

## Evidence of ventilation changes in the Arabian Sea during the late Quaternary: Implication for denitrification and nitrous oxide emission

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[1] Modern seawater profiles of oxygen, nitrate deficit, and nitrogen isotopes reveal the spatial decoupling of summer monsoon-related productivity and denitrification maxima in the Arabian Sea (AS) and raise the possibility that winter monsoon and/or ventilation play a crucial role in modulating denitrification in the northeastern AS, both today and through the past. A new high-resolution 50-ka record of  $\delta^{15}\text{N}$  from the Pakistan margin is compared to five other denitrification records distributed across the AS. This regional comparison unveils the persistence of east-west heterogeneities in denitrification intensity across millennial-scale climate shifts and throughout the Holocene. The oxygen minimum zone (OMZ) experienced east-west swings across Termination I and throughout the Holocene. Probable causes are (1) changes in ventilation due to millennial-scale variations in Antarctic Intermediate Water formation and (2) postglacial reorganization of intermediate circulation in the northeastern AS following sea level rise. Whereas denitrification in the world's OMZs, including the western AS, gradually declined following the deglacial maximum (10–9 ka BP), the northeastern AS record clearly witnesses increasing denitrification from about 8 ka BP. This would have impacted the global Holocene climate through sustained  $\text{N}_2\text{O}$  production and marine nitrogen loss.

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### 1. Introduction

[2] The Arabian Sea (AS) hosts one of the largest pools of denitrifying water in the ocean and accounts for at least one third of the loss of marine fixed N [Codispoti and Christensen, 1985]. In the near absence of  $\text{O}_2$ , denitrification converts combined, bioavailable nitrogen to gaseous  $\text{N}_2$ , strongly discriminating against the heavier isotope ( $^{15}\text{N}$ ). Monitoring denitrification intensity in this region is essential given its potential to alter surface fertility and in turn the marine carbon cycle [McElroy, 1983; Ganeshram et al., 1995; Altabet et al., 1995]. In addition, Nitrous oxide ( $\text{N}_2\text{O}$ ) is a by-product of denitrification and, once released to the atmosphere, contributes to the radiative forcing [Codispoti and Christensen, 1985].

[3] A growing body of evidence points to strong coupling between denitrification strength in the AS and climate changes at both orbital and millennial timescales [e.g., Altabet et al., 1995, 1999a, 2002; Ganeshram et al.,

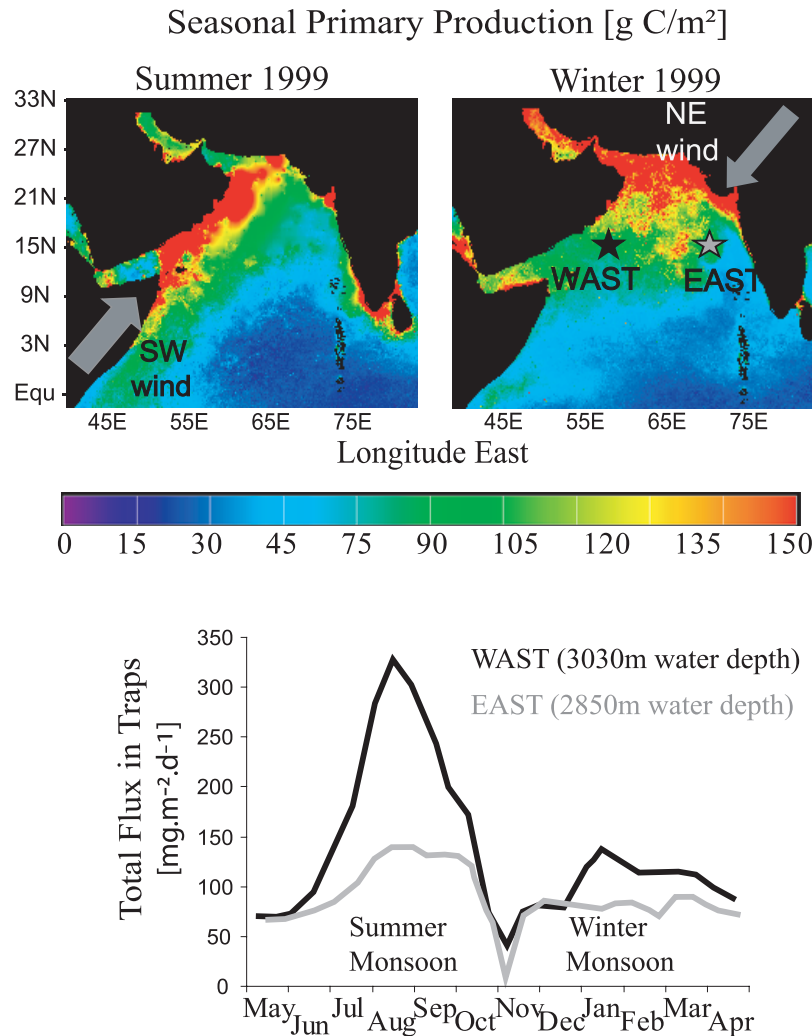
2000; Suthhof et al., 2001; Ivanochko et al., 2005]. Using sedimentary  $\delta^{15}\text{N}$  measurements, these studies show that denitrification increases during warm periods concurrent with the summer monsoon and productivity intensification [Reichart et al., 1998; Schulz et al., 1998]. Surface productivity and associated oxidant demand are therefore regarded as the main control on water column denitrification in the past [Ganeshram et al., 2000; Ivanochko et al., 2005]. Intriguingly, modern data reveal that marine productivity is maximal in the western AS while the most severe oxygen deficiency develops in the relatively oligotrophic central and northeastern AS [Olson et al., 1993; Naqvi, 1994]. Such a paradox points to  $\text{O}_2$  supply through oceanic ventilation as an additional control on denitrification. Model simulations indeed show that past changes in oxygen supply at intermediate depth by the Antarctic Intermediate Waters (AAIW) can modulate denitrification in the Oxygen Minimum Zones (OMZ) such as the AS, and hence impact the global marine N budget [Schulte et al., 1999; Galbraith et al., 2004; Meissner et al., 2005; Schmittner et al., 2007].

[4] Given the regional decoupling between denitrification and productivity in the basin today, we propose to critically explore the impact of ventilation on the intensity and distribution of the denitrifying zone through millennial-scale climate shifts and across the Holocene. One crucial limitation in assigning clear causes to denitrification changes in the OMZs lies in the lack of unequivocal proxies for either oxidant demand or  $\text{O}_2$  supply. In an attempt to

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**Figure 1.** (top) Primary productivity reconstruction (<http://marine.rutgers.edu/>) and (bottom) total fluxes in sediment traps WAST and EAST per month over 1986–1989 [Rixen *et al.*, 1996].

compensate for this limitation, we propose to compare the  $\delta^{15}\text{N}$  signals of six sediment cores distributed from the southwestern to the northeastern AS with recently documented changes in intermediate/thermocline circulation [Kuhnt *et al.*, 2004; Pahnke and Zahn, 2005].

