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26 during the Eocene at 500-3000 ppm⁵⁻⁷, and in the absence of tighter constraints
27 carbon-climate interactions over this interval remain uncertain. Here we utilise
28 recent analytical and methodological developments⁸⁻¹¹ to generate a new high-
29 fidelity record of CO₂ concentrations using the boron isotope ($\delta^{11}\text{B}$) composition
30 of well-preserved planktonic foraminifera from the Tanzania Drilling Project,
31 revising previous estimates⁶. Although species-level uncertainties make absolute
32 values difficult to firmly constrain, CO₂ concentrations during the EECO were
33 ~1400 ppm. The relative decline in CO₂ through the Eocene is more robustly
34 constrained at ~50%, with further decline into the Oligocene¹². Provided the
35 latitudinal dependency of sea-surface temperature change for a given climate
36 forcing was similar in the Eocene to the late Quaternary¹³, this CO₂ decline was
37 sufficient to drive the well documented high and low latitude cooling that
38 occurred through the Eocene¹⁴. Once the change in global temperature between
39 the pre-industrial and Eocene due to the action of all known slow feedbacks
40 (apart from those associated with the carbon cycle) are removed²⁻⁴, both the
41 EECO and late Eocene exhibit Equilibrium Climate Sensitivity relative to pre-
42 industrial of 2.1-4.6 °C per CO₂ doubling (66% confidence), which is similar to
43 the canonical range (1.5-4.5 °C¹⁵), indicating that a large fraction of the warmth
44 of the early Eocene greenhouse was driven by elevated CO₂ concentrations, and
45 that climate sensitivity was relatively constant through this period.

46

47 Throughout the last ~540 My climate has oscillated between a globally warm and
48 humid “greenhouse state” and an “icehouse state” with significant continental
49 glaciation¹⁶. The most recent of these transitions occurred between the warmest time
50 interval of the last 65 million years, the Early Eocene Climatic Optimum (EECO) ~51-

51 53 Ma ($\sim 14 \pm 3$ °C warmer than pre-industrial²) and the rapid growth of ice on
52 Antarctica in the earliest icehouse Oligocene (~ 33.6 Ma¹). It has been suggested that
53 variations in the concentration of the greenhouse gas CO₂ were responsible for both
54 the overall warmth of the Eocene and the subsequent cooling¹⁷. Recent studies have
55 documented the importance of CO₂ decline for the final step into the icehouse across
56 the Eocene-Oligocene transition^{12,18}. Despite this, the few available CO₂
57 reconstructions vary markedly between different proxy systems obscuring
58 relationships with the global cooling trend^{1,5,19,20} and therefore preventing a robust test
59 of this hypothesis (Fig. 1). Furthermore, fully coupled climate models require CO₂
60 concentrations ranging from 2x to 16x preindustrial (560-6500 ppm; e.g. Ref. 21) in
61 order to replicate reconstructed early Eocene Sea Surface and Mean Annual Air
62 Temperatures (SSTs and MATs). Clearly in order to use past warm climate states like
63 the early Eocene to evaluate models and our understanding of the climate system, new
64 and more accurate CO₂ reconstructions are needed.

65

66 Boron isotopes ($\delta^{11}\text{B}$) in marine carbonates is a well understood proxy of seawater pH
67 allowing for high-fidelity reconstructions of atmospheric CO₂ in locations where
68 surface seawater is in near equilibrium with the atmosphere regarding CO₂ (e.g. Ref.
69 9,22). Recently the isotopic fractionation factor of boron in seawater was
70 experimentally determined¹⁰, our understanding of the evolution of the ocean
71 carbonate system through time has improved^{23,24}, and it has become apparent through
72 inter-laboratory comparison studies⁸ that a potential bias relating to sample B/Ca ratio
73 may exist in the earlier studies, suggesting published Eocene $\delta^{11}\text{B}$ -CO₂ estimates^{6,7}
74 need revisiting. Here we resolve these issues by measuring the $\delta^{11}\text{B}$ of well-preserved
75 Eocene planktonic foraminifera from the Kilwa group from Tanzania using Multi

76 Collector Inductively Coupled Plasma Mass Spectrometry (MC-ICPMS)⁹, utilising a
77 greater array of species to more clearly resolve the potential for species-specific and
78 depth habitat related biases in $\delta^{11}\text{B}$.
79
80 The approach we follow to determine atmospheric CO_2 from foraminiferal $\delta^{11}\text{B}$
81 ($\delta^{11}\text{B}_c$) is detailed in full in the Methods section. We analysed foraminifera from five
82 discrete timeslices between 36.9 and 53.2 Ma recovered by the Tanzania Drilling
83 Project (Extended Data Fig. 1), and the oxygen isotopic composition ($\delta^{18}\text{O}$) of up to
84 17 different foraminifera species occupying a range of depth habitats was quantified to
85 derive the calcification temperature and hence the relative habitat depth of the taxa²⁵.
86 In each case we find a consistent decrease of $\delta^{11}\text{B}_c$ with increasing depth consistent
87 with modern ocean $\delta^{11}\text{B}_{\text{borate}}$ profiles (Fig. 2) and a clustering of the warmest species
88 with a similar $\delta^{11}\text{B}_c$. By applying species-specific $\delta^{11}\text{B}$ -pH calibrations, $\delta^{11}\text{B}_c$
89 measurements can be converted to pH reconstructions with depth. However, we lack
90 such calibrations for these extinct taxa. The $\delta^{11}\text{B}$ vital effects which are a result of
91 foraminiferal physiology (Methods) appear reduced in Eocene foraminifera compared
92 to modern taxa for both shallow symbiont-bearing and deeper non-symbiotic
93 planktonic foraminifera. We therefore calculate pH from $\delta^{11}\text{B}_c$ using two different
94 approaches to bracket the likely magnitude of $\delta^{11}\text{B}$ -pH vital effects: (i) we assume
95 $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$ (hereafter the “borate” calibration), and (ii) we assume the shallower
96 foraminifera in each time-slice has the $\delta^{11}\text{B}$ -pH calibration of the modern mixed layer
97 dwelling foraminifera *Trilobatus sacculifer*²².
98

99 The boron isotopic composition of seawater ($\delta^{11}\text{B}_{\text{sw}}$) is also required to calculate pH,
100 constrained here by two methods both based on the observation that the reconstructed
101 pH gradient from shallow to deep for a given $\delta^{11}\text{B}_{\text{borate}}$ gradient in the water column is
102 a function of $\delta^{11}\text{B}_{\text{sw}}$ because of the non-linearity of the $\delta^{11}\text{B}$ -pH proxy (Extended Data
103 Fig. 2, Methods). If we assume $\delta^{11}\text{B}_{\text{c}} = \delta^{11}\text{B}_{\text{borate}}$ we calculate $\delta^{11}\text{B}_{\text{sw}}$ to be in the range
104 38.2 to 38.7 ‰. This is slightly increased to 38.6-38.9 ‰ if we use the $\delta^{11}\text{B}$ -pH
105 calibration of *T. sacculifer* (Methods).

