### 1 Incursions of southern-sourced water into the deep North Atlantic

# 2 during Late Pliocene glacial intensification

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34 35 Marcus Gutjahr 36 National Oceanography Centre Southampton, University of Southampton, Waterfront 37 Campus, Southampton SO14 3ZH, UK. 38 Now at: GEOMAR Helmholtz Centre for Ocean Research Kiel, Wischhofstraße 1–3, 39 24148 Kiel, Germany. 40 41 The circulation and internal structure of the oceans exert a strong influence on 42 Earth's climate because they control latitudinal heat transport and the segregation of carbon between the atmosphere and the abyss<sup>1</sup>. Circulation 43 change, particularly in the Atlantic Ocean, is widely suggested<sup>2-5</sup> to have been 44 instrumental in the intensification of northern hemisphere glaciation when large 45 46 ice-sheets first developed on North America and Eurasia during the Late Pliocene, ~2.7 million years ago<sup>6</sup>. Yet the mechanistic link and cause/effect 47 48 relationship between ocean circulation and glaciation are debated. Here we 49 present new records of North Atlantic Ocean structure using the carbon and 50 neodymium isotopic composition of deep waters for both the Last Glacial to 51 Holocene (35-5 thousand years ago) and the Late Pliocene to earliest Pleistocene 52 (3.3 to 2.4 million years ago). Our data show no secular change. Instead we 53 document major southern-sourced water incursions into the deep North Atlantic 54 during prominent glacials from 2.7 million years ago. Our results suggest that 55 Atlantic circulation acts as a positive feedback rather than as an underlying 56 cause of Late Pliocene northern hemisphere glaciation. We propose that, once surface Southern Ocean stratification and/or extensive sea-ice cover was 57 58 established, cold-stage expansions of southern-sourced water such as those 59 documented here enhanced carbon dioxide storage in the deep ocean, helping to 60 increase the amplitude of glacial cycles. 61 During the Last Glacial Maximum (LGM, ~26-19.5 thousand years ago, ka) nutrient 62 63 (carbon)-poor North Atlantic Deep Water (NADW), or northern component waters 64 (NCW) were replaced at depth in the western North Atlantic by nutrient (carbon)enriched southern-component waters (SCW)<sup>8</sup>, although the precise relationship 65 between this orbitally paced change in ocean structure and Atlantic Meridional 66 Overturning Circulation (AMOC) is uncertain<sup>8,9</sup>. Ocean circulation change is also 67

suggested<sup>2-5</sup> to have played an important role in driving Earth's last major transition 68 in secular climate state, the intensification of northern hemisphere glaciation (NHG) 69 (~3.6-2.4 million years ago, Ma<sup>10</sup>). Here we test this hypothesis by constructing new 70 records of water mass mixing in the deep North Atlantic Ocean using two proxies: 71 carbon isotopes ( $\delta^{13}$ C) in benthic foraminiferal calcite and the neodymium (Nd) 72 isotope composition  $(\epsilon_{Nd})$  of fish debris. Our records come from the benchmark site 73 74 for monitoring the water mass structure of the deep Plio-Pleistocene North Atlantic, Deep Sea Drilling Project (DSDP) Site 607<sup>11-15</sup> and its reoccupation<sup>16</sup>, Integrated 75 Ocean Drilling Program (IODP) Site U1313 (~41°N and 3427 m depth) situated in the 76 77 core of modern NADW. 78 79 The first reconstructions of water mass mixing during the intensification of NHG used  $\delta^{13}$ C from Site  $607^{11,12}$  and concluded that prominent glaciations from ~2.5 Ma were 80 associated with suppression of NCW in the deep North Atlantic, but that NCW 81 82 production was always greater during NHG intensification than during the Late Pleistocene glaciations. Subsequent  $\delta^{13}$ C-based investigations<sup>3,13-15</sup> concluded. 83 84 however, that NHG intensification was not associated with any major reorganization 85 of North Atlantic circulation, but was instead characterized by the persistence of NCW in the deep Atlantic and of SCW in the deep South Atlantic throughout this 86 time. That interpretation is at odds with suggestions that either a major spin-up<sup>2</sup> or a 87 slow down<sup>5</sup> in AMOC played a key role in driving NHG intensification. 88 89 Our ability to reconstruct water mass mixing using benthic  $\delta^{13}$ C is complicated by the 90 potential for biological modification of this signal <sup>14</sup> and published estimates of the 91 relative abundance of NCW (versus SCW) bathing the deep North Atlantic (%NCW) 92 during NHG intensification are compromised by use of low resolution  $\delta^{13}$ C data and 93 unsuitable end-member records (see S1 in supplementary information). To address 94 95 these issues we present two new independent estimates of %NCW over the past 3.3 Myr based on: (i) published benthic  $\delta^{13}$ C from Site 607 (for <2.4 Ma<sup>11,12</sup>) and Site 96 U1313 for 3.3 to 2.4 Ma<sup>16</sup> (which has twice the temporal resolution of the record from 97 98 Site 607 for the NHG intensification interval; Fig. 1); and (ii) new records of the  $\varepsilon_{Nd}$ 99 of fish debris at Site U1313 for both the Last Glacial to Holocene (35–5 ka) and the 100 Late Pliocene to earliest Pleistocene (1 sample every 6 kyr, 3.3–2.4 Ma; Fig. 2). Fish 101 debris flourapatite acquires a bottom water  $\varepsilon_{Nd}$  signature during early stage diagenesis