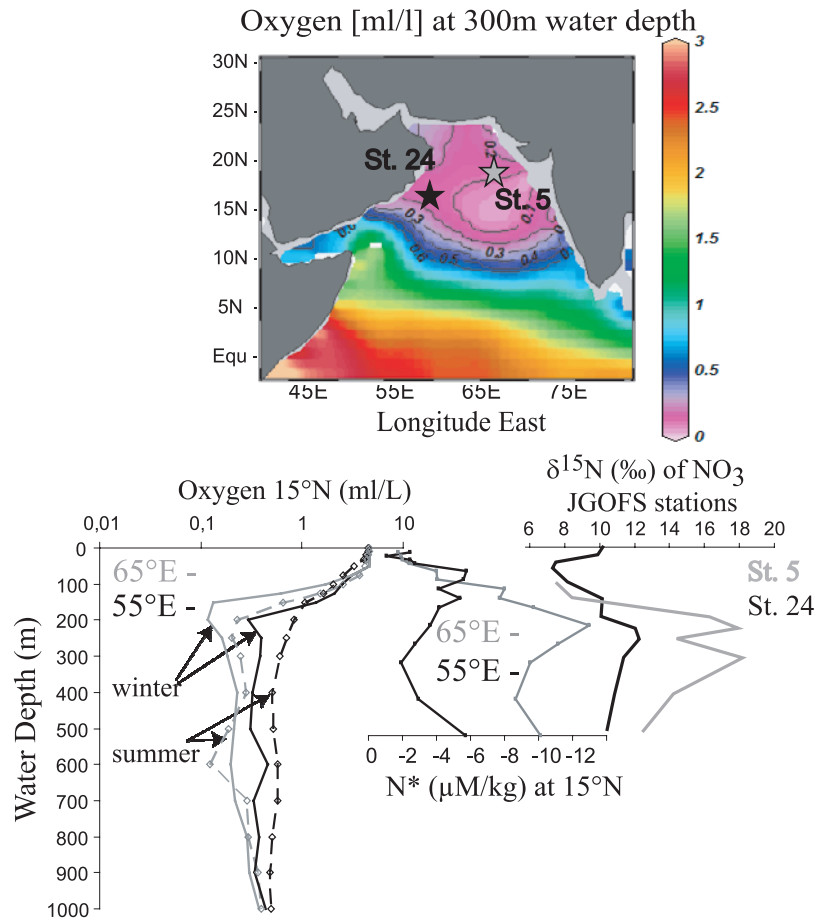
## 2. Nutrient and Oxygen Inventories in the Modern AS

### 2.1. Primary Productivity and Denitrification

[5] Latitudinal shifts in the position of the Inter-Tropical Convergence Zone over the Indian Ocean region results in drastic seasonal variation in atmospheric circulation and hence in upwelling intensity [Wyrki, 1973]. Figure 1 shows the contrast in primary productivity between summer and winter in the surface AS. Primary productivity peaks off Somali and Oman during summer monsoon and in the northern AS during winter. Intermonsoon periods exhibit low productivity conditions. Strong seasonality in particle fluxes and export production result from these alternating atmospheric and surface ocean conditions [Nair *et al.*, 1989;

Haake *et al.*, 1993; Ramaswamy and Nair, 1994]. Sediments traps record a bimodal distribution in biogenic particle fluxes with very high fluxes in summer and moderately high fluxes during winter time [Rixen *et al.*, 1996; Honjo *et al.*, 1999] (Figure 1). From 58 to 78% of the particle flux to the seafloor occur during the summer monsoon with the highest productivity observed in the southwest part of the basin [Haake *et al.*, 1993].

[6] Figure 2 shows the oxygen distribution in the AS. This figure highlights the clear west-east decrease in oxygen concentration, opposite to what might be expected given the distribution of primary productivity. Figure 2 also reveals that  $\text{O}_2$  deficiency at 100–200 m builds up during winter rather than during the highly productive summer monsoon, in agreement with de Sousa *et al.* [1996]. The OMZ is permanent in spite of drastic seasonal changes in productivity [Sarma, 2002]. This stability has been explained by invoking a balance between the biological pump (oxidant demand) and the physical pump (mixing and ventilation) of oxygen around the year [Olson *et al.*, 1993; Sarma, 2002].



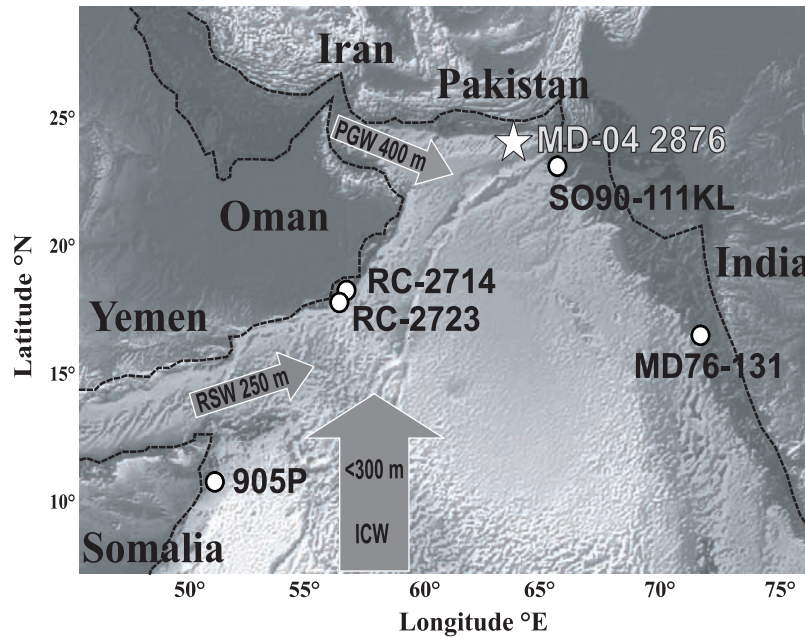
**Figure 2.** (top) oxygen concentration at 300 m water depth (World Ocean Atlas 05, ODV) and (bottom) profiles of oxygen concentrations and Nitrate deficit ( $N^* = ([NO_3] - 16[PO_4] + 2.9) \times 0.87$ ) compared to the  $\delta^{15}N$  signal of  $NO_3$  at JGOFS Station 5 and 24 [Altabet *et al.*, 1999a].

[7] Water column denitrification occurs when  $O_2$  concentrations fall below 0.15–0.2 mL/L [Naqvi and Jayakumar, 2000], a requirement which is not met within the entire OMZ but in areas restricted to the northeastern AS, mostly around 250 m water depth (Figure 2). Using hydrostation data from the World Ocean Atlas database (<http://iridl.ldeo.columbia.edu/>), we calculated nitrate deficit ( $N^*$ ) in the water column according to the formula published by Gruber and Sarmiento [1997] where  $N^*$  represents the nitrate lost through denitrification. Between 100 and 500 m depth  $N^*$  is strongly negative in the northeastern AS, and roughly coincides with  $[O_2]$  minima (Figure 2). However, oxygen content at intermediate depth in the western AS is not low enough to cause year-round denitrification at any depth.  $N^*$  is less negative here than in the eastern AS and minimum values do not correspond to maxima in  $\delta^{15}N-NO_3$  (Figure 2). This could indicate that heavy nitrate found in the western AS is not fully produced in situ but partly brought from the northeastern AS in winter, when the thermocline circulation reverses [You, 1998]. Water column denitrification causes the  $\delta^{15}N$  of nitrate to be significantly higher than the mean ocean nitrate value of 4.5–6‰ [Sigman *et al.*, 1997]. As a consequence  $\delta^{15}N-NO_3$  at 300 m depth reaches 18‰ in the

northeastern AS and 12‰ in the western AS [Brandes *et al.*, 1998; Naqvi *et al.*, 1998; Altabet *et al.*, 1999a], further highlighting the clear regional heterogeneity.

