1	Surface ocean cooling in the Eocene North Atlantic coincides with
2	declining atmospheric CO <sub>2</sub>
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#### 28 Key points:

Long-term (~4° C) decline in North Atlantic sea surface temperatures between the early
 (~53-49 Ma) and middle (~44-41 Ma) Eocene.

• This indicates that CO<sub>2</sub> was likely responsible for the onset of long-term Eocene cooling.

- However, east-west temperature gradients in the North Atlantic are decoupled, possibly
   due to additional non-CO<sub>2</sub> forcing mechanisms.
- 34

# 35 Abstract:

The Eocene (56-34 million years ago) is characterised by declining sea surface temperatures 36 (SSTs) in the low latitudes (~4°C) and high southern latitudes (~8-11°C), in accord with decreasing 37 38 CO<sub>2</sub> estimates. However, in the mid-to-high northern latitudes there is no evidence for surface water cooling, suggesting thermal decoupling between northern and southern hemispheres and 39 additional non-CO<sub>2</sub> controls. To explore this further, we present a multi-proxy (Mg/Ca,  $\delta^{18}$ O, TEX<sub>86</sub>) 40 SST record from Bass River in the western North Atlantic. Our compiled multi-proxy SST record 41 42 confirms a net decline in SSTs (~4°C) between the early Eocene Climatic Optimum (53.3-49.1 Ma) and mid-Eocene ( $\sim$ 44-41 Ma), supporting declining atmospheric CO<sub>2</sub> as the primary mechanism of 43 44 Eocene cooling. However, from the mid-Eocene onwards, east-west North Atlantic temperature 45 gradients exhibit different trends, which we attribute to incursion of warmer waters into the eastern North Atlantic and inception of Northern Component Water across the early-middle Eocene 46 47 transition.

48

## 49 Plain Language Summary

50 Over the past 541 million years, the Earth has oscillated between warm (greenhouse) and cold 51 (icehouse) climates. The most recent transition between a greenhouse and icehouse climate state 52 occurred during the Eocene (56 to 34 million years ago). This transition shows a gradual cooling, 53 previously suggested to be driven by a decline in atmospheric carbon dioxide (CO<sub>2</sub>). However, we 54 know little about this transition in the North Atlantic Ocean. Previous studies show limited cooling of 55 surface waters in this region. This suggests that changes in North Atlantic temperatures are not driven by CO<sub>2</sub>. To understand how sea surface temperature changes in the western North Atlantic, 56 we analysed the chemistry of microscopic marine fossils in sediments. Our results show a 4°C 57 decline in temperature from the early (~53 Ma) to the middle Eocene (~42 Ma). This matches 58 59 computer simulations of Eocene climate and confirms CO<sub>2</sub> was responsible for the transition. The lack of cooling observed in previous work is probably due to the development of an ancient water 60 mass known as Northern Component Water (observed today as North Atlantic Deep Water) and 61 62 changes in how the Eocene ocean transported heat.

63

## 64 Introduction

The early Eocene Climatic Optimum (EECO; 53.3 to 49.1 million years ago; Ma) (Hollis et al., 65 2019a; Zachos et al., 2001) is characterised by a long-term maximum in atmospheric  $CO_2$  (~1470 66 ppm) (Anagnostou et al., 2020), followed by a gradual decline in atmospheric CO<sub>2</sub> during the 67 middle Eocene (47.8 to 38.0 Ma) to ~800ppm (Anagnostou et al., 2020). This is consistent with 68 declining SSTs in the tropics (ca. 4°C) (Cramwinckel et al., 2018; Evans et al., 2018) and the mid-69 to-high southern latitudes (ca. 8-11 °C; Bijl et al., 2009; Hollis et al., 2009; Hollis et al., 2012). 70 71 However, SST estimates from the eastern North Atlantic suggest relatively muted surface water cooling (~1°C) between the EECO and middle Eocene (~40 Ma) (Bornemann et al., 2016). 72 73 Temperature asymmetry between the northern and southern hemisphere would not be expected 74 from a long-term decline in atmospheric CO<sub>2</sub> alone (Liu et al., 2018) and suggests that other non-CO<sub>2</sub> driving mechanisms (e.g. gateway reorganisation and/or changes in ocean circulation) may 75 76 influence regional SST patterns.

