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1	Holocene shifts in sub-surface water circulation of the North-East Atlantic inferred
2	from Nd isotopic composition in cold-water corals
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13	
14	Abstract:
15	Variations of the Sub-Polar Gyre (SPG) and the Sub-Tropical Gyre (STG) circulation during
16	the Holocene are believed to be related to regional and global climate over this time period.
17	To improve our understanding of these phenomena we provide new constraints on variations
18	in surface circulation patterns using neodymium isotopes (ENd) on precisely U-Th dated coral
19	fragments of L. pertusa. The fragments were retrieved from two sediment cores taken from
20	cold-water coral (CWC) mounds at ~ 127-134 m water depth in the Mingulay Reef Complex
21	located on the Western British continental shelf. The results have been combined with ENd
22	analyzed on seawater samples from two stations located on the continental shelf and margin
23	in order to establish whether ϵ Nd is a reliable proxy of the ocean circulation variations and

24 notably of the relative contribution of water originating from the SPG and STG. ENd values in CWCs from the Mingulay Reef Complex range from -14.5±0.4 to -11.8±0.3, highlighting two 25 26 major variations. Unradiogenic ɛNd values (-14.5±0.4) indicate a higher contribution of water from the SPG around 2.8 ka. Conversely, more radiogenic values at 3.4 ka (-11.8±0.3) point 27 to a declining SPG strength, accompanied by stronger northward penetration of STG water 28 along the western European margin transported by the Shelf Edge Current (SEC) and/or 29 30 cooler and fresher waters from the interior Seas. The eastward extension of the SPG at 2.8 ka is associated with lower ¹⁴C reservoir age (200 yrs) compared to periods associated with a 31 higher contribution of STG waters. This indicates that ¹⁴C reservoir ages are mainly a function 32 of vertical mixing of the sub-surface of the ocean. As stronger vertical ventilation is not 33 associated with a higher proportion of local radiogenic surface water, we hypothesize it could 34 represent vertical ventilation in the North-Eastern Atlantic. Active SPG is associated with a 35 better ventilation of the water masses within the SPG and warmer climatic conditions in 36 37 Northern Europe and in the Eastern Norwegian Sea linked to an intensification of the surface limb of the AMOC. 38

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40 Key words: Nd isotopic composition; cold-water corals; NE Atlantic; Sub-Polar Gyre

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42 **1. Introduction**

The Atlantic Meridional Overturning Circulation (AMOC) transports warm saline surface water masses northwards and deeper cool waters southwards (Hansen and Østerhus, 2000; Read, 2001). The upper layer of the AMOC, the North Atlantic Current (NAC), loses much of its heat to the atmosphere on its way northwards, impacting the climate over Europe (Hall and Bryden, 1982). Whereas variability of the lower branch of the AMOC is relatively well established during the Holocene, at least for the Iceland-Scotland Overflow Water (ISOW)

and the Labrador Sea Water (LSW) (e.g. Hillaire-Marcel et al., 2001; Hoogakker et al., 2011; 49 50 Thornalley et al., 2013; Mjell et al., 2015), the variations of the upper layer of the AMOC are not yet well established. This is due to the complex spatial and temporal variability of the 51 52 classical Sea Surface Temperature (SST) and Sea Surface Salinity (SSS) proxies used to track past changes of the AMOC that are strongly influenced by an orbitally-induced insolation 53 trend and the final disintegration of the residual Laurentide ice sheet during the early 54 Holocene, which induces a pronounced provincialism in the surface physical property records 55 of the North Atlantic (e.g. de Vernal and Hillaire-Marcel, 2006; Renssen et al., 2009). 56

It has been suggested that during modern times variations in the shape and position of the 57 58 subpolar gyre (SPG) (Fig. 1) play a major role in the redistribution of fresh melt-water and in pole-ward heat transport at surface and intermediate depths and subsequently have an impact 59 on the AMOC strength and the properties of waters entering the Nordic Seas (e.g. Häkkinen et 60 al., 2013). The NAC follows the boundary between the North Atlantic subpolar gyre (SPG) 61 and the subtropical gyre (STG), and its position, intensity and composition have been 62 63 modulated by SPG variations over recent decades (Hàtun et al., 2005; Berx and Payne, 2017; Zunino et al., 2017). Foukal & Lozier (2018), by using a combination of empirical orthogonal 64 functions and direct calculations of the size and strength of the SPG, have proposed that the 65 66 SPG dynamics may not control the magnitude of the STG waters flowing into the North east Atlantic, on interannual time scales. However, numerous paleohydrological records point to a 67 major role of the SPG on longer time scales (see references below). During the early 68 Holocene, a reorganization of the mid-depth (200-800 m) circulation has been highlighted at 69 about 7-6 ka, with a weakened SPG leading to a stronger northward penetration of temperate 70 water masses in the North-East Atlantic (Thornalley et al., 2009; Colin et al., 2010). Other 71 studies based on cold-water corals from the Bay of Biscay and the Rockall Trough have 72 73 highlighted an existing link between SPG intensity and the North Atlantic Oscillation (NAO),

i.e. atmospheric circulation (Copard et al., 2012; Montero-Serrano et al., 2013). Between 1950 74 and 1990, periods characterized by a dominant negative phase of NAO were associated with a 75 stronger influence of mid-depth STG water (Montero-Serrano et al., 2013). Over the last 1500 76 77 yrs, the warm Medieval Climatic Anomaly (between 1000 AD and 1250 AD), characterized by a persistently positive NAO phase, resulted in greater eastward extension of the mid-depth 78 SPG in the North Atlantic (Copard et al., 2012). Conversely, the Little Ice Age (between 1350 79 AD and 1850 AD), characterized by a persistently negative NAO phase, is associated with the 80 westward contraction of the mid-depth SPG (Copard et al., 2012). Overall, these records have 81 shown that Holocene climatic anomalies are associated with mid-depth SPG dynamics which 82 83 are in turn affected by changes in wind stress and/or freshwater input from the Labrador Sea (Thornalley et al., 2009; Colin et al., 2010; Copard et al., 2012). Subsequent variations in the 84 heat and salt budgets at intermediate depths may have contributed to changes in the properties 85 86 of North Atlantic inflow to the Nordic Seas and thus to deep-water formation.

The Mingulay Reef Complex, located on the Western British continental shelf (Fig. 1), is a 87 potential target to track past changes of the SPG extension using the ENd proxy because it 88 receives waters from the NAC. In addition, cold-water corals (CWCs) have been shown to act 89 as useful archives for identifying rapid changes in the dynamics of water masses (Lutringer et 90 al., 2005; Colin et al., 2010; van de Flierdt et al., 2010; Copard et al., 2010, 2012; Montero-91 Serrano et al., 2013; Wilson et al., 2014; Chen et al., 2015; Dubois-Dauphin et al., 2016; 92 Douarin et al., 2016). Firstly, their aragonitic skeleton can be precisely dated thanks to U-93 series disequilibrium methods (Adkins et al., 1998; Cheng et al., 2000; Douville et al., 2010). 94 95 Secondly, they do not suffer from the same problems often encountered in marine sediment cores, which are often affected by bioturbation, smoothing the original signals, and which are 96 characterized by low sedimentation rates that limit our ability to obtain records with 97 98 centennial resolutions. Furthermore, the Nd isotopic composition, expressed as ϵ Nd =

