 2003). At the same time, NH high latitude cooling promotes Northern Hemisphere ice sheet  
318 development (Lunt et al., 2008; Brierley & Fedorov, 2010). High latitude cooling and increased  
319 sea ice cover cause intensification of water-column stratification (Sigman et al., 2004) and, thus,  
320 favor enhanced carbon accumulation in the deep-sea (Marinov et al., 2008; Eggleston and  
321 Galbraith, 2018; Odalen et al., 2018). In sum, oceanic processes associated with high-latitude  
322 SST reduction were crucial to atmospheric CO<sub>2</sub> reduction and NH glacial intensification during  
323 the Late Pliocene. This is supported by a substantial S<sub>obl</sub> increase for NH STG at ~2.7 Ma (Fig.  
324 6b,c). Several studies have proposed a simple inverse correlation between global mean surface  
325 temperature and STG measures, indicating a robust and fairly universal relationship through the  
326 past 10 million years (at low resolution) and the Eocene (on eccentricity time scales; Liu et al.,  
327 2022; Gaskell et al., 2022; Fokkema et al., 2024). Similarly, we find both a notable STG increase  
328 (Fig. 6b-d) and a distinct global sea surface temperature decrease at ~2.7 Ma (Fig. 2c-m). The  
329 predictability of this relationship between global (sea) surface temperature and STG may be  
330 relevant to future climate change.

331 In contrast to the atmospheric CO<sub>2</sub> decrease from ~2.7 Ma, there is no notable long-term  
332 (>100 kyr) atmospheric CO<sub>2</sub> decrease after 1 Ma (Fig. 2o), even though SST near Antarctica  
333 continued to decrease (ODP Site 1094). We note that Arctic SST (ODP Site 982) does not show  
334 this SST decrease. Inconsistent evolution of SST changes for the two poles gives a strong  
335 indication that global ice volume increases after 1 Ma had an important Antarctic component  
336 (Fig. 2b). Mid-Pleistocene high latitude cooling may have caused persistent East Antarctic ice  
337 sheet expansion into the ocean so that it no longer contained an extensive terrestrial margin,  
338 which would favor in-phase variability of NH and SH ice sheets (Raymo et al., 2006). Ocean  
339 temperature plays an important role in controlling long-term fluctuations in ice sheet margins

340 (Joughin et al., 2012), with marine-based ice particularly susceptible to rapid changes in ocean  
341 heat supply (Mengel & Levermann, 2014). Such mass balance sensitivity to ocean temperature  
342 has been emphasized for the EAIS (Pollard & DeConto, 2009; McKay et al., 2012; Hansen et al.,  
343 2015; Hasenfrazt et al., 2019). Overall, therefore, we infer that EAIS variability may have  
344 become amplified from ~1 Ma by the development of an extensive marine ice sheet margin.

345 To test our argument about fundamental transitions in the relationships between SST, STG,  
346 and high-latitude ice sheets, we performed recurrence analyses of the NH and SH STG, as well  
347 as global ice volume ( $\delta^{18}\text{O}_{\text{sw}}$ ). Results highlight major global ice volume shifts at ~2.7 Ma and  
348 ~1 Ma (Fig. 8a), consistent with previous studies that suggest a major global climate transition  
349 associated with high-latitude ice sheet development (Westerhold et al., 2020). Recurrence  
350 analyses of STG in the North Atlantic and North Pacific (ODP Sites 662-982 and 846-1012; Fig.  
351 8b,c) show a major shift at ~2.7 Ma, suggesting a link between NH STG and iNHG. Conversely,  
352 recurrence analyses of STG in the South Atlantic and South Pacific (Sites 1082-1090 and 1239-  
353 1237; Fig. 8d,e) show a major transition at ~1 Ma, in agreement with the aforementioned concept  
354 of widespread marine AIS margin development. The recurrence analyses, therefore, support our  
355 previous interpretations that there were two key climate transitions since 4 Ma (LP/EP and MPT)  
356 that contained critical involvement of SST and STG changes with distinctly different expressions  
357 (and, hence, governing processes) between the two hemispheres.

358

#### 359 **4 Conclusions**

360 We compiled globally distributed SST records to investigate the involvement of orbital-  
361 scale SST trends and oceanic meridional temperature gradient changes in key climate transitions  
362 during the last 4 million years. We observe important orbital eccentricity and obliquity  
363 influences in the SST evolution, and infer both summer insolation and high-latitude ice volume  
364 (feedback) impacts on global SST. We find that the SST in the NH and the equatorial region  
365 shows a significant enhancement of the obliquity signal at ~2.7 Ma, and at the same time, the NH  
366 meridional SST gradient as well as its obliquity sensitivity also increases sharply at ~2.7 Ma. We  
367 infer that the enhancement of the obliquity signal in the NH and equatorial region during this  
368 period originates from large-scale NH ice sheet expansion. In addition, the increase in obliquity  
369 sensitivity of the southern hemisphere meridional SST gradient at ~1 Ma may be attributed to  
370 oceanward expansion of the Antarctic ice sheet margin. Phase analysis revealed key links  
371 between variations in SST, global ice volume, and atmospheric  $\text{CO}_2$  levels, which underpinned  
372 the major climate shifts. Changes in SST can directly affect not only the formation and  
373 development of high-latitude ice sheets, but also global temperatures and thus ice volume by  
374 controlling changes in the atmospheric  $\text{CO}_2$  concentration.

375

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381

382 **Open Research**

383 The sea surface temperature data are available at Zhang (2024a). The data analysis was carried out  
 384 in R. The complete codes reported in this study are available at Zhang (2024b).

385 **References**

- 386 Anderson, R. F., *et al.* Modern CaCO<sub>3</sub> preservation in equatorial Pacific sediments in the context  
 387 of late-Pleistocene glacial cycles. *Marine Chemistry* **111**, 30-46 (2008).
- 388 Bartoli, G., Hönisch, B., Zeebe, R. E. Atmospheric CO<sub>2</sub> decline during the Pliocene  
 389 intensification of Northern Hemisphere glaciations. *Paleoceanography* **26** (2011).
- 390 Bosmans, J. H. C., *et al.* Obliquity forcing of low-latitude climate. *Clim. Past* **11**, 1335-1346  
 391 (2015).
- 392 Brierley, C. M. *et al.* Greatly expanded tropical warm pool and weakened Hadley circulation in  
 393 the early Pliocene. *Science* **323**, 1714-1718 (2009).
- 394 Brierley, C. M., Fedorov, A. V. Relative importance of meridional and zonal sea surface  
 395 temperature gradients for the onset of the ice ages and Pliocene-Pleistocene climate  
 396 evolution. *Paleoceanography* **25** (2010).
- 397 Callahan, J., *et al.* IRISSeismic: Classes and Methods for Seismic Data Analysis [https://cran.r-](https://cran.r-project.org/web/packages/IRISSeismic/index.html)  
 398 [project.org/web/ packages/IRISSeismic/index.html](https://cran.r-project.org/web/packages/IRISSeismic/index.html) (2018).
- 399 Cao, M. M. *et al.* Mineral Dust Coupled With Climate-Carbon Cycle on Orbital Timescales Over  
 400 the Past 4 Ma. *Geophys. Res. Lett.* **48**, e2021GL095327 (2021).
- 401 Cayan, D. R. Latent and sensible heat flux anomalies over the northern oceans: driving the sea  
 402 surface temperature. *J. Phys. Oceanogr.* **22**, 859-881 (1992).
- 403 Chalk, T. B. *et al.* Causes of ice age intensification across the Mid-Pleistocene Transition. *Proc.*  
 404 *Natl. Acad. Sci. U.S.A.* **114**, 13114-13119 (2017).
- 405 Clark, P. U., *et al.* Global and regional temperature change over the past 4.5 million  
 406 years. *Science* **383**, 884-890 (2024).
- 407 Clemens, S. C., Tiedemann, R. Eccentricity forcing of Pliocene-early Pleistocene climate  
 408 revealed in a marine oxygen-isotope record. *Nature* **385**, 801-804 (1997).
- 409 De Boer, A. M., Hogg, A. M. Control of the glacial carbon budget by topographically induced  
 410 mixing. *Geophys. Res. Lett.* **41**, 4277-4284 (2014).
- 411 De Boer, B., Lourens, L. J., van de Wal, R. S. W. Persistent 400,000-year variability of Antarctic  
 412 ice volume and the carbon cycle is revealed throughout the Plio-Pleistocene. *Nat. Commun.*  
 413 **5**, 1-8 (2014).
- 414 De Vleeschouwer, D. *et al.* High-latitude biomes and rock weathering mediate climate-carbon  
 415 cycle feedbacks on eccentricity timescales. *Nat. Commun.* **11**, 5013 (2020).
- 416 Dekens, P. S., *et al.* Core top calibration of Mg/Ca in tropical foraminifera: Refining  
 417 paleotemperature estimation. *Geochem. Geophys. Geosy.* **3**, 1-29 (2002).
- 418 Dekens, P. S., Ravelo, A. C., McCarthy, M. D. Warm upwelling regions in the Pliocene warm  
 419 period. *Paleoceanography* **22** (2007).
- 420 Deser, C., *et al.* Sea surface temperature variability: Patterns and mechanisms. *Annu. Rev. Mar.*  
 421 *Sci.* **2**, 115-143 (2010).
- 422 Dowsett, H. J., Poore, R. Z. A new planktic foraminifer transfer function for estimating Pliocene  
 423 through Holocene sea surface temperatures. *Mar. Micropaleontol.* **16**, 1-23 (1990).
- 424 Dowsett, H. J., Robinson, M. M., Foley, K. M. Pliocene three-dimensional global ocean  
 425 temperature reconstruction. *Clim. Past* **5**, 769-783 (2009).

