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Late Miocene cooling coupled to carbon dioxide with Pleistocene-like climate sensitivity

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Earth's climate cooled markedly during the Late Miocene from 12 to 5 million years ago, with far-reaching consequences for global ecosystems. However, the driving forces of these changes remain controversial. A major obstacle to progress is the uncertainty over the role played by greenhouse gas radiative forcing. Here we present boron isotope compositions for planktonic foraminifera, which record carbon dioxide change for the interval of most rapid cooling, the Late Miocene Cooling event between 7 and 5 Ma. Our record suggests that CO2 declined by some 100 ppm over this two-million year-long interval to a minimum at approximately 5.9 Ma. Having accounted non-CO2 greenhouse gasses and slow climate feedbacks, we estimate global mean surface temperature change for a doubling of CO2 - Equilibrium Climate Sensitivity - to be 3.9˚C (1.8–6.7 ˚C at 95% confidence) based on comparison of our record of radiative forcing from CO2 with a record of global mean surface temperature change. We conclude that changes in CO2 and climate

were closely coupled during the latest Miocene and that Equilibrium Climate Sensitivity was within range of estimates for the late Pleistocene, other intervals of the Cenozoic, and the 21st century as presented by the Intergovernmental Panel on Climate Change.

Carbon dioxide (CO_2) is a powerful greenhouse gas. Changes in its atmospheric concentration drive major changes in global temperature and climate state on many timescales[1–3]. Model simulations and (palaeo)climate observations suggest that for a doubling of atmospheric CO_2 concentrations from the pre-industrial baseline, Earth's surface temperature will increase by 1.5 to 4.5˚C in response to CO_2-driven direct radiative forcing and its amplification by positive feedbacks associated with sea-ice extent, atmospheric water vapour loading and clouds[4,5]. However, changes in the nature of these climate feedbacks and boundary conditions greatly influence this relationship and potentially are a major source of uncertainty associated with the range of global mean surface temperatures predicted for the end of this century[6–8]. Previous attempts to constrain climate sensitivity have relied heavily on data from colder-than-present climate states and are thus potentially limited in their applicability to a warming world[9,10]. One way to better understand the nature of climate sensitivity and its dependence on background climate state is to study the geological record of warmer-than-present climate states[5,11]. A prime example is the Late Miocene (~12–5 Myrs ago): a warmer than present climate, but one for which decoupling between CO_2 and temperature has been suggested (Figure 1)[12–15].

Alkenone-derived sea surface temperature records show that, between 12 and 10 Myrs ago, sea surface temperatures (SST) were approximately 5˚C to 15°C warmer than today at tropical and high latitude sites (>60˚N) respectively, followed by an interval of sustained cooling at all latitudes culminating in the Late Miocene Cooling (7-5 Ma, LMC; Figure 1e)[16].

This global cooling appears to have triggered glacial advances on Greenland and it is suggested to have had profound implications elsewhere in driving drying and ecosystem change across large areas of Africa, Asia, and the Americas[17–19]. Yet, the underlying cause of this dramatic change in Earth's global climate state and the terrestrial ecosystem response is poorly understood and vigorously debated[15,19–21]. A reasonable working hypothesis is that these events were driven by changes in the geochemical carbon cycle because Late Miocene cooling occurred gradually and globally (Figure 1e)[16]. A major shift towards lower values in the benthic foraminiferal carbon isotope record from ~7.7–6.5 Ma (the Late Miocene Carbon Isotope Shift) and a steepening of the gradient between planktic and benthic foraminiferal $\delta^{13}C$ values after ~7 Ma also indicate major changes in carbon cycling, such as potential increases in marine primary production and deep ocean storage of carbon, though the specific mechanisms remain unclear (Figure 1e)[21–23].

While a growing data set supports the long-postulated influence of changing atmospheric CO_2 concentration on long-term changes in Earth's climate for the Late Cretaceous and early Cenozoic[24], many existing records imply that the opposite is true for the Late Miocene and Early Pliocene, with some going so far as to suggest a complete decoupling of changes in CO_2 from global climate in the Late Miocene (Figure 1b)[12,15,24]. The main problem limiting progress is the sparse CO_2 data coverage in this interval[25,26]. For the LMC, the currently available data are ambiguous as to whether there was a substantial change in CO_2. Recent development of the phytoplankton and C_3-plant based proxies have produced high-resolution CO_2 records for the Late Miocene and are suggestive of some change in CO_2 during the Late Miocene Cooling minimum[27–29]. However, these records are associated with large uncertainties, highlighting the need for more precise high-resolution records[27]. Here we use the boron isotope technique to reconstruct CO_2 and follow the approach of ref. 30 to examine the relationship between climate forcing by CO_2 change and latest Miocene temperature at high temporal resolution. This allows us to quantify the relationship between climate and CO_2 forcing 7 to 5 million years ago for the first time.

A high resolution record of late Miocene CO_2 change

We used the isotopic ratio of boron ($\delta^{11}B$) of the surface-dwelling foraminifera *Trilobatus trilobus* (*T. trilobus*) to reconstruct surface ocean pH and atmospheric CO_2 concentrations for the latest Miocene (7-5 Ma, Figure 2) at a resolution of 1 sample per ~40 kyrs, from Ocean Drilling Project (ODP) Site 926 (3˚ 43’ 16.49” N and 42˚ 54’ 47.83” W, water depth of 3,598 m, Supplementary Figure 1). Using recently published estimates of the evolution of Neogene Dissolved Inorganic Carbon (DIC) and the isotopic composition of boron in seawater ($\delta^{11}B_{sw}$)[12,31], we reconstruct absolute CO_2 from our $\delta^{11}B$ record (see Methods for more detail).