 $([(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR}] - 1) \times 10000$ (CHUR: Chondritic Uniform Reservoir 99 [Jacobsen and Wasserburg, 1980]), of living and fossil scleractinian CWCs faithfully traces 100 water mass provenance and mixing in the ocean (e.g. van de Flierdt et al., 2010; Copard et al., 101 102 2012; Struve et al., 2017). Taking into consideration that mid-depth STG and SPG water masses are characterized by contrasted Nd isotopic signatures ($\epsilon Nd \approx -10$ and $\epsilon Nd \approx -15$, 103 respectively; Lambelet et al., 2016; Dubois-Dauphin et al., 2017), CWC ENd records obtained 104 105 previously in the Rockall Trough and in the northern Bay of Biscay have been used to reconstruct the eastward extension of the mid-depth SPG (Colin et al. 2010; Copard et al., 106 2012; Montero-Serrano et al., 2011, 2013). 107

108 In this paper we present a high-resolution ENd record for the last 4500 yrs, which is derived from CWCs from the Mingulay Reef Complex, located at the sub-surface under the strong 109 influence of NAC water. Our objective is to track hydrological changes in the SPG dynamics 110 111 during the Holocene. Ambient seawater ENd was also investigated to assess seawater ENd of the Western British continental shelf and margin and to verify that ENd is a reliable proxy for 112 tracking past changes in the influence of the SPG in this region. This CWC ENd record was 113 compared to previously published Δ^{14} C performed by Douarin et al. (2016) that was 114 115 interpreted in terms of changes in water mass origin. It also complements well previous studies based on ENd in the Rockall Trough (Colin et al., 2010; Copard et al., 2012) that have 116 highlighted major variations of the mid-depth SPG dynamics throughout the Holocene. Our 117 study permits us to re-interpret Δ^{14} C with a proxy of water mass origin and provides new 118 119 evidence of abrupt changes in the composition of water masses entering the Nordic Seas; these changes are linked to variations in the shape and strength of the SPG. 120

121

122 **2.** Hydrological setting

Presently, the deepest waters (>100 m) and the waters overlying the outer parts of the shelf of 123 west Scotland are of Atlantic origin (Inall et al., 2009). At the surface, the northward flowing 124 Scottish Coastal Current (SCC) brings cooler and fresher waters from the Irish and Clyde 125 Seas (Ellett and Edwards, 1983) (Fig. 1). The Mingulay Reef Complex (56°50'N; 7°20'W; 126 100-260 m water depth) is thus primarily bathed by Atlantic water (Dodds et al., 2007). The 127 upper-layer Atlantic water off the Mingulay Reef Complex, carried by the NAC, results from 128 the mixing of saline Eastern North Atlantic Central Water (ENACW) originating from the 129 Bay of Biscay (Ellett and Martin, 1973; Ellett et al., 1986; Pollard et al., 1996) and the fresher 130 Modified North Atlantic Water (MNAW) originating in the North-West Atlantic (New and 131 132 Smythe-Wright, 2001). The MNAW is formed by the mixing of the Western North Atlantic Central Water (WNACW), which flows from the Caribbean Sea, with the Sub-Arctic 133 Intermediate Water (SAIW). The SAIW is characterized by low temperatures and salinity and 134 135 is formed in the upper layers of the southern Labrador Sea (Arhan, 1990). Both WNACW and SAIW are transported north-eastwards into the NAC where vertical mixing occurs (Arhan, 136 137 1990). The NAC is composed of three main branches (Daniault et al., 2016). The northern and central branches bifurcate west of the Rockall Trough and feed the cyclonic circulation in the 138 Iceland Basin and the Irminger Sea (Fig. 1). The southern branch penetrates into the Rockall 139 Trough (Fig. 1) 140

All along the European margin, the Shelf Edge Current (SEC) forms a northward-flowing boundary current that brings warm and saline upper water with ENACW characteristics through the Rockall Trough and the Scottish margin (Hill and Mitchelson-Jacob, 1993; White and Bowyer, 1997; Holliday et al., 2000) (Fig. 1). Below this, the Mediterranean Sea Water (MSW) flows northwards from the Gulf of Cadiz with a main core located at 1000-1200 m water depth. Nevertheless, the MSW cannot be traced through εNd, salinity and potential temperature at mid-depth further north than the Porcupine Seabight (Iorga and Lozier, 1999; McCartney and Mauritzen, 2001; New and Smythe-Wright, 2001; Lavender et al., 2005;Dubois-Dauphin et al., 2017).

The NAC strength, direction and composition respond to SPG dynamics (Hátún et al., 2005): when the SPG circulation is stronger and presents a more pronounced east-west shape the SPG water influence on the NAC is increased relative to the STG water, coupled with a strong AMOC. Conversely, when the SPG circulation is weaker and forms a more pronounced north-south shape, a lower contribution of SPG water flows to the NW European Margin relative to the STG water (Larsen et al., 2012; Hansen et al., 2015).

156

3. Material and methods

158 **3.1.1. CWC samples**

This study has investigated 23 well-preserved fossil CWCs of the species Lophelia pertusa, 159 collected in two cores from the Mingulay Reef Complex located on the Western British 160 continental shelf (Fig. 1 and Table 1). The cores, +56-08/929VE (56°49'19"N - 7°23'27"W, 161 127 m water depth) and +56-08/930VE (56°49'20"N -7°23'47"W, 134 m water depth), were 162 collected in October 2007 by the British Geological Survey during a survey (Cruise 15) on 163 board of the NERC vessel RRS James Cook (Stewart and Gatliff, 2008) (Table 1). The coring 164 sites are located within 300 m of each other at quite a similar water depth on small carbonate 165 mounds of the Mingulay Reef Complex (Figure 1; Table 1). The sediment cores are 166 composed of biogenic fragments (mainly CWC fragments) in a carbonate matrix. Three 167 additional surface videograb samples (GRAB 15.5.5.10; GRAB 56.08.928; GRAB 1151) 168 were retrieved from the seafloor with a Remotely Operated Vehicle close to the location of 169 the cores (see location in Fig. 1 and Table 1). 170

U-Th ages obtained for CWC samples collected in the Mingulay Reef Complex were
published by Douarin et al. (2016) (Table 1). They document three major peaks in growth rate
that are centered at 3.7, 4, and 4.2 ka BP. Two major reductions in growth rate were observed
at 1.75–2.8 ka and 3.2–3.6 ka BP. The coral fragments investigated in this study, from cores
+56-08/929VE and +56-08/930VE, range between 4.29±0.26 and 2.09±0.018 ka BP (Table
1). In addition, GRAB 56.08.928, GRAB 1151 and GRAB 15.5.5.10 samples have been dated
to 2.67±0.014, 1.31±0.011 and 0.02±0.013 ka BP, respectively (Table 1).

178 **3.1.2. Seawater samples**

During the R/V Atalante MINGULAY-ROCKALL cruise in June 2016, 12 seawater samples 179 were collected at 2 full water column stations MR-4 (56°37.44' N; 9°5.48' W) and MR-5 180 (56°46.04'N; 7°25.98' W) (Fig. 1, Table 2). These samples, located on the Western British 181 182 continental shelf (station MR5) and margin (station MR4), were collected in order to assess seawater ENd flowing around the Mingulay Reef Complex. The seawater samples and water 183 column hydrographic parameters (temperature and salinity) were collected using a CTD-184 185 Rosette system equipped with 12 10-liter Niskin bottles and a standard Sea-Bird SBE 911plus CTD-Rosette system. The samples were then filtered through 0.8-0.45 µm AcroPak 500 186 capsule filters and transferred to 10-liter acid-cleaned bottles, and immediately acidified to a 187 188 pH of 2 using suprapur 2 N HCl.

