Tracing water mass mixing and continental inputs in the southeastern Atlantic Ocean with dissolved neodymium isotopes

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Abstract

In contrast to the vigorous deep ocean circulation system of the north- and southwestern Atlantic Ocean, no systematically sampled datasets of dissolved radiogenic neodymium (Nd) isotope signatures exist to trace water mass mixing and provenance for the more restricted and less well ventilated Angola Basin and the Cape Basin in the southeastern Atlantic Ocean, where important parts of the return flow of the Atlantic Meridional Overturning Circulation are generated. Here, to improve our understanding of water mass mixing and provenance, we present the first full water column Nd isotope (expressed as ε_{Nd} values) and concentration data for a section across the western Angola Basin from 3° to 30° S along the Zero Meridian and along an E-W section across the northern Cape Basin at 30° S sampled during GEOTRACES cruise GA08. Compared with the southwestern Atlantic basin we find overall less radiogenic ε_{Nd} signatures reaching -17.6 in the uppermost 200 m of the Angola and Cape basins. In the western Angola Basin these signatures are the consequence of the admixture of a coastal plume originating near 13 °S and carrying an unradiogenic Nd signal that likely resulted from the dissolution of Fe-Mn coatings of particles formed in river estuaries or near the West African coast. The highly unradiogenic Nd isotope signatures in the upper water column of the northern Cape Basin, in contrast, originate from old Archean terrains of southern Africa and are introduced into the Mozambique Channel via rivers like the Limpopo and Zambezi. These signatures allow tracing the advection of shallow waters via the Agulhas and Benguela currents into the southeastern Atlantic Ocean. The Nd isotope compositions of the deep water masses in both basins primarily reflect conservative water mass mixing with the only exception being the central Angola Basin, where the signatures are significantly overprinted by terrestrial inputs. Bottom waters of the Cape Basin show excess Nd concentrations of up to 6 pmol/kg (20%), originating from resuspended bottom sediments and/or dissolution of dust, but without significantly changing the isotopic composition of the waters due to similar ε_{Nd}

values of particles and bottom waters ranging between -9.6 and -10.5. Given that bottom waters within the Cape Basin today are enriched in Nd, non-conservative Nd isotopic effects may have been resolvable under past glacial boundary conditions when bottom waters were more radiogenic.

1. Introduction

The Atlantic Meridional Overturning Circulation (AMOC) controls meridional heat and salt transport in the Atlantic Ocean and thus exerts important control on present and past global climate. It is also responsible for the transfer of gases such as O_2 and CO_2 from the atmosphere to the deep ocean and thus controls the ventilation and alkalinity of the deep Atlantic Ocean. The AMOC is known to have undergone major changes in the recent past as well as on longer time scales during the Late Quaternary, and its reconstruction based on different geochemical tools has been a major objective of paleoceanographic research (cf. Curry and Oppo, 2005; Böhm et al., 2015).

Radiogenic neodymium (Nd) isotopes are widely used as tracers for present and past large-scale water mass mixing processes. This is possible due to the quasi-conservative behavior of Nd and its average oceanic residence time of 300-1000 yr (Arsouze et al., 2009; Rempfer et al., 2011; Tachikawa et al., 2003). Nd is introduced into the oceans via particulate and dissolved loads of rivers and aeolian dust (Goldstein et al., 1984; Frank, 2002; Goldstein and Hemming, 2003) as well as through exchange with shelf and slope sediments (Lacan and Jeandel, 2001, 2005b). The Nd isotope ratio ($^{143}Nd/^{144}Nd$) of water masses is commonly expressed as $\varepsilon_{Nd} = [(^{143}Nd/^{144}Nd)_{sample}/(^{143}Nd/^{144}Nd)_{CHUR} - 1] \times 10^4$ with CHUR = 0.512638 (Jacobsen and Wasserburg, 1980).

Neodymium isotope data for the Atlantic Ocean, recently extended and refined by the international GEOTRACES program, closely track conservative intermediate and deep water mass mixing of the AMOC (Stichel et al., 2012a, b Lambelet et al., 2016, van de Flierdt et al., 2016, Zieringer et al., 2019). North Atlantic Deep Water (NADW) formed by mixing of source waters in the Labrador and Nordic Seas is characterized by an ε_{Nd} signature of -13.2 to -13.5 (Piepgras and Wasserburg, 1987, Lacan and Jeandel, 2005a, Lambelet et al., 2016). In the subtropical and tropical Atlantic, NADW mixes with Antarctic Intermediate Water (AAIW) and Antarctic Bottom Water (AABW) (Naveira-Garabato et al. 2002). AAIW is formed by mixing of Sub-Antarctic Surface Water with sinking Antarctic Surface Water in the South Atlantic Ocean and is characterized by an average ε_{Nd} signature of -8.7 (Jeandel, 1993), whereas AABW is formed by mixing of cold Antarctic Shelf Water with warmer Circumpolar Deep Water (Orsi et al., 1999) having ε_{Nd} values ranging between -8.6 and -9.6 (Jeandel, 1993; Stichel et al., 2012b).

