# Northern Sourced Water dominated the Atlantic Ocean during the Last Glacial Maximum

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## ABSTRACT

Increased carbon sequestration in the ocean subsurface is commonly assumed to have been one of the main causes responsible for lower glacial atmospheric $CO_2$ concentrations. Remineralized carbon must have been stored away from the atmosphere for thousands of years, yet the water mass structure accommodating such increased carbon storage continues to be debated. Here we present new sediment derived bottom water neodymium isotope records that allow fingerprinting of water masses and provide a more complete picture of the Atlantic overturning circulation geometry during the Last Glacial Maximum. These results suggest that the vertical and meridional structure of the Atlantic water mass distribution only experienced minor changes since the last ice age. In particular, we find no compelling evidence supporting glacial southern sourced water substantially expanding to shallower depths and farther into the northern hemisphere than today, which has been previously inferred from stable carbon isotope ($\delta^{13}C$) reconstructions. We argue that depleted $\delta^{13}C$ values observed in the deep Northwest Atlantic do not necessarily indicate the presence of southern sourced water. Instead, these values may represent a northern sourced water mass with lower than modern preformed $\delta^{13}C$ values that were further modified downstream by increased sequestration of remineralized carbon, facilitated by a more sluggish glacial deep circulation corroborating previous evidence.


## INTRODUCTION

During the Last Glacial Maximum (LGM) atmospheric $CO_2$ concentrations were ~90 ppm lower than during pre-industrial times (Monnin et al., 2001). Converging evidence suggests that a considerable amount of remineralized $CO_2$ was sequestered from the atmosphere into the deep ocean due to an overall more efficient biological carbon pump, a more stratified Southern Ocean, and globally lower sea surface temperatures increasing the solubility of $CO_2$ (Broecker, 1982; Galbraith and Jaccard, 2015). Further, it has been proposed that enhanced oceanic $CO_2$ storage was

facilitated by stronger deep water stratification along with increased ocean alkalinity (Broecker and Peng, 1989). Benthic foraminiferal carbon isotope-based reconstructions ($\delta^{13}C$ and $\Delta^{14}C$) of the Atlantic Meridional Overturning Circulation (AMOC) support the notion of an enhanced glacial deep ocean stratification by indicating that young, nutrient-rich waters occupied water depths shallower than ~2500 m, whereas older, nutrient depleted waters prevailed below (Curry and Oppo, 2005). The similarity of LGM deep North Atlantic carbon isotope compositions to those of glacial North Pacific Deep Water led to the suggestion that large parts of the glacial deep Atlantic were filled with Southern Sourced Water (SSW; Curry and Oppo, 2005; Keigwin, 2004). Such a circulation pattern differs fundamentally from the modern AMOC geometry, in which SSW advection is mostly confined to the deep Southwest Atlantic (Fig. 1). However, a major caveat of carbon isotope based paleoceanographic reconstructions is their inherent dependency on changes in the carbon cycle. For instance, the stable carbon isotope ($\delta^{13}C$) distribution of dissolved inorganic carbon reflects water mass provenance, but remineralization of organic matter can substantially alter the $\delta^{13}C$ signature of a water mass independent of mixing (Lynch-Stieglitz et al., 2007). Furthermore, $\delta^{13}C$ as well as $\Delta^{14}C$, are influenced by changes in air-sea carbon exchange (Broecker and Maier-Reimer, 1992). Thus, distinguishing water mass provenance from carbon cycle related changes constitutes a major difficulty intrinsic to these proxies.

