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Quentin Dubois-Dauphin, Christophe Colin, Mary Elliot, Arnaud Dapoigny, Eric Douville. Holocene shifts in sub-surface water circulation of the North-East Atlantic inferred from Nd isotopic composition in cold-water corals. *Marine Geology*, 2019, 410, pp.135-145. 10.1016/j.margeo.2019.01.004 . hal-02149410

HAL Id: hal-02149410

<https://hal.science/hal-02149410>

Submitted on 24 Jun 2021

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Holocene shifts in sub-surface water circulation of the North-East Atlantic inferred from Nd isotopic composition in cold-water corals

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Abstract:

Variations of the Sub-Polar Gyre (SPG) and the Sub-Tropical Gyre (STG) circulation during the Holocene are believed to be related to regional and global climate over this time period. To improve our understanding of these phenomena we provide new constraints on variations in surface circulation patterns using neodymium isotopes (ϵNd) on precisely U-Th dated coral fragments of *L. pertusa*. The fragments were retrieved from two sediment cores taken from cold-water coral (CWC) mounds at ~ 127-134 m water depth in the Mingulay Reef Complex located on the Western British continental shelf. The results have been combined with ϵNd analyzed on seawater samples from two stations located on the continental shelf and margin in order to establish whether ϵNd is a reliable proxy of the ocean circulation variations and

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

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

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](#); [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 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 trend and the final disintegration of the residual Laurentide ice sheet during the early Holocene, which induces a pronounced provincialism in the surface physical property records of the North Atlantic (e.g. [de Vernal and Hillaire-Marcel, 2006](#); [Renssen et al., 2009](#)).

It has been suggested that during modern times variations in the shape and position of the 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 on the AMOC strength and the properties of waters entering the Nordic Seas (e.g. [Häkkinen et al., 2013](#)). The NAC follows the boundary between the North Atlantic subpolar gyre (SPG) and the subtropical gyre (STG), and its position, intensity and composition have been modulated by SPG variations over recent decades ([Håtun et al., 2005](#); [Berx and Payne, 2017](#); [Zunino et al., 2017](#)). [Foukal & Lozier \(2018\)](#), by using a combination of empirical orthogonal functions and direct calculations of the size and strength of the SPG, have proposed that the 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 major role of the SPG on longer time scales (see references below). During the early Holocene, a reorganization of the mid-depth (200-800 m) circulation has been highlighted at about 7-6 ka, with a weakened SPG leading to a stronger northward penetration of temperate water masses in the North-East Atlantic ([Thornalley et al., 2009](#); [Colin et al., 2010](#)). Other studies based on cold-water corals from the Bay of Biscay and the Rockall Trough have 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 and 1990, periods characterized by a dominant negative phase of NAO were associated with a stronger influence of mid-depth STG water (Montero-Serrano et al., 2013). Over the last 1500 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 SPG in the North Atlantic (Copard et al., 2012). Conversely, the Little Ice Age (between 1350 AD and 1850 AD), characterized by a persistently negative NAO phase, is associated with the westward contraction of the mid-depth SPG (Copard et al., 2012). Overall, these records have shown that Holocene climatic anomalies are associated with mid-depth SPG dynamics which 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 heat and salt budgets at intermediate depths may have contributed to changes in the properties 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 potential target to track past changes of the SPG extension using the ϵNd proxy because it receives waters from the NAC. In addition, cold-water corals (CWCs) have been shown to act as useful archives for identifying rapid changes in the dynamics of water masses (Lutringer et al., 2005; Colin et al., 2010; van de Flierdt et al., 2010; Copard et al., 2010, 2012; Montero-Serrano et al., 2013; Wilson et al., 2014; Chen et al., 2015; Dubois-Dauphin et al., 2016; Douarin et al., 2016). Firstly, their aragonitic skeleton can be precisely dated thanks to U-series disequilibrium methods (Adkins et al., 1998; Cheng et al., 2000; Douville et al., 2010). 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 characterized by low sedimentation rates that limit our ability to obtain records with centennial resolutions. Furthermore, the Nd isotopic composition, expressed as $\epsilon\text{Nd} =$

$$\left(\frac{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{sample}}}{(^{143}\text{Nd}/^{144}\text{Nd})_{\text{CHUR}}} - 1 \right) \times 10000$$
 (CHUR: Chondritic Uniform Reservoir [Jacobsen and Wasserburg, 1980]), of living and fossil scleractinian CWCs faithfully traces water mass provenance and mixing in the ocean (e.g. van de Flierdt et al., 2010; Copard et al., 2012; Struve et al., 2017). Taking into consideration that mid-depth STG and SPG water masses are characterized by contrasted Nd isotopic signatures ($\epsilon\text{Nd} \approx -10$ and $\epsilon\text{Nd} \approx -15$, respectively; Lambelet et al., 2016; Dubois-Dauphin et al., 2017), CWC ϵNd records obtained 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., 2012; Montero-Serrano et al., 2011, 2013).

In this paper we present a high-resolution ϵNd record for the last 4500 yrs, which is derived from CWCs from the Mingulay Reef Complex, located at the sub-surface under the strong influence of NAC water. Our objective is to track hydrological changes in the SPG dynamics during the Holocene. Ambient seawater ϵNd was also investigated to assess seawater ϵNd of the Western British continental shelf and margin and to verify that ϵNd is a reliable proxy for tracking past changes in the influence of the SPG in this region. This CWC ϵNd record was compared to previously published $\Delta^{14}\text{C}$ performed by Douarin et al. (2016) that was interpreted in terms of changes in water mass origin. It also complements well previous studies based on ϵNd in the Rockall Trough (Colin et al., 2010; Copard et al., 2012) that have highlighted major variations of the mid-depth SPG dynamics throughout the Holocene. Our study permits us to re-interpret $\Delta^{14}\text{C}$ with a proxy of water mass origin and provides new 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.

2. Hydrological setting

Presently, the deepest waters (>100 m) and the waters overlying the outer parts of the shelf of west Scotland are of Atlantic origin (Inall et al., 2009). At the surface, the northward flowing Scottish Coastal Current (SCC) brings cooler and fresher waters from the Irish and Clyde Seas (Ellett and Edwards, 1983) (Fig. 1). The Mingulay Reef Complex (56°50'N; 7°20'W; 100-260 m water depth) is thus primarily bathed by Atlantic water (Dodds et al., 2007). The upper-layer Atlantic water off the Mingulay Reef Complex, carried by the NAC, results from the mixing of saline Eastern North Atlantic Central Water (ENACW) originating from the Bay of Biscay (Ellett and Martin, 1973; Ellett et al., 1986; Pollard et al., 1996) and the fresher Modified North Atlantic Water (MNAW) originating in the North-West Atlantic (New and 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 Intermediate Water (SAIW). The SAIW is characterized by low temperatures and salinity and 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, 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 Iceland Basin and the Irminger Sea (Fig. 1). The southern branch penetrates into the Rockall Trough (Fig. 1)

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).

3. Material and methods

3.1.1. CWC samples

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

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).