The application of radiogenic Nd isotopes as conservative water mass tracers is, however, not possible in places such as ocean boundaries, where significant continental inputs of Nd occur via rivers, dust or sedimentary exchange processes (Lacan and Jeandel, 2005b). Nevertheless, pronounced changes in the mixing between Southern Ocean waters and Northern Component waters in the restricted Angola Basin and the Cape Basin of the southeastern Atlantic Ocean (Fig. 1) during the Late Quaternary have been inferred from deep water ε_{Nd} signatures obtained by leaching of sedimentary Fe-Mn oxyhydroxides (Jonkers et al., 2015, Klevenz et al., 2008, Piotrowski et al., 2005, Wei et al., 2016). In the modern water column of the Angola Basin, highly unradiogenic ε_{Nd} signatures varying between -13.9 and -11.1 are found for AAIW, which cannot be explained by conservative mixing. Instead, partial dissolution of ferromanganese oxides originating from the Congo River under low oxygen conditions (Rickli et al., 2009, 2010) or near the African shelf (Zheng et al., 2016) have been invoked.

Relatively unradiogenic Nd isotope compositions of AAIW have also been found southwest of Africa (ε_{Nd} of -9.3) and have been associated with entrainment of the Agulhas Current (AC) (Stichel et al., 2012a). The lack of seawater Nd isotope and concentration data from the Angola and Cape Basins has, however, hindered a systematic assessment of the contribution of Nd from the AC, which represents the main surface return flow of the AMOC from the Indian Ocean.

Here we present the first detailed study of the distribution of dissolved Nd isotopic compositions and concentrations in the Angola Basin and in the northern Cape Basin based on filtered seawater samples from 20 full water column profiles collected during GEOTRACES cruise GA08 (Fig. 1). We constrain the potential origins of unradiogenic Nd isotopic signatures of surface and deep waters and demonstrate that dissolved Nd isotopes can serve to reliably trace deep water mass mixing in most parts of the southeastern Atlantic Ocean, where deep circulation is more restricted than that of the western South Atlantic.

1.1. Hydrography

The Benguela Current (BC) is the dominant surface current of the southeastern Atlantic Ocean, originating from the Agulhas Current and introduced via the Agulhas Leakage (Stramma and England, 1999). The BC flows north along the West African coast until it meets the Angola Current at the Angola-Benguela Front (ABF) near 15 °S. A second branch of the BC feeds into the South Equatorial Current, which flows in a northwesterly direction across the entire South Atlantic. Near the Brazilian coast it changes direction and then flows eastward across the South Atlantic as the South Equatorial Counter Current that feeds into the cyclonic Angola Gyre (AG) (Stramma and England, 1999) (Fig. 1). The AG is bordered by the Angola Front in the north and by the ABF in the south. South of the Angola Basin, between the Walvis Ridge and Cape Agulhas (34 °S), the southeasterly winds cause strong coastal upwelling (Meeuwis and Lutjeharms, 1990).

Surface waters in the northern part of the Angola Basin are dominated by Tropical Surface Water (TSW), which is characterized by temperatures near 27°C (station 24, 25) (Fig. 2) and constitutes the mixed layer of most of the tropical Atlantic. The mixed layer of the southern part of the Angola Basin and the Cape Basin is occupied by Subtropical Surface Water (STSW) as indicated by temperatures near 20°C (Fig. 2). TSW and STSW are underlain by South Atlantic Central Water (SACW) (Fig. 2) (Sverdrup et al., 1942, Stramma and England, 1999). SACW is transported into the subtropical gyre by the South Atlantic Current, which feeds into the BC. SACW in the tropical Atlantic partly originates from Indian Central Water (ICW), which is advected into the Atlantic Ocean by the Agulhas Current (Stramma and Schott, 1999). Intermediate waters are characterized by a low salinity ranging between 34.3 and 34.6, potential temperatures between 4 and 6 °C and neutral densities of $27.13\ kg/m^3 \leq \gamma^n \leq 27.55\ kg/m^3$, which are typical for nutrient-rich Antarctic Intermediate Water (AAIW) (Fig. 2) (Whitworth and Nowlin, 1987). The AAIW in the western South Atlantic originates from the surface of the Antarctic Circumpolar Current (ACC) and is subducted northwards at the Polar Front between 50° and 40°S. In the South East Atlantic Ocean (SEAO), AAIW originates from the Indian Ocean and is advected as part of the Agulhas Current leakage (Roman and Lutjeharms, 2010, Stramma and England, 1999). Northward propagation of AAIW occurs between 500 and 1200 m water depth (Talley, 1996). Below AAIW, Upper Circumpolar Deep Water (UCDW) prevails at salinities between 34.8 and 34.6, potential temperatures between 3 and 4°C and neutral densities of $27.55\ kg/m^3 \leq \gamma^n \leq 27.8\ kg/m^3$ (Fig. 2). The oxygen-poor and nutrient-rich UCDW also originates from the ACC, propagates northwards and loses its characteristics through mixing by the time it reaches the equator (Stramma and Schott, 1999). In the Angola Basin, deep and bottom waters below AAIW are dominated by North Atlantic Deep Water (NADW), characterized by higher salinities between 34.8 and 35, potential temperatures between 2 and 3 °C and neutral densities of 27.8 $kg/m^3 \leq \gamma^n \leq 28.12\ kg/m^3$ (Fig. 2). NADW is advected into the SEAO via a branch of the Deep Western Boundary Current that forms near the equator (Rhein et al., 1995) and enters the Angola Basin across the Romanche Fracture Zone. Bottom waters of the Cape Basin predominantly consist of Lower Circumpolar Deepwater (LCDW) ($\gamma^n \geq 28.12\ kg/m^3$), which is markedly distinct from the Angola Basin due to the fact that the Walvis Ridge prevents northward advection of LCDW and Antarctic Bottom Water (AABW) (Fig. 2) (e.g. Rickli et al., 2009).