Of particular relevance is the growing evidence for Northern Component Water (NCW) initiation in the North Atlantic during the early-middle Eocene (~49 and 47 Ma) (Boyle et al., 2017; Hohbein et al., 2012; Norris et al., 2001). The onset of NCW has been attributed to gateway reorganisation, specifically deepening of the Greenland-Scotland Ridge (GSR) (Boyle et al., 2017; Hohbein et al., 2012; Vahlenkamp et al., 2018), although other mechanisms have been proposed 82 such as isolation of the Arctic Ocean (Zhang et al., 2011) or restriction of the Tethys Ocean (Roberts et al., 2009). The onset of NCW is followed by a period of weaker overturning (~42 to 38 83 Ma) (Witkowski et al., 2021), before re-invigoration of NCW during the late Eocene (~38 Ma) 84 (Coxall et al., 2018) or Eocene-Oligocene transition (EOT; ~34 Ma) (Hutchinson et al., 2019). The 85 establishment of NCW can transport additional heat into the eastern North Atlantic (Vahlenkamp et 86 al., 2018), potentially muting any long-term cooling trend in this region and has been invoked to 87 explain stable temperatures in the eastern North Atlantic during the middle Eocene (Bornemann et 88 89 al., 2016). However, our understanding of long-term North Atlantic temperature change is based on a single proxy record (planktonic foraminiferal  $\delta^{18}$ O) from a single site (DSDP Site 401; Bornemann 90 91 et al., 2016) and may not be regionally representative.

92 To test whether the wider North Atlantic region exhibits stable temperatures during the Eccene, we use a multi-proxy approach ( $\delta^{18}$ O, Mg/Ca, TEX<sub>86</sub>) to reconstruct SST in the western 93 North Atlantic (Bass River; ODP Leg 174AX; ~36°N paleolatitude) during the early-to-middle 94 95 Eocene (53.7 to 42.0 Ma). We compare our new dataset with climate model simulations spanning a wide range of CO<sub>2</sub> values to explore (i) temporal and spatial patterns of cooling in the North 96 Atlantic during the Eocene and (ii) whether there is thermal decoupling between the northern and 97 southern hemisphere during the Eocene. This allows us to test whether declining CO<sub>2</sub> is the 98 primary driver of long-term Eocene cooling or whether regional forcing mechanisms are also 99 important. 100

101

# 102 Methods

### 103 Site description

The Bass River section (ODP Leg 174AX; 39°36'N, 74°26'W) consists of calcareous marls and glauconitic silty clays deposited in middle to outer neritic paleodepths between 30 and 150 m (Fung et al., 2019; Miller et al., 2003). The biostratigraphic age model was developed using planktonic foraminifera and nannofossils (following Fung et al., 2019) with datums converted to the GTS2012 (Vandenberghe et al., 2012). Sediments span the early to middle Eocene (53.7 to 42.0 Ma) and encompass the EECO (~340 to 291 m). However, there are a series of hiatuses between
~49 and 44 Ma.

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## 112 Analytical methods

113 Lipid biomarker analysis was performed on 47 sediment samples. Approximately 5-10g of sediment was extracted with an Ethos Ex microwave extraction system using 15 ml of 114 dichloromethane (DCM) and methanol (MeOH) (9:1, v/v). The total lipid extract was separated over 115 116 silica into apolar and polar fractions using hexane:dichloromethane (9:1, v/v) and dichloromethane:methanol (1:2, v/v), respectively. The polar fraction (containing isoGDGTs) was 117 dissolved in hexane/isopropanol (99:1, v/v), passed through 0.45µm PTFE filters and analysed by 118 HPLC/APCI-MS following Hopmans et al. (2016). Trace element and stable oxygen isotope ( $\delta^{18}$ O) 119 120 planktonic foraminiferal analysis was performed on multiple depth intervals (n = 8) spanning the 121 early-to-middle Eocene. Foraminiferal preservation is excellent, appearing transparent or translucent under the light microscope, with no signs of diagenetic alteration observed under SEM 122 (Figure S1; Table S1). Analysis was performed on various surface-dwelling species 123 (Acarinina praetopilensis, Morozovella formosa. Morozovelloides 124 crassatus. and 125 Pseudohastigerina wilcoxensis) and deeper, thermocline-dwelling species (Parasubbotina hagni, Parasubbotina inaequispira). Single-specimen Mg/Ca analysis was performed via slow depth-126 profiling by laser ablation-inductively coupled mass spectrometry (LA-ICPMS) (see Müller et al., 127 2009; Supplementary Information; Table S2). Mg/Ca values were determined in multiple chambers 128 (~3 to 5) within a single specimen and averaged. The same specimens were subsequently 129 analysed for  $\delta^{18}$ O using a Multiprep-Isoprime 100 dual inlet system optimised for analysis of single 130 specimens (Supplementary information). 131