The radiogenic neodymium isotopic signature (εNd), independent of the carbon cycle, is a promising proxy that behaves quasi-conservatively in the Atlantic Ocean (van de Flierdt et al., 2016). In the modern Atlantic, Northern Sourced Water (NSW) is characterized by a dissolved Nd isotope signature of -12.8 ± 0.4 whereas SSW has a distinctly more radiogenic (higher) signature of -9.0 ± 0.4 (Stichel et al., 2012). However, these εNd end members were not constant through time. During the LGM SSW exhibited higher values around -5.5 (Skinner et al., 2013), likely due to the absence of bottom water export from the Weddell Sea into the Atlantic as recently reported by Huang et al. (2020). Similarly, new reconstructions of the northern end member indicate a higher

glacial εNd value between -10 and -11 of NSW at 40°N (Zhao et al., 2019). The authors argue that the unradiogenic Canadian Shield and Greenland contributed considerably less Nd to the North Atlantic during the LGM, due to extensive ice-sheet cover of these provinces. Consideration of these εNd end members allows for a more accurate interpretation of past changes in water mass distribution of the Atlantic.

Here we present new Nd isotope reconstructions that close a crucial data gap in the deep Southwest Atlantic. We combine these new data with published time series from the West Atlantic (Howe et al., 2016; Pöppelmeier et a., 2020; Skinner et al., 2013) to generate transects for the late Holocene and LGM time slices. Building on the compilation of Howe et al. (2016), we further reconstruct the vertical water mass structure at 30°S in high resolution, assess the potential influence of benthic exchange processes on bottom water εNd by employing a box model, and by excluding regions where such processes are exacerbated by benthic nepheloid layers (Northwest Atlantic and Cape Basin, Pöppelmeier et al., 2019). Hence, we are able to robustly reconstruct glacial-interglacial changes of the deep water geometry.

## METHODS

Authigenic Nd was extracted from bulk sediment following the protocol of Blaser et al. (2016). The Nd isotopic compositions were measured on two Neptune Plus MC-ICP-MSs at GEOMAR, Kiel and the University of Erlangen-Nuernberg, Germany. Instrumental mass fractionation was corrected for by normalizing $^{146}Nd/^{144}Nd$ to 0.7219. Further, samples were bracketed by JNdi-1 standard reference solution normalized to the accepted value of $^{143}Nd/^{144}Nd$ = 0.512115 (Tanaka et al., 2000). The external reproducibility (2σ) was determined by repeated in-house standard measurements and varied between 0.12 and 0.50 ε-units in different sessions.

## RESULTS AND DISCUSSION

The spatial distribution of sediment core data is arguably sparse compared to modern oceanographic seawater sampling stations. To nonetheless allow for a comparison between Holocene-aged εNd reconstructions and modern oceanography and water mass provenance, we choose the 4000 ± 500 m water depth isobath to derive the relative proportion of SSW at a given location based on modern oceanographic parameters (silicate, N*, pot. T, and salinity; Figs. 1A and S1) and compare to core-top measurements at similar depths. This water depth represents the lower limit up to which well dated sediment cores are available in sufficient quantity, as robustly dating sediments from greater depths becomes increasingly difficult due to poor preservation of calcium carbonate and generally lower sedimentation rates (Broecker, 1982).

To calculate the proportion of SSW based on the εNd reconstructions, we used a binary mixing model with modern seawater compositions (van de Flierdt et al., 2016) and reconstructed (Huang et al., 2020; Zhao et al., 2019) end members for the Holocene and LGM, respectively. At the 4000 m isobath late Holocene εNd based reconstructions and modern oceanography both show a coherent decrease of SSW from nearly 100% at 45°S to less than 20% north of the Equator (i.e. ΔSSW ≈ 85%, Fig. 2A), which supports the validity of the Nd isotope proxy as a semi-conservative water mass tracer along the transect. Based on εNd reconstructions this meridional water mass gradient existed with nearly the same configuration during the LGM (Fig. 2B), with a decrease of the SSW proportion from 100% at 45°S to about 20% north of the Equator (ΔSSW ≈ 80%). For these calculations we consider Nd concentrations as constant due to a lack of observational data, but moderate variations of modern Nd concentrations do not lead to changes in our interpretations (Fig. S5). Such a sharp meridional deep water mass gradient strongly contrasts with nutrient proxies that suggest a substantially greater amount of SSW bathing large parts of the deep North Atlantic basin during the LGM (Curry and Oppo, 2005). This discrepancy raises the question whether the Nd isotope derived water mass gradient is at least partly affected by non-conservative processes.

