

1 **Export of nutrient rich Northern Component Water preceded early Oligocene**  
2 **Antarctic glaciation**

3

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27 **Onset of North Atlantic deep water formation is thought to have coincided with Antarctic**  
28 **ice sheet growth about 34 million years ago. However, this timing is debated, in part due to**  
29 **questions over the geochemical signature of ancient Northern Component Water formed in**  
30 **the deep North Atlantic. Here we present detailed geochemical records from North Atlantic**  
31 **sediment cores located close to sites of deep water formation. We find that prior to 36**  
32 **million years ago, the northwestern Atlantic was stratified, with nutrient-rich, low salinity**  
33 **bottom waters. This restricted basin transitioned into a conduit for Northern Component**  
34 **Water that began flowing southwards approximately one million years before initial**  
35 **Antarctic glaciation. The probable trigger was tectonic adjustments in subarctic seas that**  
36 **enabled increased exchange across the Greenland-Scotland Ridge. Increasing surface**

37 **salinity and density strengthened Northern Component Water production. The late Eocene**  
38 **deep water mass differed in its carbon isotopic signature from modern as a result of**  
39 **leakage of fossil carbon from the Arctic Ocean. Export of this nutrient-laden water**  
40 **provided a transient pulse of CO<sub>2</sub> to the Earth system perhaps causing short-term**  
41 **warming, whereas the long-term effect of enhanced NCW formation was greater**  
42 **northward heat transport that cooled Antarctica.**

43  
44 Production of deep water in the North Atlantic Ocean plays a vital role in maintaining the global  
45 meridional overturning circulation (MOC)<sup>1</sup>. North Atlantic Deep Water (NADW), the lower  
46 branch of the Atlantic part of the MOC (AMOC), forms in the Labrador and Nordic Seas as  
47 surface waters cool and densify. The sinking is largely controlled by an interplay of (i) the  
48 stratification at convection sites, determined by the balance of warm salty water from low  
49 latitudes, cold freshwater from the Arctic Ocean, and local heat and freshwater fluxes, and (ii)  
50 wind-driven upwelling in the Southern Ocean, which returns deep water to the surface<sup>2,3</sup>. Both  
51 factors likely impacted the early Cenozoic MOC state, when Atlantic bathymetry and ocean  
52 gateways were different and global temperatures were warmer than today<sup>4,5</sup>. However, resolving  
53 their interplay at the onset of NADW production, referred to here as its palaeo pre-cursor  
54 Northern Component Water (NCW), is challenging because the early history of the AMOC  
55 remains poorly constrained.

56  
57 Benthic foraminifera  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  records constrain development of global deep water  
58 circulation by giving insights into subsurface temperatures, salinity and nutrients<sup>5</sup>. A widely held  
59 view is that NCW began filling the Atlantic close to the Eocene-Oligocene greenhouse to

60 icehouse climate transition (EOT), ~34 million years ago (Ma)<sup>4,6-9</sup>, or earlier<sup>10-13</sup>. Others argue  
61 that emergence of significant NCW was delayed until the late Miocene<sup>14</sup>. Modeling studies  
62 diverge, suggesting either no NCW<sup>15</sup> during the EOT, strengthening/onset of bipolar deep water  
63 formation triggered by Drake Passage deepening<sup>16-18</sup>, or an ocean state with robust NCW  
64 throughout<sup>19</sup>. Previous data studies that argue for a late Eocene onset of NCW production  
65 assume that early Cenozoic NCW was nutrient-poor with a high ('young') benthic  $\delta^{13}\text{C}$  signature  
66 similar to modern well ventilated NADW<sup>4,10,13,20</sup>. Yet palaeo data from northerly regions suitable  
67 for characterizing NCW are lacking.

68

69 To fill this gap, we produced EOT benthic  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  records from four deep sea sites (>1000  
70 m palaeodepth) in the high latitude North Atlantic (see Methods and Supplementary  
71 Information). Of these, Site 647 in the Southern Labrador Sea (SLS; 47°N, 34 Ma palaeolatitude,  
72 ~2000-3000 m palaeodepth), is the most northerly EOT sequence containing calcareous  
73 microfossils necessary for  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  analysis<sup>21</sup>. Additional data for portions of the late  
74 Eocene were generated from DSDP Sites 112 and 612, and IODP Site U1411 (Fig. 1). The latter  
75 two sites should record NCW export in the deep western boundary current (DWBC) (Fig. 1). The  
76 data are compared against an Atlantic isotope compilation incorporating 14 previously  
77 investigated sites (Fig. 2). Records of benthic foraminifera Mg/Ca, fish tooth  $\epsilon\text{Nd}$ , and planktic  
78 foraminifera  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  from Site 647 and U1411 were also generated to provide constraints  
79 on bottom water temperature (BWT) and provenance, and water column structure (Methods).  
80 The results add unique perspectives on Atlantic end-member deep water properties and changes  
81 in circulation during the EOT.

82

83 **High nutrient content of late Eocene Southern Labrador Sea deep waters**

84 At Site 647 we recognize the typical pattern of  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  increase ( $>1.0\text{‰}$  and  $\sim 0.5$  to  $1.0\text{‰}$   
85 respectively) between 34-33.5 Ma diagnostic of early Oligocene Antarctic glaciation, including  
86 the peak in  $\delta^{18}\text{O}$  seen at other sites<sup>22</sup>, here referred to as the Early Oligocene Glacial Maximum  
87 (EOGM) (Fig. 2). The first novel observation at Site 647 is that before  $\sim 35.8$  Ma,  $\delta^{13}\text{C}$  of SLS  
88 bottom water is on average 0.5-1 ‰ lower than all southerly sites. This is opposite to the modern  
89 AMOC state, where northern deep waters have the highest  $\delta^{13}\text{C}$  due to sinking of well ventilated,  
90 nutrient poor surface waters<sup>23</sup>. The low  $\delta^{13}\text{C}$  may reflect nutrient accumulation under stratified  
91 conditions analogous to the modern North Pacific, i.e. the end of the circulation path. This could  
92 imply a southern-sourced deep water filled the SLS during the late Eocene. However, Site 647  
93 fish debris  $\epsilon\text{Nd}$ , an isotopic tracer for the origin of deep water masses, bear the fingerprint of a  
94 northern hemisphere source ( $\epsilon\text{Nd} = -11.4$  to  $-9.4$ ) throughout the studied interval (Fig. 3g).  
95 Consequently, we instead argue the low benthic  $\delta^{13}\text{C}$  reflects local bottom water sourced from  
96 surface waters with a high nutrient concentration, within the narrow, restricted North Atlantic  
97 basinal deep water circulation. A probable nutrient source is ‘fossil’ carbon leaking in from the  
98 Arctic Ocean and subarctic seas (Greenland and Norwegian Seas), which had high nutrient  
99 stocks during the Eocene because of their semi-isolation, heavily vegetated margins<sup>24</sup>, and high  
100 riverine inflow<sup>25-27</sup>.

101

102 We identify three circulation regimes based on our new Eocene-Oligocene proxy records and  
103 comparisons with published data (Figs. 2 and 3). Focusing first on  $\delta^{13}\text{C}$  (Fig. 2), under Regime 1  
104 ( $>35.8$  Ma), SLS bottom waters were isolated from the rest of the Atlantic and a distinct NCW  
105 with low  $\delta^{13}\text{C}$  bathed Site 647. After  $\sim 35.8$  Ma an approximately 1.5-2 million year-long ( $\sim 35.8$   
106 to  $\sim 33.8$  Ma) negative  $\delta^{13}\text{C}$  excursion ( $0.5$ - $1\text{‰}$   $\delta^{13}\text{C}$  decrease) is seen to varying degrees at Sites  
107 612, U1411 and 647, as well as some other Atlantic sites (1053, 1090, 366). Offsets in this

108 excursion's timing between sites are likely caused by age model differences. The phase  
109 encompassing the onset and peak of the excursion is Regime 2. Importantly, the  $\delta^{13}\text{C}$  excursions  
110 are largest (maximum 1‰) at Sites 612 and U1411 sitting in the DWBC (Fig. 1). While noted  
111 previously<sup>28</sup>, the wider significance of this  $\delta^{13}\text{C}$  excursion has not been fully explored. The  
112 observation that Sites 612 and U1411, directly down stream of Site 647, gain benthic  $\delta^{13}\text{C}$   
113 signals close to the SLS end-member suggests the signal was propagated from the north. Thus it  
114 records southward export of Arctic imprinted, nutrient-rich NCW. The increase in Atlantic  
115 benthic  $\delta^{13}\text{C}$  towards the end of Regime 2 indicates that either the pulse of NCW ended, or  
116 sufficiently ventilated surface water with higher  $\delta^{13}\text{C}$  was imported to convection sites. Regime  
117 3, described below, represents the phase where a more mature form of NCW existed.