3.1.2. Seawater samples

During the R/V Atalante MINGULAY-ROCKALL cruise in June 2016, 12 seawater samples were collected at 2 full water column stations MR-4 ($56^{\circ}37.44'$ N; $9^{\circ}5.48'$ W) and MR-5 ($56^{\circ}46.04'$ N; $7^{\circ}25.98'$ W) (Fig. 1, Table 2). These samples, located on the Western British continental shelf (station MR5) and margin (station MR4), were collected in order to assess seawater ϵ Nd flowing around the Mingulay Reef Complex. The seawater samples and water column hydrographic parameters (temperature and salinity) were collected using a CTD-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 μ m AcroPak 500 capsule filters and transferred to 10-liter acid-cleaned bottles, and immediately acidified to a pH of 2 using suprapur 2 N HCl.

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 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 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 described in detail by Copard et al. (2010). This purification process and the measurement techniques applied follow approved GEOTRACES protocols and GEOPS laboratory participated in the international GEOTRACES intercalibration study (van de Flierdt et al., 2012).

The ¹⁴³Nd/¹⁴⁴Nd ratios of seawater samples and of the CWCs from the Mingulay Reef Complex, were analyzed using a Thermo Scientific Neptune Plus MC-ICPMS installed at the 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 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 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 Jolla solutions were analyzed at concentrations similar to those of the samples (4-10 ppb). The 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

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

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, ϵNd 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).

4.2. Cold Water Coral samples

Overall, CWC ϵNd 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 ϵNd 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).

5. Discussion

5.1. Significance of the ϵNd record

The youngest CWC sample collected in the Mingulay Reef complex, with a modern age of 0.02 ± 0.013 ka BP (Douarin et al., 2016), displays an ϵNd 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 study site (between -12.9 ± 0.2 and -12.4 ± 0.2 at 101-152 m water depth; station MR-5) (Fig. 2 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 with seawater analyzed in the Rockall Trough (ICE-CTD 03; Dubois-Dauphin et al., 2017) 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 continental slope near the study site. In contrast, station MR-5, located on the continental shelf, exhibits more radiogenic values at all depths compared to the continental slope (MR-4) and the Rockall Trough (ICE-CTD 03; Dubois-Dauphin et al., 2017). The ϵNd value of warm and saline surface water (upper ENACW) flowing in the SEC is poorly constrained along the 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 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 strong influence on the Mingulay Reef Complex as seen at station MR-5.

Nevertheless, Nd isotopic signatures for the SCC that circulates from the Irish Sea and the Firth of Clyde are unknown. However, rocks and river sediment samples from the northern British Isles display a mean ϵNd value of -10.6 ± 0.4 (Davies et al., 1985), suggesting a rather 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 (station 9G, 166 m bottom depth; Fig. 1) located close to the Mingulay Reef Complex suggesting possible local desorption from resuspended sediments or pore water release to the overlying water column. Seawater ϵNd 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 ϵNd 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 $10\text{-}12^\circ\text{C}$ and $34.5\text{-}34.9$, respectively (Fig. 2). This suggests that surface water is likely affected by local inputs from the Scottish shelf and/or the interior Seas. On the other hand, the ϵNd 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 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 and NAC waters. Furthermore, the initial $\delta^{234}\text{U}$ values of the samples considered in this study ranged from 142.9 ± 2.4 to 150.0 ± 3.5 (2σ uncertainties; Douarin et al., 2013), which is in the range of the modern seawater value (146.8 ± 0.1 ; Andersen et al., 2010), while ^{232}Th values for all corals were low ($< 0.48 \pm 0.07$ ppb; Douarin et al., 2013). These arguments do not support an influence of a local source on CWC ϵNd from the Mingulay Reef Complex. However, the desorption from resuspended sediments cannot be ruled out.

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 correspondence between samples of the same age in both sites (Fig. 3). Seawater ϵNd investigated in the Rockall Trough does not indicate significant variations of Nd isotopic 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 relatively strong homogeneity of the Nd isotopic composition of the surface-to-mid water masses of the Rockall Trough. On the other hand, station MR-5 displayed more radiogenic 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 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 margin.

The ϵNd 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 ϵNd 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 ϵNd 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 ϵNd records extracted from CWCs of the Mingulay Reef Complex (this study) and the Rockall Trough (Colin et al., 2010) do not seem to be affected by this mechanism, at least not during the late 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 higher ϵNd values. Overall, a strong contribution of Nd input from volcanic material can probably be excluded, suggesting that CWCs from the Mingulay Reef Complex record ϵNd variability in the mixing between SEC and NAC water through time.

Fresh sub-surface water flowing into the SPG is characterized by unradiogenic ϵNd values (-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 that ϵNd of the NAC was dominated by the subpolar gyre signature due to higher Nd concentrations in the subpolar surface waters compared to the northern subtropical gyre waters (Lambelet et al., 2016; Dubois-Dauphin et al., 2017). Therefore, when the subpolar 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 influence of temperate subtropical water flowing northwards along the European margin could increase the ϵNd value at the study site. ϵNd record of the Mingulay Reef Complex can be associated to the spatial evolution of the interface between the SPG waters and the ENACW carried northwards by the SEC. Thus, lower ϵNd recorded (2.8 ka) in the subsurface Mingulay Reef Complex may reflect periods of intense, eastward extension of water from the SPG, while higher ϵNd (3.4 ka) should point to a westward contraction of the SPG and a subsequent stronger influence of the STG waters carried by the slope current and/or water from interior Seas.

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 samples from the Mingulay Reef Complex investigated in this study, thereby allowing us to calculate $\Delta^{14}\text{C}$ of the water (Douarin et al., 2016) so as to track changes in the circulation of 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 fresher water flowing into the SPG is depleted in radiocarbon compared to the NAC, displaying a MRE between 450 and 500 years (Eiríksson et al., 2011; Franke et al., 2008). Along the western European margin, in the inter-gyre area, the mean MRE for sub-surface water has been estimated at 380 ± 60 years (Tisnerat-Laborde et al., 2010). Consequently, high 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).

The ϵNd 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 ϵNd (Fig. 5). At 3.4, the coral ϵNd 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).

