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Antarctic intermediate water circulation in the South Atlantic over the past 25,000 years

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Key Points:
- Foraminifera successfully preserve seawater neodymium isotopic composition in the intermediate-depth South Atlantic
- There is no evidence for changes in the proportion of Antarctic Intermediate Water in the South Atlantic across the deglaciation
- More radiogenic neodymium isotopic values indicate that there was a shift in Antarctic Intermediate Water production during the mid-Holocene

Supporting Information:
- Supporting Information S1

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Abstract Antarctic Intermediate Water is an essential limb of the Atlantic meridional overturning circulation that redistributes heat and nutrients within the Atlantic Ocean. Existing reconstructions have yielded conflicting results on the history of Antarctic Intermediate Water penetration into the Atlantic across the most recent glacial termination. In this study we present leachate, foraminiferal, and detrital neodymium isotope data from three intermediate-depth cores collected from the southern Brazil margin in the South Atlantic covering the past 25 kyr. These results reveal that strong chemical leaching following decarbonation does not extract past seawater neodymium composition in this location. The new foraminiferal records reveal no changes in seawater Nd isotopes during abrupt Northern Hemisphere cold events at these sites. We therefore conclude that there is no evidence for greater incursion of Antarctic Intermediate Water into the South Atlantic during either the Younger Dryas or Heinrich Stadial 1. We do, however, observe more radiogenic Nd isotope values in the intermediate-depth South Atlantic during the mid-Holocene. This radiogenic excursion coincides with evidence for a southward shift in the Southern Hemisphere westerlies that may have resulted in a greater entrainment of radiogenic Pacific-sourced water during intermediate water production in the Atlantic sector of the Southern Ocean. Our intermediate-depth records show similar values to a deglacial foraminiferal Nd isotope record from the deep South Atlantic during the Younger Dryas but are clearly distinct during the Last Glacial Maximum and Heinrich Stadial 1, demonstrating that the South Atlantic remained chemically stratified during Heinrich Stadial 1.

1. Introduction

The Atlantic meridional overturning circulation plays an important role in the redistribution of heat, carbon, and nutrients within the Atlantic Ocean. Antarctic Intermediate Water (AAIW) is one of the primary water masses flowing northward in the Atlantic that eventually feed the production of North Atlantic Deep Water (NADW) [*Broecker and Takahashi*, 1981]. Numerous studies have focussed upon changes in NADW production during the past 25 kyr, including possible changes in the strength of overturning during the Last Glacial Maximum (LGM) [*Otto-Bliesner et al.*, 2007; *Weber et al.*, 2007; *Lippold et al.*, 2012] as well across the deglaciation [*Boyle and Keigwin*, 1987; *Rutberg et al.*, 2000; *McManus et al.*, 2004; *Piotrowski et al.*, 2004; *Roberts et al.*, 2010]. Particular emphasis has been placed upon possible "shut downs" in production of NADW or its glacial counterpart Glacial North Atlantic Intermediate Water during the Northern Hemisphere cold periods—Heinrich Stadial 1 (HS1) and the Younger Dryas (YD)—that punctuated the most recent glacial termination [*McManus et al.*, 2004]. It follows that any decrease in NADW production could be accompanied by shifts in the flux of AAIW, the water mass that flows from South to North Atlantic that is converted into NADW in the North Atlantic.

Despite these assertions, the extent to which AAIW occupied the intermediate-depth Atlantic across the most recent glacial termination remains unclear. Some studies have proposed that during abrupt Northern Hemisphere cold events AAIW may have penetrated much further into the North Atlantic [*Rickaby and Elderfield*, 2005; *Pahnke et al.*, 2008; *Thornalley et al.*, 2011], whereas others suggest the exact opposite [*Xie et al.*, 2012; *Huang et al.*, 2014]. Indeed, a recent study reinterprets North Atlantic deglacial anomalies as deriving from old water that originated in the Arctic Ocean [*Thornalley et al.*, 2015]. Studies of intermediate water circulation based upon nutrient proxies such as the Cd/Ca ratio or $\delta^{13}C$ of benthic foraminifera [*Marchitto et al.*, 1998; *Zahn and Stüber*, 2002; *Came et al.*, 2003, 2008] are complicated by the fact that the changes in overturning strength that accompanied abrupt Northern Hemisphere cold events [*McManus et al.*, 2004] could decouple these proxies from water mass mixing due to greater accumulation of respired organic matter [*Gebbie*, 2014; *Oppo et al.*, 2015; *Schmittner and Lund*, 2015]. Changes in the ventilation age of intermediate waters reconstructed from benthic-planktic radiocarbon measurements could similarly be due either to changes in the strength of overturning and thus in situ water mass aging or changes in water mass mixing proportions [*Freeman et al.*, 2015].

Neodymium isotopes offer an independent tracer of water mass mixing [*Frank*, 2002]. The neodymium isotopic composition (ε_{Nd}) is defined as $((^{143}Nd/^{144}Nd_{Sample})/(^{143}Nd/^{144}Nd_{CHUR}) - 1) \times 10^4$ where the chondritic uniform reservoir $^{143}Nd/^{144}Nd_{CHUR} = 0.512638$ [*Jacobsen and Wasserburg*, 1980]. Upper NADW has a neodymium isotopic composition (ε_{Nd}) of -13.2 [*Lambelet et al.*, 2016], whereas AAIW shows an ε_{Nd} around -8.3 [*Stichel et al.*, 2012]. Published intermediate-depth ε_{Nd} reconstructions of the past 25 kyr based upon measurements of various authigenic phases of sediment cores show conflicting results. Some records indicate a greater presence of AAIW in the Atlantic during abrupt Northern Hemisphere cold events [*Pahnke et al.*, 2008], while others suggest the exact opposite [*Xie et al.*, 2012; *Huang et al.*, 2014]. These interpretations are complicated by the fact that some of the records consist of leachate measurements [*Pahnke et al.*, 2008; *Xie et al.*, 2012], which are susceptible to detrital contamination [*Elmore et al.*, 2011; *Wilson et al.*, 2013], or come from enclosed basins [*Xie et al.*, 2012, 2014] where the seawater has been shown to be subject to modification by boundary exchange [*Osborne et al.*, 2014]. At least one study utilized cores from below the main flow of AAIW [*Pahnke et al.*, 2008]. Furthermore, the scarcity of ε_{Nd} records from the intermediate-depth South Atlantic means that there is no certainty in how the AAIW end-member neodymium composition may have changed across the past 25 kyr.