106

107 Given the strong positive relationship between $\delta^{11}\text{B}$ and pH, it is evident from our
108 $\delta^{11}\text{B}_{\text{c}}$ data alone that surface water pH increased through the Eocene (Figure 3a).
109 However, having determined both $\delta^{11}\text{B}_{\text{sw}}$ and calcification temperature, we can
110 generate quantitative estimates of upper ocean pH, and here we propagated a
111 conservative estimate of the relevant uncertainties using a Monte Carlo approach
112 (Methods). Based on these upper water column reconstructions and the borate (or *T.*
113 *sacculifer*) calibration we estimate that surface water pH increased from 7.66 ± 0.06
114 (7.59 ± 0.06) at 53.2 Ma, to 7.80 ± 0.07 (7.76 ± 0.06) by 36.9 Ma. By extending our
115 knowledge of species depth habitat and $\delta^{11}\text{B}_{\text{sw}}$ to the $\delta^{11}\text{B}_{\text{c}}$ data set of Ref. 12
116 (Methods), it is apparent that surface water pH continued to increase in the early
117 Oligocene reaching 7.88 ± 0.05 (7.84 ± 0.05) by ~33 Ma. Notably, our Eocene surface
118 water pH estimates are lower than any others reported in the Cenozoic, including
119 during the middle Miocene and Pliocene warmth (minimum pH: 7.83 Ref. 22 and 7.99
120 Ref. 26 respectively).

121

122 Ocean pH, like seawater CO_2 ($[\text{CO}_2]_{\text{aq}}$), is largely determined by the ratio of total
123 alkalinity to total dissolved inorganic carbon. Hence, to a first order, our pH estimates

124 also indicate that $[\text{CO}_2]_{\text{aq}}$ declined through the Eocene. Given the oligotrophic setting
125 of our site, this further implies that atmospheric CO_2 declined through the study
126 interval. However, quantitative constraints on early Cenozoic CO_2 evolution require a
127 second carbonate system parameter. Here we assume that the surface seawater
128 saturation state for calcite (Ω_{calc}) at our site remained between 5.5 and 7.5, a
129 conservative range that encompasses geochemical model estimates²⁷ (Methods;
130 Extended Data Fig. 3). Uncertainty was again fully propagated (details in Methods)
131 and as expected given the raw $\delta^{11}\text{B}_c$ values and calculated pH, we find that CO_2
132 decreased (Fig. 3b) for the borate (or *T. sacculifer* in Extended Data Fig. 4) calibration
133 from 1400 ± 470 (1900 ± 600) ppm during the EECO to ≤ 770 (920) ppm from 40.3
134 Ma onwards, reaching a minimum of 550 ± 190 (670 ± 210) ppm in the early
135 Oligocene. Our new data confirm that CO_2 was most likely >1000 ppm (at 95% CI)
136 during the EECO consistent with the recently revised estimates using sodium
137 carbonate mineral nahcolite¹¹. Although our younger timeslices are in good agreement
138 with alkenone based CO_2 reconstructions (Extended Data Fig. 4), our early Eocene
139 CO_2 reconstructions are clearly unprecedented in the Cenozoic. At 5-7x pre-industrial,
140 our reconstructed EECO CO_2 can provide tighter constraints on models than those that
141 have been previously available. Critically, our CO_2 reconstructions are not consistent
142 with the concentrations required by a number of coupled ocean-atmosphere climate
143 models in order to achieve a good match with Eocene temperature data²¹.

144

145 Using our new CO_2 record, the possible role of CO_2 in driving Eocene cooling can be
146 quantitatively examined. The relative change in climate forcing attributable to CO_2
147 change (ΔF_{CO_2}) through the Eocene can be estimated²⁸ as:

148

149 $\Delta F_{\text{CO}_2} = 5.32 * \ln (C_t/C_{\text{EECO}}) + 0.39 * (\ln (C_t/C_{\text{EECO}}))^2$ (1)

150

151 where C_{EECO} is the CO_2 at the EECO, and C_t refers to the CO_2 reconstruction at a
152 particular time in the Eocene. The CO_2 forcing based on our reconstructions for the
153 Eocene is well correlated with the contemporaneous proxy derived sea surface
154 cooling, albeit with considerably larger cooling at high latitudes (Fig. 4a, Extended
155 Data Fig. 5a, $R^2 = 0.8$ at low latitudes and 0.9 at high latitudes). The apparent
156 latitudinal dependency of SST change for a given change in radiative forcing has
157 recently been determined for the last 520 kyr¹³, using CO_2 , ice-sheet albedo (slow-
158 feedbacks), dust and mean annual insolation as forcing terms²⁹. Assuming that this
159 latitudinal dependency for a given forcing (Fig. 4b and Methods)¹³ is also appropriate
160 for the Eocene, we can calculate the expected SST change as a result of the climate
161 forcing, ΔF_{CO_2} (Eq. (1)), for each target timeslice in our study, for high (60 °North and
162 South; blue lines on Fig 4b) and low latitudes (30 °N to 30 °S, red box on Fig. 4b; see
163 Methods) and compare these to a recent SST compilation¹⁴. The calculated SST
164 change from our CO_2 reconstructions is within error of the observed, proxy-derived
165 SST evolution of both high and low latitudes through the Eocene (Fig. 4c, Extended
166 Data Fig. 5c) regardless of which $\delta^{11}\text{B}$ -pH calibration is used. This supports CO_2 as
167 the principal driver of long-term Eocene cooling, although feedback processes
168 involved in causing the observed polar amplification in Fig. 4a and Extended Data
169 Fig. 5a must also play a vital role in determining its pattern.

170

171 Our CO_2 reconstructions also allow us to examine the sensitivity of the Earth system
172 (i.e. temperature) to changes in radiative forcing. This is often discussed in terms of
173 Equilibrium Climate Sensitivity (ECS): the global mean surface air temperature

174 change for a doubling of CO₂ on the timescales of about a century after all the “fast”
175 feedbacks have had time to operate but before longer-timescale feedbacks act²⁹, and
176 can be calculated for the Eocene using:

177

$$178 \text{ ECS (}^\circ\text{C)} = \Delta T / \Delta F_{\text{CO}_2} * 3.87 \text{ (2)}$$

179

180 where ΔT is the global mean air temperature change compared to the pre-industrial,
181 corrected for slow-feedbacks (Fig. 4d, Extended Data Fig. 5d); ΔF_{CO_2} is calculated as
182 in Eq. (1), replacing C_{EECO} with pre-industrial CO₂ ($C_0 = 278$ ppm) as we are
183 interested in forcing change relative to the pre-industrial (Fig. 4e, Extended Data Fig.
184 5e); and 3.87 is the climate forcing of a CO₂ doubling in W*m⁻² calculated using Eq.
185 (1). The global mean annual temperature for the EECO is thought to be $\sim 14 \pm 3$ °C
186 warmer than the pre-industrial, and ~ 5 °C warmer than the late Eocene (35 Ma; Ref.
187 2,29). Recent climate modelling studies have ascribed between 4 to 6 °C of Eocene
188 warmth to changes in boundary conditions (e.g. paleotopography and continental
189 configuration) and the action of the slow climate feedbacks, principally the lack of a
190 continental ice sheet on Antarctica and vegetation change²⁻⁴. If CO₂ was the principal
191 driving mechanism for the relative warmth of the EECO and late Eocene, we would
192 expect an Eocene-ECS within the range reported by the Intergovernmental Panel on
193 Climate Change (IPCC¹⁵) based on climate modelling and observations of the (paleo-)
194 climate system. However, if other climate feedbacks were important we have not
195 accounted for in our correction of the “slow” feedbacks²⁻⁴, or if the forcing is not due
196 to CO₂ alone, then the calculated ECS would be different from that reported by the
197 IPCC. Full details of how the uncertainties in these various parameters are propagated
198 can be found in the Methods, but from Fig. 4f and Extended Data Fig. 5f it is clear that