As STG water exhibits more radiogenic values than water flowing in the SPG (see section 5.1), a shift toward more radiogenic ϵNd values at Mingulay points to a weakened SPG, promoting the northeastward advection of subtropical water to the Rockall Trough and the Mingulay Reef Complex. This conclusion is inconsistent with the previous interpretation of MRE variations in Mingulay corals (Douarin et al., 2016). Alternatively, MRE variations 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 homogenization of the water column due to enhanced wind stress could result in a younger

367 MRE from Mingulay corals, as previously observed in CWCs from the Bay of Biscay
 368 (Montero-Serrano et al., 2013). Previous studies have shown that downwelling phenomena
 369 occur at the Mingulay Reef Complex (Davies et al., 2009). A long-term influence of
 370 downwelling on the area of the Mingulay Reef Complex should have resulted in the transfer
 371 of rather radiogenic ϵNd surface waters (-11.7 ± 0.2 , station MR-5) to depth at times of lower
 372 MRE. However, this is not the case as ϵNd from Mingulay CWCs decreases when the marine
 373 age reservoir is low (i.e. at 2.8 ka) and inversely (i.e. at 3.4 ka). Similarly, inputs of Scottish
 374 fjords and coastal waters, which are likely to result in reduced regional MRE (Stuiver and
 375 Braziunas 1993) and higher ϵNd values (Davies et al., 1985), are not supported by the data
 376 trend from Mingulay CWCs. Furthermore, a moderate regional deviation in marine reservoir
 377 age (-26 ± 14 years) has been estimated for the Interior Seas (Cage et al., 2006), suggesting a
 378 limited influence on radiocarbon variations. Finally, the influences of ^{14}C -deficient carbon
 379 derived from humus or carbonate bedrock are likely to be small (Cage et al., 2006). Taken
 380 together, all the above evidences suggest that MRE variations recorded in the Mingulay Reef
 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.
 383 The more radiogenic ϵNd value between at 3.4 ka, associated with high MRE, is synchronous
 384 with storminess maxima recorded in Icelandic eolian soil deposits (Jackson et al., 2005) (Fig.
 385 6). On the basis of several paleostorm activity records, five Holocene storm periods have been
 386 defined in the North-East Atlantic during the mid- to late Holocene (5.8-5.5 ka; 4.5-3.9 ka;
 387 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.,
 388 2012). These storm periods have been shown to coincide with an enhanced upper-water-
 389 column stratification south of Iceland and increases in temperature and salinity (Sorrel et al.,
 390 2012). Enhanced stratification and changes in eastern Atlantic water properties are consistent
 391 with a westward contraction of the SPG accompanied by a stronger northward penetration of

warm and saline waters of the STG (Sorrel et al., 2012). In addition, a decrease in the 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 along the European coasts (Sabatier et al., 2012b; Sorrel et al., 2012). These observations on a multi-millennial timescale are consistent with centennial variations occurring at the Mingulay Reef Complex. Hence, we propose the ϵNd maximum recorded in the Mingulay Reef Complex at 3.4 ka, coeval with strong storms activity (Fig. 6), indicates a stronger influence of subtropical water in the NAC, leading to a higher stratification of the water column in agreement with older MRE (Fig. 6 and 7).

Conversely, the minimum ϵNd value at 2.8 ka, which is in phase with a younger MRE, is not associated with strong storm activity (Fig. 6). It is, however, characterized by periods of elevated Na^+ in the Greenland Ice Sheet Project 2 (GISP2) ice core which are interpreted as sea salt transported by strengthened westerly winds (O'Brien et al., 1995; Mayewski et al., 2004; Jackson et al., 2005) (Fig. 6). Consequently, increased wind intensity at high latitudes of the North Atlantic results in an extension of the SPG leading to more unradiogenic ϵNd 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 (Oppo et al., 2003; Thornalley et al., 2009). High temporal resolution sea surface temperature (SST) reconstructions for the late Holocene have been investigated in the Nordic Seas (core MD99-2275, Sicre et al., 2008). SST variability in the North-East Atlantic and the Nordic Seas has a strong influence on colder and warmer climate over Europe. Copard et al. (2012) have demonstrated that coral ϵNd shifted towards values indicative of a stronger SPG during the Medieval Climatic Anomaly and a weaker SPG during the Little Ice Age. Similarly, the 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

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

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

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.

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 Scottish continental shelf which is around -11.7 ± 0.2 , probably due to the influence of radiogenic interior Seas. Below 100 m ϵNd 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 North Atlantic Current (NAC), as observed in the Porcupine Seabight (Dubois-Dauphin et al., 2017). An influence of the interior Seas cannot be ruled out. In contrast, water station samples collected on the continental slope display unradiogenic ϵNd values (from -14.4 ± 0.2 to -13.8 ± 0.2), similar to the western Rockall Trough, meaning that NAC water has a strong influence along the continental slope near the study site.

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

Acknowledgements

The research leading to this paper received funding from the National Research Agency L-IPSL project (Grant ANR-10-LABX-0018) and the HAMOC project (Grant ANR-13-BS06-0003). We gratefully acknowledge the support provided by Louise Bordier during Nd isotopic composition analyses. We thank the three reviewers for their helpful and insightful comments. We also thank Marc de Batist for his editorial handling of the manuscript.

Data availability

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

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Figure captions

Fig. 1. Sampling map. The schematic circulations of surface currents are reported by black arrows based on Daniault et al. (2016) and Cage & Austin (2010). NAC, North Atlantic Current; SCC, Scottish Coastal Current; SEC, Shelf Edge Current; SPG, Subpolar Gyre; STG, Subtropical Gyre. Seawater ϵNd values at 100-150m (in white) have been reported from 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 square. Marine cores discussed in the text are indicated by black dots: a. MD99-2275 (Sicre 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., 2017); f. ICE-CTD 02 (Dubois-Dauphin et al., 2017); g. Carols Stations (Copard et al., 2011); h. Station 9G (Crocket et al., 2018). Seawater stations MR-4 (orange dot, continental slope)

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 ($^{\circ}\text{C}$), 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 CWCs (Douarin et al., 2016); (b) ϵNd 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). The blue star corresponds to the present day ϵNd value for station MR-5 at depth of the Mingulay CWCs. The green star corresponds to the present day ϵNd value for station ICE-CTD 03 (Dubois-Dauphin et al., 2017) at depth of the Rockall CWCs. (c) Reef growth rates estimated from downcore +56-08/930VE (pink) and downcore +56-08/929VE (orange) U-series chronologies; grey dots indicate ages for seabed surface *Lophelia* dated by radiocarbon and/or U-series (Douarin et al., 2013).

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.