The Agulhas Current is the largest boundary current in the world's ocean and originates from the South Equatorial Current in the tropical Indian Ocean, which is a mixture of contributions from the Tasmanian leakage, the Indonesian Throughflow, the Red Sea and the Arabian Sea (Durgadoo et al., 2017). It bifurcates near the northeastern tip of Madagascar and feeds warm and saline waters into the Madagascar and Mozambique currents (Stramma and Lutjeharms, 1997) (Fig. 1). The Agulhas Current then flows along the east coast of South Africa, detaches from the continent at the Agulhas Bank and is partly retroflected into the Indian Ocean as the Agulhas Return Current. The remaining waters are advected into the SEAO via the Agulhas Leakage, as cyclonic and anticyclonic eddies and filaments, thereby feeding the surface return flow of the AMOC (Loveday et al., 2014).

The main water masses in the Mozambique Channel contributing to the upper water column and ultimately to the Agulhas Current are Tropical Surface Water (TSW) and Subtropical Surface Water (STSW) (Ullgren et al., 2012). The TSW or Equatorial Surface Water (Sæter and Jorge da Silva, 1984) is a warm (28 °C) and low salinity water mass (<34.5), carried by the South Equatorial Current–into the central Mozambique Channel (Tomczak and Godfrey, 1994). The STSW is also a warm (21-28 °C), but highly saline water mass (35.2-35.5)

(Ullgren et al., 2012) that prevails in the upper 300 m of the southern Channel (Sæter and Jorge da Silva, 1984).

1. Methods

Seven near surface water samples were taken during Meteor Cruise M75-3 in 2008 from the runoff-influenced areas of the Zambezi and Limpopo Rivers in the Mozambique Channel. 106 water samples from the Angola and Cape Basin were taken during GEOTRACES cruise GA08 (RV Meteor cruise M121) in November/December 2015 along the Zero Meridian between 3°S and 30°S, followed by an E-W section along 30°S between 0° and 17°E near the South African coast (Fig. 1). Samples from the full water column were collected with 10 l Niskin bottles attached to a stainless steel CTD rosette, while surface water samples were recovered with a towed stainless steel fish. The samples were then treated in the onboard laboratory strictly following recommended GEOTRACES protocols (van de Flierdt et al., 2012). Each 20 L sample was filtered through a nitro-cellulose acetate filter (0.45 µm pore diameter) into an acid-cleaned LDPE-cubitainer with a peristaltic pump within 2 h after sample collection, and subsequently acidified with ~20 ml concentrated, distilled HCl. For Nd concentration measurements, 2 L aliquots from each filtered sample were collected in acid-cleaned 2-liter PE-bottles. To each large volume sample 400 µl $FeCl_3$ solution (~200 mg Fe/ml) were added and the sample was left to equilibrate for 24 h. Ammonia solution (25 %, Merck Suprapur®) was then added to raise the pH from about 2 to 7.5-8.0. After 48 h, the trace elements co-precipitated with the FeOOH settled to the bottom of the cubitainers and the supernatant was syphoned off.

The precipitates were transported to the home laboratory at GEOMAR in 2 L bottles and were centrifuged and rinsed three times with deionized water (MilliQ, 18.2 MΩcm) in 50 ml centrifuge tubes to remove major seawater ions. After dissolution in 6 M HCl/0.5 M HF and transfer into Teflon vials the samples were evaporated to dryness. To remove organic compounds, the samples were treated with aqua regia at 120 °C for 24 h. Most of the Fe was subsequently removed via liquid–liquid extraction with pre-cleaned di-ethyl ether (Stichel at al., 2012b). The rare earth elements (REEs) were chromatographically separated from matrix elements using cation exchange resin AG 50W-X8 (1.4 ml, 200-400 µm) and following a modified protocol of Münker et al. (2001). Neodymium was then separated from the other REEs for isotope measurements using Eichrom®LN-Spec resin (2 ml, 50-100 µm) following a modified protocol of Pin and Zalduengui (1997). To remove residual traces of the resin and organic compounds, the Nd cuts were treated with 100 µl quartz distilled HNO_3 and 100 µl H_2O_2 (30 wt.%, Merck Suprapur®).

For the determination of Nd concentrations, 1 L aliquots were spiked with a pre-weighed ^{150}Nd spike and then purified with the same cation column chemistry that was used for the Nd isotope separation. Nd concentrations were then determined via isotope dilution on a Nu Plasma MC-ICPMS (Nu Instruments).