## 2.2. Circulation and Oxygen Supply

[8] The observed spatial segregation between productivity and denitrification maxima is caused by the circulation of thermocline and intermediate waters in the AS and the initially very low oxygen content of water entering the basin [Olson *et al.*, 1993]. The main water sources at 200–1000 m originate both from the northwest (the highly saline Red Seawater (RSW) and the Persian Gulf Water (PGW)) and from the south (the Indian Central Water (ICW)) [Wyrki, 1973; Swallow, 1984; You, 1998] (Figure 3). The latter is a mixture of aged Antarctic Intermediate Water (AAIW) and Banda Seawater (BSW) coming from the Indonesian throughflow [Sharma, 1972; Sharma *et al.*, 1978; You, 1998], and enters the AS across the southwestern boundary during the summer monsoon with the Somali current [You, 1998]. Oxygen concentrations in the northern and southern water masses are equally low but the volume transported across the equator with the southern waters is 1 order of magnitude larger than that contained in the northern source



**Figure 3.** Map of the Arabian Sea with intermediate/thermocline water masses in the AS with approximate core depth. Sediment core studied: Core MD76 131 [Ganeshram *et al.*, 2000], core SO90-111KL [Suthhof *et al.*, 2001], core MD04-2876 (this study), cores RC27-14 and RC27-23 [Altabet *et al.*, 2002] and core 905P [Ivanochko *et al.*, 2005].

inflow [Swallow, 1984]. Oxygen replenishment occurs mostly along the southern and western boundaries of the AS and hence the northeastern part of the basin is bathed by waters severely impoverished in  $O_2$ .

### 3. Material and Method

[9] Core MD-04 2876 ( $24^{\circ}50'57N$ ;  $064^{\circ}00'49E$ ; 33 m length) was retrieved at 828 m water depth on the Pakistan Margin within the present-day OMZ. The core shows alternating sequences of varved/laminated and bioturbated/massive sediments.

[10] Nitrogen content and isotopic composition in MD-04 2876 were analyzed with a Micromass Mass Spectrometer at the University of Bordeaux on bulk sediments. To ensure comparability with other AS records measured in the School of Geosciences, The University of Edinburgh [Ivanochko *et al.*, 2005], we run a set of 10 sediment samples from MD-04 2876 as well as an internal (sediment) standard on both Mass Spectrometers. Intercomparability was found to be within  $\pm 0.3\%$ . In total, 10 samples were additionally analyzed for  $\delta^{13}C_{org}$  using the same Mass Spectrometer, in order to investigate marine versus terrestrial origin of the organic matter.

[11] The calendar chronology for core MD04-2876 is based on 15 radiocarbon ages measured by accelerator mass spectrometry on samples composed of hand-picked shells of the planktonic foraminifera *Globigerinoides ruber*. Further details on the samples in relation to foraminiferal abundance variations, on the correction for reservoir age and on the conversion in terms of calendar ages will be presented

elsewhere (E. Bard *et al.*, manuscript in preparation, 2007). The sedimentation rates calculated between each dated interval are  $53 \pm 11$  cm/ka on average.

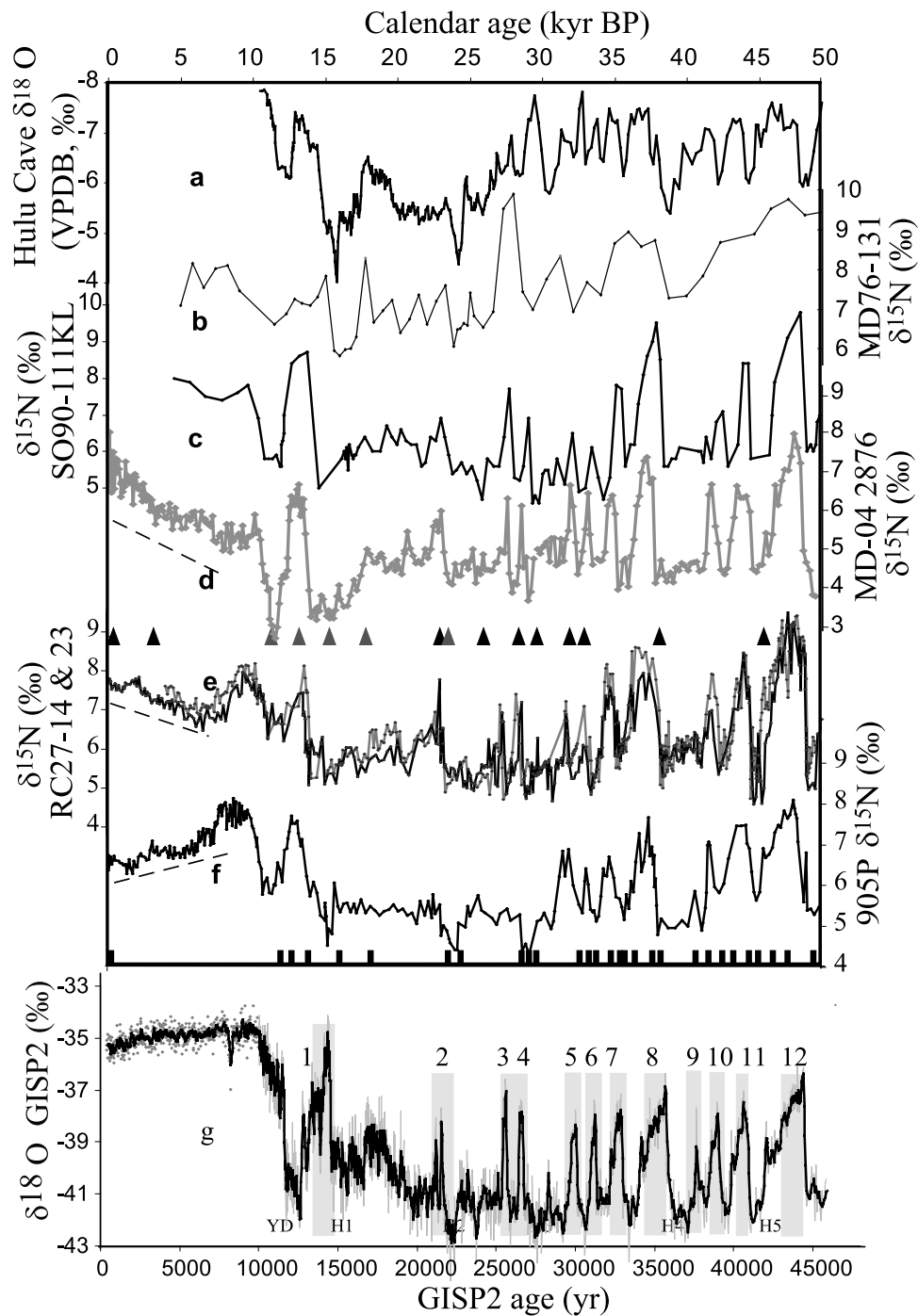
[12] The new  $\delta^{15}N$  record was compared to 5 existing records from the AS (Figure 3): MD76 131 [Ganeshram *et al.*, 2000], SO90-111KL [Suthhof *et al.*, 2001], RC27-14 and RC27-23 [Altabet *et al.*, 2002] (Figure 4e), and 905P from [Ivanochko *et al.*, 2005]. Detailed comparison between the high-resolution records RC-27 14, RC-27 23, 905P and MD-04 2876 was achieved by tuning the existing records to the well-dated MD-04 2876 record. To do so, we used 28 tie points between the MD-04 2876 record and each of the published records. Linear interpolation of the  $\delta^{15}N$  record was undertaken at 100-year intervals between measured values. The  $\Delta\delta^{15}N$  curves, defined as the difference between our new  $\delta^{15}N$  records and each of the three existing records from western AS, were obtained by subtracting these interpolated records.