189 3.2.3 Analytical procedures for Nd isotope measurements

Each coral fragment used in this study corresponded to one polyp of the *Lophelia pertusa*species, integrating only a few years of growth (Freiwald et al, 2004; Sabatier and al., 2012a).
The CWC samples were subjected to a mechanical and chemical cleaning procedure (Copard
et al., 2010). The visible contaminations, such as Fe-Mn coatings and detrital particles, were
carefully removed from the inner and outermost surfaces of the coral skeletons using a small

diamond blade. The physically cleaned fragments were ultrasonicated for 10 min. with 0.1 N
ultra-clean HCl, followed by several MilliQ water rinses and finally dissolved in 2.5 N
ultraclean HNO₃. Neodymium was separated from the carbonate matrix using Eichrom TRU
and LN resins, following the analytical procedure described in detail in Copard et al. (2010).

Neodymium was purified from seawater samples following the analytical procedures 199 200 described in detail by Lacan and Jeandel (2001) and Wu et al. (2015). Briefly, seawater REEs were preconcentrated using SepPak Classic C18 cartridges loaded with a HDEHP/H2MEHP 201 202 complexing agent. Solutions were then passed through a cationic resin (AG50W-X8) and finally Nd was extracted and purified using an Eichrom Ln-Spec resin following the method 203 204 described in detail by Copard et al. (2010). This purification process and the measurement techniques applied follow approved GEOTRACES protocols and GEOPS laboratory 205 participated in the international GEOTRACES intercalibration study (van de Flierdt et al., 206 2012). 207

The ¹⁴³Nd/¹⁴⁴Nd ratios of seawater samples and of the CWCs from the Mingulay Reef 208 209 Complex, were analyzed using a Thermo Scientific Neptune Plus MC-ICPMS installed at the 210 Laboratoire des Sciences du Climat et de l'Environnement (LSCE, Gif-sur-Yvette, France). The mass-fractionation correction was made by normalizing ¹⁴⁶Nd/¹⁴⁴Nd to 0.7219 by 211 212 applying an exponential-fractionation correction. During the analytical sessions, every two samples were bracketed with analyses of appropriate standard JNdi-1 and La Jolla Nd 213 214 solutions, which are characterized by certified values of 0.512115±0.000006 (Tanaka et al., 2000) and 0.511858±0.000007 (Lugmair et al., 1983), respectively. Standard JNdi-1 and La 215 Jolla solutions were analyzed at concentrations similar to those of the samples (4-10 ppb). The 216 217 external reproducibility (2σ) deduced from repeated measurements of the La Jolla standard and JNdi-1, ranged from 0.2 to 0.4 Epsilon units for the different analytical sessions. The 218

analytical error associated with each sample analysis is taken as the external reproducibility ofthe La Jolla standard for each session.

221

4. Results

4.1. Seawater samples

The Nd isotopic compositions of the seawater samples from stations MR-4 and MR-5 are provided in Table 2 and Figure 2. Station MR-5 is located on the continental shelf, very close to the Mingulay Reef Complex. At station MR-5, ε Nd values decrease downward from -11.6±0.2 at the surface to -13.1±0.3 at a water depth of 240 m (Fig. 2). Temperature decreases from 12 to 9.5°C between depths of 30 and 80 m, then remains constant (9.5°C) down to 240 m water depth (Fig. 2). Conversely salinity increases from 34.5 to 35.2 in the upper 100 m, then displays a constant value of 35.2 down to 240 m water depth (Fig. 2).

On the other hand, station MR-4 was collected on the continental slope. Seawater samples from station MR-4 exhibit similar εNd values at all depths, within the error bars, centered around -14 from the surface to 540 m water depth (Fig. 2). Temperature decreases from 13 to 10 °C between depths of 20 and 80 m, then displays a constant value of about 9.7°C down to 550 m water depth. Salinity displays an almost constant value of about 35.3. The upper 50 m are, however, slightly less saline (Fig. 2).

237 4.2. Cold Water Coral samples

Overall, CWC ε Nd values from the Mingulay Reef Complex range from -14.5±0.4 to -11.8±0.3 (Fig. 3). The earlier part of the record (4.5 – 3.5 ka) displays a narrow range of values between -13.1±0.3 and -12.5±0.3 (Fig. 3). An abrupt increase of the ε Nd values for corals occurred between 3.40 and 3.20 ka BP, reaching between -12.1±0.3 and -11.8±0.3 (Fig. 3). A return to lower values (-13.1±0.3 to -12.4±0.4) is observed between 3.12 and 2.82 ka
BP, followed by an abrupt and short decrease over 100 years to -14.5±0.4 at 2.75 ka BP (Fig.
3). After 2.67 ka BP, only four specimens display εNd values from -12.0±0.4 to -13.2±0.5
(Fig. 3).

246

247 **5.** Discussion

248 5.1. Significance of the εNd record

The youngest CWC sample collected in the Mingulay Reef complex, with a modern age of 249 250 0.02±0.013 ka BP (Douarin et al., 2016), displays an ENd value of -13.2±0.5 (Fig. 3) which is similar, within the error bars, to the present-day value of seawater flowing at depth of the 251 study site (between -12.9±0.2 and -12.4±0.2 at 101-152 m water depth; station MR-5) (Fig. 2 252 253 and 3). At station MR-4, located on the continental slope, seawater samples values range from -14.4±0.2 to -13.8±0.2 down to 500 m water depth (Fig. 2). These values are in agreement 254 with seawater analyzed in the Rockall Trough (ICE-CTD 03; Dubois-Dauphin et al., 2017) 255 256 the values of which range from -14.0 ± 0.2 to 13.3 ± 0.2 for the same depth interval (Fig. 4), implying that subsurface water masses brought by the NAC have a strong influence along the 257 continental slope near the study site. In contrast, station MR-5, located on the continental 258 shelf, exhibits more radiogenic values at all depths compared to the continental slope (MR-4) 259 and the Rockall Trough (ICE-CTD 03; Dubois-Dauphin et al., 2017). The ENd value of warm 260 261 and saline surface water (upper ENACW) flowing in the SEC is poorly constrained along the 262 European margin. It has been reported to be radiogenic in the Bay of Biscay (-10.7±0.3 at 140 m in CAROLS stations; Copard et al., 2011), while there are no values shallower than 400 m 263 264 published for the Porcupine Seabight (-13±0.2 at 400 m in station ICE-CTD 02; Dubois-Dauphin et al., 2017) (Fig. 4). This radiogenic water brought by the SEC is expected to have a 265 strong influence on the Mingulay Reef Complex as seen at station MR-5. 266

Nevertheless, Nd isotopic signatures for the SCC that circulates from the Irish Sea and the 267 Firth of Clyde are unknown. However, rocks and river sediment samples from the northern 268 British Isles display a mean ε Nd value of -10.6 \pm 0.4 (Davies et al., 1985), suggesting a rather 269 270 radiogenic Nd isotopic signature for seawater from interior Seas. Crockett et al. (2018) have provided Nd concentrations (20 to 60 pmol.kg⁻¹) from seawater collected in a coastal station 271 (station 9G, 166 m bottom depth; Fig. 1) located close to the Mingulay Reef Complex 272 suggesting possible local desorption from resuspended sediments or pore water release to the 273 274 overlying water column. Seawater ENd obtained in the upper 50 m of the MR-5 water station, collected above the Mingulay Reef Complex, allows us to establish for the first time the ɛNd 275 276 value of seawater flowing along the Scottish margin; this value is around -11.7±0.2 and is associated with a temperature and salinity of about 10-12°C and 34.5-34.9, respectively (Fig. 277 2). This suggests that surface water is likely affected by local inputs from the Scottish shelf 278 279 and/or the interior Seas. On the other hand, the ENd values are more unradiogenic (from -13.0±0.3 to -12.4±0.2) below 100 m at water station MR-5 (in the range of Porcupine 280 281 Seabight; Dubois-Dauphin et al., 2017) and associated with salinity of ~35.2 suggesting that CWCs from the Mingulay Reef Complex are under the influence of a mixing between SEC 282 and NAC waters. Furthermore, the initial δ^{234} U values of the samples considered in this study 283 ranged from 142.9 ± 2.4 to 150.0 ± 3.5 (2σ uncertainties; Douarin et al., 2013), which is in the 284 range of the modern seawater value (146.8±0.1; Andersen et al., 2010), while ²³²Th values for 285 286 all corals were low (<0.48±0.07 ppb; Douarin et al., 2013). These arguments do not support an influence of a local source on CWC ENd from the Mingulay Reef Complex. However, the 287 desorption from resuspended sediments cannot be ruled out. 288