conditions<sup>17</sup>. Ocean  $\varepsilon_{Nd}$  acts as a quasi-conservative water-mass tracer because the 103 ocean residence time of Nd (400–700 years <sup>17</sup>) is short relative to the whole ocean 104 105 mixing time (~1500 years) allowing water masses originating in different ocean 106 basins to bear distinct  $\varepsilon_{Nd}$  signatures (reflecting spatial heterogeneity in the age and 107 lithology of material added to surface oceans and ocean continental-margin sediment boundary exchange processes<sup>17</sup>). The unradiogenic present day composition of NCW 108 (modern NADW,  $\varepsilon_{Nd} \sim -13.5$ , Ref. 17) reflects the ancient continental crust 109 110 surrounding sites of North Atlantic Deep-Water formation, while Pacific Deep Water 111 (typically  $\varepsilon_{Nd} \sim -3$  to -6, Ref. 17) and SCW end-members (modern Antarctic Bottom 112 Water and Antarctic Intermediate Water,  $\varepsilon_{Nd} \sim -7$  to -9, Ref. 17) bear more radiogenic values. Bottom water  $\varepsilon_{Nd}$  can be modified through 'boundary exchange' with ocean 113 sediments 18 but, unlike  $\delta^{13}$ C,  $\epsilon_{Nd}$  is independent of the carbon cycle 17, which 114 underwent major change during NHG intensification <sup>19</sup>. Thus, application of two 115 116 proxies with different controls provides a way to improve our understanding of NCW 117 history during this time. 118 119 In Figure 2 we present new  $\varepsilon_{Nd}$  records from Site U1313 for the Last Glacial through 120 mid-Holocene and for NHG intensification from the deep North Atlantic and compare them to other  $\varepsilon_{Nd}$  data sets and the  $\delta^{13}C$  records from Figure 1. Our  $\varepsilon_{Nd}$  record for the 121 Last Glacial shows the same isotopic composition and closely follows published 122 123 records from the deep western North Atlantic ( $\epsilon_{Nd}$  LGM > Holocene by ~3.5  $\epsilon$ units)8,18,20, demonstrating a clear incursion of radiogenic SCW into the deep North 124 125 Atlantic during the LGM. Based on the age model of Ref. 21, and our new records of 126 coarse lithic deposition at Site U1313 (Fig. 2c), changes in bottom water  $\varepsilon_{Nd}$  suggest that the return of NADW during the last deglacial may occur at our study site during 127 128 Heinrich (H)-event 1,  $\sim$ 18 ka,  $\sim$ 3 kyrs earlier than at deeper (>4500 m) and more 129 southerly sites (Fig. 2a). This finding may be attributable to the more proximal 130 location of Site U1313 to the Labrador Sea compared to Bermuda Rise, or 131 alternatively, bottom water  $\varepsilon_{Nd}$  at Site U1313 may be influenced by unradiogenic-132 labeling of deepwaters by massive episodes of ice-rafted detrital-carbonate deposition upstream of our study site during H1<sup>22</sup> (see supplementary information, S4). 133 Regardless, our records show that, outside of H-events,  $\varepsilon_{Nd}$  data from Site U1313 134