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## 133 Temperature calibrations

134 TEX<sub>86</sub> data was screened using established indices for non-Thaumarchaeota inputs 135 (Supplementary Information; **Figure S5**) and converted to SST using a Bayesian linear calibration 136 (prior mean = 25, prior standard deviation = 10, n = 2000) (Tierney and Tingley, 2014). Planktonic

for a miniferal  $\delta^{18}$ O values were converted to SST using the bayfox Bayesian calibration (prior mean 137 = 25, prior standard deviation = 20, n = 2000). Seawater  $\delta^{18}O(\delta^{18}O_{sw})$  values were obtained via 138 the isotope-enabled Community Earth System Model version 1.2 (iCESM1.2; see below). Mg/Ca 139 values were converted into SST using a modified version of MgCaRB (Gray and Evans, 2019) 140 141 (Supplementary Information). We report pH-corrected Mg/Ca temperatures as the majority of modern foraminifer species are characterised by Mg/Ca-pH sensitivity (Gray and Evans, 2019). 142 Planktonic foraminifera were rare and thus for Mg/Ca and  $\delta^{18}$ O, we report the 'average' SST 143 estimates for a given time slice (n = 8 for  $\delta^{18}$ O, n = 7 for Mg/Ca ) by combining (i) multiple-144 specimens from multiple size fractions and (ii) all surface-dwelling species within multiple genera 145 (i.e., Acarinina praetopilensis, Acarinina pseudotopilensis, Morozovella formosa, Morozovelloides 146 crassatus, Pseudohastigerina wilcoxensis) into a single estimate, following DeepMIP protocols 147 (Hollis et al, 2019; Supplementary Information). Average 'SST' estimates comprise a minimum of 148 two samples from a single depth horizon (see Data S4-S5). When SSTs are calculated using 149 individual species (Figure S2) and size segregating species (Figure S2-S3), similar patterns in 150 long-term trends are observed. 151

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### 153 Climate model simulations

We use the water isotope-enabled Community Earth System Model version 1.2 (iCESM1.2) (Zhu 154 et al., 2020; Zhu et al., 2019) to compare with our proxy reconstruction and to provide an 155 independent estimate of  $\delta^{18}O_{sw}$ . iCESM1.2 is able to closely replicate large-scale features of early 156 157 Eocene climate, including: i) enhanced global mean surface temperature estimates (Lunt et al., 2021; Zhu et al., 2019), ii) reduced meridional temperature gradients (Lunt et al., 2021), iii) 158 changes in the hydrological cycle (Cramwinckel et al., 2023), and iv) the values and distribution of 159 planktonic foraminifera  $\delta^{18}$ O values (Zhu et al., 2020). It is also the only DeepMIP model that has 160 161 water isotopes enabled (Zhu et al., 2020). The iCESM1.2 simulations were performed following the Deep-time Model Intercomparison Project protocols (Lunt et al., 2017) with early Eocene 162 paleogeography and vegetation (56.0–47.8 Ma) (Herold et al., 2014) and atmospheric CO<sub>2</sub> levels 163