Potential changes in intermediate-depth water mass sourcing during the deglaciation are of particular interest as the abrupt Northern Hemisphere cold periods are accompanied by changes in productivity throughout the Southern Ocean and eastern Atlantic Oceans that imply significant changes in nutrient availability [*Romero et al.*, 2008; *Anderson et al.*, 2009; *Hendry et al.*, 2012; *Meckler et al.*, 2013]. The source of these nutrients and the mechanism by which they reached the surface ocean, however, remains under debate [*Meckler et al.*, 2013; *Hendry et al.*, 2016]. Greater confidence in the source of intermediate-depth water in the South Atlantic would help to determine the source of nutrients to the low-latitude Atlantic during these abrupt climate events.

This study investigates water mass sourcing, with a particular focus on the distribution of AAIW, in the intermediate-depth South Atlantic across the last 25 kyr. We present ε_{Nd} measured on leachates, detrital sediment, and foraminifera from three intermediate-depth cores collected from the southern Brazil margin. These data are used to test the ability of leachate records to archive past seawater ε_{Nd} in this location. The foraminiferal ε_{Nd} records are then compared to other published paleoceanographic reconstructions to investigate changes in water mass mixing at intermediate depths in the South Atlantic over the past 25 kyr. We find no evidence for changes in the amount of AAIW bathing the southern Brazil margin across the deglaciation but see more radiogenic neodymium isotope values at the shallower sites in the mid-Holocene, implying a fundamental shift in AAIW production at that time.

2. Materials and Methods

2.1. Core Sites

Cores GeoB2107-3, KNR159-5-36GGC, and GeoB2104-3 from 1050, 1268, and 1500 m water depth on the southern Brazil margin, respectively (Figure 1), were used to investigate changes in intermediate water circulation in the Atlantic over the past 25 kyr. The GeoB cores were collected during R/V *Meteor* cruise M23 [*Bleil et al.*, 1994]. All three cores are located at the boundary between AAIW and NADW in the modern ocean (Figure 1), with a greater influence of northern-sourced waters at the deeper sites. The age model for GeoB2107-3 is based on planktic radiocarbon dates [*Heil*, 2006] converted to calendar ages by using the MARINE13 calibration curve [*Reimer et al.*, 2013]. The age model of GeoB2104-3 comes from planktic radiocarbon dates as well as correlation of XRF Fe/Ca data to that of GeoB2107-3 [*Hickey*, 2010]. These age model assignments are supported by $\delta^{18}O$ records of *Uvigerina peregrina* from both cores [*Hickey*, 2010]. *Hendry et al.* [2012] used the MARINE09 calibration of the same radiocarbon ages and tuned the benthic oxygen isotopes to Antarctic temperature to construct the age model for GeoB2107-3. These different approaches cause shifts

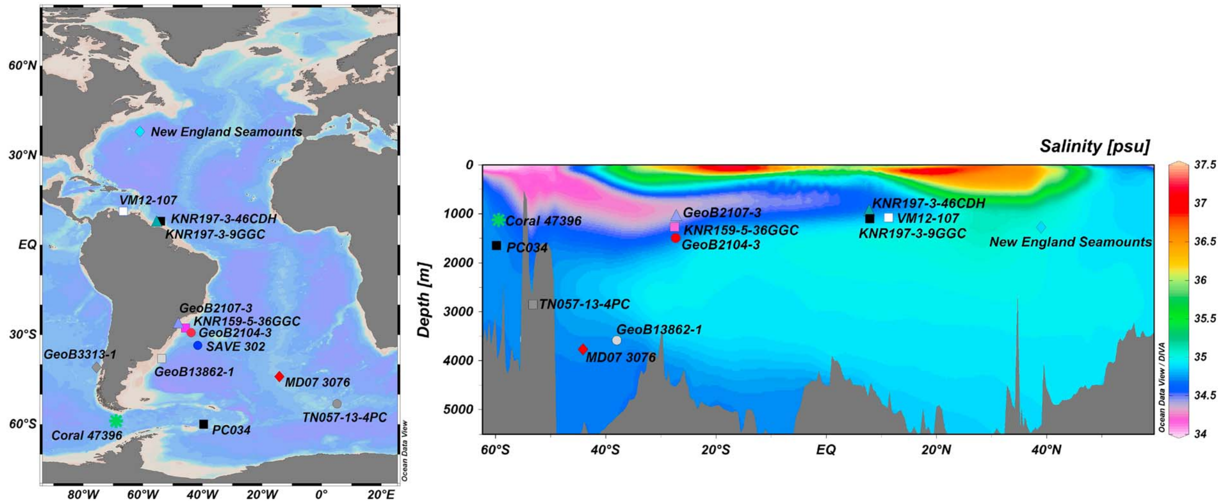

Figure 1. (left) Map showing the location of cores used in this work to reconstruct ε_{Nd} records across the past 25 kyr. GeoB2107-3 (purple triangle; 27.2°S, 46.5°W; 1050 m), KNR159-5-36GGC (pink square; 27.5°S, 46.5°W; 1268 m), and GeoB2104-3 (burgundy circle; 27.3°S, 46.4°W; 1500 m), and the location of published records to which these results are compared. (right) Profile of salinity in the western Atlantic [*Antonov et al.*, 2010] with the location of the Atlantic cores shown in Figure 1 (left). Both panels were produced with Ocean Data View [*Schlitzer*, 2016].

of at most a few hundred years during the early deglaciation, but importantly do not shift the occurrence of the peak silicon isotope values at 12.9 ka that *Hendry et al.* [2012] interpreted as being in the Younger Dryas. We note that this peak occurs at the boundary of the Bølling-Allerød and Younger Dryas; given the uncertainty of the age model of a few hundred years, however, that peak could indeed occur within the Younger Dryas itself as suggested by Hendry *et al.* [2012], and we refer to it here as an early Younger Dryas peak. The age model for KNR159-5-36GGC has been updated by *Lund et al.* [2015] using the radiocarbon dates of *Sortor and Lund* [2011]. The published leachate ε_{Nd} data from KNR159-5-36GGC from *Pahnke et al.* [2008] presented here were put onto this new radiocarbon-based age model.