- 426 Eggleston, S., Galbraith, E. D. The devil's in the disequilibrium: multi-component analysis of  
427 dissolved carbon and oxygen changes under a broad range of forcings in a general  
428 circulation model. *Biogeosciences* **15**, 3761-3777 (2018).
- 429 Eroglu, D. *et al.* See-saw relationship of the Holocene East Asian-Australian summer monsoon.  
430 *Nat. Commun.* **7**, 12929 (2016).
- 431 Etourneau, J., *et al.* Pliocene-Pleistocene variability of upwelling activity, productivity, and  
432 nutrient cycling in the Benguela region. *Geology* **37**, 871-874 (2009).
- 433 Etourneau, J., *et al.* Intensification of the Walker and Hadley atmospheric circulations during the  
434 Pliocene-Pleistocene climate transition. *Earth Planet. Sci. Lett.* **297**, 103-110 (2010).
- 435 Farmer, J. R. *et al.* Deep Atlantic Ocean carbon storage and the rise of 100,000-year glacial  
436 cycles. *Nat. Geosci.* **12**, 355-360 (2019).
- 437 Fedorov, A. V. *et al.* Patterns and mechanisms of early Pliocene warmth. *Nature* **496**, 43-49  
438 (2013).
- 439 Fedorov, A. V., *et al.* Tightly linked zonal and meridional sea surface temperature gradients over  
440 the past five million years. *Nat. Geosci.* **8**, 975-980 (2015).
- 441 Fokkema, C. D., *et al.* Polar amplification of orbital-scale climate variability in the early Eocene  
442 greenhouse world. *Clim. Past* **20**, 1303-1325 (2024).
- 443 Gaskell, D. E., *et al.* The latitudinal temperature gradient and its climate dependence as inferred  
444 from foraminiferal  $\delta^{18}\text{O}$  over the past 95 million years. *Proc. Natl. Acad. Sci. U. S. A.* **119**,  
445 e2111332119 (2022).
- 446 Hain, M. P., Sigman, D. M., Haug, G. H. The biological Pump in the Past. Reference Module in  
447 Earth Systems and Environmental Sciences, treatise on Geochemistry (Second Edition). *The*  
448 *Oceans and Marine Geochemistry* **8**, 485-517 (2014).
- 449 Han, W. X., *et al.* Climate transition in the Asia inland at 0.8-0.6 Ma related to astronomically  
450 forced ice sheet expansion. *Quat. Sci. Rev.* **248**, 106580 (2020).
- 451 Hasenfratz, A. P. *et al.* The residence time of Southern Ocean surface waters and the 100,000-  
452 year ice age cycle. *Science* **363**, 1080-1084 (2019).
- 453 Haug, G. H., *et al.* Onset of permanent stratification in the subarctic Pacific Ocean. *Nature* **401**,  
454 779-782 (1999).
- 455 Herbert, T. D., *et al.* Tropical ocean temperatures over the past 3.5 million years. *Science* **328**,  
456 1530-1534 (2010).
- 457 Hönisch, B., *et al.* Atmospheric carbon dioxide concentration across the Mid-Pleistocene  
458 transition. *Science* **324**, 1551-1554 (2009).
- 459 IPCC. Climate Change 2007: The Physical Science Basis. Contribution of Working Group I to  
460 the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, 996 pp.,  
461 Cambridge Univ. Press, Cambridge, U. K. (2007).
- 462 Ito, T., Follows, M. J. Air-sea disequilibrium of carbon dioxide enhances the biological carbon  
463 sequestration in the Southern Ocean. *Global Biogeochem. Cy.* **27**, 1129-1138 (2013).
- 464 Jian, Z., *et al.* Half-precessional cycle of thermocline temperature in the western equatorial  
465 Pacific and its bihemispheric dynamics. *Proc. Natl. Acad. Sci. U. S. A.* **117**, 7044-7051  
466 (2020).
- 467 Joughin, I., Alley, R. B., Holland, D. M. Ice-sheet response to oceanic forcing. *Science* **338**,  
468 1172-1176 (2012).
- 469 Jouzel, J., *et al.* Orbital and millennial Antarctic climate variability over the past 800,000  
470 years. *Science* **317**, 793-796 (2007).

- 471 Källén, E., Crafoord, C., Ghil, M. Free oscillations in a climate model with ice-sheet  
472 dynamics. *J. Atmos. Sci.* **36**, 2292-2303 (1979).
- 473 Karas, C., *et al.* Pliocene Indonesian Throughflow and Leeuwin Current dynamics: implications  
474 for Indian Ocean polar heat flux. *Paleoceanography* **26**, PA2217 (2011).
- 475 Kender, S., *et al.* Closure of the Bering Strait caused mid-Pleistocene transition cooling. *Nat.*  
476 *Commun.* **9**, 1-11 (2018).
- 477 Khatiwala, S., Schmittner, A., Muglia, J. Air-sea disequilibrium enhances ocean carbon storage  
478 during glacial periods. *Sci. Adv.* **5**, eaaw4981 (2019).
- 479 Kohfeld, K. E., Chase, Z. Temporal evolution of mechanisms controlling ocean carbon uptake  
480 during the last glacial cycle. *Earth Planet. Sci. Lett.* **472**, 206-215 (2017).
- 481 Kohfeld, K. E., Ridgwell, A. Glacial-interglacial variability in atmospheric CO<sub>2</sub>. *Surface ocean-*  
482 *lower atmosphere processes* **187**, 251-286 (2009).
- 483 Laskar, J. *et al.* A long-term numerical solution for the insolation quantities of the Earth. *Astron.*  
484 *Astrophys.* **428**, 261-285 (2004).
- 485 Lawrence, K. T., Liu, Z., Herbert, T. D. Evolution of the eastern tropical Pacific through Plio-  
486 Pleistocene glaciation. *Science* **312**, 79-83 (2006).
- 487 Lawrence, K. T., *et al.* North Atlantic climate evolution through the Plio-Pleistocene climate  
488 transitions. *Earth Planet. Sci. Lett.* **300**, 329-342 (2010).
- 489 Lawrence, K., *et al.* High-amplitude variations in North Atlantic sea surface temperature during  
490 the early Pliocene warm period. *Paleoceanography* **24**, PA2218 (2009).
- 491 Levy, R. H. *et al.* Antarctic ice-sheet sensitivity to obliquity forcing enhanced through ocean  
492 connections. *Nat. Geosci.* **12**, 132-137 (2019).
- 493 Li, L. *et al.* A 4-Ma record of thermal evolution in the tropical western Pacific and its  
494 implications on climate change. *Earth Planet. Sci. Lett.* **309**, 10-20 (2011).
- 495 Li, M. S., Hinnov, L., Kump, L. Acycle: time-series analysis software for paleoclimate research  
496 and education. *Comput. Geosci.* **127**, 12-22 (2019).
- 497 Li, T., *et al.* Continued obliquity pacing of East Asian summer precipitation after the mid-  
498 Pleistocene transition. *Earth Planet. Sci. Lett.* **457**, 181-190 (2017).
- 499 Liebrand, D. *et al.* Evolution of the early Antarctic ice ages. *Proc. Natl. Acad. Sci. U.S.A.* **114**,  
500 3867-3872 (2017).
- 501 Lisiecki, L. E., Raymo, M. E. A Pliocene-Pleistocene stack of 57 globally distributed benthic  
502  $\delta^{18}\text{O}$  records. *Paleoceanography* **20**, 1003 (2005).
- 503 Liu, Z. H., Herbert, T. D. High-latitude influence on the eastern equatorial Pacific climate in the  
504 early Pleistocene epoch. *Nature* **427**, 720-723 (2004).
- 505 Liu, X., *et al.* Persistent high latitude amplification of the Pacific Ocean over the past 10 million  
506 years. *Nat. Commun.* **13**, 7310 (2022).
- 507 Lourens, L. J., Wehausen, R., Brumsack, H. J. Geological constraints on tidal dissipation and  
508 dynamical ellipticity of the Earth over the past three million years. *Nature* **409**, 1029-1033  
509 (2001).
- 510 Loutre, M. F., *et al.* Does mean annual insolation have the potential to change the climate? *Earth*  
511 *Planet. Sci. Lett.* **221**, 1-14 (2004).
- 512 Lunt, D. J., *et al.* Late Pliocene Greenland glaciation controlled by a decline in atmospheric CO<sub>2</sub>  
513 levels. *Nature* **454**, 1102-1105 (2008).
- 514 Lüthi, D., *et al.* CO<sub>2</sub> and O<sub>2</sub>/N<sub>2</sub> variations in and just below the bubble-clathrate transformation  
515 zone of Antarctic ice cores. *Earth Planet. Sci. Lett.* **297**, 226-233 (2010).