## 4. Millennial-Scale Changes in Denitrification

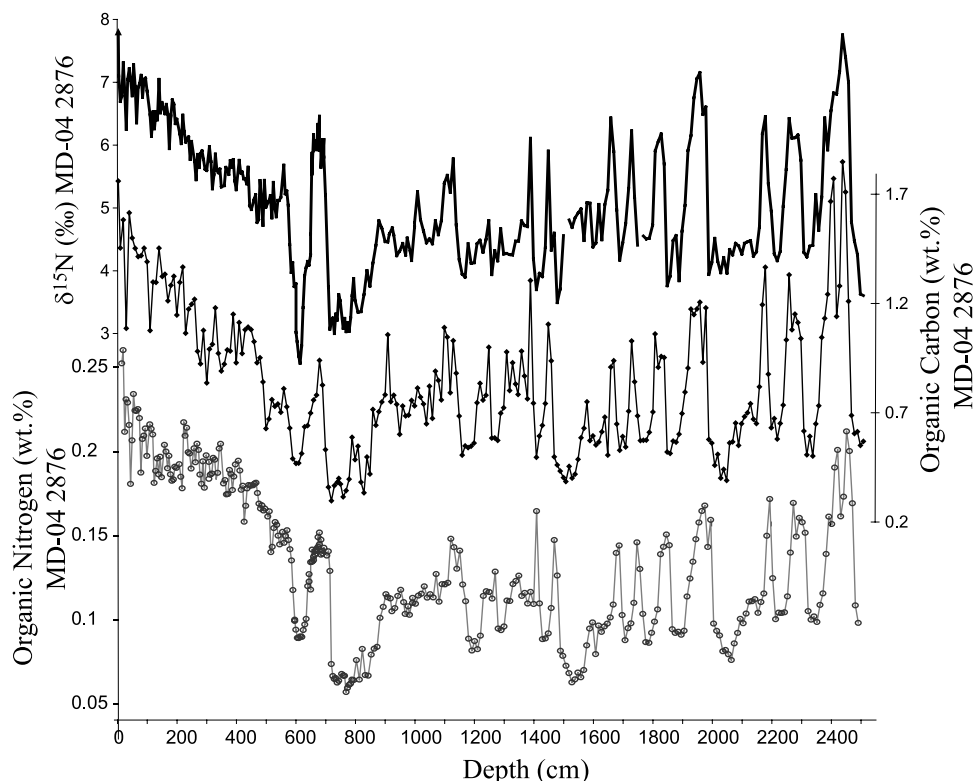
### 4.1. A New 50-ka Record of Denitrification Changes in the Northeastern AS

[13] High-resolution  $\delta^{15}N$  measurements from Core MD-04 2876 display large-amplitude, abrupt changes that faithfully mirror millennial-scale northern high-latitude climatic changes recorded in Greenland ice core  $\delta^{18}O$  and Monsoon fluctuations recorded in Hulu cave  $\delta^{18}O$  (Figure 4). The  $\delta^{15}N$  values range from around 3 to 8‰ ( $\pm 0.3\%$ ). The arithmetic mean  $\delta^{15}N$  value is 5.35‰. The  $\delta^{13}C$  of organic carbon is consistently around  $-19\%$  which confirms that





**Figure 4.** Correlation between ice core and speleothem  $\delta^{18}\text{O}$  records and sedimentary  $\delta^{15}\text{N}$  records from the Arabian Sea. (a) The  $\delta^{18}\text{O}$  of Hulu Cave stalagmite (composite record [Wang *et al.*, 2001]), (b) the  $\delta^{15}\text{N}$  records of Core MD76 131 from the India Margin [Ganeshram *et al.*, 2000], (c) core SO90-111KL from the northeastern AS [Suthhof *et al.*, 2001], (d) core MD04-2876 (grey), (e) cores RC27-14 and RC27-23 from the Oman Margin [Altabet *et al.*, 2002], (f) core 905P from the Somalia Margin [Ivanochko *et al.*, 2005], and (g) the  $\delta^{18}\text{O}$  of GISP2 ice core [Grootes and Stuiver, 1997]. Numbers from 1 to 12 designate Dansgaard-Oeschger events, labels H1 to H5 are Heinrich Events 1 to 5, and LGM means Last Glacial Maximum. Black triangles show the  $^{14}\text{C}$  dates obtained for core MD-04 2876, and black rectangles are the tie points used to tune the  $\delta^{15}\text{N}$  records to core MD04-2876.



**Figure 5.** (bottom to top) Organic carbon, organic nitrogen, and  $\delta^{15}\text{N}$  records from core MD-04 2876 versus depth.

the organic matter is pristine planktonic material irrespective of the climatic period (Table 1). Sedimentary nitrogen is enriched in  $^{15}\text{N}$  during warm periods compared to today's mean ocean nitrate, testifying to the occurrence of intense denitrification. During Stadials and Heinrich events however, the signal systematically drops down to about 3–5‰, evidencing low surface/thermocline  $\delta^{15}\text{N}\text{-NO}_3$ . These values are lower than the modern average isotopic signal of nitrate and reveal the occurrence of either incomplete N relative utilization or N fixation. Incomplete relative utilization of surface nitrate has been documented to have a very limited imprint on the  $\delta^{15}\text{N}$  signal in the AS [e.g., Schäfer and Ittekkot, 1993]. Furthermore, given that the  $\delta^{15}\text{N}$  signal is overall positively correlated to organic carbon fluxes at core site over the period studied (Figure 5) we assume that the  $\delta^{15}\text{N}$  record does not reflect changes in nitrate relative utilization off Pakistan.