The ^{143}Nd/^{144}Nd ratios of 25 samples were measured on a Nu Plasma MC-ICPMS and were corrected for instrumental mass bias to ^{146}Nd/^{144}Nd = 0.7219, using an exponential mass fractionation law. Isobaric interferences between ^{144}Sm and ^{144}Nd were corrected by measuring the abundance of the interference-free isotope ^{147}Sm and by calculating the potential ^{144}Sm contribution on mass 144 from the natural abundance of Sm. Mass bias corrected ^{143}Nd/^{144}Nd, normalized to a ^{146}Nd/^{144}Nd of 0.7219, for the JNdi-1 standard on the Nu Plasma MC-ICPMS ranged from 0.512046 to 0.512086 and on the Neptune Plus MC-ICPMS ranged from 0.512009 to 0.512080. The mass bias corrected ^{143}Nd/^{144}Nd of all samples were normalized to the accepted JNdi-1-standard value of 0.512115 (Tanaka et al., 2000). In the case of the 81 seawater samples measured on our Neptune Plus MC-ICPMS, the ^{143}Nd/^{144}Nd ratios were double-corrected for instrumental mass bias with ^{146}Nd/^{144}Nd = 0.7219 and ^{142}Nd/^{144}Nd = 1.141876 following the approach of Vance and Thirlwall (2002).

The external reproducibility of the Nd isotope measurements was estimated by repeated measurements of in-house Nd standard solutions. For the Neptune Plus, the in-house standard gave reproducibilities between 0.1 and 0.35 ε_{Nd} units (2SD). For the Nu Plasma, the reproducibility of the in-house standard was between 0.2 and 0.4 ε_{Nd} units (2SD). The external reproducibility for Nd concentration measurements was 2 % (2SD, n = 4 sample replicates).

2. Results

Neodymium isotope compositions and concentrations are listed in supplementary Table 1 and are plotted together with previously published data from one station in the Angola Basin (69/21) and one station in the Cape Basin (69/26) (Rickli et al., 2009) in Fig. 3. The data from the two cruises show consistent distributions. In addition, our data are displayed in a section plot together with Nd isotope and concentration data from the Southern Ocean obtained during Polarstern cruise ANT-XXIV/3 (Stichel et al., 2012a, b) (Fig. 4).

3.1. Surface waters

Surface waters in the Angola Basin, and Cape Basin between stations 26 and 40, have highly unradiogenic ε_{Nd} values of -14.5 to -17.6 (Fig. 3). Between stations 41 and 43, in the Benguela Upwelling area above the South African shelf, the εNd signatures are significantly more radiogenic (-8.3 to -13.5) (Fig. 3). Surface water Nd concentrations in the Angola Basin range between 9.8 pmol/kg and 36.1 pmol/kg, with highest concentrations prevailing at station 28 (Fig. 3). The surface water concentrations in the Cape Basin range between 7.5 and 21.9 pmol/kg, with the highest concentrations observed near the coast at station 43 (Fig. 3).

Surface waters off the Zambezi River mouth have ε_{Nd} signatures of -15.5 (GIK16156) and -14.7 (GIK16157) and Nd concentrations reach 50.6 and 63.9 pmol/kg, respectively. Surface waters off the Limpopo River mouth are characterized by ε_{Nd} signatures near -22 and concentrations between 54 and 97 pmol/kg (Supplementary Table 2).

2.2. Intermediate waters

Intermediate waters between 500 and 1500 m have the lowest ε_{Nd}-value of -13.2 in the central Angola Basin (station 28) and the signatures become more radiogenic southwards, reaching ε_{Nd} values of up to -10.4 at station 35 (Figs. 3, 4). Nd concentrations for these waters range between 11.6 pmol/kg (station 35) and 18.5 pmol/kg (station 28). In contrast, the ε_{Nd} signatures of intermediate waters along the Cape Basin section are essentially invariant at values around -10 and Nd concentrations near 12 pmol/kg (Figs. 3, 4).

2.3. Deep and bottom waters

Deep waters in the southern and central Angola Basin between 1500 and 4000 m depth are characterized by ε_{Nd} values between -11 (station 35) and -14.4 (station 28), respectively. In the northern part of the basin, ε_{Nd} values are near -13 and hence are close to typical values for NADW (station 24) (Figs. 3, 4). In contrast, deep Cape Basin waters in the density range of NADW are more radiogenic (ε_{Nd} = -11 to -12.5). At 1500 m water depth, Nd concentrations vary between 13.9 and 22.3 pmol/kg in the Angola Basin, while Nd concentrations of all stations in the Cape Basin are constant at ~14 pmol/kg (Fig. 4). Both basins exhibit a nearly linear increase in Nd concentration with water depth to ~25 pmol/kg at 4000 m (Fig. 4).

Bottom waters in the Angola Basin show εNd values between -11.7 and -13 and Nd concentrations between 24.6 and 29.8 pmol/kg (Fig. 3), whereas the signatures of bottom waters in the Cape Basin are significantly more radiogenic (ε_{Nd} ~ -10) and the Nd concentrations increase to 37 pmol/kg between 4000 and 5000 m water depth (Fig. 3).