εNd of CWC samples, collected at 747 m water depth at the SW Rockall Trough margin, has
been investigated by previous studies and has been shown to range from -11.9±0.3 to 14.4±0.2 over the last 4500 yrs (Colin et al., 2010; Copard et al., 2012) (Fig. 3). These values

are close to those obtained at the Mingulay Reef Complex, with a generally good 292 293 correspondence between samples of the same age in both sites (Fig. 3). Seawater ENd investigated in the Rockall Trough does not indicate significant variations of Nd isotopic 294 295 composition down to 1000 m water depth (-13.3±0.2 to -14.0±0.2; Dubois-Dauphin et al., 2017) and presents similar values to those found at station MR-4 (Fig. 4). This implies a 296 relatively strong homogeneity of the Nd isotopic composition of the surface-to-mid water 297 masses of the Rockall Trough. On the other hand, station MR-5 displayed more radiogenic 298 299 values, as the Mingulay Reef Complex is likely to experience a strong mixing with STG water. Nevertheless, both sites should be under the influence of the gyre circulation system. In 300 301 this instance, it is therefore possible to consider that the CWCs from the shallow Mingulay Reef Complex record similar gyre dynamics than the mid-depth of the SW Rockall Trough 302 303 margin.

The ε Nd record from the Rockall Trough provides evidence of negative excursions during the warm Medieval Climatic Anomaly (between 1000 AD and 1250 AD) and during the most recent period (1950 AD to 2000 AD) (Fig. 3), which have been interpreted as indicating a strengthened SPG and an increase in the subpolar water contribution to the Nordic Seas (Copard et al., 2012). CWCs from the Mingulay Reef Complex do not record these events as none of the sample ages match these periods. However, they indicate another major negative excursion of ε Nd values at about 2.8 ka BP (Fig. 3).

The presence of Ice Rafted Debris (IRD) during the Holocene is evidenced in North Atlantic sediment cores located in the Rockall Trough (cores MC52 and VM29-191; Bond et al., 2001) (Fig. 1) by peaks in hematite-stained grains and Icelandic glass. It has been shown to have a potential influence on ɛNd records during periods of northern-hemisphere ice sheet collapse throughout the MIS2, due the input of very radiogenic volcanic material (Roberts and Piotrowski, 2015). However, the Holocene is not marked by such variations in the Nd isotopic

signature of water masses (Roberts and Piotrowski, 2015). In addition, the ENd records 317 extracted from CWCs of the Mingulay Reef Complex (this study) and the Rockall Trough 318 (Colin et al., 2010) do not seem to be affected by this mechanism, at least not during the late 319 320 Holocene, as an increase in Icelandic glass concentration in the Rockall Trough (0.1-0.6 ka, 1.2-1.5 ka, 2.8-2.9 ka, 3.3-3.4 ka, 4.0-4.1 ka; Bond et al., 2001) is not linked to systematically 321 higher ENd values. Overall, a strong contribution of Nd input from volcanic material can 322 probably be excluded, suggesting that CWCs from the Mingulay Reef Complex record ENd 323 variability in the mixing between SEC and NAC water through time. 324

325 Fresh sub-surface water flowing into the SPG is characterized by unradiogenic ENd values (-326 14 to -16) while subtropical water exhibits more radiogenic values (-10 to -12) (Lacan & Jeandel, 2004, 2005; Lambelet et al., 2016; Dubois-Dauphin et al., 2017). It has been shown 327 that ENd of the NAC was dominated by the subpolar gyre signature due to higher Nd 328 concentrations in the subpolar surface waters compared to the northern subtropical gyre 329 waters (Lambelet et al., 2016; Dubois-Dauphin et al., 2017). Therefore, when the subpolar 330 331 gyre extension is enhanced, NAC seawater is expected to be even more unradiogenic than today. Conversely, during times of westward contraction of the subpolar gyre, a stronger 332 influence of temperate subtropical water flowing northwards along the European margin 333 334 could increase the ENd value at the study site. ENd record of the Mingulay Reef Complex can be associated to the spatial evolution of the interface between the SPG waters and the 335 ENACW carried northwards by the SEC. Thus, lower ENd recorded (2.8 ka) in the subsurface 336 Mingulay Reef Complex may reflect periods of intense, eastward extension of water from the 337 SPG, while higher ENd (3.4 ka) should point to a westward contraction of the SPG and a 338 339 subsequent stronger influence of the STG waters carried by the slope current and/or water from interior Seas. 340

- 341
- 5.2.Hydrological and climate implications of Nd isotope variations

U-series dating and radiocarbon ages (Douarin et al., 2016) have been obtained for CWC 342 343 samples from the Mingulay Reef Complex investigated in this study, thereby allowing us to calculate Δ^{14} C of the water (Douarin et al., 2016) so as to track changes in the circulation of 344 345 sub-surface water masses during the Holocene. The Marine Reservoir Effect (MRE) of the NAC and derived branches averages about 400 years (Stuiver et al., 1998). However, the 346 fresher water flowing into the SPG is depleted in radiocarbon compared to the NAC, 347 displaying a MRE between 450 and 500 years (Eiríksson et al., 2011; Franke et al., 2008). 348 Along the western European margin, in the inter-gyre area, the mean MRE for sub-surface 349 water has been estimated at 380±60 years (Tisnerat-Laborde et al., 2010). Consequently, high 350 351 MRE in the Mingulay CWCs has been interpreted as indicating a stronger influence of subpolar water (Douarin et al., 2016), linked to the extension of the SPG (Hatun et al., 2005). 352

The ϵ Nd record obtained in this study co-varies with the reconstructed MRE values for the Mingulay CWCs (Douarin et al., 2016), as older (younger) MRE is synchronous with more (less) radiogenic ϵ Nd (Fig. 5). At 3.4, the coral ϵ Nd record shifts towards more radiogenic values (~ -11.8) associated with an increase in MRE (up to 592±41 years). Conversely, low MRE values (down to 207±38 years) occur at about 2.8 ka and are associated with more unradiogenic values (~ -14.5) (Fig.3 and 5).