Our CO_2 estimates are in the same range (Figure 2c) as those estimated from sparse pre-existing $\delta^{11}B$-derived data (280-440 ppm), but our higher temporal resolution reveals considerably more structure than previously documented. Our $\delta^{11}B$ record shows a ~1.2‰ increase, equivalent to a ~100 ppm reduction in CO_2 and ~2 W m^{-2} reduction in radiative forcing of CO_2 (ΔF_{CO2}), from 6.2 to 5.9 Ma (Figure 2a, b and d). Overall, the mean $\delta^{11}B$ increases from 17.80‰ between 7 and 6 Ma to 18.22‰ between 6 and 5 Ma (Figure 2a), equivalent to a significant change in mean CO_2 from ~360 ppm to ~330 ppm (Student’s t-test, $p < 0.01$). The data therefore suggest an overall reduction of ~30 ppm in mean CO_2 from 7 to 5 Ma with a pronounced minimum at ~6 Ma (Figures 2a, c and d).

As in other $\delta^{11}B$ studies for this time interval[12,31,32], changes in $\delta^{11}B_{sw}$ are the largest source of uncertainty in our absolute CO_2 estimates. Proxy reconstructions propose that $\delta^{11}B_{sw}$ was within the range of 39-40‰ during the latter half of the Late Miocene[31,33]. Supplementary Figure 2 illustrates the sensitivity of our absolute CO_2 and ΔF_{CO2} estimates to this range in $\delta^{11}B_{sw}$ at 6.4 Ma, 5.9 Ma and 5.0 Ma. This treatment confirms that ΔF_{CO2} shows minimal sensitivity to $\delta^{11}B_{sw}$, illustrates that CO_2 is well constrained to fall between 280 and 500 ppm (see Methods, Supplementary Figure 2), and indicates that the reconstructed amplitude of CO_2 decline across the study interval is robust.

Our estimates of CO_2 during the Late Miocene are near or below the hypothesised thresholds for the Late Miocene C_4 expansion (200–500 ppm) and divergence of coccolithophore size and $\delta^{13}C$ signatures (~375–500 ppm)[12,26,34–36]. Recently published high resolution phytoplankton[27] and C_3 plant[29] derived CO_2 reconstructions document a similar CO_2 minimum centred on ~6 Ma and similar absolute CO_2 levels (Figure 1b; 2c)[27,37,38]. While absolute estimates from recent phytoplankton CO_2 estimates from 7–5 Ma agree fairly well with the absolute estimates using boron isotopes presented in this study (Figure 2c)[27], the C_3 plant derived CO_2 estimates for this time slice are on average ~100 ppm lower, though both are within error (Figure 2c). This is possibly attributable to uncertainties in $\delta^{11}B_{sw}$ for the $\delta^{11}B$ proxy (see above, Supplementary Figure 2)[31] and/or differences in Late Miocene hydroclimate influencing the C_3 plant-based proxy, particularly if any samples were affected by water stress[39]. The similarity in the concentrations calculated across a range of $\delta^{11}B_{sw}$ values, and broad agreement with the phytoplankton $\delta^{13}C$-based CO_2 record of ref. 27, increases confidence in our absolute CO_2 estimates and in the reconstructed CO_2 minimum at ~6 Ma and limits the range in absolute CO_2 for this minimum to between 250 (± 70) ppm and 330 (± 90) ppm. This marked change in CO_2 correlates with the LMC, changes in the carbon cycle tracked by $\delta^{13}C$ and inferred expansion of the Greenland ice sheet[17,22,23,40–42]. Together these lines of evidence suggest at least some degree of coupling between CO_2 and climate during the latest Miocene. This finding contrasts with those of previous studies in which CO_2 and climate were inferred to have decoupled during the late Miocene[12,13,15].

Strong latest Miocene CO_2-climate coupling

The high fidelity of the boron isotope-based estimates of change in the radiative forcing of CO_2 irrespective of changes in $\delta^{11}B_{sw}$ (Supplementary Figure 2), coupled with the improved temporal resolution provided by our $\delta^{11}B$ record allows us to examine the sensitivity of Earth's climate system to CO_2 during an interval of change when Earth's global climate was warmer than today[16]. By using the approach described in ref. 30 to calculate ΔF_{CO2} from changes in $\delta^{11}B$, uncertainties in $\delta^{11}B_{sw}$ and DIC do not add to the uncertainty in ΔF_{CO2} (Supplementary Figure 2), and the influence of temperature uncertainty is greatly reduced (see ref. 30 for details). To better observe long term trends, we smooth the ΔF_{CO2} record to remove short term variability using a spline function (Figure 2d).