of x1, x3, x6, and x9 preindustrial values (284.7 ppmv). Seawater  $\delta^{18}$ O in the simulations was 164 initialized from a constant value of -1.0% to account for the absence of ice sheets in a hothouse 165 climate (Shackleton and Kennett, 1975; Hollis et al., 2019). Previous studies at Bass River have 166 suggested sea level changes through the middle Eocene on the order of 20-30 m, that have been 167 attributed to changes in Antarctic ice volume (Fung et al., 2019). We do not adjust  $\delta^{18}O_{sw}$  in this 168 study for middle Eocene ice volume fluctuations, as the timing and magnitude of these ephemeral 169 glaciations are currently poorly constrained and our planktonic  $\delta^{18}$ O data from Bass River are from 170 intervals where water depth was greatest (i.e. ice volume was minimal). Our model results indicate 171 only minor changes in  $\delta^{18}O_{sw}$  at the Bass River location through the early-middle Eocene (~0.2‰ 172 change between x1 and x9 CO<sub>2</sub> simulations using iCESM1.2; **Table S3**). As such, we use the 173 average  $\delta^{18}O_{sw}$  value (-0.54‰) to calculate planktonic foraminiferal  $\delta^{18}O$ -derived SST estimates. 174 See Zhu et al. (2019; 2020) and Zhang et al. (2022) for further details of the experimental setup 175 176 and equilibration state.

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#### 178 Results

During the EECO (53.3 to 49.1 Ma), TEX<sub>86</sub> SST estimates average ~33°C (Figure 1a). Between 179 the EECO and the middle Eocene (44-41 Ma), TEX<sub>86</sub> SST estimates decline by ~5°C (Figure 1a). 180 Oxygen isotope SST estimates during the EECO from surface-dwelling planktonic foraminifera 181 average ~32°C (Figure 1a). Surface-dwelling species yield higher temperatures (up to ~5 °C 182 higher) than thermocline-dwelling species but exhibit a similar magnitude of cooling (~4°C) 183 between the EECO and the middle Eocene (44-41 Ma). During the early Eocene, Mg/Ca SST 184 estimates (calculated using the *G. ruber* calibration) average ~27°C (Figure 1a). These values are 185 lower than  $\delta^{18}$ O and TEX<sub>86</sub> SST estimates by ~5°C and ~6°C, respectively; Figure 1a) but agree 186 within the propagated calibration uncertainties. Mg/Ca SST estimates increase by ~3°C between 187 188 the EECO and middle Eocene (44-41 Ma; Figure 1a). However, the absolute values (~30°C) are comparable to middle Eocene-aged TEX<sub>86</sub> and  $\delta^{18}$ O SST estimates (28°C and 29°C, respectively) 189 and agree within the propagated calibration uncertainties. 190

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# 192 Discussion

193 Long-term cooling in the western North Atlantic during the Eocene

The use of multiple proxies provides more robust long-term temperature records than a single 194 195 proxy. The consistency between Mg/Ca, oxygen isotopes and TEX<sub>86</sub> values in the EECO and late Eccene is encouraging and indicates that each proxy is recording the same environmental signal 196 (i.e. SST). TEX<sub>86</sub> and  $\delta^{18}$ O values indicate very high SSTs at Bass River during the EECO (~32 to 197 33°C). These values are in agreement with existing low-resolution TEX<sub>86</sub> estimates generated at 198 Bass River (de Bar et al., 2019) and nearby South Dover Bridge (~34°C; Inglis et al., 2015). Mg/Ca 199 SST estimated are also relatively high (~27°C; **Figure 1**) but are lower than TEX<sub>86</sub> and  $\delta^{18}$ O-200 derived SST estimates by ~5-6°C. Between the EECO and middle Eocene (44-41 Ma), TEX<sub>86</sub> and 201 202  $\delta^{18}$ O values indicate gradual surface water cooling (5 and 4°C, respectively; **Figure 1a**), coherent with declining TEX<sub>86</sub> SSTs (~7°C) at South Dover Bridge between the EECO and middle Eocene 203 (~42 Ma). Evidence of cooling in two independent proxies (TEX<sub>86</sub>,  $\delta^{18}$ O) and locations provides the 204 205 first compelling evidence for surface ocean cooling in the (western) North Atlantic between the early and middle Eocene, which is in parallel with the inferred deep-ocean cooling in benthic 206 foraminifera  $\delta^{18}$ O record (Figure 1b; Westerhold et al., 2020). 207