3. Discussion

4.1. Sources of unradiogenic Nd

The highly unradiogenic ε_{Nd} values of surface waters in the Angola Basin and Cape Basin of up to -17.6 cannot be explained by water mass mixing processes, and must at least partly be the result of regional terrestrial Nd inputs originating from old continental source rocks. Below we discuss the potential sources of such unradiogenic Nd inputs.

4.1.1. Aeolian dust

A possible source of terrestrial material with unradiogenic neodymium isotope signatures is aeolian dust (Goldstein et al., 1984, Tachikawa et al., 1997, 1999). The Sahara-Sahel Dust Corridor (SSDC) between 12°N and 28°N is the world's largest source of desert-derived dust, reaching an annual production of 400-700 million tons/year (Middleton and Goudie, 2001, Moreno et al., 2006). The dust is transported across the Atlantic Ocean by the trade wind belts (Grousset et al., 1988). However, the deposition of Saharan dust mainly occurs between 10° N and 30° N (Karyampudi et al., 1999, Mahowald et al., 2005, Moreno et al., 2006) and does not pass the inter tropical convergence zone to reach the South Atlantic Ocean. Furthermore, ε_{Nd} signatures of -8.5 and -14.6 in the dust (Goldstein et al., 1984, Grousset et al., 1988) are too radiogenic to explain the surface water signatures of the Angola and Cape Basin. While dust from the Namib and Kalahari desert in southern Africa is transported into the SEAO, the dust concentrations above this region are relatively low (0.06 to 0.23 µg/m^3) (Chester et al., 1972) compared to the Sahara (Mahowald et al., 2005). Most importantly, ε_{Nd} signatures of dust collected above the Angola Basin range between -9.1 and -10.4 and are thus also far too radiogenic to explain the observed highly unradiogenic surface seawater values (Goldstein et al., 1984, Grousset, et al., 1988, Rickli et al., 2010). Based on these observations, aeolian dust input is unlikely to be a major source of the unradiogenic dissolved surface water Nd isotope signatures in the Angola Basin.

4.1.2. Sedimentary Fe-Mn oxides in the Angola Basin

Fe-Mn oxide coatings of marine sediment particles are important carriers of Nd and other REEs and release these elements to ambient seawater under low oxygen conditions (Haley and Klinkhammer, 2003). Nd isotope compositions of Fe-Mn oxides in Congo fan sediments and in Congo River borne shelf and slope sediments further south at 13 °S have an average ε_{Nd} value of -16.3 and -22.9, respectively (Bayon et al., 2004, 2009). Lateral advection of Nd released by the reduction of Fe-Mn coatings of suspended particles in the oxygen minimum zone of the Angola Basin, between 100 and 700 m water depth (Rickli et al., 2010), is a suitable potential source of unradiogenic Nd to the waters of the upper water column and provides a viable explanation for the high surface Nd concentrations at station 28. This is further supported by the observation of a trace metal enriched plume originating from the African coast and extending 2500 km into the subtropical gyre between 11 and 15 °S (Noble et al., 2012, Zheng et al., 2016).

4.1.3. Surface waters of the Cape Basin

With ε_{Nd} values of up -17.6, surface waters in the Cape Basin are less radiogenic than those of the Angola Basin. However, along the Cape Basin transect there is no evidence for a trace metal enriched plume extending from the coast. The Nd isotope compositions of shelf and slope sediments from the Cape Basin (ε_{Nd} = -13.3) and of dust particles (ε_{Nd} = -10.9) (Bayon et al., 2009) are too radiogenic to cause the observed near surface water signatures. Similarly, with ε_{Nd} values varying between -14 to -12 (Franzese et al. 2006), the Nd isotope compositions of sediments along the proximal South African coast are too radiogenic to explain the observed highly unradiogenic signatures. However, Nd isotope signatures of -9 to -13 of surface waters at stations 41-43 are likely caused by partial dissolution of suspended sediments from the Orange River, which have similar ε_{Nd} signatures (Weldeab et al., 2013).

4.1.4. The Mozambique Channel and the Agulhas Current

Surface waters near southwestern South Africa are marked by highly unradiogenic ε_{Nd} signatures as revealed by one sample yielding a value of -18.9 (S1, Fig. 1), which is consistent with the influence of the Agulhas Current receiving its unradiogenic signature from particle dissolution close to the eastern coast of southern Africa (Stichel et al., 2012a). A potential source region for these particles is the Mozambique Channel, which receives large amounts of sediments from the Zambezi and Limpopo Rivers. Surface waters of the Zambezi discharge area have ε_{Nd} signatures between -14.7 (GIK16157) and -15.5 (GIK16156) (Supplementary Table 2), whereas the river suspended load is less radiogenic and has a mean ε_{Nd} value of -16.7. Surface sediments directly at the river mouth have ε_{Nd} values as negative as -17.7 (van der Lubbe et al., 2016). The dissolved and particulate river loads are only slightly more radiogenic than the isotopic signatures of the near surface Cape Basin. It is, however, likely that the signatures released, for example from resuspension induced by deep reaching eddies, can also be less radiogenic depending on the exact origin of the sediment particles transported by the Agulhas Leakage into the Cape Basin either in particulate or dissolved form.