that is retained on geological timescales under widely divergent preservation

135 track the pattern of Last Glacial SCW incursion into the deep North Atlantic well documented by other proxy records<sup>9</sup>. 136 137 Our new Nd isotope record for NHG intensification exhibits a range of  $\varepsilon_{Nd}$  values (-138 139 9.6 to -13.1 = 3.5  $\varepsilon$ -units) similar to the amplitude of change (-11 to -14.5 = 3.5  $\varepsilon$ -140 units) documented for the Last Glacial transition (Fig. 2e,f). During the mid-141 Piacenzian Warm Period (~3.3-3 Ma), Site U1313 is characterized by unradiogenic 142 values (Fig. 2b) similar to those observed for NCW as observed in contemporaneous 143 Fe-Mn crust records precipitated in the shallow North Atlantic (1.9-2.65 km). In 144 contrast in a number of cold stages during NHG intensification (MIS M2, G16, G6, 145 100 and 96) and one interglacial (MIS K1), our Nd record shows prominent 146 radiogenic excursions that must reflect either significant changes in NCW  $\varepsilon_{Nd}$  or times 147 of increased influence of SCW on bottom waters at Site U1313 (see S2-4 in 148 supplementary information). Analogy to our Last Glacial record strongly supports the 149 latter explanation, thus providing the first evidence for a number of LGM-magnitude 150 incursions of SCW into the deep North Atlantic during NHG intensification. To 151 explore these events further we employ simple binary mixing equations (see methods. and supplementary information, S1-2) to both the  $\varepsilon_{Nd}$  and  $\delta^{13}$ C records from Site 152 153 607/U1313. 154 155 Both of our %NCW records (Figs. 1b, 2g-h) show significant excursions towards SCW values during NHG intensification (with reductions in %NCW down to 60%) 156 consistent with published reconstructions<sup>11,12</sup>, albeit not as clearly expressed in the 157 lower resolution Site 607  $\delta^{13}$ C record (Fig. S4e). Comparison of our %NCW $_{\delta 13C}$  and 158 %NCW $\varepsilon_{Nd}$  records shows that the significant radiogenic excursions in  $\varepsilon_{Nd}$  at Site 159 U1313 during MIS M2, 3.3 Ma, and MIS K1, ~3.07 Ma, (Fig. 2b,h) are not supported 160 by our  $\delta^{13}$ C-based record (Fig. 2h,j). Similarly, a number of cold stages between  $\sim 3$ 161 162 and 2.75 Ma show %NCW $_{\delta 13C}$  reductions that are not evident in %NCW $_{\delta Nd}$ . Our understanding of the history of %NCW prior to ~2.72 Ma will improve when our 163 164 knowledge of NCW and SCW end-member compositions is refined. Meanwhile, the 165 good correspondence between our two records from ~2.72 Ma onwards strongly 166 suggests that SCW incursions during MIS G6, 100 and 96 were comparable to the 167 LGM. This inference is supported by LGM-magnitude cooling of bottom water