To examine climate-CO_2 coupling, we first estimate global mean surface temperature change (ΔGMST) from 7 to 5 Ma, relative to the modern mean. This was achieved by creating a stack of alkenone-derived SST change (ΔSST) records (relative to the modern mean annual SST), with original SST values recalibrated with the BAYSPLINE model[43] to improve estimates at high temperatures and records checked to ensure that values were not compromised by alkenone saturation (see Methods)[44]. We used the ΔGMST:ΔSST ratio of 1.5 (1.3–1.9 at 95% confidence) to estimate ΔGMST[45,46]. This ratio was taken from multiple model simulations for the mid-Piacenzian Warm Period (3–3.0 Ma), a climate similar in terms of global ΔSST to the late Miocene. This treatment suggests that global surface temperatures were 2–6 ˚C warmer in the latest Miocene than the pre-industrial, in broad agreement with previous studies (Figure 2g,f)[16].

To determine the sensitivity of global temperature to CO_2 forcing in the latest Miocene, we regress ΔGMST against smoothed ΔF_{CO2} (see Methods). The $\Delta GMST/\Delta F_{CO2}$ regression slope describes the average change in temperature in ˚C (ΔT) per watt per square metre of forcing (ΔF), otherwise referred to as the climate sensitivity (˚C $W^{-1}m^2$) (Figure 3a, bold black line). Because we are using palaeoclimate data, our calculated climate sensitivity incorporates all feedbacks, both slow and fast, and is thus defined as the 'palaeo-sensitivity' (S_p, ˚C $W^{-1}m^2$)[9]. By multiplying S_p by 3.7 W m^{-2} (the change in radiative forcing from a doubling of CO_2) we calculate Earth System Sensitivity (ESS, mean warming expected for a doubling of CO_2 following action of all climate feedbacks)[9]. Variations in ESS between different time intervals are to be expected and can be attributed to the presence/absence of climate feedbacks, such as the presence/absence of large continental ice sheets, and boundary conditions such as global vegetation distribution[9,47]. Note that the inclusion of all climate feedbacks makes both S_p and ESS different to true climate sensitivity (S_a, ˚C $W^{-1}m^2$) or Equilibrium Climate Sensitivity (ECS, the warming expected for a doubling of CO_2, following the action of only fast climate feedbacks). This distinction is important in the context of discussions of anthropogenic climate change, which is concerned primarily with ECS given that it encompasses climate changes and feedbacks on human time scales.

We estimate ESS for the latest Miocene to be 9.3 ˚C per CO_2 doubling (5.5–13.1 ˚C at 95% confidence; Figure 3a). This broadly overlaps with the range of ESS estimates for the Late Pleistocene (6–9 ˚C per CO_2 doubling, Figure 3)[8,11,48–51]. The similarity to Late Pleistocene estimates, a climate system associated with the presence of significant slow acting climate feedbacks from changing ice sheet albedo and vegetation, suggests, in relative terms, a similarly significant contribution from slow feedbacks during the latest Miocene.

There are several potential amplifying slow feedbacks that may have operated in the latest Miocene. The appearance of ice rafted debris (IRD) in northern basins of the Atlantic, changes in sedimentation, and cosmogenic beryllium and aluminium isotopes around the Greenland coast from 7 Ma onwards suggest a contribution from land snow/ice albedo feedbacks at this time[17,40–42,52]. Elsewhere, high Late Miocene ESS estimates have been attributed to the influence of Late Miocene geography (e.g. potentially reduced elevation of various mountain ranges like the Andes and North American Rocky Mountains), ocean gateways (e.g. closure of the Atlantic-Mediterranean exchange), and vegetation cover (e.g. expansion of grasslands), supported by proxy-based reconstructions[53–57]. Model simulations of the Late Miocene suggest an expansion of grasslands at high latitudes and reduction in deciduous tree cover with declining CO_2[54]. This would increase global albedo, which, when combined with moderate ice sheet forcing, could explain large sensitivities observed[54]. Additionally, closure of the Atlantic-Mediterranean gateway is proposed to enhance North Atlantic overturning and increase interactions between deep and surface ocean reservoirs making the high latitude climate more sensitive to changes in CO_2[57]. Attempts to disentangle the relative roles of palaeogeography and vegetation on climate sensitivity suggest that late Miocene vegetation distribution has a three times stronger impact on global temperature than palaeogeography during this time[54,55]. Therefore, although the exact cause remains uncertain, there are several potential reasons why ESS was elevated during the Late Miocene, and importantly, they are all consistent with strong climate feedbacks coupling changes in CO_2 to the global climate in the Late Miocene.

Constraints on Equilibrium Climate Sensitivity

To determine ECS during the Late Miocene we must account for the slow feedbacks discussed above including the non-CO_2 greenhouse gases. We can assume that vegetation and land ice albedo feedbacks had meaningful effects on global climate but given the lack of information on the appropriate magnitude of the various feedbacks involved, we are unable to quantify them from the geological record in the same way as is possible for the Late Pleistocene[58].

We therefore apply an ECS:ESS ratio of 1.66 (1.11–2.85 at 95% confidence) as estimated across multiple model simulations (n = 16) for the mid-Piacenzian Warm Period (2.97–3.29 Ma) to estimate latest Miocene ECS from ESS[59] to account for all the slow climate feedbacks (e.g. sea-ice and cloud albedo, water vapour, vegetation, and aerosols)[10,59]. The mid-Piacenzian Warm Period is similar in terms of temperature and continental arrangement and overall climate to the latest Miocene. By using the mean ECS:ESS ratio from a suite of models, we account for a wide variation in climate feedbacks. Thus, in the absence of more robust climate models for the Late Miocene and scant proxy reconstructions of slow climate feedbacks, this method provides an appropriate estimate for ECS from ESS. This is supported by the observation that the estimated ECS:ESS for the mPWP being within range for the ECS:ESS from a late Miocene model simulation (1.4, ref. 53). We then use the relationship between ΔF_{CO2} and radiative forcing from all greenhouse gases (ΔF_{GHG}) defined by: $\Delta F_{GHG} = \Delta F_{CO2} \times 1.4$ (±0.1), from ref. 60 (recently confirmed by ref. 61).