Benthic Nd addition represents the main process capable of altering the Nd isotopic composition of bottom waters independent of water mass mixing (Haley et al., 2017). To quantitatively test the influence of a potential benthic Nd source on the observed εNd gradient of the LGM we employ a simple box model (Fig. S6). In the model, we only consider the flow path between the Rio Grande Rise and the Equatorial Atlantic, because εNd reconstructions farther south show a homogeneous signal. This is a first indication that basin-wide benthic Nd addition only played a subordinate role. The reasoning for this is that a strong benthic flux would produce an εNd gradient along the flow path of SSW, assuming that the signature matched not exactly the initial bottom water εNd value, which is not observed here. For conservative parameter estimates of the advection rate and benthic Nd flux (<4 Sv and 30 pmol/cm$^2$/year (Abbott et al., 2015) and $\varepsilon Nd_{detrital}$ = -7 (Pöppelmeier et al., 2020), respectively) the model results suggest that non-conservative benthic Nd addition may have produced about 0.5 ε-units of the total meridional ~4.5 ε-units gradient, which translates into a negligible decrease of the SSW gradient (from ΔSSW ≈ 80% to ~75%; Fig. S8). Even for an unrealistically, extreme scenario that this non-conservative process contributed 2 of the 4.5 ε-units meridional Nd isotope gradient (< 2 Sv and $\varepsilon Nd_{detrital}$ = -10), the decrease in the SSW gradient amounts to only about 10% (from ΔSSW ≈ 80% to ~70%). Thus, we conclude that the meridional gradient in past bottom water Nd isotopes is robust, which implies dominance of NSW over SSW in the glacial deep Equatorial and North Atlantic.

In order to not only constrain the latitudinal spread of glacial SSW but also the vertical extent of this water mass, we reconstructed past seawater εNd for a depth transect near 30°S (Figs. 3). In this region, the modern water mass boundary between SSW and NSW is situated at depths between 3500 and 4000 m (as defined by the 50% mixing ratio (Fig. 1A)). Modern seawater and reconstructed Holocene εNd data capture this feature well. During the LGM the Nd isotope data were generally more radiogenic than modern, due to the more radiogenic end members, but at ~3500 m water depth a sharp 2.5 ε-units step is observed. Since the investigated sites are not only

located at the continental margin but are also in pelagic environments (Fig. 1B), local detrital input effects cannot account for the observed εNd depth trend, but instead must represent a more integrated regional signal. Therefore, the most plausible explanation for the marked εNd gradient at ~3500 m is the presence of a pronounced water mass boundary between NSW (less radiogenic) above and SSW (more radiogenic) below. Since non-conservative processes cannot account for the meridional and vertical εNd gradients observed here, the data clearly indicate that similar to modern proportions of NSW bathed the entire deep Northwest Atlantic during the LGM. This calls into question the nutrient-proxy based interpretation of a glacial deep Atlantic dominated by SSW.