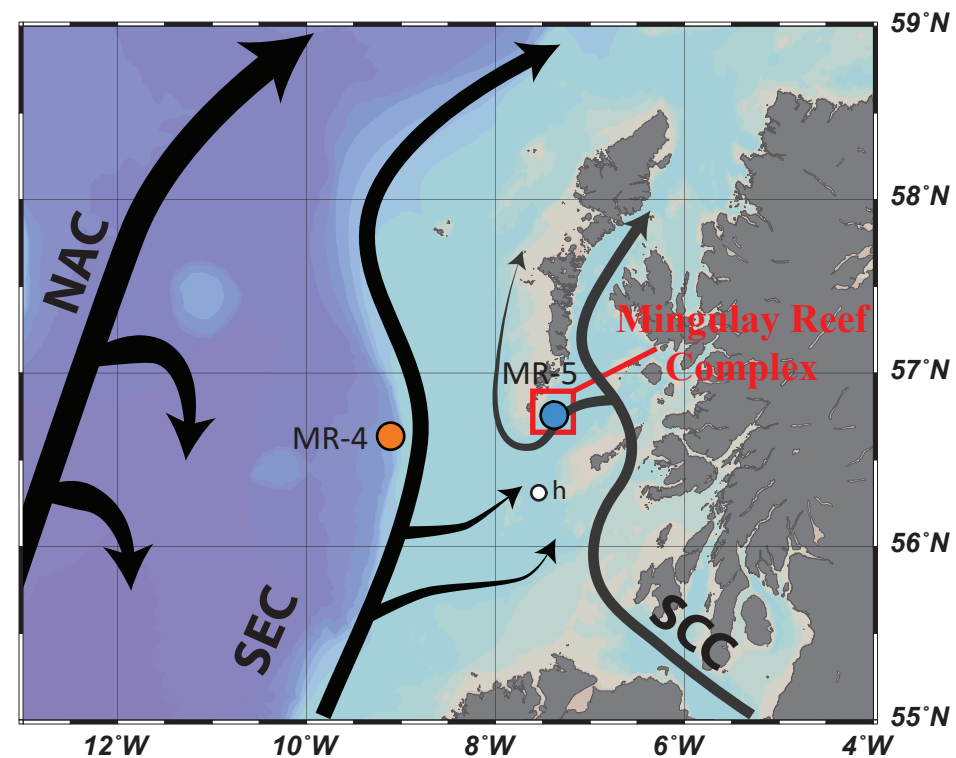
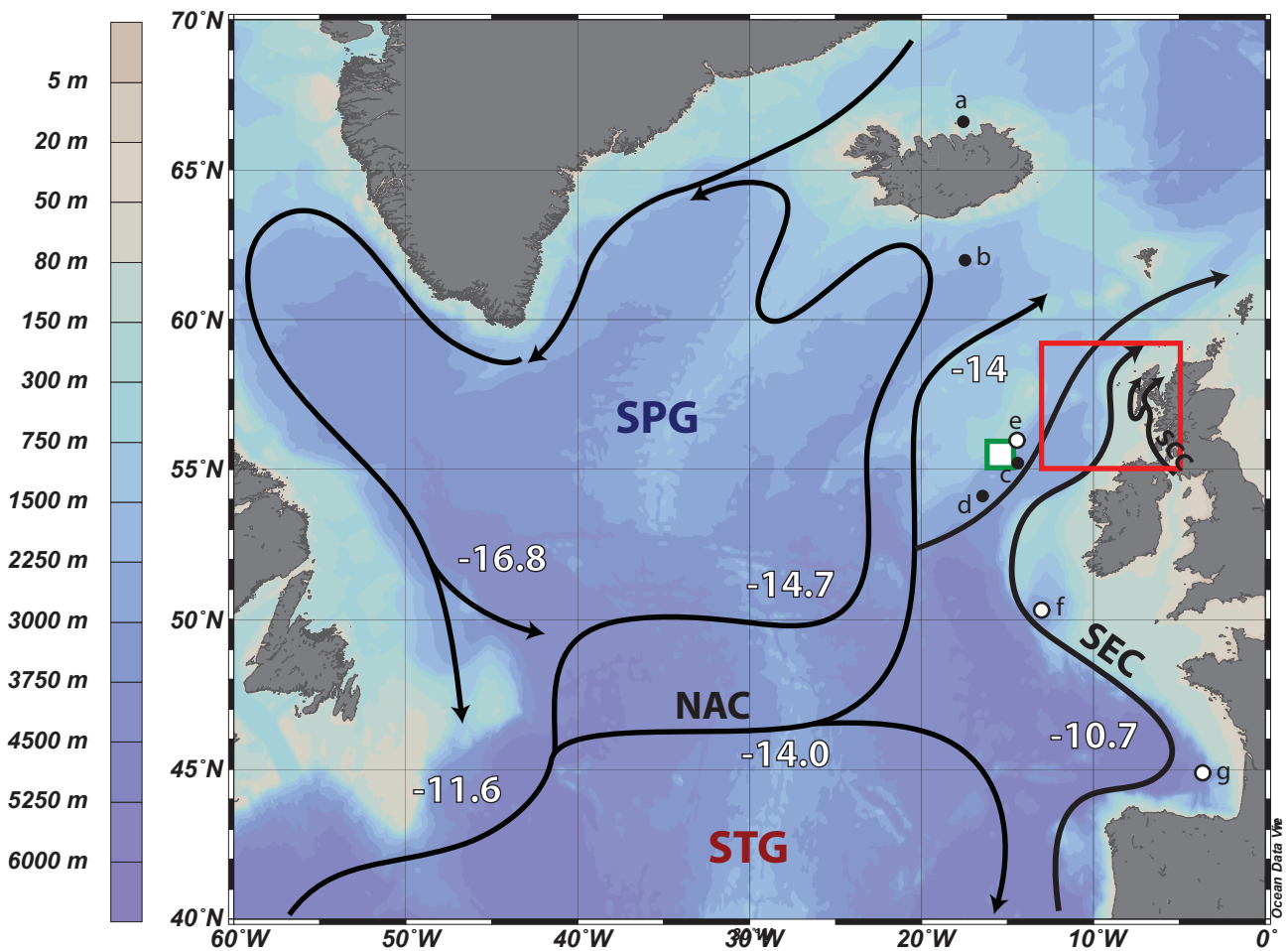
Fig. 6. (a) High resolution Sea surface temperature (in $^{\circ}\text{C}$) derived from alkenone paleothermometry (MD99-2275; Sicre et al., 2008). Temporal resolution of the blue curve ranges from 2 to 5 years. The superimposed red curve is a 5 points running mean; (b) ϵNd record from the Mingulay Reef 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