168 temperatures at Site 607 during these three glacials, presumably reflecting the arrival 169 of cold SCW in the deep North Atlantic (Fig. 3a, b). We also note that these events 170 correlate with pronounced cooling of mid-latitude and/or Arctic surface temperatures 171 (Fig. 3a-c). 172 173 Our evidence for Southern Ocean incursions into the deep North Atlantic provides 174 fresh insight into the relationship between North Atlantic circulation and NHG 175 intensification (Fig 3). Numerical models of ocean circulation suggest that any 176 substantial secular slowdown in overturning of the NCW cell during NHG intensification would be registered by shoaling of its lower boundary<sup>23</sup> and hence a 177 change in  $\varepsilon_{Nd}$  at Site U1313 on a corresponding timescale. Instead the most prominent 178 feature of our records is that they exhibit enhanced excursions during cold-stages 179 180 rather than any long-term secular change (Fig. 2). We also observe the development of a lag in  $\epsilon_{Nd}$  relative to changes in benthic  $\delta^{18}O$  at Site U1313 from  $\sim\!\!2.9$  Ma 181 (typically of 4-9 kyr, Fig. S11), analogous to the Late Pleistocene (e.g., Ref. 8). These 182 183 observations suggest a response of Atlantic circulation to, rather than secular forcing 184 of glaciation. 185 186 Long-term expansion of the Antarctic Ice Sheet and coastal sea-ice between ~3.3 and 2.6 Ma<sup>4,5,24</sup>, bi-polar surface water stratification from ~2.7 Ma<sup>7</sup> and increased heat 187 transport from the northern hemisphere to the deep Pacific from this time<sup>4</sup> during 188 189 warm but not cold stages (this study) helped to precondition the northern hemisphere 190 for significant continental glaciation by cooling the Arctic and by enhancing storage of carbon dioxide in the abyss. We suggest that, once these preconditions were met 191 192 (Fig. 3g-h), incursions of SCW into the deep North Atlantic acted as a positive 193 feedback to amplify glacial cycles from ~2.72 Ma through increased sequestration of carbon in the deep Atlantic<sup>1</sup>, contributing to the observed<sup>4,5</sup> tighter coupling of inter-194 195 hemisphere climate on orbital timescales from this time (Fig. 3). One prediction of 196 this hypothesis is that further improvements to the proxy record of atmospheric CO<sub>2</sub> levels<sup>19</sup> will reveal an amplified signal at the obliquity scale from ~2.7 Ma. 197 198 199 It is debated whether Southern Ocean water incursions into the deep Atlantic during 200 the Last Glacial Cycle were driven by southern polar ocean conditions, notably

- deepwater densification through increased sea-ice formation (a proposed driver of
- 202 LGM NADW shoaling<sup>25</sup>) or by reductions in NCW formation during the LGM
- because of sensitivity to freshwater forcing<sup>8</sup> in the North Atlantic and/or sea ice
- 204 expansion in the Nordic Seas. For NHG intensification, the close association between
- 205 major SCW incursions at Site U1313 and evidence for the onset of major ice-rafted
- debris (IRD) deposition across both the Nordic Seas<sup>26</sup> and North Atlantic<sup>6</sup> from ~2.72
- 207 Ma onwards (Fig. 3f) suggests that SCW incursions into the deep North Atlantic were
- 208 perhaps driven by northern forcing agents. On the other hand, step-wise expansion of
- 209 the Antarctic Ice Sheet ~3.3 Ma is thought<sup>5</sup> to have triggered the initiation of quasi-
- permanent Antarctic sea ice formation (Fig 3g,h). Hence, if as suggested by our  $\varepsilon_{Nd}$
- record, major incursions of SCW into the deep Atlantic date to MIS M2 (i.e., well
- before the onset of major NHG<sup>6,27,28</sup>; Fig. 3e) then a southern hemisphere forcing
- 213 mechanism is implied.

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#### 322 Author contributions

- 323 IB and PAW designed the study. DCL picked, prepared and analysed fish debris for
- 324 their Nd isotope compositions with guidance from MG. IB generated the Last Glacial-
- Holocene IRD record. DCL made initial interpretations of the data. IB and PAW
- wrote the main text with input from GLF and MG. TBC and GLF performed Monte
- 327 Carlo simulations and statistical analyses. All authors discussed the results.