199 the average ECS we calculate for the Eocene lies for the borate (*T. sacculifer*)
200 approach in the range 2.6 to 4.6 °C (2.1 to 3.6 °C) per CO₂ doubling (66% confidence)
201 with maximum probability for the EECO of 3.8 °C (3.3 °C). This range (2.1 to 4.6 °C
202 per CO₂ doubling) is similar to the IPCC range (1.5 to 4.5 °C at 66% confidence¹⁵).
203 Therefore, the 65% of early and late Eocene warmth attributed to fast feedbacks^{2-4,29} is
204 consistent with it being driven by higher atmospheric CO₂. This supports the likely
205 importance of atmospheric CO₂ variability in determining both the warmth of the
206 early Eocene and the cooling that ultimately led to the establishment of the Late
207 Cenozoic icehouse.

208

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289

290 **Supplementary Information and Extended Data Figures** are linked to the online
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292

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302

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304 calculations, and drafted the manuscript. E.H.J. and K.M.E. prepared foraminifer
305 samples and conducted the stable isotope analysis. P.N.P. led the fieldwork, taxonomy
306 and prepared foraminifer samples. A.R. provided cGENIE model results. P.N.P. and
307 G.L.F. designed the study and all authors discussed the results and contributed to the
308 final text.

309

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314

315 **Figure 1. Currently available Eocene atmospheric CO₂ records and benthic**
316 **foraminiferal $\delta^{18}\text{O}$ values. a.** CO₂ reconstructions with the 2 s.d. uncertainties. **b.**
317 Benthic foraminiferal $\delta^{18}\text{O}$ ¹. Line shows five point moving average, with data age
318 modified to account for the revised timing of the MECO³⁰. Lower benthic $\delta^{18}\text{O}$ values

319 denote warmer bottom water temperatures (or less ice volume, which is assumed to be
320 minimal in the Eocene). Error bars on $\delta^{18}\text{O}$ are smaller than the symbols (Methods).

321

322 **Figure 2. Eocene planktonic foraminiferal multi-species stable isotope arrays. a.**

323 to **e.** Foraminiferal $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ and **f. to j.** Foraminiferal $\delta^{11}\text{B}$ values vs. $\delta^{18}\text{O}$

324 inferred water depths for each timeslice. Panels **c.** and **h.** show the anomalous

325 behaviour of *Globigerinatheka* spp. (in parentheses) in both $\delta^{13}\text{C}$ and $\delta^{11}\text{B}$ (Methods).

326 The arrow in panel **j.** shows the correction of *T. cerroazulensis* $\delta^{11}\text{B}$ to shallower

327 water values (Methods). Shaded areas in **f. to j.** surround the species used in Fig. 3a.

328 Errors on $\delta^{13}\text{C}$ and $\delta^{18}\text{O}$ are smaller than symbols. Error bars on $\delta^{11}\text{B}$ are 2 s.d. based

329 on the long-term precision (Methods).

330

331 **Figure 3. New atmospheric CO₂ reconstructions from shallow planktonic**

332 **foraminiferal $\delta^{11}\text{B}$.** **a.** $\delta^{11}\text{B}$ of shallower foraminifera (symbols as in shaded

333 rectangles of Fig. 2). Blue squares and orange circles represent the warmest species

334 used in panel b. Ref. 12 data are offset (Supplementary Table 2, Methods). **b.**

335 Atmospheric CO₂ assuming $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$ using the warmest species of each

336 timeslice. **c.** Benthic foraminiferal $\delta^{18}\text{O}^1$ (as in Fig. 1b). Age error bars (panel b) are

337 based on the ages of the nearest datums, $\delta^{11}\text{B}$ errors as in Fig. 2, and CO₂ uncertainty

338 is based on 95% confidence of Monte Carlo error propagation (Methods).

339

340 **Figure 4. CO₂ as a driver of latitudinal cooling in the Eocene, and Equilibrium**

341 **Climate sensitivity (ECS) analyses of the EECO and late Eocene timeslices. a.**

342 Evolving relationship between SST¹⁴ for high and low latitudes and the CO₂ forcing of

343 each of our timeslices relative to the EECO, with linear regression fits and coefficients

344 of determination (R^2). **b.** Apparent latitudinal SST sensitivity for the last 520 kyr¹³.
345 Dashed line is second order polynomial through the SST sensitivity data (grey
346 crosses) of Ref. 13. Red rectangle surrounds the SST sensitivity estimates averaged as
347 a low-latitude mean, and blue line for the high latitude mean. **c.** Reconstructed (lines)
348 and estimated (symbols) SST relative to 53.2 Ma. Symbols are estimated using the
349 respective CO₂ reconstructions and the average low and high latitude SST sensitivities
350 of b. Bold lines show the reconstructed long-term mean TEX₈₆ SST estimates at high
351 (blue >55°) and low (red <30°) latitudes¹⁴ relative to ~53 Ma SST. Error bars represent
352 full propagation of errors at 95% confidence. **d.** Mean Air Temperature range for early
353 and late Eocene corrected for changes due to slow feedbacks^{2,4,21,29}. **e.** Forcing
354 compared to pre-industrial calculated using our CO₂ reconstructions for the 53.2 Ma
355 (early Eocene) and 36.9 Ma (late Eocene) timeslice. **f.** Probability density functions of
356 ECS for the early and late Eocene compared to IPCC estimates. Shaded bands around
357 lines throughout show 95% confidence intervals.

358

359 **METHODS**

360 **Site information.** To obtain optimal records of oxygen, carbon, and boron isotopes,
361 we used well-preserved ‘glassy’ carbonate microfossils from the Paleogene of the
362 Kilwa Group material drilled by the Tanzanian Drilling Project (TDP)^{31,32}. The core
363 sites were located on the outer shelf-upper continental slope approximately 50-70 km
364 offshore³³, at a paleolatitude of 19 °S, and bathyal paleowater depths of >300 m³¹.
365 Seismic data, sedimentary facies, nannofossil and planktic foraminiferal assemblages
366 as well as low concentration of dinoflagellates and lack of marine biomarkers
367 indicative of high export productivity confirm that these sediments were deposited
368 under open-ocean conditions with waters derived from the Indian Ocean subtropical

369 gyre³⁴⁻³⁶, supporting the assumption that [CO₂]_{aq} offshore Tanzania was in (near)
370 equilibrium with the atmosphere with respect to CO₂. This is supported by cGENIE
371 estimates with Eocene boundary conditions for offshore Tanzania (Extended Data Fig.
372 6)³⁷, nevertheless, to include the uncertainty of potential disequilibrium with the
373 atmosphere we added ± 40 ppm CO₂ uncertainty into our estimates of atmospheric
374 CO₂ using δ¹¹B.