118

### 119 **Northern deep water cooling, salinification and destratification**

120 A second prominent feature is the pattern of SLS benthic  $\delta^{18}\text{O}$ . The majority of pre-Oligocene  
121  $\delta^{18}\text{O}$  data south of Site 647 range between 0.4-1.2‰ (Fig. 2). Strikingly, in the SLS during  
122 Regime 1, benthic  $\delta^{18}\text{O}$  is 1-3‰ lower than the ensemble. The primary controls on benthic  $\delta^{18}\text{O}$   
123 are BWT and the  $\delta^{18}\text{O}$  composition of seawater, the latter reflecting global glacial ice volume  
124 and local salinity. Assuming minimal ice before 34 Ma, the relatively low  $\delta^{18}\text{O}$  in SLS benthos  
125 indicates a considerably warmer or fresher water mass bathing the seafloor compared to southern  
126 stations. Benthic  $\delta^{18}\text{O}$  from Sites 647 and 112 increased gradually from 36.0 to 35.4 Ma, then  
127 again from ~34.6-34.4 Ma, and had converged close to the dominant Atlantic trend by ~34.3 Ma,  
128 i.e. coincident with or just lagging the Atlantic-wide  $\delta^{13}\text{C}$  minimum. Diagenetic alteration of Site  
129 647 benthic fossils<sup>29</sup> is ruled out due to (i) the excellent fossil calcite preservation<sup>21</sup>  
130 (Supplementary Information), and (ii) the similarity of our new planktic  $\delta^{18}\text{O}$  values from Sites

131 647 and U1411 (Fig. 3). Moreover, a similar pattern of decreasing benthic  $\delta^{18}\text{O}$  is seen in a  
132 North Sea record<sup>30</sup>, although at shallower depths (~500m).  
133  
134 Mg/Ca BWT help deconvolve temperature and salinity influences on  $\delta^{18}\text{O}$  (Methods and  
135 Supplementary Information). Across the EOT, Site 647 Mg/Ca data suggest BWT cooling of  
136  $\sim 1^\circ\text{C}$  combined with a  $\sim 0.6\text{‰}$  increase in  $\delta^{18}\text{O}_{\text{sw}}$ , in agreement with previous studies<sup>31</sup> (Fig. 3a).  
137 From this, we estimate gradual bottom water cooling in the SLS of  $\sim 3\text{-}4^\circ\text{C}$  between 37.5 Ma to  
138 35 Ma, which is similar to observed northern high latitude sea surface coolings<sup>32</sup>. Substituting  
139 the Mg/Ca BWTs into a  $\delta^{18}\text{O}$  palaeotemperature equation (Methods) yields ice-free  $\delta^{18}\text{O}_{\text{sw}}$   
140 estimates of between -3 to -4 ‰ SMOW during this interval. Based on these  $\delta^{18}\text{O}_{\text{sw}}$  constraints,  
141 we estimate late Eocene SLS bottom salinities using relevant modern  $\delta^{18}\text{O}_{\text{sw}}$ -sea surface salinity  
142 (SSS) relationships<sup>33</sup> (Fig. 3c).  
143  
144 Applying a modern SSS- $\delta^{18}\text{O}_{\text{sw}}$  relationship from eastern Greenland, today a conduit for low  
145 salinity (32 PSU) Arctic outflow, implies Site 647 bottom water salinity of 30-32 PSU before 36  
146 Ma, increasing by 2-3 PSU from 36 to 34 Ma. The salinity change is similar when a Laptev Sea  
147 (today fed by large Siberian rivers)  $\delta^{18}\text{O}_{\text{sw}}$ -salinity regression is applied. While SSS- $\delta^{18}\text{O}_{\text{sw}}$   
148 relationships are spatially widely variable, and modern relationships are only loose analogues for  
149 the Eocene, our inferred values are compatible with modern temperature-salinity fields.  
150 Therefore, we suggest that (i) before 36 Ma SLS bottom waters were relatively fresh, and (ii)  
151 SLS bottom salinity increased from Regime 1 to 2. This conclusion does not change even if  
152 samples older than 35 Ma are biased to higher Mg (Supplementary Information) since the  
153 salinity signal is embedded in the benthic  $\delta^{18}\text{O}$ , which is independent of Mg/Ca. This

154 interpretation is consistent with that for pre-formed nutrients—they are both likely derived from  
155 the Arctic Ocean. Proxies and models agree that the Arctic had a thick freshwater cap during the  
156 Palaeogene as a consequence of a strong hydrological system and high fluvial inputs under  
157 greenhouse forcing, combined with restricted salt input<sup>26,33-35</sup>. With no Pacific Ocean outlet at  
158 this time, major surface discharge occurred through the Nordic Seas<sup>34,36</sup>.

159

160 Site 647 planktic foraminifera  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  add information on the upper water column.  $\delta^{18}\text{O}$  of  
161 mixed layer (surface) species is 1-2 ‰ lower than deep-dwellers (subthermocline), consistent  
162 with a stratified upper ocean and calcification of mixed-layer dwellers high in the water column  
163 or during the warmest season (Fig. 3b). During Regime 1, and before 34.5 Ma,  $\delta^{18}\text{O}$  of the deep-  
164 dwelling planktic species is indistinguishable from benthic foraminifera, reflecting influence of  
165 relatively fresh deep water at sub-thermocline levels in the SLS. Site 647 planktic data are sparse  
166 before 35 Ma due to low foraminiferal abundance and coring gaps. However, after 34.5 Ma,  
167 deep-dwelling planktic and benthic  $\delta^{18}\text{O}$  records separate coincident with the appearance of deep  
168 water with temperature and salinity properties similar to typical Atlantic values. Additionally, a  
169 progressive collapse in the planktic – benthic  $\delta^{13}\text{C}$  gradient (Fig. 3d, e) is documented that  
170 captures the SLS water column transitioning from being well-stratified with large vertical  $\delta^{13}\text{C}$   
171 differences (1-1.5 ‰) during Regime 1, to a state with a smaller  $\delta^{13}\text{C}$  gradient (0.5‰)  
172 comparable to better-mixed modern North Atlantic convection sites<sup>23</sup> by ~34.3 Ma. Both  
173 observations are consistent with increasing NCW volume. An abrupt shift in Site 647 benthic  
174 assemblages at 34.3 Ma from agglutinated species tolerant of carbonate-poor, nutrient-rich  
175 environments, to calcareous species suited to stronger current flow<sup>37</sup> coincident with other  
176 changes (Fig. 3f), provides further evidence for increased convection. Circulation Regime 3

177 begins at 34.3 Ma, when a saltier, denser form of NCW with higher  $\delta^{13}\text{C}$  is exported through the  
178 SLS.

179

### 180 **Deep water sources and sinking**

181 Our Site 647 fish debris  $\epsilon\text{Nd}$  data behave as a conservative tracer of northern sourced deep water (see  
182 Supplementary Information) and can be compared to published ocean references<sup>38,39</sup> to identify probable  
183 NCW source regions (Fig. 3g). While we do not reconstruct Nd directly for Regime 1, our sample from  
184 39 Ma is similar to the rest of the record, implying no systematic change in  $\epsilon\text{Nd}$  and thus bottom water  
185 provenance as NCW evolved. The comparison suggests that the Southern Ocean, which has the highest  
186 end-member  $\epsilon\text{Nd}$  signature in our compilation, was not the source of bottom waters at Site 647.

187 Moreover we do not find evidence for the prominent EOT shift to high  $\epsilon\text{Nd}$  values found in Southern  
188 Ocean records<sup>40</sup>. Previous studies have suggested that prior to deepening of the Greenland Scotland  
189 Ridge (GSR) NCW was sourced from the Labrador Sea<sup>10,13</sup>. Modern Labrador Sea deep water, however,  
190 has characteristically low  $\epsilon\text{Nd}$  ( $\sim -14$ ), reflecting erosional inputs from the cratonic hinterland<sup>41</sup>. In  
191 contrast, Site 647 Nd is significantly more radiogenic ( $\epsilon\text{Nd} = -11.4$  to  $-9.4$ ), and a closer match to the  
192 range of values measured in Nordic sea overflows ( $\epsilon\text{Nd} = \sim -12.0$  to  $-8.4$ )<sup>41,42</sup> and proximal Arctic Ocean  
193 basins above 500 m ( $\epsilon\text{Nd} = -11.7$  to  $-8.8$ )<sup>43</sup>. A Palaeogene presence of Tethys-sourced deep water at Site  
194 647 is another possibility, since the Tethyan  $\epsilon\text{Nd}$  signature ( $\epsilon\text{Nd} = -10.0$  to  $-9.3$ <sup>44,45</sup>) is  
195 indistinguishable from that of North Atlantic water masses. However, palaeogeographic reconstructions  
196 suggest that water mass exchange between European Tethys and Nordic Seas was limited during the  
197 middle to late Eocene<sup>25,46</sup> making this unlikely.

198



199 The only connections between the Arctic Ocean and Nordic Seas during the Palaeogene were  
200 shallow<sup>27,46,47</sup>. Transfer of freshened, nutrient-rich waters from the Arctic would have occurred  
201 via a proto Greenland Current. Similarity between the North Sea (Kysing-4, borehole) and Site  
202 647 benthic  $\delta^{18}\text{O}$ , as well as independent evidence for low salinities in the Nordic Seas<sup>34,48</sup>, is  
203 consistent with this picture. Transport from the subarctic seas to the Atlantic Ocean was also  
204 shallow, and sinking of Arctic imprinted NCW, must have taken place south of the GSR until it  
205 subsided. With sufficient cooling in the subarctic seas, the density contrast of brackish Arctic  
206 waters with warmer saltier North Atlantic surface waters permitted sinking, resulting in the  
207 distinct bottom water recorded at Site 647.

208

209 Importantly, before  $\sim 36$  Ma, Arctic imprinted NCW deep water formation was minimal,  
210 implying regular stratification and stagnation in the SLS. This is consistent with the considerable  
211 noise in  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  during Regime 1. How this deep water remained isolated in the SLS at  
212 depths of around 2000 m under Regime 1 remains uncertain. One possibility is that production  
213 and export rates of local deep waters in the North Atlantic were high compared to the influx of  
214 southern-sourced deep waters, and subsequently increased further as the cooler, saltier NCW  
215 started being produced. Alternatively, bathymetric highs associated with the now extinct  
216 Labrador Sea spreading ridges and the West Thulean igneous province to the south, may have  
217 isolated SLS subsurface waters from the overall Atlantic during the early Palaeogene<sup>7</sup>. In this  
218 case, cessation of Labrador Sea spreading close to the EOT was likely important, allowing ridges  
219 to subside and enabling enhanced deep water export.