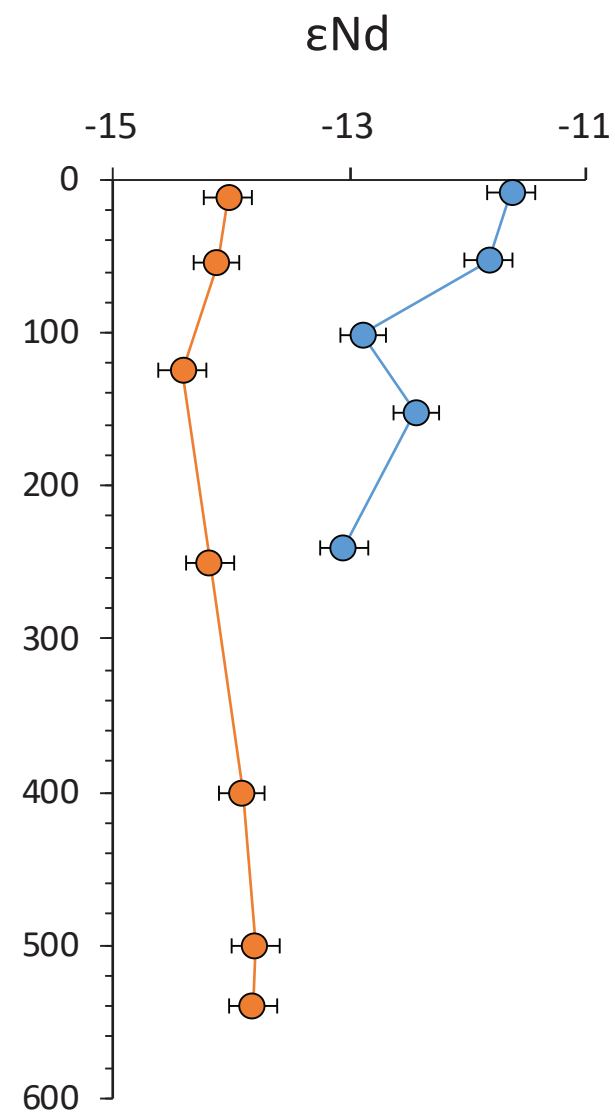
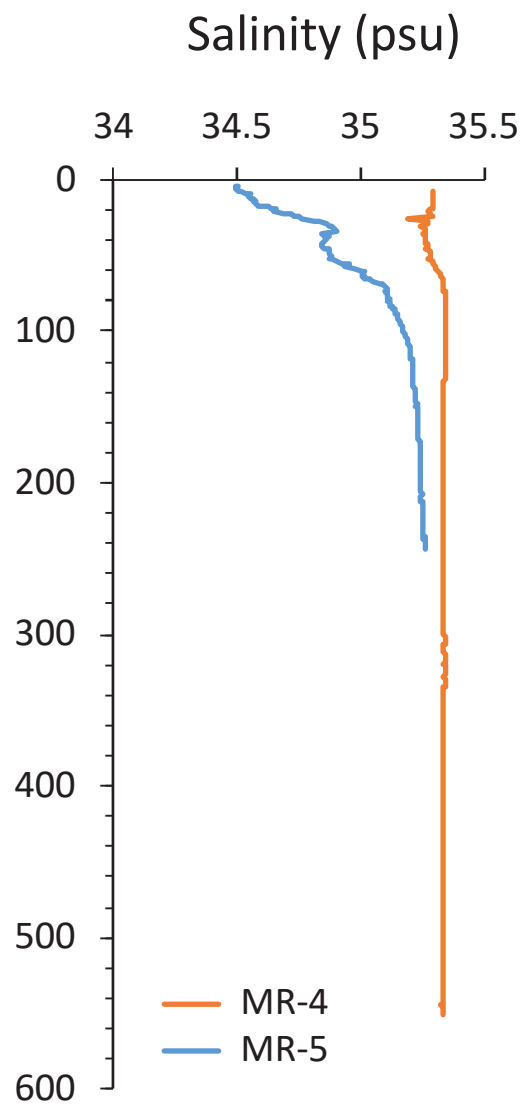
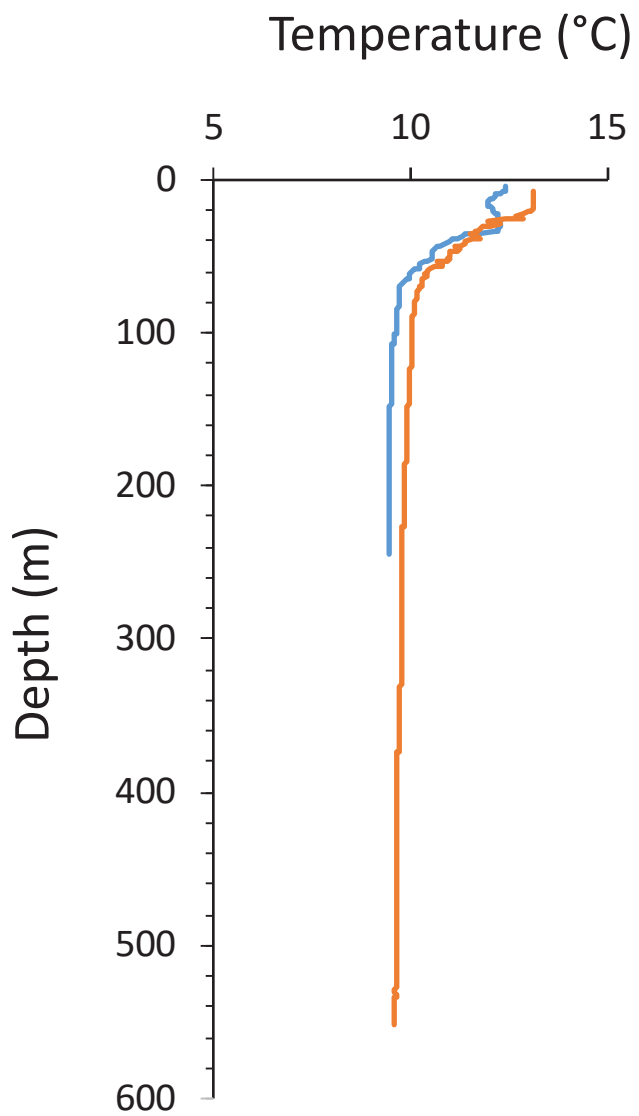
(red line) and at thermocline level (blue line) (RAPiD12-1K ; Thornalley et al., 2009).(e)
Mean grain size (μm) measured in south Icelandic loess (Jackson et al., 2005). (f) Greenland
Ice Sheet Project 2 sea-salt sodium as a proxy of westerly winds (O'Brien et al., 1995). MCA:
Medieval Climatic Anomaly. Dashed lines underline warm events in North Iceland.