This treatment of our data yields a latest Miocene ECS of 3.9 ˚C warming per doubling of CO_2 (1.8–6.7 ˚C at 95% confidence, Figure 3b). This value falls in the range of published estimates for the Late Pleistocene, those suggested by the IPCC for the 21st century, and other intervals of Earth history (Figure 3c). In the larger geological context, this finding implies that the scaling of fast feedbacks to radiative forcing from CO_2 is broadly consistent across different climate states, at least up to climates 2–6 ˚C warmer than the pre-industrial. In other words, our data suggest that there is little detectable state dependency to ECS in the latest Miocene.

Implications

Our CO_2 and resulting ΔF_{CO2} records are the best resolved records from the Late Miocene to date and reveal a decline of ~100 ppm at ~6.0 Ma associated with an interval of global cooling. Contrary to previous suggestions, we find that latest Miocene climate was strongly coupled to CO_2 (see Figure 3a)[12–15]. The ESS we calculate for the late Miocene is like that determined for the Late Pleistocene and points to a relatively strong slow feedback component (e.g., land ice/snow fields and/or vegetation). Using a ratio of ECS:ESS derived from multiple model experiments to account for slow feedbacks, and a scaling factor to calculate ΔF_{GHG} from ΔF_{CO2}, we estimate ECS to overlap with the IPCC range for the 21st century. This implies that the fast feedbacks between CO_2 and global climates operate with a consistent efficacy at least for the last 7 million years with no resolvable state dependency in slightly warmer climates than today. Thus, the IPCC range of estimates of ECS (1.5 to 4.5 ˚C per doubling) is likely appropriate for warming of ~2–6 ˚C.

Author contributions

RMB and TBC provided primary data based on their laboratory work. GLF, TBC, and PAW conceived the project. RMB produced the first draft of the manuscript and all authors contributed to the final text. RMB performed all statistical data analysis and compiled the data with input from AJC on chronology.

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Data Availability

The experimental data are available in the Pangea public repository XXXXX.

Competing Interests

The authors declare no competing interests.

Figures

Figure 1: Neogene and Late Miocene climate changes. a) Benthic foraminifera $\delta^{18}O$ stack for the Neogene[97]. **b)** Collection of published CO_2 estimates for the last 20 Myr using phytoplankton ($\delta^{13}C$ of phytoplankton compounds, blue)[13,25–27,37,38,98–101], boron isotopes (orange)[12,32,50,68,98,100,102–105], C_3 plants (green)[29] , paleosols (teal)[106–110] and stomata (purple)[111–113] . Error bars denote reported 2sd. Data compilation built on previous CO_2 compilation by ref. 35 **c)** 20-point running mean $\delta^{18}O$ and **d)** $\delta^{13}C$ of benthic foraminifera

records from ODP 999 (green)[114], IODP U1338 (light blue)[22], ODP 982 (red)[40] and ODP 1147 (dark blue)[21]. An equilibrium correction (+0.64‰) applied to all $\delta^{18}O$ records[115]. **e)** SST records used in Late Miocene temperature stack recalibrated using BAYSPLINE[43]. Records are coloured according to latitude with >50˚N (pink)[16], 30-50˚N (yellow) [16], the tropics (green)[16,87–89], and 30–50˚S (purple)[16]. Site information and citations for all datasets are available in Supplementary Table 1.

Figure 2: Records of late Miocene climate change. **a)** $\delta^{11}B$ of *T. trilobatus* (this study, blue circles). Dashed line represents mean $\delta^{11}B$ for 7–6 Ma and 6–5 Ma. Previous planktic foraminifera $\delta^{11}B$ data from ODP 926 (red circles) and ODP 1000 (red triangles)[12]. Error bars denote 2sd. **b)** Mg/Ca-SST estimates from ODP 926 (filled blue circles, this study), ODP 1146 (yellow[21]; orange[81]) and IODP U1338 (empty blue circles)[23]. **c)** CO_2 estimates (this study, ble filled circles) using $\delta^{11}B$ (data in 2a). Previous $\delta^{11}B$ - CO_2 estimates from ODP 926 (red circles) and ODP 1000 (red diamonds)[12]. Error bars denote 2sd. Previous estimates using phytoplankton as shown in 1b (light purple circles, error bars denote 2sd) and recently published phytoplankton estimates (light purple squares, errors omitted but displayed in Figure 1b)[27]. **d)** Radiative forcing of CO_2 (ΔF_{CO2}) using $\delta^{11}B$ (data in 2a, blue filled circles) with smoothing spline (bold blue line, see Methods). **e)** ΔSST stack. **f)** ΔGMST stack. Error bands encompass 68% (dark red/blue) and 95% (light red/blue) of 10,000 Monte Carlo simulations for $\delta^{11}B$-derived CO_2 and ΔF_{CO2} estimates and ΔSST/ΔGMST stacks (see Methods for full details). See Supplementary Table 1 for full ΔSST/ΔGMST output and Supplementary Table 2 for raw data and CO_2/Mg/Ca-SST/ΔF_{CO2} estimates.