Surface waters at the Limpopo River mouth have ε_{Nd} signatures as negative as -22.4 (GIK16152, Supplementary Table 2). According to the T-S-relationships these mix with water of Station S1 (Stichel et al., 2012a) and SACW encountered at stations 36-40 of the Cape Basin (Fig. 2). We selected Station S1 as an endmember and calculated mixing relationships between S1 (Stichel et al., 2012a), SACW of the Cape Basin and TSW/STSW of the Angola Basin (Fig. 5). For the Agulhas Current endmember represented by station S1 (Stichel et al., 2012a) we chose an ε_{Nd} signature of -18.9 and a salinity of 35.47. For SACW, an ε_{Nd} value of -9.6 and a salinity of 34.79 was adopted (Jeandel, 1993). For TSW/STSW an ε_{Nd} of -12.8 and a salinity of 36.39 was used (Zieringer et al., 2019). As a result, we find that a mixture between SACW from the Cape Basin and waters from S1 can explain the unradiogenic surface water signatures, noting that waters from S1 are likely influenced by even less radiogenic waters from the Mozambique Channel (Fig. 5). Station 43 in the Cape Basin consists of almost pure SACW, whereas station 40 located only 164 nautical miles further offshore to the west is most influenced by waters of the Agulhas Current (Fig. 5). The Nd isotopic compositions of surface waters from the western Cape Basin and Angola Basin are less radiogenic than the mixing line between SACW and TSW/STSW (Fig. 5) and clearly indicate admixture of the unradiogenic Agulhas Current (Fig. 5). The compositions of stations 27 to 30 and fish 44 (Rickli et al., 2009) are close to the mixing line between TSW/STSW and S1, indicating that these stations are also strongly influenced by the Agulhas Current. Overall, our Nd data track the Agulhas Current entering the Cape Basin at station 40, flowing across the Angola Basin, passing the location of fish 44 (Rickli et al., 2010) and exiting the basin close to station 30 (Fig. 1).

4.2. Intermediate waters

Mixing relationships between the intermediate and deep water masses NADW, AAIW and UCDW were calculated in order to evaluate to what extent the Nd isotope and concentration data reflect conservative mixing in the Angola and Cape basins (Figs. 6a, b). We chose regional water mass endmembers prevailing at the northern and southern borders of the research area, which mix at the corresponding depths according to the TS-relationships (Fig.

2). The northernmost station of the Angola Basin (station 24, 2000 m, ε_{Nd} = -12.8, Nd = 17.5 pmol/kg, salinity = 34.95) was selected for the regional NADW endmember signature. The regional AAIW- and UCDW-endmember compositions (ε_{Nd} = -8.2, Nd = 10.9 pmol/kg, salinity = 34.28 and ε_{Nd} = -8.5, Nd = 11.8 pmol/kg, salinity = 34.37, respectively) were adopted from station 101 in the southern Cape Basin (Stichel et al. 2012b). In the Angola Basin the contribution of NADW decreases southward, reflecting gradual dilution with southern sourced AAIW and UCDW (Fig. 6a). However, all stations have a less radiogenic ε_{Nd} signature than expected from the mixing lines, except station 35 (1248 m) (Fig. 6a). To quantify the variability not related to conservative water mass mixing, we calculated the difference $\Delta\varepsilon_{Nd}$ between measured ε_{Nd} values and the corresponding ε_{Nd} values resulting from pure water mass mixing of the two previously defined regional endmembers. The same was done for Nd concentrations (Δ_{Nd}). We defined deviations from conservative mixing exceeding ±0.5 ε_{Nd} units and ±0.8 pmol/kg Nd as indicating non-conservative behavior. This is based on the max 2SD uncertainties of the Nd isotope measurements and on a 2 % uncertainty of the highest measured Nd concentration. The patterns of $\Delta\varepsilon_{Nd}$ and Δ_{Nd} for all samples below 500 m are shown in Fig. 7. Intermediate waters of the Angola Basin are 1 to 2.5 ε_{Nd} units less radiogenic and reach elevated Nd concentrations of up to 30 % (1 to 5 pmol/kg) (Fig. 7), compared with calculated values assuming conservative mixing. The highest $\Delta\varepsilon_{Nd}$ and Δ_{Nd} occur in the northern and central part of the basin above 3500 m water depth, where low oxygen conditions are likely responsible for the reduction of Fe-Mn oxyhydroxides near the coast and the associated release of unradiogenic Nd to intermediate waters (Rickli et al., 2010) (see 4.1.2).

The intermediate waters of the Cape Basin constitute a well constrained mixture of 90 % AAIW and 10 % NADW between 650 and 800 m water depth and a mixture of 60-80 % AAIW/UCDW and 40-20 % NADW between 1000 and 1200 m water depth (Fig. 6b).

However, water samples between 650 and 800 m do not exactly fall on the mixing line and are slightly less radiogenic (ΔεNd ~ -1), whereas Nd concentrations remain constant (Fig. 7). These intermediate waters likely acquired their unradiogenic Nd isotope signatures remotely via reversible scavenging (Siddall et al., 2008), partial dissolution of sinking particles or boundary exchange processes in the Mozambique Channel and subsequent advection of this unradiogenic Nd into the Cape Basin via the Agulhas Leakage.