- 516 Mann, M. E., Lees, J. M. Robust estimation of background noise and signal detection in climatic  
517 time series. *Clim. Change* **33**, 409-445 (1996).
- 518 Marinov, I., *et al.* Impact of oceanic circulation on biological carbon storage in the ocean and  
519 atmospheric pCO<sub>2</sub>. *Global Biogeochemical Cycles* **22** (2008).
- 520 Martin, P., Archer, D., Lea, D. W. Role of deep sea temperature in the carbon cycle during the  
521 last glacial. *Paleoceanography* **20** (2005).
- 522 Martínez-Botí, M., *et al.* Plio-Pleistocene climate sensitivity evaluated using high-resolution  
523 CO<sub>2</sub> records. *Nature* **518**, 49-54 (2015).
- 524 Martínez-García, A., *et al.* Southern Ocean dust-climate coupling over the past four million  
525 years. *Nature* **476**, 312-315 (2011).
- 526 Martínez-García, A., *et al.* Subpolar link to the emergence of the modern equatorial Pacific cold  
527 tongue. *Science* **328**, 1550-1553 (2010).
- 528 Marwan, N. Cross Recurrence Plot Toolbox for MATLAB®, Ver. 5.22 (R32.4), [http://tocsy.pik-](http://tocsy.pik-potsdam.de/CRPtoolbox/)  
529 [potsdam.de/CRPtoolbox/](http://tocsy.pik-potsdam.de/CRPtoolbox/) accessed 2020-12-09 (2020).
- 530 Marwan, N., *et al.* Recurrence Plots for the Analysis of Complex Systems. *Phys. Rep.* **438**, 237-  
531 329 (2007).
- 532 Max, L., *et al.* Evaluation of reconstructed sea surface temperatures based on U<sup>K'</sup><sub>37</sub> from  
533 sediment surface samples of the North Pacific. *Quaternary Sci. Rev.* **243**, 106496 (2020).
- 534 McKay, R. *et al.* Pleistocene variability of Antarctic ice sheet extent in the Ross  
535 embayment. *Quaternary Sci. Rev.* **34**, 93-112 (2012).
- 536 Mengel, M., Levermann, A. Ice plug prevents irreversible discharge from East Antarctica. *Nat.*  
537 *Clim. Change* **4**, 451-455 (2014).
- 538 Monnin, E., *et al.* Atmospheric CO<sub>2</sub> concentrations over the last glacial termination. *Science* **291**,  
539 112-114 (2001).
- 540 Müller, P. J., *et al.* Calibration of the alkenone paleotemperature index U<sup>K'</sup><sub>37</sub> based on core-tops  
541 from the eastern South Atlantic and the global ocean (60°N - 60 °S). *Geochem. Cosmochim.*  
542 *Acta* **62**, 1757-1772 (1998).
- 543 Naish, T. *et al.* Obliquity-paced Pliocene West Antarctic ice sheet oscillations. *Nature* **458**, 322-  
544 328 (2009).
- 545 O'Brien, C. L. *et al.* High sea surface temperatures in tropical warm pools during the  
546 Pliocene. *Nat. Geosci.* **7**, 606-611 (2014).
- 547 Odalen, M., *et al.* The influence of the ocean circulation state on ocean carbon storage and CO<sub>2</sub>  
548 drawdown potential in an Earth system model. *Biogeosciences* **15**, 1367-1393 (2018).
- 549 Pälike, H., *et al.* The heartbeat of the Oligocene climate system. *Science* **314**, 1894-1898 (2006).
- 550 Pena, L. D., Goldstein, S. L. Thermohaline circulation crisis and impacts during the mid-  
551 Pleistocene transition. *Science* **345**, 318-322 (2014).
- 552 Pépin, L., *et al.* Hemispheric roles of climate forcings during glacial-interglacial transitions as  
553 deduced from the Vostok record and LLN-2D model experiments. *Journal of Geophysical*  
554 *Research: Atmospheres* **106**, 31885-31892 (2001).
- 555 Petit, J. R., *et al.* Climate and atmospheric history of the past 420,000 years from the Vostok ice  
556 core, Antarctica. *Nature* **399**, 429-436 (1999).
- 557 Pollard, D., DeConto, R. M. Modelling West Antarctic ice sheet growth and collapse through the  
558 past five million years. *Nature* **458**, 329-332 (2009).
- 559 Raymo, M. E., Lisiecki, L. E., Nisancioglu, K. H. Plio-Pleistocene ice volume, Antarctic climate,  
560 and the global δ<sup>18</sup>O record. *Science* **313**, 492-495 (2006).

- 561 Raymo, M. E., Nisancioglu, K. H. The 41 kyr world: Milankovitch's other unsolved  
562 mystery. *Paleoceanography* **18** (2003).
- 563 Raynaud, D., *et al.* The record for marine isotopic stage 11. *Nature* **436**, 39-40 (2005).
- 564 Ridgwell, A., Zeebe, R. E. The role of the global carbonate cycle in the regulation and evolution  
565 of the Earth system, *Earth Planet. Sci. Lett.* **234**, 299-315 (2005).
- 566 Rohling, E. J., *et al.* Sea level and deep-sea temperature reconstructions suggest quasi-stable  
567 states and critical transitions over the past 40 million years. *Sci. Adv.* **7**, eabf5326 (2021).
- 568 Rohling, E. J., *et al.* Sea surface and high-latitude temperature sensitivity to radiative forcing of  
569 climate over several glacial cycles. *J. Climate* **25**, 1635-1656 (2012).
- 570 Rosell-Melé, A. Interhemispheric appraisal of the value of alkenone indices as temperature and  
571 salinity proxies in high-latitude locations. *Paleoceanography* **13**, 694-703 (1998).
- 572 Samtleben, C., Bickert, T. Coccoliths in sediment traps from the Norwegian Sea. *Mar.*  
573 *Micropaleontol.* **16**, 39-64 (1990).
- 574 Schinkel, S., Dimigen, O., Marwan, N. Selection of recurrence threshold for signal detection.  
575 *European Physical Journal Special Topics* **164**, 45-53 (2008).
- 576 Siegenthaler, U. R. S., *et al.* Supporting evidence from the EPICA Dronning Maud Land ice core  
577 for atmospheric CO<sub>2</sub> changes during the past millennium. *Tellus B: Chemical and Physical*  
578 *Meteorology* **57**, 51-57 (2005a).
- 579 Siegenthaler, U., *et al.* Stable carbon cycle–climate relationship during the Late Pleistocene.  
580 *Science* **310**, 1313-1317 (2005b).
- 581 Sigman, D. M., Hain, M. P., Haug, G. H. The polar ocean and glacial cycles in atmospheric CO<sub>2</sub>  
582 concentration. *Nature* **466**, 47-55 (2010).
- 583 Sigman, D. M., Jaccard, S., Haug, G. H. Polar ocean stratification in a cold climate. *Nature* **428**,  
584 59-63 (2004).
- 585 Smith, Y. M. *et al.* Icebergs in the Nordic Seas Throughout the Late Pliocene. *Paleoceanogr.*  
586 *Paleoclimatol.* **33**, 318-335 (2018).
- 587 Stap, L. B. *et al.* CO<sub>2</sub> over the past 5 million years: Continuous simulation and new  $\delta^{11}\text{B}$ -based  
588 proxy data. *Earth Planet. Sci. Lett.* **439**, 1-10 (2016).
- 589 Tsutsui, H., *et al.* Nineteen-year time-series sediment trap study of *Coccolithus pelagicus* and  
590 *Emiliana huxleyi* (calcareous nannoplankton) fluxes in the Bering Sea and subarctic Pacific  
591 Ocean. *Deep-Sea Res. Part II Top. Stud. Oceanogr.* **125**, 227-239 (2016).
- 592 Vázquez Riveiros, N., *et al.* Mg/Ca thermometry in planktic foraminifera: Improving  
593 paleotemperature estimations for *G. bulloides* and *N. pachyderma* left. *Geochem. Geophys.*  
594 *Geosy.* **17**, 1249-1264 (2016).
- 595 Westerhold, T., *et al.* An astronomically dated record of Earth's climate and its predictability  
596 over the last 66 million years. *Science* **369**, 1383-1387 (2020).
- 597 Willeit, M., *et al.* Mid-Pleistocene transition in glacial cycles explained by declining CO<sub>2</sub> and  
598 regolith removal. *Sci. Adv.* **5**, eaav7337 (2019).
- 599 Yamamoto, M., *et al.* Increased interglacial atmospheric CO<sub>2</sub> levels followed the mid-  
600 Pleistocene Transition. *Nat. Geosci.* **15**, 307-313 (2022).
- 601 Yehudai, M., *et al.* Evidence for a Northern Hemispheric trigger of the 100,000-y glacial  
602 cyclicity. *Proc. Natl. Acad. Sci. U.S.A.* **118** (2021).
- 603 Yu, J., *et al.* Deep South Atlantic carbonate chemistry and increased interocean deep water  
604 exchange during last deglaciation. *Quaternary Sci. Rev.* **90**, 80-89 (2014b).
- 605 Yu, J., *et al.* Last glacial atmospheric CO<sub>2</sub> decline due to widespread Pacific deep water  
606 expansion. *Nat. Geosci.* **13**, 628-633 (2020).

- 607 Yu, J., *et al.* More efficient North Atlantic carbon pump during the Last Glacial Maximum. *Nat.*  
608 *Commun.* **10**, 2170 (2019).
- 609 Yu, J., *et al.* Sequestration of carbon in the deep Atlantic during the last glaciation. *Nat. Geosci.*  
610 **9**, 319-324 (2016).
- 611 Yu, J., Anderson, R. F., Rohling, E. J. Deep ocean carbonate chemistry and glacial-interglacial  
612 atmospheric CO<sub>2</sub> changes. *Oceanography* **27**, 16-25 (2014a).
- 613 Zhang, Z. Sea surface temperature data reconstructed by unsaturated alkenone index and  
614 foraminiferal Mg/Ca since 4 Ma. Version 2 [Dataset]. 4TU. ResearchData  
615 (2024a). <https://doi.org/10.4121/20486157>.
- 616 Zhang, Z. R code for Obliquity sensitivity and phase analysis. Version 1 [Software]. 4TU.  
617 ResearchData (2024b). <https://doi.org/10.4121/2904d0f4-0f50-45f4-8363-69574ac1b2e5>.
- 618

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620 **Figure 1. Map of sites (black dots) from which data were retrieved (data are from:**

621 **Lawrence et al., 2006, 2009, 2010; Brierley et al. 2009; Etourneau et al. 2009, 2010; Dekens**  
622 **et al. 2007; Herbert et al., 2010; Liu & Herbert, 2004; Karas et al., 2011; Martínez-Garcia**

623 **et al. 2010; Hasenfratz et al., 2019), superimposed on a map of modern annual mean SST**

624 **(contours and colours after Fedorov et al., 2013). Numbers indicate sites from the Deep Sea**

625 **Drilling Project (DSDP), the Ocean Drilling Program (ODP), the Integrated Ocean Drilling**

626 **Program and International Ocean Discovery Program (both IODP), and the International Marine**

627 **Global Change Study (IMAGES). Black arrows indicate modern global ocean circulation.**

628 **Acronyms: Atlantic Drift (NAD), Gulf Stream (GS), North Equatorial Current (NEC), South**

629 **Equatorial Current (SEC), Humboldt Current (HC), Labrador Current (LC), California Current**