In contrast, our new Mg/Ca SSTs increase by ~3°C between the EECO and middle 208 Eocene. Although middle Eocene (44-41 Ma) SST estimates are in excellent agreement with TEX<sub>86</sub> 209 and  $\delta^{18}$ O values (Figure 1) and alkenone-derived SST estimates (~29-30°C; Liu et al, 2018) from 210 nearby site IODP Site 1404, the temporal trends are inconsistent with regional observations (this 211 paper) (de Bar et al., 2019; Inglis et al., 2015) and declining global bottom water temperature 212 estimates inferred via changes in benthic foraminiferal  $\delta^{18}$ O values (Figure 1b) (Westerhold et al., 213 2020). To explore this mismatch further, we compared our proxy-derived temperature estimates 214 (TEX<sub>86</sub>, Mg/Ca,  $\delta^{18}$ O) from the EECO (53.3 to 49.1 Ma; Hollis et al. 2019) and middle Eocene (44 215 to 41 Ma) alongside iCESM1.2 simulations with different CO<sub>2</sub> scenarios (x1 to x9 pre-industrial 216 217 CO<sub>2</sub>) (Figure S6). These two intervals are chosen as they contain SST estimates from multiple

proxies (Mg/Ca,  $\delta^{18}$ O and TEX<sub>86</sub>) and exhibit a similar sampling density. iCESM1.2 simulated SSTs 218 at the Bass River are 31 and 27 °C in the ×6 and ×3 PI CO<sub>2</sub> simulations, respectively (Figure 2b), 219 which overlaps with proxy reconstructions (Figure 2a; Figure S6). For a two-fold decrease in 220 atmospheric CO<sub>2</sub> (i.e., from x6 to x3 PI CO<sub>2</sub>), the model predicted decrease in SST of  $\sim$ 4°C is 221 comparable to the magnitude of cooling captured by TEX<sub>86</sub> and  $\delta^{18}$ O (5 and 4 °C, respectively; 222 Figure S6) between the EECO and middle Eocene, but is inconsistent with warming observed in 223 Mg/Ca values. Given that proxy-derived CO<sub>2</sub> estimates decline from ~1470 ppm (~x5 PI CO<sub>2</sub>) to 224 ~800ppm (~x3 PI CO<sub>2</sub>) during this interval (Anagnostou et al., 2020), this implies additional non-225 thermal controls on Mg/Ca values at this site. 226

The choice of Mg/Ca calibration remains uncertain when working with extinct species. 227 However, the discrepancy between Mg/Ca-derived SSTs and other proxy data is insensitive to the 228 choice of Mg/Ca calibration approach (Supplementary Information). This is because seawater pH 229 230 was substantially lower than modern throughout the Eocene (Anagnostou et al., 2020), such that choosing a G. ruber or T. sacculifer-like calibration has a minor effect on the long-term Mg/Ca-231 derived trend in our dataset (Figure S3). Seawater Mg/Ca is also well-constrained for the Eocene 232 233 (Evans et al., 2018; Gothmann et al., 2015) and is broadly invariant across this interval, such that it 234 is very unlikely that unidentified changes mask cooling. Given that this site was targeted for its exceptional foraminiferal preservation and diverse assemblages (Figure S1), this potentially points 235 236 towards either an evolutionary control on Eocene planktonic foraminifera Mg incorporation, or a 237 shift in seawater carbonate chemistry at this site that substantially differs from the existing pH records (Anagnostou et al., 2020; Rae et al., 2021; see Supplementary Information for more 238 discussion). Resolving this issue and exploring any other additional controls (e.g., local 239 240 hydrographic variability; c.f. Thornalley et al., 2011) will require further data and is beyond the scope of this study. We continue to include the Mg/Ca SST estimates in our assessment of the 241 thermal evolution of Bass River (**Figure 2a**) and note that mismatches in  $\delta^{18}$ O and Mg/Ca derived 242 SSTs are not unique to deep-time species. Furthermore, this discrepancy in inorganic geochemical 243 temperature reconstructions may ultimately stem from a small number of Mg/Ca analyses in the 244

early Eocene, highlighting the benefit of working with a larger numbers of specimens, wherepossible.