2.2. Neodymium Measurements

Foraminifera from all three cores were prepared following the protocol of *Roberts et al.* [2010]. In brief, mixed planktic foraminifera were picked from the coarse fraction (>63 μm) for neodymium isotope measurements. Foraminifera tests were broken open and washed with sonication, and the supernatant decanted, rinsed, and this process repeated until the water remained clear and all clays were removed. The crushed foraminiferal samples were then dissolved in 1 mol L^{-1} reagent grade acetic acid.

Leaching was carried out on samples from GeoB2107-3 and GeoB2104-3 following the procedure used by *Pahnke et al.* [2008]. In short approximately 5 cm^3 of sediment was repeatedly decarbonated overnight using a buffered acetic acid solution until no carbonate remained. Decarbonated samples were MilliQ water rinsed then leached for 1 h with a 0.02 mol L^{-1} solution of hydroxylamine hydrochloride in 25% acetic acid (vol/vol).

For detrital analysis, after applying the leaching protocol described above, the remaining sediment fraction was leached two further times, for 6 h then overnight, with the 0.02 mol L^{-1} hydroxylamine hydrochloride solution. Samples were then MilliQ rinsed and dried down in an oven at 50°C before being roasted overnight at 900°C to remove organic material. Roasted sediment were transferred to Teflon hexnut vials and sequentially bombed overnight in qHF/qHNO_3, qHCl, and qHNO_3 [*Bayon et al.*, 2002; *Noble et al.*, 2012].

For all three phases the rare earth elements were extracted from other elements by using Eichrom TRUspec™ resin in 100 μL Teflon columns. Neodymium was then separated from the other rare earth elements by using Eichrom LNspec™ resin on volumetrically calibrated Teflon columns.

Samples from GeoB2104-3 and GeoB2107-3 were analyzed for isotopic composition on either a Nu Plasma or a Neptune Plus multicollector inductively coupled plasma–mass spectrometer (MC-ICP-MS) in the Department of Earth Sciences at the University of Cambridge. Samples from KNR159-5-36GGC were

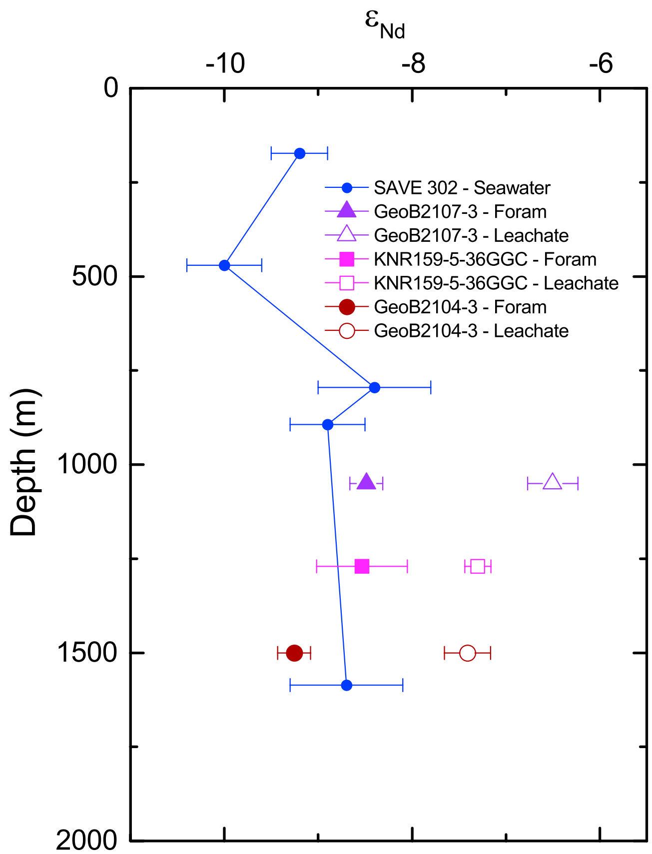

Figure 2. Comparison of core top ε_{Nd} measurements made on uncleaned foraminifera (filled symbols) and leachates (open symbols) from GeoB2107-3 (purple triangles; 27.2°S, 46.5°W; 1050 m), KNR159-5-36GGC (pink squares; 27.5°S, 46.5°W; 1268 m; leachate data from *Pahnke et al.* [2008]), and GeoB2104-3 (burgundy circles; 27.3°S, 46.4°W; 1500 m) with nearby seawater ε_{Nd} (blue circles; SAVE 302, 33.6°S, 41.6°W [*Jeandel*, 1993]). The error bars are 2σ external error.

analyzed on a Neptune MC-ICP-MS at Woods Hole Oceanographic Institute (WHOI) [*Huang et al.*, 2012]. Isotopic ratios were corrected to a $^{146}Nd/^{144}Nd$ of 0.7219 by applying an exponential mass correction. Samples were bracketed with a concentration-matched solution of reference standard JNdi-1, the measured composition of which varied between runs but was corrected to the accepted value of $^{143}Nd/^{144}Nd = 0.512115$ [*Tanaka et al.*, 2000]. The error bars correspond to 2σ of external reproducibility of the bracketing standards except for when the internal error was larger than the external error, in which case the error quoted is the combined internal and external errors. The average ε_{Nd} external reproducibility was ±0.15 for the Neptune Plus and ±0.40 for the Nu Plasma and ±0.40 for the Neptune; this was corroborated by the reproducibility of secondary standards. Samples measured on the Neptune Plus and Nu Plasma at the University of Cambridge were at least 5 ng in size, whereas sample sizes were as low as 1 ng for those samples run on the Neptune at WHOI. All results are listed with errors in Tables 1–3 in the supporting information.