630 **(CC), Equatorial Counter Current (ECC), Thousand Islands Cold Current (TICC), Kuroshio**

631 **(Kuro.), Canary Cold Stream (CCS), Benguela Current (BC), Somali Warm Current (SWC), and**

632 **Western Australian Cold Stream (WACS).**

633

634 **Figure 2. SST and CO<sub>2</sub> concentration records shown with benthic foraminiferal  $\delta^{18}\text{O}$  and**

635 **seawater  $\delta^{18}\text{O}_{\text{sw}}$  for the past 4000 kyr (4 Myr). a) Benthic foraminiferal  $\delta^{18}\text{O}$  (LR04 stack;**

636 Lisiecki & Raymo, 2005). b) Seawater  $\delta^{18}\text{O}_{\text{sw}}$  (proxy for global ice volume; Rohling et al.,  
637 2021). The direction of the arrow indicates increasing ice volume. c) Sea surface temperature of  
638 ODP Sites 982 (North Atlantic; Lawrence et al., 2009). d) Sea surface temperature of DSDP Site  
639 607 (North Atlantic; Lawrence et al., 2010). e) Sea surface temperature of ODP Site 1012 (North  
640 Pacific; Brierley et al., 2009). f) Sea surface temperature of ODP Site 722 (Arabian Sea; Herbert  
641 et al., 2010). g) Sea surface temperature of ODP Site 1239 (Eastern equatorial Pacific; Etourneau  
642 et al., 2010). h) Sea surface temperature of ODP Site 662 (Equatorial Atlantic; Herbert et al.,  
643 2010). i) Sea surface temperature of ODP Site 846 (Eastern equatorial Pacific; Liu & Herbert,  
644 2004; Lawrence et al., 2006). j) Sea surface temperature of ODP Site 709C (Indian Ocean; Karas  
645 et al., 2011). k) Sea surface temperature of ODP Site 1237 (South Pacific; Dekens et al., 2007). l)  
646 Sea surface temperature of ODP Site 1082 (South Atlantic; Etourneau et al., 2009). m) Sea  
647 surface temperature of ODP Site 1090 (Subantarctic; Martínez-García et al., 2010). n) Sea  
648 surface temperature of ODP Site 1094 (Antarctic; Hasenfratz et al., 2019). o) Estimated  
649 atmospheric  $\text{CO}_2$  concentrations (Yamamoto et al., 2022; Stap et al., 2016; Martínez-Botí et al.,  
650 2015; Bartoli et al., 2011; Hönisch et al., 2009).

651

652 **Figure 3. Astronomical evolution of SSTs from ocean drilling sites at different latitudes in**  
653 **the Northern Hemisphere.** Spectrograms with 600 kyr sliding windows with  $2\pi$  multi-taper  
654 (MTM) power spectra of (a) SST of ODP Site 982 since 4 Ma, (b) SST of DSDP Site 607 since 4  
655 Ma, (c) SST of ODP Site 1012 since 4 Ma, (d) SST of ODP Site 722 since  $\sim 3.3$  Ma. SSTs from  
656 these sites were reconstructed using  $\text{U}^{\text{K}'}_{37}$ . The red dotted line on the left side of the  
657 spectrograms are the 95% confidence levels. The ticks on the power x-axes indicate the strength  
658 of the periodic signal in the spectrogram. The colors in the sliding window spectrogram indicate

659 the strength of the periodic signal, with blue-grayish blue-green indicating an increasingly strong  
660 periodic signal. The white shaded band indicates the enhancement of the obliquity signal at ~2.7  
661 Ma.

662

663 **Figure 4. Astronomical evolution of SST from ocean drilling sites in the equatorial region.**

664 Spectrograms with 600 kyr sliding windows with  $2\pi$  multi-taper (MTM) power spectra of (a)  
665 SST of ODP Site 662 since 4 Ma, (b) SST of ODP Site 1239 since 4 Ma, (c) SST of ODP Site  
666 846 since ~3.9 Ma, (d) SST of ODP Site 709C from ~4.9 to ~2.2 Ma. SST of ODP Sites 709C  
667 were reconstructed using foraminiferal Mg/Ca, and the remaining SST temperatures were  
668 reconstructed using  $U^{K'}_{37}$ . The red dotted line on the left side of the spectrograms are the 95%  
669 confidence levels. The ticks on the power x-axes indicate the strength of the periodic signal in  
670 the spectrogram. The colors in the sliding window spectrogram indicate the strength of the  
671 periodic signal, with blue-grayish blue-green indicating an increasingly strong periodic signal.  
672 The white shaded band indicates the enhancement of the obliquity signal at ~2.7 Ma.

673

674 **Figure 5. Astronomical evolution of SST from ocean drilling sites at different latitudes in**

675 **the Southern Hemisphere.** Spectrograms with 600 kyr sliding windows with  $2\pi$  multi-taper  
676 (MTM) power spectra of (a) SST of ODP Site 1237 since 4 Ma, (b) SST of ODP Site 1082 since  
677 ~3.5 Ma, (c) SST of ODP Site 1090 since ~3.6 Ma, (d) SST of ODP Site 1094 from 1.5 to 0.9  
678 Ma. ODP Sites 1094 were reconstructed using foraminiferal Mg/Ca, and the remaining SST  
679 temperatures were reconstructed using  $U^{K'}_{37}$ . Note that the absence of an obliquity signal in the  
680 SST record at ODP Site 1237 is likely due to this record's lower temporal resolution (~14 kyr).  
681 The red dotted line on the left side of the spectrograms are the 95% confidence levels. The ticks

682 on the power x-axes indicate the strength of the periodic signal in the spectrogram. The colors in  
 683 the sliding window spectrogram indicate the strength of the periodic signal, with blue-grayish  
 684 blue-green indicating an increasingly strong periodic signal.

685

686 **Figure 6. Meridional SST gradients (STG) and STG obliquity sensitivity ( $S_{obl}$ ) for the**  
 687 **North Atlantic, North Pacific, South Atlantic and South Pacific since 4 Ma.** a) Seawater  
 688  $\delta^{18}O_{sw}$  (proxy for global ice volume; Rohling et al., 2021). The black arrow indicates increasing  
 689 ice volume. b) Meridional SST gradients between ODP Sites 846 (3°S, 91°W; Liu & Herbert,  
 690 2004; Lawrence et al., 2006) and 1012 (32°N, 118°W; Brierley et al. 2009) for North Pacific and  
 691 obliquity sensitivity ( $S_{obl}$ ). c) Meridional SST gradients between ODP Site 662 (1°S, 12°W;  
 692 Herbert et al., 2010) and 982 (58°N, 16°W; Lawrence et al., 2009) for North Atlantic and  
 693 obliquity sensitivity ( $S_{obl}$ ). d) Meridional SST gradients between ODP Sites 1239 (1°S, 82°W;  
 694 Etourneau et al. 2010) and 1237 (16°S, 76°W; Dekens et al. 2007) for South Pacific and  
 695 obliquity sensitivity ( $S_{obl}$ ). e) Meridional SST gradients between ODP Sites 1082 (21°S, 12°E;  
 696 Etourneau et al. 2009) and 1090 (43°S, 9°E; Martínez-García et al. 2010) for South Atlantic and  
 697 obliquity sensitivity ( $S_{obl}$ ). The thick black lines are  $S_{obl}$ .

698

699 **Figure 7. Phase analysis of SST- $\delta^{18}O_{sw}$  and SST- $CO_2$  concentrations at the 100-kyr**  
 700 **eccentricity scale over the past 4 Ma.** A typical confidence interval is  $\pm 6^\circ$ . The  $\delta^{18}O_{sw}$  data are  
 701 from Rohling et al. (2021), atmospheric  $CO_2$  concentrations are obtained from reconstruction  
 702 based on leaf wax  $\delta^{13}C$  at IODP Site U1446 (0-1.46 Ma; Yamamoto et al., 2022). Note that the  
 703 red symbols and lines indicate SSTs in the NH, the green symbols and lines indicate SSTs in the

704 equatorial regions, and the blue symbols and lines indicate SSTs in the SH. The time is based on  
705 the centre of the window.

706

707 **Figure 8. Recurrence plots of Global ice volume ( $\delta^{18}\text{O}_{\text{sw}}$ ) and STG in the NH and SH.** a-e

708 show recurrence plots of  $\delta^{18}\text{O}_{\text{sw}}$  (global ice volume), STG in the North Atlantic, STG in the  
709 North Pacific, STG in the South Atlantic and STG in the South Pacific, respectively. The red  
710 arrows on top of the recurrence plots indicate points in time when transitions occurred.

711 Transitions in these recurrence plots from darker areas to white areas indicate prominent systems  
712 changes (Marwan et al., 2007), see also Section 2.5 for further details. Global ice volume shows  
713 changes at  $\sim 2.7$  and  $\sim 1$  Ma. NH STG (ODP 662-982; ODP 846-1012) show a change at  $\sim 2.7$   
714 Ma; SH STG (ODP 1239-1237; ODP 1082-1090) show a change at  $\sim 1$  Ma. Note that subplot b  
715 has a different time axis than all the other subplots due to the lack of SST data between  $\sim 1.5$ - $0.5$   
716 Ma of ODP 662 in the North Atlantic.