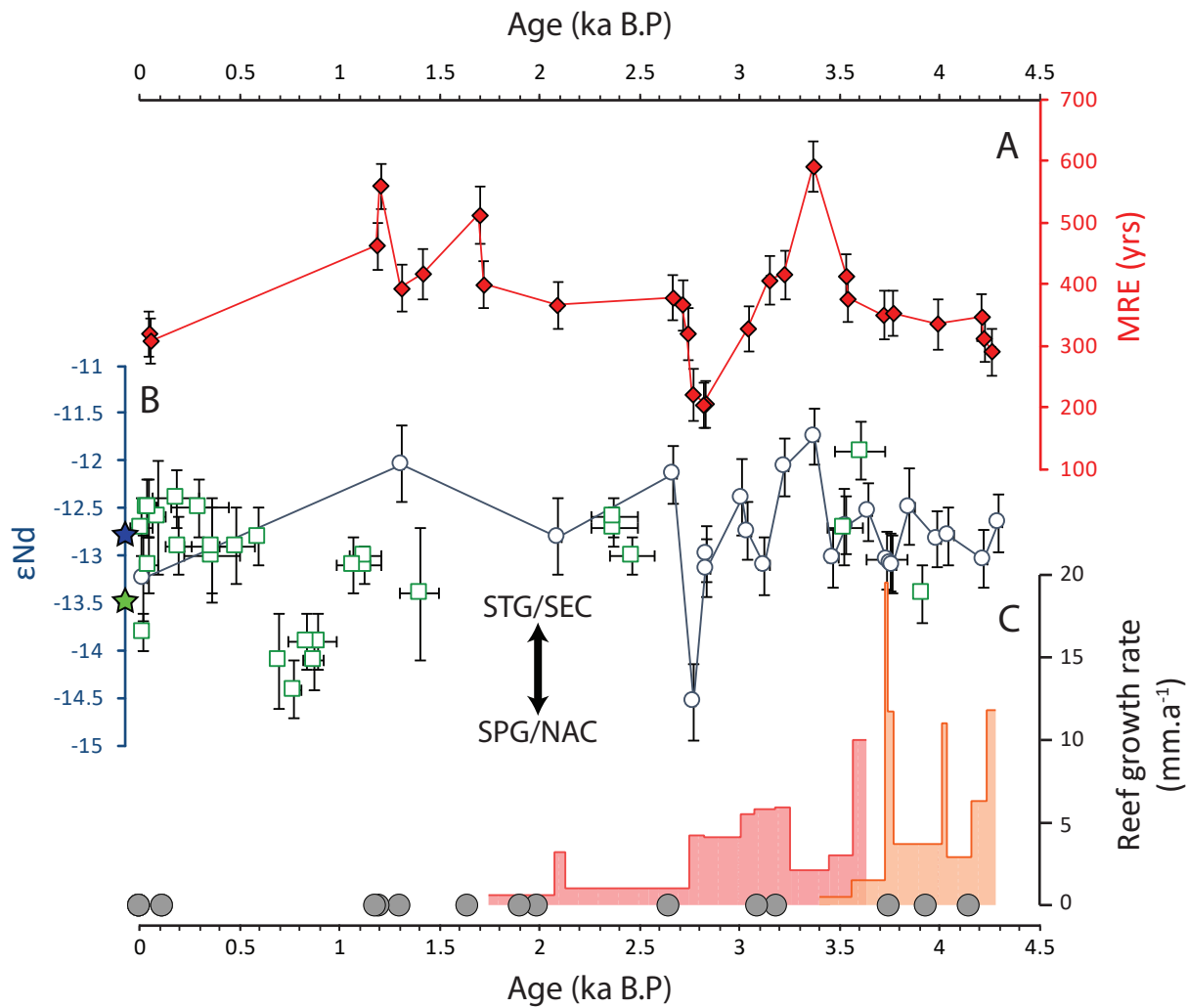
Fig. 7. Schematic representations of the dynamic of sub-surface circulation in the North
Atlantic during the 2.8 event (a) and 3.4 event (b).

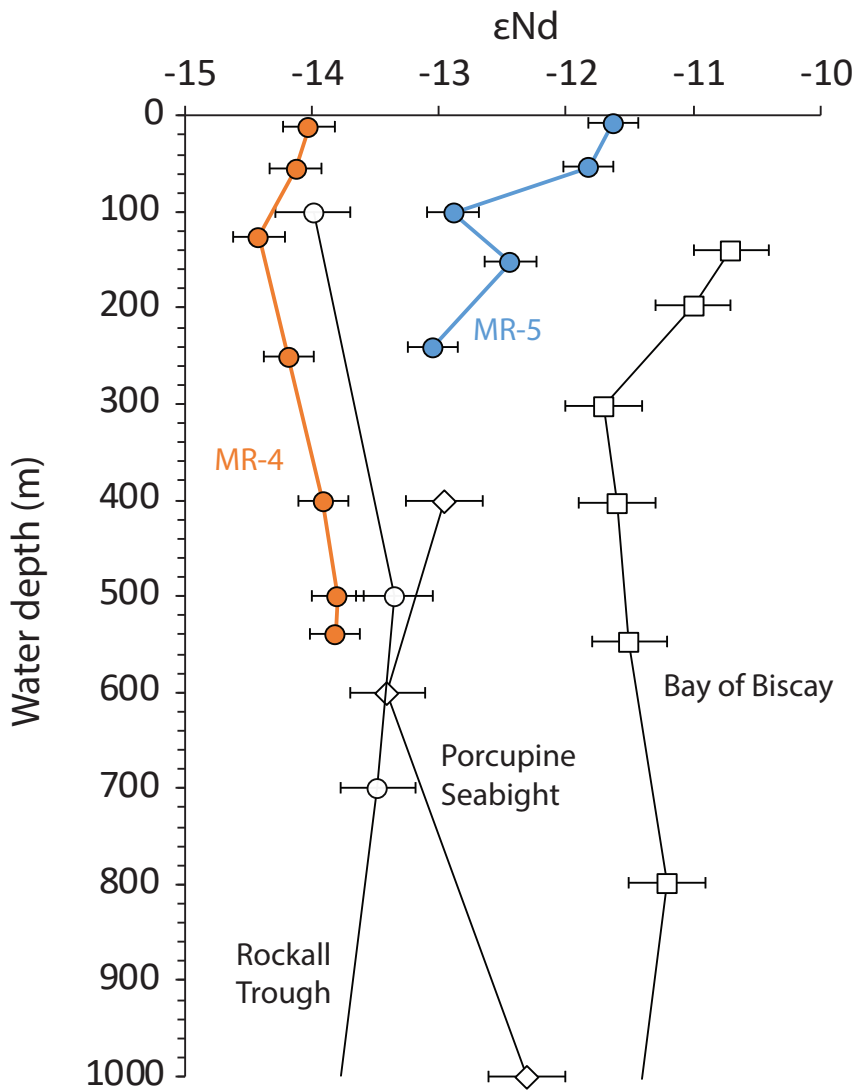
Table 1. ϵNd values obtained for cold-water corals from the Mingulay Reef Complex.
Internal error (2 SE) corresponds to error measurement based on 90 cycles of measurements.
External error (2 SD) corresponds to external reproducibility (2σ standard deviation) derived
from repeated measurements of La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