3. Results

The core top leachate ε_{Nd} values of GeoB2107-3, KNR159-5-36GGC, and GeoB2104-3 are approximately 2 epsilon units more radiogenic than nearby seawater measurements (Figure 2) [*Jeandel*, 1993]. In contrast, the core top foraminiferal ε_{Nd} values from all three cores agree with linear interpolation of the nearest seawater measurements within analytical error (Figure 2). The low-resolution leachate ε_{Nd} record of GeoB2107-3 shifts from −5.6 to −6.5 (Table S1), and the record of GeoB2104-3 from −6 to −7.4 (Table S2), between the LGM and the Holocene (Figure 3). These records bracket the published leachate ε_{Nd} record of KNR159-5-36GGC [*Pahnke et al.*, 2008] with the shallower site (i.e., GeoB2107-3) offset to more radiogenic values and the deeper site (i.e., GeoB2104-3) to less radiogenic values (Figure 3). The foraminiferal ε_{Nd} records of GeoB2107-3, KNR159-5-36GGC, and GeoB2104-3 are all less radiogenic than their corresponding leachate records and are remarkably stable with ε_{Nd} values near −9 for most of the last 25 kyr (Figure 3). The only excursion outside of analytical error occurs in the mid-Holocene when the records of GeoB2107-3 and KNR159-5-36GGC shift to more radiogenic values, with GeoB2107-3 reaching an ε_{Nd} of −7.8. Throughout most of the last 25 kyr, the foraminiferal ε_{Nd} record of GeoB2107-3 is offset to slightly more radiogenic values than the other two records (Figure 3). Although KNR159-5-36GGC often displays the least radiogenic values, those data points are almost entirely within analytical error of the measurements from GeoB2104-3 (Figure S1 in the supporting information), indicating that the offset of the intermediary site to the least radiogenic values is unlikely to be a real phenomenon. The bulk detrital ε_{Nd} values of GeoB2107-3, KNR159-5-36GGC,

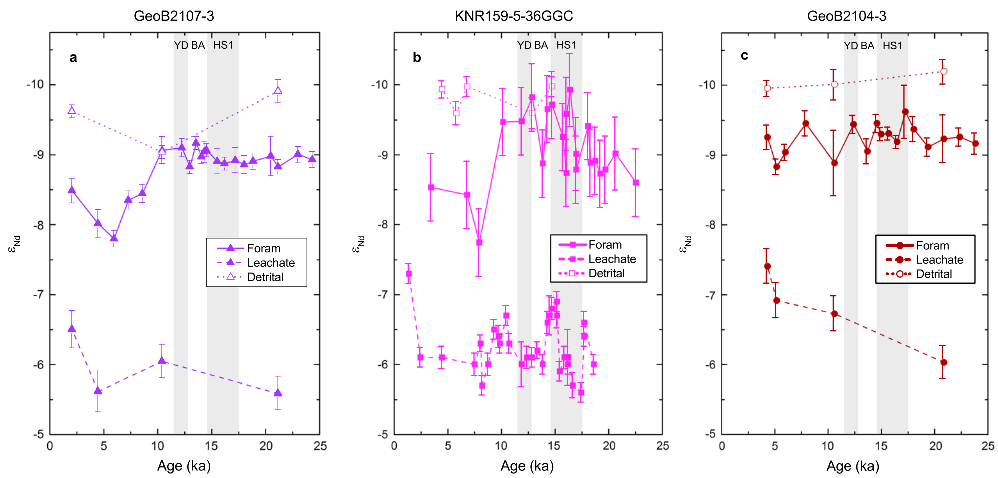

Figure 3. ε_{Nd} of uncleaned foraminifera (filled symbols and solid lines), leachates (filled symbols and dashed lines), and detrital material (hollow symbols and dotted lines) from (a) GeoB2107-3 (purple triangles; 27.2°S, 46.5°W; 1050 m), (b) KNR159-5-36GGC (pink squares; 27.5°S, 46.5°W; 1268 m; leachate and detrital data from *Pahnke et al.* [2008]), and (c) GeoB2104-3 (burgundy circles; 27.3°S, 46.4°W; 1500 m). Climate periods labeled are the Younger Dryas (YD), Bølling-Allerød (BA), and Heinrich Stadial 1 (HS1). See Figure 1 for the location of all records.

and GeoB2104-3 show very little change across the last 25 kyr with all values for the three sites falling between −9 and −10.2 and are less radiogenic than the leachate values as well as most of the foraminiferal values (Figure 3).

4. Discussion

4.1. Intermediate-Depth Leachates

The leachate records of GeoB2107-3 and GeoB2104-3 bracket the leachate ε_{Nd} record of site KNR159-5-36GGC (Figure 3). The similar values of these records is to be expected as we used the same decarbonation and strong chemical leaching method as *Pahnke et al.* [2008]. All three leachate records are more radiogenic than the modern ε_{Nd} of AAIW of around −8.3 [*Stichel et al.*, 2012] throughout the past 25 kyr (Figure 3). *Pahnke et al.* [2008] stated that the detrital fraction could not be causing the offset of the core top measurement to more radiogenic values than modern seawater (Figure 2) as the bulk detrital ε_{Nd} composition of the core was less radiogenic than the seawater values. Since then, however, it has been shown that leachates are susceptible to contamination by detrital material [*Elmore et al.*, 2011] and that this contamination may not reflect the bulk detrital ε_{Nd} composition as more radiogenic material may be preferentially leached [*Wilson et al.*, 2013].

The core top foraminiferal ε_{Nd} values of all three cores agree with the linearly interpolated modern seawater values (Figure 2); although higher-resolution seawater data would improve confidence, this agreement suggests that the decarbonated leachates were, in fact, contaminated by radiogenic neodymium. Furthermore, this contamination must have come from a subcomponent of the detrital material being preferentially leached as the bulk detrital ε_{Nd} values are less radiogenic than the foraminiferal values (Figure 3). This pattern of leachates being more radiogenic than foraminiferal or fish debris ε_{Nd} values but bulk detrital values being less radiogenic has also been reported for intermediate-depth cores on the Demerara Rise further north in the Atlantic [*Huang et al.*, 2014] and in the southern Caribbean Sea [*Xie et al.*, 2014]. It is also clear that the offset between leachates and foraminifera is not constant through time (Figure 3), and therefore, a simple correction cannot be applied to the leachate results to convert them to seawater ε_{Nd} values. This changing offset may result from variations in the sample size to leachate volume ratio [*Wilson et al.*, 2013], as this ratio was not kept constant in this work. The stability of the foraminiferal ε_{Nd} records reveals that the deglacial peaks in the leachate record from KNR 159-5-36GGC (Figure 3) are not due to changes in AAIW penetration into the South Atlantic [*Pahnke et al.*, 2008]. These observations support the assertion that decarbonated leachate ε_{Nd} records should be verified by other authigenic phases such as foraminifera or fish debris before