**Figure 1.**

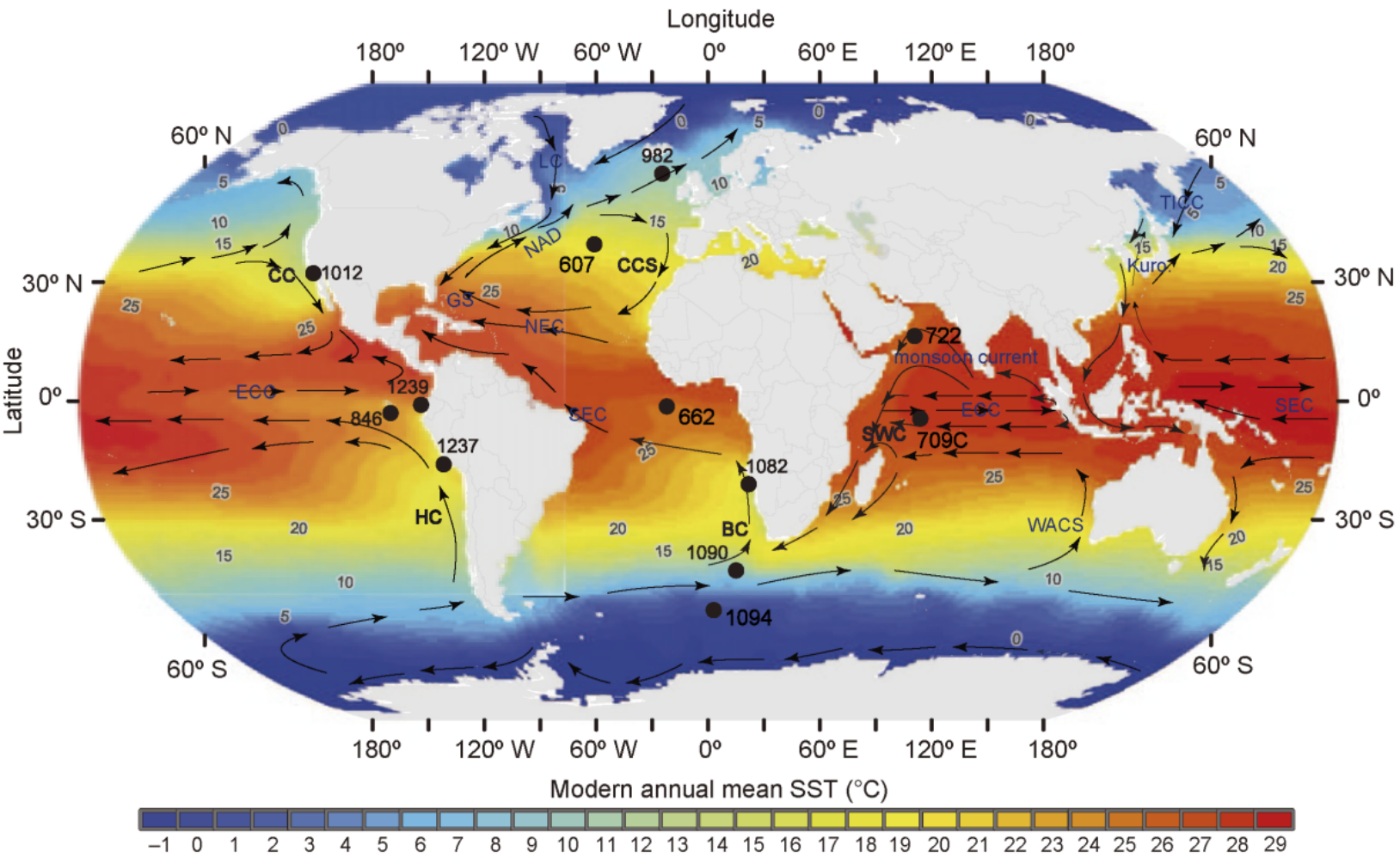


Figure 2.

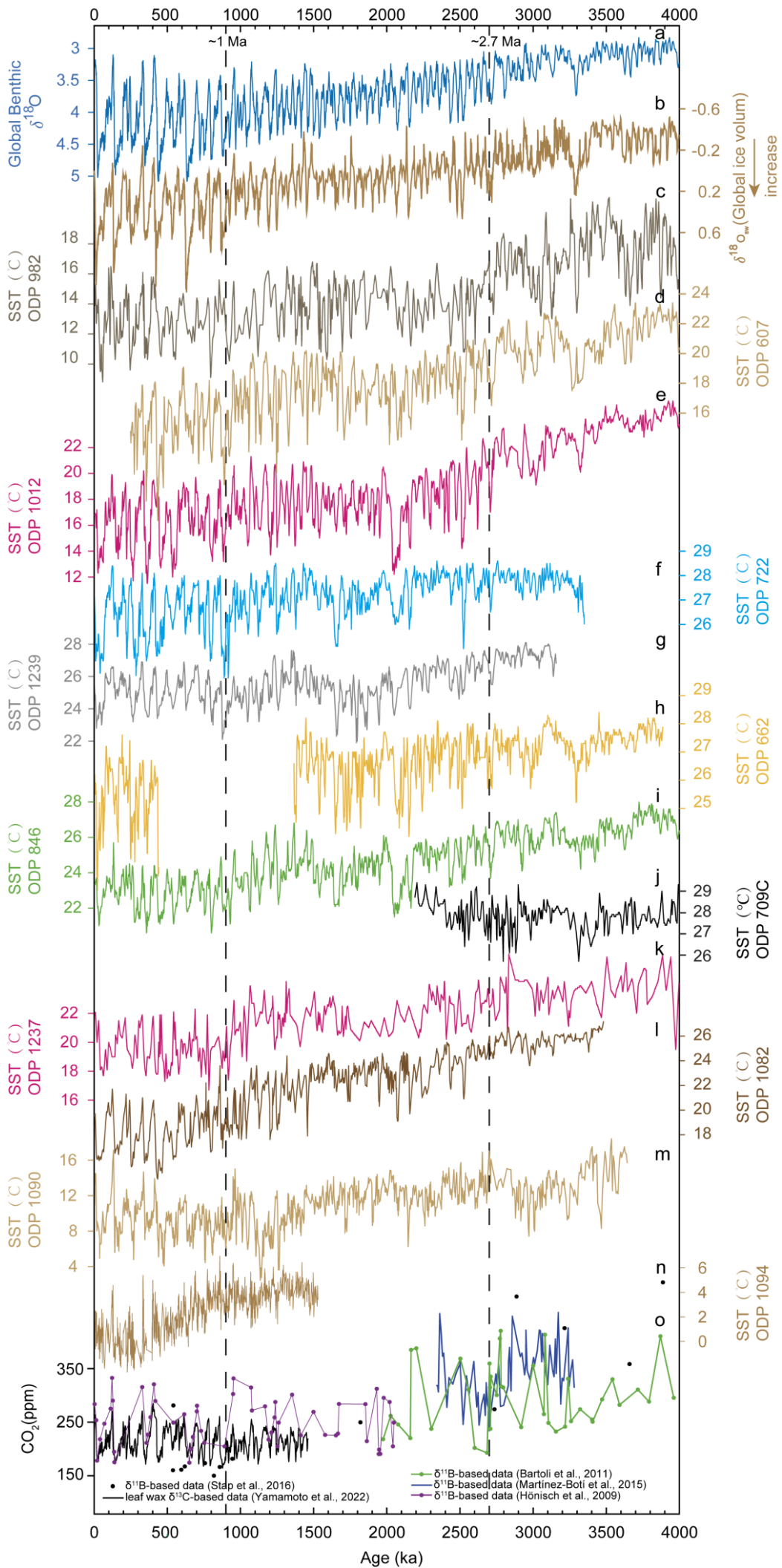


Figure 3.

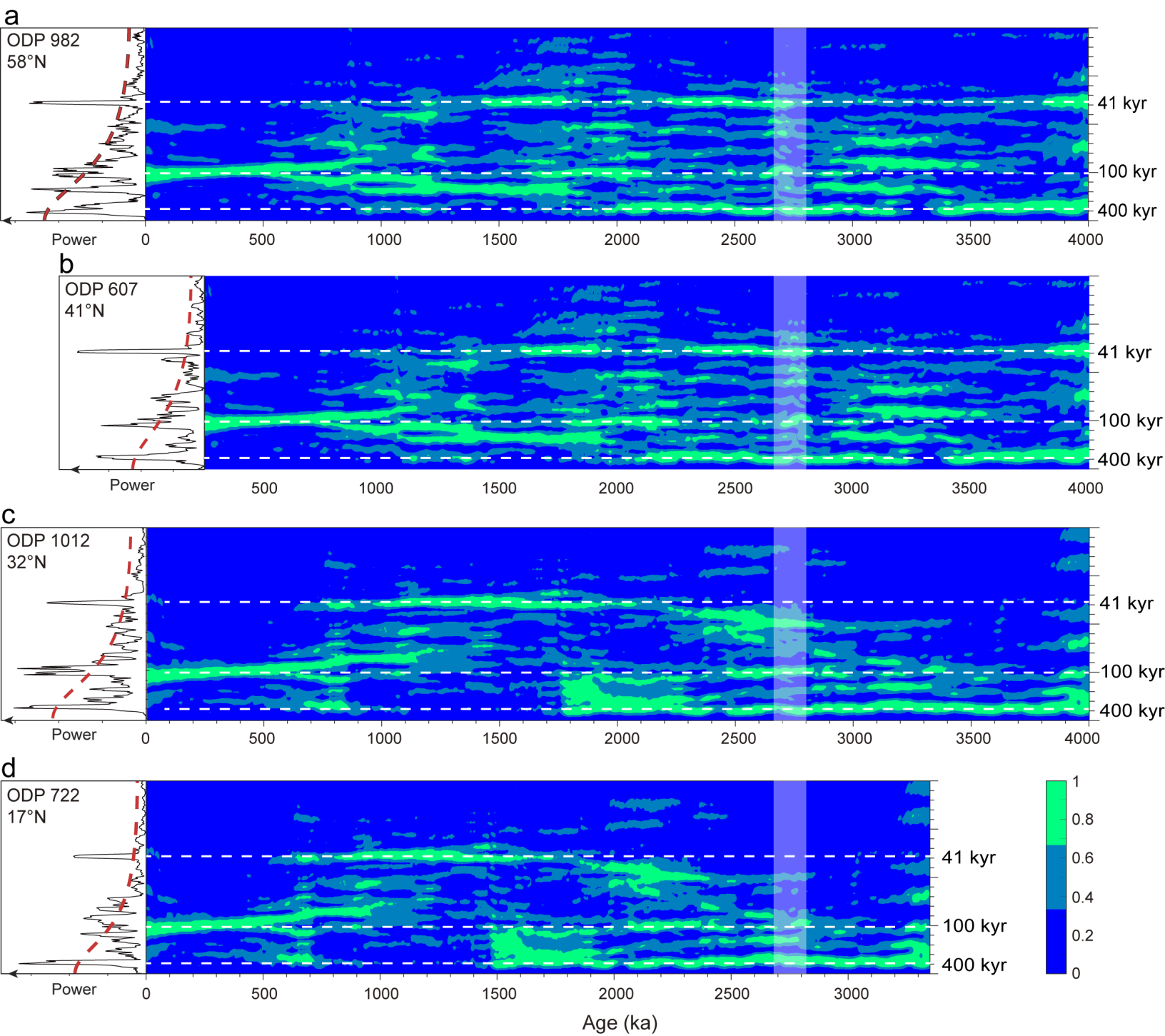


Figure 4.