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# 248 Divergent zonal temperature gradients in the North Atlantic during the early-to-middle Eocene

To determine the long-term mean SST evolution at Bass River, we fit LOESS regressions to our 249 multi-proxy dataset (TEX<sub>86</sub>, Mg/Ca,  $\delta^{18}$ O) (Supplementary Information). This approach indicates 250 net cooling (~4°C) in the western North Atlantic between the EECO and middle Eocene (Figure 251 **2a**). Our data from the western North Atlantic contrasts with existing planktonic foraminifera  $\delta^{18}$ O-252 derived SST estimates from the eastern North Atlantic (~37° N; DSDP Site 401; Bornemann et al., 253 2016) that indicate minimal (<1°C) or no cooling between the EECO and late middle Eocene (ca. 254 42-40 Ma) (Figure 2a). CESM1.2 model simulations show that the magnitude of cooling at Bass 255 256 River inferred via proxies is consistent with a halving of  $CO_2$  (Figure 2c) but that the magnitude of proxy-inferred cooling at DSDP Site 401 is much lower than expected (Figure 2b-c). The east-257 west zonal mean temperature gradient inferred via proxy estimates (~15-20°C) is also larger than 258 inferred via model simulations (~3°C; Figure 2b). As the model simulations are identical with the 259 exception of changes in CO<sub>2</sub>, this implies that non-CO<sub>2</sub> controls influence SSTs in the eastern 260 North Atlantic (DSDP Site 401; Bornemann et al., 2016) during the Eocene. 261

262 Planktonic foraminifera at Bass River exhibit excellent preservation (Supplementary Information) and tests are translucent and 'glassy' (Figure S1) whereas Hollis et al. (2019) 263 classified post-PETM planktonic foraminifera at DSDP Site 401 as 'recrystallized'. However, post-264 PETM foraminifera at DSDP Site 401 exhibit good preservation (Bornemann et al., 2016) and show 265 266 limited evidence for recrystallization. If planktonic foraminifera had been subject to significant postdepositional alteration, they would be "reset" towards deep-sea temperatures and would track 267 changes in benthic foraminiferal  $\delta^{18}$ O values (Pearson et al., 2007). However, planktonic 268 for a miniferal  $\delta^{18}$ O values at DSDP Site 401 do not co-vary with benthic  $\delta^{18}$ O values, either at this 269 site (Bornemann et al., 2016) or elsewhere (Westerhold et al., 2020). Therefore, this is unlikely to 270

explain the observed trends (Figure 2a). However, additional SST records from the North Atlantic
are required to explore regional variations further.

Alternatively, changes in ocean circulation could have modulated regional temperature 273 patterns in the eastern North Atlantic during the middle-to-late Eocene, specifically the onset of 274 275 Northern Component Water (NCW) formation. None of the DeepMIP models (including CESM1.2) show deep overturning circulation (> 2,000 m) in the North Atlantic during the early Eocene (Zhang 276 et al., 2022), consistent with proxy evidence (e.g., benthic foraminifera  $\delta^{13}$ C and fish teeth  $\epsilon_{Nd}$ 277 278 values; see Zhang et al. (2022). Instead, most of the DeepMIP models (and CESM simulations 279 with x1 to x3 CO<sub>2</sub>) suggests that deep water formation is likely to form in the Southern Ocean, which also broadly agrees with proxy-based evidence from the early Eocene (Zhang et al., 2022). 280 281 CESM does simulate a North Atlantic deep/intermediate water formation at 1x PI CO<sub>2</sub>, suggesting that NCW formation represents a delicate balance between multiple factors such as global or 282 283 regional cooling, widening of the Atlantic basin, closure of the Arctic-Atlantic gateway (Hutchinson et al., 2019) and/or deepening/opening of the Greenland-Scotland Ridge (Vahlenkamp et al., 2018; 284 Straume et al., 2022). The lack of deep water formation in iCESM1.2 at high CO<sub>2</sub> concentrations 285 (i.e. x6 CO<sub>2</sub>) is likely related to the initial condition and short integration length (see Zhang et al., 286 287 2022 for further discussion). We speculate that this limitation in the iCESM1.2 simulation at  $6x CO_2$ would have a minor impact on the surface ocean of the North Atlantic, where regional ocean-288 atmosphere coupling and wind driven circulation are more important in determining the SSTs. 289