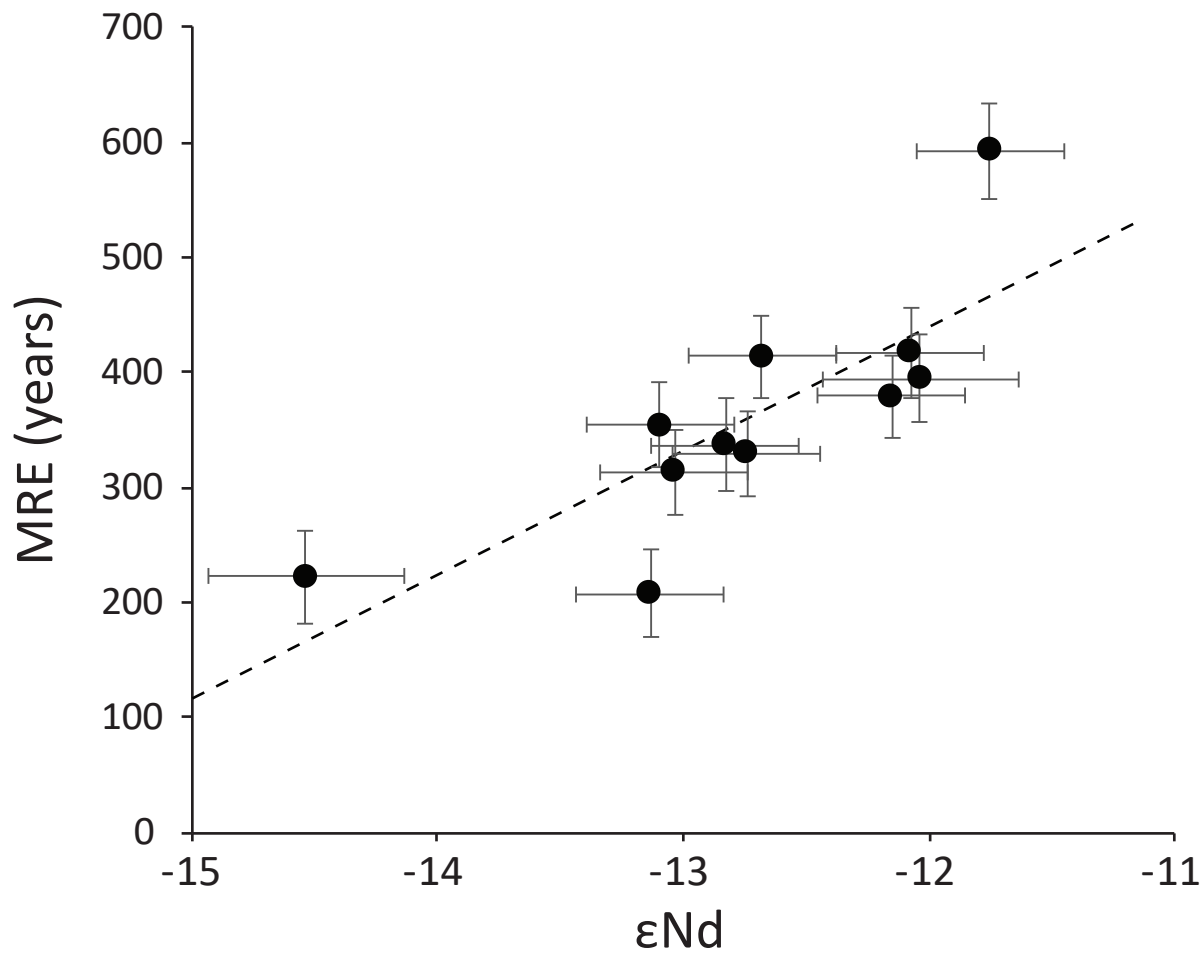
Table 2. Locations, water depth (m), potential temperature (θ , $^{\circ}\text{C}$), salinity (psu), potential
density (σ , kg.m^{-3}), $^{143}\text{Nd}/^{144}\text{Nd}$ and ϵNd values of seawater samples investigated in this
study. Error bars reported for ϵNd values correspond to external reproducibility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