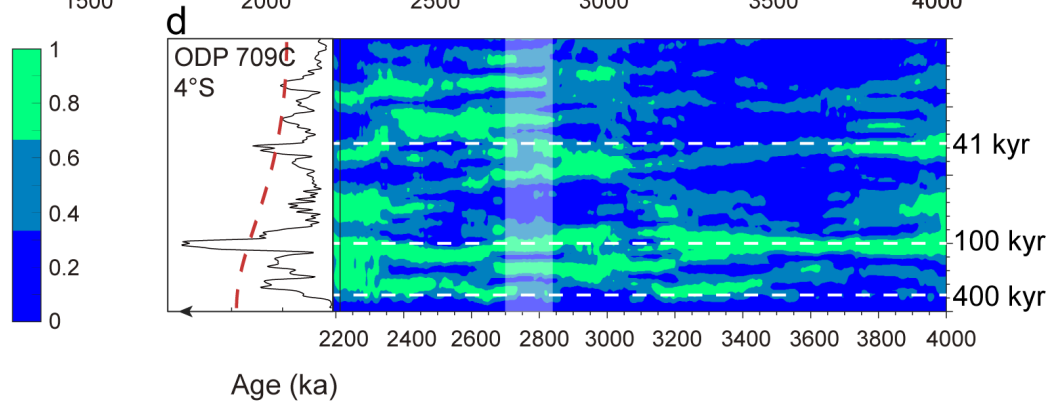
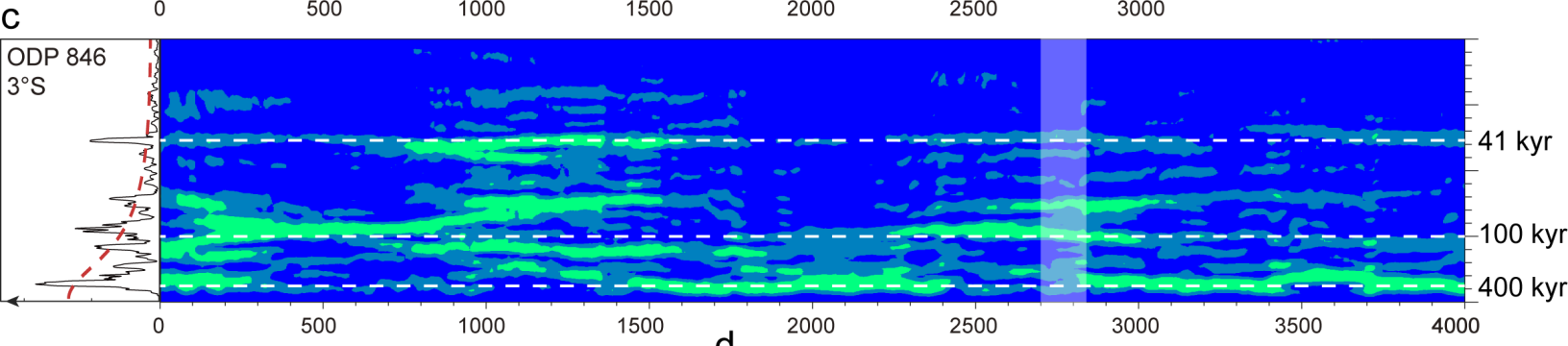
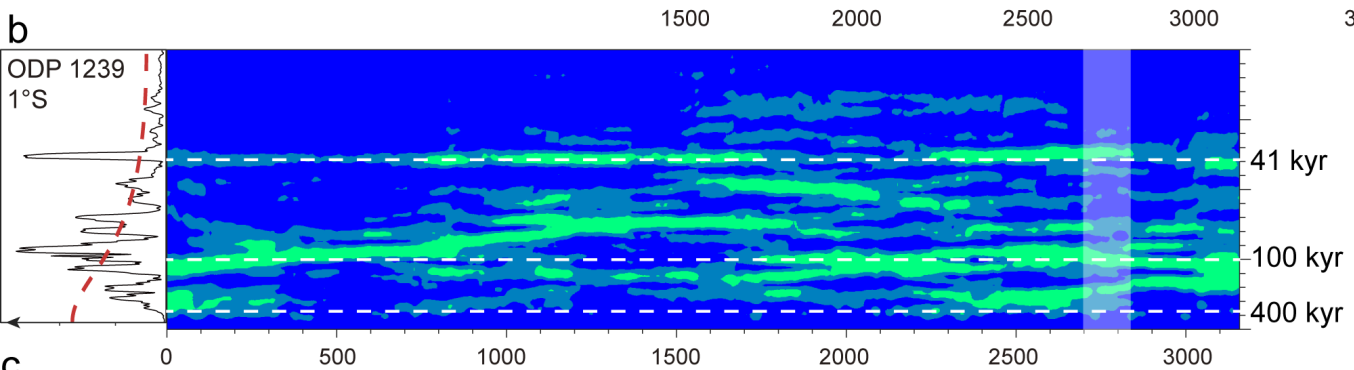
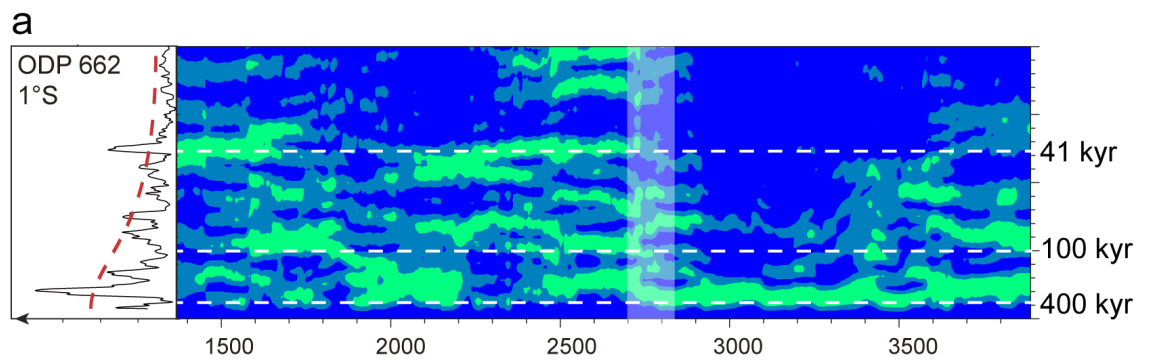


Figure 5.

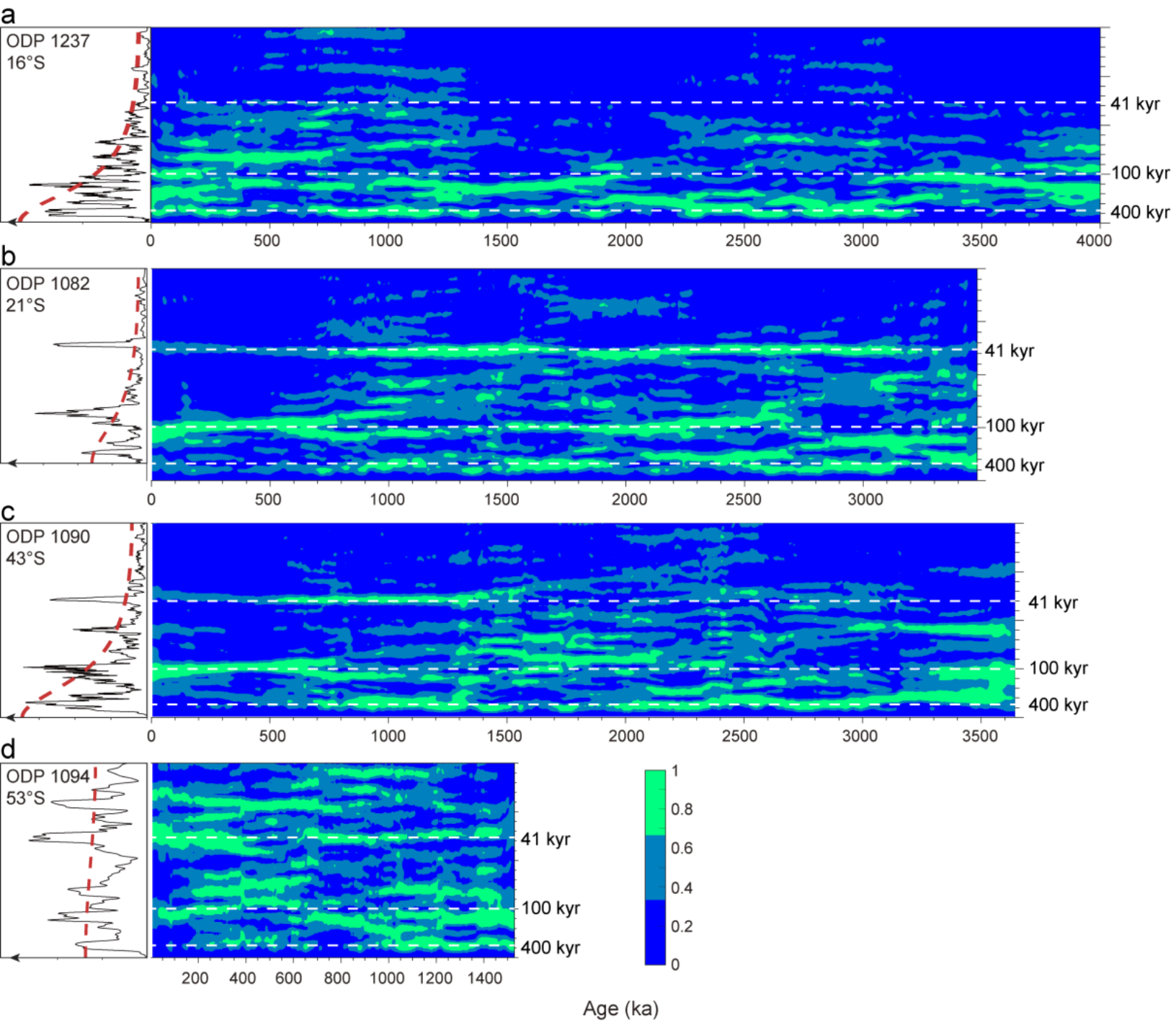


Figure 6.