375

376 **Sample preparation.** Approximately 3-4 mg of mono-specific foraminiferal material
377 of a narrow size fraction (e.g. ~300 individuals from the 212-250 µm sieve size
378 fraction) were separated from ~10 cm of core material (24 cm for the 53.2 Ma
379 timeslice) for tandem analyses of all isotopes (δ¹⁸O, δ¹³C, and δ¹¹B) and trace element
380 composition. Identification of planktonic foraminifera followed Ref. 35. Each sample
381 was gently crushed and divided into two parts: one for δ¹¹B and elemental analyses
382 and another for δ¹⁸O and δ¹³C measurements (~100 µg, ~3% per weight). For the 53.2
383 Ma and 40.3 Ma timeslices, *Cibicidoides* sp. were also picked for analyses (~1 mg).
384 Splits for elemental and δ¹¹B analyses were cleaned following established methods³⁸⁻
385 ⁴⁰. Trace element ratios were determined as in Ref. 9 and in all cases Al/Ca ratios were
386 <120 µmol/mol (typically <100 µmol/mol) evidencing efficient foraminiferal
387 cleaning^{9,40}.

388

389 **δ¹⁸O and δ¹³C analyses, temperature reconstructions and relative habitat depth**
390 **assignments.** For all δ¹⁸O and δ¹³C analyses, we followed the analytical approach
391 described elsewhere⁴¹, and values are reported relative to the Vienna Pee Dee
392 Belemnite (VPDB) standard, with 1 s.d. standard external precision of ±0.06 ‰ and
393 ±0.07 ‰ respectively. All temperature reconstructions are based on the equation of

394 Ref. 42, corrected for changes in global ice volume of -0.8‰ ⁴³ and paleolatitude of
395 $+0.83\text{‰}$ ⁴⁴ (following Ref. 45). Any further secular trends in seawater $\delta^{18}\text{O}$, regional
396 variations in the oxygen isotope ratio of seawater, or differing carbonate ion
397 concentrations are not known with sufficient confidence, therefore no additional
398 corrections were applied. The lowest $\delta^{18}\text{O}$ value in each multi-species depth profile
399 was used to calculate the maximum SST for each timeslice with a conservative
400 uncertainty of $\pm 2\text{ °C}$ that encompasses the differences among variable carbonate $\delta^{18}\text{O}$
401 equations (Ref. 45), and includes potential uncertainties in $\delta^{18}\text{O}_{\text{sw}}$ (Ref. 43,46).

402

403 Identification of foraminiferal depth habitats is based on $\delta^{18}\text{O}$ following Ref. 25,35.
404 This method could carry large uncertainties in the absolute depth reconstructions, but
405 the relative depth habitats of surface and deep taxa are clearly distinguishable from the
406 $\delta^{13}\text{C}$ vs. $\delta^{18}\text{O}$ cross-plots (e.g. Ref. 25,47,48-50,45,51-54), and supported by
407 comparison of $\delta^{11}\text{B}$ vs. $\delta^{18}\text{O}$ profiles (Fig. 2). Further support for our approach is
408 given by the depth assignment of benthic foraminifera *Cibicidoides* based on $\delta^{18}\text{O}$
409 values at 53.2 and 40.3 Ma, which at 300-320 m agrees well with the site's paleowater
410 depth³¹. Any uncertainty in the absolute depth assignment of foraminifera as a result
411 of the 4 °C range ($\pm 2\text{ °C}$) contributes $< 0.1\%$ uncertainty to the carbonate system
412 calculations and thus is not included, although the temperature uncertainty is fully
413 propagated into our carbonate system estimates (see below).

414

415 **Boron isotope proxy and analyses.** Boron isotopes in planktonic foraminifera
416 residing close to the seawater surface have been used extensively to reconstruct past
417 ocean pH and thus CO_2 concentrations^{9,22,55-57}. External reproducibility of $\delta^{11}\text{B}_c$
418 analyses is calculated using the approach of Ref. 40 and the relationship of Ref. 41.

419

420 To reconstruct seawater pH and thus CO₂ concentrations on million year timescales
421 using foraminiferal δ¹¹B values, we must take some necessary steps that include:
422 correcting for any vital effects in extinct species; constraining the δ¹¹B_{sw}, which, given
423 the 10-20 Ma residence time for boron in the ocean is likely different from modern
424 (39.61 ‰)^{58,59}; and estimating a second carbonate system parameter to infer aqueous
425 CO₂ concentration ([CO₂]_{aq}) from pH. Other necessary steps include reconstructing
426 the calcification temperature and relative habitat depth of foraminifera; knowing the
427 CO₂ disequilibrium between surface waters and atmosphere at the study site; and that
428 primary δ¹¹B values are not altered by diagenesis (mitigated here by using ‘glassy’
429 foraminifera).

430

431 **Vital effects corrections associated with the boron isotope proxy.** Offsets between
432 the boron isotopic composition of ambient seawater borate ion (δ¹¹B_{borate}) and that of
433 foraminiferal calcite (δ¹¹B_c) are known as vital effects. Such vital effects develop in
434 response to respiration, calcification and photosynthesis generating a pH gradient
435 within the micro-environment (the diffusive boundary layer, DBL) around
436 foraminifera^{60,61}. For extant species these effects can be accounted for using species-
437 specific δ¹¹B-pH calibrations (e.g. Ref. 9,62-64). However, all Eocene foraminifera
438 species are extinct and thus vital effects can only be assessed indirectly. To
439 supplement our existing understanding of modern vital effects^{22,26,62,64,65} we generated
440 core top (Holocene) planktonic foraminiferal δ¹¹B, δ¹⁸O, and δ¹³C values (Extended
441 Data Fig. 7c and Supplementary Table 1) from offshore Tanzania (Glow 15, Ref. 66).
442 Modern foraminifera from offshore Tanzania show a significantly less coherent
443 relationship with δ¹⁸O than our Eocene samples (Extended Data Fig. 7 and Fig. 2).

444 This increased spread is at least partly due to the known large vital effects of modern
445 foraminifera, where offsets of 0 to 4 ‰ are required for the $\delta^{11}\text{B}$ of our modern
446 species to match the $\delta^{11}\text{B}$ of seawater borate (Extended Data Fig. 7c).
447
448 If we apply published modern symbiont-bearing planktonic foraminifera $\delta^{11}\text{B}$ -pH
449 calibrations^{22,62,64} for the shallower Eocene species, and symbiont-barren $\delta^{11}\text{B}$ -pH
450 calibrations of modern planktonic foraminifera^{26,65} for the Eocene deep dwellers, an
451 unrealistic picture emerges (Extended Data Fig. 8). Specifically, there is increased
452 scatter between the shallowest and deepest planktonic foraminifera and inversion in
453 pH gradients with depth. Additionally, there is clear disagreement between
454 *Cibicidoides*, that we know exhibits minimal vital effect for $\delta^{18}\text{O}$, $\delta^{11}\text{B}$ and $\delta^{13}\text{C}$ ^{40,67},
455 and deeper planktonic foraminifera of similar depth (Extended Data Fig. 8), which are
456 otherwise overlapping in $\delta^{11}\text{B}_c$ (for the 53.2 Ma timeslice, *S. rosnaesensis* $\delta^{11}\text{B} =$
457 12.96 ± 0.38 ‰ is in agreement with *Cibicidoides* which average 13.15 ± 0.53 ‰, and
458 for the 40.3 Ma timeslice, average *Subbotina* $\delta^{11}\text{B} = 12.63 \pm 0.38$ ‰ is within
459 uncertainty of the *Cibicidoides* $\delta^{11}\text{B} = 12.71 \pm 0.30$ ‰). This suggests that modern
460 vital effects are likely not applicable to Eocene planktonic foraminifera. Given that
461 modern vital effects appear to be unsuitable for the Eocene as explained above, we
462 argue by extension that the reduced scatter in the Eocene, and notably the agreement
463 in $\delta^{11}\text{B}$ among the mixed layer species targeted in the Eocene (which is in stark
464 contrast with the modern, Extended Data Fig. 7c), suggests a more minor role for vital
465 effects in determining $\delta^{11}\text{B}_c$, and thereby that the targeted Eocene foraminifera must
466 more closely reflect the $\delta^{11}\text{B}$ of borate ion. Furthermore, different size fractions of the
467 Eocene foraminifera belonging to the genus *Acarinina* from this study (Supplementary
468 Table 1), and for Early Eocene/Paleocene *Morozovella velascoensis* and *A.*