Figure 3: Latest Miocene climate sensitivity regression and key climates sensitivity studies throughout the last 70 million years. a) Cross-plot of smoothed ΔF_{CO2} against ΔGMST for the latest Miocene with 95% confidence interval (error bars). Regression lines fitted by SIMEX regression (bold black line) with 95% confidence interval (blue dashed line, see Methods)[117]. b) Probability density function of Latest Miocene ECS calculated when scaling ESS to ECS. Bold, regular and dashed vertical lines denote the median and 66% and 95% confidence. c) Most likely ECS and ESS and their distributions for the present day (ECS: red[5,118]), Last Glacial Maximum (ECS: orange[10,58,119–124]; ESS: pale orange[11]), Pleistocene (ECS: yellow[8,9,49,50]; ESS: pale yellow[48,50,51,125]), Pliocene (ECS: green[50,59,126]; ESS: pale green[50,127,128]), Miocene (ECS: blue[54,129]; ESS: pale blue[54], arrows denote estimates from this study), and the rest of the Cenozoic (>20 Ma, ECS: purple[130–132]; ESS: pale purple[3,9,9,47,133]). Boxes and whiskers represent reported 66% and 95% confidence interval respectively. Black lines represent reported most likely ECS/ESS. Black square denote estimates incorporating modelling data. ECS from the IPCC for the 21st century (1.5–4.5˚C warming per doubling of CO_2 at 66% confidence, red vertical lines). See Supplementary Table 3 for data and references.

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Materials and Methods

Site details

We present a highly resolved (1 sample per ~40 kyrs) $\delta^{11}B$-derived atmospheric CO_2 record from ODP 926 (Ceara Rise, 3˚ 43' 16.49" N and 42˚ 54' 47.83" W, water depth of 3,598 m) where modern surface water is close to equilibrium with the atmosphere, with respect to CO_2 (Supplementary Figure 1b)[62]. Studies have shown that changes in $\delta^{11}B$ at ODP 926 are consistent with variations in $\delta^{11}B$ from other core sites throughout the Neogene, strongly implying that changes in CO_2 are the primary control on $\delta^{11}B$ of borate in seawater at this location (Supplementary Figure 3)[12,32,63].

Sampling and stratigraphy

53 bulk sediment samples were taken at 0.75 m intervals between 151 and 194 composite metres below the seafloor (cmbsf) at ODP 926. We targeted precessionally-paced carbonate-rich sediments to ensure minimal dissolution and the highest number of well-preserved foraminifera retrieved[64]. Approximately 20 cm^3 of core material was taken from the most carbonate-rich horizons, however, from 169–190 cmbsf, the carbonate content of the sediment was lower, hence 40 cm^3 of core material was taken to ensure sufficient sample material.

All samples were dried at 40˚C and weighed before being washed with 15 MΩ cm MilliQ water over a 63 µm sieve. The coarse fraction (>63 µm) of the sediment was dried at 50˚C, and the fine fraction (<63 µm) was dried at 40˚C before being weighed and stored. *Cibicidoides wuellerstorfi* (*C. wuellerstorfi*) and *Trilobatus trilobus* (*T. trilobu*s) were then picked from the 212-500 µm and 300–355 µm size fractions respectively. Only whole, unbroken foraminifera showing no obvious signs of dissolution or alteration were selected to minimise the effects of diagenesis.

We present our data on a previously published age model from ODP 926 based upon astronomical tuning of magnetic susceptibility, XRF and core images[64,65]. As an additional check on the validity of this age model, we generated benthic foraminiferal stable isotope records to compare to other published records. Between 1–6 *C. wuellerstorfi* tests were homogenised, with ~50 µg analysed for stable carbon ($\delta^{13}C$) and oxygen ($\delta^{18}O$) isotopes by a Thermo Scientific Kiel IV Carbonate device coupled with a MAT253 isotope ratio mass spectrometer at the University of Southampton. $\delta^{13}C$ and $\delta^{18}O$ data derived from only one foraminiferal test are marked in Supplementary Table 2. Both $\delta^{18}O$ and $\delta^{13}C$ records captured the characteristic structure of the Late Miocene (Supplementary Figure 4)[64,65]. Comparison of $\delta^{13}C$ from benthic foraminifera at ODP 926 with other high-resolution $\delta^{13}C$ records from cores used in the sea surface temperature stack illustrates strong degrees of similarity supporting the validity of the astronomically tuned age model at ODP 926 (Supplementary Figure 5).