220

221 We illustrate the isotopic evidence and sequence of EOT oceanic changes using natural  
222 neighbour re-gridding (Methods) of compiled isotopic data to produce south–north Atlantic  
223 depth transects during time windows centered on circulation Regimes 1-3 (Fig. 4, see  
224 Supplementary Information for data sources, additional transects and maps). Before ~36 Ma  
225 (Regime 1), a strong isotopic  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  depletion effects water masses down to ~2000 m  
226 above 40°N (Fig. 4a and b), corresponding to small amounts of low salinity, high nutrient Arctic  
227 imprinted NCW in the SLS. The rest of the Atlantic is filled with deep waters with more  
228 homogenous  $\delta^{18}\text{O}$  sourced from southerly and possibly low latitude regions<sup>10,13</sup>. Increasing  
229 subarctic  $\delta^{18}\text{O}$ , reflecting progressive salinification and densification of Nordic surface waters, is  
230 accompanied by a 0.5 – 1.0 Myr pulse of NCW export during Regime 2 (Figs. 4 c to h). By 33.3-  
231 34.3 Ma the ‘fresh’ SLS deep water signal no longer exists, bottom water  $\delta^{13}\text{C}$  increased, the  
232 acute phase of low  $\delta^{13}\text{C}$  export is over, and a better-ventilated NCW is exported (Regime 3).  
233 Importantly, the initial pulse of NCW export under Regime 2 is recorded by decreasing  $\delta^{13}\text{C}$   
234 signals in deep waters down stream of the SLS. The presence of late Eocene NCW in the  
235 Atlantic has not been identified in previous  $\delta^{13}\text{C}$  records<sup>10,13,20</sup> because NCW was assumed to  
236 have high  $\delta^{13}\text{C}$  signature similar to modern NADW.

237

### 238 **Causes and consequences of Northern Component Water export**

239 Deepening of the GSR in the late Eocene, for which there is geological evidence<sup>4,34</sup>, would have  
240 increased Nordic overflows, thus strengthening NCW production. Modelling suggests sill  
241 deepening to 50 m would initiate a threshold switch from lagoonal to estuarine circulation,  
242 salinifying the Nordic Seas sufficiently to intensify northern deep water production<sup>34</sup>. While this  
243 idea is consistent with our findings, the bathymetric history of the GSR is currently too crude to

244 accurately date such a change. Moreover, we propose that contemporaneous restrictions to the  
245 Arctic-Nordic Sea exchange also played a role. Geological evidence suggests that the Barents  
246 Sea-Arctic passageway shoaled in the latest Eocene<sup>46,47</sup> and that relative sea-level variations in  
247 the Arctic were decoupled from global trends from the late Eocene to early Miocene<sup>49</sup>. This  
248 palaeogeographic Arctic isolation enhanced salinification in the Nordic Seas as brackish Arctic  
249 outflows were gradually cut off.

250

251 Changes in NCW production had varied and competing effects. Its onset presumably impacted  
252 poleward heat transport in both hemispheres<sup>17,18</sup>. Initial export of nutrient rich Arctic imprinted  
253 NCW may have generated a short-lived pulse of CO<sub>2</sub>, on the order of 100-200 ppm, which is  
254 consistent with proxy compilations showing a temporary reversal in the falling CO<sub>2</sub> trend  
255 between ~34 to ~35 Ma<sup>50</sup>. On the other hand, strengthening of NCW production, and enhanced  
256 northward ocean heat transport, could have played a role in longer-term CO<sub>2</sub> drawdown due to  
257 an accompanying increase in rainfall over land and associated CO<sub>2</sub>-weathering feedbacks<sup>16</sup>. The  
258 circulation change timing, 1-2 million years prior to Antarctic glaciation, reinforces the idea that  
259 onset of NCW played a role in preconditioning the late Eocene Earth system for the greenhouse  
260 to icehouse transition.

261

## 262 **Methods**

263 Methods, including statements of data availability and any associated accession codes and  
264 references, are available in the online version of this paper.

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267

268 **References**

- 269 1 Broecker, W. S. Paleocean circulation during the last deglaciation: A bipolar seesaw?  
270 *Paleoceanography* **13**, 119-121 (1998).
- 271 2 de Boer, A. M., Toggweiler, J. R. & Sigman, D. M. Atlantic dominance of the meridional  
272 overturning circulation. *J. of Phys. Oceanog.* **38**, 435-450 (2008).
- 273 3 Toggweiler, R. & Samuels, B. Effect of Drake Passage on the global thermohaline  
274 circulation. *Deep Sea Res., Pt. 1* **42**, 477-500 (1995).
- 275 4 Abelson, M. & Erez, J. The onset of modern-like Atlantic meridional overturning  
276 circulation at the Eocene-Oligocene transition: Evidence, causes, and possible  
277 implications for global cooling. *Geochem., Geophys., Geosystems* **18**, 2177-2199 (2017).
- 278 5 Cramer, B. S., Toggweiler, J. R. T., Wright, J. D., Katz, M. E. & Miller, G. H. Ocean  
279 overturning since the Late Cretaceous: Inferences from a new benthic foraminiferal  
280 isotope compilation. *Paleoceanography* **24**, PA4216 (2009).
- 281 6 Davies, R., Cartwright, J., Pike, J. & Line, C. Early Oligocene initiation of North Atlantic  
282 Deep Water formation. *Nature* **410**, 917-920 (2001).
- 283 7 Egloff, J. & Johnson, G. L. Morphology and structure of the Southern Labrador Sea.  
284 *Canadian J. of Earth Sci.* **12**, 2111-2133 (1975).
- 285 8 Miller, K. G. & Tucholke, B. E. in *Structure and development of the Greenland-Scotland*  
286 *Ridge* (eds M.H.P. Bott, J. Thiede, S. Saxov, & M. Talwani) 549-589 (Plenum Press,  
287 1983).
- 288 9 Via, R. K. & Thomas, D. J. Evolution of Atlantic thermohaline circulation: Early  
289 Oligocene onset of deep-water production in the North Atlantic. *Geology* **34**, 441-444  
290 (2006).

- 291 10 Borrelli, C., Cramer, B. S. & Katz, M. E. Bipolar Atlantic deepwater circulation in the  
292 middle-late Eocene: effects of Southern Ocean gateway openings. *Paleoceanography* **29**,  
293 308-327 (2014).
- 294 11 Boyle, P. R. *et al.* Cenozoic North Atlantic deep circulation history recorded in contourite  
295 drifts, offshore Newfoundland, Canada. *Marine Geology* **385**, 185-203 (2017).
- 296 12 Hohbein, M. W., Sexton, P. F. & Cartwright, J. A. Onset of North Atlantic Deep Water  
297 production coincident with inception of the Cenozoic global cooling trend. *Geology* **40**,  
298 255-258 (2012).
- 299 13 Langton, S. J., Rabideaux, N. M., Borrelli, C. & Katz, M. E. Southeastern Atlantic deep-  
300 water evolution during the late-middle Eocene to earliest Oligocene (Ocean Drilling  
301 Program Site 1263 and Deep Sea Drilling Project Site 366). *Geosphere* **12**, 1032-1047  
302 (2016).
- 303 14 Wright, J. D. & Miller, K. G. Control of North Atlantic Deep Water circulation by the  
304 Greenland-Scotland Ridge. *Paleoceanography* **11**, 157-170 (1996).
- 305 15 Sijp, W. P., England, M. H. & Huber, M. Effect of the deepening of the Tasman Gateway  
306 on the global ocean. *Paleoceanography* **26**, PA4207 (2011).
- 307 16 Elsworth, G., Galbraith, E., Halverson, G. & Yang, S. Enhanced weathering and CO<sub>2</sub>  
308 drawdown caused by latest Eocene strengthening of the Atlantic meridional overturning  
309 circulation. *Nat. Geosci.* **10**, 213-216 (2017).
- 310 17 Tigchelaar, M., von der Heydt, A. S. & Dijkstra, H. A. A new mechanism for the two-  
311 step  $\delta^{18}\text{O}$  signal at the Eocene-Oligocene boundary. *Clim. Past* **7**, 235-247 (2010).
- 312 18 Zhang, Z. *et al.* Tropical seaways played a more important role than high latitude  
313 seaways in Cenozoic cooling. *Clim. Past* **7**, 801-813 (2011).

- 314 19 Huber, M. & Sloan, L. C. Heat transport, deep waters, and thermal gradients: Coupled  
315 simulation of an Eocene greenhouse climate. *Geo. Res. Lett.* **28**, 3481-3484 (2001).
- 316 20 Pusz, A. E., Thunell, R. C. & Miller, K. G. Deep water temperature, carbonate ion, and  
317 ice volume changes across the Eocene-Oligocene climate transition. *Paleoceanography*  
318 **26**, PA2205 (2011).
- 319 21 Firth, J. V., Eldrett, J. S., Harding, I. C., Coxall, H. K. & Wade, B. Integrated  
320 biomagnetostratigraphy for the Palaeogene of ODP Hole 647A: Implications for  
321 correlating palaeoceanographic events from high to low latitudes. *Geol. Soc. Spec. Pub.*  
322 **373**, 29-78 (2013).
- 323 22 Coxall, H. K. & Wilson, P. A. Early Oligocene glaciation and productivity in the eastern  
324 equatorial Pacific; insights into global carbon cycling. *Paleoceanography* **26**, PA2221  
325 (2011).
- 326 23 Kroopnick, P. The distribution of  $^{13}\text{C}$  of  $\Sigma\text{CO}_2$  in the world oceans. *Deep Sea Res.* **32**,  
327 57-84 (1985).
- 328 24 Golovneva, L. B. Early Palaeogene floras of Spitsbergen and North Atlantic floristic  
329 exchange. *Acta Universitatis Carolinae, Geologica* **44**, 39-50 (2000).
- 330 25 Akhmetiev, M. A. & Beniamovski, V. N. Paleogene floral assemblages around  
331 epicontinental seas and straits in Northern Central Eurasia: proxies for climatic and  
332 paleogeographic evolution. *Geologica Acta* **7**, 297-309 (2009).
- 333 26 Gleason, J. D. *et al.* Early to middle Eocene history of the Arctic Ocean from Nd-Sr  
334 isotopes in fossil fish debris, Lomonosov Ridge. *Paleoceanography*. **24**, PA2215 (2009).