469 *soldadoensis* in Ref. 68 have $\delta^{11}\text{B}_c$ compositions within analytical uncertainty. This
470 contrasts with the modern symbiont-bearing planktonic foraminifera *T. sacculifer* and
471 *G. ruber* that exhibit large changes in $\delta^{11}\text{B}$ with size (1 ‰ to 2.3 ‰ Ref. 64,69), again
472 supporting our assertion that vital effects are reduced in the Eocene foraminifera.

473

474 Although we do not yet have a full mechanistic understanding of the causes of $\delta^{11}\text{B}$ -
475 vital effects in planktonic foraminifera a reduction in vital effects for the Eocene
476 foraminifera would most likely result from a reduction in the relative magnitude of
477 one or more of the following physiological processes: calcification, photosynthesis,
478 respiration. For instance, for smaller modern foraminifera the magnitude of the
479 influence of symbiont photosynthesis on the pH of the DBL is reduced, with smaller
480 size fraction foraminifera recording $\delta^{11}\text{B}_c$ closer to that of seawater borate^{64,69}. Here,
481 the size of our shallower Eocene foraminifera is mostly within 212-250 μm , which is
482 reduced compared to the modern foraminifera analysed here (300-355 μm) and
483 elsewhere (e.g. $\geq 425 \mu\text{m}$ Ref. 70). Also, vital effect offsets are pH dependent (e.g.
484 Extended Data Fig. 9) for symbiont bearing foraminifera^{62-64,71}. Eocene symbiotic
485 planktonic foraminiferal composition is 15.5-14.5 ‰, and for this range the *Trilobatus*
486 *sacculifer* (300-355 μm , Ref. 22) calibration has a minimal effect on pH estimates, as
487 it requires only 0.1-0.3 ‰ correction on $\delta^{11}\text{B}_c$ for deriving $\delta^{11}\text{B}_{\text{borate}}$.

488

489 Importantly, whilst the modern symbiont bearing foraminifera calibrated to date have
490 $\delta^{11}\text{B}$ composition offset from the seawater borate curve, only $\delta^{11}\text{B}$ -pH calibrations for
491 three dinoflagellate hosting species have so far been published^{9,62-64}. Our new depth
492 profile for the modern Indian Ocean shown in Extended Data Fig. 7c shows that some
493 extant species (*Globigerinella siphonifera* and *Globorotalia menardii*) have minimal

494 $\delta^{11}\text{B}_c$ offsets from seawater borate $\delta^{11}\text{B}$.

495

496 Additional support for minimal vital effects in Eocene foraminifera is also given by
497 the relative agreement with independently calculated Eocene $\text{CO}_2^{5,11,19}$ when the
498 borate $\delta^{11}\text{B}$ -pH calibration is used (Extended Data Fig. 4). Nonetheless, we also apply
499 the modern *T. sacculifer* calibration as this provides an upper limit on the likely extent
500 of $\delta^{11}\text{B}$ vital effects in Eocene foraminifera. Notably, our calculated pH and CO_2
501 estimates for both approaches are largely within uncertainty (Extended Data Fig. 4),
502 highlighting that such assumptions have little influence on the conclusions drawn (Fig.
503 4 and Extended Data Fig. 5).

504

505 **Constraints on $\delta^{11}\text{B}_{\text{sw}}$.** Geochemical box models⁵⁸ and existing records of benthic
506 foraminiferal $\delta^{11}\text{B}_c$, coupled with assumptions regarding deep water pH evolution⁷²,
507 indicate $\delta^{11}\text{B}_{\text{sw}}$ is unlikely to have changed from its modern value by more than 2.5 ‰
508 since the EECO (with a likely maximum rate of change of 0.1 ‰/Ma)⁵⁸. Here, we use
509 new, empirically constrained, upper and lower bounds on Eocene $\delta^{11}\text{B}_{\text{sw}}$ from several
510 independent approaches. The underlying methodology for quantifying a maximum
511 and minimum $\delta^{11}\text{B}_{\text{sw}}$ is based on the observation that the $\delta^{11}\text{B}$ vs. pH is non-linear
512 (Extended Data Fig. 2), and its curvature is affected by $\delta^{11}\text{B}_{\text{sw}}$; when $\delta^{11}\text{B}_{\text{sw}}$ is higher,
513 for any given $\delta^{11}\text{B}_c$ gradient the reconstructed pH change is larger than when $\delta^{11}\text{B}_{\text{sw}}$ is
514 lower. Furthermore, the curvature in the $\delta^{11}\text{B}$ -pH relationship results in there being a
515 pH below which the $\delta^{11}\text{B}$ system becomes unresponsive, the lowest foraminiferal
516 $\delta^{11}\text{B}_c$ recorded in this study therefore provides the first constraint on the $\delta^{11}\text{B}_{\text{sw}}$, which
517 then places an additional upper bound on $\delta^{11}\text{B}_{\text{sw}}$. The lowest $\delta^{11}\text{B}_c$ values we

518 recovered are for *Subbotina crociapertura* of 11.54 ± 0.29 ‰ at 45.6 Ma, and *S.*
519 *eocaena* of 11.97 ± 0.22 ‰ at 40.3 Ma. If $\delta^{11}\text{B}_{\text{sw}} \geq 39.5$ ‰, these samples do not give
520 a valid mathematical solution for the $\delta^{11}\text{B}$ vs. pH relationship, therefore Eocene $\delta^{11}\text{B}_{\text{sw}}$
521 must be lower than the modern (39.6 ‰⁵⁹).