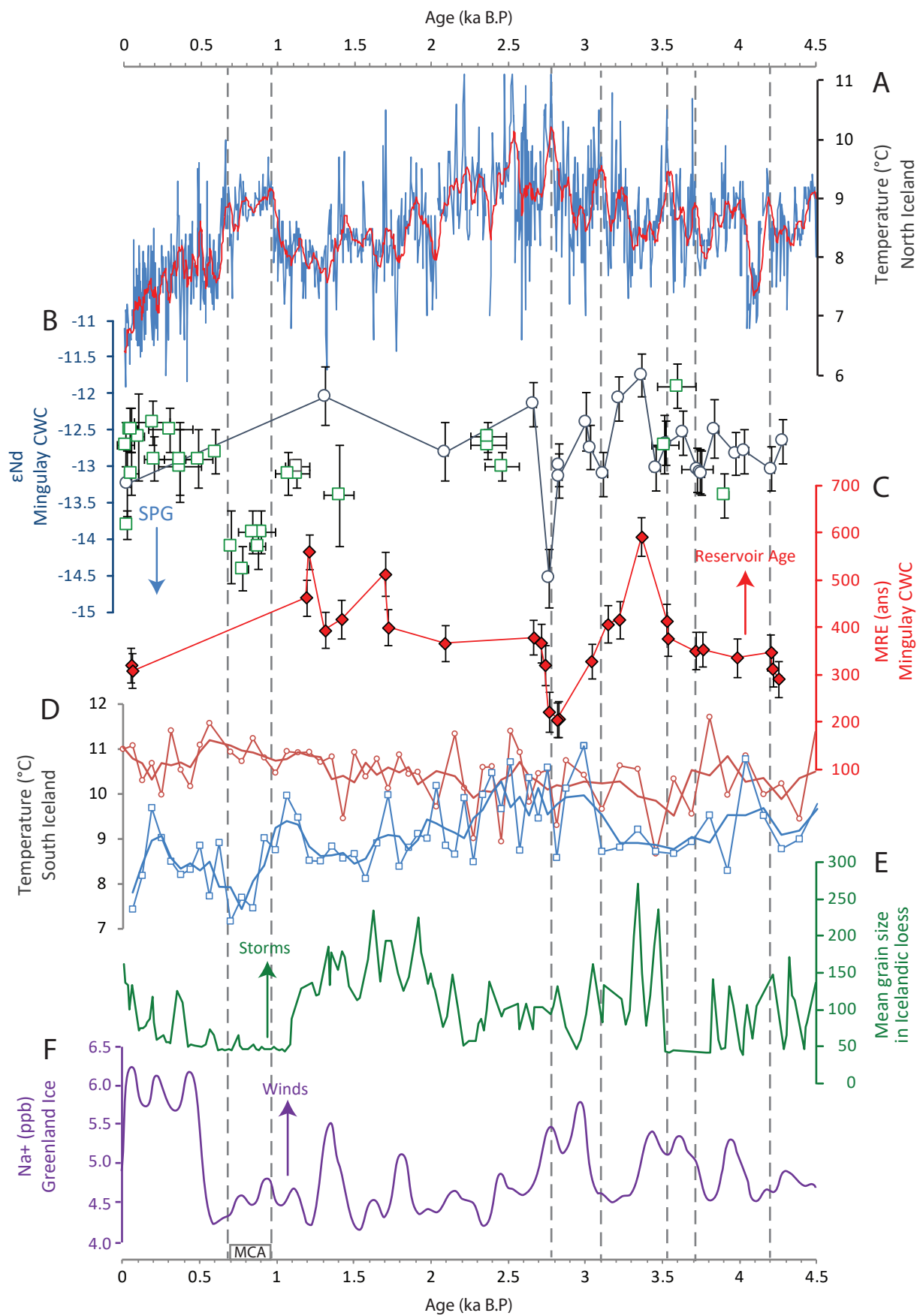


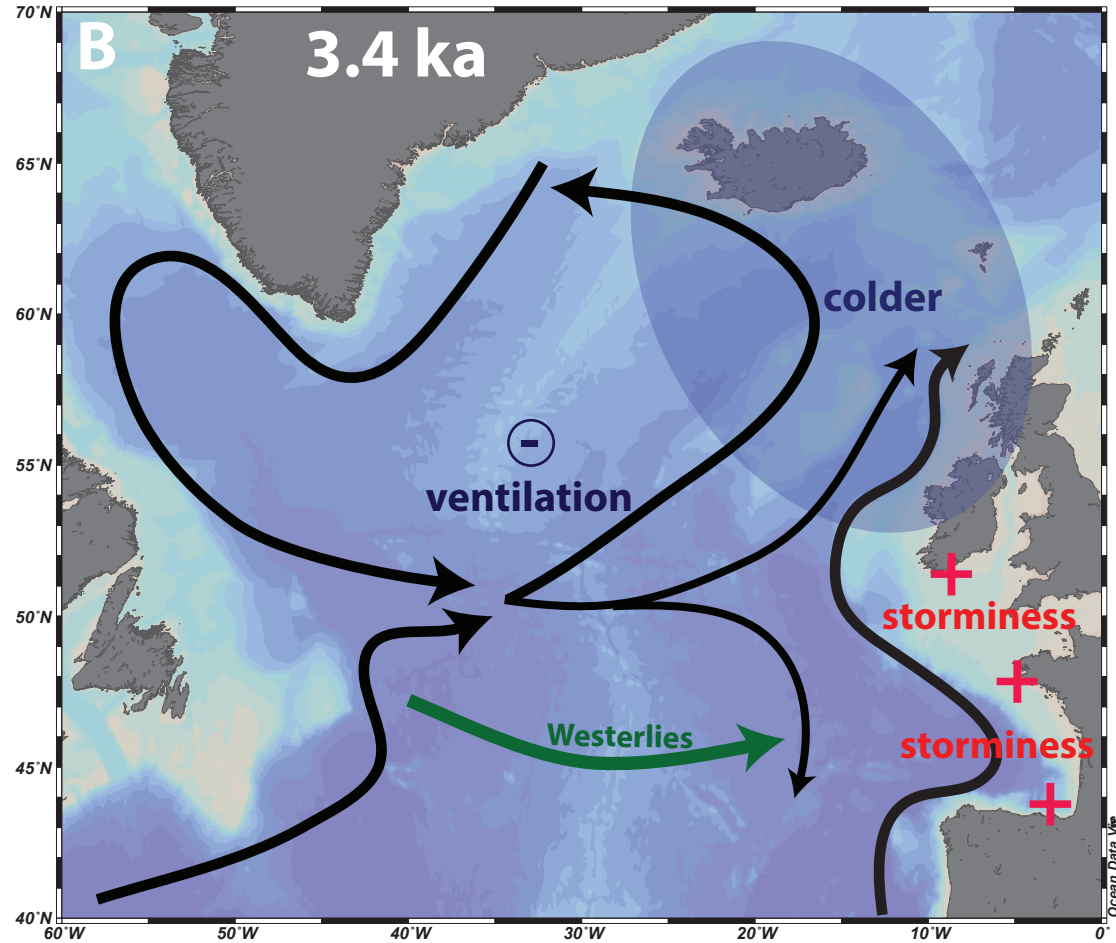
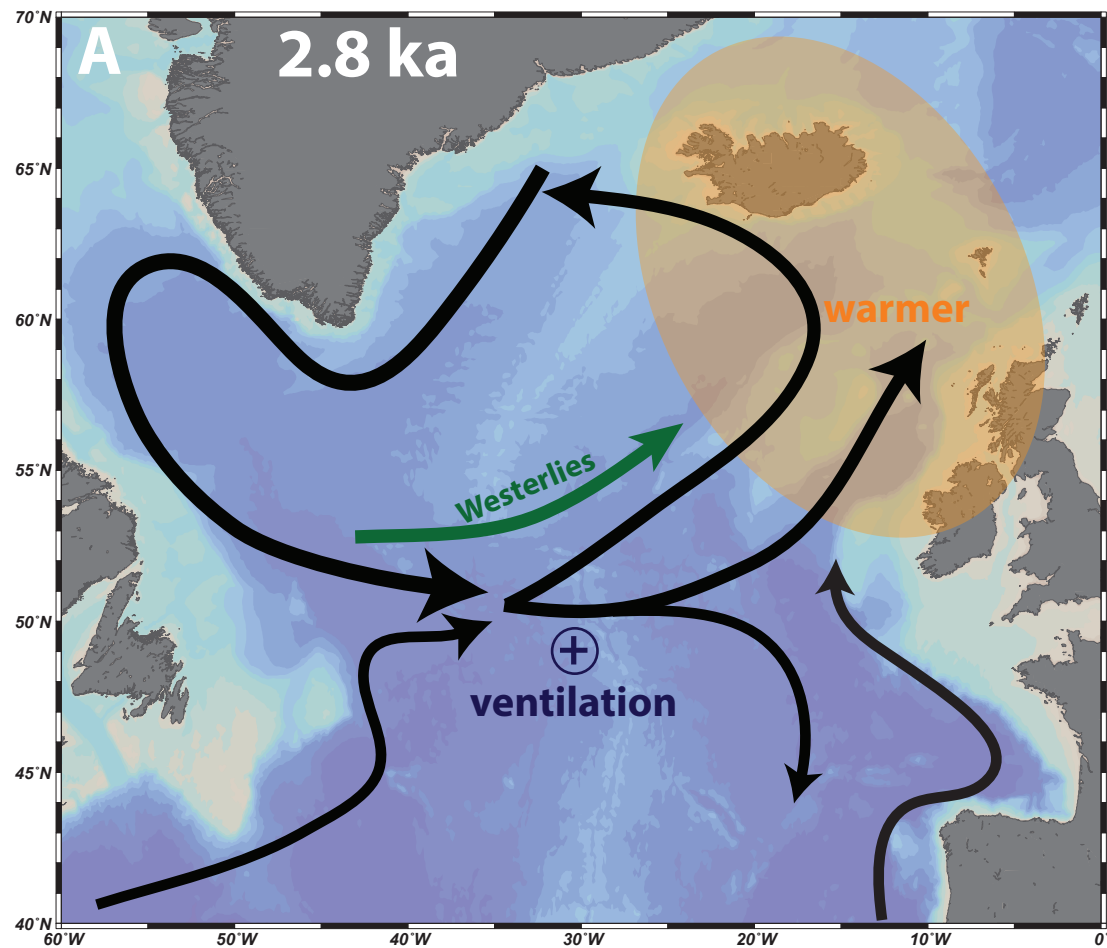












Sample Name	Latitude	Longitude	Depth (m)	Age (yrs)	$^{143}\text{Nd}/^{144}\text{Nd}$	error (2 SE)	ϵNd	error (2 SE)	error (2 SD)
<i>GRAB 15.5.5.10</i>	56°47'08" N	7°25'48" W	seafloor	21 ±13	0.511959	±0.000023	-13.24	±0.45	±0.40
<i>GRAB 1151</i>	56°49'08" N	7°23'36" W	seafloor	1312 ±11	0.512021	±0.000021	-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
<i>GRAB 56.08.928</i>	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	±0.000008	-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)	ϵ Nd	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