Orbital-scale denitrification changes in the Eastern Arabian Sea during the last 800 kyrs

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Denitrification in the Arabian Sea is closely related to the monsoon-induced upwelling and subsequent phytoplankton production in the surface water. The δ15N values of bulk sediments collected at Site U1456 of the International Ocean Discovery Program (IODP) Expedition 355 reveal the orbital-scale denitrification history in response to the Indian Monsoon. Age reconstruction based on the correlation of planktonic foraminifera (Globigerinoides ruber) δ18O values with the LR04 stack together with the shipboard biostratigraphic and paleomagnetic data assigns the study interval to be 1.2 Ma. Comparison of δ15N values during the last 800 kyrs between Site U1456 (Eastern Arabian Sea) and Site 722B (Western Arabian Sea) showed that δ15N values were high during interglacial periods, indicating intensified denitrification, while the opposite was observed during glacial periods. Taking 6‰ as the empirical threshold of denitrification, the Eastern Arabian Sea has experienced a persistent oxygen minimum zone (OMZ) to maintain strong denitrification whereas the Western Arabian Sea has undergone OMZ breakdown during some glacial periods. The results of this study also suggests that five principal oceanographic conditions were changed in response to the Indian Monsoon following the interglacial and glacial cycles, which controls the degree of denitrification in the Arabian Sea.

Denitrification occurs when nitrate is used as an alternate electron acceptor to break down organic matter when dissolved oxygen concentration is lower than 0.2 ml/L. During the denitrification, N2O and N2 are released into the atmosphere2,3. Thus, denitrification in the ocean plays an important role in the marine nitrogen cycle and global climate because it produces a greenhouse gas (N2O). OMZ develops in several oceans including the Eastern Tropical North Pacific, Eastern Tropical South Pacific, and Arabian Sea5,6. The OMZ in the Arabian Sea occurs between 200 to 1000 m in depth and accounts for more than 30% of the global denitrification process2,6. Denitrification in the Arabian Sea is generally associated with the monsoon climate, which leads to a seasonal reversal of wind pattern6–9. In the Western Arabian Sea, denitrification intensifies due to the enhanced primary production caused by strong upwelling which was induced by the southwest monsoon during summer7,10. During denitrification, lighter 14N-nitrate is utilized preferentially for the degradation of organic matter in the water column by bacteria, resulting in the increase of seawater nitrate δ15N values11. The controlling factors for δ15N value of marine sediments are diverse such as incomplete nitrate consumption12, Rayleigh isotopic fractionation13, early diagenesis14, and water column denitrification15. In general, the average δ15N value of global deep water is ~4.8‰ and increases with the degree of denitrification16. As a result, the δ15N values of seawater nitrate are higher than the average of global deep water when the degree of denitrification is strong in the Arabian Sea17. This change is reflected in the sediments below the water column by sinking of δ15N of sediment organic matter8,18. Previous studies reported that the empirical approach assigned ~6‰ as a reference or threshold δ15N value of sediment organic matter to judge the occurrence of denitrification19,20.

It is has been well known that the degree of denitrification fluctuates in response to the monsoon activity at seasonal-, millennial-, and orbital-scales in the Arabian Sea2,6,9,16,20–25 (Supplementary Figure S1). The Indian Monsoon system is one of the most distinctive features in the Arabian Sea. The southwest monsoon during summer develops by the low pressure in Peninsula India delivers rain26 and induces upwelling along the Oman Margin27. As a result, primary productivity increases28, seasonal development of the OMZ occurs2, and denitrification intensifies2. In contrast, the northeast monsoon prevails during winter26, causing upwelling along the west margin of India in a narrow portion of the Eastern Arabian Sea29. Thus, the δ15N value of sediment organic...

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matter in the Arabian Sea increased during the interstadial and interglacial periods when the summer monsoon was enhanced to cause denitrification and the expansion of OMZ. In contrast, denitrification was relatively weakened and the OMZ was reduced during the stadial and glacial periods, when the winter monsoon was strong, resulting in a low $\delta^{15}$N value of sediment organic matter near ~4‰. However, previous studies in the Arabian Sea that revealed the relationship between the $\delta^{15}$N value of sediment organic matter and the degree of denitrification were limited spatially to shallow marine areas and temporally to millennial-scales 6,17,20,22,23,25. In 2015, IODP Expedition 355 drilled the Laxmi Basin in the Eastern Arabian Sea in order to obtain long-term pale-oceanographic records related to development of the Arabian Sea monsoon (i.e., Indian Monsoon)30. This study is the first to report the orbital-scale denitrification change associated with the monsoon activity throughout the Mid-Pleistocene in the Eastern Arabian Sea and discusses their differences between the Western and Eastern Arabian Seas in terms of diverse oceanographic factors that control the degree of denitrification.