Idealised modelling experiments show that deepening of the Greenland-Scotland Ridge 290 and/or closure of the Arctic-Atlantic gateway (Hutchinson et al., 2019) can initiate NCW formation 291 in the North Atlantic and increase SST in the eastern North Atlantic by up to 7 °C (Vahlenkamp et 292 al., 2018), thus muting any long-term CO<sub>2</sub>-driven cooling at DSDP Site 401. Importantly, deepening 293 of the Greenland-Scotland Ridge has only a minimal influence (< 1°C) on SSTs in the western 294 North Atlantic (i.e., where Bass River is located) (Vahlenkamp et al., 2018). There is growing 295 geochemical and sedimentological evidence placing the initial onset of NCW between ~49 and 47 296 297 Ma, coincident with changes in zonal temperature gradients between the eastern and western 298 North Atlantic. Evidence for onset of NCW between ~49 and 47 Ma includes development of contourite drifts in the western North Atlantic (Boyle et al., 2017), changes in biosiliceous sedimentation (Witkowski et al., 2021) and a collapse in  $\delta^{13}$ C gradients between the North and South Atlantic (Hohbein et al., 2012). These changes would also influence local hydrography within the eastern North Atlantic and could exert an additional control on  $\delta^{18}O_{sw}$  values at DSDP Site 401.

303 Proxy-based reconstructions during the Middle Eocene Climatic Optimum have argued that 304 northward expansion of the North Atlantic subtropical gyre could also act as a mechanism to increase SSTs within the North Atlantic (Van Der Ploeg et al., 2023). However, details of the gyre 305 306 heat transport and the impact of this large-scale process on regional SSTs (especially near 307 coastlines) requires further investigation. Thus, although diverging zonal temperature gradients in 308 the North Atlantic are consistent with the initial early onset of NCW during the early-middle Eocene, 309 additional proxy data and isotope-enabled model simulations are required to test this further. From 310 a model-based perspective, simulations with higher resolution and longer simulation length are 311 required to explore the equilibrium state of the modelled ocean circulation and any possible regional features that may be missed by the relatively coarse ( $\sim 1-2^{\circ}$ ) resolution model. 312

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314 Synchronous surface water cooling in the northern and southern hemispheres during the Eocene

315 To explore whether long-term cooling is globally synchronous, we compiled TEX<sub>86</sub>-derived SST estimates that span the early (55 Ma) to late (34 Ma) Eocene. To avoid relying on single proxy 316 records, we focus on regions with two or more TEX<sub>86</sub> records. Our data compilation spans three 317 regions: (i) the equatorial Atlantic (0-30° N/S) (Cramwinckel et al., 2018; Zhang et al., 2013; Inglis 318 et al., 2015; Liu et al., 2009), (ii) the northwest Atlantic (30-50 °N) (this study; Keating-Bitonti et al., 319 2011; Inglis et al., 2015; Cramwinckel et al., 2020a; van der Ploeg et al., 2023) and (iii) the 320 southwest Pacific (>50°S) (Bijl et al., 2013; Bijl et al., 2009; Crouch et al., 2020; Hollis et al., 2009; 321 Inglis et al., 2015; Cramwinckel et al, 2020b; Liu et al., 2009). (Figure 3; Supplementary 322 Information). 323

Our results suggest that the onset of long-term cooling occurs ~49 to 48 million years ago in the North Atlantic and southwest Pacific (i.e. following the termination of the EECO; **Figure 3a-c**) and coincides with an increase in the latitudinal SST gradient from 49 to 44 Ma (**Figure 3d**). Our 327 study indicates that the onset of Eocene cooling is a global feature and thus consistent with a decline in atmospheric CO<sub>2</sub> as a forcing mechanism for cooling. However, there is a relative lack of 328 data in the North Atlantic from ~49 to 48 Ma, such that additional records are required to determine 329 330 the exact onset of long-term cooling. Proxy records have also suggested that ocean gateways may 331 have played an important role at this time (e.g., Hohbein et al., 2012; Bijl et al., 2009; Bijl et al., 332 2013). Previous work argues that the Tasman Gateway was open to shallow circulation at this time (~49 to 46 Ma) (Bijl et al., 2013) and deepening of the Tasman Gateway would initiate regional 333 334 surface water cooling (Sijp et al., 2011; Sijp et al., 2016) and may account for declining SSTs in the SW Pacific between the termination of the EECO and middle Eocene (~44 Ma). However, as 335 surface ocean cooling occurs in multiple basins (Figure 3a-c) at a comparable time (~49-48 Ma), it 336 337 suggests that  $CO_2$  was likely responsible for the majority of long-term Eocene cooling.