4.3. Deep waters

Deep water mixing relationships were calculated between NADW and UCDW/LCDW in the Angola Basin, (Fig. 6c, d). For mixing calculations at 2000 m water depth, we again used the regional NADW endmember from the northernmost station of the Angola Basin (station 24, 2000 m depth) and the regional UCDW endmember from station 104 (1200 m, ε_{Nd} = -8.5, Nd = 13.82 pmol/kg, salinity = 34.63) (Stichel et al. 2012b). For mixing calculations at 3000 m water depth, NADW at station 24 at 3000 m water depth (ε_{Nd} = -12.8, Nd = 20.89 pmol/kg, salinity = 34.9) and LCDW at station 113 (2400 m depth) (ε_{Nd} = -8.5, Nd = 24.51 pmol/kg, salinity = 34.68) (Stichel et al. 2012b) were used as regional endmembers. While deep waters in the northern Angola Basin are composed of almost pure NADW, the southern Angola Basin waters represent a conservative mixture of 30-40 % UCDW and LCDW (Fig. 6c). Deep water masses of the central Angola Basin (station 28, 30) exhibit an unradiogenic Nd isotope excess ($\Delta\varepsilon_{Nd}$ ~ -1.5) (Fig. 7). At the same time Nd concentrations are slightly elevated by a maximum of 10 % (ΔNd ~ +1 pmol(kg) between 2000 and 3000 m water depth and are essentially in agreement with the water mass mixing relationship and similar to modeled ΔNd values from the Angola Basin at ~12 °S (cf. Zheng et al., 2016). This can be explained by partial dissolution of Fe-Mn oxides originating from surface waters (see 4.1.2). Lateral transport of dissolved Nd originating from sediments of the African shelf may also contribute to the observed patterns (Zheng et al., 2016). In contrast, deep waters of the northern and southern Angola Basin are not marked by significant $\Delta\varepsilon_{Nd}$ and ΔNd values and thus are dominantly controlled by preformed REE concentrations thus reflecting essentially conservative water mass mixing (Figs. 6c, 7).

Deep waters of the eastern Cape Basin contain a NADW fraction of up to 80-90 %, whereas NADW in deep waters of the western Cape Basin (station 36) is mixed with up to 40 % UCDW/LCDW (Fig. 6d). This is consistent with the notion that these deep waters from 3000 m depth in the Cape Basin are directly mixed with NADW (Rickli et al., 2009), which can be explained by exchange across gaps in the Walvis Ridge. Differences between measured and calculated Nd isotopic compositions and concentrations in deep waters of the Cape Basin are within the defined uncertainty range ($\Delta\varepsilon_{Nd}$ = ±0.5, ΔNd = ± 0.8), again suggesting essentially conservative water mass mixing (Fig. 7). However, at ~3000 m depth near the slope area, a significant Nd loss of up to 18 % (ΔNd ~ -4) is observed, which likely results from Nd scavenging by resuspended shelf sediments.

4.4. Bottom waters

For mixing relationships of the bottom waters, the same NADW regional endmember as for the deep waters was chosen and the LCDW regional endmember was adopted from station 104 (4440 m depth, ε_{Nd} = -8.7, Nd = 27,9 pmol/kg, salinity of 34.69 (Stichel et al. 2012b). Bottom waters of the Angola Basin are composed of almost pure NADW (Fig. 6e), but are slightly less radiogenic than expected from the conservative mixing relationships and exhibit excess Nd concentrations of up to 20 % (ΔNd = ~+5 pmol/kg) (Fig. 7). This excess can be explained by release of REEs from particles sinking from surface waters into the hydrographically isolated deep Angola Basin (Rickli et al., 2009, Zheng et al. 2016).

Cape Basin bottom waters are composed of pure LCDW, based on hydrographic parameters (Stichel et al., 2012b) and on Nd isotope compositions (Fig. 6h) but also show an excess Nd concentration (ΔNd = ~+6 pmol/kg) (Fig. 7), which is about 20 % higher than the calculated expected concentration for conservative LCDW (Fig. 6h). However, at the same time no significant change of the Nd isotope composition ($\Delta\varepsilon_{Nd}$) is observed (Fig. 7). The Nd excess likely originates from partial dissolution of resuspended sediments and/or dust particles from the Namib or Kalahari deserts, which have isotope compositions of -9.8 to -11.4 and -9.3 to -10.9 (Bayon et al., 2004), respectively, similar to those of Cape Basin bottom waters (-9.6 to -10.5) (Fig. 6). A simple mass balance calculation reveals that the dissolved sediment or dust would need to have an ε_{Nd} signature above -7.2 or below -12.7 to change the isotope composition of LCDW beyond ±0.5 ε_{Nd} units, assuming that particle dissolution results in similar excess Nd concentrations mixed with LCDW as today. This indicates that terrigenous inputs with extremely low or high Nd isotope compositions are required to significantly alter the Nd isotope compositions of the bottom waters in the study area in the modern regional deep water mass configuration. Available evidence from detrital sediment compositions in the Cape Basin (Dausmann et al., 2017) suggests that the ε_{Nd} range between -7.2 and -12.7 was not exceeded over the past 12 Myr. Thus, while local additions affect the Nd concentration in the deep Cape Basin, water mass mixing exerts the key control over the Cape Basin bottom water Nd isotope composition in the modern regional deep water mass configuration. However, the non-conservative Nd isotope effect may have been more pronounced under glacial boundary conditions such as the Last Glacial Maximum (LGM), when NADW contributions to Cape Basin deep water were significantly lower resulting in a more radiogenic signature of the bottom waters of ~-6 as extracted from sediments (Piotrowski et al., 2004). Without non-conservative Nd addition in the Cape Basin, the ambient glacial bottom water value may hence have been somewhat more radiogenic than -6.