Stable carbon isotope reconstructions have a long-standing history as a water mass tracer and a recent study confirmed the good correlation between core top and seawater $\delta^{13}C$ on a global scale (Schmittner et al., 2017). However, Holocene-aged core top reconstructions in key regions such as the deep Northwest Atlantic partly deviate from seawater $\delta^{13}C$, potentially related to the regionally elevated accumulation of organic matter (Fig. S8, Keigwin, 2004). $\delta^{13}C$ reconstructions from sites that exhibit these core top – seawater offsets have also been used for interpretation of past seawater provenance (Keigwin, 2004). $\delta^{13}C$ signatures at these sites have been interpreted as high SSW contributions during the LGM (Keigwin, 2004), which may at least partly be biased by the same obfuscating effects that produce the core top – seawater offset at these locations today. Furthermore, model simulations suggest that the shoaling of NSW, previously inferred from nutrient-like proxies, may have been limited to the core of the water mass (defined as the depth with the strongest advection) and may not have affected the boundary between NSW and SSW to the same extent (Gebbie, 2014; Oppo et al., 2018). Yet ocean circulation models are inconsistent on this issue, with roughly half of the models within the Paleoclimate Model Intercomparison Project 2 (PMIP2) showing a shallower NSW while the others yield deep NSW during the LGM comparable to the modern geometry (Weber at al., 2007). On the other hand, the majority of models in the later PMIP3 favor a circulation pattern similar to modern (Muglia and Schmittner, 2015). One possibility

to reconcile a modern-like extent of NSW in the Northwest Atlantic with the observed $^{13}C$ depletions relates to increased accumulation of respired carbon originating from remineralization of organic matter (Gebbie, 2014). This remineralization may have been facilitated or amplified by the less vigorous Atlantic deep water circulation of the LGM as indicated by Pa/Th (McManus et al., 2004; Ng et al., 2018) and generally colder ocean temperatures (Matsumoto, 2007). Likewise, $^{14}C$ data suggest a higher ventilation age of the deep Atlantic during the LGM allowing for deep water to accumulate more respired carbon and thus lowering its $\delta^{13}C$ signature (Skinner et al., 2017). In addition, these $^{14}C$ data show aging of deep waters from North to South as expected from a deep northern sourced water mass propagating southward.

These consistent observations lead us to the conclusion that the glacial deep water mass distribution was not substantially different from today (Fig. 4). Our new Nd isotope reconstructions suggest that glacial SSW may have penetrated slightly farther northward than today, but was still mostly confined to the deep South Atlantic with a vertical expansion similar to the modern Atlantic. Thus, as a direct consequence, NSW must have dominated the deep North Atlantic also with a spatial expansion similar to today. We hence argue that the shallower stratification observed in the North Atlantic with nutrient-based proxies was instead likely the result of differences in the contribution of remineralized organic matter to the upper and lower limb of NSW due to different advection rates of both limbs (Ng et al., 2018; Skinner et al., 2017). This is in accordance with the rather steady change of $\delta^{13}C$ with depth that do not indicate a clear water mass boundary (Fig. S10). Since Nd is not affected by biological cycling, this chemical divide did not affect the εNd distribution. The deep Atlantic-wide evidence for elevated deep water Pa/Th (Ng et al., 2018) and the prolonged glacial deep-to-surface supply of Southern Ocean waters (Hasenfratz et al., 2019) nevertheless suggests LGM circulation modes substantially different from the Holocene situation.

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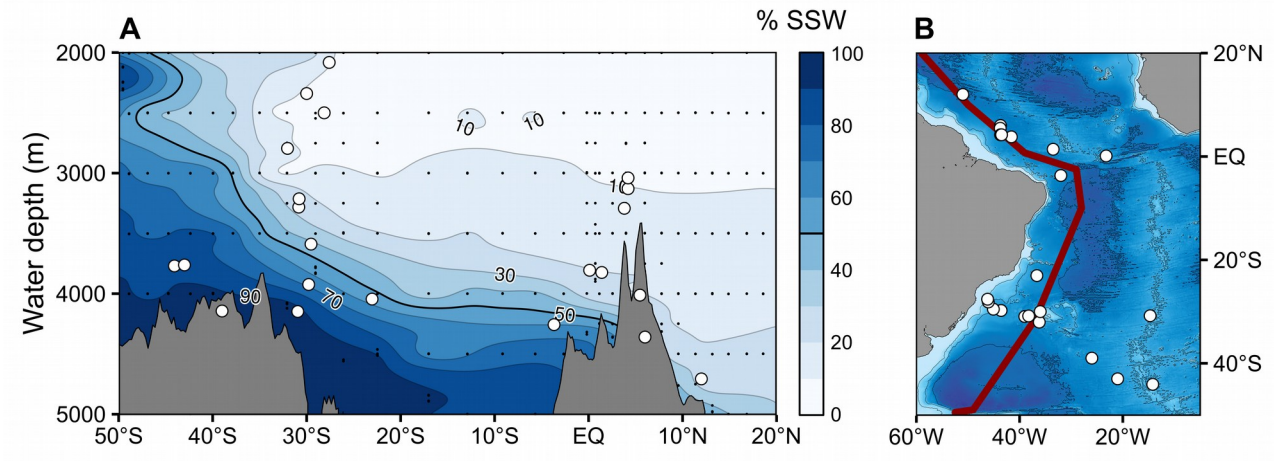