522

523 The lower bound for $\delta^{11}\text{B}_{\text{sw}}$ is constrained using the modern pH gradient (corrected for
524 anthropogenic DIC) within the top ~300 m of the water column in the tropics and
525 assuming that this represents the likely minimum pH gradient in Eocene. This
526 assumption is justified by 1) a positive linear relationship between $\delta^{13}\text{C}$ and pH in
527 modern²² and Eocene seawater (Extended Data Fig. 10 based on cGENIE output⁷³),
528 and 2) larger $\delta^{13}\text{C}$ gradients between surface and deep dwelling Eocene planktonic
529 foraminifera than in the modern because of enhanced metabolic rates due to warmer
530 ocean temperatures²⁵. As a result, Eocene pH gradients were most likely larger than
531 modern equivalent and therefore our approach is conservative. Modern tropics, in non-
532 upwelling regions, have an average pH gradient of the first 300 m equal to 0.17 (Ref.
533 74) with an anthropogenic CO₂ correction resulting in an uncertainty of 0.003 pH
534 units (GLODAP v1.1). The 53.2 Ma timeslice is used for this treatment because it has
535 full upper water column depth coverage and the smallest $\delta^{11}\text{B}_c$ gradient (hence it will
536 provide the smallest pH difference and thus the minimum $\delta^{11}\text{B}_{\text{sw}}$ value). A Monte
537 Carlo propagation of uncertainties in pH, temperature, and composition of seawater
538 results in the pH difference (assuming $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$) between the shallowest and
539 the deepest planktonic dwellers (ΔpH) at 53.2 Ma falling below 0.17 (the modern
540 equivalent) when $\delta^{11}\text{B}_{\text{sw}} < 38.2$ ‰ (with 86% of simulations resulting in ≥ 0.17 ΔpH).
541 Therefore the minimum $\delta^{11}\text{B}_{\text{sw}}$ is calculated at 38.2 ‰ for the whole Eocene. This
542 exercise was repeated using the alternative methodology for dealing with $\delta^{11}\text{B}$ -vital

543 effects (assuming $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$ for the deeper dwelling non symbiotic
544 foraminifera, which have $\delta^{11}\text{B}_c$ within error of co-occurring *Cibicicoides*, and
545 assuming that the shallowest foraminifera has the $\delta^{11}\text{B}$ -pH calibration of modern *T.*
546 *sacculifer*). The minimum constraint from 86% of potential Monte Carlo solutions is
547 then 38.6 ‰.

548

549 To further constrain the maximum Eocene $\delta^{11}\text{B}_{\text{sw}}$ we used restrictions offered by
550 estimates of Apparent Oxygen Utilization (AOU), adapted from previous approaches⁷.
551 AOU describes the amount of dissolved oxygen consumed by the remineralisation of
552 sinking organic carbon between a selected depth and saturated surface waters. The
553 surface seawater oxygen concentration ($[\text{O}_{2\text{-sw}}]_{\text{sat}}$) depends on SST as a result of
554 Henry's law. Here, $\delta^{18}\text{O}$ derived SSTs are ~30-33 °C (Supplementary Table 1),
555 therefore corresponding $[\text{O}_{2\text{-sw}}]_{\text{sat}}$ are ~191-182 $\mu\text{mol}/\text{kg}$ ⁷⁵, with potential mean
556 oversaturation of ~13 $\mu\text{mol}/\text{kg}$ (low latitude maximum: 0.2-0.4 ml/L^{76,77}). To calculate
557 the upper estimate of $\delta^{11}\text{B}_{\text{sw}}$, the minimum SST of the Eocene timeslices is selected,
558 and thus maximum $[\text{O}_{2\text{-sw}}]_{\text{sat}}$ is calculated at 204 $\mu\text{mol}/\text{kg}$. Additionally, we assume
559 Eocene planktonic foraminifera have an oxygen minimum tolerance threshold of 44.7
560 $\mu\text{mol}/\text{kg}$, analogous to the low oxygen accustomed *Hastigerinella digitata*⁷⁸ in the
561 modern Monterey Bay (California). Therefore, we assume Eocene deeper dwelling
562 planktonic foraminifera were bathed in waters with oxygen concentrations equal to or
563 higher than the minimum oxygen level of *H. digitata* ($\text{AOU} = 204 - 44.7 = 159.3$
564 $\mu\text{mol}/\text{kg}$). To calculate AOU for each studied timeslice, we modified $\delta^{11}\text{B}_{\text{sw}}$ (similarly
565 following the Monte Carlo approach explained above) from the minimum constraint
566 of 38.2 ‰ to modern seawater (39.6 ‰⁵⁹). We then first calculate the difference
567 between the calculated (initially assuming that $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$) dissolved inorganic

568 carbon (DIC) at the depth occupied by our deepest dwelling foraminifera (average),
569 and compare this to the DIC of the shallowest ~50 m averaged dwelling planktonic
570 foraminifera. The difference in DIC is then transformed into AOU using Redfield
571 ratios (106:138, CO₂:O₂). The AOU is $\leq 159.3 \mu\text{mol/kg}$ when $\delta^{11}\text{B}_{\text{sw}}$ is at maximum
572 38.5 ‰ for the 45.6 Ma timeslice and 38.7 ‰ for the 40.3 Ma timeslice, resulting in
573 86% of potential solutions with our Monte Carlo approach satisfying the AOU
574 constrain. The same approach was used applying the *T. sacculifer* calibration for the
575 shallowest planktonic foraminifera, resulting in a maximum $\delta^{11}\text{B}_{\text{sw}}$ of 38.8 ‰ and
576 38.9 ‰ for the 45.6 and 40.3 Ma timeslice respectively. To be conservative, we use
577 the maximum of the two estimates for each calibration method. In summary, although
578 we are calculating the lower $\delta^{11}\text{B}_{\text{sw}}$ bound at 53.2 Ma and the upper at 40.3 and 45.6
579 Ma, we apply this range across the whole of the study interval (53.2-36.9 Ma).

580

581 **Second carbonate parameter.** After calculating seawater pH using $\delta^{11}\text{B}_{\text{sw}}$ and $\delta^{11}\text{B}_{\text{c}}$,
582 an additional carbonate parameter is required to calculate CO₂ concentrations at any
583 given seawater salinity and temperature. Here the second parameter we use is the
584 saturation state of calcium carbonate (surface $\Omega_{\text{calc}} = [\text{CO}_3^{2-}]/[\text{CO}_3^{2-}]_{\text{saturation}}$). Typically
585 for the pre-industrial tropical surface ocean Ω_{calc} is ~6, while for Eocene Tanzania
586 waters it is ~7 (Ref. 27; Extended Data Fig. 3). In support of the narrow range of
587 potential Ω_{calc} , a variety of modelling studies of the early Cenozoic show that surface
588 water Ω_{calc} remains within ± 1 , essentially constant in the early Cenozoic independent
589 of model boundary conditions^{23,79-81}. Therefore here we assume surface Ω_{calc} was
590 equal to 6.5 ± 1 throughout the Eocene.