Study Area
IODP Expedition 355 Site U1456 is located in the Laxmi Basin of the Eastern Arabian Sea, within the mid-fan of the Indus Fan, which is the second largest fan system in the world (Fig. 1). The Indus Fan delta system is characterized by the repetition of turbidites that were formed by sediments supplied from various rivers adjacent to the Indian continent31. The Indus, Narmada and Tapti rivers in western India discharge more than 15,000 m$^3$/s of freshwater into the Eastern Arabian Sea annually32. The warm-saline oxygen-depleted Red Sea (Persian Gulf) Intermediate Water (RSIW or PGIW) flows into the intermediate depth of the Arabian Sea 33,34. The oxygen-rich Antarctic Intermediate Water (AAIW) also flows into the intermediate depth of the Arabian Sea from the south 34. The intrusion of AAIW into the Arabian Sea is inhibited by lateral development of low-salinity and oxygen-rich Banda Sea Intermediate Water during the warm or interglacial periods, and vice versa during cold or glacial periods35.

Results
The sedimentary sequence drilled at Site U1456 was divided onboard into four lithologic units based on visual descriptions, magnetic susceptibility and colour spectral analysis30. The uppermost Unit I consisted mostly of hemipelagic to pelagic ooze with occasional turbidites composed of sand, silt, clay, and mixtures (Supplementary Figure S2). These turbidite layers are characterized by poor preservation of planktonic foraminifera and erosional contact of sediments, which may be obstacles to construction of a precise age model. Under such circumstances, the age of Unit I was mainly determined by the correlation of $\delta^{18}$O values of planktonic foraminifera (Globigerinoides ruber) with the LR04 stack36 supplement to the consideration of shipboard biostratigraphic and paleomagnetic data (Fig. 2). The 37 tie points are determined by correlating $\delta^{18}$O values to the LR04 stack36, with an additional correlation of $\delta^{15}$N values to ODP Leg 117 Site 722B 38 in the Western Arabian Sea (Supplementary Figure S3). Although the age reference of Site 722B is different from the LR04 stack37, the minor offset between them seems insignificant. The precise age determination is described in detail in Supplementary Figure S3. As a result, the age of Unit I spans as old as 1.2 Ma, covering up to Marine Isotope Stage (MIS) 36 (Fig. 2). The oxygen isotope stratigraphy of Unit I shows that the quasi-cyclic pattern is characterized by 100-ka periodicity (Supplementary Figure S4), which is prominently more visible since the Mid-Pleistocene Transition (MPT). Thus, this study focuses particularly on the last 800 krys during which the glacial-interglacial cycles are distinctly discernible.

The $\delta^{15}$N values of bulk sediments at Site U1456, representing the degree of denitrification in the Eastern Arabian Sea, range from 5.3 to 10.1‰ with an average of 7.5‰ during the last 800 krys (Fig. 3). The $\delta^{15}$N values...
at Site 722B in the Western Arabian Sea fluctuate similarly between 4.1 and 10.8‰ with an average of 6.6‰ (Fig. 3). The δ¹⁵N variation between Site U1456 and Site 722B is comparable following glacial-interglacial changes, being characterized by high δ¹⁵N values during the interglacial periods indicating active denitrification and showing the opposite during glacial periods. Such data of δ¹⁵N from Site U1456 shows the 100-ka periodicity that is displayed in glacial-interglacial variations that are possible to be correlated to the orbital-scale eccentricity cycle. Such cyclicity is evident especially before the MPT for both δ¹⁸O and δ¹⁵N.