Figure 1. (A) Water mass distribution based on four independent conservative water mass properties (Fig. S1) along the GEOTRACES GA02 transect (Schlitzer et al., 2018; red line in panel b). Black dots indicate water sampling stations. (B) Overview map of the tropical and South Atlantic. The red line marks the GA02 transect. Sediment core sites used in this study are marked by open circles in both panels.

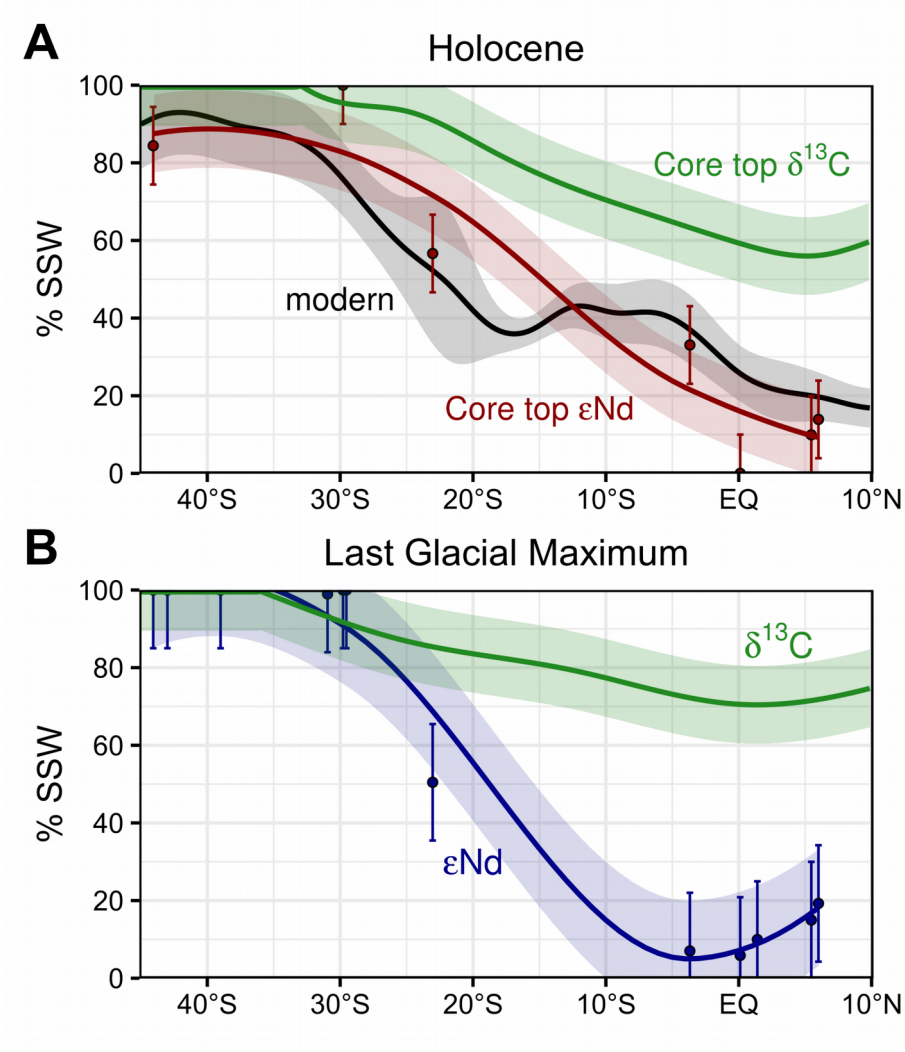
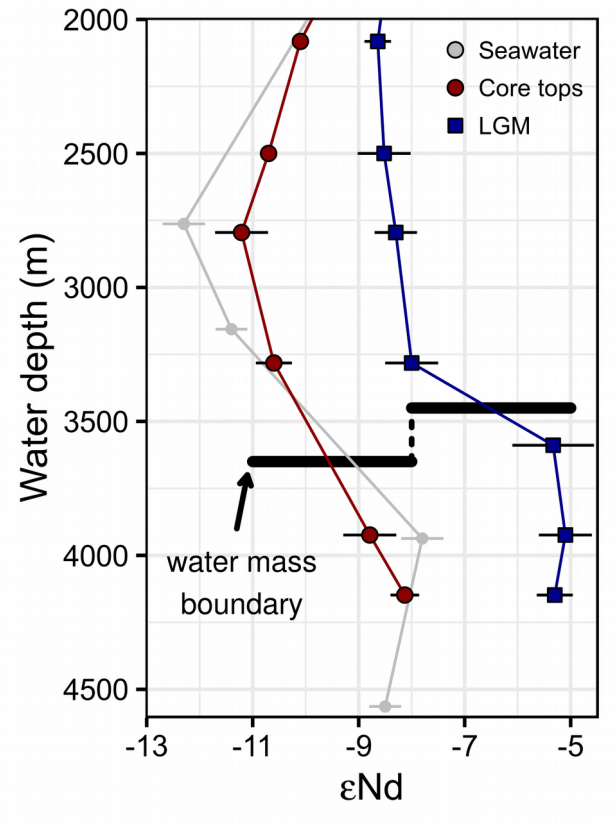
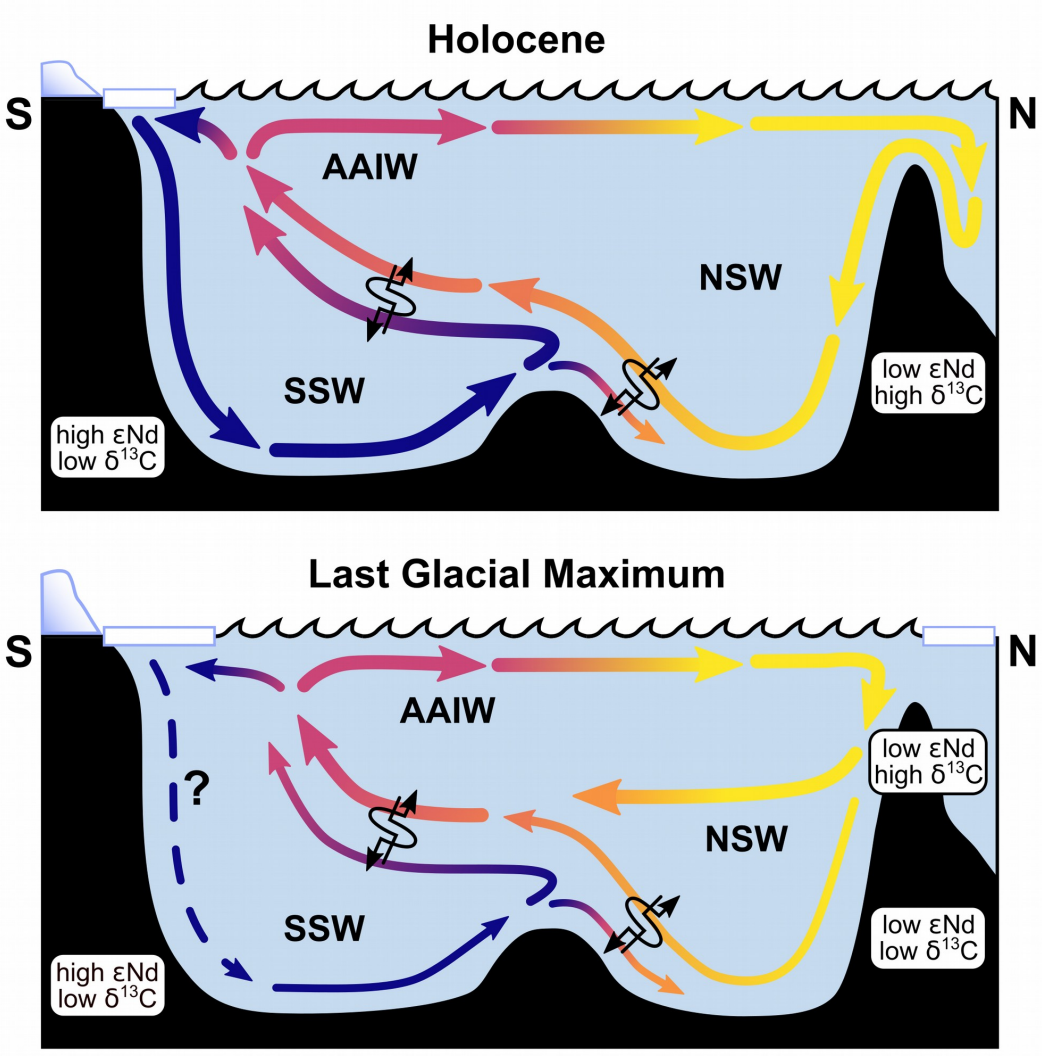