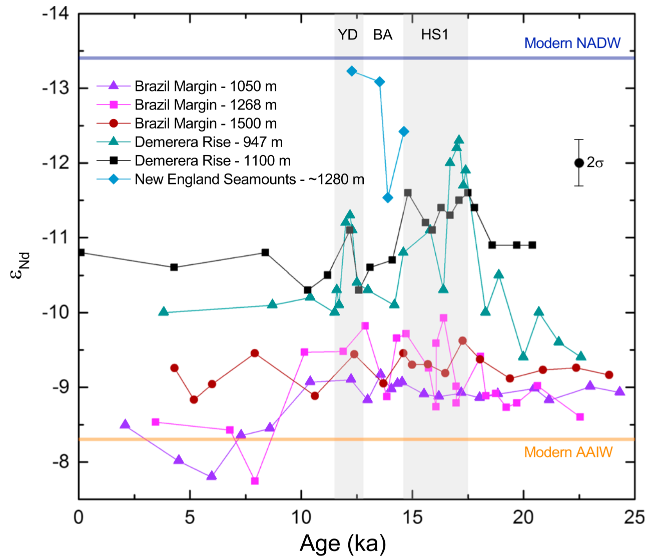

Figure 4. Deglacial records of ε_{Nd} from uncleaned foraminifera from GeoB2107-3 (purple triangles; 27.2°S, 46.5°W; 1050 m), KNR159-5-36GGC (pink squares; 27.5°S, 46.5°W; 1268 m), GeoB2104-3 (burgundy circles; 27.3°S, 46.4°W; 1500 m) , KNR197-3-46CDH (green triangles; 7.8°N, 53.7°W; 950 m [*Huang et al.*, 2014]), and KNR197-3-9GGC (black squares; 7.9°N, 53.6°W; 1100 m [*Huang et al.*, 2014]). Coral records from the New England Seamounts (light blue diamonds; ~39°N, ~61°W; ~1280 m [*van de Flierdt et al.*, 2006; *Wilson et al.*, 2014]). The orange line gives the ε_{Nd} of modern Antarctic Intermediate Water (AAIW) [*Stichel et al.*, 2012], and the purple line that of modern upper North Atlantic Deep Water (NADW) [*Lambelet et al.*, 2016]. 2σ gives the average analytical error climate periods labeled are the Younger Dryas (YD), Bølling-Allerød (BA), and Heinrich Stadial 1 (HS1). See Figure 1 for the location of all records.

being interpreted as a hydrographic signal [*Piotrowski et al.*, 2012; *Xie et al.*, 2014] and indicate that unverified leachate records should no longer be included in studies that model changes in ε_{Nd} of Atlantic seawater during millennial-scale climate events [*Friedrich et al.*, 2014].

It is interesting to note, however, that the excursions in the leachate record of KNR159-5-36GGC (Figure 3b) do appear to correlate with deglacial climate changes [e.g., *Clark and Shakun*, 2012]. We propose that this similarity may be due to changes in the neodymium isotopic composition of the detrital fraction as is seen at sites further north in the Atlantic Ocean (Figure S2) (*Huang et al.* [2014] and this study). These excursions in the detrital records could be driven by changes in the main sources of terrigenous sediments from South America reaching the western Atlantic [*White et al.*, 1985; *McDaniel et al.*, 1997; *de Mahiques et al.*, 2008; *Zhang et al.*, 2015] associated with shifts in the climate [e.g., *Clark and Shakun*, 2012]. This hypothesis could be tested by measuring higher-resolution detrital records from these southern Brazil margin sites since the composition and origin of the terrigenous sediments are well known [*de Mahiques et al.*, 2008; *Razik et al.*, 2015], although that is outside the scope of the present work.

4.2. Deglacial Intermediate-Depth Circulation in the Atlantic

The offset of the GeoB2107-3 (1000 m) foraminiferal ε_{Nd} record to more radiogenic values than GeoB2104-3 (1500 m) throughout the last 25 kyr (Figure 4) suggests that our shallowest site (GeoB2107-3) was bathed by a greater proportion of more radiogenic AAIW. Only during the Holocene, however, is this offset consistently outside of analytical error. The foraminiferal ε_{Nd} record of KNR159-5-36GGC varies from being within analytical error of both the GeoB2107-3 and GeoB2104-3 records during the LGM, to mostly within the error of GeoB2104-3 during the deglaciation and then closer to the record of GeoB2107-3 in the Holocene (Figure S1). These shifts may reflect subtle changes in the depth of the water mass boundary between AAIW and upper NADW; however, the only observation that appears to be significantly outside of analytical error is that there is more overlap between the two shallower sites (GeoB2107-3 and KNR159-5-36GGC) during the Holocene than during the deglaciation (Figure S1).

The stability of the three foraminiferal ε_{Nd} records from 25 to ~10 ka (Figure 3) suggests an unchanged proportion of AAIW at these sites across that time. It must be noted, however, that the end-member composition of northern- and southern-sourced intermediate water masses may have varied through time. Coral measurements in the intermediate-depth North Atlantic reveal that northern-sourced intermediate-depth waters showed similar-to-modern ε_{Nd} values during HS1 (Figure 4) [*van de Flierdt et al.*, 2006; *Wilson et al.*, 2014]. In contrast, foraminiferal records from the Demerara Rise exhibit unradiogenic ε_{Nd} peaks during HS1 and the YD that are not seen at the Brazil margin sites (Figure 4). Although these unradiogenic peaks in the Demerara Rise records should be interpreted with some caution as two of the foraminiferal records show a strong correlation with the corresponding detrital values (Figure S2), that correlation is deemed unlikely to be due to postdiagenetic detrital overprinting of the foraminiferal signal ([*Taylor and McLennan*, 1985; *Gutjahr et al.*, 2007; *Kraft et al.*, 2013] see supporting information). Given that the North Atlantic coral values do not suggest a change in the northern-sourced end-member was responsible for the peaks at the Demerara Rise, these peaks are more likely to represent changes in water mass mixing as they were originally interpreted [*Huang et al.*, 2014] or else it is possible that slow circulation during these Northern Hemisphere cold events may have led to local modification of seawater values ([*Lacan and Jeandel*, 2005; *Siddall et al.*, 2008; *Bradtmiller et al.*, 2014; *Roberts and Piotrowski*, 2015] further discussion in the supporting information). Either way, the Demerara Rise records always exhibit less radiogenic values, indicating a lower proportion of AAIW, than the southern Brazil margin sites throughout the past 25 ka.