### 329 Competing financial interests

330 The authors declare no competing financial interests.

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#### 332 Figure captions 333 Fig. 1: Mixing history of northern-versus southern-sourced waters in the deep North 334 Atlantic (~3400 m) for the past 3.3 Myr based on benthic foraminiferal carbon isotopes: (a) benthic $\delta^{13}$ C records from the shallow North Atlantic (ODP Site 982<sup>14</sup>), 335 deep North Atlantic (composite of Chain 82-24-23PC<sup>29</sup>, DSDP Site 607<sup>11</sup>, and 336 U1313<sup>16</sup>) and deep Pacific (ODP Site 849<sup>30</sup>) (**b**) %NCW estimated using records 337 shown in (a) (purple line best estimate, shading 95% confidence interval, see 338 methods); (c) benthic for aminiferal $\delta^{18}{\rm O}$ data from Site U1313 (purple<sup>31</sup>) and global 339 benthic $\delta^{18}$ O stack<sup>10</sup> (black). 340 341 342 Fig. 2: Mixing history of northern-versus southern-sourced waters in the deep North Atlantic, for 3.3-2.4 Ma and the Last Glacial-Holocene (35-5 ka): (a & b) fish debris 343 $\varepsilon_{Nd}$ from IODP Site U1313 (purple; this study) and, in (a), $\varepsilon_{Nd}$ of uncleaned planktic 344 foraminifera (brown<sup>18</sup>) and bulk sediment leachate (green<sup>8,20</sup>) from Bermuda Rise 345 (error bars = $2\sigma$ uncertainty); (c & d) U1313 IRD concentration (40-5 ka, this study; 346 NHG intensification $^{16,31}$ ); $\varepsilon_{Nd}$ deviation from mean (35-5 ka) (e) and 3.3-2.4 Ma (f); (g 347 & h) %NCW at U1313 using $\varepsilon_{Nd}$ (top) from (a & b, this figure) and $\delta^{13}$ C (bottom) 348 349 from Fig. 1b; purple line best estimate, shading 95% confidence interval; (i-l) benthic for aminiferal $\delta^{13}$ C, $\delta^{18}$ O and $\epsilon_{Nd}$ from U1313<sup>16</sup>/82-24-23PC<sup>29</sup> and benthic $\delta^{18}$ O 350 stack<sup>10</sup>. Sources of crust and Cape Basin $\varepsilon_{Nd}$ records used to define NCW and SCW 351 352 $\varepsilon_{Nd}$ given in Tab. S3. Pink(blue) zones in (a) and (b) reflect $\varepsilon_{Nd}$ range of NCW(SCW) 353 end-members used to estimate %NCW shown in (g) & (h) (see methods). Red bar in 354 (a) = range of modern NADW $\varepsilon_{Nd}^{17}$ . 355 Fig. 3: Incursions of southern-sourced water into the deep North Atlantic (~3400 m) 356 and relation to NHG intensification: (a) %NCW $\epsilon_{Nd}$ (purple line best estimate, shading 357 95% confidence interval); (b) alkenone-SST at Site U1313<sup>32</sup> and Mg/Ca bottom water 358 temperature at Site 607<sup>29</sup>; (c) Air temperature in Arctic NE Russia<sup>28</sup> (d) age range and 359 2σ uncertainty for oldest advance of Laurentide Ice Sheet into Missouri<sup>27</sup>; (e) onset of 360 major NHG<sup>6,27,28</sup>; (**f**) Ice-rafting to U1313<sup>16,31</sup>, Site 611<sup>6</sup> (52.5°N) and Site 907<sup>26</sup> 361 (69°N); (g) Antarctic sea-ice evolution based on Site 1096 opal mass accumulation<sup>24</sup> 362 363 and on AND-1B diatom species<sup>5</sup>; (h) glacial regime of marine Antarctic Ice Sheet in Ross Embayment (AND-1B)<sup>5</sup>; (i) benthic $\delta^{18}$ O stack<sup>10</sup>. 364