Discussion

Tripathi et al. referred to 6‰ as indicative of the initiation of denitrification during the Pliocene and Miocene at Site U1456 in the Eastern Arabian Sea. Based on our results, the δ¹⁵N values at Site U1456 rarely fall below 6‰ (Fig. 3), which clearly indicates that denitrification occurred continuously during the last 800 kyr in the Eastern Arabian Sea. On the contrary, the δ¹⁵N values at Site 722B are less than 6‰ during sporadic intervals, corresponding to the glacial periods. These findings indicate that denitrification has collapsed frequently during cold periods in the Western Arabian Sea.

Nitrate utilization in the surface water affects the resultant δ¹⁵N value of sediment organic matter. The nitrate concentration of the surface water between the Western and Eastern Arabian Sea offsets ~2 μM.
reflecting almost complete nitrate utilization during the phytoplankton bloom. The offset may increase seasonally and during the production, suggesting that variability of δ¹⁵N values would be predominantly driven by changes in nitrate consumption and not the initial nitrate δ¹⁵N value (partially set by water column denitrification). Thus, the δ¹⁵N values of sediment organic matter were influenced by the extent of denitrification within the OMZ, although nitrate utilization during the glacial periods was assumed to be similar to the present-day utilization. Hence, we accept the δ¹⁵N values of bulk sediments to compare the degree of denitrification between the Western (Site 722B) and Eastern (Site U1456) Arabian Sea. Because early diagenesis causes an increase of δ¹⁵N value during the degradation of sediment organic matter, the possible effects of early diagenesis need to be evaluated to confirm that the alteration of δ¹⁵N values was caused solely by water column denitrification. For example, Pichevin et al. reported that the δ¹⁵N value of sediment organic matter was affected by the depth of sedimentation and the degree of sediment organic matter degradation, both of which are related to early diagenesis. At Site U1456, the effect of early diagenesis was estimated by comparing the δ¹⁵N value with the total nitrogen (TN) content (Supplementary Figure S5). The lack of correlation between two parameters demonstrates that early diagenesis has not been a significant factor to raise δ¹⁵N values. Hence, the δ¹⁵N values of bulk sediments at Site U1456 are related to the degree of denitrification and development of OMZ within the water column.

Denitrification in the Eastern Arabian Sea during the last 800 kyrs has clearly varied between glacial and interglacial periods, which is closely related to the orbital-scale global climate changes (Fig. 3). Hence, we compared several monsoon-related proxies to evaluate the correspondence between denitrification and monsoon intensity. The degree of denitrification at Site 722B in the Western Arabian Sea has also changed comparably to that in the Eastern Arabian Sea. XRF-scanned Ba counts from core MD04-2881 collected in the Northeastern Arabian Sea.
increased during warm interglacial periods because the southwest monsoon drove the intense mixture between the surface and subsurface waters, resulting in enhanced primary productivity. The increased production of organic matter promotes denitrification via more consumption of dissolved oxygen during degradation process. This relationship between surface water productivity and denitrification has also been recognized in the Eastern Tropical North Pacific and the Eastern Tropical South Pacific. The Chinese loess plateau in south central China records glacial-interglacial fluctuations of the East Asian Monsoon which is closely linked to the Indian Monsoon. Magnetic susceptibility (MS) during arid and cold glacial periods was low due to increased wind-blown loess, whereas MS during wet and warm interglacial periods was high because of soil-producing weathering. Methane concentrations measured from EPICA Dome C (Antarctica) are closely linked to monsoon precipitation. The strong global monsoon-induced precipitation expanded swamps and wetland in continents to release more methane into the atmosphere. Thus, global paleoclimate proxies related to the southwest monsoon were maximized during warm interglacial periods in the Arabian Sea, while those related to the northeast monsoon were minimized during cold glacial periods, which supports our finding that δ¹⁵N values of bulk sediments indicate the degree of denitrification.