In terms of the southern-sourced end-member, a measurement from a single coral in the Drake Passage suggests that intermediate-depth water in the Southern Ocean may have been more radiogenic during HS1 [*Robinson and van de Flierdt*, 2009]. However, this coral was retrieved from a site located to the south of the AAIW formation area in the modern ocean (Figure 1, green star); therefore, it may not be representative of past AAIW ε_{Nd}. Although this uncertainty in the end-member composition of water masses during the deglaciation precludes calculation of water mass mixing proportions, the comparison of the southern Brazil margin records with intermediate-depth ε_{Nd} records measured on foraminifera and corals from the North Atlantic (Figure 4) clearly shows that intermediate water masses in the North and South Atlantic were isotopically distinct throughout the deglaciation. It seems unlikely that the stability of the ε_{Nd} values from the southern Brazil margin sites across the deglaciation is simply a coincidence due to competing end-member changes, although this possibility cannot be ruled out without improved end-member constraints, especially from the South Atlantic.

The divergence of the ε_{Nd} records from the Brazil margin and Demerara Rise during HS1 and the YD argues against the greater northward penetration of AAIW during these intervals, which has been concluded by some studies [*Rickaby and Elderfield*, 2005; *Pahnke et al.*, 2008], but rather, indicates a lesser influence of AAIW in the Northern Hemisphere during abrupt Northern Hemisphere cold events, as concluded by other studies [*Came et al.*, 2003, 2008; *Xie et al.*, 2012; *Huang et al.*, 2014]. Furthermore, we see no evidence for AAIW being denser than, and therefore underlying, northern-sourced intermediate waters during HS1, as has been suggested by some studies [*Keeling and Stephens*, 2001; *Rickaby and Elderfield*, 2005]. If this density reversal had occurred, the seawater bathing GeoB2107-3 (1000 m) would have been less radiogenic than that bathing GeoB2104-3 (1500 m), which is not observed (Figure 4).

4.3. Holocene Antarctic Intermediate Water Production

The shift of the southern Brazil margin foraminiferal ε_{Nd} to more radiogenic values during the mid-Holocene (Figure 4) suggests a change in the nature of AAIW production relative to the deglaciation. The KNR159-5-36GGC ε_{Nd} record, from the core located in the middle in terms of depth (1268 m), shifts from being largely within error of the record from the deeper site (GeoB2104-3; 1500 m) during the deglaciation to agreeing better with the shallower record (GeoB2107-3; 1050 m) during the middle- to late-Holocene (Figure S1). This implies an expansion in the depth range of AAIW during the mid-Holocene relative to the deglaciation. This shift may have been caused in part by stronger AAIW production during the Holocene increasing the proportion of AAIW in the intermediate-depth South Atlantic relative to the deglaciation.

As the ε_{Nd} values of GeoB2107-3 become more radiogenic (−7.8) during the mid-Holocene than the typical value of AAIW in the modern ocean (~ −8.3) [*Stichel et al.*, 2012], however, an end-member shift in the ε_{Nd} of AAIW must also be considered (Figure 4). Other intermediate-depth ε_{Nd} records from the Atlantic do not sample the Holocene at the same resolution as this work [*Huang et al.*, 2014; *Xie et al.*, 2014]; thus, no comparison can be made to investigate whether this signal was propagated further north into the tropical Atlantic during the mid-Holocene. A record from the intermediate-depth Caribbean Sea (Figure 5b) shows higher coarse fraction percentages during the mid-Holocene that were interpreted as a greater influence of more corrosive AAIW at that time [*Schmidt et al.*, 2012; *Xie et al.*, 2014]. However, the neodymium isotope analyses from that core are not of sufficient resolution to confirm this interpretation [*Xie et al.*, 2014].