365 366 Methods 367 To reconstruct the provenance of bottom waters bathing Site U1313 during NHG 368 intensification between 2.4 to 3.3 Ma (MIS MG1–95) we sampled on average every 369 30 cm (~6 kyr) from the shipboard primary splice (between 114.3 and 155.2 metres composite depth, mcd) used to generate the published benthic  $\delta^{18}$ O (Ref. 31) and  $\delta^{13}$ C 370 (Ref. 16) stratigraphies for our study site. Fish debris from sediments deposited at Site 371 372 U1313 during MIS 100, and from six samples on which detrital sediment  $\varepsilon_{Nd}$  values were previously published<sup>16</sup>, were hand-picked from samples taken from the 373 374 secondary splice after placing the secondary splice onto the depth scale of the primary 375 splice via manual graphical correlation of shipboard-derived sediment lightness 376 records (L\*, here driven by variability in the sediment calcium carbonate content<sup>16</sup>, Fig. S10). The age model for our record is based on tuning<sup>31</sup> the Site U1313 benthic 377  $\delta^{18}$ O stratigraphy to the LR04 stack<sup>10</sup>. To generate a Last Glacial to Holocene  $\varepsilon_{Nd}$ 378 379 record for Site U1313 we sampled Holes C and D of this site every 7 to 14 cm from 380 0.2-1.76 mcd, (n = 22, see also Fig. S9) corresponding to an average sampling resolution of ~1.3 kyr. The published age model<sup>21</sup> that we use for our Last Glacial 381 382 record is based on correlation of H-layers in the Site U1313 stratigraphy (tracked by 383 XRD dolomite/calcite ratio data) to X-ray fluorescence-based Ca/Sr elemental data 384 that track the position of H-layers in the stratigraphy of northeast Atlantic Ocean IODP Site U1308 (~50°N), which has a benthic  $\delta^{18}$ O stratigraphy on LR04 ages<sup>33</sup>. 385 386 387 We hand-picked fish debris (10 to 30 pieces of bones and teeth, depending on 388 availability) from the >150µm fraction of disaggregated sediment. Samples were 389 cleaned with two steps of ultrasonication in methanol, followed by three rinses in 390 MilliQ H<sub>2</sub>O. Organic material was removed by bathing each sample in an oxidising 391 solution (1% H<sub>2</sub>O<sub>2</sub> – 0.1M NH<sub>4</sub>OH) at 80°C for 30 minutes, while sonicating for 30 392 seconds every 10 minutes. A weak-acid (0.001M HNO<sub>3</sub>) leach was applied to ensure 393 removal of any reabsorbed contaminants, before samples were dissolved in 0.1ml 1.75M HCl. Pure samples of Nd were separated following standard procedures<sup>34,35</sup>.

Nd isotope ratios (143Nd/144Nd) were measured at the University of Southampton using a multi-collector inductively coupled plasma mass spectrometer (MC-ICP-MS, Thermo Scientific Neptune). Neodymium isotope compositions were obtained using

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the method of Ref. 36 through adjustment to a  $^{146}$ Nd/ $^{144}$ Nd ratio of 0.7219 and a secondary normalization to  $^{142}$ Nd/ $^{144}$ Nd = 1.141876 (Ref. 36). Total procedural blanks averaged 35 pg for fish debris. External reproducibility of the JNdi standard<sup>37</sup> was  $\pm 0.000006$  (2 s.d.), corresponding to an external error of  $\pm 0.13 \, \epsilon_{Nd}$  (2 s.d.). In all cases we plot the external error, unless the internal error is larger than the external error when we plot a combined error calculated as  $\sqrt{\text{(external error}^2 + internal error}^2)}$ . All Nd isotope ratios are reported in epsilon notation as:

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$$406 \qquad \varepsilon_{Nd} = \left[ \frac{\frac{143}{Nd} \frac{144}{Nd} \frac{Nd}{sample}}{\frac{143}{Nd} \frac{144}{Nd} \frac{Nd}{c_{HUR}}} - 1 \right] x 10^4$$

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Radiogenic ingrowth of <sup>143</sup>Nd is corrected using a typical <sup>147</sup>Sm/<sup>144</sup>Nd of 0.125 (Ref.

409 38). Corrections are within analytical uncertainty ( $< 0.06 \, \epsilon_{Nd}$  for our NHG

intensification data, and negligible for Last Glacial-Holocene data), reflecting the

411 young age of our samples relative to the half-life of <sup>147</sup>Sm. We note that fish debris

412 <sup>147</sup>Sm/<sup>144</sup>Nd ratios typically vary within a narrow range and that our results are

insensitive to the choice of this value.