[14] Biological N fixation develops either in stratified, oligotrophic surface waters or in denitrifying zones owing to relative P enrichment in thermocline waters that favors N fixing biota over other producers [Karl *et al.*, 2002]. Today, fixed N may account for a significant part of surface nitrate in the AS where denitrification is exceptionally intense [Brandes *et al.*, 1998; Deutsch *et al.*, 2007], hence partially cancelling out the isotopic enrichment caused by denitrification. It is, however, questionable whether N fixation was more active in the northeastern AS during Stadials and Heinrich events because (1) denitrification was reduced if

not shut down [Reichert *et al.*, 2004] and (2) surface waters were neither oligotrophic nor stratified during cold periods [Reichert *et al.*, 2004]. In addition, Schenau *et al.* [2005] documented reduced phosphorus regeneration in the northeastern AS during Heinrich 1 and the Younger Dryas, conditions that would hinder N fixation. Deutsch *et al.* [2007] suggest that N fixation rates are bound to increase with enhanced denitrification. Simple mass balance calculation shows that today, fixed nitrate accounts for 30% of the total N exported to the seafloor in the AS [Brandes *et al.*, 1998]. The same calculation also predicts that the  $\delta^{15}\text{N}$  signal of surface nitrate (to be used by the phytoplankton and exported to the sediment) always increases with the intensity of water column denitrification, even for low denitrification rates such as expected during cold periods. The only way to avoid this conclusion involves applying unreasonably high contributions of fixed N (>50%), i.e., increasing N fixation with decreasing denitrification rate. Hence we propose that although N fixation occurs in the AS and explains the occurrence of N isotopic values <4.5‰ during cold phases, the general relationship remains that the  $\delta^{15}\text{N}$  ratio of surface  $\text{NO}_3$  decreases with decreasing water column denitrification.

#### 4.2. Regional Comparison of $\delta^{15}\text{N}$ Records

[15] While the regional biogeochemical cycles are essentially controlled by the monsoon in the western AS, nitrogen and oxygen budgets in the northeastern AS are also prone to respond to slight ventilation changes at intermediate-

**Table 1.** Organic Carbon Isotopic Signal in Core MD-04 2876

Depth in Core, cm	Calendar Age, Years BP	$\delta^{13}\text{C}$ of Organic Matter, ‰
315	4300	-20.19
605	11,400	-20.30
615	11,600	-20.13
680	13,300	-17.81
1020	21,100	-18.39
1170	24,000	-19.76
1390	27,900	-18.24
1520	30,000	-19.55
1651	32,200	-19.85
1830	35,300	-19.71

thermocline depth, owing to the area being located at the end of the oxygenated water route. To document potential regional heterogeneity in the denitrification response within the AS through the past, we compare the  $\delta^{15}\text{N}$  signals of 6 cores (Figure 3) distributed across the AS.

[16] Although the  $\delta^{15}\text{N}$  records are comparable in terms of overall structure over the period studied (Figure 4), significant differences in arithmetic mean  $\delta^{15}\text{N}$  values are observed between core sites, with a maximal difference of 2.3‰ between MD76 131 (Indian Margin [Ganeshram *et al.*, 2000]) and MD-04 2876 (Pakistan Margin). *Gaye-Haake et al.* [2005] demonstrated that the diagenetic decay of organic compounds at the seafloor causes the  $\delta^{15}\text{N}$  of surface sediments to be 2–3‰ higher than the overlying deep trap values in the AS, with the exception of the northeastern AS (where Core MD 02-2876 has been retrieved) which shows no significant enrichment in  $^{15}\text{N}$ . This study further confirms the impact of export and burial rates on diagenetic N isotope fractionation [Lehmann *et al.*, 2002; Brummer *et al.*, 2002] and implicates that for a given initial  $\delta^{15}\text{N}$  signal carried to the seafloor, the recorded isotopic signal will be substantially higher (by 2–3‰) in areas characterized by low sedimentation rates than for those with rapidly deposited sediments. Accordingly, negligible isotopic effect during settling in the water column has been reported in high export fluxes, low oxygen environments [Altabet *et al.*, 1999b; Thunell *et al.*, 2004].

[17] Figure 6 illustrates the negative correlation between arithmetic mean  $\delta^{15}\text{N}$  values and mean accumulation rates calculated for the six cores studied. The difference in the average  $\delta^{15}\text{N}$  values between cores depends on the rate with which sediments are deposited and buried at each specific site, and is independent of local denitrification intensity. Moreover, radiocarbon dating on cores MD-04 2876 (Figure 4), 905 [Ivanochko *et al.*, 2005] and MD76 131 (T. Ivanochko, personal communication, 2006) reveal nearly constant sedimentation rates throughout these cores, including across Stadial/Interstadial transitions. We are therefore confident that millennial-scale  $\delta^{15}\text{N}$  variations from a given core are not caused by diagenetic effects attributable to temporal changes in sedimentation rate.

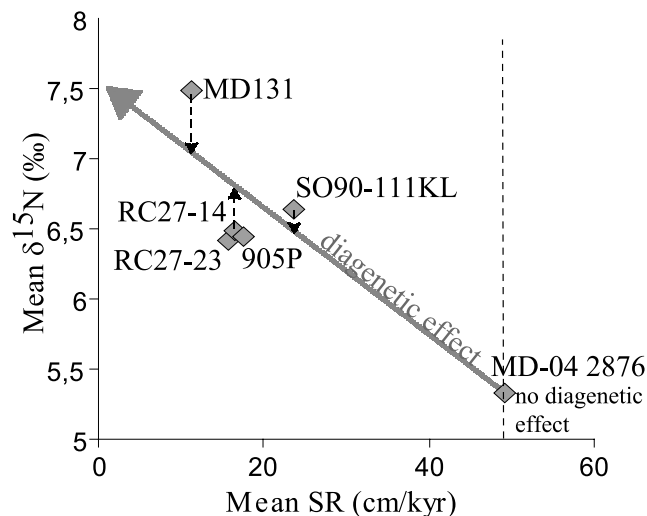
#### 4.3. Spatial and Temporal Heterogeneity in Denitrification

[18] Superimposed on the clear pattern of basin-wide changes in denitrification rate over millennial-scale in the