591

592 **Monte Carlo pH-CO₂ estimates from planktonic foraminiferal $\delta^{11}\text{B}_c$.** Atmospheric
593 CO₂ was calculated using a Monte Carlo approach to solving the relevant carbonate
594 system equations with 10,000 iterations, deriving mean, upper and lower bounds of
595 95% of the simulations. We use the following constraints for our calculations: The
596 average seawater composition for $[\text{Ca}]_{\text{sw}} = 17 \text{ mmol/kg}^{82-86}$, $[\text{Mg}]_{\text{sw}} = 38 \pm 3 \text{ mmol/}$
597 kg^{83} (see also Supplementary Table 2), salinity = 35 ± 2 , and temperature (derived
598 from $\delta^{18}\text{O}$ of the shallowest dwelling species $\pm 2 \text{ }^\circ\text{C}$). All simulations are iterated
599 assuming Gaussian distribution of these parameters within the stated 2 s.d.. A similar
600 distribution is also applied to the $\delta^{11}\text{B}_c$ of the warmest and thus shallowest dwelling
601 species for each timeslice (Supplementary Table 2). We did not have sufficient
602 shallow dwellers in the 36.9 Ma timeslice for boron isotope work. Thus, the
603 intermediate dweller *Turborotalia cerroazulensis* was used. *T. cerroazulensis* was also
604 analyzed in the 40.3 Ma timeslice, and its $\delta^{11}\text{B}_c$ and $\delta^{18}\text{O}$ offset (and uncertainty) from
605 *M. lehneri*, (the shallowest dweller in the same timeslice) was applied to estimate the
606 shallowest temperature at 36.9 Ma (Supplementary Table 2). The temperature offsets
607 between species beyond the $\pm 2 \text{ }^\circ\text{C}$ uncertainty applied to our calculations are
608 considered unlikely. Note that a Gaussian distribution is not applicable to $\delta^{11}\text{B}_{\text{sw}}$
609 because there is equal likelihood that it laid between the minimum and maximum
610 constraints; we therefore applied a uniform “flat” probability $\delta^{11}\text{B}_{\text{sw}}$ for the Monte
611 Carlo simulations. The relative magnitude of each component of our error propagation
612 is shown in Extended Data Fig. 4.

613

614 **Correction to pH-CO₂ estimates from *Turborotalia ampliapertura* $\delta^{11}\text{B}_c$ in**
615 **published $\delta^{11}\text{B}_c$ -CO₂ records for the EOT¹².** We compared $\delta^{18}\text{O}$ measurements of
616 the surface dwellers *Planoglobanomalina* and *Pseudohastigerina* in the Eocene and

617 early Oligocene^{12,87} with that of *T. ampliapertura* used in Ref. 12, and confirmed that
618 *T. ampliapertura* gives a slightly cooler temperature than *Pseudohastigerina*. On
619 average, when contemporaneous shallow dwelling planktonic foraminifera are
620 available, *T. ampliapertura* are offset by $\sim +0.3$ ‰ in $\delta^{18}\text{O}$ and therefore occupy a
621 shallower water depth than *T. cerroazulensis* (whose $\delta^{18}\text{O}$ value is consistently ~ -0.53
622 ‰ higher than contemporaneous Eocene shallow dwelling planktonic foraminifera).
623 Further, considering the $\delta^{11}\text{B}$ offset between *T. cerroazulensis* and the shallow
624 dwelling *Morozovelloides lehneri* in the 40.3 Ma timeslice, we apply a $+1$ ‰
625 correction to *T. ampliapertura* $\delta^{11}\text{B}$ values of Ref. 11 to estimate surface seawater
626 values.

627

628 **Climate sensitivity calculations**

629 Our new CO_2 reconstructions allow us to examine their role in driving long-term
630 Eocene cooling. Climate forcing compared to the early Eocene timeslice at 53.2 Ma is
631 calculated from Eq. (1)²⁸ for our two CO_2 scenarios. We then assume that the apparent
632 latitudinal SST sensitivity for the Eocene was similar to that of the last ~ 520 ky¹³.
633 Estimates of SST change per $\text{W}\cdot\text{m}^{-2}$ forcing for each degree latitude were generated
634 using the second order polynomial fit through the data of Ref. 13 (dotted line Fig. 4b,
635 Extended Data Fig. 5b); as a high latitude SST sensitivity we used the average of
636 estimates for 60° North and South ($1.63 \text{ K}/\text{W}\cdot\text{m}^2$), and as low latitude estimates the
637 average of values between 30 °S and 30 °N ($0.36 \text{ K}/\text{W}\cdot\text{m}^2$). We use the ratio of high
638 or low latitude SST sensitivity and forcing for each timeslice to calculate the SST
639 change expected as a result of our reconstructed CO_2 forcing. The product gives the
640 estimated SST change of Fig. 4c and Extended Data Fig. 5c for high (blue symbols)
641 and low latitudes (red symbols) driven by CO_2 . The uncertainties (Fig. 4c, Extended

642 Data Fig. 5c) are the product of 1,000 realizations of each CO₂ estimate within its
643 uncertainty envelope (95% confidence intervals).
644
645 To test the importance of CO₂ to early and late Eocene warmth, we calculated the
646 sensitivity of the Earth system to radiative forcing, the Equilibrium Climate
647 Sensitivity (ECS). For ΔT we use MAT for the EECO and late Eocene, where EECO-
648 MAT is thought to be $\sim 14 \pm 3$ °C warmer than the pre-industrial, and ~ 5 °C warmer
649 than the late Eocene (35 Ma)^{2,29}. Of this ΔT , 4 to 6 °C is attributed to changes in
650 boundary conditions (paleotopography and continental configuration of the Eocene)
651 and the action of the slow climate feedbacks (mainly the lack of a continental ice sheet
652 on Antarctica and vegetation change²⁻⁴), for both the early and late Eocene. The
653 uncertainties in Fig. 4d to 4f (and Extended Data Fig. 5d to 5f) are the products of
654 1,000 realizations of the 53.2 Ma and 36.9 Ma CO₂ and MAT estimate based on
655 randomly sampling each variable within its 95% CI uncertainty envelope. The
656 uncertainty for the CO₂ and MAT estimates follows a Gaussian distribution, and for
657 the slow feedbacks correction on MAT follows a uniform “flat” probability.
658

659 **Data.** The underlying data of this manuscript may be found in the Supplementary
660 Tables 1 and 2.

661

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844

845 Extended Data Figure Legends

846 **Extended Data Figure 1: Paleogeography and $\delta^{18}\text{O}$ - derived temperature against**
847 **foraminiferal calcification depth. a.** Approximate paleoposition of TDP sites studied
848 here (map generated from www.odsn.de). **b.** Reconstructed temperature and relative
849 depth of each foraminifera within each time slice. Pale blue line represents the output
850 of a General Circulation Model (GCM) simulation run with Eocene boundary
851 conditions^{88,89}, while the other coloured lines show the GCM output offset to intersect
852 with the warmest temperature at depth “0” for each timeslice. Note that depth
853 assignments are approximate (see Methods). Also shown are the reconstructed
854 temperatures from core top planktonic foraminiferal $\delta^{18}\text{O}$ (Supplementary Table 1),
855 where *G. ruber* is assigned a depth “0” and the rest of the planktonic foraminifera are
856 offset to reproduce the CTD temperature profile of GLOW 15^{51,66}. For Holocene
857 temperature reconstructions we used the modern site latitude of 9 °S, and $\delta^{18}\text{O}_{\text{sw}}$
858 (SMOW) of 0 ‰.

859

860 **Extended Data Figure 2: $\delta^{11}\text{B}$ vs. pH as a function of $\delta^{11}\text{B}_{\text{sw}}$.** Increasing $\delta^{11}\text{B}_{\text{sw}}$
861 results in lower pH for the same $\delta^{11}\text{B}$. However for the same $\delta^{11}\text{B}$ range, the
862 reconstructed pH range is larger for higher $\delta^{11}\text{B}_{\text{sw}}$ (see brown shaded regions).