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A record of sand IRD abundance in sediments deposited at Site U1313 during NHG intensification already exists <sup>16,31</sup>. To assess the concentration of IRD in sediments for the Last Glacial at our study site we performed new coarse lithic counts on the >150 µm size fraction between 0.2-2.43 mcd. To generate a statistically significant record of sand IRD abundance in Site U1313 sediments (expressed as IRD per gram of dry sediment) we identified the composition of at least 300 coarse lithics or where 300 grains were not present we counted all grains in the sample (where there were typically only 1-30 total grains in Last Glacial sediments deposited outside of Heinrich-events). In each case, our counts differentiated between volcanics, cream coloured detrital limestone and other coarse lithics (predominantly quartz and

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To estimate the relative contribution of NCW bathing Site 607/U1313 over the past

428 3.3 Ma using benthic  $\delta^{13}$ C datasets we used the following binary mixing equation:

feldspar, plus minor numbers of other rock clasts).

429 %NCW<sub>$$\delta_{13C}$$</sub> = 100 \*  $\left(\frac{\delta_{13C_{(607/U1313)}} - \delta_{13C_{(SCW)}}}{\delta_{13C_{(NCW)}} - \delta_{13C_{(SCW)}}}\right)$ 

- Where  $\text{%NCW}_{\delta 13C}$  is the relative contribution of NCW to waters bathing the deep
- North Atlantic Ocean (%SCW = 100 %NCW),  $\delta^{13}C_{(607/U1313)}$  is the carbon isotope
- composition of benthic foraminiferal calcite (predominantly determined from the
- 433 epifaunal benthic foraminifera Cibicidoides wuellerstorfi) from the deep North
- Atlantic (Site 607/U1313), and  $\delta^{13}C_{(NCW)}$  and  $\delta^{13}C_{(SCW)}$  are the equivalent carbon
- isotope signatures of NCW (ODP Site 982, Ref. 14) and SCW (ODP Site 849, Ref.
- 436 30), respectively (see supplementary information, S1, for further details).

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- To estimate the relative contribution of NCW and SCW to waters bathing IODP Site
- 439 U1313 during NHG intensification and the last deglacial using Nd isotopes we used
- the following binary mixing equation constrained by our current understanding of
- end-member compositions and their Nd concentrations:

442 %NCW<sub>ENd</sub> = 
$$\frac{\frac{fnCs}{fsCn}}{1 + \frac{fnCs}{fsCn}} \times 100$$

- Where %NCW<sub>ENd</sub> is the relative contribution of NCW to waters bathing Site U1313
- 444 (%SCW = 100 %NCW<sub>ENd</sub>) and Cs = concentration of Nd in SCW, Cn =
- concentration of Nd in NCW. fn = fraction of Nd coming from the north, such that:

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$$fn = \frac{\varepsilon_{NdU1313} - \varepsilon_{NdSCW}}{\varepsilon_{NdNCW} - \varepsilon_{NdSCW}}$$

- where  $\varepsilon_{NdU1313}$  is the Nd isotope value of fish debris from Site U1313,  $\varepsilon_{NdSCW}$  is the
- 450 southern end member isotope composition and  $\varepsilon_{NdNCW}$  is the northern end member
- isotope composition; fs is then the fraction of Nd coming from the south which is
- equal to 1-fn in our binary mixing model (i.e. everything that is not from a northern
- 453 source). To estimate the  $\varepsilon_{Nd}$  of NCW and SCW during NHG intensification we took
- the average value of all published Fe-Mn crust  $\varepsilon_{Nd}$  (and their 2s.d.) that span ~3.3-2.4
- 455 Ma (Fig. 2b and Tab. S3). For our Last Glacial-Holocene record, following Ref. 39,
- 456 NCW  $\varepsilon_{Nd}$  was defined as the average of our Holocene data (<14 ka; MIS 2/1
- boundary as defined by Ref. 10) from Site U1313 (and their 2s.d.) (Fig. 2a and Tab.
- 458 S3). We define SCW  $\varepsilon_{Nd}$  for this time interval using a splice of Site RC11-83 &