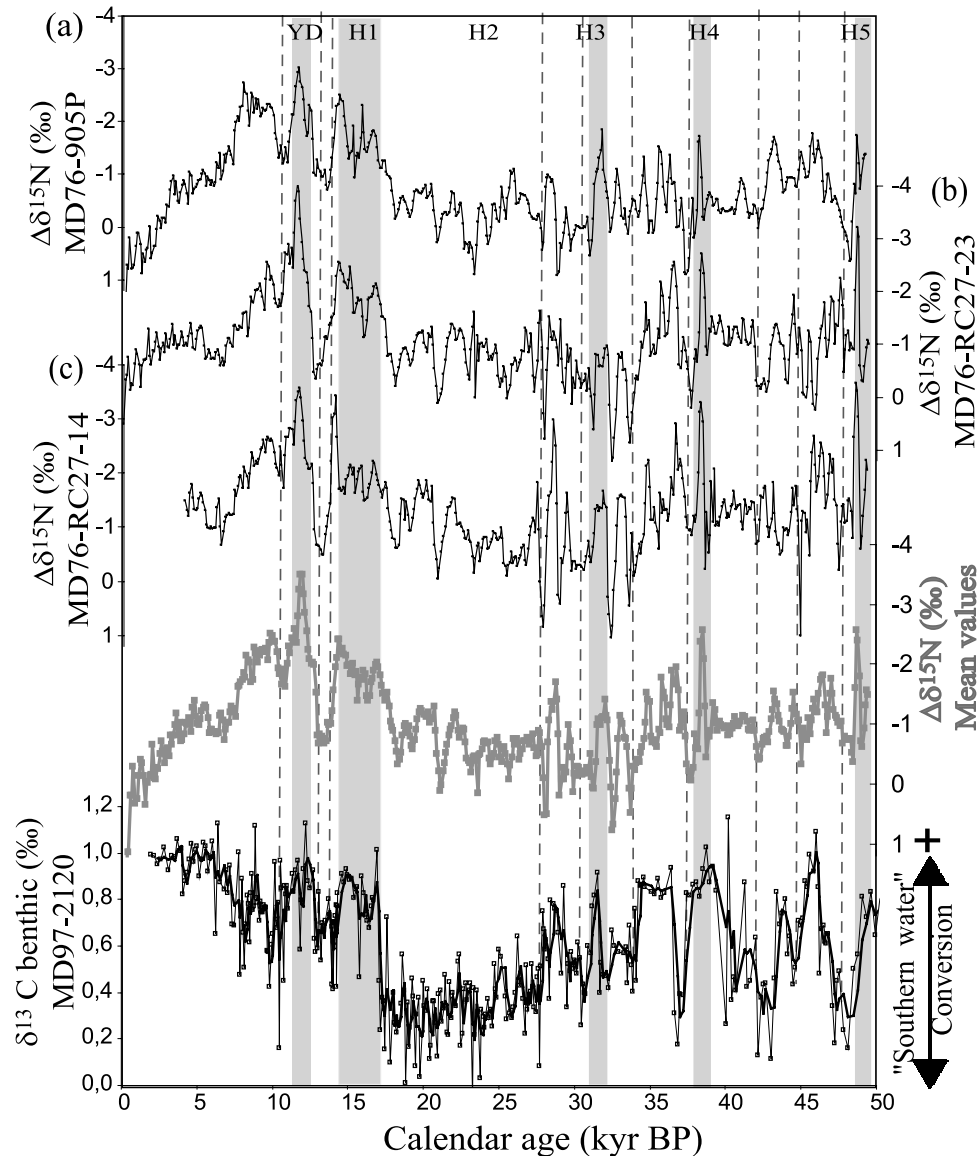
AS, we identify several differing trends of interest between the six records (Figure 4). The most conspicuous discrepancies between the records appear during Heinrich events 1, the Younger Dryas and the Holocene. The Holocene trend clearly decreases toward present in the Somalia Margin record, while it constantly increases in Core MD-04 2876. The Oman Margin cores exhibit an intermediate pattern. Further comparison is prevented by the absence of the late Holocene in cores MD76 131 and SO90-111KL. Sedimentary  $\delta^{15}\text{N}$  has also been found to increase over the Holocene until 2 ka BP in other records from the Indian margin [Banakar *et al.*, 2005; R. S. Ganeshram, unpublished data, 2004], corroborating the observed intensification of denitrification in the north and eastern AS since the early Holocene.

[19] In order to better quantify the differences between western AS  $\delta^{15}\text{N}$  records (i.e., Somalia, Oman margin sites) and the northeastern AS record, we calculate the  $\Delta\delta^{15}\text{N}$  by subtracting the 905P, RC17-23, and RC17-14 records from the MD-04 2876 record (Figure 7). The three calculated curves show strongly consistent variations, both in timing and amplitude of changes. The overall amplitude of the  $\Delta\delta^{15}\text{N}$  variations is  $>4.5\text{‰}$  in each case, comparable to the variation within the  $\delta^{15}\text{N}$  records themselves and much greater than both the long-term trends discussed above and the uncertainties associated with  $\delta^{15}\text{N}$  measurements. Over the past 50 ka, these changes occur at millennial timescales (Figure 7).

[20] Negative  $\Delta\delta^{15}\text{N}$  values indicate a less positive  $\delta^{15}\text{N}$  signal in the northeastern AS core relative to the western sites. More positive anomalies are found during Interstadials and the Late Holocene and more negative anomalies during Stadials and Heinrich events (and particularly the cold spells surrounding Termination 1). We interpret these variations as arising owing to east to west shifts of the OMZ from warm to cold periods, i.e., a collapse of the OMZ in the northeastern AS and the existence of small and probably seasonal denitrification areas confined to the western AS during



**Figure 6.** Arithmetic mean  $\delta^{15}\text{N}$  versus mean sedimentation rates (SR) for the six cores studied.



**Figure 7.**  $\Delta\delta^{15}\text{N}$  curves representing the difference in denitrification response between the northeastern site (MD-04 2876) and western sites (a) 905P, (b) RC27-23, and (c) RC27-14. In grey is shown the  $\Delta\delta^{15}\text{N}$  stack constructed by averaging the three aforementioned curves. (bottom) The  $\delta^{13}\text{C}$  of benthic foraminifera from core MD-97 2120 (45°S–32.06°S, 174°E–55.85°E) evidences millennial-scale changes in AAIW formation rate [Pahnke and Zahn, 2005].

Stadials (Figure 8). We propose that these swings were governed principally by ventilation changes in the AS.