359 As STG water exhibits more radiogenic values than water flowing in the SPG (see section 5.1), a shift toward more radiogenic ENd values at Mingulay points to a weakened SPG, 360 promoting the northeastward advection of subtropical water to the Rockall Trough and the 361 Mingulay Reef Complex. This conclusion is inconsistent with the previous interpretation of 362 MRE variations in Mingulay corals (Douarin et al., 2016). Alternatively, MRE variations 363 364 could be interpreted as changes in gas exchange rates between the atmosphere and the surface mixing layer (Ascough et al., 2009; Tisnérat-Laborde et al., 2010; Wanamaker et al., 2012). A 365 homogenization of the water column due to enhanced wind stress could result in a younger 366

MRE from Mingulay corals, as previously observed in CWCs from the Bay of Biscay 367 368 (Montero-Serrano et al., 2013). Previous studies have shown that downwelling phenomena occur at the Mingulay Reef Complex (Davies et al., 2009). A long-term influence of 369 370 downwelling on the area of the Mingulay Reef Complex should have resulted in the transfer of rather radiogenic ɛNd surface waters (-11.7±0.2, station MR-5) to depth at times of lower 371 MRE. However, this is not the case as ENd from Mingulay CWCs decreases when the marine 372 age reservoir is low (i.e. at 2.8 ka) and inversely (i.e. at 3.4 ka). Similarly, inputs of Scottish 373 374 fjords and coastal waters, which are likely to result in reduced regional MRE (Stuiver and Braziunas 1993) and higher ENd values (Davies et al., 1985), are not supported by the data 375 trend from Mingulay CWCs. Furthermore, a moderate regional deviation in marine reservoir 376 age (-26±14 years) has been estimated for the Interior Seas (Cage et al., 2006), suggesting a 377 limited influence on radiocarbon variations. Finally, the influences of ¹⁴C-deficient carbon 378 379 derived from humus or carbonate bedrock are likely to be small (Cage et al., 2006). Taken together, all the above evidences suggest that MRE variations recorded in the Mingulay Reef 380 381 Complex are the result of changes in vertical mixing occurring in the NAC and/or SEC and 382 that the source of the young MRE anomaly should be water of subpolar origin.

The more radiogenic ENd value between at 3.4 ka, associated with high MRE, is synchronous 383 with storminess maxima recorded in Icelandic eolian soil deposits (Jackson et al., 2005) (Fig. 384 6). On the basis of several paleostorm activity records, five Holocene storm periods have been 385 defined in the North-East Atlantic during the mid- to late Holocene (5.8-5.5 ka; 4.5-3.9 ka; 386 3.3-2.4 ka; 1.9-1.05 ka and 0.6-0.25 BP) with a frequency of about 1,500 years (Sorrel et al., 387 2012). These storm periods have been shown to coincide with an enhanced upper-water-388 column stratification south of Iceland and increases in temperature and salinity (Sorrel et al., 389 390 2012). Enhanced stratification and changes in eastern Atlantic water properties are consistent with a westward contraction of the SPG accompanied by a stronger northward penetration of 391

warm and saline waters of the STG (Sorrel et al., 2012). In addition, a decrease in the 392 393 temperature gradient of the North Atlantic has been linked to a southward shift of the westerlies and low-pressure systems, leading to southward displacement of the storm tracks 394 395 along the European coasts (Sabatier et al., 2012b; Sorrel et al., 2012). These observations on a multi-millenial timescale are consistent with centennial variations occurring at the Mingulay 396 Reef Complex. Hence, we propose the ENd maximum recorded in the Mingulay Reef 397 Complex at 3.4 ka, coeval with strong storms activity (Fig. 6), indicates a stronger influence 398 of subtropical water in the NAC, leading to a higher stratification of the water column in 399 agreement with older MRE (Fig. 6 and 7). 400

401 Conversely, the minimum εNd value at 2.8 ka, which is in phase with a younger MRE, is not 402 associated with strong storm activity (Fig. 6). It is, however, characterized by periods of 403 elevated Na⁺ in the Greenland Ice Sheet Project 2 (GISP2) ice core which are interpreted as 404 sea salt transported by strengthened westerly winds (O'Brien et al., 1995; Mayewski et al., 405 2004; Jackson et al., 2005) (Fig. 6). Consequently, increased wind intensity at high latitudes 406 of the North Atlantic results in an extension of the SPG leading to more unradiogenic εNd 407 values and lower MRE in the Mingulay Reef Complex (Fig. 7).

The variability in north Atlantic gyre circulation could in turn impact the AMOC dynamics 408 (Oppo et al., 2003; Thornalley et al., 2009). High temporal resolution sea surface temperature 409 (SST) reconstructions for the late Holocene have been investigated in the Nordic Seas (core 410 MD99-2275, Sicre et al., 2008). SST variability in the North-East Atlantic and the Nordic 411 Seas has a strong influence on colder and warmer climate over Europe. Copard et al. (2012) 412 413 have demonstrated that coral ENd shifted towards values indicative of a stronger SPG during the Medieval Climatic Anomaly and a weaker SPG during the Little Ice Age. Similarly, the 414 415 2.8 ka event recorded in CWCs from the Mingulay Reef Complex is coeval with a temperature maximum in the Nordic Seas, while lower SSTs are associated with the 3.4 ka 416

event (Sicre et al., 2008) (Fig. 6). These findings are in agreement with a previous study that 417 shows a linkage between LSW formation, SPG strength and climate periods in North-West 418 Europe (Moffa-Sanchez and Hall, 2017). In addition, the lower ENd from subtler variations of 419 the Mingulay corals are often associated with higher temperatures north of Iceland (see 420 dashed lines in Fig. 6). We conclude that an extension (contraction) of the SPG and the 421 subsequent intensification (weakening) of the AMOC results in a higher (lower) latitudinal 422 heat flux in the North-East Atlantic and the Norwegian Seas (Fig. 7), as has been evidenced 423 during the last millennium (Wanamaker et al., 2012). By comparing SST variability in the 424 Nordic Seas and the subpolar Atlantic, Miettinen et al. (2012) have demonstrated a SST 425 seesaw between the two regions that could be a surface expression of the variability of the 426 NAC. 427

428 Furthermore, there is a link between the growth rate of Mingulay corals and the ENd of the CWCs (Fig. 3). Small decreases in the ENd values are often associated with an increase in the 429 reef growth rate (Fig. 3). The sensitivity of corals and coral reefs to abrupt climatic and 430 oceanic changes has been pointed out in previous studies (Dodds et al., 2007; Davies et al., 431 2008; Eisele et al., 2011; Frank et al., 2011; Thiagarajan et al., 2013; Henry et al., 2014; 432 Hebbeln et al., 2014; Douarin et al., 2014; Bonneau et al., 2018). This suggests that the 433 Mingulay Reef Complex is very sensitive to the origin of the water mass. A higher 434 contribution of water originating from the strongly ventilated SPG (such as that observed 435 around 2.8 ka) could cause favorable conditions for CWC growth. Further investigation will 436 be necessary to explain this relationship between changes in the origin of the water masses 437 438 and coral growth on the Mingulay reef.

439

440 **6.** Conclusion

εNd of seawater and cold-water coral (CWC) samples, collected at depths of 127-134 m in the
Mingulay Reef Complex on the Scottish continental shelf, have been investigated in order to
constrain subpolar gyre (SPG) dynamics over the past 4500 years.

444 εNd values obtained in the upper 30 m water depth collected above the Mingulay Reef Complex permit us to establish, for the first time, the surface Nd isotopic signature of the 445 Scottish continental shelf which is around -11.7±0.2, probably due to the influence of 446 447 radiogenic interior Seas. Below 100 m ENd values vary from -13±0.3 to -12.4±0.2, pointing to a mixing between the Shelf Edge Current (SEC) water and the subpolar water brought by the 448 North Atlantic Current (NAC), as observed in the Porcupine Seabight (Dubois-Dauphin et al., 449 450 2017). An influence of the interior Seas cannot be ruled out. In contrast, water station samples collected on the continental slope display unradiogenic ENd values (from -14.4±0.2 to -451 13.8±0.2), similar to the western Rockall Trough, meaning that NAC water has a strong 452 influence along the continental slope near the study site. 453

εNd values from the Mingulay Reef Complex show two major variations at 2.8 ka and 3.4 ka, 454 which are synchronous with changes in ${}^{14}C$ reservoir age. An unradiogenic ϵNd value of -455 14.5±0.4 at 2.8 ka indicated an eastward extension of the SPG associated with a lower ¹⁴C 456 reservoir age (200 yrs). Conversely, a more radiogenic value at 3.4 ka (-11.8±0.3) pointed to a 457 458 stronger northward penetration of the SEC water along the western European margin, combined with a higher ¹⁴C reservoir age (600 yrs). The link between ɛNd and ¹⁴C records 459 highlights the fact that ¹⁴C reservoir age cannot be used to track water mass origin as it has 460 been previously interpreted (Douarin et al., 2016) but rather vertical ventilations that occurred 461 462 in the NAC and the SEC.