Trace element and boron isotope methodology

About ~300 *T. trilobus* tests from each sample (total mass ~3 mg) were cracked open and any contaminating clays were removed by ultrasonication in a water bath. The tests were then oxidatively cleaned and analysed for $\delta^{11}B$ and a 20 µl aliquot was taken for trace element analysis (e.g. B/Ca, Mg/Ca, Al/Ca) at the University of Southampton following established procedures[66–68]. Boron isotopes were measured on a Thermo Scientific Neptune multicollector inductively coupled mass spectrometer at the University of Southampton according to methods described elsewhere[69]. The external precision of the $\delta^{11}B$ determinations was estimated from the reproducibility of measurements of an in-house standard (Japanese Coral *Porites* = 24.2 ‰)[70] and calculated according to ref. 71.

$$2\sigma = 1.87*exp(-20.6*[^{11}B]) + 0.22*exp(-0.43*[^{11}B])$$ (eq. 1)

where $[^{11}B]$ is the intensity of the ^{11}B signal in volts measured on 10^{12} Ω resistors. We recorded a typical external reproducibility of 2sd = 0.20‰. pH estimates were calculated following equation 2:

$$pH = pK_B* - log(-\frac{\delta^{11}B_{sw} -^{11} B_{borate}}{(\delta^{11}B_{sw} - \alpha_B \times \delta^{11}B_{borate} - (\alpha_B - 1) \times 1000})$$

(eq. 2)

where $\delta^{11}B_{borate}$ represents boron isotopic composition of borate in sea water and was calculated using the *T. trilobus* $\delta^{11}B$-borate calibration from ref. 12, pK_B* is the disassociation constant for boric acid at in situ temperature, salinity and pressure and was calculated using the 'seacarb' package in R[72] using ref. 73,74 , $\delta^{11}B_{sw}$ is the mean $\delta^{11}B_{sw}$ for 7-5 Ma (39.8‰ ±0.3)[31], and α_B is the isotopic fractionation between the two species of boron in seawater, reported as 1.00272 ±0.0006[75].

Elemental analyses were performed on a Thermo-Fisher Scientific Element 2 single collector ICPMS at University of Southampton following established methods[69]. For this study, analytical reproducibility for Mg/Ca was ±5 % (2sd). Sea surface temperatures (SST) were estimated from Mg/Ca values using the calibration of ref. 76 corrected for core depth according to ref. 77 (equation 3), with a correction for seawater Mg/Ca[78] following ƒ92

79 using 0.41 as the power constant[80] (equation 4).

$$SST = ln\left(\frac{Mg/Ca_{trilobus}}{Mg/Ca_{corrected} * 0.37}\right) \div 0.09 + 0.36 \times core\ depth\ (km)$$

(eq.3)

$$Mg/Ca_{corrected} = \left(\frac{Mg/Ca_{sw} * age}{Mg/Ca_{modern}}\right)^{0.41}$$ (eq.

4)

Mg/Ca derived SST estimates are consistent with previously collected Mg/Ca temperature estimates for this time interval[19,30,84], showing an increase of 2˚C from ~6 Ma to ~5 Ma (Figure 2b) and are within the same range as Mg/Ca and U^{K}_{37}' SST estimates published at other low latitude core sites suggesting that our temperature record is consistent with regional trends[21,81]. Trace element data were used to check clay removal efficiency. All samples had Al/Ca ratios of <250 µmol mol^{-1}, and typically <70 µmol mol^{-1}. No correlation was observed between Al/Ca and Mg/Ca, Al/Ca and $\delta^{11}B$ or Al/Ca and B/Ca, confirming that all samples were sufficiently cleaned.

Estimation of CO_2 and ΔF_{CO2}

In the ocean, CO_2 (aq) and pH are closely linked such that pH estimates derived from $\delta^{11}B$ can be used to calculate paleo-CO_2[82]. Although pH and CO_2 are closely coupled, a second carbonate parameter is needed to calculate absolute CO_2[82]. The marine carbonate system is described by four equations with six parameters such that knowledge of any two, as well as temperature, salinity and pressure is required to definitively constrain the system[82]. The parameters easiest to use in this regard are total alkalinity (ALK) and dissolved inorganic carbon (DIC). As pH reflects the ratio of ALK to DIC and any change in DIC (ALK) is counteracted by a complimentary change in ALK (DIC) such that at a constant pH, a 10% change in DIC (ALK) will result in a ~10% change in CO_2[50]. We used the most recent DIC reconstructions from Sosdian et al. (2018) described according to equation 5:

$$DIC_s = \frac{-20.1}{1000} \times Ma + \frac{2027.8}{10^6}$$

(eq. 5)

where Ma is the age in millions of years of the sample and DIC_s is the DIC for the sample with 1sd uncertainty of ± 250 µmol/kg.

CO_2 (aq) was calculated from DIC and pH using equation 6 and a Monte Carlo approach to fully propagate the uncertainty in the input parameters[83]. Atmospheric CO_2 (ppm) was then calculated with equation 7 according to Henry's Law assuming an equilibrium offset at the sample site between the seawater surface and atmosphere (30 ppm, Supplementary Figure 1a, ref. 62):

$$\boldsymbol{CO}_2(\mathrm{aq}) = \frac{\boldsymbol{DIC}}{1 + \frac{\boldsymbol{K}_1^*}{[\boldsymbol{H}^+]} + \frac{\boldsymbol{K}_1^*\boldsymbol{K}_2^*}{[\boldsymbol{H}^+]^2}} \quad \text{(eq. 6)}$$

$$\boldsymbol{CO}_2(\mathrm{ppm}) = \frac{CO_2(aq)}{K_0} - 30\,ppm \quad \text{(eq. 7)}$$

Where DIC is DIC_s for the sample calculated with equation 5, $[H^+]$ is the concentration of protons calculated from $\delta^{11}B$-pH, K_0 is Henry's constant, and K_1 and K_2 are the first and second disassociation constants calculated with ref. 84. All constants were calculated with the 'seacarb' package in R[72]. Because our focus here is on the relative change in ΔF_{CO2} (see below) we have chosen not to correct these carbonate system constants for changing Mg and Ca of seawater. The agreement with ref. 12, where these changes were considered, confirms they are of secondary importance when estimating CO_2 using the boron system in the Late Miocene.