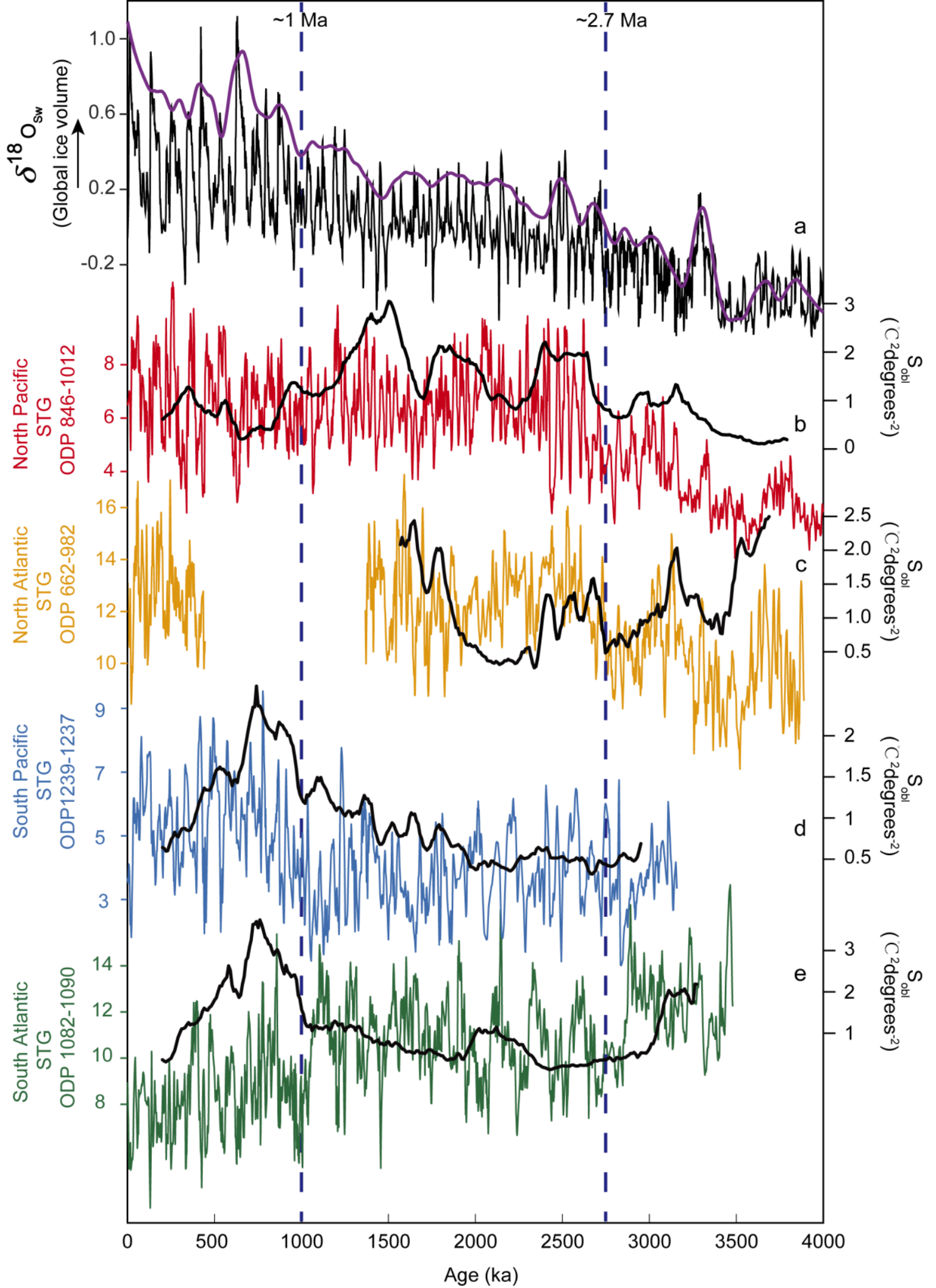


Figure 7.