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## 339 Conclusions

Here we present the first multi-proxy (Mg/Ca,  $\delta^{18}$ O, TEX<sub>86</sub>) SST record from the western North 340 Atlantic spanning the early-to-middle Eocene. Our results indicate very high SSTs during the early 341 Eocene Climatic Optimum ( $\sim$ 27-33°C), in agreement with high atmospheric CO<sub>2</sub> concentrations. 342 343 Our compiled dataset reveal a net decline (~4°C) in SSTs between the early Eocene Climatic Optimum (53.3-49.1 Ma) and the middle Eocene (44-41 Ma), consistent with long-term decrease in 344 atmospheric CO<sub>2</sub>. However, east-west zonal temperature gradients in the North Atlantic are likely 345 decoupled during the early-to-middle Eocene. This may be related to inception of Northern 346 Component Water at the early-middle Eocene transition and incursion of warmer waters into the 347 eastern North Atlantic, but additional datasets are required to test this further. We also 348 demonstrate that the onset of long-term Eocene cooling in the western North Atlantic (~49-48 Ma) 349 occurs synchronously in other ocean basins (e.g., N. Atlantic vs S. Pacific) and across different 350 latitudinal bands, implying that CO2 was likely responsible for the onset of long-term Eocene 351 352 cooling.

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### 369 **Open Research**

Inorganic and organic geochemical data and associated sea surface temperature estimates are
 available at OSF (Inglis et al., 2023). The transformation of measured Mg/Ca into temperature was
 calculated using MgCaRB (Evans et al., 2023). Loess regressions were calculated using R
 (http://www.R-project.org).

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# 375 Captions

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Figure 1. a) SST reconstructions from Bass River during the early-middle Eocene inferred via TEX<sub>86</sub> (blue), planktonic foraminifera  $\delta^{18}$ O (dark orange) and Mg/Ca (light orange). Error bars represent the 95% confidence intervals. b) Atmospheric CO<sub>2</sub> reconstructions inferred via planktonic foraminifera  $\delta^{11}$ B (blue circles) and alkenone  $\delta^{13}$ C (blue squares). Error bars represent ±1 standard deviation (Rae et al., 2021). c) benthic foraminifera  $\delta^{18}$ O values (Westerhold et al., 2020).

Figure 2: Divergent zonal temperature gradients in the North Atlantic during the early-to-middle 383 Eocene. a) proxy-derived SST reconstructions for Bass River (this study; blue symbols) and DSDP 384 Site 401 ( $\delta^{18}$ O only; orange symbols) (Bornemann et al., 2016) fitted with a LOESS regression. 385  $\delta^{18}$ O values from DSDP Site 401 re-calculated for surface-dwelling foraminiferal genera (*Acarinina* 386 and Morozovella spp.) using the bayfox Bayesian calibration ( $\delta^{18}O_{sw} = -0.81$ , prior mean = 25, prior 387 standard deviation = 20, n = 2000).  $\delta^{18}O_{sw}$  values obtained via iCESM1.2 (Table S3). Error bars 388 389 represent the 95% confidence intervals. b) iCESM1.2-derived SST estimates for Bass River (blue symbols) and DSDP Site 401 (orange symbols) under different CO<sub>2</sub> concentrations, c) iCESM1.2-390 derived  $\Delta$ SST estimates (x6 PI CO<sub>2</sub> - x3 PI CO<sub>2</sub>) with proxy-derived cooling between the early- to 391 392 middle Eocene shown for each site

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394 Figure 3: Long-term evolution of surface ocean temperatures during the Eocene inferred via TEX<sub>86</sub> in the (a) equatorial Atlantic (Cramwinckel et al., 2018; Inglis et al., 2015; Zhang et al., 2013), b) 395 396 North Atlantic (this study; Inglis et al., 2015; de Bar et al., 2019), and c) the southwest Pacific (Bijl et al., 2013; Bijl et al., 2009; Crouch et al., 2020; Hollis et al., 2009; Inglis et al., 2015). Panel (d) 397 398 shows the SST gradient between the equatorial Atlantic and the North Atlantic (dark blue line) and southwest Pacific (light blue line). To determine the long-term mean SST evolution for the low-, 399 mid-, and high-latitudes, nonparametric LOESS regressions were fitted using the fANCOVA 400 software package (http://www.R-project.org/). 401

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Figure 1.



Figure 2.



Figure 3.