As described above, changes in seawater pH are tightly correlated with changes in CO_2. Thus the radiative forcing of CO_2 (ΔF_{CO2}) can be calculated directly from relative changes pH (ΔpH) according to the method described in ref. 30 using equations 8 and 9 and a Monte Carlo approach (n = 10,000) with the simulations calculating uncertainties for 95% and 68% envelopes:

$$\Delta pH = \left(pK_B{*} - pK_{B_0}\right) - log_{10}(1 + \frac{(\delta^{11}B_{borate} - \delta^{11}B_0)}{\delta^{11}B_{sw} - \alpha_B \times \delta^{11}B_{borate} - (\alpha_B - 1) \times 1000} \times \frac{(\alpha_B - 1) \times \delta^{11}B_{sw} - (\alpha_B - 1) \times 1000}{\delta^{11}B_0 - \delta^{11}B_{sw}})$$

(Eq. 8)

$$\Delta F_{CO2} = \Delta pH \times m$$

(Eq. 9)

Where pK_{B0} is the mean pK_B^* for the record calculated using the mean Mg/Ca derived temperature (25.8 ± 0.5 ˚C), $\delta^{11}B_0$ is the mean $\delta^{11}B$ for the record (18.4 ± 0.1 ‰) and *m* is the ΔpH-to-ΔF_{CO2} conversion coefficient. As the magnitude of the ΔpH-to-ΔF_{CO2} conversion coefficient is dependent on the dominant carbon cycle processes driving the change, we set *m* to range between -16 and -10.6, the maximum range of potential ΔpH-to-ΔF_{CO2} conversion coefficient values in ref. 30, with a flat probability (that is, an equal probability of ΔpH-to-ΔF_{CO2} being any value in the range)[30]. We therefore did not ascribe weight to the assumption that dominant carbon cycling processes remain constant, but rather fully explored the likely range given the available empirical constraints.

As discussed below, offsets in the age models used to construct the ΔSST/ΔGMST stack introduced a degree of smoothing, removing the short-term climate variability shown in our $\delta^{11}B$, CO_2 and ΔF_{CO2} records. To account for such smoothing in the ΔSST/ΔGMST record, we smoothed the ΔF_{CO2} with a smoothing spline function in R (smoothing parameter = 0.53, determined by cross validation)[85] using a Monte Carlo approach (n = 10,000) taking into account the ΔF_{CO2} uncertainty of each data point with the simulation calculating uncertainties for the spline fit with 95% and 68% envelopes.

ΔSST and ΔGMST stack

A global stack of mean annual surface temperature change relative to the modern day (ΔSST) is needed to investigate the climatic temperature response to changes in CO_2. Our ΔSST stack uses only alkenone temperature records to avoid any influence of inter-proxy offsets and biases. We omitted records with a temporal resolution lower than 80 kyrs per sample to give a final selection of records with a relatively high temporal resolution. All selected records are also continuous from 7–5 Ma. Temperature records that fulfil these criteria are: ODP 907[16], ODP 982[16], ODP 883/4[16], ODP 1021[34], IODP U1338[86], ODP 1208[34], ODP 1010[34], ODP 846[16,87,88], IODP U1337[89], ODP 1088[16], ODP 1125[16] and DSDP 594[16] (see

Table 1, Supplementary Figure 1a). We recalculated temperature records with the BAYSPLINE model[43] (prior standard deviation = 5, Figure 1e). This calibration accounts for non-linear effects towards the upper end of the alkenone range that have previously resulted in temperature underestimates[44]. We note that while BAYSPLINE is able to account for some non-linearities, given the temperature sensitivity issues imposed on the alkenone SST proxy at the warm end of the range, it is likely that Late Miocene warmth is underestimated at ODP 722 meaning that the full magnitude of temporal cooling is also likely underestimated. ΔSST was then calculated by subtracting the alkenone SST estimates from the annual modern mean SST at each site (Figure 1e, Supplementary Figure 6)[16].

To create an average of these records, each record was interpolated to a 1kyr resolution. The uncertainty for the stack was estimated using a Monte Carlo approach wherein 1,000 realisations were made for each record within the error bounds to simulate the uncertainty in the calibration and analysis (± error calculated in the BAYSPLINE calibration for each record). The 1,000 simulations were then averaged together to create 1,000 iterations of the SST stack. The mean and 95% and 68% confidence intervals were taken from the distribution of these 1,000 simulations.

ΔSST is used to estimate global mean surface temperature change relative to modern day (ΔGMST). ΔGMST covers a larger range than ΔSST as it includes change in temperature on the continents, which can be both colder and much warmer than those observed in the surface oceans. Here, we used a ΔGMST:ΔSST ratio of 1.5 (1.3–1.9 at 95% confidence) as estimated across multiple model simulations from PlioMIP2 to estimate latest Miocene ΔGMST from ΔSST[59].