Figure 2. Deep water provenance of SSW at 4000 ± 500 m water depth. (A) Modern and Holocene SSW contribution. Black line depicts % SSW at 4000 m derived from modern oceanographic properties of Fig. 1A. The εNd based reconstructions (red line) are Holocene averages of the time period from 0 to 6.5 ka. $\delta^{13}C$ data are based on Schmittner et al. (2017) (B) εNd based reconstruction of the deep water mass provenance during the LGM with adjusted end member compositions. $\delta^{13}C$ data are based on Oppo et al. (2018). Ribbons depict the uncertainty estimated to ± 10% for the Holocene εNd reconstructions and ± 15% for the LGM (SI). The uncertainty of the modern hydrography (panel a, black line) is the 1σ deviation of the calculation for Fig. 1A.




290 Figure 3. εNd depth gradient at ~30°S. Core tops, representative of the mid to late Holocene are
291 depicted in red, modern seawater (Jeandel, 1993) in gray, and averaged LGM values in blue. Core
292 sites are from the Brazil Margin as well as the open ocean (near the Rio Grande Rise). The modern
293 water mass boundary is derived from the 50% mixing line at 30°S of Fig. 1A.




294 Figure 4. The Holocene overturning (top) shows two main circulation cells, with NSW forming in
295 the Nordic Seas and Labrador Sea and SSW forming near Antarctica. During the Last Glacial
296 Maximum (bottom) the proposed circulation scheme consisted of three cells. The reduced deep
297 water formation in the Nordic Seas produced less deep water and an intermediate depth northern
298 sourced water mass formed (Glacial North Atlantic Intermediate Water) but at the same time the
299 formation of SSW was also reduced. The width of the arrows roughly corresponds to proposed
300 volume transports.

301

[1]GSA Data Repository item 201Xxxx, Southwest Atlantic hydrography; sample sites and neodymium isotope data; estimates of benthic flux influence, is available online at www.geosociety.org/pubs/ft20XX.htm, or on request from editing@geosociety.org.