- 335 27 O'Regan, A. M., Williams, C. J., Frey, K. E. & Jakobsson, M. A Synthesis of the long-  
336 term paleoclimatic evolution of the Arctic. *Oceanography, The changing Arctic Ocean,*  
337 *Special Issue on the International Polar Year (2007–2009)* **24**, 66-80 (2011).
- 338 28 Pusz, A. E. *et al.* in *Special Paper of the Geol. Soc. of America.* **452**, 83-95 (2009).
- 339 29 Arthur, M. A. *et al.* in *Proceedings of the Ocean Drilling Program, Scientific Results*  
340 Vol. **105** (eds S.P. Srivastava, M.A. Arthur, B. Clement *et al.*) 111-135 (Ocean Drilling  
341 Program, 1989).
- 342 30 Nielsen, S. B. *et al.* The evolution of western Scandinavian topography: A review of  
343 Neogene uplift versus the ICE (isostasy–climate–erosion) hypothesis. *J. Geodynam.* **47**,  
344 72-95 (2009).
- 345 31 Lear, C. H., Bailey, T. R., Pearson, P. N. P., Coxall, H. K. & Rosenthal, Y. Cooling and  
346 ice growth across the Eocene-Oligocene transition. *Geology* **36**, 251-254 (2008).
- 347 32 Liu, L. *et al.* Global cooling during the Eocene-Oligocene climate transition. *Science* **323**,  
348 1187-1190 (2009).
- 349 33 Waddell, L. M. & Moore, T. C. Salinity of the Eocene Arctic Ocean from oxygen isotope  
350 analysis of fish bone carbonate. *Paleoceanography*, PA1S12 (2008).
- 351 34 Stärz, M., Jokat, W., Knorr, G. & Lohmann, G. Threshold in North Atlantic-Arctic Ocean  
352 circulation controlled by the subsidence of the Greenland-Scotland Ridge. *Nat. Comm.* **8**,  
353 15681, doi:10.1038/ncomms15681 (2017).
- 354 35 Roberts, C. D., LeGrande, A. N. & Tripathi, A. K. Climate sensitivity to Arctic seaway  
355 restriction during the early Paleogene. *Earth Planet. Sci. Lett.* **286**, 576-585 (2009).
- 356 36 Brinkhuis, H. *et al.* Episodic fresh surface waters in the Eocene Arctic Ocean. *Nature*  
357 **441**, 606-609 (2006).

- 358 37 Kaminski, M. & Ortiz, S. The Eocene-Oligocene turnover of deep-water agglutinated  
359 foraminifera at ODP Site 647, Southern Labrador Sea (North Atlantic).  
360 *Micropaleontology* **60**, 53-66 (2014).
- 361 38 Burton, K. W., Ling, H. F. & O'Nions, R. K. Closure of the Central American isthmus  
362 and its effect on deep-water formation in the North Atlantic. *Nature* **386**, 382-385 (1997).
- 363 39 O'Nions, R. K., Frank, M., von Blanckenburg, F. & Ling, H. F. Secular variation of Nd  
364 and Pb isotopes in ferromanganese crusts from the Atlantic, Indian and Pacific Oceans.  
365 *Earth Planet. Lett.* **155**, 15-28 (1998).
- 366 40 Scher, H. D. & Martin, E. E. Timing and climatic consequences of the opening of Drake  
367 Passage. *Science* **312**, 428-430 (2006).
- 368 41 Lambelet, M. *et al.* Neodymium isotopic composition and concentration in the western  
369 North Atlantic Ocean: Results from the GEOTRACES GA02 section. *Geochem.*  
370 *Cosmochim. Acta* **177**, 1-29 (2016).
- 371 42 Lacan, F. & Jeandel, C. Acquisition of the neodymium isotopic composition of the North  
372 Atlantic Deep Water. *Geochem., Geophys., Geosystems.* **6** (2005).
- 373 43 Porcelli, D. *et al.* The distribution of neodymium isotopes in Arctic Ocean basins.  
374 *Geochim. Cosmochim. Acta* **73**, 2645-2659 (2009).
- 375 44 Grandjean, P., Cappetta, H., Michard, A. & Albarede, F. The assessment of REE patterns  
376 and  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios in fish remains. *Earth Planet. Sci. Lett.* **84**, 181-196 (1987).
- 377 45 Stille, P. & Fischer, H. Secular variation in the isotopic composition of Nd in Tethys  
378 seawater. *Geochim. Cosmochim. Acta* **54**, 3139-3145 (1990).
- 379 46 Kharin, G. S. & Lukashina, N. P. Paleogeography of the Norwegian-Greenland and  
380 northwestern European Sea basins in the Paleogene. *Oceanology* **50**, 226-239 (2010).



- 381 47 Musatov, E. E. & Pogrebetskij, Y. E. Late Mesozoic-Cenozoic evolution of the Barents  
382 Sea and Kara Sea continental margins. *Polarforschung* **68**, 283-290 (2000).
- 383 48 Andreasson, F. P., Schmitz, B. & Spiegler, D. in *Proceedings of the Ocean Drilling*  
384 *Program, Scientific Results*. Vol. **151** (eds J. Thiede *et al.*) 583-591 (Ocean Drilling  
385 Program, 1996).
- 386 49 Hegewald, A. & Jokat, W. Relative sea level variations in the Chukchi region - Arctic  
387 Ocean - since the late Eocene. *Geophys. Res. Lett.* **40**, 2013 (2013).
- 388 50 Anagnostou, E. *et al.* Changing atmospheric CO<sub>2</sub> concentration was the primary driver of  
389 early Cenozoic climate. *Nature* **533**, 380-384 (2016).

390

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403 **Author Contributions** HKC and JB conceived the project. HKC directed the research,  
404 generated the stable isotope data for Sites 112, 647 and U1411, compiled the proxy records and  
405 led writing of the paper. AL produced the new Site 612 data and age model. CHL conducted the  
406 trace metal analysis. CH produced and interpreted the Nd data with the help of TF. MO produced  
407 the palaeogeographic map for Figure 1 and conducted the subsidence modeling. KS helped  
408 produce the Site 647 age model. MH helped with the interpretative framework and produced the  
409 interpolated Atlantic depth isotopic transects and maps. JZ and AdB helped interpret the data. All  
410 authors contributed to writing the manuscript.

411

#### 412 **Additional information**

413 Supplementary information is available in the online version of this paper. The data can be  
414 obtained from the ‘Bolin Centre for Climate Research’ database: [http://bolin.su.se/data/Coxall-](http://bolin.su.se/data/Coxall-2018)  
415 [2018](http://bolin.su.se/data/Coxall-2018). Reprints and permissions information is available online at [www.nature.com/reprints](http://www.nature.com/reprints).

416

#### 417 **Competing financial interests**

418 The authors declare no competing financial interests.

419

#### 420 **Figure captions**

421

422 **Figure 1. Site locations of sections included in this study.** Stars identify the new data sets  
423 presented here. Map annotations: Red line = Mid Ocean Ridge 34 Ma; white line = position of  
424 56 Ma isochron; black line = continent-ocean crust boundary. See Methods Section for details of  
425 the palaeogeographic framework and inset map. LS = Labrador Sea, NGS = Norwegian

426 Greenland Sea; T = Tethys Ocean; GSR = Greenland Scotland Ridge. Inset panel shows the path  
427 of major deep (blue) and surface currents today: Denmark Strait Overflow Water (DSOW);  
428 Iceland-Scotland Overflow Water (ISOW); Labrador Sea Water (LSW); North Atlantic Current  
429 (NAC) and the Deep Western Boundary Current (DWBC).

430

431 **Figures 2. New and published Atlantic benthic  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  (*Cibs.* adjusted).** The EOT  
432 (fine dashed horizontal lines) and the EOGM event are identified by the step-increase and  
433 maximum in  $\delta^{18}\text{O}$  in the earliest Oligocene respectively. Trend lines represent smoothed curve  
434 fits that incorporate a geometric weighting. Regimes 1-3, separated by blue long-dash lines, are  
435 phases of ocean circulation defined here based on proxy data. Pale aqua shading represents  
436 transition phases. Vertical black arrows identify time windows gridded in Fig. 4. See  
437 Supplementary Information for data sources, age modeling and an expanded figure with  
438 additional data.

439

440 **Figure 3. Sites 647 and U1411 multiproxy data.** a) Mg/Ca BWT; paler blue symbols =  
441 maximum BWTs due to potential Fe contamination (Supplementary information). b), d): planktic  
442 and benthic  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  (black symbols=equatorial Pacific EOT chemostratigraphic  
443 reference<sup>22</sup>, open symbols =U1411). c) Estimated Site 647 bottom salinity based on modern  
444  $\delta^{18}\text{O}_{\text{sw}}$ - relationships; (LS) Laptev Sea, (EG) Eastern Greenland. Error envelopes are based on  
445 2STD of the Mg/Ca BWT. e) =Site 647 planktic-benthic  $\delta^{13}\text{C}$  difference and modern gradients<sup>23</sup>.  
446 f) Site 647 agglutinated benthic foraminifera<sup>37</sup>. g) Site 647 fish tooth Nd and EOT ocean  
447 signatures, including Artic ranges (error bars =  $2\sigma$  standard reproducibility). Annotations as in  
448 Fig. 2.