To ensure that any single SST record does not have undue influence on the stack (n= 13), each record was removed from the stack sequentially and the stack recalculated (a process known as jack-knifing). We find no substantial difference between the various stacks, strongly suggesting that no one record exerts undue influence on the stack (Supplementary Figure 7). To ensure that the ΔSST/ΔGMST calculated in the stack is reflective of global mean temperature change, we carried out two tests: 1) we compared the mean ΔSST calculated from historic data (from the HadISST dataset located at our sampling locations)[90] with global mean ΔSST for the past 140 years. The mean historic ΔSST (0.55 ± 0.45˚C, 2sd) from the sites used in the ΔSST stack does not substantially deviate from global mean ΔSST calculated from the entire dataset (0.50 ± 0.13 ˚C, 2sd). (2) We then compared a ΔGMST stack for the last 100 kyr generated with sites used in the Late Miocene stack (or nearby sites where Late Quaternary data was absent) to existing ΔGMST multi-proxy reconstructions[51,91]. This allows us to test whether our stack is able to accurately reconstruct ΔSST/ΔGMST when compared to more robust methods for a known timeframe. Our ΔGMST stack for the last 100 kyr is generated using high resolution (<5kyr between samples) alkenone temperature records using the same method and sample sites as described previously. High resolution records for the last 100 kyr were only available for sites ODP 982[16] and ODP 846[92]. For sites where high-resolution records were not available, records were supplemented from nearby cores sites within 1˚: records from U938[93], ODP 1090[94], ODP 1012[95] and ODP 882[94] are used to supplement ODP 1125, ODP 1088, ODP 1010, and ODP 883/884 respectively (Supplementary Figure 8a, Supplementary Table 1). For sites DSDP 594, ODP 907, ODP 1208, IODP U1338 and IODP U1337 there are no acceptable sites nearby these locations, and they are omitted from this comparison (Supplementary Figure 8a). When data for the last 100 kyr are stacked together in a similar way to the Late Miocene stack ($\Delta GMST_{100kyr}$, red on Supplementary Figure 8b), the multi-proxy stack of ref. 51 (blue on Supplementary Figure 8b) and recent comprehensive estimates of LGM temperature change from ref. 91 (black circle on Supplementary Figure 8b) compare well with our $\Delta GMST_{100kyr}$ (black dashed line on Supplementary Figure 9b)[91]. The last 100 kyr stack from ref. 51 is based on 60 temperature proxy records covering all latitudes while the LGM stack from ref. 91 is based on 956 temperature proxy values with an isotope-enabled climate model ensemble. These are the most accurate and comprehensive temperature reconstructions of the last glacial cycle to date. The strong similarity between previously published temperature stacks and our own for the last 100 kyr suggests that our temperature stack for the Late Miocene is representative of global temperature change.

Despite only 60% of Late Miocene core sites being present in the last 100 kyr, omission of these records does not significantly alter the magnitude or temperature changes observed in the Late Miocene stack (Supplementary Figure 8c) and as such, we do not believe their omission hinders the use of the last 100kyr stack in validating the latest Miocene stack. Additionally, both Late Miocene and last 100 kyr temperature stacks have an asymmetric distribution of temperature records, with a bias towards the northern hemisphere. The strong agreement between the last 100 kyr stack with those of ref. 51 and ref. 92, both of which have much greater latitudinal cover than the stacks presented in this paper, suggests that the asymmetric distribution of sites in the Late Miocene stack does not skew the resulting stack to either hemisphere.

Age model uncertainties

The $\delta^{13}C$ data of benthic foraminifera from ODP 926 are consistent with orbitally resolved records in our SST stack, strongly suggesting that broad comparisons between the presented ΔF_{CO2} and $\Delta GMST$ records are valid (Supplementary Figure 5). However, of the temperature records used in generating the ΔSST/ΔGMST stack, eight have age models based on a combination of magnetic reversals and biostratigraphic tie points (ODP 846, ODP 883/4, ODP 1010, ODP 1021, ODP 1125, ODP 1208 and DSDP 594)[16]. This introduces uncertainties of up to ±250 kyr[16], and may lead to substantial temporal offsets in the ΔSST/ΔGMST stack when compared to other orbitally resolved climate records. It is possible that in the construction of our ΔSST/ΔGMST stack these age model errors may lead to an underestimation of the correlation between ΔF_{CO2} and ΔGMST. In an attempt to put a maximum constraint on palaeo-sensitivity (S_p), we forced our ΔF_{CO2} to align with our ΔGMST stack by tying the smoothed ΔF_{CO2} minimum to the ΔGMST minimum using Analyseries[96]. This treatment gives us a theoretical maximum S_p of 3.1 ± 0.4 ˚C (vs. 2.3 ± 0.5˚C, Supplementary Figure 9). We note the possibility of this higher sensitivity, but without further evidence, we utilise the age models as published.

These large uncertainties also make it difficult to determine whether ΔGMST lagging ΔF_{CO2} by ~100 kyr is real or an artefact of relatively poor age control. In many of the ΔSST records, the Late Miocene Cooling minimum is centred between 6.0–5.5 Ma (Figure 1e, Supplementary Figure 6). Indeed, recent improvements in age models have tended to change the timing of the Late Miocene Cooling minimum such that it agrees better with the observed minimum in our CO_2/ΔF_{CO2} record. For instance, recently, ref. 27 updated the age model for the Late Miocene section on ODP 1088, shifting the Late Miocene Cooling minimum back ~250 kyrs, in good agreement with our CO_2 minimum. Discussions regarding the nature of potential offsets between the ΔGMST and ΔF_{CO2} are therefore difficult in the absence of higher quality age models and will not be attempted here.

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