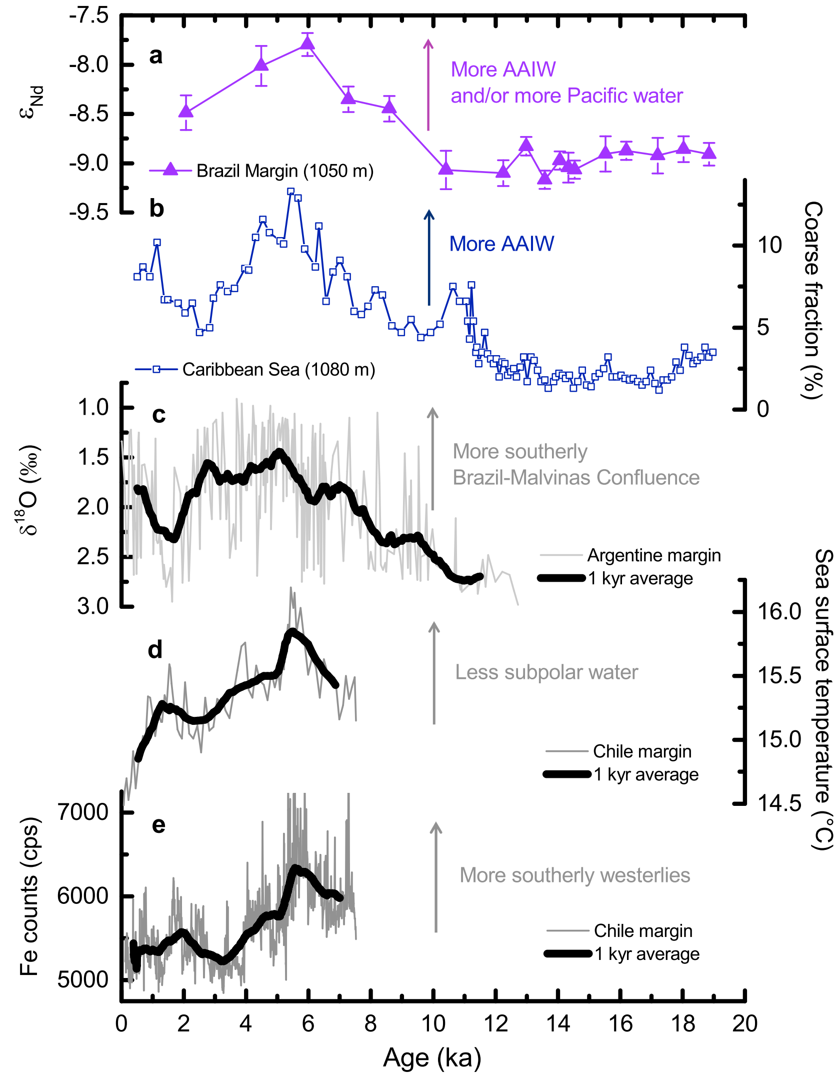

Figure 5. (a) Deglacial record of ε_{Nd} from uncleaned foraminifera from GeoB2107-3 (27.2°S, 46.5°W; 1050 m); more radiogenic (positive) values imply more Antarctic Intermediate Water (AAIW) or more radiogenic AAIW due to a greater proportion of Pacific-derived intermediate water. (b) Coarse fraction percentage from VM12-107 (11.3°N, 66.6°W; 1079 m [*Schmidt et al.*, 2012; *Xie et al.*, 2014]) where a higher coarse fraction percentage represents a greater proportion of corrosive AAIW dissolving fine grained particles. (c) Stable oxygen isotope data of *Globorotalia inflata* (sea level corrected) for sediment core GeoB13862-1 (38.0°S, 53.7°W; 3588 m) on the Argentine margin. A lower $\delta^{18}O$ represents a more southerly position of the Brazil-Malvinas Confluence, and an inferred more southerly position of the Southern Hemisphere westerlies [*Voigt et al.*, 2015]. (d) Sea surface temperature reconstruction from GeoB3313-1 (41.0°S, 74.5°W; 852 m), where warmer temperatures represent a lesser influence of cooler subpolar surface waters [*Lamy et al.*, 2002]. (e) Iron counts from GeoB3313-1 controlled by rainfall and therefore indicating the relative position of the Southern Hemisphere westerlies [*Lamy et al.*, 2001]. See Figure 1 for the location of all records.

In the modern ocean, AAIW formation occurs in the Southern Ocean between the Polar Front and the Subantarctic Front [*Meredith et al.*, 1999]. The timing of the mid-Holocene values at site GeoB2107-3 that are more radiogenic than modern AAIW (Figure 5a) coincides with evidence for a more southerly position of the Southern Hemisphere westerlies as inferred from (i) iron counts from a core collected from the Chilean margin (Figure 5e) [*Lamy et al.*, 2001] and (ii) stable oxygen isotope values from a core collected from the Argentinean margin (Figure 5c) [*Voigt et al.*, 2015]. Furthermore, sea surface temperature reconstructions from the Chilean margin site show warmer temperatures during the mid-Holocene (Figure 5d) that were interpreted as a lesser influence of subpolar waters in the South Pacific [*Lamy et al.*, 2002]. This correlation suggests that the southward position of the westerly winds and the associated fronts in the Southern

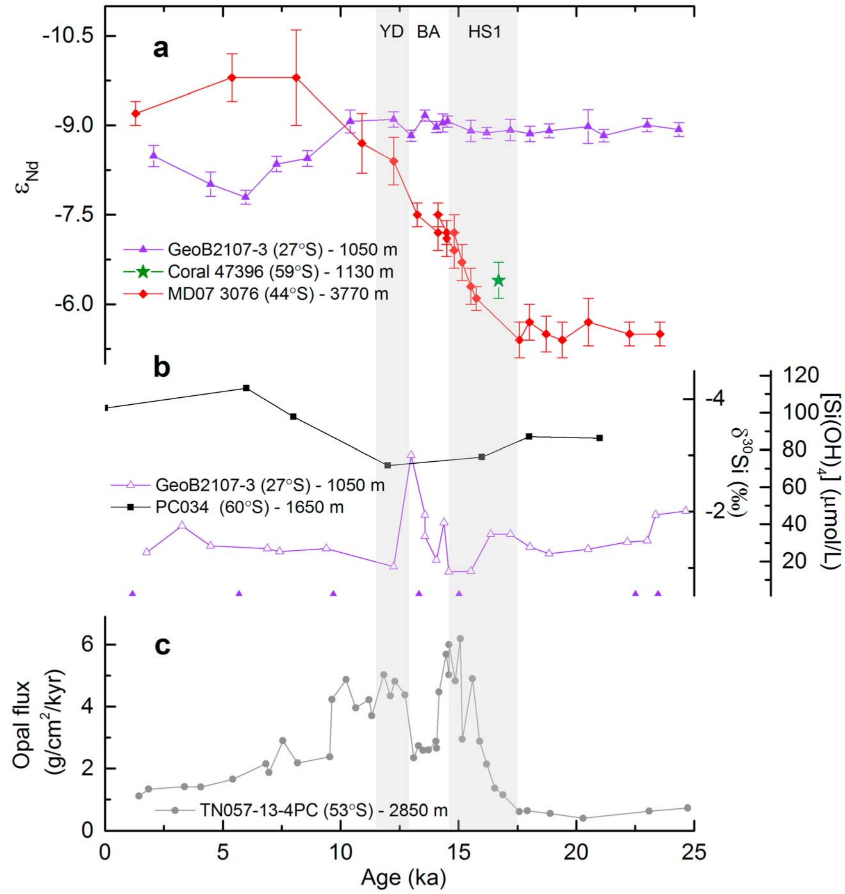

Figure 6. (a) Deglacial ε_{Nd} records measured on foraminifera from the intermediate-depth southern Brazil margin core GeoB2107-3 (purple triangles; 27.2°S, 46.5°W; 1050 m), deep South Atlantic core MD07-3076 (red diamonds; 44.1°S, 14.2°W; 3770 m [*Skinner et al.*, 2013]), and an intermediate-depth Drake Passage coral (green star; ID: 47396, 59.4°S, 68.5°W; 1130 m [*Robinson and van de Flierdt*, 2009]). The error bars are 2σ external error. (b) Deglacial silicon isotope measurements from deep-sea sponge spicules also from the intermediate-depth Brazil margin core GeoB2107-3 (open purple triangles [*Hendry and Robinson*, 2012]) and middepth Southern Ocean (black squares; PC034; 59.8°S, 39.6°W; 1650 m [*Hendry et al.*, 2010]). Conversion of δ^{30}Si to ((Si(OH)$_4$) taken from *Hendry et al.* [2010]. Age control points from planktic radiocarbon measurements for GeoB2107-3 are shown by the filled purple triangles [*Heil*, 2006]. (c) Opal flux from the Southern Ocean core TN057-13-4PC (grey circles; 53.2°S, 5.1°E; 2850 m [*Anderson et al.*, 2009]). Climate periods labeled are the Younger Dryas (YD), Bølling-Allerød (BA), and Heinrich Stadial 1 (HS1). See Figure 1 for the location of all records.