4.5. Comparison of the eastern and western South Atlantic Basins

We compare our Nd isotope compositions from the Angola and Cape basins with Nd isotope measurements from full water profiles at 30°0’ S, 1°25’W (station 217, eastern South Atlantic) and at 33°15’S, 41°45’W (station 302, western South Atlantic) (Jeandel 1993). Although station 217 is located at a distance of only 80 nautical miles from our station 35, surface waters at station 217 are three ε-units more radiogenic than at station 35 but interestingly match the isotope compositions of surface waters in the eastern Cape Basin (Fig. 8). AAIW at station 217 (εNd -6) also shows a more radiogenic value than presented in Stichel at al. (2012b) and the values we detected for the Angola and Cape basins, but for NADW similar values at all locations are observed (Fig. 8). The large differences between the AAIW measurements may be due to the fact that Jeandel (1993) at the time used unfiltered seawater samples, so that release from more radiogenic particles during seawater acidification may have contributed to these more positive signatures. The western South Atlantic water masses, except NADW, are generally more radiogenic than the Angola and Cape basins in the east. This can be explained by the unrestricted admixture of more radiogenic Pacific waters advected through the Drake Passage (Jeandel, 1993, Rickli et al. 2009).

4.6. Nd isotope composition of seawater and sediments from the Walvis Ridge and Cape Basin

Comparison between the Nd isotope compositions of benthic foraminifera from core top sediments from the Cape Basin side of the Walvis Ridge (Klevenz et al., 2008) and seawater of our stations 36 and 37 in the Cape Basin from similar depths reveal a close correspondence (Fig. 9). This suggests that the authigenic fraction in these sediments faithfully reflects today’s Nd isotope distribution of the main water masses in the Cape Basin. Core top sediments from the southern Cape Basin record ε_{Nd} values of -9.9 in good agreement with extracted Holocene NADW signatures (Wei et al., 2016), which are only slightly more radiogenic than the modern signature of the core of NADW we find in the southern Angola

Basin and eastern Cape Basin (ε_{Nd} -11). Overall, the comparison of our water column Nd isotope data with the surface sediment signatures indicates that authigenic sedimentary Nd isotopes in the SE Atlantic reliably reflect paleo water mass compositions.

5. Conclusions

Dissolved Nd isotopic compositions and Nd concentrations were determined along a N-S and an E-W full water column section across the Angola Basin and the northernmost Cape Basin in the SE Atlantic, which were sampled during GEOTRACES cruise GA08.

We found ε_{Nd} signatures as unradiogenic as -17 in surface waters of the Angola Basin and -17.6 in surface waters of the Cape Basin, which must originate from local or regional terrestrial inputs. In the central Angola Basin sediment particles from the African shelf and slope are transferred into the oxygen minimum zone and release unradiogenic Nd by reductive dissolution of their Fe-Mn oxyhydroxide coatings resulting in altered surface and intermediate water Nd isotope signatures. In contrast, surface waters of the Cape Basin are dominated by unradiogenic Nd isotope signatures originating from dissolved and particulate inputs of large rivers in the Mozambique Channel, from where they are advected by the Agulhas Current.

Deep water Nd isotope compositions in the northern and southern Angola Basin and in the western Cape Basin between 2000 and 4000 m essentially suggest that compositions are controlled by conservative water mass mixing, whereas the Nd isotopic composition of the deep central Angola Basin is overprinted by unradiogenic Nd likely released by dissolution of Fe-Mn oxides on sinking particles. At the deep eastern margin of the Cape Basin Nd is scavenged and removed from ambient seawater without significantly altering the Nd isotope compositions, most likely a consequence of resuspension of shelf and slope sediments.

Due to the hydrographic isolation of bottom waters in the Angola Basin, unradiogenic Nd released from sinking particles accumulates below 4000 m water depth. Bottom waters of the well ventilated Cape Basin hint at excess Nd concentrations below 4000 m water depth, originating from resuspended bottom sediments and/or dissolution of particles, increasing the Nd concentrations of the bottom waters. However, this addition does not significantly affect the seawater Nd isotope compositions expected from conservative mixing, due to similar ε_{Nd} values of the sediments and bottom waters, which today range from -9.6 to -10.5.

We conclude that Nd isotopes are a reliable quasi-conservative tracer of present and past deep water mass mixing in the southern and northern Angola Basin and in the Cape Basin, whereas the Nd isotope compositions of surface and bottom waters of the Angola Basin, as well as the entire water column of the central Angola Basin are affected by non-conservative addition of Nd from terrestrial inputs. The non-conservative additions to the Cape Basin water column are too small to significantly modify its Nd isotope compositions today, but may have been more significant in the past such as during glacial maxima.

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