The degree of denitrification in the Arabian Sea is primarily controlled by several oceanographic factors linked with glacial-interglacial cycles. Figure 4 provides a schematic model demonstrating that the degree of denitrification changed between the interglacial and glacial periods in the Arabian Sea. First, the driving force of OMZ development is closely related to the seasonally-reversing monsoon winds. The southwest monsoon during summer causes seasonal upwelling in the Western Arabian Sea. Similarly, during the warm interglacial periods, the degree of denitrification increased in the Western Arabian Sea. However, the prevailing northeast monsoon during the cold glacial periods caused downwelling in the Western Arabian Sea and upwelling in the Eastern Arabian Sea. Thus, the monsoon-derived upwelling in the Eastern Arabian Sea maintained the perennial OMZ, while the seasonal OMZ collapsed in the Western Arabian Sea. Second, the clockwise-flowing Somali Current plays an important role in transporting oxygen-depleted water throughout the Arabian Sea during the

**Figure 4.** Model of interglacial-glacial denitrification change in association with development of the OMZ in the Arabian Sea. The intensity of the OMZ differs between interglacial and glacial periods. A thicker OMZ and intensified denitrification occurred during interglacial periods, and while the opposite occurred during glacial periods. Because the seasonal OMZ during the glacial periods was weak, denitrification collapsed in the Western Arabian Sea. In contrast, the perennial OMZ has maintained strong denitrification in the Eastern Arabian Sea as indicated by δ¹⁵N values greater than 6‰.
summer monsoon28,30 (Fig. 1). Thus, the surface current during the interglacial periods promoted wide OMZ for
denitrification throughout the basin27. However, the role of the Somali Current was reduced during glacial peri-
ods due to the northeast monsoon. The third component controlling the degree of denitrification is the supply of
oxygen−depleted waters by the RSIW. During interglacial periods, the RSIW influx from the Arabian Peninsula
supplied oxygen−depleted waters to the intermediate layer in the Arabian Sea, which elevated the degree of deni-
trification10,33,34. However, during glacial periods when the sea level was lower than the sill depth of the straits,
the input of the RSIW ceased, resulting in the reduced denitrification. The fourth factor is the low contribution
of relatively oxygen−rich AAIW to the Arabian Sea during the interglacial periods as a result of development of
the Banda Sea Intermediate Water. However, when the Banda Sea Intermediate Water was weak during the cold
glacial periods, denitrification decreased because of the greater influence of the AAIW, which was able to sup-
ply the dissolved oxygen14,51,52. Finally, salinity stratification due to increased monsoon precipitation during the
southwest monsoon limited the oxygen exchange between the subsurface water and the atmosphere, intensifying
the degree of denitrification. In contrast, such stratification was weakened when riverine freshwater discharge
decreased during the arid winter monsoon19.

In conclusion, the OMZ and denitrification have developed actively to a greater extent during interglacial
periods, which are mainly characterized by a strong southwest monsoon in the Arabian Sea. The seasonal OMZ
in the Western Arabian Sea collapsed due to the diverse environmental factors during the glacial periods. In
contrast, perennial OMZ was maintained in the Eastern Arabian Sea, as indicated by most δ15N values of bulk
sediments at Site U1456 being higher than the δ15N denitrification threshold during the last 800 kyr. The deni-
trification in the Arabian Sea has also changed in response to climatic fluctuations following the 100-ka cyclicity
represented by global δ18O values and other multi-proxy signals. Our results emphasize that the Eastern Arabian Sea
has experienced persistent denitrification without prominent collapse throughout the Mid−Pleistocene.

Methods

For the present study, a total of 260 samples were collected from a composite section of Unit I consisting of
Holes U1456A and U1456C at Site U1456. The δ18O values were measured from planktonic foraminifera
(Globigerinoides ruber) with a test size between 250 and 355 μm using IsoPrime at Center for Advanced Marine
Core Research of Kochi University (Japan). Cleaning of foraminifera tests was conducted according to Barker
et al.34. Oxygen isotope ratios were calibrated to the V−PDB standard using international standard NBS19. The
analytical precision of the δ18O values is ±0.06‰. The δ15N values of 173 bulk sediments from Unit I were mea-
ured using EA−IRMS at Iso−Analytical Ltd. (UK). All δ15N values were calibrated to δ15Nair and the precision was
about ±0.1‰. The total nitrogen (TN) content of bulk sediments was measured using CHN Elemental Analyzer
(Flash 2000 Model) at Pusan National University (Korea). The analytical precision of TN was ±0.1‰. All analyti-
cal data are summarized in Supplement Data File.

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B.K.K. designed the research and collected the samples onboard *JOIDES Resolution* aided by IODP Expedition 355 Scientists; J.E.K., M.I., and J.L. contributed to the sample processing and analytical measurement; J.E.K. and B.K.K. wrote the manuscript, which was discussed with M.I. and J.L.

**Additional Information**

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