449  
450 **Figure 4. Depth-latitude compilation of Atlantic benthic  $\delta^{13}\text{C}$  and  $\delta^{18}\text{O}$  during the late**  
451 **Eocene to early Oligocene constructed using natural neighbor interpolation.** Data are plotted  
452 at their 34 Ma positions (dots indicate core palaeopositions). The five time slices illustrate the  
453 transition through the three circulation regimes identified here (R1, R2 and R3; see Fig. 2). See  
454 Supplementary information for data sources and a more extensive set of gridded time slices and  
455 late Eocene maps.

## 456 457 **METHODS**

458  
459 **Palaeogeographic plate reconstructions and modern Atlantic Ocean circulation inset**  
460 Palaeogeographic plate reconstructions used in production of main text Figure 1 were performed  
461 using G-plates, with coastlines adapted from E-O reconstructions (34 Ma) of Ron Blakey,  
462 Colorado Plateau Geosystems, Arizona USA. The inset figure showing modern North Atlantic  
463 surface and deep current paths is based on the schematics of ref-<sup>51</sup>.

### 464 465 **Age framework**

466 Site 647 age control is based on biomagnetostratigraphy<sup>21</sup>, adjusted here using Site 647  $\delta^{18}\text{O}$   
467 chemostratigraphy (See Supplementary information for further details for Site 647 and the other  
468 sites). Site 112 ages are estimated from biostratigraphy<sup>52</sup>. Site U1411 ages are based on IODP  
469 Exp. 342 shipboard magnetostratigraphy<sup>53</sup>. The Site 612 age model is based on the  
470 biostratigraphy of ref-<sup>54</sup>. In all cases, datum events are calibrated or rescaled using linear  
471 interpolation to the chronology of ref-<sup>55</sup> to permit comparison with the Atlantic benthic isotope  
472 stack (after refs-<sup>5,56,57</sup>), much of which exists on this common time scale.

### 473 474 **Stable isotopes**

475 Planktic and benthic foraminifera are present throughout the EOT interval of Site 647, 112 and  
476 U1411, although heavily diluted by terrestrial clay. Planktic and benthic foraminifera are  
477 somewhat more common at Site 612. Tests are exceptionally well preserved at all sites  
478 throughout the studied intervals (see Supplementary Information for further details).

479  
480 Foraminiferal  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  for Sites 647 and 112, was derived from the benthic foraminifera  
481 taxa *Oridorsalis umbonatus* (shallow infaunal) and *Cibicidoides* spp. (epifaunal), where  
482 available, both shown to be a reliable deep-sea tracer in previous studies<sup>22,58</sup> (see Supplementary  
483 Information). Site U1411 measurements are on *Cibicidoides* spp. and the new Site 612  
484 measurements on *Hanzawaia ammophila*. Sites 647 and U1411 planktic foraminiferal analyses  
485 were made on *Turborotalia ampliapertura* and *Catapsydrax unicavus*, representing surface  
486 mixed layer and thermocline/subthermocline habitats respectively<sup>59</sup>. Site 647 stable isotope  
487 analysis was performed at Cardiff University using a ThermoFinnigan MAT252 mass

488 spectrometer equipped with an automated KIEL III carbonate preparation unit. Additional  
489 samples were run at the National Oceanographic Centre, Southampton University, using a  
490 Europa Geo 20–20 mass spectrometer equipped with a CAPS automatic carbonate preparation  
491 system. Standard external analytical precision quoted at Cardiff was better than 0.05‰ for  $\delta^{18}\text{O}$   
492 and 0.03‰ for  $\delta^{13}\text{C}$ , and  $\pm 0.08\text{‰}$  for  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$  at Southampton. Site 612 analyses were  
493 measured at the Department of Geological Sciences, Stockholm University on a ThermoFinnigan  
494 MAT 252 IRMS coupled with a Finnigan Gasbench II device. Standard external analytical  
495 precision, based on replicate analysis of in-house standards calibrated to international standards  
496 (NBS19, IAEA-CO-1 and IAEA-CO-8), was better than 0.07‰ for  $\delta^{13}\text{C}$  and 0.15‰ for  $\delta^{18}\text{O}$ . All  
497 results are reported relative to the VPDB standard. Our Site 647 *O. umbonatus* data have been  
498 adjusted to *Cibicidoides* values (believed to be close to ambient seawater) by subtracting -0.28‰  
499 for the  $\delta^{18}\text{O}$  following ref-<sup>60</sup>, and by addition of 1.4‰ to the  $\delta^{13}\text{C}$ , following ref-<sup>61</sup> (consistent  
500 with a species comparison study in a restricted basin in the western North Pacific which closely  
501 matches our few Site 647 *Cibicidoides-Oridorsalis umbontatus*  $\delta^{13}\text{C}$  pairs). The different species  
502 are differentiated in Figs. 2 and 3 by dark red (*O. umbontatus*) and bright red (*Cibicidoides* spp.)  
503 symbols. For Site 612 the following adjustments (after ref-<sup>60</sup>) were used when integrating the  
504 new *H. ammophila* data:  $(\delta^{18}\text{O}_{H. ammophila} - 0.16)/0.62 = \delta^{18}\text{O}_{Cibicidoides}$ , and  $\delta^{13}\text{C}_{H. ammophila} + 0.08 =$   
505  $\delta^{13}\text{C}_{Cibicidoides}$ . The planktic-benthic  $\delta^{13}\text{C}$  gradient ( $\Delta\delta^{13}\text{C}$ ) was generated by resampling the  
506 planktic and benthic foraminifera  $\delta^{13}\text{C}$  curves to provide paired samples. Our new  $\delta^{18}\text{O}$  and  $\delta^{13}\text{C}$   
507 are compared with 21 other Atlantic data sets that build on the compilations of refs-<sup>5,57</sup>. The new  
508 data produced in this study are presented in Supplementary Data file S1). See Supplementary  
509 Table S1 for the full list of sites meta data and sources used in our Atlantic compilation.  
510

#### 511 **Trace metal analysis and Mg/Ca foraminiferal bottom water palaeothermometry**

512 Trace metal content (Mg/Ca, Mn/Ca, Fe/Ca) was analyzed on Site 647 *O. umbonatus*. Prior to  
513 analysis, benthic foraminifera samples were cleaned following the protocol of refs-<sup>62,63</sup> leaving  
514 out the reducing step due to the scarcity of material but including contaminant removal under  
515 binocular microscope following the oxidative step<sup>64</sup>. Samples were subjected to one weak acid  
516 leach prior to dissolution and dilution. Analysis was carried out at Cardiff University on a  
517 Thermo Element XR ICP-MS against standards with equivalent Ca concentration. Multi-element  
518 standards were made in-house from single element standards supplied by Greyhound  
519 Chromatography and Allied Chemicals. Analytical precisions determined from separate  
520 consistency standards over the course of a year are 0.5% for Mg/Ca, and 2% for Mn/Ca and  
521 Fe/Ca (rsd). Mg/Ca paleo- bottom water temperatures (BWT) were calculated using the  
522 exponential calibration of ref-<sup>58</sup> (See Supplementary Information and Supplementary Data file  
523 S2).  
524

525 Our *O. umbonatus* Mg/Ca record is noisy and high Mg/Ca ratios are often associated with high  
526 Fe/Ca (correlation coefficient  $r^2 = 0.4$ ) (Supplementary Data file S2). By excluding the samples  
527 with  $\text{Fe/Ca} \gg 900 \mu\text{mol/mol}$ ,  $r^2$  was reduced to 0.18. The subset of data, with lower Fe/Ca,  
528 largely the upper portion of the core in samples younger than 34.5 Ma (Supplementary Table  
529 S4), may be regarded as most reliable. However, despite the higher Fe/Ca in the older samples,  
530 we believe the Mg/Ca data from the lower part of the core are not entirely flawed since the Fe/Ca  
531 vs Mg/Ca  $r^2$  value based on the full sample set is still relatively low and there is Mg/Ca overlap  
532 of higher and lower Fe/Ca Mg/Ca around the EOT (Main Figure 3A, paler blue symbols). Thus,  
533 the older Mg/Ca should provide realistic palaeo-bottom water temperatures (within the

534 uncertainties of the method), and are thus included in the down core record to provide ballpark  
535 BWTs and allow salinity reconstructions in the initial part of the late Eocene.

536

### 537 **Bottom Water Salinity reconstruction**

538 The pattern of progressive benthic  $\delta^{18}\text{O}$  increase and maximum 4°C BWT cooling between 37.5-  
539 35.5 Ma implies that sea water  $\delta^{18}\text{O}$  ( $\delta^{18}\text{O}_{\text{sw}}$ ) was changing over this period. To explore this  
540 further bottom water palaeosalinity was reconstructed based on modern sea surface salinity  
541 (SSS) -  $\delta^{18}\text{O}$  relationships. This was performed in two steps. First,  $\delta^{18}\text{O}_{\text{sw}}$  values were calculated  
542 by substituting the Site 647 Mg/Ca BWTs into the  $\delta^{18}\text{O}$ -benthic foraminifera palaeotemperature  
543 equation of ref-<sup>65</sup>. Due to the noise in our estimated benthic bottom water temperatures we used  
544 broad ‘BWT brackets’, based on mean BWT values for three intervals (Supplementary Table  
545 S5).