- 459 TN057-21  $\varepsilon_{Nd}$ ; Ref. 39; Tab. S3), which we resampled onto the ages of our Site
- U1313 record (see S2 in supplementary information for further details). Confidence 460
- intervals (95%) for %NCW are estimated using a Monte Carlo simulation (n=10,000) 461
- performed in R<sup>40</sup>, fully propagating analytical uncertainties, and uncertainties in the 462
- $\varepsilon_{Nd}$  of NCW and SCW and their Nd concentrations<sup>41</sup> (see S2 in supplementary 463
- information). To perform this exercise we make the following assumptions: (i) Nd 464
- 465 isotopes exhibit conservative behaviour, which is an acceptable approximation in
- abyssal open ocean settings, (ii) mixing of NCW and SCW at Site U1313 is binary, 466
- and (iii) that the modern day end-member [Nd] has remained constant over the past 467
- 468 ~3.3 Ma (see S2 in supplementary information).
- To determine the lag-correlation of Site U1313  $\epsilon_{Nd}$  (this study), benthic  $\delta^{13}C$  (Ref. 16) 470
- and Site U1313 benthic  $\delta^{18}$ O (Ref. 31) during NHG intensification (Fig. S11) we used 471
- an auto-correlation function in R<sup>40</sup> every 50 kyr using a 200 kyr window. Before 472
- 473 analysing the data all records were resampled at a common 1 kyr resolution.

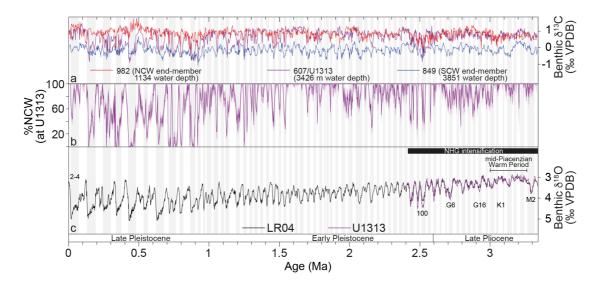
#### **Methods References**

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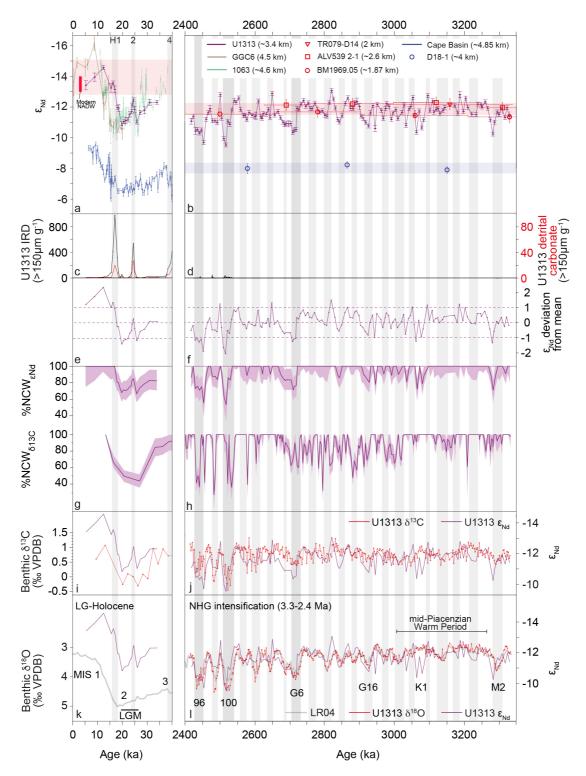
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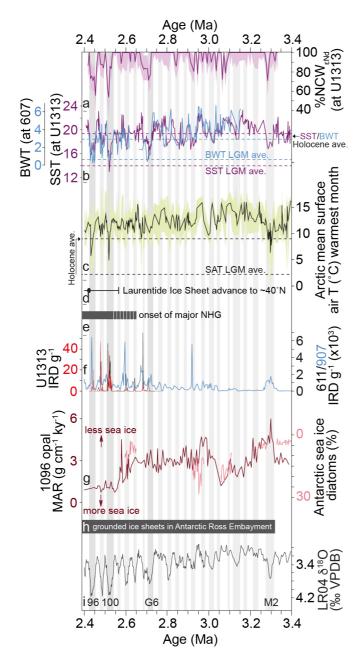




509 Figure 1.



510511512 Figure 2.



513514515 Figure 3.516