The major negative excursion at 2.8 ka as well as subtler examples recorded in CWCs are systematically associated with warmer climatic conditions in Northern Europe and the Eastern Norwegian Sea linked to an intensification of the surface limb of the AMOC, as has been previously observed for the Medieval Climatic Anomaly (Copard et al., 2012). Such
hydrological conditions have stimulated the growth rate of the Mingulay Reef Complex.

468

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475 **Data availability**

476 Data related to this article are all available in Tables 1 and 2.

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768

769 Figure captions

Fig. 1. Sampling map. The schematic circulations of surface currents are reported by black 770 arrows based on Daniault et al. (2016) and Cage & Austin (2010). NAC, North Atlantic 771 Current; SCC, Scottish Coastal Current; SEC, Shelf Edge Current; SPG, Subpolar Gyre; STG, 772 773 Subtropical Gyre. Seawater ENd values at 100-150m (in white) have been reported from 774 Dubois-Dauphin et al. (2017) and Lambelet et al. (2016). The location of CWCs investigated by Colin et al. (2010) and Copard et al. (2012) in the Rockall Trough are indicated by a green 775 square. Marine cores discussed in the text are indicated by black dots: a. MD99-2275 (Sicre 776 777 et al., 2008); b. RAPiD-12-1K (Thornalley et al., 2009); c. VM29-191 and d. MC52 (Bond et al., 2001). Water stations are indicated by open dots: e. ICE-CTD 03 (Dubois-Dauphin et al., 778 2017); f. ICE-CTD 02 (Dubois-Dauphin et al., 2017); g. Carols Stations (Copard et al., 2011); 779 h. Station 9G (Crocket et al., 2018). Seawater stations MR-4 (orange dot, continental slope) 780

and MR-5 (blue dot, continental shelf) investigated in this study are reported on the insert
map. The Mingulay Reef Complex is indicated by a red square.

Fig. 2. Potential temperature (°C), salinity and εNd value depth profiles for stations MR-4
(continental slope) and MR-5 (continental shelf).

Fig. 3. (a) Marine Reservoir Effect (MRE) reconstructed from Mingulay Reef Complex 785 CWCs (Douarin et al., 2016); (b) ENd record from the Mingulay Reef Complex CWCs (blue 786 787 dots, this study) and from the Rockall Trough (green squares, Colin et al., 2010; Copard et al., 2012). The blue star corresponds to the present day ENd value for station MR-5 at depth of the 788 Mingulay CWCs. The green star corresponds to the present day ENd value for station ICE-789 CTD 03 (Dubois-Dauphin et al., 2017) at depth of the Rockall CWCs. (c) Reef growth rates 790 estimated from downcore +56-08/930VE (pink) and downcore +56-08/929VE (orange) U-791 792 series chronologies; grey dots indicate ages for seabed surface Lophelia dated by radiocarbon and/or U-series (Douarin et al., 2013). 793

- Fig. 4. Seawater εNd profiles for stations MR-4 and MR-5 compared with previous ones
 published for the Rockall Trough and the Porcupine Seabight (Dubois-Dauphin et al., 2017),
 as well as for the Bay of Biscay (Copard et al., 2011)
- Fig. 5. Crossplot of MRE (Douarin et al., 2016) versus εNd (this study) of the Mingulay Reef
 Complex.

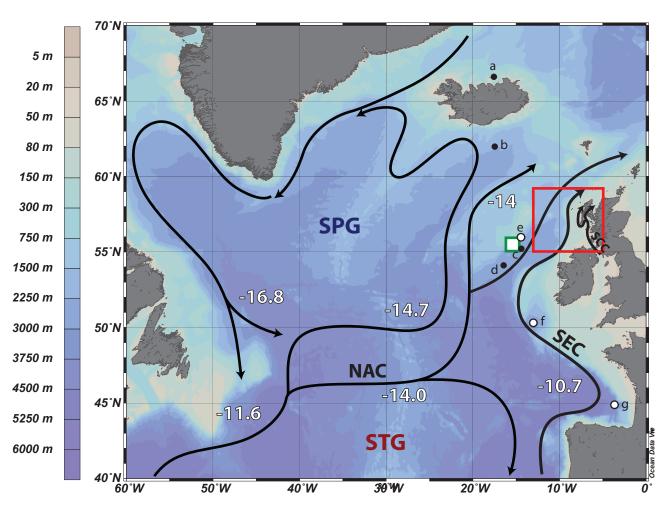
Fig. 6. (a) High resolution Sea surface temperature (in °C) derived from alkenone paleothermometry (MD99-2275; Sicre et al., 2008). Temporal resolution of the blue curve ranges from 2 to 5 years. The superimposed red curve is a 5 points running mean; (b) ϵ Nd record from the Mingulay Reef Complex CWCs (blue dots, this study) and from the Rockall Trough (green squares, Colin et al., 2010; Copard et al., 2012) (c) MRE reconstructed from Mingulay Reef Complex CWCs (Douarin et al., 2016); (d) Sea surface temperatures at surface (red line) and at thermocline level (blue line) (RAPiD12-1K ; Thornalley et al., 2009).(e)
Mean grain size (µm) measured in south Icelandic loess (Jackson et al., 2005). (f) Greenland
Ice Sheet Project 2 sea-salt sodium as a proxy of westerly winds (O'Brien et al., 1995). MCA:
Medieval Climatic Anomaly. Dashed lines underline warm events in North Iceland.

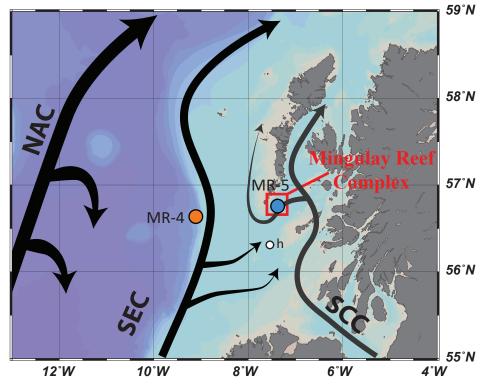
Fig. 7. Schematic representations of the dynamic of sub-surface circulation in the NorthAtlantic during the 2.8 event (a) and 3.4 event (b).