## 5. Evidence for Circulation and Ventilation Changes

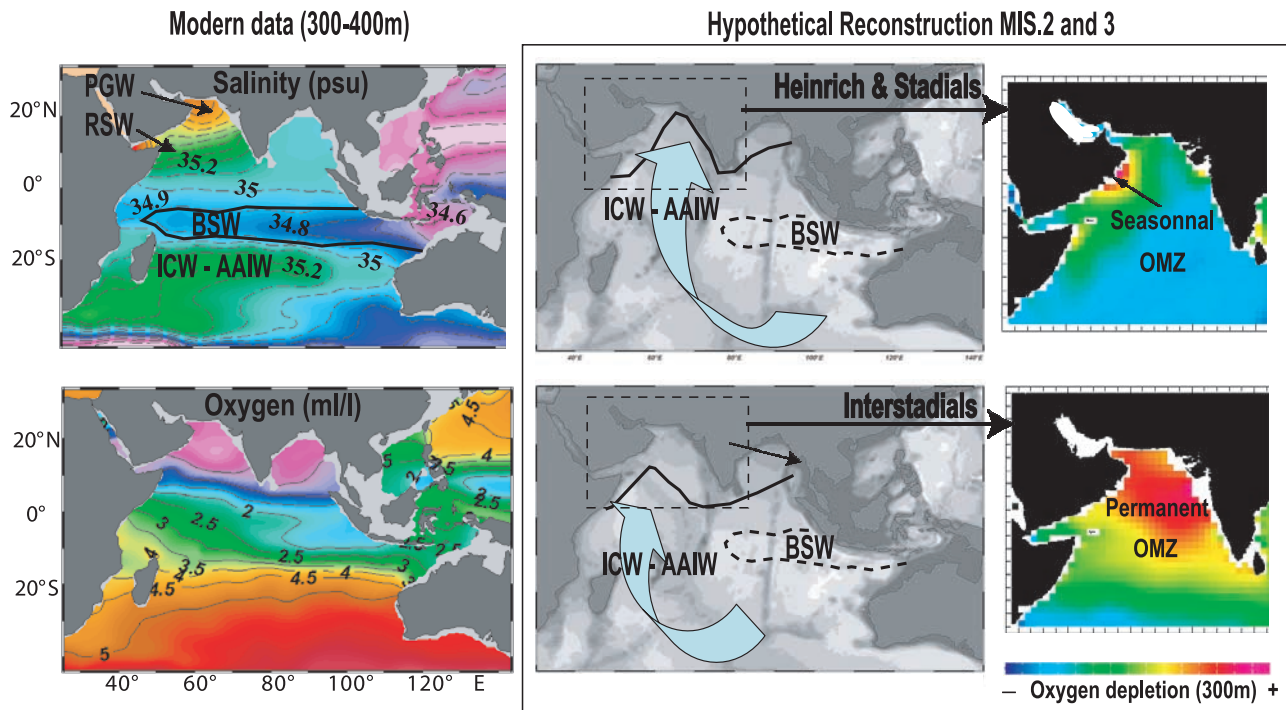
### 5.1. Millennial-Scale Changes in Circulation

[21] Recent studies document invigorated winter monsoon conditions during cold spells in the eastern AS [e.g., Reichert *et al.*, 2004; Higginson *et al.*, 2004]. Greater oxidant demand due to higher productivity related to the winter monsoon in the northeastern AS would result in shifting the denitrification maximum to the northeast in

these periods. However, the  $\Delta\delta^{15}\text{N}$  index shows the opposite trend, pointing to another scenario.

[22] Pahnke and Zahn [2005] demonstrate that the ocean circulation was arguably as reactive as the atmospheric circulation to abrupt climatic changes and adjusted rapidly to perturbations in North Atlantic Deep Water (NADW) formation. According to the authors, the conversion of AAIW and Subantarctic Mode Water in the Southern Ocean increased substantially during Heinrich events (and perhaps more generally during Stadials) and in turn increased the  $\text{O}_2$  inventory in the ventilated thermocline and intermediate depth ocean. Such a northward expansion of the AAIW





**Figure 8.** Hypothetical reconstruction of ventilation patterns at around 300 m depth in the Indian Ocean. (left) Modern annual mean salinity and Oxygen (mL/L) at depth range 300–400 m (WOA05), (middle) reconstruction of BSW and southern waters extension according to the extreme Interstadial and Stadial/Heinrich event scenarios, and (right) qualitative reconstruction of abrupt swings of the OMZ.

following perturbation of the Thermohaline Circulation (typical of Heinrich events) has been previously modeled and found to influence  $O_2$  inventory at low latitudes [Schmittner *et al.*, 2007] and particularly in the AS [Schulte *et al.*, 1999]. Likewise, the influence of the AAIW on the  $O_2$  budget in the modern AS is well known [e.g., Sharma, 1972; Olson *et al.*, 1993].

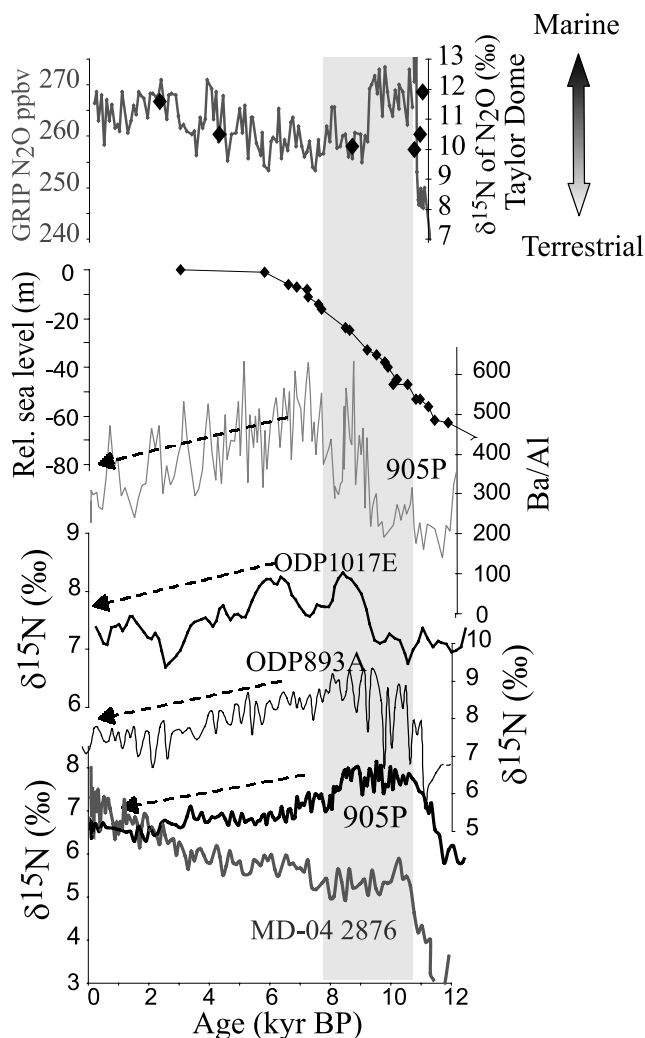
[23] Comparison between the  $\Delta\delta^{15}N$  curves and the  $\delta^{13}C$  signal of benthic foraminifera from a Southern Ocean core [Pahnke and Zahn, 2005] reveals that northeast to southwest swings of the OMZ (negative  $\Delta\delta^{15}N$  anomalies) are concurrent with episodes of increased AAIW formation (Figure 7). Today, the AAIW spreads northward in the Indian Ocean up to  $5^\circ N$  where it feeds the ICW [You, 1998]. This meridional progression of oxygen-rich, southern water masses to the AS is hampered owing to (1) the low-salinity front created along the equator by the zonal BSW flow (<500 m water depth) and (2) the presence of very saline waters from the marginal seas north of the equator (Figure 8). During MIS 2 and probably MIS 3, however, both the inflows from the marginal seas and the BSW were strongly reduced owing to sea level change [Kuhnt *et al.*, 2004], favoring northward circulation of southern waters in the AS and hence a greater contribution of the AAIW to the AS  $O_2$  inventory. The influence of changing AAIW flux on the AS is indeed clearly seen in the pre-Holocene  $\Delta\delta^{15}N$  curves whereas the observed correlation weakens after Termination I (Figure 7).