546

547 Second, bottom water palaeosalinity was reconstructed based on the modern surface salinity  $\delta^{18}\text{O}$   
548 relationships for the Laptev Sea and East Greenland Current<sup>66,67</sup>, regions with relatively low  
549  $\delta^{18}\text{O}_{\text{sw}}$  linked to the Arctic Ocean or Arctic outflows respectively (the Laptev Sea is an Arctic  
550 shelf sea that receives large volumes of river run-off from Siberian rivers (22-34 PSU), while the  
551 East Greenland Current carries low salinity surface waters (32 PSU) out of the Arctic Ocean). It  
552 was assumed that these relationships remained the same down core.

553

554 Laptev Sea:            Salinity= ( $\delta^{18}\text{O}_{\text{sw}} - 18.86$ ) = 0.5             $R^2 = 0.98$  ref-<sup>66</sup>  
555 East Greenland:       Salinity= ( $\delta^{18}\text{O}_{\text{sw}} - 35.02$ ) = 1.01            ref-<sup>67</sup>

556

557 The resulting curve, which is plotted as a ‘smoothed curve-fit’ in KaleidaGraph® with error  
558 envelopes representing limits determined by  $2\sigma$  of the BWT brackets (Main Fig. 3C), provides  
559 coarse constraints on the evolution of Site 647 bottom water salinity in the late Eocene. As  
560 discussed above, its possible that the decreasing Mg/Ca between 37.5-35.5 Ma represents  
561 decreasing trace metal contamination rather than BW cooling. If this is the case then the BWTs  
562 around 37 Ma are too high, which would bias the salinities to too salty values. Thus, including or  
563 excluding the older Mg/Ca data does not change the conclusions. We have confidence in the  
564 Mg/Ca after 34 Ma and thus have  $\delta^{18}\text{O}_{\text{sw}}$  constrained there. The important point is that the  
565 subsurface densification signal is seen in the benthic foraminifera  $\delta^{18}\text{O}$  record (increasing  $\delta^{18}\text{O}$   
566 from 37.5-35.5 Ma). It is impossible that this is an artifact of the Mg/Ca data.

567

### 568 **Neodymium isotope methodology**

569 Fish teeth and bone debris were hand-picked from the  $>63\mu\text{m}$  fraction of sieved sediment and  
570 cleaned to remove adhering debris. Initial experiments (see Supplementary information)  
571 indicated that a the ‘simple cleaning method’ of ref-<sup>68</sup> was sufficient. All samples were dissolved  
572 in 2M HCl, dried and converted to nitrate form prior to column chemistry. A standard two-stage  
573 ion chromatography procedure was used, which first isolated the REEs from the sample matrix  
574 using TRU Spec resin (100-120 $\mu\text{m}$  bead size) and then separated Nd from the other REE’s with  
575 Ln-Spec resin (50-100 $\mu\text{m}$  bead size) (after ref-<sup>69</sup>). Neodymium isotope ratios were measured on  
576 a Nu Plasma MC-ICP-MS at Imperial College London in static mode. Instrumental mass bias  
577 was corrected for using a  $^{146}\text{Nd}/^{144}\text{Nd}$  ratio of 0.7219. Samarium interference can be adequately  
578 corrected if the  $^{144}\text{Sm}$  signal contributes less than 0.1% of the  $^{144}\text{Nd}$  signal. The Sm contribution  
579 in all our samples was well below this level. Chemistry blanks were consistently below 10pg

580 Nd. Replicate analyses of the Nd standard JNdi yielded  $^{143}\text{Nd}/^{144}\text{Nd}$  ratios from  $0.512060 \pm$   
581  $0.000015$  to  $0.512251 \pm 0.000015$  ( $n=116$ ), dependent on daily running conditions over 12  
582 months. The external reproducibility of our chemistry and mass spectrometry procedure was  
583 monitored using a fossil bone composite standard supplied by Clive Trueman (University of  
584 Southampton) yielding a  $^{143}\text{Nd}/^{144}\text{Nd}$  ratio of  $0.512377 \pm 0.000028$  ( $n=8$  over 18 months), which  
585 agrees within error with previously published values<sup>70,71</sup>. We note that the standard material was  
586 digested for analysis following the methods in ref<sup>70</sup> for analytical consistency. Briefly, 50 mg of  
587 material was digested in 3M HNO<sub>3</sub> in a Teflon beaker at 130°C. Any material remaining after  
588 this step was subjected to a further 48 hours digestion in a 3:1 mixture of 15M HNO<sub>3</sub> to 27M HF.  
589 To correct for the decay of  $^{147}\text{Sm}$  to  $^{143}\text{Nd}$  within the fish teeth over time we use rare earth  
590 element concentrations obtained in two samples from this site. Derived  $^{147}\text{Sm}/^{143}\text{Nd}$  ratios of  
591 0.129 and 0.134 are consistent with values reported in other studies<sup>72-74</sup>. We applied an average  
592 value of our measurements ( $^{147}\text{Sm}/^{143}\text{Nd} = 0.132$ ) to all remaining samples. All Nd isotope ratios  
593 are reported in epsilon notation ( $\epsilon_{\text{Nd}}$ ) (Supplementary Table 6). Comparative  $\epsilon_{\text{Nd}}$  data in Figure 3  
594 are listed in Supplementary Table S7.

595

### 596 **Atlantic isotopic depth sections**

597 We used standard techniques in ocean data assimilation to create homogenized and interpolated  
598 maps of isotopic values based on the sparse data (see Supplementary Table S1 for site  
599 information, data sources and a more complete set of maps; SI Figs 9A-L). For the transects the  
600 proxy data were placed on to a regular latitude-depth grid based on their reconstructed depths  
601 and palaeo-locations, and the natural neighbor regridding routines implemented in the NCAR  
602 Command Language were used to create interpolated, gridded values  
603 (<https://www.ncl.ucar.edu/Document/Functions/Built-in/natgrid.shtml>). Extrapolation beyond the  
604 convex hull of the kernel was not used. The background palaeogeography is based on Ref-<sup>75</sup>  
605 (hotspot reference frame). The differences in interpretation for our paper using the alternative  
606 Palaeomag reference frame described in the same study are negligible.

607

608 For the maps, the proxy records interpolated using the technique of Barnes-Cressman iterated  
609 objective analysis implemented in the NCAR Command Language  
610 ([www.ncl.ucar.edu/Document/](http://www.ncl.ucar.edu/Document/Functions/Contributed/obj_anal_ic_Wrap.shtml) Functions/Contributed/obj\_anal\_ic\_Wrap.shtml). In this  
611 approach, each observation is assigned a circular radius of influence, R. Here, we used  
612 successive values of R of 9°, 8°, 6°, 4°, 2°, 1°. A first guess of the value at every grid point was  
613 made by including all observations within the region of influence of that grid point. A distance-  
614 weighted average of the differences between the first-guess fields and the actual observations  
615 was made, and this anomaly was added into the first-guess fields to calculate a second-guess  
616 field. Thus, observations from beyond the radius of influence were ignored in updating the field,  
617 and other observations closer to the initial observation were given greater weight. This was done  
618 for all grid points under consideration. The resulting fields were used as the basis for the next  
619 iteration, which was carried out with a smaller region of influence.

620

### 621 **Data availability statement**

622 The authors declare that lists of the sources of previously published data used in this study are  
623 available within the article and its Supplementary Information files. The new data are available  
624 at the Bolin Centre for Climate Research Database: <http://bolin.su.se/data/Coxall-2018>.

625

626 **References**

- 627
- 628
- 629 51 Schmitz, W. J. & McCartney, M. S. On the North Atlantic Circulation. *Rev. of Geophys.*  
630 **31**, 29-49 (1993).
- 631 52 Laughton, A. S. *et al.* Site 112, in *Initial Reports of the Deep Sea Drilling Project*, Vol.  
632 **12** (eds A.S. Laughton, W. A. Berggren *et al.*) 161-253 (US Government Printing Office,  
633 1972).
- 634 53 Norris, R. D., Wilson, P. A., Blum, P. & the Expedition Scientists, *Proceedings of the*  
635 *Integrated Ocean Drilling Program*, **342**, doi:10.2204/iodp.proc.342.2014 (Integrated  
636 Ocean Drilling Program, 2014).
- 637 54 Miller, K. G. & Katz, M. E. in *Initial Reports of the Deep Sea Drilling Project*, Vol. **95**  
638 (eds C. W. Poag, A. B. Watts *et al.*) 267-298 (U.S. Government Printing Office, 1987).
- 639 55 Cande, S. C. & Kent, D. V. Revised calibration of the geomagnetic polarity timescale for  
640 the Late Cretaceous and Cenozoic. *J. Geophys. Res.* **100**, 6093-6095 (1995).
- 641 56 Zachos, J. C., Pagani, M., Sloan, L. C., Thomas, E. & Billups, K. Trends, rhythms, and  
642 aberrations in global climate 65 Ma to present. *Science* **292**, 686-693 (2001).
- 643 57 Zachos, J. C., Dickens, G. R. & Zeebe, R. E. An early Cenozoic perspective on  
644 greenhouse warming and carbon-cycle dynamics. *Nature* **451**, 279-283 (2008).
- 645 58 Lear, C. H. *et al.* Neogene ice volume and ocean temperatures: Insights from infaunal  
646 foraminiferal Mg/Ca paleothermometry. *Paleoceanography* **30**, 1437-1454 (2015).
- 647 59 Pearson, P. N. *et al.* Warm tropical sea surface temperatures in the Late Cretaceous and  
648 Eocene epochs. *Nature* **413**, 481-487 (2001).
- 649 60 Katz, M. E. *et al.* Early Cenozoic benthic foraminiferal isotopes: species reliability and  
650 interspecies correction factors. *Paleoceanography* **18**, 1024 (2003).
- 651 61 Rathburn, A. E., Corliss, B. H., Tappa, K. D. & Lohmann, K. C. Comparisons of the  
652 ecology and stable isotopic compositions of living (stained) benthic foraminifera from the  
653 Sulu and South China Seas. *Deep Sea Res. Pt 1. Oceanographic research papers* **43**,  
654 1617-1646 (1996).
- 655 62 Boyle, E. A. Manganese carbonate overgrowths on foraminifera tests. *Geochim.*  
656 *Cosmochim. Acta* **47**, 1815-1819 (1983).
- 657 63 Boyle, E. A. & Keigwin, L. D. Comparison of Atlantic and Pacific paleochemical records  
658 for the last 215,000 years: changes in deep ocean circulation and chemical inventories.  
659 *Earth Planet. Sci. Lett.* **76**, 135-150 (1985).
- 660 64 Barker, S., Greaves, M. & Elderfield, H. A study of cleaning procedures used for  
661 foraminiferal Mg/Ca paleothermometry. *Geochem., Geophys., Geosystems* **4**,  
662 doi:10.1029/2003GC000559 (2003).
- 663 65 Marchitto, T. M. *et al.* Improved oxygen isotope temperature calibrations for  
664 cosmopolitan benthic foraminifera. *Geochim. Cosmochim. Acta* **130**, 1-11 (2014).
- 665 66 Mueller-Lupp, T., Erlenkueser, H. & Bauch, H. A. Seasonal and interannual variability of  
666 Siberian river discharge in the Laptev Sea inferred from stable isotopes in modern  
667 bivalves. *Boreas* **32**, 292-303 (2003).
- 668 67 Fairbanks, R. G., Charles, C. D. & Wright, J. D. in *Radiocarbon After Four Decades* (ed  
669 R. E. Taylor) 473-500 (Springer, 1992).
- 670 68 Martin, E. E. & Haley, B. A. Fossil fish teeth as proxies for seawater Sr and Nd isotopes.  
671 *Geochim. Cosmochim. Acta* **64**, 835-847 (2000).