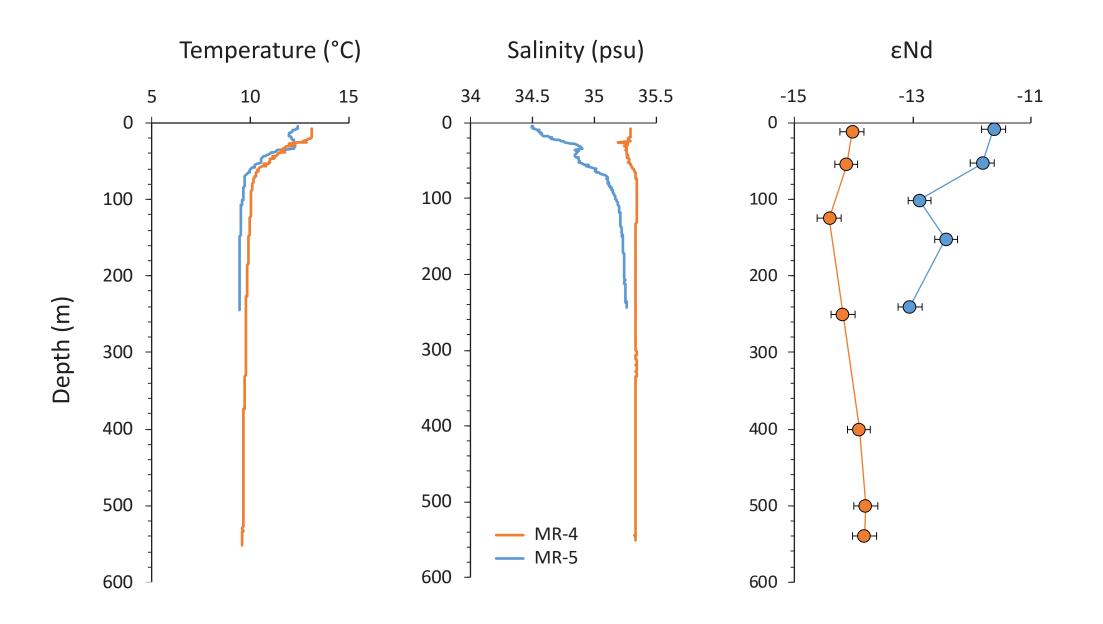
Table 1. ε Nd values obtained for cold-water corals from the Mingulay Reef Complex. Internal error (2 SE) corresponds to error measurement based on 90 cycles of measurements. External error (2 SD) corresponds to external reproducibility (2 σ standard deviation) derived from repeated measurements of La Jolla standard for each analytical session. When internal error is larger than external error, internal error is used in the figures.

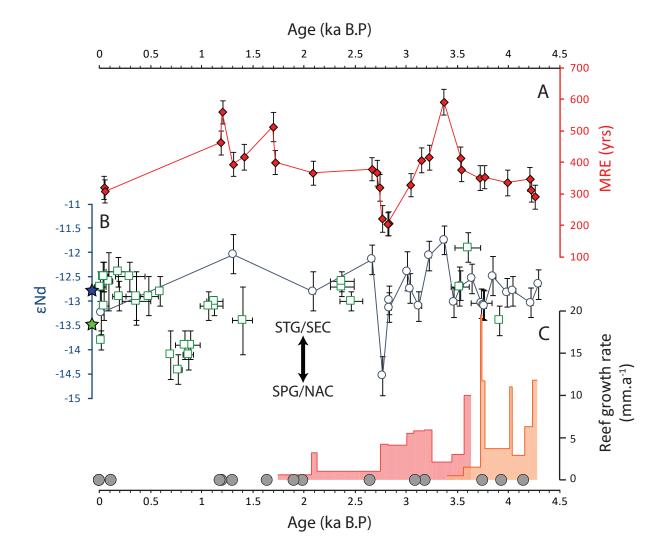
Table 2. Locations, water depth (m), potential temperature (θ , °C), salinity (psu), potential density (σ , kg.m-3), ¹⁴³Nd/¹⁴⁴Nd and ε Nd values of seawater samples investigated in this study. Error bars reported for ε Nd values correspond to external reproducibility (2σ standard deviation) derived from repeated measurements of La Jolla standard for each analytical session.