Ocean caused stronger AAIW formation and/or resulted in more Pacific-like radiogenic water being incorporated into Atlantic AAIW. The deep Atlantic can be ruled out as the source of the radiogenic peak, as deglacial records from the deep Atlantic tend to show unradiogenic peaks in the early Holocene (Figure 6a) [*Piotrowski et al.*, 2004; *Roberts et al.*, 2010]. The different mid-Holocene ε_{Nd} values at intermediate and deep sites suggest that the ε_{Nd} homogeneity of South Atlantic seawater in the modern ocean [*Stichel et al.*, 2012] may be coincidence rather than intrinsic to this region. It has been suggested that the less radiogenic values in the deep Atlantic during the early to mid-Holocene (Figure 6a) may correspond to stronger NADW production [*Lippold et al.*, 2016]. This could indicate a coupling between NADW production in the mid-Holocene and the strong intermediate water production we observe here in the South Atlantic.

4.4. Deglacial South Atlantic Nutrient Sourcing

A reconstruction of bottom water silicic acid concentration from GeoB2107-3 [*Hendry et al.*, 2012] revealed peaks in silicic acid concentration during HS1 and the early YD, with the early YD peak resembling the concentration of a Southern Ocean site [*Hendry et al.*, 2010] (Figure 6b). These silicic acid peaks coincide, within the error of the age models (Figure 6b), with periods of elevated opal accumulation in the Atlantic sector of the Southern Ocean (Figure 6c) that were interpreted as evidence of enhanced upwelling in the Southern

Ocean during HS1 and the YD [*Anderson et al.*, 2009]. Similar opal accumulation peaks have been reported in the lower latitude Atlantic Ocean [*Romero et al.*, 2008; *Meckler et al.*, 2013], indicating an enhanced supply of silicic acid to the surface ocean in those regions during HS1 and the YD. The source of those nutrients to the surface waters of the low-latitude Atlantic is, however, debated [e.g., *Meckler et al.*, 2013; *Hendry et al.*, 2016].

Hendry et al. [2012] interpreted the early YD overlap between the $\delta^{30}Si$ records of GeoB2107-3 and PC034 (Figure 6b) to indicate the entrainment of unaltered upwelled Circumpolar Deep Water/Antarctic Bottom Water into intermediate water formed in the Atlantic sector of the Southern Ocean. The intersection of the ε_{Nd} records of bottom water at GeoB2107-3 and at deep South Atlantic site MD07-3076 during the YD (Figure 6a) [*Skinner et al.*, 2013] could be taken to support this assertion. It should, however, be noted that the similarity of the ε_{Nd} values of the two sites is not due to an excursion in the intermediate-depth ε_{Nd} values—which are constant across this time period—to deepwater values, but rather the ε_{Nd} of the deep South Atlantic, which had been steadily trending toward less radiogenic since early in the deglaciation, reached values similar to those seen at intermediate depths during the YD; thus, the overlap could be coincidence. However, benthic foraminiferal $\delta^{13}C$ records also show a breakdown in the strong chemical stratification between the intermediate and deep South Atlantic during the YD [*Roberts et al.*, 2016].

Unlike the YD, the HS1 silicic acid peak at GeoB2107-3 (Figure 6b) was interpreted as a possible shoaling of the Si nutricline (the depth at which silica redissolved), resulting in the site sitting under the core of higher silicic acid AAIW [*Hendry et al.*, 2012]. The lack of change in ε_{Nd} at GeoB2107-3 argues against any significant change in water mass mixing proportions during HS1, but does not negate the possibility of greater dissolution of silica at that depth during HS1. *Meckler et al.* [2013] concluded that the low-latitude opal accumulation peaks arose due to the shoaling of nutrient-rich, southern-sourced, deep water to the bottom of the thermocline during abrupt Northern Hemisphere cold events. Although our study sites are at higher latitudes, the 2 to 3 epsilon unit offset between the intermediate and deep South Atlantic during HS1 (Figure 6a) reveals that intermediate water in the South Atlantic remained chemically distinct from the deep South Atlantic at this time. Evidence for direct upwelling of deep water is only seen by a coral ε_{Nd} measurement (Figure 6a) from intermediate depths further south in the Southern Ocean (Figure 1). Foraminiferal radiocarbon data show that the intermediate-depth South Atlantic was better ventilated than the deep South Atlantic during HS1 [*Skinner et al.*, 2010; *Sortor and Lund*, 2011; *Freeman et al.*, 2015]. This finding suggests that the low-latitude opal accumulation peaks are more likely due to enhanced silicic acid concentrations near the base of the thermocline that were transported to the surface water by greater upwelling and/or weakened stratification [*Romero et al.*, 2008; *Hendry et al.*, 2016] than to direct upwelling of deep southern-sourced waters [*Meckler et al.*, 2013].

5. Conclusions

This study presents new records of leachate, detrital, and foraminiferal ε_{Nd} from cores GeoB2107-3 and GeoB2104-3, and foraminiferal ε_{Nd} from core KNR159-5-36GGC, collected from intermediate depths on the southern Brazil margin in the South Atlantic. The leachate measurements are offset to more radiogenic values than the foraminiferal ε_{Nd} values, revealing that strong chemical leaching following decarbonation is not suitable for reconstructing seawater ε_{Nd} in this location. The bulk detrital values from the same cores are less radiogenic than both the leachates and foraminiferal values, indicating that these leachates must be contaminated by a subcomponent of the detrital material.

The foraminiferal ε_{Nd} records exhibit little change across the last deglaciation and, when compared with North Atlantic intermediate-depth records, confirm that AAIW did not extend further into the South Atlantic during HS1 and the YD. The only significant change in the foraminiferal ε_{Nd} records from the southern Brazil margin is a mid-Holocene peak of more radiogenic values in the two shallower cores. These more radiogenic values suggest that during the mid-Holocene AAIW formation may have been stronger than during the deglaciation and/or that Pacific waters with a more radiogenic ε_{Nd} signal had a greater influence on Atlantic AAIW, possibly due to a more southerly position of the Southern Hemisphere westerlies.

Comparison of the new intermediate-depth ε_{Nd} records with a deep South Atlantic site and with silicon isotope data helped to place constraints on the source of nutrient pulses to the low-latitude Atlantic during abrupt climate events of the last deglaciation. They argue against the direct upwelling of deep southern-sourced waters to intermediate depths in the South Atlantic during HS1.

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