672 69 Pin, C. & Zalduegui, J. S. Sequential separation of light rare-earth elements, thorium and  
673 uranium by miniaturized extraction chromatography: application to isotopic analyses of  
674 silicate rocks. *Analyt. Chimica Acta* **339**, 79-89 (1997).

675 70 Chavagnac, V. *et al.* Towards the development of a fossil bone geochemical standard: An  
676 inter-laboratory study. *Analyt. Chimica Acta* **599**, 177-190 (2007).

677 71 Scher, H. D. & Delaney, M. L. Breaking the glass ceiling for high resolution Nd isotope  
678 records in early Cenozoic paleoceanography. *Chem. Geol.* **269**, 329-338 (2010).

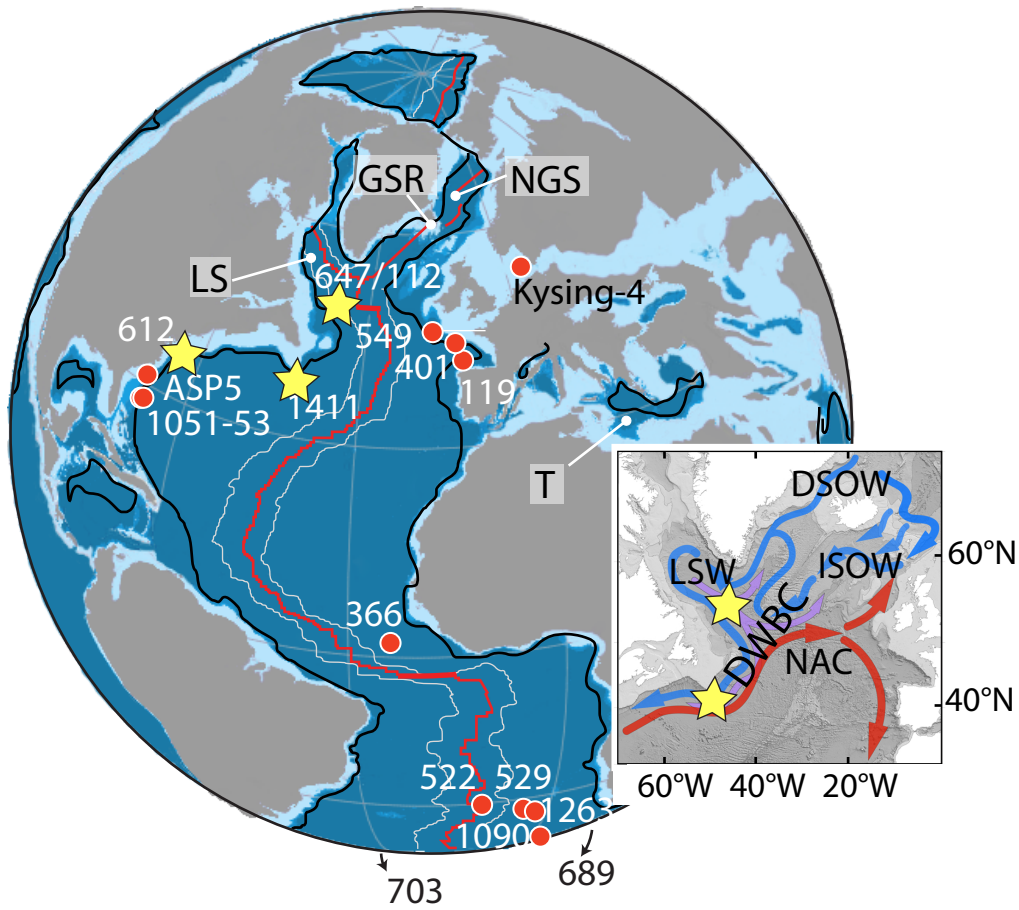
679 72 Thomas, D. J., Bralower, T. J. & Jones, C. E. Neodymium isotopic reconstruction of late  
680 Paleocene-early Eocene thermohaline circulation. *Earth Planet. Sci. Lett.* **209**, 309-322  
681 (2003).

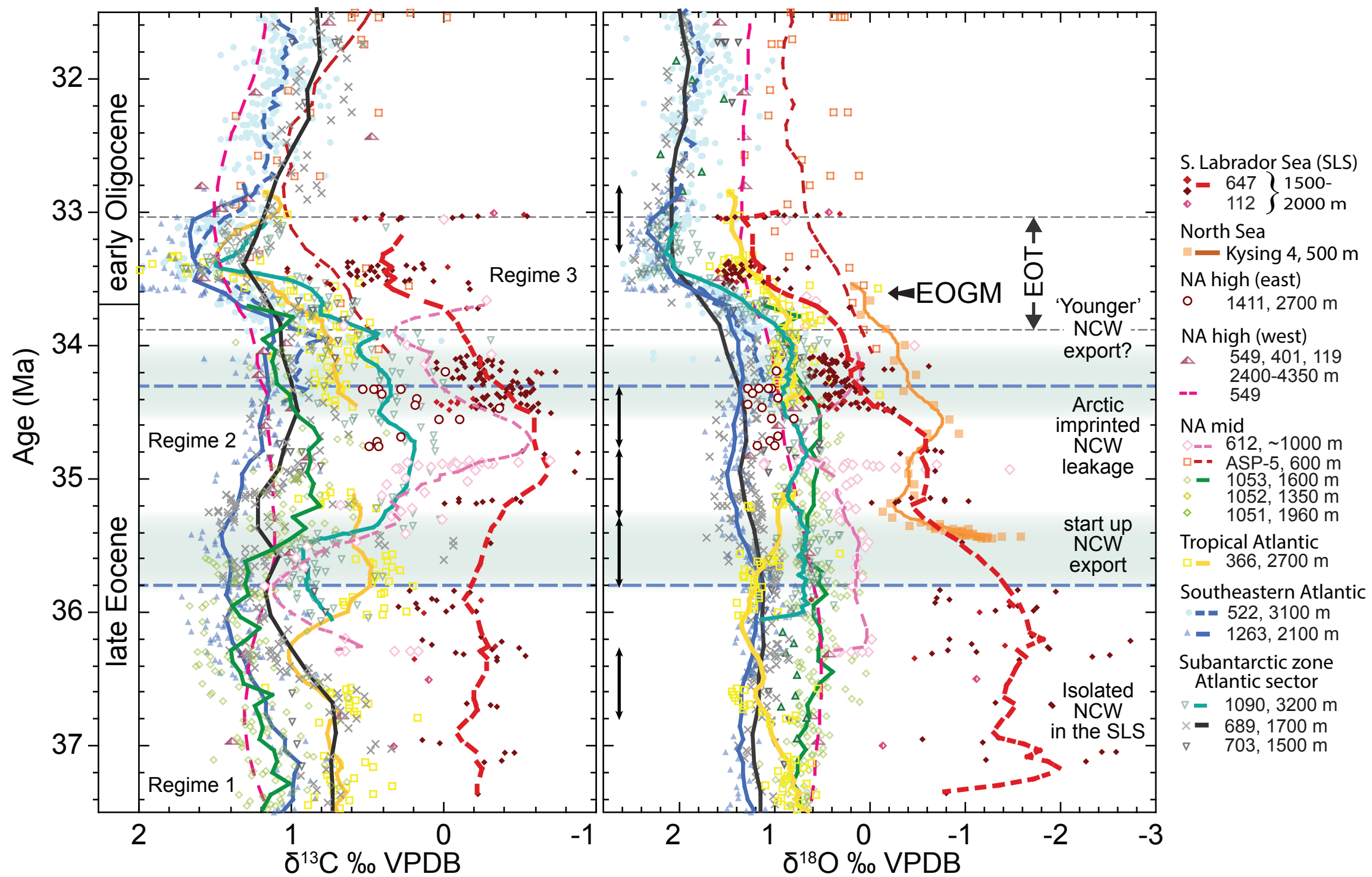
682 73 Martin, E. E. & Scher, H. A Nd isotopic study of southern sourced waters and Indonesian  
683 throughflow at intermediate depths in the Cenozoic Indian Ocean. *Geochem. Geophys.*  
684 *Geosystems* **7**, Q09N02 (2006).