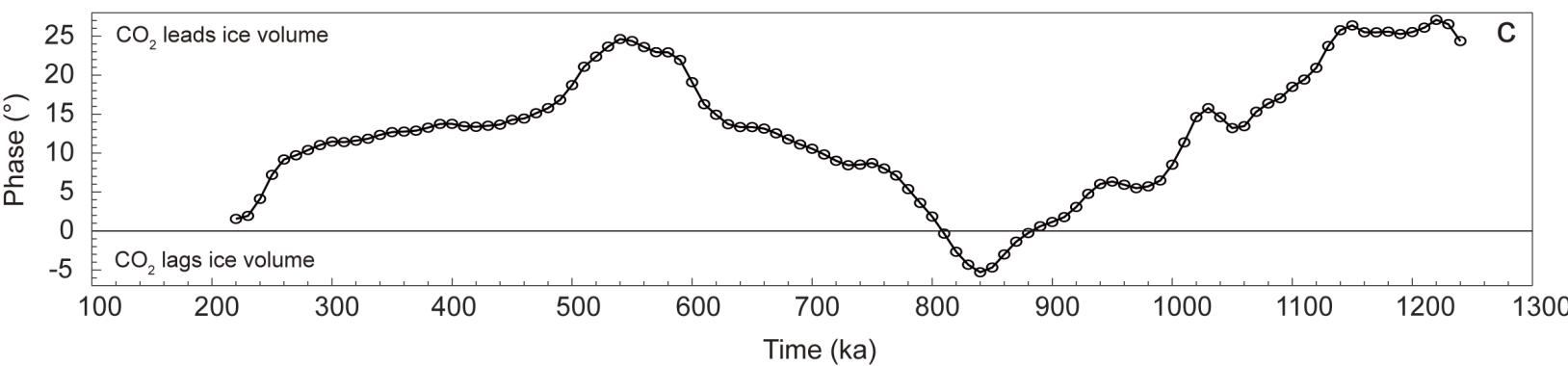
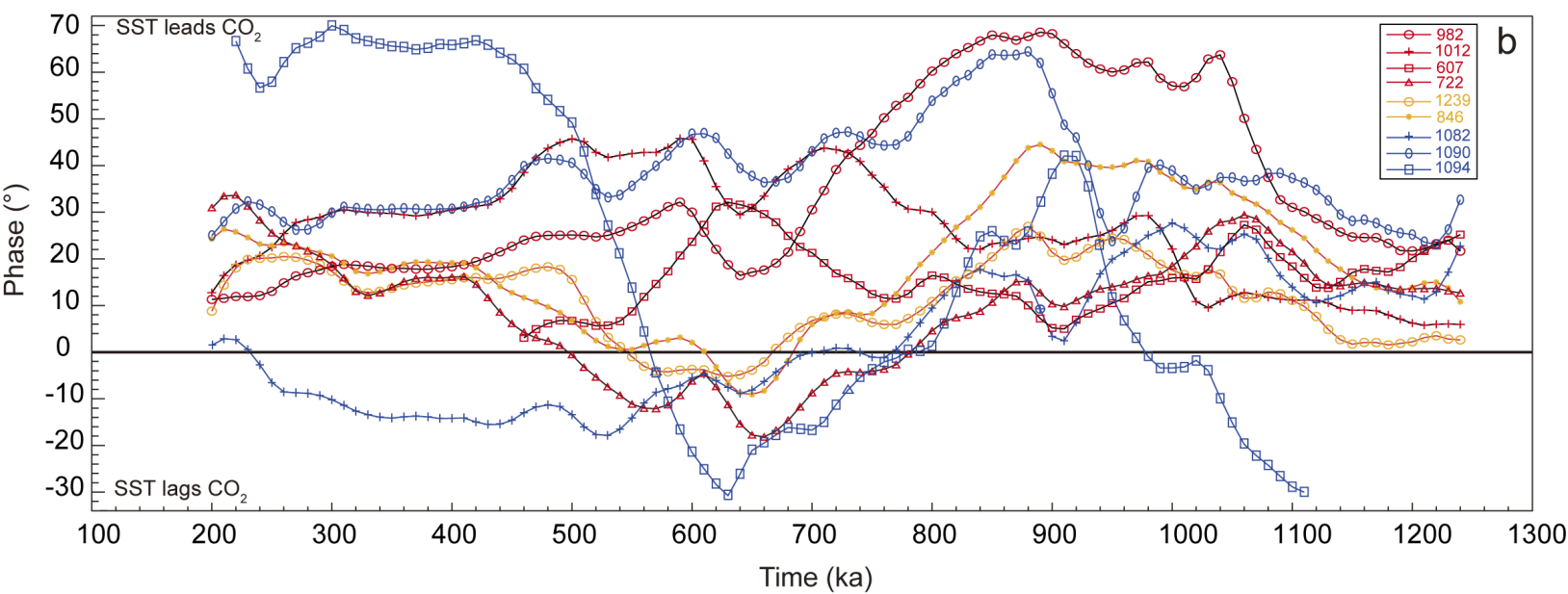
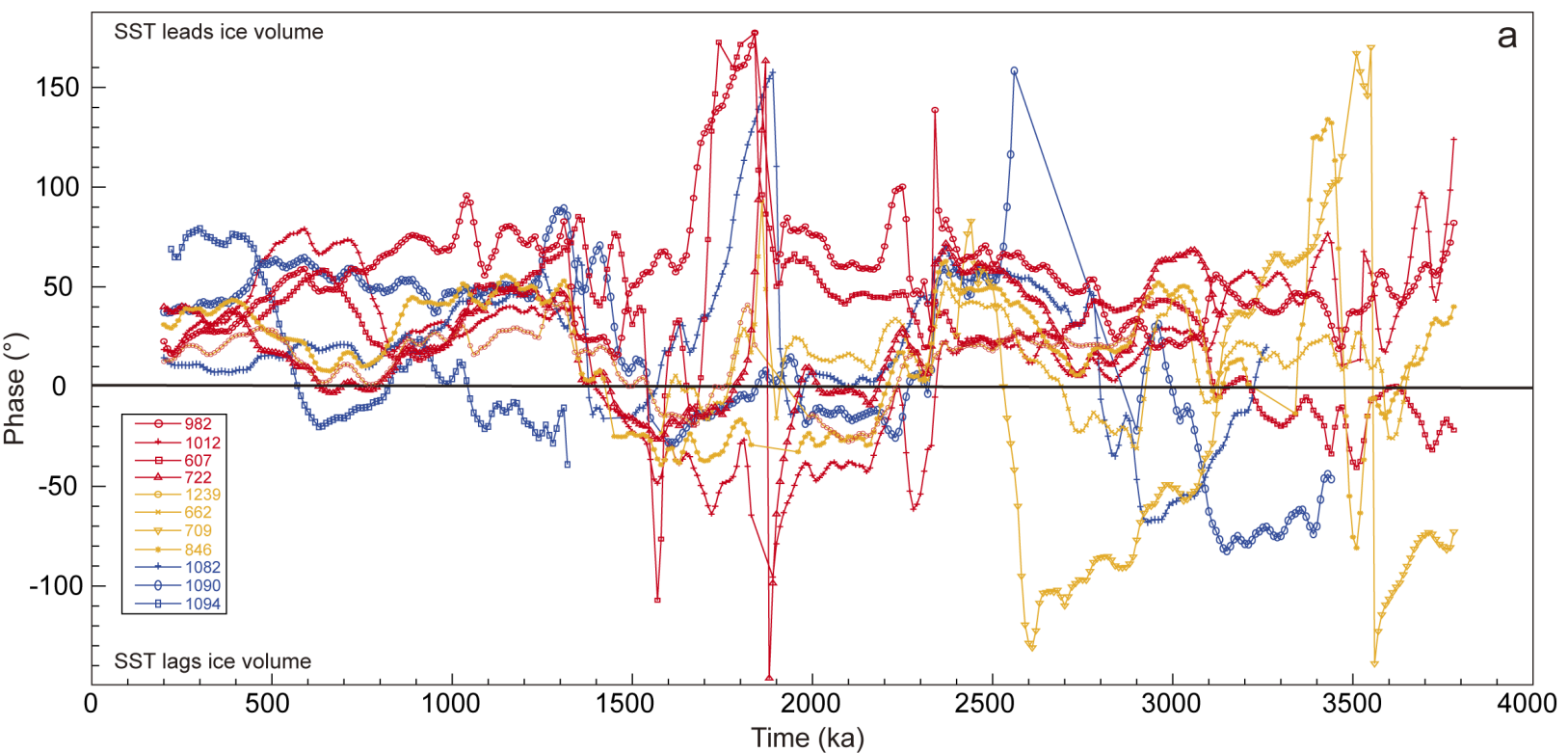
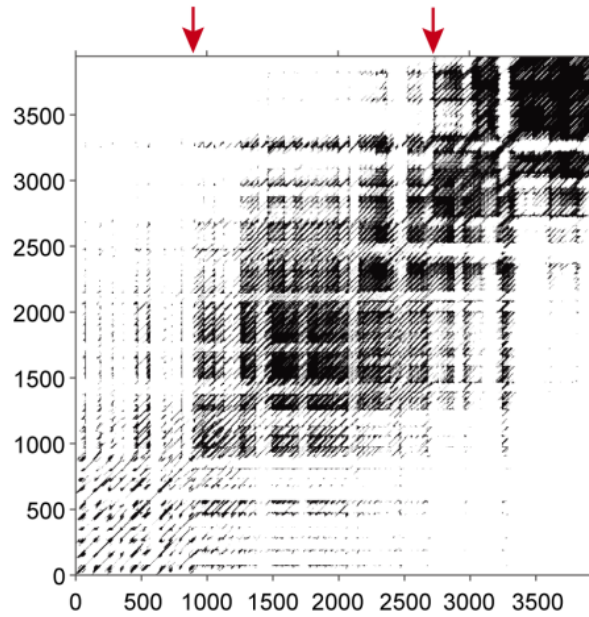
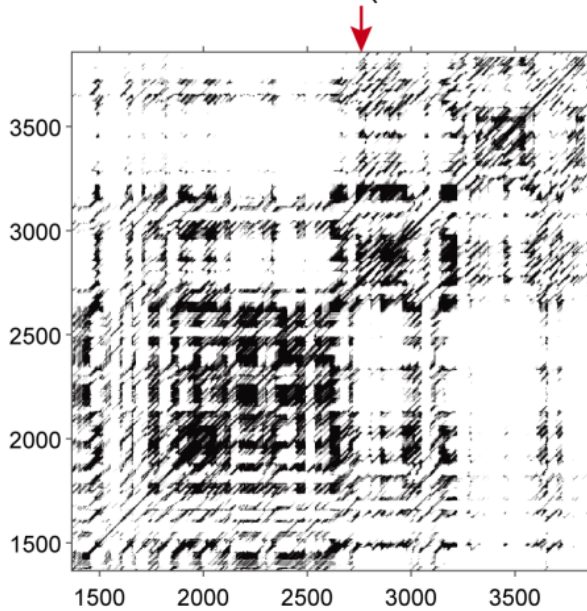


Figure 8.

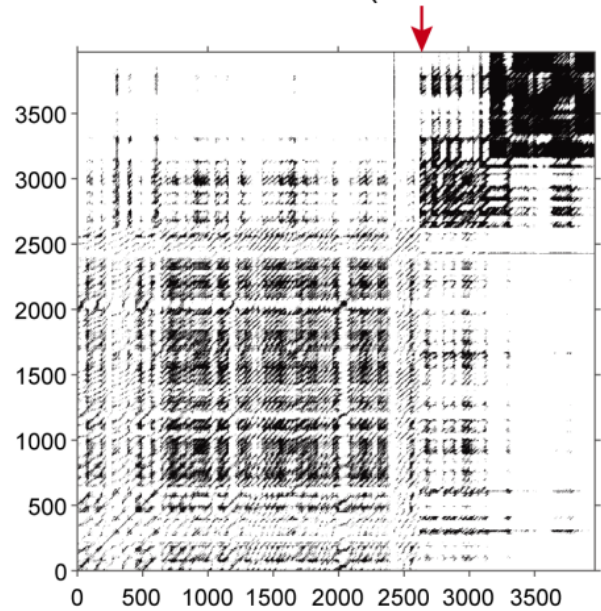
a Global ice volume



b North Atlantic STG (ODP 662-982)

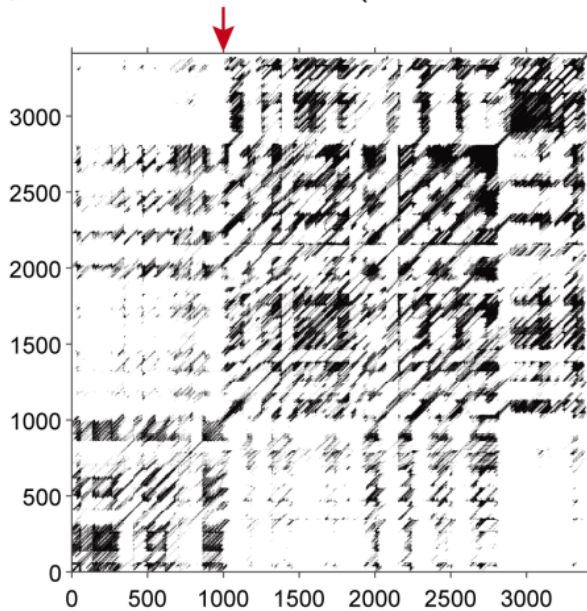


c North Pacific STG (ODP 846-1012)

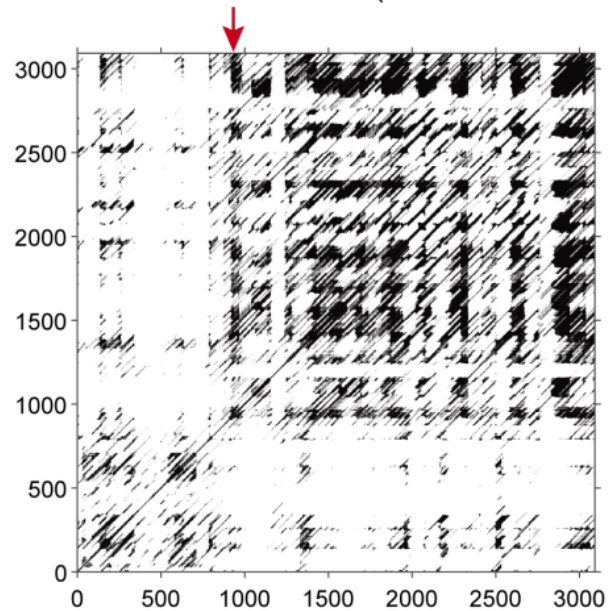


Age (ka)

d South Atlantic STG (ODP 1082-1090)



e South Pacific STG (ODP 1239-1237)



Age (ka)