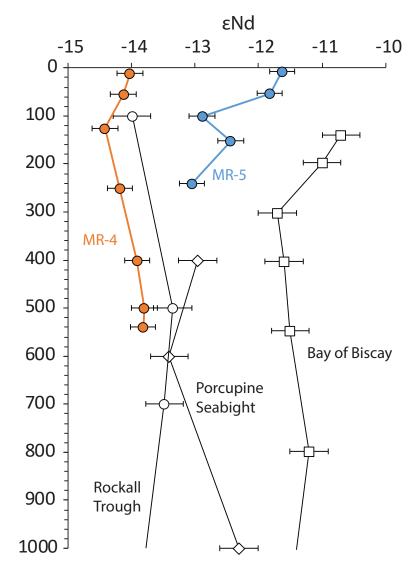
821



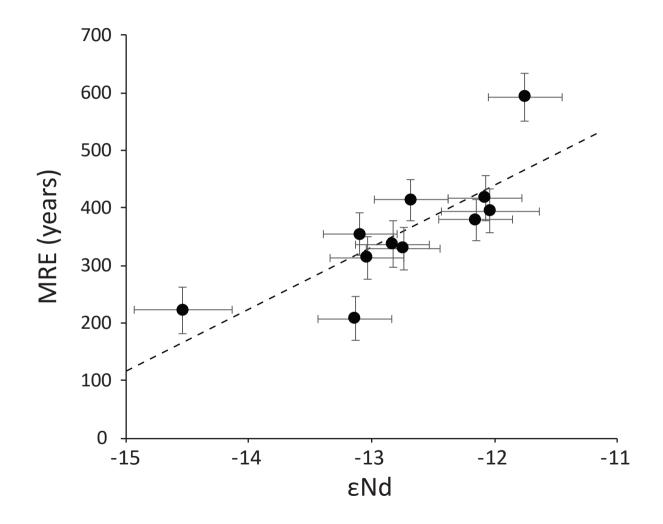


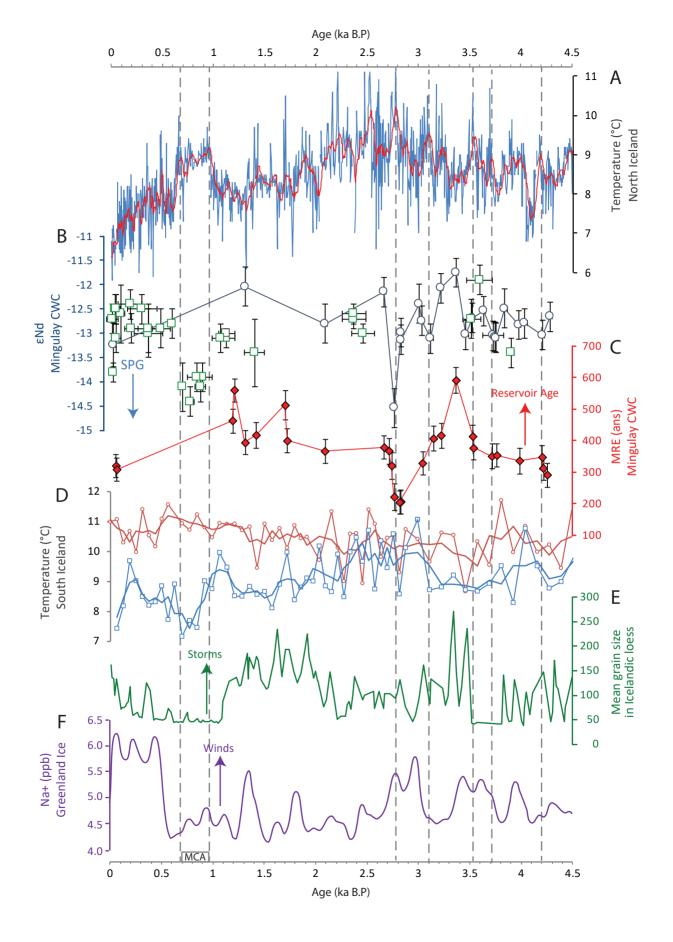


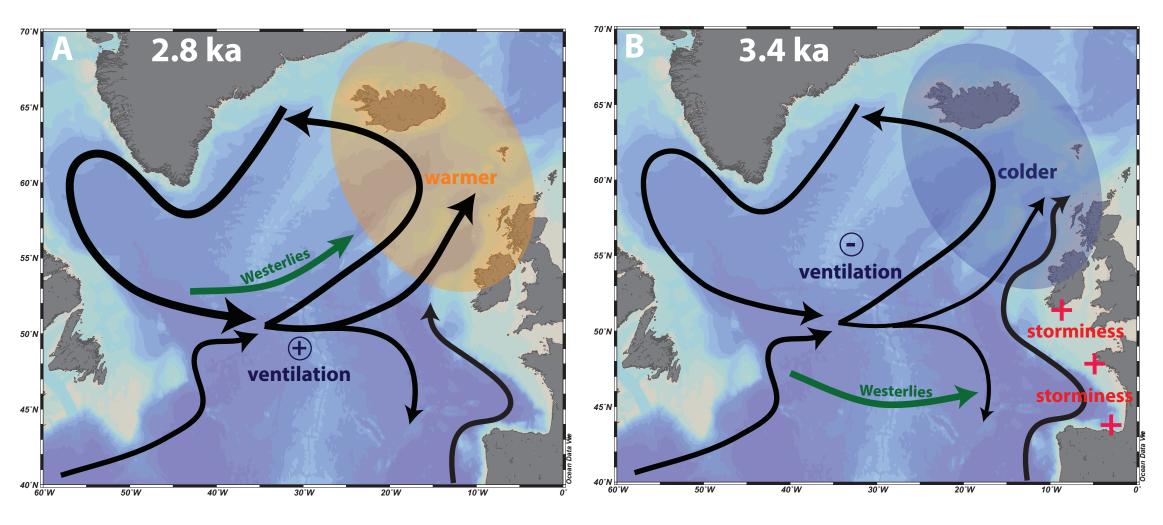




Water depth (m)







Sample Name	Latitude	Longitude	Depth (m)	Age (yrs)	¹⁴³ Nd/ ¹⁴⁴ Nd	error (2 SE)	εNd	error (2 SE)	error (2 SD)
GRAB 15.5.5.10	56°47'08" N	7°25'48" W	seafloor	21 ±13	0.511959	±0.000023	-13.24	±0.45	±0.40
GRAB 1151	56°49'08" N	7°23'36" W	seafloor	1312 ±11	0.512021	±0.000021	-12.04	±0.40	±0.40
930 A6 42-45	56°49′20″ N	7°23′47″ W	134	2088 ±18	0.511982	±0.000018	-12.80	±0.36	±0.40
GRAB 56.08.928	56°47'08" N	7°25'48" W	seafloor	2666 ±14	0.512015	± 0.000008	-12.15	±0.15	±0.30
930 B6 142-146	56°49′20″ N	7°23′47″ W	134	2765 ±16	0.511893	±0.000021	-14.53	±0.40	±0.40
929 A4 100-101	56°49′19″ N	7°23′27″ W	127	2828 ±13	0.511973	± 0.00008	-12.98	±0.15	±0.30
930 B6 134-137	56°49′20″ N	7°23′47″ W	134	2830 ±19	0.511965	±0.000016	-13.13	±0.31	±0.30
930 C6 214-217	56°49′20″ N	7°23′47″ W	134	3012 ±17	0.512003	±0.000018	-12.39	±0.36	±0.40
930 C6 251-254	56°49′20″ N	7°23′47″ W	134	3040 ±11	0.511985	±0.000016	-12.74	±0.31	±0.30
930 D6 388-392	56°49′20″ N	7°23′47″ W	134	3119 ±26	0.511966	± 0.000008	-13.11	±0.15	±0.30
930 D6 356-358	56°49′20″ N	7°23′47″ W	134	3221 ±18	0.512019	±0.000012	-12.07	±0.23	±0.30
929 A4 3-9 cm	56°49′19″ N	7°23′27″ W	127	3367 ±18	0.512036	±0.000011	-11.75	±0.21	±0.30
930 D6 377-379	56°49′20″ N	7°23′47″ W	134	3461 ±27	0.511970	±0.000011	-13.03	±0.22	±0.30
929 A4 12-17	56°49′19″ N	7°23′27″ W	127	3530 ±16	0.511988	± 0.000008	-12.68	±0.16	±0.30
930 F6 505-510	56°49′20″ N	7°23′47″ W	134	3641 ±22	0.511995	± 0.000008	-12.54	±0.15	±0.30
929 A4 38-41	56°49′19″ N	7°23′27″ W	127	3734 ±104	0.511970	±0.000009	-13.04	±0.18	±0.30
929 A4 64-67	56°49′19″ N	7°23′27″ W	127	3747 ±17	0.511968	±0.000015	-13.07	±0.29	±0.30
929 A4 90-91	56°49′19″ N	7°23′27″ W	127	3765 ±13	0.511967	±0.000010	-13.09	±0.19	±0.30
930 E6 459-461	56°49′20″ N	7°23′47″ W	134	3847 ±24	0.511998	±0.000019	-12.49	±0.37	±0.40
929 B4 188-189	56°49′19″ N	7°23′27″ W	134	3986 ±18	0.511980	±0.000012	-12.83	±0.24	±0.30
929 C4 213-216	56°49′19″ N	7°23′27″ W	127	4043 ±23	0.511982	±0.000009	-12.79	±0.18	±0.30
929 C4 284-290	56°49′19″ N	7°23′27″ W	127	4216 ±16	0.511970	±0.000010	-13.03	±0.19	±0.30
929 D4 349-353	56°49′19″ N	7°23′27″ W	127	4287 ±26	0.511989	±0.000011	-12.66	±0.22	±0.30

Table 1

Station	Latitude	Longitude	Depth (m)	θ (°C)	S (psu)	σ (kg.m ⁻³)	¹⁴³ Nd/ ¹⁴⁴ Nd	error (2 SD)	εNd	error (2 SD)
MR-4	56°37.44' N	9°5.48'W	12	13.10	35.29	26.60	0.511919	±0.000010	-14.0	±0.20
MR-4	56°37.44' N	9°5.48'W	55	10.78	35.30	27.05	0.511914	±0.000014	-14.1	±0.30
MR-4	56°37.44' N	9°5.48'W	125	9.99	35.34	27.22	0.511899	±0.000011	-14.4	±0.20
MR-4	56°37.44' N	9°5.48'W	250	9.80	35.34	27.25	0.511911	±0.000012	-14.2	±0.20
MR-4	56°37.44' N	9°5.48'W	400	9.65	35.33	27.27	0.511925	±0.000011	-13.9	±0.20
MR-4	56°37.44' N	9°5.48'W	500	9.63	35.33	27.27	0.511931	±0.000011	-13.8	±0.20
MR-4	56°37.44' N	9°5.48'W	540	9.60	35.33	27.28	0.511930	±0.000012	-13.8	±0.20
MR-5	56°46.04' N	7°25.98'W	8	12.33	34.51	26.15	0.512042	±0.000011	-11.6	±0.20
MR-5	56°46.04' N	7°25.98'W	53	10.39	34.89	26.80	0.512032	±0.000011	-11.8	±0.20
MR-5	56°46.04' N	7°25.98'W	101	9.60	35.18	27.16	0.511977	±0.000012	-12.9	±0.20
MR-5	56°46.04' N	7°25.98'W	152	9.47	35.23	27.22	0.512000	±0.000011	-12.4	±0.20
MR-5	56°46.04' N	7°25.98'W	240	9.43	35.26	27.25	0.511969	±0.000014	-13.0	±0.30

Table 2