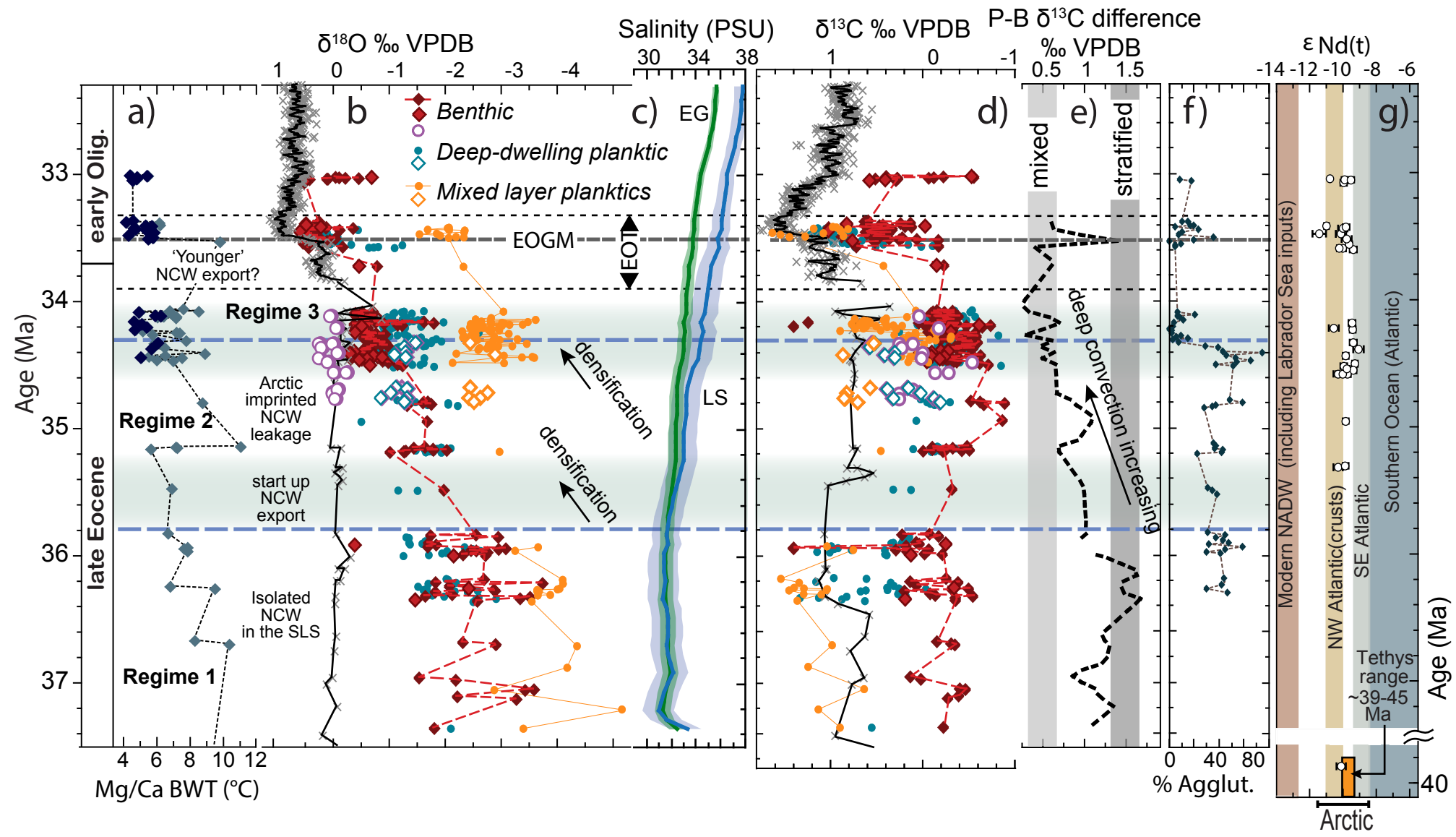
685 74 Moiroud, M. *et al.* Evolution of the neodymium isotopic signature of neritic seawater on  
686 a northwestern Pacific margin: new constrains on possible end-members for the  
687 composition of deep-water masses in the Late Cretaceous ocean. *Chem. Geol.* **356**, 160-  
688 170 (2013).

689 75 Baatsen, M. *et al.* A generalised approach to reconstructing geographical boundary  
690 conditions for palaeoclimate modelling. *Clim. Past.* **12**, 4917-4942 (2016).

691

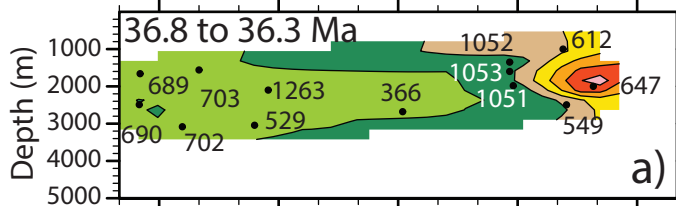




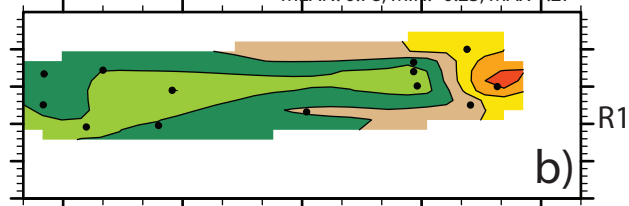


$\delta^{18}\text{O}$ 

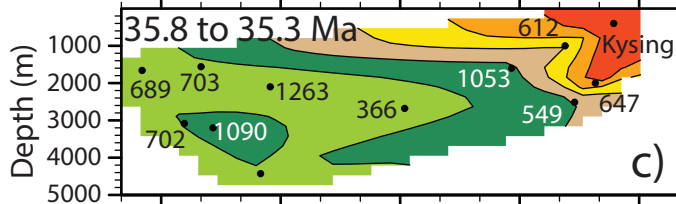
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 $\delta^{13}\text{C}$ 

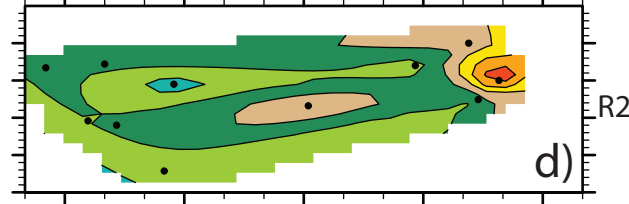
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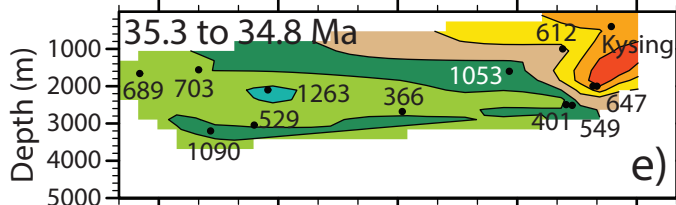
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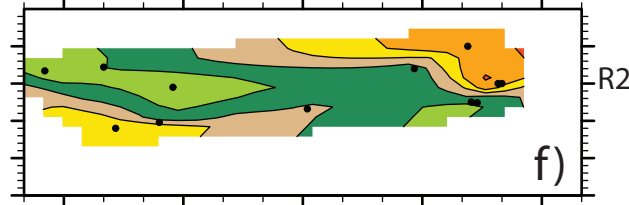
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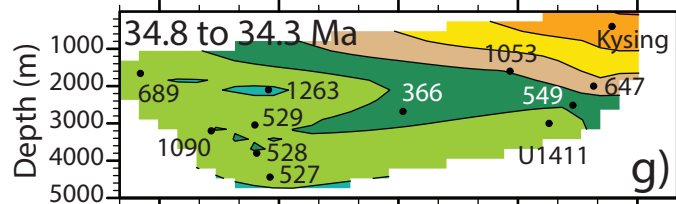
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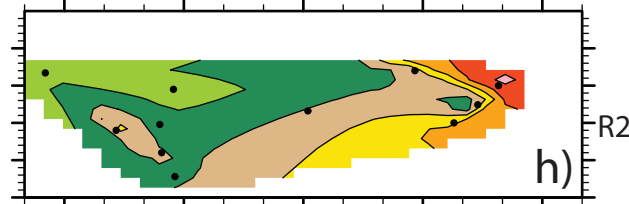
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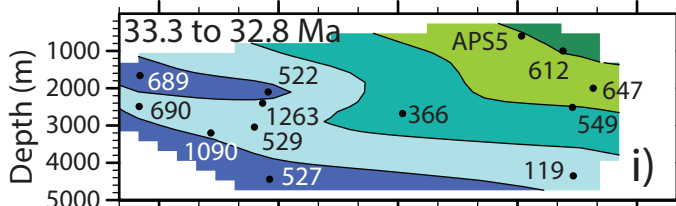
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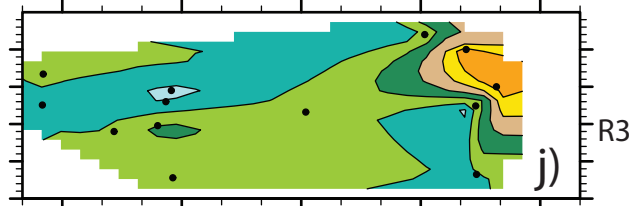
MEAN: 0.64; MIN: -0.37; MAX 1.17



MEAN: 1.64; MIN: 0.88; MAX 2.3



MEAN: 0.99; MIN: 0.01; MAX 1.58



60S 30S 0 30N 60N



60S 30S 0 30N 60N