## 5.2. Holocene Trend

[24] The most prominent regional heterogeneity revealed by this multisite comparison occurs during the Holocene (Figures 4 and 9): Denitrification seems to intensify in the northeastern AS while it declines in the western AS. The continuous rise observed in our new  $\delta^{15}N$  record from around 8 ka BP contradicts evidence reported elsewhere [Deutsch *et al.*, 2004] that globally, the world's OMZ gradually retreated during the Holocene following the last deglacial apex, around 10 ka ago [Deutsch *et al.*, 2004, and references therein]. Such a retreat is attributed to an overall decline in productivity driven by the reduction of the marine nitrate inventory at the end of Termination I [Deutsch *et al.*, 2004]. Our results reveal that N cycling in the AS did not uniformly respond to the denitrification feedback and that the influence of local/regional productivity and circulation changes has to be considered.

[25] Reconstructions of paleoproductivity in the western AS [Naidu, 2004; Ivanochko *et al.*, 2005] and summer monsoon strength [Hong *et al.*, 2003] show declining trends from 5 ka to present (Figure 9). On the other hand, winter monsoon and productivity in the eastern AS were reduced during the Holocene until 3 ka BP (A. Singh *et al.*, manuscript in preparation, 2007) and hence cannot explain the Holocene increase in denitrification in the northeastern AS. We therefore invoke a causal role for ventilation changes.

[26] As proposed earlier (section 5.1), deglacial relative sea level rise caused a profound reorganization of the



**Figure 9.** (top to bottom) Nitrous oxide ( $\text{N}_2\text{O}$ ) concentration [Flückiger et al., 1999, 2004] and  $\delta^{15}\text{N}$  of  $\text{N}_2\text{O}$  (black diamonds [Sowers et al., 2003]) from ice cores, late deglacial changes in relative sea level [Bard et al., 1996], Ba/Al ratio in core 905P [Ivanochko et al., 2005], and  $\delta^{15}\text{N}$  records from cores ODP1017 [Hendy et al., 2004], ODP893 [Emmer and Thunell, 2000] from the North Pacific OMZ, and MD-04 2876 and 905P [Ivanochko et al., 2005] from the Indian OMZ.

intermediate/thermocline circulation in the northern AS; i.e., (1) increased inflow of oxygen-poor, saline waters from the southwest at crucial depths (200–300 m) with respect to denitrification in combination with (2) the expansion of a strong frontal system along the equator preventing oxygen-rich southern water masses from reaching the northern Indian Ocean (Figure 7). We suggest that this reorganization lowered the postglacial  $\text{O}_2$  inventory in the northeastern AS. The Red Sea and the Persian Gulf are separated from the open ocean by shallow and narrow sills (137 and <100 m water depth, respectively). Because of this sill, the Red Sea outflow was reduced by 85% during the last glacial period [Rohling and Zachariasse, 1996] and by around 50% at

10 ka BP compared to today, and increased progressively with sea level rise until it reached its present state at around 6 ka BP, gradually altering the  $\text{O}_2$  budget in the AS. We propose that the very sluggish circulation of Red Sea and Persian Gulf Waters further prevented the reduced southern source water from ventilating the northern AS and enables a continuous built up of oxygen deficiency in the northeastern part of the basin since the beginning of the Holocene.

## 6. Implication for Climatic Changes and Nitrous Oxide Emissions

[27] The global ocean accounts for one half of the total global  $\text{N}_2\text{O}$  source, with denitrifying regions such as the AS being the most important marine source [Suntharalingam et al., 2000]. Today, measurements of  $\text{N}_2\text{O}$  flux from the surface ocean to the atmosphere show that most of the AS emissions originate from the north and eastern part of the basin [Lal et al., 1996; Bange et al., 2001].

[28] Atmospheric [ $\text{N}_2\text{O}$ ] inferred from ice core records spanning the last hundred of ka have experienced large changes at both orbital and suborbital scales [Flückiger et al., 1999, 2004; Sowers et al., 2003]. These changes occurred in phase with denitrification variations in the world's OMZs, including the AS, implying a tight causal link between denitrification intensity,  $\text{N}_2\text{O}$  emissions and the Earth's climate through the past. During Dansgaard-Oeschger 9, contrary to preceding Dansgaard-Oeschger events, both denitrification intensity and atmospheric  $\text{N}_2\text{O}$  concentration remain low while Greenland ice cores record a prominent temperature increase at the time (see Figure 4 and Flückiger et al. [2004]). This decoupling emphasizes that the emission of greenhouse gas such as  $\text{N}_2\text{O}$  and  $\text{CH}_4$  contributes only partly to suborbital climate changes [cf. Flückiger et al., 2004]. The nitrogen isotopic signature of atmospheric  $\text{N}_2\text{O}$  reveals that in spite of these high-amplitude variations, the relative contributions of continental and oceanic sources have been fairly constant over the last glacial with the exception of the end of the YD, when continental source probably increased compared to marine  $\text{N}_2\text{O}$  production [Sowers et al., 2003].

[29] Over the Holocene, denitrification rates in most of the suboxic ocean declined in response to deglacial reduction in marine productivity [Deutsch et al., 2004]. This global decline in the organic carbon rain rate is expected to cause a decrease in  $\text{N}_2\text{O}$  production in the well-ventilated Ocean as well. Surprisingly, from 10 ka BP to present, [ $\text{N}_2\text{O}$ ] increases by 10 ppbv, while  $\delta^{15}\text{N}$ - $\text{N}_2\text{O}$  signal supports unchanged relative contribution of continental and marine sources or a slightly increased ocean source (Figure 9). These results suggest that the marine  $\text{N}_2\text{O}$  source increased locally in order to compensate for the overall decline in marine denitrification, and to explain the relative increase in [ $\text{N}_2\text{O}$ ] from 8 ka BP. The clear denitrification increase observed in the northeastern AS since ~8 ka BP may have had the potential to promote Nitrous oxide production in the context of global ocean productivity and denitrification slow down. This raises the possibility that the northeastern AS represents an important feedback on the

Holocene climate through sustained increase in N<sub>2</sub>O production and marine N loss.

## 7. Conclusions

[30] The regional comparison of denitrification records from across the AS illustrates the occurrence of regional heterogeneity in denitrification intensity within the basin, both across millennial-scale climate shifts and during the Holocene. We show that the OMZ experienced east-west swings across the abrupt climate changes and through the Holocene epoch. Ventilation changes due to millennial-scale variations in AAIW formation as well as circulation reorganization in the north Indian Ocean following sea level rise are probable causes of the observed regional differences in denitrification.

[31] This study further strengthens the evidence that AAIW exerts an important control on denitrification in the AS and on the N cycle as a whole [Schulte et al., 1999; Galbraith et al., 2004; Meissner et al., 2005]. Our study is based on very high resolution records and demonstrates both the very tight coupling between the ocean and the atmosphere across climate transitions and between circulation changes and denitrification in the AS.

[32] Moreover, our new  $\delta^{15}\text{N}$  record clearly shows intensified denitrification from about 8 ka BP in the northeastern AS whereas denitrification in world's OMZs including the western AS generally decreased following the deglacial maximum. We suggest that the northeastern AS influenced the global Holocene climate through sustained N<sub>2</sub>O production and marine nitrogen loss, partly offsetting the global ocean trends.

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