863

864 **Extended Data Figure 3: cGENIE estimates of calcite saturation (Ω_{calc}) in surface**
865 **waters.** Comparison for pre-industrial times (PI; blue) and 55 Ma (red) at different
866 latitudes.

867

868 **Extended Data Figure 4: Compilation of several CO_2 records for the Eocene in**
869 **comparison to this study.** The contribution of different parameters to the uncertainty
870 on our CO_2 reconstructions is colour-coded; sequentially from bottom to top, red is
871 from $\delta^{11}\text{B}_{\text{sw}}$, blue is from Ω_{calc} uncertainty, green is from $\delta^{11}\text{B}_c$ error, black is 40ppm
872 uncertainty in the event of disequilibrium with the atmosphere. Other parameters
873 contribute $< 10\%$ uncertainty to the CO_2 calculations and are not shown. Note data
874 from Pearson et al. ¹² (orange circles) are corrected as in Fig. 3, and there are two
875 scenarios included for the Tanzania records of Ref. 12 and this study: one with $\delta^{11}\text{B}_c =$
876 $\delta^{11}\text{B}_{\text{borate}}$ (closed blue and orange symbols) and the other applying *T. sacculifer* (open
877 blue and orange symbols) corrections to the shallowest symbiotic planktonic
878 foraminifera (Methods). Y-axis is in log-scale. Error bars are representative of each
879 proxy's reconstruction uncertainty (typically at 95% confidence). For the $\delta^{11}\text{B}$
880 reconstructions in this study, the errors are based on Monte Carlo propagation of
881 relevant errors (Methods).

882

883 **Extended Data Figure 5. CO_2 as a driver of latitudinal cooling in the Eocene, and**
884 **Equilibrium Climate sensitivity (ECS) analyses of the EECO and late Eocene**

885 **timeslices: case for the *T. sacculifer* calibration applied to shallowest planktonic**
886 **foraminifera. a.** Evolving relationship between SST¹⁴ for high (blue) and low (red)
887 latitudes and the CO₂ forcing of each of our timeslices relative to the EECO. Linear
888 regression fits and coefficients of determination (R²) are also shown, with the 95%
889 confidence interval (shaded bands). **b.** Apparent latitudinal SST sensitivity for the last
890 520 kyr¹³. Dashed line is second order polynomial through the SST sensitivity data
891 (grey crosses) of Ref. 13, and grey lines show 95% confidence interval. Red rectangle
892 surrounds the SST sensitivity estimates averaged as a low-latitude mean, and blue line
893 for the high latitude mean (see text and Methods). **c.** Reconstructed (lines) and
894 estimated (symbols) SST relative to 53.2 Ma. Symbols show each of our timeslices,
895 and calculated using the respective CO₂ reconstructions and the average low (red) and
896 high (blue) latitude SST sensitivities of b. Bold lines and shaded uncertainty band (at
897 95% confidence) show the reconstructed long-term mean TEX₈₆ SST estimates at high
898 (blue color >55°) and low (red color <30°) latitudes¹⁴ relative to ~53 Ma SST. Error
899 bars represent full propagation of errors at 95% confidence¹⁴. **d.** Mean Air
900 Temperature range for early (green) and late (black) Eocene corrected for changes due
901 to slow feedbacks^{2,4,21,29}. **e.** Forcing compared to pre-industrial calculated using our
902 CO₂ reconstructions for the 53.2 Ma (early Eocene) and 36.9 Ma (late Eocene)
903 timeslice (Methods). **f.** Probability density functions of ECS for the early (green) and
904 late (black) Eocene compared to IPCC estimates (dashed lines show the 95%
905 confidence interval (solid pink line)).

906

907 **Extended Data Figure 6. cGENIE estimates of air-seawater CO₂ disequilibrium.**

908 The model uses Eocene boundary conditions and positive values mean seawater is a
909 source of CO₂ (in ppm).

910

911 **Extended Data Figure 7. Comparison of Eocene and Modern planktonic**
912 **foraminiferal $\delta^{11}\text{B}$ and $\delta^{13}\text{C}$ with $\delta^{18}\text{O}$. a. and b.** Analyses from the 53.2 Ma and
913 40.3 Ma timeslices respectively (as in Fig. 2 and Supplementary Table 1).
914 *Cibicidoides* sp. are shaded. **c.** Core-top (Holocene) offshore Tanzania foraminiferal
915 measurements. Seawater $\delta^{11}\text{B}_{\text{borate}}$ and $\delta^{18}\text{O}$ was calculated from temperature,
916 Alkalinity and Dissolved Inorganic Carbon measurements (GLODAP Cruise 18 and
917 23, Station 17742 and 23037, 1995-07-24 and 1986-4-13, 53.959 °E -7.04 °N and
918 52.367 °E -6.33 °N respectively), correcting for anthropogenic carbon input. Black
919 line in panel c. represents seawater derived $\delta^{18}\text{O}$ and $\delta^{11}\text{B}_{\text{borate}}$ data (Methods).
920 Symbols key for the 53.2 and 40.3 Ma timeslices as in Fig. 2. Note the change in
921 scales for the x-axis between the Eocene and Holocene panels. Errors in $\delta^{11}\text{B}$
922 represent 2 s.d. of long-term precision (Methods).

923

924 **Extended Data Figure 8. Reconstructed pH using different combinations of**
925 **published symbiont-bearing and non-symbiotic foraminiferal vital effect**
926 **calibrations for the two most complete with depth timeslices.** Panel **a** shows the
927 case where no vital effect corrections were applied for comparison. The vital effect
928 corrections for deeper asymbiotic planktonic foraminifera are based on either
929 *Globigerina bulloides*²⁶ (panels **b** and **d**) and *Neogloboquadrina pachyderma*⁶⁵ (panels
930 **c** and **e**) calibrations. For shallow symbiont bearing planktonics we used calibrations
931 specific to modern shallow, symbiont-bearing *Trilobatus sacculifer*²² (panels **b** and **c**)
932 and *Orbulina universa*⁶² (panels **d** and **e**) (as recalculated by Ref. 64 and offset by -3
933 ‰ to account for analytical differences between NTIMS and MC-ICPMS

934 instrumentation^{8,9}). For comparison we also show the pH reconstructions if we assume
935 $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$.

936

937 **Extended Data Figure 9. The effect of seawater composition on boron isotope**
938 **calibrations in foraminifera.** The example of *T. sacculifer* calibration²² and seawater
939 borate curve (“borate” calibration: $\delta^{11}\text{B}_c = \delta^{11}\text{B}_{\text{borate}}$) for modern and Eocene seawater
940 composition as in Methods and furthered detailed in Supplementary Table 2.

941

942 **Extended Data Figure 10: cGENIE output of seawater pH vs. $\delta^{13}\text{C}$ for the top**
943 **~300 m of the ocean.** The scenarios explored are from offshore Tanzania (triangles),
944 Walvis Ridge (crosses), and the Global Ocean (circles) at three different atmospheric
945 CO_2 concentrations (modern, 3x pre-industrial and 16x pre-industrial). An additional
946 scenario showing 3x pre-industrial CO_2 but considering the temperature effect on
947 remineralisation is also shown as blue squares and triangles.







